Jurnal Akademika Kimia, 14(1): 8-12, February 2025
ISSN (online) 2477-5185 | ISSN (print) 2302-6030
http://jurnal.fkip.untad.ac.id/index.php/jak/

Analysis of Bioaccumulation of Mercury (Hg) in Yuyu Crabs (Parathephusa Convexa) in The Janja River, Malomba Village, Dondo District, Tolitoli Regency

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Received 13 November 2024, Revised 13 January 2025, Accepted 03 February 2025
doi: 10.22487/j24775185.2025.v14.i1.pp8-12

Abstract

The phenomenon of environmental damage due to unlicensed gold mining activities harms aquatic ecosystems. Mercury (Hg) waste from the mining process has the potential to cause bioaccumulation in aquatic biota such as fish and crabs, which can lead to mercury contamination in the human body through the food chain. The bioaccumulation process can also produce toxic organic compounds that harm human health. Therefore, this research aims to determine whether mercury bioaccumulation occurs in yuyu crabs in the Janja River, Malomba Village, Dondo District, Tolitoli Regency, due to gold mining activities. The method used in the research is the calibration curve method with Cold Vapor Atomic Absorption Spectrophotometry (CV-AAS) analysis at a wavelength of 253.7 nm. The results obtained from this research indicate that bioaccumulation has occurred in yuyu crabs that live in the Janja River. The highest accumulated mercury metal levels were obtained at upstream locations with an average of 5.24 ppb; the lowest was in the midstream, which is 1.53 ppb. At the downstream location, it has a mercury concentration of 4.53 ppb. So it shows that the mercury contamination in the yuyu crab samples is below the quality standard threshold, Indonesian National Standard No. 7387 of 2009, for the heavy metal mercury in other types of crabs and crustaceans is 1.0 ppm or 1000 ppb.

Keywords: Mercury (Hg), bioaccumulation, yuyu crab, Tolitoli

Introduction

The damage or decline in environmental quality occurs in various sectors, including the mining sector. As a sector with high environmental risks, mining is always in the public spotlight. Unlicensed gold mining is a mining activity carried out without a permit. Unlicensed gold mining is generally carried out traditionally and uses the amalgamation method, which uses mercury (Hg) as a binder to separate gold ore from other metals or minerals. Thus, the waste produced can cause a decrease in environmental quality, and rivers can be polluted by mercury.

Mercury (Hg) and its derivatives have hazardous toxic properties, so their presence in the aquatic environment can cause harm. This is because mercury (Hg) easily dissolves and is bound in the body tissues of aquatic organisms. Besides, water pollution by mercury (Hg) also harms the local ecosystem. This is due to the stable nature of mercury in sediment, its low solubility in water, and its ability to be absorbed and collected in aquatic organisms' tissues through bioaccumulation or biomagnification processes (Silalahi et al., 2020).

Bioaccumulation refers to an increase in the concentration of certain chemical elements, for example, the metal ion Hg, in the bodies of living creatures and the trophic levels in the food pyramid. Heavy metals can accumulate through the food pyramid, where the higher an organism's position in the food pyramid, the higher the accumulation of heavy metals in its body (Jais et al., 2020). One of the heavy metals that often pollutes the environment is mercury (Hg), especially in the industrial sector or gold mining environments that use mercury in the production process (Pelu et al., 2022). If mercury enters the water, it will bind with chlorine and form an Hg-Cl bond, which causes the mercury to enter the plankton and move to other aquatic biota easily. Humans can also accumulate mercury through consuming contaminated food such as fish and shellfish (Narasiang et al., 2015).

Mercury is generally used as an extracting agent in gold mining activities, and the waste resulting from the process is usually discharged into rivers or other water bodies (Rasul & Musafira, 2022). Mercury waste that enters water bodies accumulates in sediment, or the mercury reacts with chlorine in water to form inorganic mercury (Hg-

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Cl), which microorganisms can then consume. Furthermore, other organisms can eat these microorganisms and cause bioaccumulation and biomagnification processes (Mulyadi et al., 2020).

Mercury compounds in Hg^{2+} can bind to cysteine residues in human proteins, causing them to lose activity. Besides Hg^{2+}, organic mercury compounds such as methyl and phenyl mercury are the most dangerous for health. They can damage the human nervous system through the bloodstream because they are very reactive and move quickly compared to Hg^0 and Hg^{2+}. Accumulation of mercury in the body tissues of organisms that live in areas polluted by this heavy metal can occur through metabolic processes (Abdullah et al., 2018).

One of the gold mining activities without a permit in Central Sulawesi province is in Dondo District, Malomba Village, Tolitoli Regency, which started in 2007. The gold ore processing process still involves using traditional tools such as pans and the amalgamation method, namely the extraction using an extracting agent, mercury metal (Hg) (Mulyadi et al., 2020).

The gold amalgamation process, traditionally carried out by the community, can release mercury into the environment, especially during washing and burning. Waste that generally still contains mercury is often thrown directly into rivers and has the potential to come into contact with aquatic biota. The gold processing process is carried out in the Janja River, which is a river used in daily life by the people of Malomba Village, such as a source of clean water and watering plants, as a source of drinking water for livestock around the river, and as a habitat for biota. Who lives in the river (Rosita et al., 2022).

Because the gold mining process is still carried out traditionally and uses mercury (Hg) as an amalgam or gold binding medium, which can cause environmental pollution, it is imperative to identify the concentration of mercury in the biota that lives around the mining area to find out how much bioaccumulation there is or how much pollution has already occurred.

Biota that can be used as a bioindicator to detect heavy metal mercury contamination in the Janja River is crustacean biota, such as crabs and shrimp. Crabs can be bioindicators to analyze heavy metal contamination because they can accumulate relatively high levels of heavy metals compared to other biota. After all, crabs move relatively slower than fish. Crabs are also aquatic biota that move and search for food at the bottom of rivers, so they are more easily affected by metal pollution in the water (Florentina & Ambarwati, 2017)

This research aims to determine whether mercury bioaccumulation occurs in yuyu crabs (Parathelphusa convex) in the Janja River, Malomba Village, Dondo District, Tolitoli Regency, due to gold mining activities.

Methods

Tools and materials

The tools used in this research were 100 mL sample bottle, 100 mL measuring cup, beaker, funnel, spatula, mortar and pestle, 50 mL measuring flask, dropper pipette, label paper, an electric bath, a set of Cold Vapor Atomic Absorption Spectrophotometry (CV-AAS) GBC Scientific Avanta tools, and the materials used were samples of yuyu crab (Parathelphusa convexa) that live in the Janja River, HNO_3 65 % solution, $HClO_4$ 70 – 72 % solution, H_2SO_4 95 - 97 % solution, distilled water, filter paper, and mercury standard solution 1000 ppb.

Sampling and preparation sample

Samples of yuyu crabs (*Parathelphusa convexa*) that live in the Janja River were taken from three sampling points: the upstream, midstream, and downstream points. At each point, three samples were taken every 5 meters in a vertical downward direction following the river water flow, so nine samples were taken. In the sampling process, the researchers referred to research conducted by Lige et al. (2022) using the cruising method, tracing river flows, and catching crabs directly using their hands. The captured samples are placed in plastic samples that have been treated with a small amount of seawater so that the samples obtained remain alive when taken to the laboratory research location. After that, the samples were taken for further analysis.

Sample preparation in this research refers to the procedure by Emelda et al. (2017), in that the yuyu crab (*Parathelphusa convexa*) sample is first cleaned. The sample is dried to reduce the water content; then, it is ground until it becomes flour. After that, the finely ground sample was weighed at 0.5 gram, then put into an Erlenmeyer for the sample destruction process, then 1 mL of distilled water was added, 2 mL of HNO_3:HCl solution in a ratio of 1: 1, and 5 mL of H_2SO_4 sequentially, then the sample is heated until the solution is clear, then the solution is cooled to room temperature, then diluted with distilled water in a 50 mL volumetric flask to the limit mark. After that, the sample was filtered using filter paper. Then the sample is ready to be analyzed for mercury concentration.

Preparation of standard solutions and calibration curves

Standard solutions were made in the series 10 ppb, 50 ppb, 100 ppb, 150 ppb, 200 ppb, and 250 ppb from a standard solution of 1000 ppb mercury. Then the absorption of the standard solution was measured, and a calibration curve was created for mercury metal.

Analysis using cold vapor atomic absorption spectrophotometry (CV-AAS)

The samples that have been obtained will have their mercury metal determined by measuring their absorption using Cold Vapor Atomic

Absorption Spectrophotometry (CV-AAS) at a wavelength of 253.7 nm. Furthermore, the absorption data obtained can be used to obtain the concentration of mercury metal in the sample by creating a calibration curve.

Data analysis techniques

Determination of mercury levels using calibration curves and linear regression. Systematically, it can be written with the equation (Purnama et al., 2020):

$$y = bx + a \rightarrow x = \frac{y-a}{b} \quad (1)$$

Explanation:
y: Sample absorbance
b: slope
x : concentration
a: Intercept

Results and Discussion

Preparation of standard solution and calibration curves

Standard solutions with various concentrations are 10 ppb, 50 ppb, 100 ppb, 150 ppb, 200 ppb, and 250 ppb. A calibration curve for the Mercury (Hg) standard solution was created. The regression equation is y = 0.0009x + 0.0112 with a correlation coefficient (R) 0.9996. This indicates that this meets the requirements of the Indonesian National Standard (SNI) 6989.84 (2009), which states that the allowable linear regression correlation coefficient is R ≥ 0.995 (Rusdianto et al., 2023). The calibration curve can be seen in **Figure 1.**

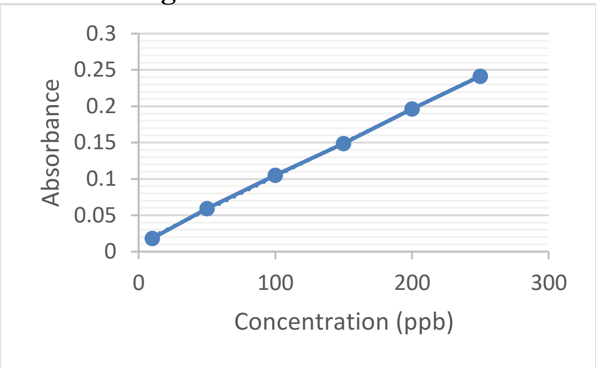

Figure 1. Calibration curve

Mercury concentration (Hg) of yuyu crab (parathelphusa convexa) samples

Based on this equation's calculations, data on mercury levels in yuyu crab (*Parathelphusa convexa*) were obtained as shown in **Table 1.**

Table 1. Concentration of Hg in yuyu crab (*Parathelphusa convexa*)

Location	Sample	Absorbance	Mercury (Hg) concentration (ppb)	Average Mercury (Hg) concentration (ppb)
	Point_1A	0.0152	4.37	
Upstream	Point_1B	0.0195	9.03	5.24
	Point_1C	0.0133	2.31	
	Point_2A	0.0154	4.59	
Midstream	Point_2B	0.0002	Not detected	1.53
	Point_2C	0.0002	Not detected	
	Point_3A	0.0179	7.29	
Downstream	Point_3B	0.0008	Not detected	4.53
	Point_3C	0.017	6.32	

The results obtained show the consentrations of the heavy metal mercury at location 1 is the upstream location of the river, three samples were taken at three different points, are point 1, point 2 and point 3 and the mercury (Hg) levels was obtained respectively is 4.37 (ppb), 9.03 (ppb), 2.31 (ppb), then at location 2 is the midstream, there are three samples with three different points are point 1, point 2, and point 3 which have a concentration of 4.59 (ppb). Two other locations were not detected, and at location 3 is the downstream location of the river, there were three samples taken at three different points, are point 1, point 2 and point 3 respectively, the mercury consentrations are 7.29 (ppb), 6.32 (ppb), and one location was not detected.

Based on the analysis results, the concentrations produced from upstream, midstream, and downstream locations show varied concentrations. This can occur because it depends on metabolic activities and the average life span of aquatic organisms or biota that live in that environment (Putra et al., 2021). The largest concentration of mercury was found in samples at location 1, or the upstream location of the river, with an average concentration of 5.24 ppb. This condition occurs because many mining locations are scattered around the upstream of the river. In addition, the close distance between the mining site and the sampling site can cause high mercury concentrations in the area. Intensive mining in upstream areas of rivers also contributes significantly to the increase in mercury levels detected in samples, considering that waste and toxic materials from mining activities tend to pollute surrounding water flows directly. Followed by mercury levels in downstream locations, the locations closest to residential areas with average mercury concentrations of 4.53 ppb. The mercury concentration in downstream locations is not much different from upstream locations because this area also has mining locations. Apart from that, this condition is also influenced by the downstream location being lower than the upstream and midstream locations, so the mercury contained in these two locations will be carried downstream following the river flow, which can cause mercury levels to increase. Then, the lowest mercury levels were found in the midstream location, with an average concentration of 1.53 ppb. This was because there was no mining activity at this location; the mercury levels obtained were due to the accumulation of mercury carried by the river flow from the upstream location.

The results obtained in this research are in line with the research conducted by (Yulis, 2018) in analyzing mercury (Hg) levels in river water affected by illegal gold mining, where the results obtained were that the highest average mercury levels were found in the upstream location of the river because there were many mining locations, then followed by the downstream location of the river, and the lowest mercury levels were obtained at the midpoint of the river. This research also further strengthens the results of the research on the analysis of mercury (Hg) metal in the Janja River water in the mining area which has been carried out by (Rosita et al., 2022), where the results obtained stated that it was true that the Janja River had been polluted by mercury metal.

This research shows that the biota, in this case the yuyu crab (*Parathelphusa convexa*) that lives in the Janja River, has been proven to experience mercury bioaccumulation in its body. The maximum limit for heavy metal contamination is based on Indonesian National Standard No. 7387 of 2009, for the heavy metal mercury in crabs and other crustaceans is 1.0 ppm or 1000 ppb. This shows that the samples of yuyu crab (*Parathelphusa convexa*) in the Janja River in this research had mercury concentrations below the maximum limit.

This is something to be wary of because the mercury concentration in the crab's body can continue to increase. This increase can occur due to the bioaccumulation process, where the mercury entering the crab's body will continue to increase from the surrounding environment, such as through the food pyramid system, where the concentration of methyl mercury ions that accumulate in the bodies of aquatic biota tends to continue to increase (Wahyudi et al., 2021). In addition, mercury concentrations in biota can increase because mercury in sediment can be broken down by bacteria that live in the sediment. This process can then cause bioaccumulation of mercury in biota either through the food pyramid or directly (Irsan et al., 2020).

Mercury generally enters rivers in the form of elemental Hg (Hg^0). This mercury will accumulate in sediments or settle at the bottom of the river and, with bacterial activity, will change into organic mercury, namely methyl mercury (CH_3Hg). Under certain environmental conditions, mercury (Hg°) can change into ethyl and methyl mercury compounds. Mercury is very dangerous for the environment in the form of methyl and ethyl mercury. These compounds hurt the environment, ultimately negatively impacting human health (Sumarjono, 2020). When mercury enters the water, it will bind to chlorine, forming an Hg-Cl. Mercury easily enters plankton and spreads to other aquatic organisms in this form. Humans can accumulate mercury through the consumption of contaminated foods, such as fish, crabs, and shellfish (Masruddin & Mulasari, 2021)

Consuming contaminated crabs can expose humans to mercury. Consuming these crabs will cause bioaccumulation or buildup of exposure to methyl mercury, which accumulates in the human body and is very dangerous for health.

The information obtained from this research can support sustainable and appropriate environmental management. This data can help design environmental protection policies, identify areas that require remediation, and monitor the effectiveness of management actions implemented to reduce mercury exposure in gold mining in Malomba Village.

Conclusions

Based on the results of research that has been carried out, the yuyu crab (*Parathelphusa convexa*) that lives in the Janja River has accumulated mercury metal with the highest concentration obtained at the upstream location with an average of 5.24 ppb, then the lowest concentration of mercury is found in the midstream is 1.53 ppb, and downstream mercury levels were 4.53 ppb. So based on these results, it shows that the mercury level contamination in the yuyu crab (*Parathelphusa convexa*) samples is below the quality standard threshold set by the Indonesian National Standard No. 7387 of 2009, for the heavy metal mercury in crabs and other crustaceans is 1.0 ppm or 1000 ppb.

Acknowledgment

The authors would like to thank all those who have contributed so that this research can be done well.

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