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CASE STUDY

Harbor water pollution by heavy metal concentrations in sediments

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ABSTRACT

BACKGROUND AND OBJECTIVES: *The Belawan Harbor is the third largest port, which is located in an estuary, causing the port water area to be vulnerable to pollution, especially heavy metals. Conflicts between the community and the port authorities often occur due to pollution. Heavy metals are dangerous contaminants for waters, and total organic carbon in waters is needed but will cause eutrophication if the concentration is excessive in the environment. The level of heavy metal pollution in the waters of the Belawan Harbor and the factors that cause the pollution should be analyzed, because the level of heavy metal pollution has not been measured in the sediments of harbor waters. This study can be used as a reference for the actions of related agencies in dealing with heavy metal pollution in waters.

METHODS: *Sampling of sediments was performed at 10 locations, starting before the harbor activity began and moving toward the open sea. Sampling was conducted using Van Veen grab. Heavy metal concentrations were analyzed in the laboratory using the atomic absorption spectrometer method to assess the essential heavy metal copper and non-essential heavy metal lead, cadmium, and mercury. Heavy metal pollution in sediments was assessed by analyzing sediment pollution index. The multivariate statistical analysis on the relationship among factors was conducted using Pearson correlation matrix method, principal component analysis, and cluster analysis.

FINDINGS: *The environmental quality standards used indicate average concentration of heavy metals; lead (28,869 milligram per kilogram) and copper (8,003 milligram per kilogram) are below the quality standard. The mercury concentrations are undetectable (<0.00011 milligram per kilogram) at each station. By comparison, the concentration of cadmium (1,455 milligram per kilogram) exceeded the Interim Sediment Quality Guidelines from the Canadian Council of Ministers of the Environment. Results of the index analysis show that the average value of the pollution factor of copper is −0.177 (low contamination), that of lead is −1.433 (moderate contamination), and that of cadmium is −4.850 (high contamination); the geoaccumulation index value of copper is −5.328. (not polluted), that of lead is −0.190 (unpolluted), and that of cadmium −1.657 (moderately polluted). As mercury concentration in sediments is relatively low, it is not considered when calculating pollution levels. Overall, on the basis of a pollution index of 1.033 ($1 < \text{pollution load index} \leq 2$), this condition indicates that the waters of the Belawan Harbor are categorized as not polluted to lightly polluted. The highest total organic carbon is at the estuaries of the Belawan and Deli Rivers. The sediment fraction is 72.2 percent sandy, 16.4 percent sludge, and 11.4 percent clay substrate.

CONCLUSION: Pollution in the waters of the Belawan Harbor is in the category of not polluted to slightly polluted. Although the pollution is still in the light category, this must be of particular concern to the relevant agencies, especially the local government, to make the right policies to overcome this pollution immediately. Pollution problems increase with the anthropogenic activities around coastal areas, as well as activities in the Belawan and Deli River watersheds, because the pollutant will flow from the upstream to the estuary area.

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INTRODUCTION

The Belawan Harbor is Indonesia's third busiest harbor and is the gateway to the economy of Medan City, North Sumatra. The working area (DLKr) is 12072.33 hectare (ha), which has more than one base and more than one terminal. On the basis of the existing data in the Indonesian central statistic agency; Badan Pusat Statistik BPS, in 2019, The Belawan Harbor loads and unloads 11,979,268 tons per year, and it has 128,396 passengers overall in annual arrivals and departures. Harbors have various supporting facilities, such as passenger terminals, international container terminals, ship repair sites, storage tanks for hazardous and toxic materials, and offices. Various harbor activities have a negative impact on the environment, especially harbor waters. Pollutants from various sources, such as manufacturing processes, passenger waste, oil spills, terminal construction, and ballast water, can affect the aquatic environment. The maintenance and repair of ships near harbors also contribute to water pollution (Pourabadehei and Mulligan, 2016a; Salam et al., 2019). The Belawan Harbor is unique, because it is located between two major river estuaries, namely, Belawan and Deli Rivers. The pollution in the waters of the Belawan Harbor is caused by anthropogenic activities, either directly or indirectly, directly or indirectly, in the watersheds of Belawan and Deli Rivers. These activities include industrial, agricultural, and urban activities. Human interference, tourism, and fisheries are potential long-term sources of organic and inorganic pollutants that can degrade harbor sediments and increase the levels of polycyclic aromatic hydrocarbons and metals (Edge *et al*., 2021; Ojemaye and Petrik, 2019). Due to its location at the confluence of estuaries, the waters of the Belawan Harbor are vulnerable to pollution because of the settling of various pollutant types (Hanggara *et al*., 2021). The Belawan Harbor, which is located in the river estuary area, is often faced with social conflicts from the community because it is considered a contributor to pollution around the waters. For example, communities complain about the difficulty in catching fish around Belawan waters, which makes it challenging for fishermen to support their livelihoods. Heavy metal contamination in waters will make endemic fish migrate to uncontaminated waters, but species such as clams, shrimp, and crabs will survive in that environment. If humans consume contaminated aquatic species, then they will accumulate in their bodies and eventually have an adverse effect on human health (Pourabadehei and Mulligan, 2016a; Zhu *et al*., 2022). According to Chen *et al*. (2016), the steel, chemical, and plastic industries—all of which are located in watersheds that also produce industrial wastewater and domestic waste—are the main sources of heavy metal pollution (Araiza-Aguilar *et al*., 2020; Karbassi *et al*., 2015). Medan industrial Park and other urban activities can be found along Belawan and Deli Rivers, which may result in the entry of heavy metals in to Belawan Harbor's waters, and heavy metal particles will be deposited in the sediment due to tidal activity (Mignard *et al*., 2017). The dangers associated with heavy metal contamination, which will have an impact on continuing social conflicts, will be in conflict with the goals of sustainable development, which considers environmental, economic, and social aspects of society. Hence, whether Belawan Harbor's waters are contaminated by heavy metals must be determined. The analysis in question calculates heavy metal concentrations in sediments because sediment deposits, as opposed to water columns, are more reliable indicators of heavy metal contamination in waters (Rochyatun and Rozak, 2008; Banu *et al*., 2013). Given that concentration analysis and studies on heavy metal pollution have never been conducted in the sediments of Belawan Harbor waters, the concentration of heavy metals in sediments, the level of heavy metal pollution, and the influence of factors affecting heavy metal contaminants in sediments must be investigated. Non-essential heavy metals have essentially less benefit from being present in water. Thus, only one form of necessary heavy metal, namely, copper (Cu), and three types of non-essential heavy metals, lead (Pb), cadmium (Cd), and mercury (Hg), were chosen to be measured. This study aims to 1) analyze the concentration of heavy metals in the sediments of Belawan Harbor waters; 2) assess the level of heavy metal pollution in sediments; and 3) examine the factors that affect heavy metal contamination in harbor water sediments. The study was conducted in the Belawan Medan Harbor in November 2018 and January 2019.

MATERIALS AND METHODS

The geographical location of Belawan Harbor block extends from 03°47′N to 98°42′E. Sediment samples were collected from 10 locations in Belawan Harbor, Medan, North Sumatra (Fig. 1). Ten locations

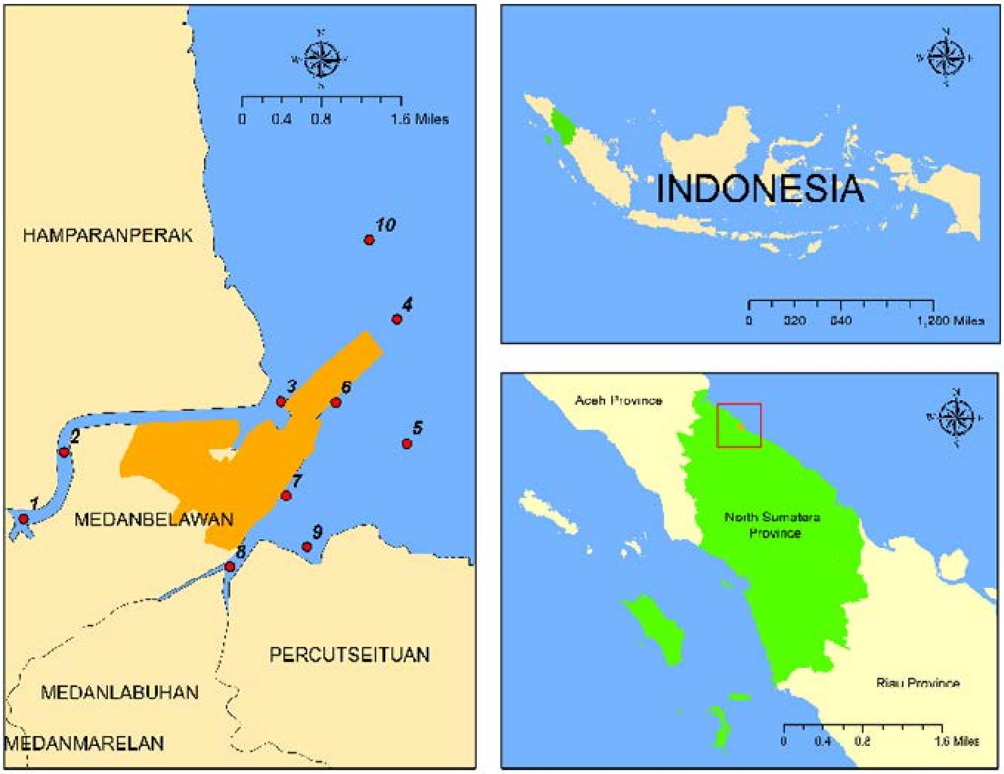

Fig. 1: Geographic location of the study area in Belawan Medan Harbor along with the sediment sampling locations for heavy metals

Table 1: Location of water sediment parameter collection

Site	Description	Coordinates	
		Latitude	Longitude
1	Belawan River Estuary	3°46’27, 39”NL	98°40’26, 15”EL
2	Passenger terminal-LANTAMAL	3°47’02, 63”NL	98°40’47, 55”EL
3	Unloading dock	3°47’29, 11”NL	98°42’41, 42”EL
4	Reclamation project	3°48’12, 20”NL	98°43’42, 50”EL
5	Deli River Sea	3°47’07, 10”NL	98°43’47, 70”EL
6	Pacific Ocean Tour	3°47’28, 60”NL	98°43’10, 30”EL
7	Belawan fishing harbor (PPS)	3°46’39, 40”NL	98°42’44, 30”EL
8	Deli River Estuary I	3°46’02, 28”NL	98°42’14, 77”EL
9	Deli River Estuary II	3°46’12, 70”NL	98°42’55, 20”EL
10	Belawan River Sea	3°48’53, 90”NL	98°43’27, 90”EL

in Belawan Medan Harbor’s DLKr and DLKp waters served as sampling points. Details description of the site sites seen in Table 1.

Targeted sampling was used to pinpoint the area between the Belawan and Deli rivers. The areas encompass the sea around Belawan Harbor and span the time before and after harbor activities. Global positioning system coordinates were used to pinpoint the locations of samples.

Sampling and handling sediment

At low tide, sediment samples were collected using the Van Veen grab from a boat. The tool was lowered to the bottom of the water, and sediment samples were then taken from the upper layers (depths between 0 centimeter: cm and 250 cm). Sediment samples were packed in polyethylene containers with a volume of 250 mL, which had been sterilized beforehand with 95% alcohol and distilled water and then put into an ice box. The sample was then taken to the laboratory for analysis. After being put in the laboratory in the freezer with a temperature of −20 °C, the heavy metal content was measured using a spectrophotometer using the ASTM C1301-95 Reav 2001 testing method. The sediment sample was used for chemical analysis, which consisted of digesting aliquots of 5 g by adding 2 mL of 4:1 of nitric acid: perchloric acid (HNO_3: $HClO_4$) solution to the samples for 3 h at 140 °C (Yap and Pang, 2011). After the sample had cooled, a 250 mL Erlenmeyer tube was prepared, in which the sample was added and then diluted with 20 mL of double distilled water and shaken gently. The sediment sample was filtered with Whatman paper, and the filtered results were put into a 100 mL volumetric flask and up to 50 mL of distilled water was added. Then, the sediment sample was ready to be measured using atomic absorption spectrometry (AAS). The AAS method is generally used for the analysis of heavy metal content because it is characterized by good precision and accuracy, as well as a percentage recovery of more than 90% of the amount of analyte (Feist *et al*., 2007). Strict quality control was maintained during the experiment. The blank samples and standard substances were progressed using the same method. Four duplicates were used for each sample. The repeat sample analysis error and the analytical precision for replicate samples were below 2% the relative standard deviation (SD). Method of blank operation showed no detectable metals (Men *et al*., 2018).

Pollution index calculation

Heavy metal pollution evaluation was conducted using Pb, Cu, Cd, and Hg geoaccumulation index (Igeo), pollution load index (PLI), and contamination factor (CF). The Igeo value is calculated using Eq. 1 (Shams El-Din *et al*., 2014; Rabee *et al*., 2011; Veerasingam *et al*., 2012).

$$\text{Igeo} = \log_2\left[\frac{C_n}{1.5B_n}\right], \tag{1}$$

where Cn is the measured concentration of metal n in the sediment, and Bn is the geochemical background values of the metal *n*. Factor 1.5 compensates for possible fluctuations in the background values for a given substance in the environment, as well as highly anthropogenic influences. The Igeo consists of seven classes: Igeo ≤ 0 is Class 0 (unpolluted); 0 < Igeo < 1 is Class 1 (unpolluted to moderately polluted); 2 < Igeo < 3 is Class 3 (moderately to heavily polluted); 2 < Igeo < 4 is Class 4 (heavily polluted); 4 < Igeo < 5 is Class 5 (heavily to extremely polluted); and 5 > Igeo is Class 6 (extremely polluted; Ke *et al*., 2017; Dai *et al*., 2018). For each observation site, the level of CF is calculated using Eq. 2 (Lars, 1980).

$$\text{CF} = \text{C heavy metal}/\text{C background}. \tag{2}$$

Where, C heavy metal is the concentration of metals in the sediment samples, and C background is the ambient environmental metal concentration (background metal concentration; Earth's crust; Effendi *et al*., 2017; Ogbeibu *et al*., 2014). Comparison between normal concentrations of metals (C background) in nature according by Turekian and Wedepohl (1961), namely Cu = 45 mg/kg, Pb = 20 mg/kg, Cd = 0.3 mg/kg, and Hg = 0.4 mg/kg. The CF values were interpreted as follows: low contamination (CF < 1), moderate contamination (3 < CF < 6), and extremely high contamination (CF > 6). PLI is calculated using Eq. 3 (Sivakumar *et al*., 2016).

$$\text{PLI} = (\text{CF1} \times \text{CF2} \times \text{CF3} \times \ldots\ldots \text{CFn})^{1/n}. \tag{3}$$

TomLinson *et al.* (1980) were the first to use PLI. The CF multiplied by the number of heavy metals n yields CF * n. The PLI method was used in the study of heavy metal contamination. The PLI values were classified into four groups: no pollution load (PLI < 1), low pollution load (1 < PLI < 3), moderate pollution load (3 < PLI < 6), and high pollution load (6 < PLI).

Statistical analysis

Pearson's correlation matrix is a statistical technique for illustrating associations between sets of data. The degree of significance is used in decision making when the significance value is correlated ($p < 0.05$) or when it is not correlated ($p > 0.05$). If R^2 equals 1, then the relationship is completely positive; if it equals −1, then the relationship is completely negative. Low (0.00–0.20), moderate (0.40–0.60), high (0.60–0.80), and extremely high (1.0–1.0) are the typical ranges assigned to correlation coefficients (0.80–1.00; Bush and Guilford, 1956). Principal component analysis (PCA) is widely used in conducting research on the environment (Abollino *et al.*, 2003; Lucho-Constantino *et al.*, 2005; Pourabadehei and Mulligan, 2016b; Rognerud and Fjeld, 2001; Sundaray *et al.*, 2011). Descriptive statistics, such as PCA, are useful for elucidating as much information as possible inside a data matrix. Specifically, the accompanying data matrix has rows that represent study sites and columns, which represent quantitative data for environmental factors (physical chemistry). By comparing the contamination intensity (Igeo) of sediment samples, cluster analysis (CA) was used to classify contaminated and clean sites. Statistics were calculated using Excel 2016 and XLSTAT 2022.

RESULTS AND DISCUSSION

Table 2 displays the percentage results of the sediment fraction. The proportion of sand (72.20% on average) is the most dominant in aquatic sediments. Total organic carbon (TOC) is important for analysis; it is considered a modifier of sediment toxicity because it can change contaminant distribution and bioavailability (Watson-Leung and Picard, 2016). The TOC concentration in the sediment around Belawan Harbor waters varies between 0.26% and 2.74%. The categories of TOC assessment in sediments are low = TOC ≤ 1%, moderate = 1 < TOC <3%, and high = TOC > 3% (USEPA, 2002). The average TOC value measured in the Belawan Harbor waters is in the low category at 1.74%. The highest TOC value is at Station 2; this station is at the DLKr, that is, at the passenger terminal. The high TOC concentration in this area is caused by the changes in the flow of river water toward the sea resulting from bends in the river flow. The existence of river water bends in the estuary due to harbor activities can affect the river water discharge, and the sediment transported from the upstream river will significantly be retained in that area (Miao *et al.*, 2010; Zheng *et al.*, 2019). Generally, marine organic matter has TOC/total nitrogen (TN) values of 4–10, and the TOC/TN values of terrestrial plants are 20 and above (Meyers, 1994; Lamb *et al.*, 2006). The TOC/TN values of marine phytoplankton are close to 6.6 (Redfield *et al.*, 1963). The TOC/TN ratios of fresh vascular plants and their degraded detritus are 23–1,560 and 14–47, respectively (Tyson, 1995; Usui *et al.*, 2006). The TOC/TN ratios in soils generally range from 8 to 15 (Kendall *et al.*, 2001). The highest average TOC concentration at each sites does not have considerably different values except at Sites 4 and 5, which are reclamation areas and seas off. The sediment substrate affects the TOC concentration, as observed in sites that have sandy substrates with a lower TOC concentration. This phenomenon will be analyzed further in a multivariate statistical analysis. Extracted organic matter may come from natural or manmade activities. Natural sources of organic compounds can be terrestrial and aquatic organisms

Table 2: Physicochemical concentration of sediment in Belawan Harbor waters

Site	Heavy metal concentration (mg/kg)				Sand	Sludge	Clay	TOC
	Pb	Cu	Cd	Hg				
1	20.99	5.64	0.90	<0.00011	67	15	18	2.10
2	24.69	5.80	1.37	<0.00011	69	17	14	2.74
3	19.82	3.42	1.32	<0.00011	67	19	14	1.42
4	15.50	6.86	1.22	<0.00011	99	1	0	0.26
5	23.97	1.68	1.30	<0.00011	79	9	12	0.76
6	31.90	4.53	1.53	<0.00011	61	29	10	2.09
7	28.00	16.24	1.35	<0.00011	69	19	12	1.89
8	68.43	21.49	2.10	<0.00011	61	23	16	2.65
9	29.46	9.82	1.92	<0.00011	77	15	8	1.96
10	25.93	4.55	1.54	<0.00011	73	17	10	1.54
Minimum (Min.)	15.5	1.68	0.9	<0.00011	61	1	0	0.26
Maximum (Max.)	68.43	21.49	2.1	<0.00011	99	29	18	2.74
Average	28.869	8.003	1.455	<0.00011	72.200	16.400	11.400	1.741
SD	14.712	6.234	0.3446	-	11.1235	7.5454	4.9933	0.7778
Interim Sediment Quality Guidelines (ISQG)								
CCME (2002)	ISQG	30.2	18.7	0.13	0.7			
	PEL	112	108	0.7	4.2			
ANZECC/ARMCANC (2000)	Low	50	65	0.15	1.5			
	High	220	270	1	10			

(Gao *et al*., 2012).

Belawan Harbor water sediments contain Pb concentrations of 15.5–68.43 mg/kg, Cu concentrations of 1.68–21.49 mg/kg, and Cd concentrations of 0.9–2.1 mg/kg; conversely, Hg concentrations are not detected because the value is relatively small (below 0.00011 mg/kg), but the concentration is not 0 in the sediment. The data obtained show that in this case, Hg is considered not to pollute and not harmful to the aquatic life in Belawan Harbor. The heavy metal concentrations of Pb, Cu, Cd, and Hg were compared with the sediment quality standards from the Canadian Council of Ministers of the Environment (CCME, 2002) and the quality standard guidelines from Australia and New Zealand (ANZECC/ARMCANZ, 2000). The obtained metal concentrations were compared using the two quality standard guidelines from other countries because Indonesia does not have its own quality standard guidelines yet. A small set of quality benchmarks for the heavy metal concentration represents the heavy metal pollutant load. The heavy metal concentrations of Pb (28,869 mg/kg) and Cu (8,003 mg/kg) were below the quality limits, according to the CCME and ANZECC/ARMCANZ (2000) sediment quality standards. Moreover, the average Cd concentration (1,455 mg/kg) exceeds the CCME (2002) ISQG and is becoming close to the low thresholds of ANZECC/ARMCANZ.

The concentration levels in other metal-polluted harbors from different regions of the world are shown in Table 3 to understand the extent of metal pollutions in the harbor area of this study. The contaminant of Cd in the Belawan Harbor sediments for all observation stations is above the quality standard threshold of the CCME (2002) compared with other heavy metals. The Cd concentration in this study is not considerably different from that in the Gironde Estuary, Yangtze River Estuary. Two harbors on the Egyptian Mediterranean Coast with a relatively high Cd content compared with other harbors worldwide exhibit different characteristics in their Cd concentration. This finding supports the claim that sediment contamination in Belawan Harbor waters is influenced by the rate of pollutants produced from anthropogenic activities that enter through the Belawan and Deli Rivers. Overall, the Pb, Cu, Cd, and Hg concentrations in the sediments of Belawan Harbor are relatively lower compared with other harbors in the world. This phenomenon is influenced by the level of harbor activities and dense urban activities around harbors, such as Rize Harbor, 12 South Korea harbors, Eastern Harbor, and Kaohsiung Harbor. The Belawan Harbor is still the third busiest harbor in Indonesia. Thus, heavy metal pollution can be caused by development in many sectors (e.g., industrial, agriculture, urbanization, and navigation), accompanied by the high rate of population growth that acts as environmental stressors on aquatic systems, particularly for coastal countries (Ebeid *et al*., 2022). Overall, the extent of metal pollution in the sediments of the Belawan Harbor is comparable to those found in other metal-contaminated harbors worldwide. Table 4 shows the Pb, Cu, and Cd values for pollution factor, geological accumulation index, and pollution index.

The average CF value of Pb is 1.4437, which is more than 1 (1 < CF < 3), so the Pb contamination level is moderate. The Igeo of Pb typically falls in the range of −0.1907 to 0; the sediment has an unpolluted status because this value is less than 0 (Igeo < 0, Class 1). The CF value of Cu has an average value of 0.1778, which is less than 1 (CF < 1), so the sediment contamination of Cu in Belawan Harbor is considered

Table 3: Concentration ranges of heavy metals reported in Belawan Harbor compared with other regions of the world

Regions	Pb	Cu	Cd	Hg	References
Gironde Estuary, France	5.0–84	0.5–40	0.01–2.1	0.001–0.37	Larrose *et al*. (2010)
Rize Harbor, Turkey	15.9–33.0	33.9–279.1	0.1–1.4	0.01–0.07	Gedik and Boran (2012)
12 South Korea harbors	20–599	5–2,360	0.03–15	0.003–3	Choi *et al*. (2012)
Eastern Harbor, Egypt	1.3–112	3.80–129	0.3–1.8	-	Ghani *et al*. (2013)
Kaohsiung Harbor, Taiwan	14–219	12–2,840	0.05–7.3	0.03–10	Chen *et al*. (2016)
Yangtze River Estuary, China	1.12–2.20	1.12–71.80	0.02–2.20	0.001–0.556	Li *et al*. (2019)
Pearl River Estuary, China	36.1–51.2	26–47.7	0.29–0.82	-	Ye *et al*. (2020)
2 harbors in the Egyptian Mediterranean Coast	50.5–134.4	828–1,537	26.1–59.5	-	Ebeid *et al*. (2022)
Belawan Harbor, Indonesia	15.5–68.43	1.68–21.49	0.9–2.1	<0.00011	This study

to be relatively low. The Igeo of Cu has a mean value of −3.4327, which is lower than 0, so the sediment does not contain Cu contaminant (Igeo < 0, Class 1). The CF value of Cd is 4.850, which is greater than 4 (3 < CF < 6), indicating a highly contaminated sediment level. The mean Igeo value is 1,657, which is greater than 1 and less than 2, so Belawan Harbor's sediment is considered to be highly contaminated (1 < Igeo < 2, Class 3). The obtained values for Igeo can be negative or positive. The results demonstrate that the sediment in these waters is cleaner (less polluted) when the Igeo is negative, whereas a higher value indicates that the sediment is more contaminated (polluted). The PLI analysis result, with values ranging from 0.579 to 2.2253, show an average value of 1.033, which is between 1 and 3, so the Belawan Harbor has a low pollution load. The human activities in DLKr and DLKp in Belawan Harbor have been increasing every year. As a local industrial cluster develops in the Deli River watershed, a harbor reclamation project (site 4) is changing the shoreline in a manner that traps airflow and sedimentary material at the river mouth. The Deli River watershed, from which the wastewater flows into the harbor area, has three of the four indicated pollutants. These contaminants may have been introduced by illicitly generated industrial output effluent (Sites 7–9). The Environment Agency of North Sumatra estimated in 2019 that there would be 54 industrial and 27 household waste canals along the Deli River. Businesses from various sectors (e.g., painting and electroplating, loose iron, food processing, and food and beverage sectors) have established themselves along the Deli River. One way in which manufacturing contributes to air pollution is through the use of Cd in the raw material trade or as a carrier material in the industry (Masum and Pal, 2020). Of all Cd, 75% of it is used to make batteries (especially Ni–Cd batteries). Pigment mixtures containing this metal are used in the production of ceramics, electroplating, low-melting-point alloys, nuclear-reaction cleavage, paint, metal coatings, semiconductors, PVC stabilizers, and cigarettes; but they are not useful in the production of phosphate fertilizers or anti-malarial drugs, such as those used to treat syphilis and malaria (Bonberg *et al*., 2017; Hill *et al*., 2010; Rodrigues *et al*., 2020). After entering the river, this toxic metal is carried by the tides to the estuary's sediments. Restoring a harbor can affect more than just the shoreline; it can also modify the oxygen content of the soils and sediments below the water. Hence, in biologically polluted estuaries (Sites 7–9), heavy metals are absorbed by the sediment substrate; especially, the silt sediment fraction has better absorption (Chen and Jiao, 2008). Heavy metal contamination in sediments is highly influenced by anthropogenic activities, which include allowing the construction of houses and businesses near watersheds and orienting most buildings almost fully away from the river. The procurement of domestic wastewater treatment plants (WWTPs) has also not

Table 4: Analysis results of CF, Igeo, and PLI of Belawan Harbor sediments

Site	Pb		Cu		Cd		PLI
	CF	Igeo	CF	Igeo	CF	Igeo	
1	1.050	−0.515	0.125	−3.581	3.000	1.000	0.733
2	1.235	−0.281	0.129	−3.541	4.567	1.606	0.899
3	0.991	−0.598	0.076	−4.303	4.400	1.553	0.692
4	0.775	−0.954	0.152	−3.299	4.067	1.439	0.783
5	1.199	−0.324	0.037	−5.328	4.333	1.531	0.579
6	1.595	−0.089	0.101	−3.897	5.100	1.766	0.936
7	1.400	−0.100	0.361	−2.055	4.500	1.585	1.315
8	3.422	1.190	0.478	−1.651	7.000	2.222	2.253
9	1.473	−0.026	0.218	−2.781	6.400	2.093	1.272
10	1.297	−0210	0.101	−3.891	5.133	1.775	0.876
Min.	0.775	−0.954	0.037	−5.328	3	1	0.579
Max.	3.422	1.190	0.478	−1.651	7	2.222	2.253
Rerata	1.443	−0.190	0.177	−3.432	4.85	1.657	1.033
SD	0.735	0.561	0.138	1.071	1.148	0.341	0.489

Note: The level of Hg contamination was not further analyzed, because the Hg concentration is extremely low (undetected at <0.00011 mg/kg) at all observation sites and considered not to contaminate the sediment.

Table 5: Personnel correlation matrix parameter of sediment quality in Belawan Harbor waters

Parameter		Pb	Cu	Cd	Sand	Sludge	Clay	TOC
Pb	Pearson Correlation	1						
	Sig. (2-tailed)							
Cu	Pearson Correlation	0.779**	1					
	Sig. (2-tailed)	0.008						
Cd	Pearson Correlation	0.783**	0.594	1				
	Sig. (2-tailed)	0.007	0.070					
Sand	Pearson Correlation	−0.519	−0.261	−0.259	1			
	Sig. (2-tailed)	0.125	0.466	0.475				
Sludge	Pearson Correlation	0.522	0.275	0.405	−0.928**	1		
	Sig. (2-tailed)	0.122	0.441	0.246	0.000			
Clay	Pearson Correlation	3.67	0.166	−0.036	−0.826**	0.556	1	
	Sig. (2-tailed)	2.97	0.647	0.922	0.003	0.095		
TOC	Pearson Correlation	0.564	0.451	0.390	−0.815**	0.751	0.679*	1
	Sig. (2-tailed)	0.089	0.191	0.265	0.004	0.012	0.031	

been implemented, especially in controlling household waste pollution. This situation has resulted in the community to consider disposing household waste (*e.g.,* plastic, food waste, cable scraps, and corrosion of water pipes) directly into river water bodies the most effective technique to handle their waste problem. Industrial waste also contributes to contamination in Belawan Harbor waters. According to BPS data in 2018, Deli Serdang Regency had 444 industries, Karo Regency had 7, and Medan City had 270. The Deli and Belawan Rivers flow in these regencies. The data from the Belawan Ular Padang resource management pattern by the Minister of Public Works in 2012 indicate that many industries have a WWTP capacity smaller than the waste they produced, such that their waste did not meet the established quality book. Anthropogenic impacts accelerate the rate of toxicity in the food web in water sediments and reduce the quality of the water environment through the transportation of pollutants through land by humans (Leung *et al.*, 2021; Azizi *et al.,* 2016).

Pearson correlation matrix analysis

The correlations between heavy metals are depicted by the Pearson matrix (Table 5). Heavy metals, sediment fraction, and TOC, as well as the significance level of the correlation matrix, are described. Having a value of 1 for the degree of significance indicates an extremely high level of relevance.

The Pearson correlation value for several metals is presented in Table 5. Pb and Cd have the strongest heavy metal correlation value (R^2 = 0.783, p = 0.007), as well as Pb and Cu (R^2 = 0.779, p = 0.008). A significant degree of association is observed. The lowest value of correlation is found between Cu and Cd (R^2 = 0.594, p = 0.070). For all metals, a p value of 0.05 is considered significant. Thus, a relationship exists between these factors. No statistically significant difference exists between the sediment and heavy metal fractions and TOC ($p > 0.05$). Sludge has the highest R^2 values for Pb, Cd, and Cu among the sediment fractions studied (0.522, 0.122, 0.405, 0.26; 0.275). After Pb and Cu, clay exhibits a weak negative association with Cd (R^2 = −0.036, p = 0.922) and a moderately positive correlation with Pb (R^2 = 0.367, p = 0.297). Thus, the Cd output decreases with the increase in the fraction of clay sediment. Pb (R^2 = −0.519, p = 0.125), Cu (R^2 = -0261, p = 0.466), and Cd (R^2 = −0.259, p = 0.471) have a negative connection with sand fraction. Given that sand cannot bind with heavy metals, as the sand percentage in the water becomes more concentrated, the heavy metal content will also drop. Heavy metals in TOC have a significance level of $p > 0.05$. The significance threshold for heavy metals in sediment fractions is equivalent to this finding. Considerable differences between the two are not observed. Pb (R^2 = 0.564, p = 0.089), Cu (R^2 = 0.451, p = 0.191), and Cd (R^2 = 0.390, p = 0.265) have the highest Pearson correlation values of TOC with the highest heavy metals. Heavy metal concentrations tend to be greater in fine-grained, high-organic-matter sediments (Dan *et al.*, 2022). There exists a high degree of relationship between TOC and sludge (R^2 = 0.751, p = 0.12), and there exists a similarly high correlation value between TOC and clay (R^2 = 0.679, p=0.31). The degree of connection between TOC and sand fraction is extremely strong (R^2 = −0.815, p=0.004). Thus, the TOC concentration in sediment

Table 6: Principal component análisis

Parameters	F1	F2
Pb	0.672	0.227
Cu	0.385	0.365
Cd	0.359	0.462
Sand	0.786	0.188
Sludge	0.726	0.068
Clay	0.473	0.326
TOC	0.779	0.038
Eigenvalue	4.180	1.674
Variance (%)	59.708	23.915
Cumulative (%)	59.708	83.623

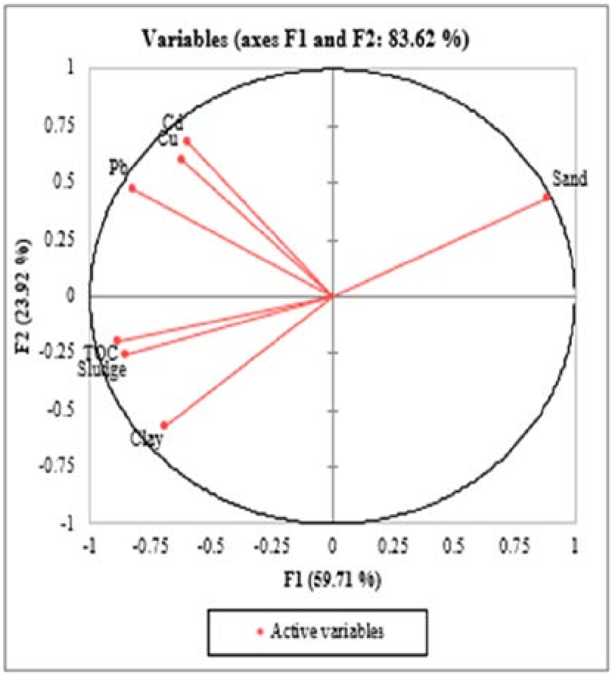

Fig. 2: Analysis of the main components of sediment quality parameters in the waters

decreases with the increase of sand content. The sludge fraction often contains a high concentration of organic and inorganic materials (Quaty *et al.*, 2022).

Principal component analysis

PCA should be performed on the normalized data to compare the compositional patterns between sediment samples and identify the factors that influence each sample. Results of PCA analysis of this study can be seen in Table 6 and Fig. 2.The PCA has revealed two principal components with eigenvalues greater than 1, with a contribution of 83.63%. The first component (F1) contains 59.71% of Pb, Cu, sand, mud, clay, and TOC. The second component (F2) contains 23.92% of Cd. F1, which accounts for 59.708% of the total dispersion, has strong positive charges (>0.70) on Pb, Cu, sand, sludge, clay, and TOC. F2, which accounts for 23.915% of the total dispersion, has a strong positive charge on Cd. Cd is the most polluting heavy metal in Belawan Harbor water based on CF, Igeo, and PLI calculations. The correlation analysis has revealed that Cd has the lowest Pearson correlation with heavy metals and sediment fractions, TOC, and other heavy metals.

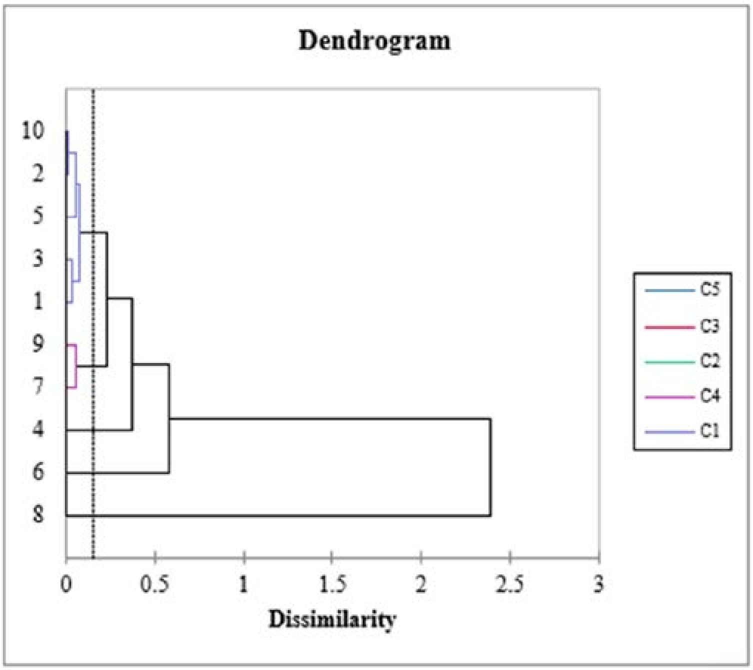

Fig. 3: Cluster analysis of observation sites

Thus, the environmental conditions for Cd and its compounds are found in many layers. Cd is known to be common in landfills, stormwater streams, and sewage (Wardani et al., 2018).

Cluster analysis

Similarity between data points is used as a criterion in the CA. The correlation between the sites is investigated using CA and the centroid linkage technique (Fig. 3). The spatial CA is shown in a dendrogram, where the 10 sediment sample locations have been partitioned into five groups.

Cluster 5 (Site 8) is located in a highly polluted area, which receives the most waste disposal. This finding is in accordance with the PLI results, which indicates that Site 8 is the most polluted area of the other sites. Cluster 4 (Sites 7 and 9) is where the two sites are at the same estuary. Cluster 3 (Site 6) is where the difference in clusters is a Pacific Ocean tourism area, and the tourism management here, in relation to waste management, is also still subpar. Cluster 2 (Site 4) is a harbor reclamation area, so the sand substrate in this area is extremely high compared with other sites. Finally, in Cluster 1 (Sites 1, 2, 3, 5, and 10), Sites 1, 2, 3, and 10 are in the same stream (*i.e.,* Belawan River), whereas Site 5 is in the sea area. Therefore, river flow influences sediment concentration in these waters.

CONCLUSIONS

The content of heavy metals in the sediments of Belawan Harbor varies; that is, essential heavy metal Cu does not contaminate the sediments. Conversely, non-essential heavy metals Cd and Pb contaminate them, but different from the concentration of Hg, which has a value greater than 0 but is undetected in the sediment. The content of heavy metals and TOC will be absorbed in the muddy sediment substrate rather than the sand substrate. Given that the smaller pores in the mud substrate are smaller, they can better trap heavy metals and TOC into the sediment. As a result, heavy metal pollution is often detected in estuary and coastal areas rather than the open sea, which has sand substrate. Harbor pollution levels are in the following order: Site 8 > Site 7 > Site 9 > Site 6 > Site 2 > Site 10 > Site 4 > Site 1 > Site 3 > Site 5. The highest pollution is at the mouth of the Deli River; this area is on the DLKp not the DLKr, which shows that harbor activities do not contribute significantly to heavy metal pollution in the waters. The high TOC in the DLKr area is due to changes in the river trajectory area. On the basis of the results of a study on heavy metal pollution in sediments, water pollution at Belawan Harbor is included in the category of lightly polluted levels. This study can be used as a reference for the government to make regulations or even wiser actions, especially related to the management of the aquatic environment in estuary areas. If not taken seriously, the pollution level will increase with the anthropogenic activities along the Belawan River and coastal watersheds every year. The long-term impact that can occur is that many endemic biota will disappear due to not being able to survive in an environment exposed to heavy metals. Thus, people who depend on their lives as fishermen will find it increasingly difficult to find fish, and social conflicts between the community and local companies will occur. Spatial planning in watersheds, especially in residential areas, might raise concerns by preventing the construction of settlements that face away from the river. Indonesia’s Ministry of Environment Regulation No. 03 of 2010 concerns wastewater quality standards for industrial areas. If these regulations are obeyed by every company, then heavy metal pollution in waters can be reduced. The reason is several research results claim that the largest heavy metal contaminants come from industrial waste and residential areas. The need for periodic monitoring of all companies and the provision of communal WWTP in each settlement area is a considerably important concern for creating sustainable management of the aquatic environment. This research suggests Indonesia to have its own sediment quality standard guidelines to have a more accurate results of the analysis of pollution levels. Quality standards are made by the state with various aspects related to the country. The lower the value of the set quality standard, the more serious the pollution that can occur in the country. For future research, this study can be continued to measure the concentration of other heavy metals. In addition to Cd and Pb, the concentration of other heavy metals that mostly pollute waters can be investigated.

AUTHOR CONTRIBUTIONS

The corresponding author, L. Sulistyowati, helped with data analysis and writing. Y. Yolanda reviewed the relevant literature, gathered relevant data, and examined the extent to which water was polluted. N. Andareswari analyzed statistically the principal component analysis and cluster analysis.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

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PUBLISHER’S NOTE

ABBREVIATIONS

%	Percent
^{0}C	Degree celsius
‘E	East
‘N	North
AAS	Atomic absorption spectrometry
ANZECC	Australia New Zealand Environment and Conservation Council
ARMCANZ	Agriculture and Reource Management Council of Australian and New Zealand

BPS	Badan Pusat Statistik: Central statistic agency in Indonesia
CCME	Canadian for Toxic Substance and Disease Registry
CA	Cluster analysis
Cd	Cadmium
CF	Contamination factor
cm	Centimeter
Cu	Copper
DLKr	Work Authority area
DLKp	Interest Authority area
EL	East longitude
Eq.	Equality
e.g.	Exempli gratia (for example)
F	Component of principal component analysis
GPS	Global positioning system
ha	Hectare
$HClO_4$	Perchloric acid
Hg	Mercury
HNO_3	Nitric acid
i.e.	Id est (that is)
Igeo	Geoaccumulation index analysis
ISQG	Interim Sediment Quality Guidelines
Max.	Maximum
mg/kg	Milligram per kilogram dry wight
Min.	Minimum
mL	Mililiter
NL	North latitude
p	Significance value
PAH	Polycystic aromatic hydrocarbons
ppb	part per billion
Pb	Lead
PCA	Principal component analysis
PEL	Probable effect levels
PLI	Pollution load index
r	Pearson's correlation coefficient
Site	Station sampling
SD	Standard deviation
TOC	Total organic carbon
TN	Total nitrogen
USEPA	United States Environmental Protection Agency
WWTP	Wastewater treatment plants

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