Distribution of dissolved heavy metals Hg, Pb, Cd, and As in Bojonegara Coastal Waters, Banten Bay

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Abstract. Heavy metals in the waters come from many sources such as anthropogenic activities and could significantly impact the environment. This study aimed to determine the distribution of dissolved heavy metals of Hg, Pb, Cd, and As in the coastal waters of Bojonegara. Sampling was conducted from October to December 2019 at six stations. Data analysis consisted of distribution of heavy metals, heavy metal correlation with water quality, and comparison of actual heavy metals concentration to heavy metals quality standard concentration. Based on results, the concentrations of Hg, Pb, Cd, and As at six stations were below the detection limit, except Pb in November. The high Pb concentration was found the most in the river area and the concentration getting lower towards the sea. Based on correlation analysis results, heavy metals concentration is influenced by temperature, salinity, pH, and DO. According to Ministerial Decree of Environment No. 51 of 2004 concerning seawater quality standards for biota, the concentration of Hg, Pb, Cd, and As in coastal waters of Bojonegara still in good condition for aquatic life.

Keywords: Bojonegara Coastal; heavy metals; water quality

1. Introduction

Banten Bay is one of the waters in Banten Province, located in the northern part of Java Island, Serang City, and Cilegon with an area of approximately 150 km² [1]. This bay has 12 large and small islands and has seven river basins that empty into this bay, namely the Cibanten, Cikamayung, Ciujung Lama, Cilengkong, Ciranjiang, Ciluncing, and Ciujiang Rivers [2]. In the last two decades, land use around the bay has increased along with population growth.

The growth of the human population and increasing demand for various types of goods have resulted in the development of various industries in the mainland area of the bay. This condition has the potential to pollute the surrounding environment. One of the environments that can be polluted is the aquatic environment. If the aquatic environment is polluted, the living things in the environment will be affected by various negative impacts. Pollution of the aquatic environment can be caused by various things, including land use, rock weathering, rainfall, and anthropogenic [3].

Chemical, environmental pollution is one of the most dangerous pollutions, because the resulting waste can dissolve in water and cannot be seen and has a hazardous impact on living things. Heavy metals are an example of chemical waste that can cause pollution to the aquatic environment, and one of the sources of this waste can be generated from industrial waste. Heavy metals have permanent
properties in the environment, are challenging to be broken down biologically, accumulate in the body, and accumulate in sediments so that the level of toxicity will continue to increase and the negative impact will also increase on living things [4]. The source of heavy metal waste in the coastal waters of Bojonegara is might be more dominantly produced from anthropogenic activities or human activities. There are several industrial activities in the coastal waters of Bojonegara that presumably have the potential to contribute to heavy metal waste, such as PT. Angel Products, PT. Samudera Marine Indonesia, PT. Anugerah Buana Marine, PT. Duta Sugar Internasional, and PT. Batu Alam Makmur [5].

Research on dissolved heavy metals has been carried out in several locations in the waters of Banten Bay, including Teluk Lada [6], the estuary of the Cisadane River [7], and Banten Bay [8]. The results of these studies indicate that the largest source of heavy metals comes from anthropogenic activities. In particular, studies regarding the content of heavy metals in the coastal waters of Bojonegara, Banten Bay have never been conducted. The coastal waters of Bojonegara are one of the areas that have a fairly rapid industrial development; thus, an assessment of water quality needs to be carried out. This research aimed to determine the distribution of dissolved heavy metals of Hg, Pb, Cd, and As in the coastal waters of Bojonegara.

2. Methods

2.1. Time and location

This research was conducted in coastal waters of Bojonegara, Banten Bay, Bojonegara District, Banten Province (figure 1) from October to December 2019. The data was collected three times at intervals of once a month at six observation stations. Analysis of heavy metal content was carried out at the PT. Bumi Ventila Indonesia. TSS and turbidity analysis was carried out at the Laboratory of Environmental Aquaculture, Department of Aquaculture, Faculty of Fisheries and Marine Sciences, IPB University.

Figure 1. Research location in coastal waters of Bojonegara, Banten Bay.
Sampling was done based on area representation using the purposive sampling method. Stations 1 and 6 are characterized by the existence of a fish auction place (TPI), a place for fishing boats to rest, and close to residential areas. Station 2 is characterized by shipbuilding activities. Stations 3 and 4 are industrial areas. Station 5 is characterized by the presence of steam-fired power plant (PLTU) activities.

2.2. Data collection
Data collection was carried out in situ and the laboratory for analysis. Data were collected every month by observing and measuring several water quality parameters at six observation stations. The parameters measured include temperature, pH, transparency, depth, dissolved oxygen, salinity, turbidity, and total suspended solids (TSS). Heavy metal parameters used as the primary data were mercury (Hg), lead (Pb), cadmium (Cd), and arsenic (As). The lead, cadmium, arsenic, and mercury content were analyzed using the Atomic Absorption Spectrophotometry (AAS) PinAAcle 900H.

Water samples were taken from the surface layer. A water sample of approximately 250 ml was put into a polyethylene bottle that had been given 1 ml of concentrated nitric acid (HNO\(_3\)) solution with a concentration of 96%. The bottle is put into a cool box and then taken to the laboratory. The heavy metal analysis was performed using the flame method, which refers to the American Public Health Association [9].

2.3. Data analysis
2.3.1. Analysis of water quality conditions
Analysis of water quality conditions was used to determine the condition of the heavy metals of mercury, lead, cadmium, and arsenic obtained. The quality standard used to analyze heavy metals' condition refers to the Decree of the State Minister of the Environment Number 51 of 2004 concerning Sea Water Quality Standards for Biota.

2.3.2. Distribution of heavy metals and TSS
The distribution of TSS and the heavy metal content of mercury (Hg), lead (Pb), cadmium (Cd), and arsenic (As) were analyzed using the interpolation method, by combining the heavy metal content with the sampling coordinates. Heavy metal content data is displayed in a distribution image by displaying a color scale as information on heavy metal content.

2.3.3. Correlation coefficient
Correlation analysis is used to determine the relationship between water quality parameters consisting of temperature, dissolved oxygen, suspended solids, salinity, and pH with heavy metal parameters of Hg, Pb, Cd, and As. According to Sugiyono [10], the determination of correlation can be done with the following formula:

\[
rx_{xy} = \frac{n \Sigma x_i y_i - \Sigma x_i \Sigma y_i}{\sqrt{(n \Sigma x_i^2 - (\Sigma x_i)^2)(n \Sigma y_i^2 - (\Sigma y_i)^2)}}
\]

Where,

- \(rx_{xy}\) = Correlation coefficient
- \(x_i\) = Independent variable
- \(y_i\) = Dependent variable
- \(n\) = Amount of data

The results of the calculation will provide several alternatives. If the correlation value (r) approaches 0, then the correlation between the two variables is very low. If the correlation value (r) approaches +1, then the correlation between the two variables is strong and unidirectional or positive. If the correlation value -1, then the correlation between the two variables is strong but opposite or negative [10]. Intervals and descriptions of correlation levels are presented in table 1.
Table 1. Interval of the correlation value.

| Coefficient interval | Correlation level |
|----------------------|-------------------|
| 0.00-0.199           | Very weak         |
| 0.20-0.399           | Weak              |
| 0.40-0.599           | Medium            |
| 0.60-0.799           | Strong            |
| 0.80-1.000           | Very strong       |

Source: Sugiyono [10].

3. Result

3.1. Water quality coastal waters of Bojonegara
Salinity coastal waters with a range of 10-37 psu. Dissolved oxygen (DO) ranged from 5.3 to 8.0 mg/L; the degree of acidity (pH) of the waters during the observation ranged from 5.00 to 7.66 (table 2). Based on the analysis results, the heavy metal content of mercury, cadmium, and arsenic during the observation was below the detection limit, except for the heavy metal lead in November, which ranged from 0.003-0.006 mg/L (table 3).

Table 2. Value of water quality parameters in coastal waters of Bojonegara.

| Parameters       | Month          |
|------------------|----------------|
|                  | October        | November      | December      |
| Temperature (ºC) | 31.4-35.0      | 31.9-36.2     | 32.0-39.0     |
|                  | (32.73 ± 1.32) | (34.17 ± 1.73)| (35.25 ± 2.36)|
| Transparency (m) | 0.19-1.55     | 0.3-1.5       | 0.21-1.53     |
|                  | (0.78 ± 0.56)  | (0.89 ± 0.49) | (0.89 ± 0.55) |
| Turbidity (NTU)  | 7.00-162.0     | 9.80-96.0     | 4.20-80.00    |
|                  | (44.60 ± 59.52)| (30.63 ± 32.87)| (19.32 ± 29.83)|
| TSS (mg/L)       | 7.1-123.0      | 6.4-61        | <3-28         |
|                  | (37.03 ± 44.37)| (22.07 ± 20.83)| (17.50 ± 14.85)|
| pH               | 5.0-8.0        | 5.80-6.33     | 6.41-7.66     |
|                  | (6.17 ± 0.31)  | (6.04 ± 0.24) | (7.12 ± 0.48) |
| Salinity (psu)   | 21-37          | 30-36         | 10.0-32.0     |
|                  | (31 ± 5.76)    | (34 ± 2.10)   | (27.67 ± 8.71)|
| DO (mg/L)        | 5.7-6.5        | 5.7-7.6       | 5.3-8.0       |
|                  | (6.17 ± 0.31)  | (6.67 ± 0.69) | (6.63 ± 1.02) |

Table 3. Heavy metal concentrations.

| Heavy Metals | Month          |
|--------------|----------------|
|              | October        | November      | December      |
| As           | <0.002         | <0.002        | <0.002        |
| Cd           | <0.001         | <0.001        | <0.001        |
| Pb           | <0.002         | 0.003-0.006   | <0.002        |
| Hg           | <0.0002        | <0.0002       | <0.0002       |

3.2. Distribution of total suspended solids (TSS)
The highest TSS concentration is obtained at station 1 (river area), and its concentration tends to decrease towards the sea. The highest concentration is in October and the lowest is in December (figure 2).
Figure 2. The temporal distribution of TSS in coastal waters of Bojonegara (a) October, (b) November, and (c) December.
3.3. Distribution of heavy metal lead (Pb) in November

The concentration of heavy metal lead in coastal waters of Bojonegara in October and December was below the detection limit, while in November, it ranged from 0.003-0.006 mg/L. The highest Pb concentration is at station 1 and tends to decrease towards the sea (figure 3).

Figure 3. Spatial distribution of heavy metal lead in November.

3.4. Correlation coefficient

The correlation analysis was conducted for water quality parameters and the heavy metal lead content in November (table 4). Temperature and TSS have a stronger relationship, whereas salinity and DO had a lower relationship.

| Parameters | R²    | r   | Annotation   |
|------------|-------|-----|--------------|
| TSS        | 0.234 | 0.483 | Medium       |
| Temperature| 0.393 | 0.627 | Strong       |
| Salinity   | 0.033 | 0.182 | Very weak    |
| DO         | 0.092 | 0.303 | Weak         |
| pH         | 0.171 | 0.413 | Medium       |

4. Discussions

The coastal waters of Bojonegara are one of the waters in Bojonegara District, Banten Bay, Serang Regency. These waters are waters adjacent to housing, industry, and port areas. High anthropogenic activities around in coastal waters of Bojonegara can contribute to waste, both organic and inorganic waste.

The wastes that enter the coastal waters of Bojonegara may contain various types of hazardous materials that can cause pollution to the aquatic environment. One of these is heavy metals, one of the most dangerous wastes for the aquatic environment [11]. Heavy metals have properties that are difficult to decompose so that they can accumulate and endanger biota in the aquatic environment [12]. The
waste from industrial activities that enter the coastal waters of Bojonegara can result in a high TSS value. The TSS value can also be affected by low rainfall during the observation.

Spatially, the TSS content obtained during observations at six stations generally fluctuated. The highest TSS concentration is at station 1 (river area). This condition can be caused by a large amount of household waste input at the station. TSS concentrations will be higher in areas close to pollutant sources and will experience a decrease in concentration towards the sea due to dilution [13]. Helfinalis [14] states that one of the factors that can increase the TSS value in waters is the input of waste from the land.

The TSS concentration obtained is directly proportional to the turbidity value. The highest turbidity value is obtained at station 1 (river area). High turbidity values in waters can be influenced by high waste input and supported by shallow waters [15]. The physical conditions at station 1 are relatively shallow, and the water transparency value tends to be low. Based on Ministerial Decree of Environment No. 51 of 2004 concerning Sea Water Quality Standards for Biota, the TSS value at station 1 has exceeded the quality standard, where the quality standard set is 80 mg/L.

Temporally, the highest TSS concentration was obtained in October and the lowest in December. The high TSS concentration value in October can be caused by high waste input and low rainfall. The relatively small volume of water can cause the accumulation of organic and inorganic waste. The TSS value, which tends to be lower in December, can be influenced by high rainfall since the diluting factor's increasing volume. TSS in an aquatic environment presumably can affect the concentration of heavy metals. It is because TSS can contain various types of hazardous materials, one of which is heavy metals.

Based on the analysis results, the heavy metal content of Hg, Pb, Cd, and As tended to be below the detection limit. The highest concentration of heavy metal Pb is found at stations 1 and 6 (river area), and its concentration tends to decrease towards the sea. The heavy metal concentration, which tends to be below the detection limit, is thought to be caused by the very low concentration of heavy metals in water. Therefore, the tool cannot read the concentration (the concentration is less than the tool's readability). Low heavy metal concentration values can be caused by currents that cause water mass transfer, both moving in and out of one water to another [16]. Wisha et al. [17] stated that surface currents in Banten Bay moved away from the coast caused by tidal events. Wulan et al. [18] also stated that the concentration of heavy metals in waters could vary; this depends on activities around the aquatic environment and conditions of the aquatic environment.

Temporally, heavy metal concentrations were generally not significantly different, except for the heavy metal lead in November, which experienced an increase. The increase in lead concentration in November can be affected by low rainfall so that the waste that enters these waters can experience accumulation. The heavy metal lead concentration decreased in December; this could be caused by high rainfall, resulting in a high dilution process [19].

The presence of heavy metals in the aquatic environment can come from nature or anthropogenic activities. The heavy metal lead (Pb) in waters can come from the use of paint used to coat metals to slow down the occurrence of rusting and accelerate the dry rate of paint [20]. The use of lead in fuel can also be a lead source in the aquatic environment [21]. Verma et al. [22] stated that the heavy metal lead could also come from power plants that use coal as fuel.

Heavy metal mercury (Hg) can naturally come from volcanic gases that exist at the bottom of the waters. In contrast, by anthropogenic activities, heavy metal mercury can come from the metal casting industry and the use of pesticides [23]. The heavy metal cadmium that can pollute the aquatic environment can come from the use of cadmium as the main ingredient in the alloy industry, refining zinc metal (Zn), and in pesticides [24]. Authman et al. [23] stated that cadmium metal in the aquatic environment could also come from the battery industry and the plastic industry. Heavy metal arsenic (As) can come from arsenic use in industrial activities, such as the metal ore processing industry, the mining industry, the metal plating industry, and the paint removal process [25].

Based on the correlation analysis results, it was found that the water quality parameters influenced the concentration of heavy metals. Wisha et al. [26] stated that water quality parameters, such as temperature, pH, DO, and water salinity, can affect the concentration of heavy metals in water. In
general, high temperatures will cause heavy metals to tend to be in the sediment [27]. A high dissolved oxygen concentration will make it easier for heavy metals to dissolve in water, while a low DO value will tend to increase the levels of heavy metals in the sediment [28]. A low pH value will reduce the solubility of heavy metals in water, which increases heavy metal levels in the bottom of the water [28]. High water salinity can lead to the formation of clumps of organic matter and accelerate the deposition of heavy metals so that the concentration of heavy metals in water will decrease [29]. Usman and Adel [30] also say that waters with high salinity will cause alkaline and alkaline cations to tend to be present in solid particles.

The correlation analysis results showed that the temperature and TSS parameters had a greater relationship and influence on heavy metal concentrations. The water temperature values obtained during the observation ranged from 31.4 to 39.0 °C. Water temperature that is high enough during data collection can cause heavy metal concentrations in the water to be low. This is because high temperatures will result in the formation of heavy metal ions so that heavy metals will tend to settle and enter the sediment [27]. One of the factors that cause the high temperature of waters is the data collection carried out during the day, which is from 10.00 am to 03.00 pm. Furthermore, the high-temperature value at station 5 is because the location is adjacent to the PLTU, and the high temperature of wastewater is suspected entered the waters.

The dissolved oxygen and salinity had a lower effect on the concentration of heavy metals. Based on the Decree of the State Minister of Environment No. 51 of 2004, DO and salinity are following quality standards. The dissolved oxygen value obtained during the study ranged from 5.3 to 8.0 mg/L, and the average value of the water salinity was 30.89 psu. DO concentrations according to quality standards in waters can be influenced by photosynthetic activities that occur during the day and the movement of water masses, which results in the diffusion of oxygen into the waters [31]. The salinity of waters can be affected by water evaporation and mixing by other water. The oxygen diffusion process will be faster because Banten Bay's waters have tides twice a day, and the waters tend to be shallow [32].

The concentration of heavy metals in the waters can be affected by the pH of the water. The pH value obtained during observation ranges from 5.00-7.66. It shows that the coastal waters of Bojonegara have pH, which tends to be acidic. A low pH value will reduce the solubility of heavy metals in water, which increases heavy metal levels in the bottom of the water [28]. Li et al. [33] also stated that the low acidity level (4-7) would accelerate the release of heavy metals into the sediment. The pH value that tends to be acidic in the coastal waters of Bojonegara is thought to be caused by the entry of high levels of waste or organic material through the river [34].

5. Conclusion
Based on Decree of the State Minister of Environment No. 51 of 2004 concerning Sea Water Quality Standards for Biota, the concentration of heavy metals Hg, Pb, Cd, and As in the surface layer in coastal waters of Bojonegara was still under quality. The high concentration of heavy metal lead the most found in the river area, and the concentration was getting lower towards the sea. Based on Ministerial Decree of Environment No. 51 of 2004 concerning seawater quality standards for biota, the concentration of Hg, Pb, Cd, and As in coastal waters of Bojonegara is meet the quality standard. It means that Bojonegara's coastal waters are still in good condition.

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