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Assessment of Heavy Metal Pollution (Hg, Cd, Pb) in The Pasuruan Sea: Status and Countermeasures

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ABSTRACT

Pasuruan Regency is a major industrial center in Indonesia, and its heavy metal industrial waste, accompanied by household waste and agricultural activities, pollutes Pasuruan waters. The purpose of this study was to determine the status of heavy metal pollution in the Pasuruan Sea in the coastal areas of Kraton, Lekok, and Nguling and to find out how to overcome it. The survey method was used in this study to analyze the levels of heavy metals in the waters and calculate the water pollution status based on the pollution index and water quality. Water sampling is carried out at a depth of between 2-15 meters at 3 stationary lift nets (*bagan tancap*) at each location. The results showed that the highest levels of heavy metals were found in Lekok waters. The results of the calculation of the pollution index (IP) show that Lekok water has a heavy pollution status for all types of heavy metals. The pollution status in Nguling and Kraton waters based on the Hg and Cd pollution index is classified as heavily polluted. Meanwhile, Pb's heavy metal levels still meet water quality standards. The results of measurements of temperature, pH, dissolved oxygen, salinity, and current velocity are classified as optimum for aquatic biota. Based on the results, areas of Pasuruan waters are heavily polluted. Therefore, better water quality management is needed to meet the needs of biota by planting *Rizhophora mucronata* mangroves.

Keywords: Heavy metal; Pollution index; Pasuruan Regency

INTRODUCTION

Sea water is a component that can be relied on in the terrestrial environment, where the waste disposal from the land will emptied into the sea. The waste containing these pollutants will enter the coastal and marine ecosystems. Some are dissolved in the air, some sink to the bottom and are concentrated into sediments, and some enter the body tissues of marine organisms (Ika et al., 2012). The water environment in Pasuruan Regency that has the potential to be contaminated by heavy metals is the coastal area in Lekok, Nguling, and Keraton Districts. Lekok Beach has the potential to be polluted because it gets input from the Rejoso River and several tributaries, where each part of the tributary has residential areas. These industrial and agricultural activities have the potential to dispose of waste into the river (Haryono et al., 2017). In the end, the waste or organic and inorganic waste from the tributary will flow and be buried in Lekok Beach.

The presence of heavy metals in marine waters can come from various sources, including mining, household, agricultural waste, and industrial waste (Masriadi et al., 2019). Marine pollution is limited as a negative impact (harmful effects) on the life of biota, resources, the comfort of marine ecosystems, and human health caused by the disposal of materials or waste directly or indirectly from human activities (Yennie & Martini, 2005). There are two types of heavy metals, namely essential heavy metals needed by organisms such as Zn, Cu, Fe, Co, and Mn; the second is non-essential heavy metals that are toxic, such as Cd, Pb, and Cr (Said, 2010). High pollution of Pb, Cd, and Hg from rivers contaminated with industrial, residential, and agricultural wastes. One of the rivers contaminated with heavy metals Pb, Cd, and Hg is the Rejoso and Wangi River, which empties into the Lekok beach because, around the river, there is a factory that has the potential to produce waste containing Pb, Cd, and Hg (Adam et al., 2018). The decline in environmental quality due to heavy metal

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pollution has an impact on decreasing productivity and hygiene of fishery commodities, so it has a negative impact on decreasing marketing value (Siregar & Martini, 2008).

Heavy metal pollution in the waters results in health problems for consumers who consume fish from polluted waters and will negatively impact marine ecosystems. Biomagnification is a process that causes pollutants to increase in concentration along with the increasing position of living things in a food chain. Bacteria, phytoplankton, and zooplankton absorb heavy metals in water and sediment, and fish eat the microorganisms. In the fish body, metals are absorbed by the blood and bind to blood proteins, which are then distributed to all body tissues. (Wicaksono et al., 2013).

Yellowstripe scad (*Selaroides leptolepis*) is one of the fish caught in Pasuruan waters, and it has a high economic value. The last recorded catch of wild fish in Lekok waters (in May 2019) was 117 kg with a selling price of Rp23,000,-. Apart from being consumed by the community, Yellowstripe scad fish catches in Pasuruan waters are also marketed between cities (PPIP Lekok, 2019). The purpose of this study was to determine the status of heavy metal pollution in the Pasuruan Sea in the coastal areas of Kraton, Lekok, and Nguling and to find out how to take countermeasures.

MATERIALS AND METHODS

Study site

Pasuruan Regency is a district in the province of East Java, Indonesia, which is located in the Golden Triangle area because it is located on the axis of economic distribution in 3 areas, namely the Surabaya - Jember - Banyuwangi - Bali, Surabaya - Malang, and Malang - Jember routes. Pasuruan Regency is located between Mojokerto Regency, SIdoarjo Regency, Probolinggo Regency and Malang Regency. Pasuruan Regency lies at 7°30'- 8°30' south latitude and 112°30' - 113°30' east longitude. Pasuruan Regency is an area that has both flat and mountainous areas with an altitude of 0 to more than 1,000 m above sea level. Areas that have an average height of up to 100 m above sea level (above sea level) are 14 districts, namely Kejayan, Wonorejo, Gempol, Beji, Bangil, Rembang, Kraton, Pohjentrek, Gondangwetan, Rejoso, Winongan, Grati, Lekok and Nguling (Agustini & Winarni, 2014). The coordinates can be seen in Table 1, and a map of the research sampling location can be seen in Figure 1.

Sample collection

The research method used was a marine survey in the Pasuruan district. Yellowstripe scad samples were taken from charts in the waters of Nguling, Lekok, and Kraton as regional representatives in the Pasuruan Sea because the three locations are close to industrial factories and community settlements. This research was conducted for two months, namely from August to September 2019.

By taking biweekly data from the fishermen's catch. Fish were caught using fishing gear called "bagan tancap" which was operated at each research location. Fish and water samples are taken at 10:00 am in sunny weather to facilitate the fishing process and in-situ measurement of water quality parameters as research support parameters (temperature, pH, salinity, current velocity, and dissolved oxygen) with a multiparameter measurement tool, namely AAQ. AAQ is a tool called the chlorotic probe. This chlorotic probe consists of a series of sensors and monitors. The sensor circuit consists of a temperature sensor, salinity, dissolved oxygen, pH, turbidity, depth, and chlorophyll-*a*. The research method uses a survey method with probability sampling.

Fish were taken randomly from each study site and then analyzed. Fish that are caught with the same effort are carried out randomly by taking samples of fish as many as 6 fish from each sampling point and 18 fish from each station with a minimum number of 54 to represent the waters. Water sampling for heavy metal analysis in water and fish for heavy metal analysis in the kidneys was carried out at 3 points in

Table 1. Coordinate points of research sampling locations

Location	Sampling Point (Coordinates of a point)		
	1	2	3
Nguling	7 ° 39.039'S and 113 ° 5.501'T	7 ° 36.431'S and 113 ° 5.022'T	7 ° 37.431'S and 113 ° 6,022'T
Lekok	7 ° 37.395'S and 112 ° 59.772'T	7 ° 35.409'S and 113 ° 0.812'T	7 ° 35.409'S and 113 ° 0.812'T
Kraton	7 ° 35.446'S and 112 ° 55.558'T	7 ° 33.555'S and 112 ° 54.890'T	7 ° 32.555'S and 112 ° 44.890'T

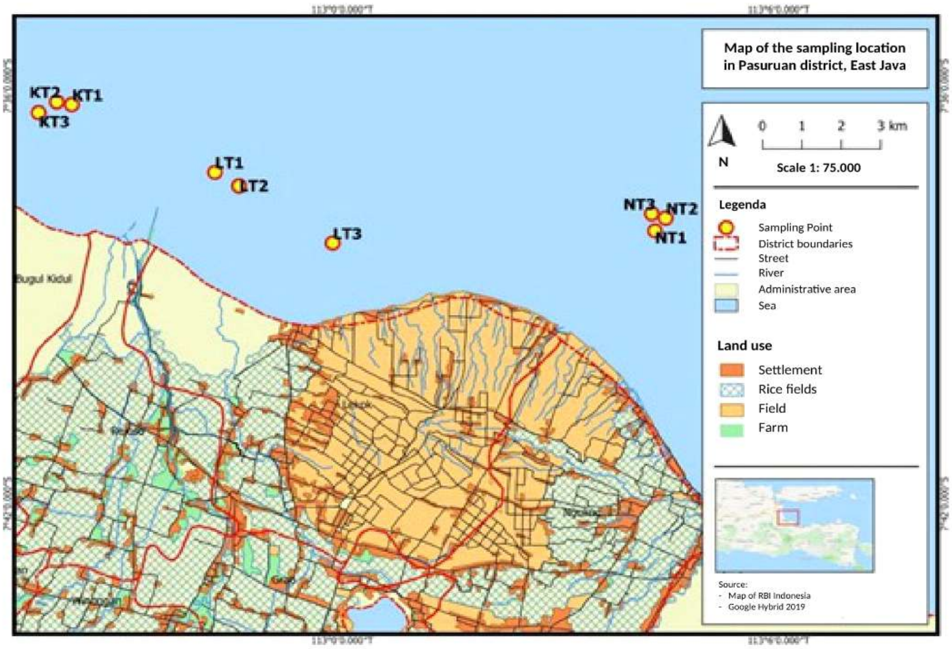

Figure 1. Fish and water sampling point

each study location. Fish kidneys are one of the first organs exposed to water pollution. Kidneys play a major role in metabolic excretion, digestion, and storage of various elements. Kidneys function to filter and excrete materials that are not needed by the body, including toxic heavy metals (Taukhid et al., 2007). Causes the kidneys to often experience histological damage due to metal toxicity. Determination of water sampling points is based on the number of charts at each location with a depth of 5 to 15 meters. Point 1 at a depth of 5 meters, point 2 at a depth of 10 meters, and point 3 at a depth of 15 meters from the water surface to determine the mixing of the water at a certain depth. Water and fish samples were put into a cool box filled with ice cubes for preservation for a 3-hour trip to the laboratory. Furthermore, the fish is taken from the kidney and then put into a plastic clip to analyze the levels of heavy metal Hg in the kidneys and levels of heavy metals such as lead (Pb), Mercury (Hg), and Cadmium (Cd) in water which was conducted at the Chemical Laboratory of the Faculty of Mathematics and Science. Natural Sciences, Brawijaya University, Malang.

The status of heavy metal pollution at the research location is determined using the pollution index method with the following formula:

$$IPj = \sqrt{\frac{\left(\frac{Ci}{Lij}\right)_M^2 + \left(\frac{Ci}{Lij}\right)_R^2}{2}}$$

IPj = pollution index for designation j
Ci = concentration of water quality parameters i
Lij = concentration of water quality parameters I stated in the water designation standard j
M = Maximum
R = Average

The value of IP water quality is determined from the maximum result value and the average value of the concentration ratio per parameter to the quality standard value. There are 4 IP index classes:
Score $0 \leq Pij \leq 1.0$ (good)
Score $1{,}0 < Pij \leq 5.0$ (slightly polluted)
Score $5{,}0 < Pij \leq 10$ (fairly polluted)
Score $Pij > 10$ (heavily polluted).

RESULTS AND DISCUSSION
Results

The results of heavy metal measurements at the 3 research locations showed that the average Hg level in Lekok waters was 1.33 mg/l, Kraton waters 0.52 mg/l, and Nguling waters 0.18 mg/l (Figure 2a). The highest levels of heavy metal Hg were found in Lekok waters at all sampling points, namely at point 1 at 1.39 mg/l, point 2 at 1.18 mg/l, and point 3 at 1.41 mg/l. The results of the measurement of Cd heavy metal at the 3 research locations showed that the average Cd level in Lekok waters was 1.22 mg/l, Kraton waters were 0.44 mg/l, and Nguling waters was 0.37 mg/l (Figure 2b). The highest Cd level on the Pasuruan coast was found in the coastal area of

Lekok, namely at point 1 at 1.51 mg/l, point 2 at 0.93 mg/l, and point 3 at 1.22 mg/l. The results of the measurement of Pb heavy metal at the 3 research locations obtained the average Pb level in Lekok waters of 0.49 mg/l, Kraton waters of 0.00 mg/l, and Nguling waters of 0.00 mg/l (Figure 2c). The highest Pb level on the Pasuruan coast was found in the coastal area of Lekok, namely at point 1 at 0.60 mg/l, point 2 at 0.49 mg/l, and point 3 at 0.38 mg/l

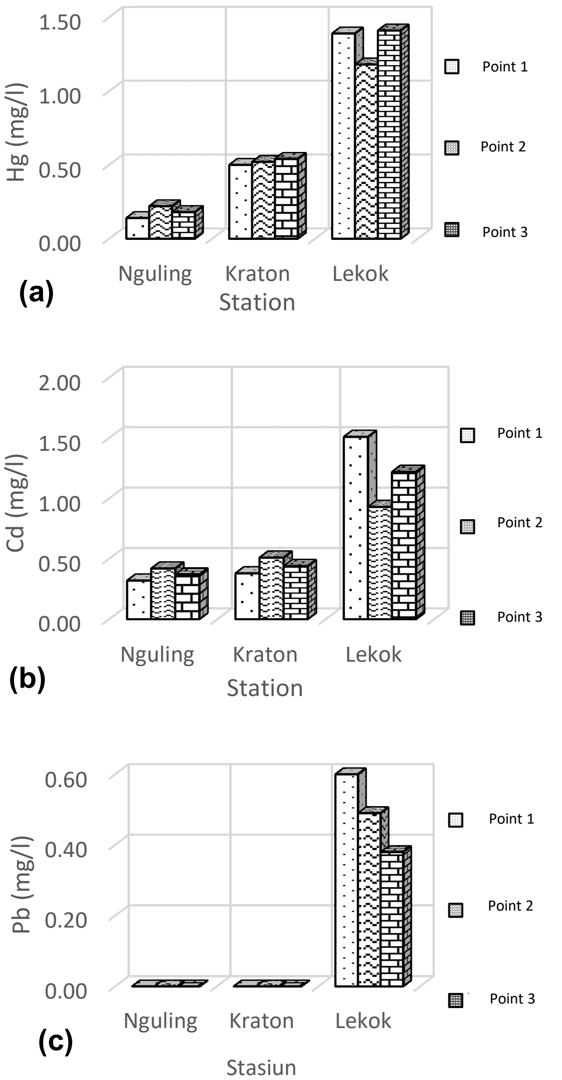

Figure 2. Results of Measurement of Heavy Metals in Pasuruan and Waters: (a) Hg, (b) Cd, (c) Pb

The results of measurements of accumulated Hg levels in Yellowstripe scad kidneys (*Selaroides leptolepis*) were obtained from Pasuruan Waters at sampling points in Nguling, Lekok, and The Kraton ranges from 1.57 - 8.13 mg/l (Figure 3). Hg levels in Selar fish kidneys in Nguling waters from three locations have an average of 1.92 mg/l, Lekok Waters 6.88 mg/l, and Kraton Waters 3.11 mg/l. The highest levels of Hg are found in Yellowstripe scad fish kidneys and Lekok Waters.

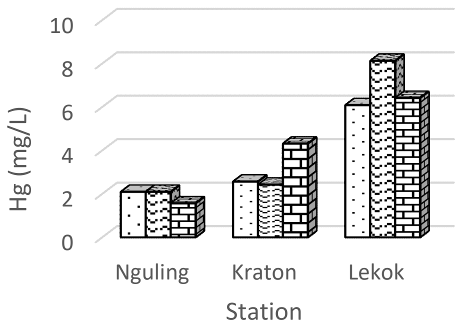

Figure 3. Results of Measurement of Hg Levels in the Kidneys of yellow stripe scad (*Selaroides leptolepis*)

The pollution status in Pasuruan waters is determined by observing the value of heavy metal levels. in the waters using the pollution index method. The results of the analysis of the pollution index value of each research location can be seen in Table 2. Based on the results of the calculation of the pollution index (IP), it was found that all research locations were in the heavily polluted category with an average pollution index of 100.50 - 1372.68 (IP> 10), which is a very high value and is categorized as heavily polluted, except for heavy metal Pb at the Nguling and Kraton locations with good status (0 d" IP d" 1,0). The waters at the Lekok research location have a very high IP value because the Lekok area is the center of fisheries in which there are activities in the field of fisheries such as fish auctions, ports, and also human activities that dispose of waste directly into the river which will end up in the Lekok.

Temperature and current velocity are physical parameters used to measure water quality (Table 3). The results of the water temperature measurements obtained the highest temperature value at the Lekok location at point 3, namely 30.5^{o}C. In contrast, the lowest temperature value was obtained at the Nguling location at point 1, namely 28.3^{o}C. The temperature obtained at 3 different locations did not show significant or relatively the same differences between the observation locations, ranging from 28.3 ^{o}C - to 30.5 ^{o}C. The current Velocity in Pasuruan waters ranges from 0.03 - 0.08 m/sec. The highest current velocity was obtained at the Nguling point 1 location of 0.08 m/sec, while the lowest value was obtained at the Kraton point 2 location of 0.03 m/sec.

The highest pH value occurred at Location Nguling

Table 2. Pollution Index Value in Pasuruan Waters

No.	Research Site	Heavy Metal Types	Pollution Index	Quality Standard Status	Category
1	Nguling	Hg	100.50	IP > 10	Heavily Polluted
		Pb	0.00	0 ≤ IP ≤ 1.0	Good
		Cd	395.79	IP > 10	Heavily Polluted
2	Kraton	Hg	183.85	IP > 10	Heavily Polluted
		Pb	0.00	0 ≤ IP ≤ 1.0	Good
		Cd	477.83	IP > 10	Heavily Polluted
3	Lekok	Hg	469.05	IP > 10	Heavily Polluted
		Pb	68.47	IP > 10	Heavily Polluted
		Cd	1372.68	IP > 10	Heavily Polluted

Table 3. Measurement of water quality

No.	Parameter	Nguling			Kraton			Lekok		
		1	2	3	1	2	3	1	2	3
1	**Tempeture (°C)**	28.3	29.2	29.1	29.7	30.3	29.3	30.4	29.8	30.5
2	**Current velocity (m/sec)**	0.08	0.06	0.07	0.04	0.03	0.04	0.05	0.04	0.05
3	**Power of Hydrogen/pH**	8.20	8.32	8.03	7.8	7.93	8.21	8.13	7.82	8.10
4	**Dissolved oxygen (mg/L)**	8.0	8.2	9.0	8.2	8.0	7.2	8.8	8.0	8.5
5	**Salinity (ppt)**	33	30	30	26	25	26	32	34	30

point 2 at 8.32, while the lowest pH value occurred at Kraton location point 1 at 7.8. The pH values obtained at all stations were relatively the same, ranging from 7.8 to 8.82. The amount of dissolved oxygen obtained during the study ranged from 7.40 to 9.05 mg/l. The highest dissolved oxygen value occurred at Location Nguling point 3, which was 9 mg/l, while the lowest dissolved oxygen value occurred at Kraton location point 3 at 7.2 mg/l. The salinity values obtained during the study ranged from 25 - 34 ppt. Salinity in Kraton waters is lower between the Lekok and Nguling points of the collection because these waters are close to the river mouth.

Discussion

Measurement of heavy metals Hg, Cd, and Pb in all locations had levels above the water quality standard except for Cd levels in the Kraton and Nguling waters. Heavy metal quality standards for marine waters are Hg of 0.002 mg/l, Cd of 0.001 mg/l, and Pb of 0.008 mg/l (Wulandari et al., 2012; Harmesa et al., 2020). The highest heavy metals are found in Lekok because these waters are used as fishing ports, and there are also community activities. According to (Amin et al., 2011), at this port, there are many human activities. The human activity in question is the activity of ships going in and out of the port in order to carry out loading and unloading activities as well as replacing fuel oil by ships. Port activities can be a source of heavy metal pollution in the surrounding waters.

Industrial wastes and a byproduct of the use of mercury compounds in agriculture generally cause high levels of mercury in waters. Mercury can exist in the form of metals, inorganic compounds, and organic compounds. Mercury fluctuations in the marine environment, especially in estuarine and coastal areas, are determined by sedimentation, flocculation, and adsorption-desorption reactions. Mercury transfer and transformation can be carried out by phytoplankton and bacteria because these two organisms relatively dominate the waters, as well as by seaweed. The bacteria can convert mercury to methylmercury and free mercury from the sediment (Hananingtyas, 2017). No pb levels were found at the Nguling location, and the palace was suspected of having no port at that location, so fuel oil spills rarely occurred; only anthropogenic waste was discharged from the river, which then entered the Nguling and the palace. According to Ricolleau et al. (2019), fuel oil generally gets tetraethyl additives containing Pb to improve quality, so the waste from these vessels can cause

Pb levels in these waters to be high. The heavy metal Pb contained in the fuel as an anti-oil breaking (such as Pb tetraethyl and tetramethyl) is then released into the atmosphere through the exhaust fume, and this part is then dissolved in the sea (Yumun, 2017). In addition, human activities that occur on land, such as household waste disposal through metabolic waste and corrosion of water pipes containing heavy metals, can also contribute significantly to the entry of heavy metals into marine waters. There are two main sources of heavy metal cadmium (Cd) contamination in the aquatic environment, namely through the earth's layer and human (anthropogenic) activities. Heavy metal cadmium is often used in human activities as a paint dye, and PVC / plastic as a nickel cathode. The main sources of metal cadmium contamination are industrial areas and pesticides produced in rice fields that flow into rivers, as well as spilled fuel from fishing boats (Masriadi et al*.,* 2019).

Based on the results of measurements of heavy metals in water, the heavy metal that has high levels is Hg, so what is studied in the kidney is heavy metal Hg. The content (amount and type) of metal in *yellowstripe* fish is determined by the absorption of fish during life in water, which is also related to the amount of heavy metals Hg dissolved in the water and its food chain. Metal pollutants that enter the aquatic environment can come from the air or anthropogenic activities on land. Water pollution by heavy metals Hg influences the local ecosystem due to its stable nature in sediment, low solubility in water, and its ease of being absorbed and collected in the body tissues of aquatic organisms, either through bioaccumulation or biomagnification processes, namely through the food chain (Vinodhini & Narayanan, 2009). Research conducted by Zodape et al. (2011) on marine fish in India showed that the absorption of heavy metals Hg by fish was greater in tissues such as the liver and gills than absorption in fish meat, so only a small portion of the heavy metal content in fish meat was detected. The presence of a higher content of heavy metals in the gills and liver is due to the fact that more of what enters the body is absorbed in the soft tissues (kidneys and liver) than is absorbed into other tissues. The order of heavy metal content in fish organs is highest to lowest in the internal organs (liver and kidney), bones, skin, and meat.

Based on the results of the calculation of the pollution index (IP), it was found that all research locations were in the heavily polluted category, except for heavy metals Pb in the Nguling and Kraton waters, which were included in the category of meeting quality standards with a pollution index value of 0.00. The waters of Nguling and Kraton have heavy metal levels of Pb below the water quality standard, possibly because these waters have begun to replant the mangrove ecosystem. Jenis mangrove yang ditanami adalah jenis Avicennia marina. Mangrove plants are coastal plants that can accumulate heavy metals in water areas (Sofian et al*.,* 2001). Absorption and retention of heavy metals by the rhizosphere layer around the roots will cause a sharp decrease in the concentration of heavy metals on the upper surface of the sediment layer, which can prevent the displacement of the surrounding coastal waters (Supriyantini et al*.*, 2017).

Water quality (temperature, flow velocity, pH, DO, and salinity) in Pasuruan waters are classified as good and optimum. Heavy metals are thought to be influenced by human activities, industrial activities, and activities at ports. Based on the statement of Flora et al. (2007), Lead (Pb) occurs naturally in the earth's crust. The presence of lead can also come from the results of human and industrial activities, which are 300 times more than natural Pb found in the earth's crust. Based on the statement of Patra et al. (2011), the presence of Cd and Hg in sediments and water is thought to originate from natural processes such as abrasion from rivers and community activities, such as disposal of market waste and household waste, as well as agricultural activities. The increase in water pH will cause the solubility of heavy metals to be smaller.

Furthermore, he states that the increase in temperature, low salinity, and pH values in these waters causes a greater level of bioaccumulation of heavy metals (Riani, 2015). The presence of heavy metals is also influenced by salinity in coastal and marine waters. The process of the solubility of heavy metals entering river estuaries into the sea is also influenced by temperature. In this case, the temperature of the waters is high, both at the mouth of the river and the sea, and the solubility of heavy metals is also getting higher. At the mouth of the river to the sea, the temperature water shows a lower value of 32.8-30.5^{o}C, so the solubility of pollutants in the waters is lower. In contrast, the heavy metal content in the river estuary is lower in height compared to the sea, causing the rate of biodegradation at high temperatures carried out by aerobic decomposing bacteria to increase and able to release chemicals into the air (Riani et al*.,* 2014).

Countermeasures in Lekok waters that have high heavy metal content include planting mangroves in the Nguling and Kraton areas. One type of mangrove that can absorb heavy metal content is *Rizhophora mucronata.* According to Pahalawattaarachchi et al.

(2009), explaining that *Rizhophora mucronata* mangroves can act as a heavy metal bioremediation agent because they can absorb and accumulate metals. *Rizhophora mucronata* has supporting roots. In addition to its function of helping trees stand upright, this type of root can also hold and stabilize soil sediment, thereby preventing the spread of pollutants to a wider area. Therefore, this type of mangrove root can more optimally absorb heavy metals. Various studies have shown that *R. mucronata* can accumulate heavy metals and act as a bioremediation agent. However, research on the bioaccumulation of heavy metals Hg and Pb, specifically in *R. mucronata*, has yet to be widely carried out, so it still needs to be studied.

CONCLUSION

The conclusion obtained is that the pollution status of heavy metals Hg, Pb, and Cd in Lekok waters, based on the IP results, is categorized as heavily polluted. The status of Hg and Cd heavy metal pollution at Nguling and Kraton based on IP results was categorized as heavily polluted. In contrast, heavy metal Pb at the Nguling and Kraton locations met the daily quality standards. In general, based on the pollution index, Pasuruan waters are heavily polluted waters, so there is a need for management of the Pasuruan coast, especially in Lekok waters, by planting *Rizhophora mucronata* types of mangroves, which are found in many Nguling and Kraton locations.

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