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Distribution of Heavy Metals in Surface Seawater and Sediment of Cirebon Coastal Area, West Java

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ABSTRACT

Heavy metals in water and sediment are one of the most dangerous pollutants that can have toxic effects on the environment. This research was conducted to evaluate the concentrations of heavy metals (Cd, Cr, Cu, Hg, Pb, and Zn) from PPN Kejawanan and PPI Gebang Mekar, Cirebon, West Java. For heavy metals in PPN Kejawanan waters, Pb has exceeded the threshold (Pb = 0.137), while the concentration of Zn is still within safe limits (Zn = 0.014). Based on the STORET method, water quality status at all stations was recorded with varying results from Class A to Class C. In sediments, cadmium (Cd) was found at high concentrations at all stations (Cd = 2.273–2.867mg/ kg), exceeding the threshold. With respect to the potential ecological risk index (RI) analysis, Cd at all stations was recorded with an extremely high ecological risk, thereby threatening the life of aquatic ecosystems (Cd= 341.10-428.7). Heavy metals under evaluation have a low to moderate ecological risk (RI = 0-64). The Pearson correlation analysis used in this research indicated a potential source of the heavy metal groups Cu-Hg-Zn, Cr-Pb, and Cr-Cu originating from several sources of domestic waste, industry, agriculture, and port activities.

INTRODUCTION

Cirebon is a regency on the northern coast of West Java, with an area spanning from 108°40' to 108°48' E and between 6°30' to 7°00' S. Cirebon Waters is a place of active industrial and port activities due to their strategic location along shipping routes **(Dwianti *et al.*, 2017)**. In 2013, there were 864 types of industries in Cirebon, including large, small, and medium-sized industries **(Zahroh *et al.*, 2019)**. However, according to the 2014 Environmental Status Report of Cirebon Regency, only 10% of registered companies had adopted wastewater treatment facilities, resulting in the discharge of liquid industrial waste directly into the rivers and sea.

According to the Minister of Maritime Affairs and Fisheries Decree No. 109 of 2021 on the National Fishery Port Master Plan, there are nine registered ports in the Cirebon area. These include the Nusantara Kejawanan Fishery Port located in the western part of Cirebon Regency and the Gebang Mekar Fish Landing Base on the eastern side. In support of the industrialization process in West Java Province, the Directorate General of Fisheries Capture at the Indonesian Ministry of Maritime Affairs and Fisheries is working to enhance the success of the fisheries industry program in the Cirebon region through these ports **(Nurhayati & Putri, 2019)**. Industrial activities, while beneficial for the economy and the well-being of the population, can also be a source of hazardous waste pollution in the surrounding waters, particularly with industries such as fisheries, oil and gas in addition to chemicals.

One of the hazardous pollutants for marine ecosystems is heavy metals. Heavy metals are metallic elements with a density greater than 5g/ cm^3. Naturally, heavy metals are found in the environment at very low concentrations (<10ppm) **(Tchounwou *et al.*, 2014)**. Generally, heavy metals are divided into essential and non-essential categories. Essential heavy metals like Cu and Zn play a role in the growth, development, and metabolism of living organisms. On the other hand, non-essential heavy metals, in concentrations exceeding certain thresholds, can be toxic to living organisms. Non-essential heavy metals like Cd, Cr, Pb, and Hg are highly toxic to the environment, and their roles and benefits for living organisms are not well understood **(Manullang *et al.*, 2017)**. Heavy metals can enter the environment through both internal and external sources. Internally, heavy metals are naturally present in the Earth's crust, rocks, soil, air, and water **(Ross *et al.*, 2017)**. External sources of heavy metals come from anthropogenic activities, both intentional and unintentional, that can lead to changes in the condition of water bodies **(Zhou *et al.*, 2020)**. Heavy metals like Cu, Pb, Cd, Cr, Hg, and Zn can be found in water bodies as a result of various industrial activities, fossil fuel combustion, agriculture, maritime transportation, and household waste **(Herawati *et al.*, 2000)**. Heavy metals are persistent materials, resistant to environmental conditions, and have high bioaccumulation and toxicity levels, making them one of the most dangerous pollutants in the environment **(Ariman & Bakan, 2008)**.

Hazardous substances from heavy metals entering the water can be deposited in sediments, accumulating in the bodies of organisms over time **(Permanawati *et al.*, 2016)**. In addition to being toxic to marine organisms, the presence of heavy metals in the environment can also pose risks to human health through various chronic health issues if consumed **(Rehman *et al.*, 2018)**. Studies suggest that the level of activity around a water body or coastal area correlates with the concentration of heavy metals in the water **(Nurhamiddin & Ibrahim, 2018)**. Pollution status in water and sediments is regulated by national and international regulations, including the Ministry of Environment and Forestry Decree No. 51 of 2004 on Sea Water Quality Standards and international standards for sediment quality released by the United States Environmental Protection Agency (USEPA) and ANZECC/ARMCANZ (2000).

Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) is one of the methods used to test heavy metal concentrations and has developed over the past few years. ICP-OES specializes in determining multi-element heavy metal concentrations. The key differences between ICP-OES and other heavy metal detection devices like ICP-MS, AAS, and UV-VIS are its better detection limits and high atomization temperature. Additionally, ICP-OES offers a more inert environment for detecting more than 30 types of metals. ICP-OES can determine concentrations as low as 10ppb (parts per billion) using emission techniques **(Raju *et al.*, 2012)**. Generally, ICP-OES is a more sensitive and specific method for determining heavy metal levels, but it can be more expensive than other methods.

Research on six different heavy metals, namely Cd, Cr, Cu, Pb, Hg, and Zn in water and sediments in the Cirebon Waters with ICP-OES has not been previously conducted. The use of ICP-OES in Indonesia has only seen recent developments over the past few years. Therefore, the purpose of this research was to evaluate the heavy metal concentrations and pollution status in water and sediments in the Cirebon Waters due to the lack of data sources. Additionally, this study analyzed the relationship between water quality and heavy metal concentrations.

MATERIALS AND METHODS

This research was conducted in February 2023 at the Nusantara Kejawanan Fishery Port and Gebang Mekar Fish Landing Base in Cirebon Regency, West Java. Sample collection was carried out using purposive sampling, which involved selecting locations based on the characteristics of the target population **(Campbell *et al.*, 2020)**. Heavy metal analysis (Cu, Pb, Cd, Cr, Hg, and Zn) in water and sediment from the research sites is performed using an ICP-OES instrument in the Central Laboratory of Universitas Padjadjaran.

Fig. 1. Research sites map

This research was conducted at three stations, each at different distances, where samples were collected and *in-situ* observations were carried out. Sample collection at these stations was performed to understand the characteristics of heavy metals (Cu, Pb, Zn, Hg, Cd, and Cr) in the surface water and sediments at Nusantara Kejawanan Fishery Port (PPN Kejawanan) and Gebang Mekar Fish Landing Base (PPI Gebang Mekar). The selection of these stations was based on the results of an initial location survey, as shown in Table (1).

Table 1. Sampling stations

Station	Location	Latitude	Longitude
1	Ciberes River estuary	-6.806017	-108.729954
2	PPN Kejawanan Harbor Pool	-6.732831	108.584905
3	PPN Kejawanan Mangrove Ecosystem	-6.733983	108.585205

During the sampling process, marking plots were established using GPS (Global Positioning System) at each station. Water samples, 500mL at each station, were collected with three repetitions from the water's surface using sample bottles. Sediment samples, 500g at each station, with three repetitions were collected using a shovel. The collected water and sediment samples were placed in a cool box. Water quality parameters measured in this research included pH, DO (Dissolved oxygen), temperature, and salinity. The heavy metal content in the water was evaluated according to the Sea Water Quality Standards established by the Indonesian Ministry of Environment and Forestry Decree No. 51 of 2004. The sediment was analyzed for grain size using Gradistat software.

Water sample quality was determined using the storage and retrieval method (STORET) based on a scoring system referring to Ministerial Decree No. 115 of 2003 from the Ministry of Environment and Forestry. The STORET method, based on Ministerial Decree No. 115 of 2003, was adopted from the United States Environmental Protection Agency's (USEPA) method for determining pollution status in 2002. This method was used to assess water quality status by comparing the data obtained with the water quality standards according to the classes defined in Government Regulation No. 82 of 2001 on water quality management and pollution control. The scoring criteria are presented in Table (4).

The STORET method involves statistical analysis to classify the minimum, maximum, and average values of the tested heavy metals. Table (4) provides a description of the minimum, maximum, and average value system of the STORET method. To determine the pollution status of heavy metals, water quality status was divided into four classes: Class A: Very good, score = 0 → meets standards; Class B: Good, score = -1 to -10 → slight pollution; Class C: Moderate, score = -11 to -30 → moderate pollution; Class D: Poor, score ≥ 31 → heavy pollution.

The results of heavy metal content in sediments was evaluated according to the standards set by the Australian and New Zealand Environment and Conservation Council (ANZECC) and Agriculture and Resource Management Council of Australia and New Zealand (ARMCANZ) water quality guidelines (2000). Factors such as contamination factor (CF), geoaccumulation index (Igeo), and potential ecological risk index (PERI) were also used. Pearson correlation analysis was employed to identify potential sources of heavy metal contamination in sediments **(Jiang *et al.*, 2023)**.

The contamination factor (CF) in this research was used to describe the contamination status of a heavy metal in sediments **(Taylor, 1964)**. The geoaccumulation index (Igeo) was employed to assess the level of accumulation and pollution of heavy metals in sediments in an environment while considering the effects of lithospheric variations with a background value by Turekian's method. The ecological risk index (PERI) was used to evaluate the impact of toxic heavy metals in sediments on biota and the surrounding water **(Rahmayanti, 2018)**. The following are the calculations used:

$$CF = \frac{C_{si}}{c_{bi}} \tag{1}$$

CF = *contamination factor*
C_{si} = *concentration of heavy metals in sediment (mg/kg)*
C_{bi} = *reference value (mg/kg)*

The contamination factor can be interpreted into categories as follows: CF < 1 indicates low contamination; 1 ≤ CF < 3 stands for moderate contamination; 3 ≤ CF < 6 indicates high contamination, and CF > 6 represents very high contamination.

$$I_{geo} = Log2\frac{C_n}{1{,}5\ x\ Bn} \tag{2}$$

I_{geo} = *geoaccumulation index*
C_n = *concentration of heavy metals in sediment (mg/kg)*
B_n = *reference value (mg/kg)*

The results in this research have been interpreted into categories by Muller as follows: Igeo < 0 indicates no pollution (class 0); 0 < Igeo < 1; indicates unpolluted to moderately polluted (class 1); 1< Igeo < 2 indicates moderately polluted (class 2); 2 < I geo < 3 indicates moderately to strongly polluted (class 3); 3 < Igeo < 4 strongly polluted (class 4); 4 < Igeo < 5 strongly to extremely polluted (class 5); Igeo > 5 extremely polluted (class 6).

$$E_f^i = C_f^i x\, T_f^i \tag{3}$$

E_f^i = *ecological risk index of heavy metal*
C_f^i = *contamination factor*
T_f^i = *response coefficient of heavy metals toxicity*
(Pb= 5; Hg= 40; Cd= 30; Cu= 5; Cr= 2; Zn= 1)

The ecological risk index (RI) for each type of heavy metal can then be interpreted into several categories as follows: $E_f^i < 40$ indicates low risk index; $40 \leq E_f^i < 80$ indicates moderate risk index; $80 \leq E_f^i < 160$ indicates fairly high risk index; $160 \leq E_f^i < 320$ indicates high risk index; $E_f^i \geq 320$ indicates very high risk index. The ecological risk index (RI) for a combination of several types of heavy metals can be calculated using the following formula:

$$RI = \sum E_f^i \tag{4}$$

The ecological risk index (RI) for a combination of several types of heavy metals can then be interpreted into several categories as follows: RI < 150 indicates a low risk index; 150 ≤ RI < 300 indicates a moderate risk index; 300 ≤ RI < 600 indicates a fairly high risk index; RI ≥ 600 indicates a serious or very high risk index.

Table 2. Seawater quality standards

	Ministry of Environment and Forestry Decree No.51/2004 (mg/l)		
Metals	Port	Tourism	Biota
Cu	0.05	0.05	0.008
Pb	0.05	0.005	0.008
Cd	0.01	0.002	0.001
Cr	-	0.002	0.005
Hg	0.003	0.002	0.001
Zn	0.1	0.095	0.05

Table 3. Sediment quality standards

	Sediment quality standards (mg/kg atau parts per million/ppm)	
Metals	US EPA	ANZECC/ARMCANZ
Cu	0.05	0.05
Pb	0.05	0.005
Cd	0.01	0.002
Cr	-	0.002
Hg	0.003	0.002
Zn	0.1	0.095

Table 4. STORET method system **(Decree No.115/2003)**

Number of samples	Value	Parameter		
		Physical	Chemical	Biological
< 10	Maximum	-1	-2	-3
	Minimum	-1	-2	-3
	Average	-3	-6	-9
≥ 10	Maximum	-2	-4	-6
	Minimum	-2	-4	-6
	Average	-6	-12	-18

1. Water sample preparation

A volume of 10ml water sample was collected and placed in an Erlenmeyer flask. Then, 10ml of HNO_3 65% was added to the water sample, and heated on a hot plate until boiling. The sample was then cooled and diluted with distilled water. The sample was filtered using filter paper, and 25ml of the filtered solution was placed in a vial for testing using ICP-OES.

2. Sediment sample preparation

Sediment samples were first dried and sieved using a sieve shaker. 1g of fine sieved sediment was placed in a glass beaker. The sample was then transferred to an Erlenmeyer flask to which reagents were added. The sediment sample was treated with a 10ml solution of HNO3 65% and HClO4 60% in a 5:1 ratio, respectively. The sample was then heated using a hot plate in the acid room. After boiling, the sample was allowed to cool to room temperature and was then diluted with distilled water and filtered. A 25ml of the filtered solution was placed in a vial and was ready for testing.

a. Determination of heavy metal levels in water and sediment using ICP-OES

The determination of heavy metal levels (Cu, Pb, Cd, Cr, Hg, and Zn) using an ICP-OES instrument was performed according to the SNI 6989-82-2018 method for water samples and the SNI 8910-2021 method for sediment samples. The specific wavelengths for each metal are as follows: Cu at 283.563nm, Pb at 220.353nm, Cd at 226.502nm, Cr at 283.563nm, Hg at 253.652nm, and Zn at 213.857nm.

The ICP-OES testing procedure begins by turning on the instrument and launching the ICP Expert II Agilent 725-ES Instrument Software (Version 2.0). Next, the argon gas cylinder is opened, and a water cooler is used to chill the gas to approximately -24°C. Once the plasma is ignited, the tube is inserted into testing vials containing the samples to be analyzed.

RESULTS AND DISCUSSION

1. Water quality

Based on the Indonesian Ministry of Environment Decree No. 51 of 2004 regarding sea water quality standards, the optimal temperature for marine biota ranges from 28 to 30°C for coral reef and seagrass ecosystems and from 28 to 32°C for mangrove ecosystems. As seen in Table (5), the *in-situ* results indicate that the average water temperature in the Cirebon area at station 1 (Gebang Mekar) is 30.67°C, at station 2 (Kejawanan Port) is 28°C, and at station 3 (Kejawanan mangrove area) is 30°C. These findings reflect normal temperatures that comply with the established quality standards. This temperature range is also sufficient to support coastal ecosystem life.

The salinity measurements reveal average salinity at station 1 (Gebang Mekar) to be 1.167 PSU. Station 2 (Kejawanan Port) shows a result of 24 PSU, and station 3 (Kejawanan mangrove area) averages 7 PSU. Station 1 is directly connected to a brackish water source, namely the Ciberes River estuary. This results in very low salinity measurements in that area. Station 2, located in the harbor area near the coast, has higher salinity levels due to its proximity to the sea. Station 3, despite being near the coast, has low salinity levels because it is located in a mangrove ecosystem area closer to the land.

Dissolved oxygen (DO) measurements at the three research stations show varying results, closely related to different water characteristics. Station 1 has an average DO level of 4.76mg/ L, which is the lowest among the three stations. DO at station 1 falls below water quality standards, indicating poor water quality. Station 2 at Kejawanan Port, recorded an average DO level of 6.3mg/ L. Station 3 demonstrated the best DO measurements among the three stations, with an average DO level of 8.96 mg/L. The high average DO at station 3 is influenced by the sampling location within the Kejawanan mangrove ecosystem.

pH measurements at station 1 showed an average of 6.31, which is below water quality standards. The results at this station represent the lowest pH observed among the three research stations. Acidic pH values in a water body indicate water disturbance. The acidity of this water is closely related to high concentrations of waste and pollutants in the sampling area. Station 2 and station 3 each have pH values of 6.87 and 6.85, respectively. These pH values indicate relatively good water pH conditions that meet the established quality standards **(Simanjuntak, 2012)**.

Table 5. Water quality parameter and threshold values

Station	Temperature (°C)	Salinity (ppt)	DO (mg/L)	pH
1	30.67 ± 0.4714	1.17 ± 0	4.76 ± 0.0471	6.31 ± 0.0535
2	28 ± 0	24 ± 2.8284	7.13 ± 0.1886	6.87 ± 0.0499
3	30 ± 0	7 ± 0.8165	8.96 ± 0.2867	6.85 ± 0.0411
Treshold	28 - 31	28 – 33	>5	6.5 – 8.5

2. Heavy metals content in water

The levels of Cd, Cr, Cu, and Hg in the water at all three stations are reported as <0.0001mg/ L, indicating that these metal groups were not detected in the water and are below the maximum allowable concentration (NAB) according to the Ministry of Environment Decree No. 51 of 2004. The NAB values for Cd, Cr, Cu, and Hg are 0.01, 0.005, 0.05, and 0.003mg/ L, respectively. The low detection of these four metals could be attributed to various factors, one of which is the timing of the sample collection. Different sea conditions and high current velocities in the sampling locations during different seasons can affect water quality measurements **(Nurhayati & Putri, 2019)**.

In this study, the presence of the heavy metal Zn was detected at stations 2 and 3 with levels of 0.0199 and 0.0419mg/ L, respectively. The Zn content falls within safe limits as it remains below the NAB of 0.1mg/ L. The Zn levels measured at the three stations range from <0.0001 to 0.0419mg/ L, with the lowest levels found at station 1. Since Zn serves as an essential metal, it is required in specific quantities as a cofactor in enzymatic processes of organisms. However, external sources of Zn can come from motor vehicles and inputs of liquid waste, such as paint, metal, and oil **(Rahmayanti, 2018)**.

The levels of Pb show more variation compared to other heavy metals, which mostly have homogeneous results. The Pb levels found at stations 1 and 3 fall within safe limits, while at station 2, they exceed the NAB. Pb pollutants can enter the environment through gasoline emissions, corrosive water pipes, industrial activities, and lead-containing paint **(Gusnita, 2012)**. Station 3, located in the mangrove ecosystem area, allows mangroves to capture and store metals and nutrients in the sediment **(Alongi, 2020)**.

Research on the levels of heavy metals in Cirebon Waters in the past 10 years has included measurements of Cu, Zn, Cd, Pb, and Hg conducted in the Kejawanan area **(Sudirman & Husrin, 2014)**, Bondet **(Nurhayati & Putri, 2019; Wahyuningsih, 2021)**, and Cirebon **(Nurhayati & Putri, 2019)**. Research on Pb in the Bondet area showed already contaminated results, with the maximum range of those results still lower than those measured in this study. Research in other areas, such as in the coastal area of Tugu Semarang **(Suryono *et al.*, 2016)**, showed higher Pb levels in water, ranging from 0.74 to 0.82mg/ L. In Jakarta Bay, the results showed lower levels, ranging from 0.005 to 0.011mg/ L **(Permanawati *et al.*, 2016)**.

Table 6. Heavy metals concentration in water and the threshold values

		Results (mg/L)					
No	Code	Cd	Cr	Cu	Hg	Pb	Zn
1	1A	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
2	1B	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
3	1C	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
	average	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**
4	2A	<0.0001	<0.0001	<0.0001	<0.0001	0.4108	0.0315
5	2B	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	0.0083
6	2C	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
	average	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**	**0.1369**	**0.0199**
7	3A	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
8	3B	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	0.0419
9	3C	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
	average	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**	**0.0140**
Indonesian Ministry of Environment Decree No.51/2004		**0.01**	**0.005**	**0.05**	**0.003**	**0.05**	**0.1**

3. Heavy metals content in sediment

In the sediment analysis as seen in Table (7), the Gradistat software was used to determine the sediment fractions at each research station. Station 1, which is near the mouth of the Ciberes River, has sandy gravel sediment, which is the coarsest sediment type among the three research stations. Stations 2 and 3 have gravelly sand sediment, with fine sandy characteristics dominant due to their closer proximity to the coast. Station 3 has sediment that is somewhat coarser than that of station 2, attributed to its mangrove substrate. The sediment tendency shows finer textures at station 2, fine textures at station 3, and coarser textures at station 1.

Table 7. Sediment type

		Composition (%)			
No	Station	Gravel	Sand	Mud	Sediment type
1	Station 1	48.2 – 59.2	40.8 – 51.8	0	Sandy gravel
2	Station 2	24.6 – 30.4	69.6 – 75.4	0	Gravelly sand
3	Station 3	27.8 – 46.4	53.6 – 72.2	0	Gravelly sand

Based on the analysis of Cd concentrations in the sediment at the three research stations, it was found that the Cd content in all samples exceeded the ANZECC/ARMCANZ sediment quality guidelines. The safe Cd metal content is expected at a concentration of 1.5mg/ kg. The high Cd concentrations exceeding the sediment quality threshold at all three research stations indicate significant contamination of heavy metal Cd in the Cirebon Waters, which poses a potential risk. Inputs of heavy metal Cd into the water can come from various industrial activities, chemical manufacturing, and household waste.

The concentration of heavy metal Cr in the sediment of the three stations is still within the safe range and is below the threshold established by ANZECC/ARMCANZ, which is 80mg/ kg. The highest Cr metal levels were found in the sediment of station 1, which is near the mouth of the Ciberes River (Station 1 > Station 3 > Station 2). Based on these results, the higher Cr input at station 1 may be attributed to ship paint coatings, wood coatings, and industrial waste in the station's vicinity. The discharge of untreated industrial waste is likely the primary reason for the high input of heavy metal Cr in the sediment at station 1, as this area lacks a wastewater treatment plant (WWTP) like station 2.

The analysis of Cu heavy metal levels in the sediment shows that the sediments at the three stations are still below the threshold set by ANZECC/ARMCANZ, which is 65mg/ kg. The heavy metal Cu at stations 1, 2, and 3 can originate from port activities, household waste, metal industry waste, and electroplating in the station's vicinity. However, a significant difference can be observed in the Cu levels at station 2 compared to the other stations. Generally, fine muddy sediments in the harbor basin can cause metals to readily bond with finer sediment particle sizes **(Leksono *et al.*, 2013)**, as observed at station 2.

The analysis of heavy metal Hg concentrations in the sediment of the three stations shows results still below the ANZECC/ARMCANZ threshold of 1.5mg/ kg. On the other hand, mercury is considered a highly toxic metal and can originate from various anthropogenic sources such as pharmaceutical industry, electrical equipment, agriculture, and wastewater irrigation **(Haryati *et al.*, 2023)**. Activities with the potential to produce higher levels of Hg input at station 2 than at station 1 are observed due to the high activity of the port, shipyard, and domestic waste in the vicinity.

Pb levels in the sediment of the three stations fall within the range of 28.4-45.7mg/ kg. These levels are still categorized as safe, being below the maximum allowable concentration set by ANZECC/ARMCANZ, which is 50mg/ kg. Lead is known to readily bond with organic material and sulfide, making it bond easily with sediment **(Yuan *et al.*, 2004)**. Some activities that could potentially increase the levels of Pb heavy metal in water and sediment include metal industry, paint, ceramics, glass, and atmospheric deposition from leaded fuel vehicles in the research area **(Patty *et al.*, 2018)**.

According to the ANZECC/ARMCANZ threshold, the maximum Zn metal levels in sediment are 200mg/ kg. Therefore, the Zn metal levels at the three research stations remain below the threshold and are considered safe. Although this metal is naturally abundant in the environment due to its role in the metabolism of living organisms, Zn can also originate from anthropogenic activities, such as vehicle fuel emissions, detergent waste, cosmetics, and metal smelting activities **(Wang *et al.*, 2010)**.

Research on heavy metals in the sediment of the Cirebon Waters in the last 10 years has been conducted for Hg **(Patty *et al.*, 2018; Nurhayati & Putri, 2019)**, Pb **(Nurhayati & Putri, 2019; Wahyuningsih, 2021)**, and Cd **(Nurhayati & Putri, 2019)**. However, the concentrations of Cu, Zn, and Cr heavy metals in the Cirebon Waters sediment have not yet been previously studied. According to previous research, the sediment in the Cirebon Waters area shows Pb and Hg levels that are considered safe and still below the ANZECC/ARMCANZ threshold, consistent with the findings of this study.

The measurement of Pb heavy metal in this research is still lower compared to the results found in the estuary of Way Kuala River, Bandar Lampung **(Hidayat, 2011)** and Jakarta Bay **(Permanawati *et al.*, 2016)**. On the other hand, the research on the Cd content in sediment in the Cirebon Waters has exceeded the threshold set by ANZECC/ARMCANZ, as found in this study. Research in other areas has shown Cd levels below the threshold, ranging from 0.008-0.062mg/ kg in Banyuasin estuary, Semarang, and 0.012 – 0.750mg/ kg in Jakarta Bay **(Permanawati *et al.*, 2016)**.

Table 8. Heavy metal concentration in sediment and threshold values

		Results (mg/kg)					
No	Code	Cd	Cr	Cu	Hg	Pb	Zn
1	1A	2.1095	52.461	16.5222	0.0374	32.5961	102.4308
2	1B	2.5215	53.0557	18.7862	0.0849	78.4951	136.1169
3	1C	2.1887	46.2809	17.9767	0	26.2298	99.8907
	average	**2.2732**	**50.5659**	**17.7617**	**0.0408**	**45.7737**	**112.8128**
4	2A	2.5461	43.0518	49.3523	0.2317	33.9811	199.6936
5	2B	2.5312	36.2831	35.9411	0	34.5382	182.5137
6	2C	2.7036	40.5985	29.1015	0.152	34.4962	4.8439
	average	**2.5936**	**39.9778**	**38.1316**	**0.1279**	**34.3385**	**129.0171**
7	3A	3.1399	45.6086	16.6774	0	29.5531	116.5157
8	3B	2.9061	47.7248	20.7774	0	27.9508	129.3716
9	3C	2.5269	41.8804	22.3458	0	27.7914	95.8865
	average	**2.8576**	**45.0713**	**19.9335**	**0**	**28.4318**	**113.9246**
ANZECC / ARMCANZ (2000)		**1.5**	**80**	**65**	**0.15**	**50**	**200**

4. Comparison of heavy metals content in water and sediment

In general, sediment is a good indicator for detecting metal levels in water **(Wahyuningsih, 2021)**. This is observed in Fig. (2), where heavy metals tend to be found at higher levels in sediment than as a dissolved form in the water. The vast and dynamic conditions of water bodies can cause metal levels in the water to fluctuate **(Arifin *et al.*, 2012)**. Metal levels in the water can fluctuate due to the solubility of metals in seawater and ocean currents. In contrast, heavy metal levels in sediment are relatively more stable and tend to increase over time.

As seen in Fig. (2), the Pb levels in the water at station 2 are higher than at station 1, even though the Pb levels in the sediment at station 2 are lower than at station 1. This suggests that heavy metal Pb at station 2, located in the Kejawanan Harbor, experiences resuspension from sediment back into the water column. Heavy metals bound to sediment may potentially be released back into the water column due to specific physicochemical conditions of the water.

Station 1, located near the mouth of the Ciberes River, shows a consistent pattern of not detecting heavy metals (Cu, Cd, Cr, Hg, Pb, or Zn) in its water. However, this station shares a similar pattern with other stations in terms of heavy metal contamination in its sediment. This indicates that the input of heavy metals at station 1 is primarily deposited in the estuarine sediment, and some of it is carried by currents toward the Gebang Sea.

In contrast to the other two stations, station 3 presents an interesting profile as it is the only station where no detectable levels of heavy metal Hg are found in both its water and sediment. This may be because station 3 is an area of a mangrove ecosystem. Heavy metals can be accumulated in the structure of mangrove plants **(Parvaresh *et al.*, 2010)**. This is further supported by the role of mangroves as traps for various metal elements and nutrients originating from land and sea **(Silva *et al.*, 2006)**. Hg, which naturally occurs in small concentrations in the environment, becomes even less detectable within the mangrove ecosystem. This may also be attributed to the absence of anthropogenic input containing Hg in the station 3 area.

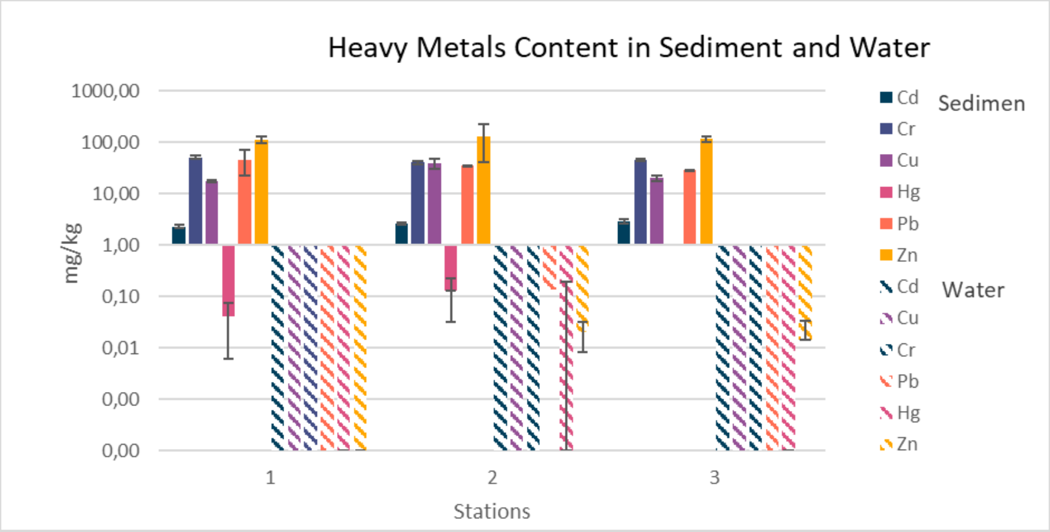

Fig. 2. Comparison of heavy metals content in water and sediment

5. Contamination levels in water and sediment

The analysis using the STORET method for the water at three stations includes eight parameters: pH, DO, and the metals Cd, Cr, Cu, Hg, Pb, and Zn. The analysis results indicate the water quality status at station 1, the Ciberes River estuary, Gebang Mekar, with a total score of -20, consisting of -10 for the quality of each parameter pH and DO. From the total score, it can be concluded that the water quality status at station 1 falls into Class C or moderately polluted. In station 1, the parameters not meeting the water quality standards are pH and DO, resulting in an overall poor water quality status.

The analysis at station 2 shows the water quality status for the Kejawanan National Fishery Port with a score of -8. The total score falls into Class B, which is slightly polluted. The score of -8 is obtained from the measurement of Pb levels in the harbor waters exceeding the specified threshold. Other parameters such as pH, DO, and the metals Cd, Cr, Cu, Hg, and Zn still meet the water quality standards and are given a score of 0.

In the waters of station 3, located in the Kejawanan mangrove ecosystem, the analysis shows a water quality status with a score of 0. A score of 0 indicates that the water at station 3 falls into Class A or meets the water quality standards. The results for all eight parameters meet the water quality standards, and each parameter is given a value of 0, resulting in a total final score of 0. This total score indicates that the waters of the mangrove ecosystem at station 3 have a healthy water quality status and are in the best condition among the three stations. The detailed results of the water quality status for each station are observed in Table (9).

Table 9. Water quality status

No	Nam3	Score	Water quality status
1	Station 1	-20	Class C
2	Station 2	-8	Class B
3	Station 3	0	Class A

Referring to Hakanson's interpretation for heavy metal contamination factors in sediment, the concentration of heavy metal Cd in the sediment at all stations is at a very high contamination level, with all CF values greater than 6. This category represents the highest level of contamination, confirming that the sediment at all three stations is heavily contaminated with Cd (CF = 11.37-14.29). The contamination factor (CF) for all heavy metals in the sediment can be observed in Table (10).

Table 10. Contamination factor of heavy metals in sediment

Heavy metals	CF			Average	Status
	Station 1	Station 2	Station 3		
Cd	11.37	12.97	14.29	12.88	Very high contamination
Cr	0.51	0.40	0.45	0.45	Low contamination
Cu	0.32	0.69	0.36	0.46	Low contamination
Hg	0.51	1.60	0	0.70	Low contamination
Pb	3.66	2.75	2.27	2.89	Moderate contamination
Zn	1.84	1.84	1.63	1.77	Moderate contamination

Heavy metals Cr and Cu in the sediment at all stations are classified as having low contamination levels, with CF values ranging from 0.32 to 0.69. Heavy metal Hg in the sediment at the three stations has different contamination levels, with a moderate contamination level at station 2 (CF = 1.60) and low to no contamination at station 1 (CF = 0.51) and station 3 (CF = 0). The observed contamination levels for heavy metal Pb in the sediment also show variation, with high contamination at station 1 (CF = 3.66) and moderate contamination at station 2 (CF = 2.75) and station 3 (CF = 2.27). The CF values for heavy metal Zn do not differ significantly, falling into the moderate contamination level at all three stations (CF = 1.63-1.84).

Several conclusions can be drawn from the presented data. Heavy metal Cd in the sediment at all stations has Igeo values greater than 2, indicating that the level of heavy metal Cd contamination in the sediment at all stations falls into Class 3, which represents a fairly severe contamination category. On the other hand, heavy metals Cr, Cu, Hg, and Zn have indices ranging from 0 to -3.88. These results suggest that heavy metals Cr, Cu, Hg, and Zn fall into Class 0, indicating no contamination to low-level contamination. The Igeo calculation results for heavy metal Pb differ at each station, with a moderate contamination level or Class 1 at station 1 (Igeo = 0.61) and station 2 (Igeo = 0.19). Station 3 shows the lowest Igeo value for heavy metal Pb (Igeo = -0.08) and falls into Class 0, indicating no contamination to low-level contamination.

Table 11. Geoacumulation index of heavy metals in sediment

	I_{geo}					
Station	Cd	Cr	Cu	Hg	Pb	Zn
1	2.34	-1.42	-1.93	-3.88	0.61	-0.15
2	2.53	-1.76	-0.82	-2.23	0.19	-0.14
3	2.67	-158	-1.76	0	-0.08	-0.32

In the results presented in Table (12), from an individual risk factor perspective, the heavy metal Cd at all stations has an index value falling into the severe or very high-risk category (E_f^i = 341,10-428,7). Hg at station 2 falls into the moderate ecological risk category with an index value of (E_f^i = 64). On the other hand, heavy metals Cr, Cu, Pb, Zn, and Hg (for stations 1 and 3) all have safe ecological risk index values ranging from E_f^i = 0-20,40 (Cd > Hg > Pb > Cu > Zn > Cr). As a result, the combined ecological risk index calculated in this study yields an RI ranging from 384.26 to 444.38, placing all three stations in the high ecological risk index category and potentially posing indirect threats to aquatic biota and humans.

Table 12. Ecological risk index of heavy metals in sediment

			Risk factor	
No	Heavy Metals	Station 1	Station 2	Station 3
1	Cd	341.10	389.1	428.7
2	Cr	1.02	0.8	0.9
3	Cu	1.60	3.45	1.8
4	Hg	20.40	64	0
5	Pb	28.30	13.75	11.35
6	Zn	1.84	1.84	1.63
Ecological Risk Index (RI)		**384.26**	**472.94**	**444.38**

6. Potential contamination sources

In this study, Pearson correlation analysis is used to identify potential sources of heavy metal content, including Cd, Cr, Cu, Hg, Pb, and Zn, in both the water and sediment. Previous research on the relationships between heavy metals has used Pearson correlation analysis to identify similar transfer patterns between the different types of metals found in sediments. Based on the results of the Pearson correlation analysis, several pairs of heavy metals were found to have relationships marked by values of $r > 0.5$. Pairs of heavy metals with positive relationships include Cr - Pb ($r = 0.465$) in the moderate relationship category, Cu - Zn ($r = 0.483$) in the moderate relationship category, and Cu - Hg ($r = 0.706$) in the strong relationship category. Negative relationships, indicating inverse relationships (non-linear), can be observed in the pair of heavy metals Cr - Cu ($r = -0.603$). According to Table (13), it can be concluded that the pair of heavy metals Cr - Cu has a strong relationship.

The significance level ($\alpha < 0.05$) for the relationships Cr - Pb, Cu - Zn, Cu - Hg, and Cr - Cu is $\alpha = 0.208$, $\alpha = 0.188$, $\alpha = 0.034$, and $\alpha = 0.085$, respectively. These values indicate that the relationships between the pairs of heavy metals Cr - Pb, Cu - Zn, and Cr - Cu are not significant. Meanwhile, the pair of heavy metals Cu - Hg has a significant relationship. The percentage of mutual influence between the concentrations of heavy metals in the sediment can be explained through the Coefficient of Determination (R^2). The calculated Coefficients of determination for the four pairs of heavy metals are 21.6% (Cr - Pb), 23.3% (Cu - Zn), 49.8% (Cu - Hg), and 36.4% (Cr - Cu).

The source of heavy metals in the sediments in this study can be predicted through the grouping of pairs of heavy metals that are related to each other. From this grouping, the common behaviour and potential sources of each group of heavy metals can be determined. In this study, the first group consists of the heavy metals Cu, Hg, and Zn, which have positive correlations with each other. This group likely originates from the same anthropogenic sources. These three metals are associated with domestic waste, industrial waste, and agricultural runoff due to the disposal of organic materials **(Lawson, 2014)**.

The second group consists of the heavy metals Cr and Pb, which are potentially derived from the metal industry, steel, and fuel types such as gasoline and ship fuel spills. The research area located in the harbor allows for the observation of associations between these two metals. The third group includes heavy metals Cr and Cu, which have a strong relationship and likely originate from the same anthropogenic sources. Heavy metals Cr and Cu have a strong relationship because they originate from the metallurgy, fertilizer, and pesticide industries **(Xu *et al.*, 2023)**. This can be observed in the Cirebon region itself, which has a high population density and industrial activity **(Patty *et al.*, 2018)**.

Table 13. Pearson correlation analysis of heavy metals in sediment

	Cd	Cr	Cu	Hg	Pb	Zn
Cd	1					
Cr	-0.221	1				
Cu	0.013	-0.603	1			
Hg	-0.042	-0.072	**0.706**	1		
Pb	-0.070	**0,465**	-0.090	0.247	1	
Zn	-0.017	-0.048	**0.483**	0.063	0.139	1

CONCLUSION

The presence of heavy metals in the research area of PPN Kejawanan and PPI Gebang Mekar is detected in the water, including Pb and Zn, while the sediment contained Cd, Cr, Cu, Hg, Pb, and Zn. The water quality measurements using the STORET method classified the area near the Ciberes River estuary in Gebang Mekar as Class C (Moderate Pollution), the Kejawanan fishing port area as Class B (Light Pollution), and the Kejawanan Mangrove ecosystem as Class A (Compliant with Standards). The concentration of heavy metal Cd in the sediment at all stations showed the highest concentration among all the metals studied, with potential ecological risks that could be harmful to aquatic ecosystems (RI = 341.10-428.7). The ecological risk index for heavy metals at all stations followed this order: Cd > Hg > Pb > Cu > Zn > Cr.

Pearson correlation analysis of heavy metals in sediment revealed potential sources of some groups of strongly correlated heavy metals. The Cu-Hg-Zn group of heavy metals identified in this study likely originates from domestic waste, industrial waste, and agricultural runoff due to the disposal of organic materials. The Cr-Pb group of heavy metals may come from port activities and ship painting that uses paint containing Cr, while the Cu-Cr group of heavy metals likely originates from metallurgy and agricultural waste. Research on heavy metals in regions with active industries in Indonesia is essential to monitor and assess the input of heavy metals into water bodies that could potentially harm marine ecosystems.

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