Original Article

Heavy metal contents in reef fishes caught in the waters of Lurang Village, North Wetar, Southwest Maluku, Indonesia

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Abstract

Heavy metal is one of the contaminant materials which hard to degrade and has high toxicity; entering the water with a particular concentration can cause pollution.The objective of this study was to analyze the heavy metal contents of mercury (Hg), Copper (Cu), Cadmium (Cd), Lead (Pb), and Zinc (Zn) in several types of reef fishes caught in the waters of Lurang village, North Wetar Subdistrict, Southwest Maluku Regency, Indonesia and to analyze the potential risks evaluation based on heavy metal contents in caught consumption fish. This study result revealed that the heavy metal contents of Hg, Cd, Cu, Pb, and Zn in the caught reef fish tissue based on the reference of several fish as processed fishery products was included in the category still below the allowable metal contamination limit. Meanwhile, for the reference of PTWI (Provisional tolerated weekly intake), the calculation results showed that the fish in the coastal waters of Lurang and Uhak Villages achieved the HI value < 1. This means that the population that consumed fish containing heavy metals of Cu, Pb, and Zn was not at risk of experiencing non-carcinogenic effects. According to the calculations, there were no heavy metal concentrations that exceeded the acceptable intake for fish samples. The acceptable daily intake is highly dependent on the consumer's body weight and the amount of fish intake per day. The daily intake may vary among consumers.

Key words: reef fish, heavy metals, coral reefs, safety of consumption.

1. Introduction

Heavy metal is a pollutant that is difficult to degrade, has a high toxicity, if it enters the waters with a certain concentration, it can cause pollution. Heavy metals are metal elements that have corrosive properties and are very dangerous (Tchounwou et al., 2012). If they are in sea water or on land (soil or sediment) in large quantities, then they will enter the body of living organisms who live in that habitat. All aquatic organisms have the ability to absorb and accumulate heavy metals, both essential and non-essential (Rajeshkumar & Li, 2018;Chan et al., 2021). Although metals are in similar environments, they have different bioavailability characteristics and responses to the absorption of these heavy metals (Balali-Mood et al., 2021).

The nature of heavy metals that are difficult to degrade tends to accumulate into the body tissues of organisms (Briffa et al., 2020). Fish body tissues that are often found to accumulate heavy metals are meat, liver, and gills (Pertiwi et al., 2021). Nerve disorders, paralysis, and congenital defects in infants are examples of diseases that can be caused by heavy metal contamination (Jaishankar et al., 2014). Toxic heavy metals have a negative effect on the lives of living organisms, which can cause various diseases or disorders of the immune, respiratory, excretory, reproductive, and growth systems (Maresca et al., 2022). Based on the level of toxicity, the most toxic heavy metals are Pb, Cu and Zn, respectively (Jeong, 2022)

The working mechanism of heavy metals that enter the waters and the sediment will be greatly influenced by the dynamics of the currents that help the distribution of these metals (Tchounwou et al., 2012). Metals that are in the water or sediment which is the habitat of aquatic organisms will then be distributed through the food chain (Pandiyan et al., 2021;Razak et al., 2021). This ecosystem is inhabited by various organisms associated with it including reef fish (Roth et al., 2021; Helder et al., 2022). Reef fish communities use coral reefs as a place to find food, spawn, and shelter (Helder et al., 2022;Kar et al., 2022). Most of fish that live in coral reef areas are classified into ornamental fish and consumption fish (Bradley et al., 2020). The types of reef fish that have important economic value are from the families Serranidae (grouper) (Efendi et al., 2020), Lutjanidae (menggeru), Lethrinidae (lencam), and Holocentridae (swanggi). The objective of this study was to analyze the contents of heavy metals in several types of reef fish caught in the waters of Lurang village, North Wetar Subdistrict, Southwest Maluku Regency, Indonesia and to analyze the potential risks evaluation based on the heavy metal contents in caught consumption fish.

2. METHODOLOGY

2.1. Sampling procedure and preparation

This study was conducted between November 2021 to February 2022 at the waters neighboring area of PT Batutua Tembaga Raya and PT Batutua Karisma Permai, a copper mining company in Southwest Maluku District (Figure 1). The heavy metals content was analyzed at the Productivity and Environmental Laboratory of Bogor Agricultural University. There were seven sampling stations chosen for this analysis. For heavy metal content analysis, coral reef fish was obtained from local fishermen fishing in the coastal area near the mining field. The fish was caught by a traditional speargun. The fish sample was then transferred into the cool box filled with dried ice to maintain the freshness of the fish. The small fish was directly frozen while the large fish was separated then 200 g of tissue per individual was taken with a sterilized dissecting set for heavy metals content analysis. This amount of tissue was taken in anticipation of weight loss arising from freezing and incineration. The sample handling procedure before the analysis was conducted based on APHA standard method 23rd ed. (2017).

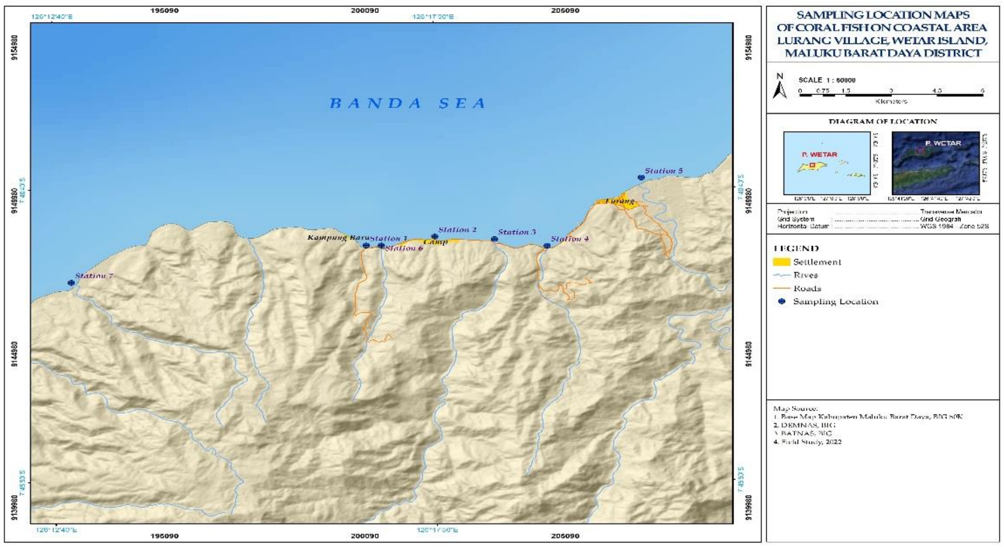

Figure 1. Sampling location

2.2. Data Analysis

2.2.1. The concentration of heavy metals

Standard preparation of each type of metal for analysis with standard curves as a reference determines the range of concentrations read by the results of the AAS analysis (APHA standard methods 23rd ed., (2017)).Preparation of laboratory analysis methods for each metal with analytical tools, methods used and references referred to as stated in Table 1.

Table 1. Preparation and measurement heavy metals and reference methods

No.	Metals	Preparation Method	Measurement	Reference Methods
1	Hg	Acid digestion + $NaBH_4$ reduction	Hydride Cold Vapor	APHA, 23rd Edition, 2540G, 3030-E, 3112-B, 2017
2	Cu	Acid digestion	FAAS (Air-A cetylene	APHA, 23rd Edition, 2540G, 3030-E, 3111-B, 2017
3	Cd	Acid digestion	FAAS (Air-A cetylene	APHA, 23rd Edition, 2540G, 3030-E, 3111-B, 2017
4	Pb	Acid digestion	FAAS (Air-A cetylene	APHA, 23rd Edition, 2540G, 3030-E, 3111-B, 2017
5	Zn	Acid digestion	FAAS (Air-A cetylene	APHA, 23rd Edition, 2540G, 3030-E, 3111-B, 2017

Explanation

FAAS : Furnace Atomic Absorption Spectroscopy

APHA : American Public Health Assotiation

2.2.2. Standard criterions and risk evaluation

The heavy metal analysis result was then used to determine the safety or unsafety concentration level of coral reef fish consumed by the local community. The standard concentration criterion for mercury (Hg) was 0.5 mg kg-1 following the National Food and Drug Supervisory Board (Regulation of The Drug and Food Control Agency Number 9 of 2022), while for fish and shellfish following Indonesian National Standard 7387:2009. whereas for food safety of fish and mollucs used SNI 7387:2009. The mercury (Hg), cadmium (Cd), lead (Pb), chromium (Cr), and zinc (Zn) quality standard for fish following FAO (1983). The result from the AAS analysis will then be used to evaluate the risk potency of fish consumed with acceptable concentration (min-max) both per day and per month (Table 2).

Table 2. Standards of use for the potential risk evaluation in the consumed fish

Heavy metal	Unit	DL	Standards	Safety Standard for Consumption
Cd	mg kg^{-1}	0.05	1.0 [a] / 0.5 [c(i)]	PTMI 0.025 mg kg^{-1}bw [d]
Cu	mg kg^{-1}	1.20	20.0 [b]	PMTDI 0.05-0.5 mg kg^{-1}bw [d]
Pb	mg kg^{-1}	0.23	0.3 [a] / 0.4 [a(i)] / 0.5 [c] / 0.3 [d]	NA
Hg	mg kg^{-1}	0.004	0.5 [a] / 1.0 [a(i)] / 0.5-1 [(iiii)] / 0.5 [d] / 1 [d(i)]	PTWI 0.0016 mg kg^{-1} bw [d]
Zn	mg kg^{-1}	0.67	100.0 [b]	PMTDI 0.3-1 mg kg^{-1}bw [d]

Remarks: NA (Not available) , BMDL (Benchmark Dose Lower Limit), TDI (Tolerable Daily Intake), PTMI (Provisional Tolarable Monthly Intake), PMTDI (Provisional Maximum Tolarable Daily Intake), PTWI (Provisional Tolarable weekly Intake), BW (Body Weight)

2.2.3. Monitoring of health risk

Health risk analysis is needed to identify how vast a person's health risks consuming fish accumulated with heavy metals to assess the risk potency of non-carcinogenic and carcinogenic occurs. The calculation comprises a) estimated daily intake (EDI); b) non-carcinogenic health risk calculated based on hazard quotient (HQ) and hazard index (HI); c) carcinogenic health risk calculated following lifetime cancer risk (LCR).

a. Hazard Quotient (HQ) and Hazard Index (HI)

The hazard quotient (HQ) calculates the health risk resulting from consuming fish contaminated by one heavy metal. In comparison, hazard index (HI) is the calculation of full risk results from more than one heavy metal. Fish consumption that has not given health risk when the ratio is < 1, and if the ratio is ≥ 1, the potential health risk might occur.

$$HQ = \frac{C_{heavy\ metal} W_{fish\ meat}}{RfD \times B_0}$$

$$HI = \text{Total number of } HQ$$

Where;

- Cheavy metal = heavy metal concentration
- Wfish meat (kg) = daily fish consumption (adult 160 gr; children 108 gr)
- Bo (kg) = individual weight measured (adult: 60 kg; children: 12 kg)
- RfD (mg kg-1) = heavy metal dosage reference

b. Lifetime Cancer Risk (LCR)

Lifetime cancer risk (LCR) calculates the lifetime health risk probability of an individual exposed to carcinogenic heavy metals. According to the US EPA level of cancer risk limit (LCR) range from 10-6 to 10-4.

CR = CDI x CSForal

` $$CDI = \frac{C_{heavy\ metal} \times W_{flesh} \times EF \times ED}{B_0 \times AT}$$

Where:

- CDI is a chronic daily intake or Pb daily intake
- CSForal heavy metal (Pb) = 0,0085 mg kg-1 /day
- Cheavy metal (mg kg-1) is the concentration of heavy metal in fish and shellfish flesh. In this study is a lead (Pb)
- Wflesh (kg) daily fish consumption (adult 160 gr; children 108 gr)
- Exposure Frequency (EF) = 365 days
- ED (exposure duration) is time exposure to heavy metal through oral; 30 years for adult and 6 years for children
- Bo (kg) is the body weight of the calculated population (adults: 60 kg; children: 12 kg).

- AT is the average time for carcinogenic effect i.e. 25,550 days

3. Results and Discussion.

3.1. The Mercury (Hg) Concentration in Fish Meat

The time when the water body or sediment is exposed to metals will determine the amount of heavy metal concentration absorbed by aquatic organisms living in contaminated environments. The analysis of heavy metal mercury in fish tissue (fish meat) showed average concentrations that had exceeded the detection limit (DL=0.004 mg kg-1), in station 1-7. The avergae concentration of mercury in fish tissue ranged from 0.01-0.02 mg kg-1 (Table 3). However, based on SNI 7387:2009, the permissible limit for mercury (Hg) contamination in fish fishery food and its processed products is 0.5 mg kg-1 ; while for predatory fish, it is 1.0 mg kg-1. Therefore, the mercury concentration in the analyzed fish samples was still far below the permissible level of mercury contamination, so it does not endanger the surrounding community who use the fish.

3.2. Heavy Metal Cadmium (Cd) Concentration in Fish Meat

The analysis of cadmium (Cd) in fish tissue (meat) showed a concentration that had exceeded the detection limit (DL) of 0.05 mg kg-1 (EPA, 2017). The concentration of Cd that passed through the DL was in station 1-7. The range of Cd metal average concentrations obtained was 0.02-0.15 mg kg-1 (Table 3). PTWI Cd toxicity is 0.007 mg kg-1. However, based on SNI 7387:2009 for Food, the maximum limit for Cd metal contamination for the category of fish food and its processed products is 0.1 mg kg-1, and some individual fish caught have passed the metal contamination limit for food. However, the PTWI food product category approved by Indonesia and several other countries for predatory fish is 0.5 mg kg-1. Heavy metal Cd can also come from human activities, such as market waste and household waste, marine transportation activities, and ship repair activities (Yan et al., 2018).

3.3. Heavy Metal Copper (Cu) Concentration in Fish Meat

The results of this study found that from approximately 32 individual fish analyzed, the average had exceeded the DL (1.20 mg kg-1), ranging from 1.95-4.4 mg kg-1. The lowest concentration was found in station 2 while the highest concentration was found in station 6 at 4.4 mg kg-1 (Table 3). Reference dose of heavy metals Copper in Fishery Food Products (US-EPA, 2004) maximum limit of consumption.

Heavy metals Cu and Pb naturally exist in waters with very low levels, both from the weathering process of soil, rock and from volcanic activity (Tchounwou et al., 2012). The presence of Cu in waters can also come from industrial areas around these waters. Heavy metals enter the body tissues of marine biota through several ways, i.e., the respiratory tract (gills), digestive tract (intestines, liver, kidneys), and penetration through the skin (Mazon et al., 2002;Rajeshkumar and Li, 2018). Biota that have been exposed to heavy metal contamination, if eaten by humans, will be very dangerous for human health.

Dawood (2022) mentioned that Cu is a micro element that is needed by both terrestrial and aquatic organisms, but in small amounts. Several researchers found that the Cu content in sediments tends to be high, and this is due to the nature of heavy metals in the water column that settle for a certain period of time and accumulate at the bottom of sedimentary waters (Algül & Beyhan, 2020). Metals will easily settle because the metal specific gravity is higher than the water density (Zhang et al., 2014).

3.4. Heavy Metal Lead (Pb) Concentration in Fish Meat

The results of the analysis of 32 individual fish showed average concentrations of Pb metal contamination ranging from 0.23-0.36 mg kg-1 (Table 3). Mean while, the maximum limit for Pb metal contamination for fish food and its processed categories is 0.3 mg kg-1, predatory fish is 0.4 mg kg-1, so that the limit for the category of fishery food has exceeded the maximum limit. The lowest Pb concentration in fish tissue was found at station 2, excepted 1,4 and 5, while the highest Pb concentration was found in station 4 at 0.36 mg kg-1, However, for PTWI fishery products recognized by Indonesia, the limit is 2.0 mg kg-1. Therefore, in terms of PTWI, the caught fish can still be consumed by the community.

Heavy metal lead is one of the most important pollutants that affect the waters and the biota. Heavy metal contamination is thought to trigger structural and functional damage to various fish organs. One organ that is sensitive to pollution is the gills (Winter et al., 2012;McRae et al., 2018). The metal enters the fish's body through the gill membranes, the food chain, the skin and mucous layer which will be transported by blood, and then accumulates in the heart and kidneys (Mahboob et al., 2016). The level of heavy metal accumulation in fish body tissues from large to small was found in the liver, kidneys, gills, and meat. The types of fish that are generally susceptible to heavy metal exposure are in habitats that are close to pollutant sources and have low mobility (Maurya et al., 2019).

3.5. Heavy Metal Zinc (Zn) Concentration in Fish Meat

The analysis showed that the lowest concentration of Zn in the liver tissue was found in station 4 at 38.2 mg kg-1, while the highest concentration was obtained in station 1 at 88.89 mg kg-1 (Table 3). Based on the Decree of the Director General of Indonesian Food and Drug Authority No.03725 of 1989, the maximum limit of Zn metal contamination is 100 mg kg-1. Therefore, all fish samples were still below the maximum Zn metal limit.

Zinc contamination comes from natural sources in the form of mineral rock erosion residues along river flows and zinc particles carried through the air (Hussain et al., 2022). Fish can show reactions to physical changes in water, as well as to pollutant agents dissolved within certain concentration limits (Yap & Al-Mutairi, 2022). Fish will adapt to the presence of various types of metals and changes in metal concentrations in the water (Rajkowska & Protasowicki, 2013). The degree of protection against pollution varies greatly and depends on the species, so that in a contaminated state, the ecological balance will decline and only organisms that have a high tolerance can survive.

Table 3. The average heavy metal concentration in fish meat

No.	Heavy metal	Unit	Detection Limit	Station1	Station 2	Station 3	Station 4	Station 5	Station 6	Station 7
1.	Hg	mg kg^{-1}	0.004	0.01±0.004	0.01±0.002	0.02±0.01	0.01±0.01	0.02±0.004	0.01±0.004	0.02±0.01
2.	Cd	mg kg^{-1}	0.05	0.11±0.10	0.08±0.03	0.13±0.02	0.05±0.02	0.15±0.08	0.02±0.01	0.05±0.04
3.	Cu	mg kg^{-1}	1.2	2.05±0.43	1.95±0.57	2.30±0.33	2.18±0.55	2.4±0.52	4.4±2.21	2.3±0.60
4.	Pb	mg kg^{-1}	0.23	0.35±0.07	0.23±0.10	0.31±0.13	0.36±0.11	0.32±0.13	0.30±0.14	0.32±0.08
5.	Zn	mg kg^{-1}	0.67	88.89±40.7	66.4±18.9	56.7±35.6	38.2±2.93	66.4±44.7	54.2±23.8	53.1±14.4

3.6. Safety Standard for Consumption

Regarding safety standards for consumption, acceptable intake standards from several international regulations can be applied. In principle, an acceptable daily intake is the estimated amount of a food additive, expressed on a body weight basis, that can be consumed daily for a lifetime without significant health risk (Environmental Health Criteria No. 70, JECFA) (WHO, 1987). The calculation of health risk was performed with the assumptions: The average consumer weight is 55 kg and average daily fish consumption is 86 g.

3.7 Health Risk Monitoring

The results of the HQ calculation of heavy metal Cu in children showed the number of 0.020-0.15; while in adults, it showed the number of 0.006-0.015, with the highest result being fish meat at station 6. The HQ calculation of heavy metal Pb in children shows the number of 0.018-0.020; while in adults, it showed the number of 0.0053-0.0058, with the highest value at station 4. Furthermore, the results of the HQ calculation of heavy metal Zn in children showed the number of 1.58-1.92; while in adults, it showed the number of 0.47-0.57. The calculation of health risks obtained the result of < 1, so it is concluded that the exposed population was not at risk of experiencing carcinogenic effects. However, the heavy metal content of Zn for children was at risk of experiencing carcinogenic effects.

The results of the HI calculation for heavy metals in children showed the number of 0.02-1.98; while in adults, it showed the number 0.006-0.58, with the highest result at station 1. The calculation results showed that the fish in the coastal waters of Lurang and Uhak Villages achieved the HI value < 1. This means that the population of people who consumed fish that contain heavy metals Cu, Pb, and Zn were not at risk of experiencing non-carcinogenic effects as a result of consuming marine fish. However, the condition of children was very at risk of experiencing non-carcinogenic effects. In addition, for the river habitats, the results of the heavy metal content of Cu and Pb in children showed a range of 0.094-0.116; while for adults, it ranged from 0.028-0.034.

The calculation results generated the LCR value in the adult category for fish meat at the four stations in order of 5.4x10-5, 5.4 x 10 -5, 5.5 x 10 -5, and 5.1x10-5, respectively. Meanwhile, the LCR results in the children category for fish meat were 3.6x10-5, 3.6x10-5, 3.7x10-5, and 3.4x10-5, respectively. The calculations in the category of children and adults for fish meat showed the LCR value that does not exceed 10-4. This value does not exceed the US-EPA acceptable risk level for carcinogenic effects, so that the population of children and adults who consumed fish meat from Lurang and Uhak villages were not at risk of carcinogenic effects due to lead exposure.

4. Conclusion

The results of this study revealed that the heavy metal contents of Hg, Cd, Cu, Pb, and Zn in the caught reef fish tissue based on the reference of several fish as processed fishery products was included in the category still below the allowable metal contamination limit. Meanwhile, for reference PTWI (Provisional tolerate weekly intake), the heavy metal contents of Cu and Zn in some fish tissue samples had exceeded the tolerated intake. Human populations that use reef fish caught as fish for consumption have not shown a risk of carcinogenic effects due to exposure to metal contamination, and no heavy metal content exceeds the acceptable intake for fish samples.

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