

## Ecological risks assessment of fishery commodities from heavy metal in The East Java Province, Indonesia

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**Abstract:** Heavy metals are a crucial group of chemicals extensively used in materials to meet human needs, eventually leading to contamination of aquatic ecosystems and accumulation in organism's tissues. Heavy metals enter the aquatic ecosystem from various sources. Those metals that pollute aquatic waters are deposited in sediments, remain dissolved in water or accumulate in the food web of aquatic organisms. Benthic biota is believed to accumulate the highest levels of these metals, while other aquatic animals, such as fish, shrimp, and macroalgae, serve as significant sources of heavy metals intake through food and the environmental exposure. In a study conducted in and around the east Java, Indonesia, the concentration of heavy metals in commercially important fish species, shellfish, and macroalgae were evaluated using an inductive couple plasma-mass spectrometer (ICP-MS). This study is very important because fishery resources are not only vital for exports but also serve as a primary source of essential food for local communities. The results showed that heavy metal concentration (Pb, Cd, and Hg) in samples were 12.3 mg·dm<sup>-3</sup>, 0.171 mg·dm<sup>-3</sup>, and undetectable, respectively. This study showed that different metals were present in the samples at different levels, all of which fell within the maximum residual levels set by the EU and USFDA. The results of an analysis of food safety based on the Hazard Index showed values below 1 point, indicating that fishery products (fish, shrimp, and macroalgae) from the East Java Province are generally safe for human consumption.

**Keywords:** accumulation, diversity, fisheries, food safety, heavy metal, macroalgae, pollutant

### INTRODUCTION

The fisheries sector plays a crucial role in the Indonesia's economy, contributing to food security, livelihoods of coastal communities, and employment opportunities (Zulkarnain, Purwanti and Indrayani, 2014). The rapid development of the fisheries sector is marked by an increase in production in 1980–

2012, positioning Indonesia as the fourth largest fishery producer in the world. With a contribution of 4.6%, and a total value of 3,067,660 Mg, Indonesia is a prominent player in animal protein production derived from the fisheries sector (Yusuf and Trondsen, 2013). The supply of animal protein in white meat and fishery products continues to rise in Indonesia, especially in East Java. This growth is not limited to freshwater fisheries, which

produce species like catfish (*Clarias gariepinus*) and tilapia (*Oreochromis niloticus*), but also extends to brackish and marine fisheries, where essential products continue to be developed.

Among the five primary commodities of Indonesia's fishery exports (shrimp, tuna, seaweed, crab, and squid), white shrimp (*Litopenaeus vannamei*) generates the highest revenue per kilogram, which accounts for about USD 2,040.2 mln or 39% of the total export value. Meanwhile, tuna and octopus contributed 14 and 10%, respectively. Moreover, seaweed and other fishery commodities accounted for 5 and 25%, respectively (MFM, 2020). However, shrimp production has witnessed a decline in several parts of Southeast Asia, including Indonesia, in recent years due to deteriorating water and environmental quality, leading to increased disease outbreaks (Pauly and Zeller, 2019).

Various sources of pollutants contribute to the degradation of water quality, leading to the decline in shrimp production. The accumulation of heavy metals (Pb, Cd, and Hg) affects aquatic biota such as shrimp and can have adverse effects in humans who consume them. Modern industrialisation exacerbates metal exposure and concentrations in water and sediment bodies. Contamination of water and air by toxic metals is a global environmental concern, affecting millions worldwide (Balali-Mood *et al.*, 2021), especially children (Ahmed *et al.*, 2019). Heavy metals are toxic to living organisms, difficult to degrade, and can accumulate in the body of organisms to cause death in high concentrations (Prasetyo *et al.*, 2018). High exposure to heavy metals, especially mercury and lead, can cause severe health complications, such as chronic abdominal pain, bleeding, diarrhea, and kidney failure (Bernhoft, 2012; Jaishankar *et al.*, 2014).

Several organisms have been found to accumulate heavy metal contamination, including tilapia (Hidayah, Purwanto and Soeprbowati, 2014; Mahboob *et al.*, 2020; Ghannam, 2021), milkfish (*Chanos chanos*) (Martuti, 2012; Murthy *et al.*, 2016; Muslim *et al.*, 2022), catfish (Ersoy, 2011; Mark *et al.*, 2019), grouper (*Epinephelus sp.*) (Heba *et al.*, 2014; Palupi, Andayani and Fadjar, 2016) and shrimp (Afolayan, Moruf and Lawal-Are, 2020; Arisekar *et al.*, 2022). Many studies have highlighted that an increase in pollution from heavy metals in water bodies that serve as the primary water sources for various fishery activities. Several areas in East Java have been the focus of research on water pollution, including; Lamongan (Sugiyanto, Yona and Kasitowati, 2016; Awaliyah, Yona and Pratiwi, 2018), Pasuruan (Salsabila, 2021; Purnomo, 2022), and Banyuwangi (Setyaningrum *et al.*, 2018; Yona *et al.*, 2018). The findings from these studies reveal the presence of various heavy metals in almost all coastal areas of East Java. However, research on the impact of heavy metal accumulation on the food safety of fishery products remains limited.

Fishery production plays a crucial role in meeting the demand for white meat animal protein (Susanto *et al.*, 2020), which is believed to be healthier than red meat protein. Fisheries products are rich in essential nutrients, as amino acids and unsaturated fatty acids, making them valuable in overcoming stunting and malnutrition issues that affect many countries (Djunaidah, 2017; Rachim and Pratiwi, 2017). Several studies have shown that poor nutrition can hinder growth and lead to stunting (Rachim and Pratiwi, 2017; Beal *et al.*, 2018; Budiastutik and Nugraheni, 2018). Therefore, ensuring food safety is an important issue (Putri, 2018), especially with efforts to increase consump-

tion of fishery products (Vergis *et al.*, 2021). Thus, it is essential to monitor the accumulation of contaminants in freshwater (Milošković *et al.*, 2016) and marine fishery commodities (Bosch *et al.*, 2016). At the same time, this practice contributes to sustainable fisheries management and the overall health of the aquatic environment.

This monitoring is expected to provide accurate information about heavy metal contamination currently affecting public water areas, with ramifications felt by the fisheries sector (Junianto, Zazidah and Apriliani, 2017). The focus of this study in East Java is on detecting and monitoring the accumulation of heavy metals in key fishery products such as shrimp and fish. Additionally, we assess how pollution affects food safety using several indices, including the potential ecological risk index (*PERI*), estimated daily intake (*EDI*), target hazard quotient (*THQ*), and target cancer risk (*TR*). This report is essential for ensuring food safety in fishery products and forms part of routine monitoring aimed at addressing heavy metal contamination in aquatic biota.

## MATERIALS AND METHODS

The research sampling was carried out across several regencies in East Java, a province experiencing rapid economic development and boasting significant fisheries potential, particularly in shrimp and fish production. Situated along the coast, East Java holds a strategic position that facilitates the growth of the fisheries sector, making it a cornerstone of local economic activity.

Fish and shrimp sampling locations include: Malang Regency (8°04'34"S, 112°28'44"E), Ngawi (7°26'27"S, 111°28'44"E), Madiun (7°47'41"S, 111°31'11"E), Blitar (8°05'28"S, 112°11'31"E), Kediri (7°45'00"S, 112°11'24"E), Pasuruan (7°34'48"S, 112°45'16"E), Mojokerto (7°27'50"S, 112°25'17"E), Tuban (7°01'59"S, 112°09'49"E), Bangkalan (6°53'05"S, 113°01'38"E), Lamongan (6°52'21"S, 112°14'27"E), and Banyuwangi (8°12'39"S, 114°22'32"E) – Figure 1. These locations have been chosen based on their status as the highest producers of fisheries products comparing with the other region in the East Java Province. Samples include white species such as shrimp (*Litopenaeus vannamei*), grouper (*Epinephelus sp.*), catfish (*Clarias gariepinus*), tilapia (*Oreochromis niloticus*), milkfish (*Chanos chanos*), and macroalgae sea-grape (*Caulerpha racemosa*). Each sample from each region weighs more than 300 g (for fish samples). Shrimp samples were collected from multiple vendors located in different traditional markets in each region. Additionally, samples were taken from sampling sites involved in aquaculture, including those related to fish, seaweed, water, and sediments, across East Java.

Fisheries products were sampled directly from farming ponds and immediately placed in a cool box filled with ice cube crumbs. Subsequently, all samples were transported to the laboratory for further examination, focusing on heavy metal contamination and adhering to standardised sample handling procedures. The fish samples were captured using nets and weighed approximately 300 g each. In addition to fish and shrimp, water and sediment samples were also collected for contamination assessment, with three replications at each sampling site. Sediment samples were obtained from fish and shrimp farming ponds at eight in the morning using Ekman grab size L 152×152×230 mm. The collected sediment was then placed



Fig. 1. Distribution of sampling site in East Java; source: own elaboration

in a 10 dm<sup>3</sup> bucket, put into one litre plastic bag, properly labelled, and stored in a cool box for further analysis.

All samples have been sent to private laboratories for heavy metal analysis by Inductive Couple Plasma – Mass Spectrometer (ICP-MS Thermo Electron Corporation, X SERIES, East Lyme, CT, USA) (Gajek *et al.*, 2022). The instrument was utilised to detect lead (Pb), cadmium (Cd), and mercury (Hg) contained in the analyte of the sample. These three metals were selected due to their harmful characteristics. Mercury (Hg), lead (Pb), and cadmium (Cd) are among heavy metals known to cause significant detrimental impacts, even when present in trace amounts (Henriques *et al.*, 2017; Zamani-Ahmadmohmoodi *et al.*, 2020). ICP-MS is capable of quantifying metal analytes in water in units of µg·dm<sup>-3</sup> (Rice *et al.* (eds.), 2012).

Data analysis was carried out to compute various indices serving as benchmarks in determining the ecological risk. The contribution of metals originating from lithogenic or anthropogenic sources was determined using the enrichment factor index (EF) (Nowrouzi and Pourkhabbaz, 2014; Gargouri *et al.*, 2018). An EF index value exceeding 1.5 indicates that heavy metal contamination in an area is caused by anthropogenic activity (Zhang and Liu, 2002). The geoaccumulation index (I-geo) was also employed to determine and evaluate the pollution degree (Muller, 1969; Kabir *et al.*, 2011). In the data analysis, particular attention was given to the concentrations of Co and Pb (McLennan, 2001) as well as Cd (Chonokhuu *et al.*, 2019), considering the naturally varying concentrations of these metals (Mohiuddin *et al.*, 2010). To determine the status of contamination, the contamination factor index (CF) was evaluated, representing a comparison of metal concentrations in the sample with those in their environment (Satapathy and Panda, 2015). Additionally, the potential ecological risk index (PERI) was also calculated to assess the ecological risk by multiplying the toxic-response factor (TR) of the metal and the contamination factor (CF) (Kumar, Pandita and Setia, 2022; Akarsu, Sönmez and Sivri, 2023), as follows:

$$E_r^i = T_r^i \cdot C_f^i \tag{1}$$

where:  $E_r^i$  = coefficient of potential ecological risk metal  $i$ ,  $T_r^i$  = toxic coefficient of metal  $i$ , and  $C_f^i$  = accumulation

coefficient (contamination factor) of metal  $i$ ; the toxic-response factor (Hakanson, 1980); the TR values for three heavy metals are as follows: Pb = 5, Cd = 30, and Hg = 40 (Hakanson, 1980).

Ecological risk values are grouped into five classes. Class 1 (low potential ecological risk) if  $EI \leq 40$ , class 2 (moderate potential ecological risk) if  $40 < EI < 80$ , class 3 (considerable potential ecological risk) if  $80 < EI < 160$ , class 4 (high potential ecological risk) if  $160 < EI < 320$ , and class 5 (significantly high potential ecological risk) if  $EI \geq 320$  (Hakanson, 1980; Weissmannová and Pavlovský, 2017).

The quality of fishery products can significantly impact human health. In recognition of this, regulatory authorities established quality standards to safeguard public health, providing authoritative assessments of health risks associated with exposure to health hazards through consumption (Tab. 1). Under the jurisdiction of the Ministry of Fisheries and Marine Affairs, the Indonesian government has implemented Regulation 37/PERMEN-KP/2019 concerning residual monitoring in fish farming for consumption. This regulation ensures compliance with heavy metal concentration limits in various fishery products.

Table 1. Maximum residue level (MRL) of heavy metal in shrimp, fish, and seaweed (mg·kg<sup>-1</sup>)

Metal	Fishery product	MRL	Reference
Hg	fish and shrimp	0.5	Peraturan (2019)
	seaweed	0.01	Commission Regulation (2006)
Pb	fish	0.3	Peraturan (2019)
	shrimp	0.5	Peraturan (2019)
	seaweed	0.1	Commission Regulation (2006)
Cd	fish	0.05	Peraturan (2019)
	shrimp	0.5	Peraturan (2019)
	seaweed	0.05	Commission Regulation (2006)

Source: own elaboration based on Indonesian and European law.

Additionally, we also analysed data to ensure of food safety, including some parameters such as estimated daily intake (*EDI*), target hazard quotient (*THQ*), the hazard index (*HI*), and carcinogenic risk.

The estimated daily intake (*EDI*) was calculated using the following equation to quantify the health risk associated with heavy metals absorbed due to consumption of fish, shrimp, and seaweed:

$$EDI = \frac{EF \cdot ED \cdot IR \cdot C_m}{WAB \cdot TA} \quad (2)$$

where: *EF* = exposure frequency (365 days per year); *ED* = exposure duration (70 years, equivalent to the average lifespan) (Anandkumar *et al.*, 2018; Keshavarzi *et al.*, 2018; Liu, Liao and Shou, 2018); *IR* = ingestion rate of fish ( $\text{g}\cdot\text{d}^{-1}$ ); *C<sub>m</sub>* = concentration of metal in fishery commodities ( $\text{mg}\cdot\text{kg}^{-1}$  DW); *WAB* = average adult body weight (kg); *TA* = average lifetime – it is equal to 365 days per year multiplied by the number of exposure years ( $365 \text{ d}\cdot\text{y}^{-1}\cdot 70 = 25,550$ ).

The consumption rate was determined to be 42.45 kg per person per year (equivalent to  $116.3 \text{ g}\cdot\text{d}^{-1}$ ) according to the Statistics Indonesia database report on fish and shrimps consumption for East Java (BPS, 2017). Additionally, the *WAB* value in Asia (including Indonesia) was estimated to be 57.7 kg (Walpole *et al.*, 2012; Hidayati *et al.*, 2020).

In this study, we also calculated the target hazard quotient (*THQ*), serving as a basis for estimating non-carcinogenic health risks associated with consuming fishery products, especially for individuals who rely on seafood as a vital protein supply. The *THQ* value was calculated based on previous research findings (Soegianto *et al.*, 2020):

$$THQ = EDI/RfD \quad (3)$$

where: *RfD* = oral reference dose, it is a value that shows the risk of each heavy metal.

The *RfD* values of Pb, Cd and Hg used the results of previous studies, namely  $3.5 \mu\text{g}\cdot\text{kg}^{-1}\cdot\text{BW}^{-1}\cdot\text{d}^{-1}$  (Hang *et al.*, 2009),  $1.0$  and  $0.3 \mu\text{g}\cdot\text{kg}^{-1}\cdot\text{BW}^{-1}\cdot\text{d}^{-1}$  (Soegianto *et al.*, 2020). The hazard index (*HI*) was also calculated, indicating the cumulative non-carcinogenic risk. The *HI* is the sum of the *THQ* that expresses

non-carcinogenic risk from individual heavy metals (Kumar *et al.*, 2020).

Using a target cancer risk value, the carcinogenic risk was calculated as the incremental probability of an individual acquiring cancer over a lifetime exposure to a possible carcinogen (USEPA, 1989). The carcinogenic health hazards associated with the consumption of fisheries products were assessed using the target cancer risk (*TR*), which was calculated as follows (Alipour, Pourkhabbaz and Hassanpour, 2015):

$$TR = EDI \cdot CSFo \cdot 10^{-3} \quad (4)$$

where: *CSFo* = oral carcinogenic slope factor from the Integrated Risk Information System database.

The slope values for Cd and Pb were  $15 \text{ mg}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$  and  $0.0085 \text{ mg}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , respectively. According to the New York State Department of Health (NYSDOH), the risk threshold *TR* is as follows:  $TR < 10^{-6}$  indicates very low risk,  $10^{-6} \leq TR < 10^{-4}$  – less low risk,  $10^{-4} \leq TR < 10^{-3}$  – moderate risk,  $10^{-3} \leq TR < 10^{-1}$  – high risk, and  $TR \geq 10^{-1}$  – very high risk (Javed and Usmani, 2016).

## RESULTS AND DISCUSSION

Pollution levels and ecological risk were assessment based on the concentrations of Pb, Cd and Hg in water and sediment samples. Comprehensive testing was carried out exclusively on seawater samples for both sediment and water. In freshwater samples, analysis was limited to rearing media water samples, revealing no detection of Hg. Results indicate significantly elevated levels of Pb ( $12.3 \text{ mg}\cdot\text{kg}^{-1}$ ) and Cd ( $0.171 \text{ mg}\cdot\text{kg}^{-1}$ ) in sediment samples. Furthermore, Hg was not detected in water and sediment samples (Tab. 2).

Table 2 shows that Pb, Cd, and Hg were not detected in water samples, remaining below the water quality standard set by environmental protection and management regulations (Peraturan, 2021). According to the government regulation, metal concentrations in water bodies should not exceed  $0.002 \text{ mg}\cdot\text{dm}^{-3}$  (Hg),  $0.01 \text{ mg}\cdot\text{dm}^{-3}$  (Cd), and  $0.03 \text{ mg}\cdot\text{dm}^{-3}$  (Pb). However, this regulation does not explicitly address metal contamination in

**Table 2.** Heavy metal concentration in water and sediment matrix in East Java

Heavy metal	Heavy metal concentration ( $\text{mg}\cdot\text{kg}^{-1}$ )					
	catfish	tilapia	shrimp	milkfish	grouper	seaweed
<b>Water</b>						
Cd	ND	ND	ND	ND	ND	ND
Pb	ND	ND	ND	ND	ND	ND
Hg	ND	ND	ND	ND	ND	ND
<b>Sediment</b>						
Cd	ND	ND	$0.171 \pm 0.001$	ND	$0.171 \pm 0.001$	$0.171 \pm 0.001$
Pb	ND	ND	$12.30 \pm 0.002$	ND	$12.30 \pm 0.002$	$12.30 \pm 0.002$
Hg	ND	ND	ND	ND	ND	ND

Explanation: ND = not detected.

Source: own study.

sediments. Many studies have also reported the absence of metal elements in their research, despite no detection in water samples. Interestingly, we observed imbalanced concentrations of Cd ( $0.171 \pm 0.001 \text{ mg}\cdot\text{kg}^{-1}$ ) and Pb ( $12.30 \pm 0.002 \text{ mg}\cdot\text{kg}^{-1}$ ). Pb concentrations were notably higher compared to other metals. Heavy metals tend to accumulate in sediments, forming complex compounds by binding with organic substances, eventually settling down to the ocean floor (Marchand *et al.*, 2006).

The presence of heavy metals in water or sediments poses a significant threat to aquatic biota (Prayoga, Utomo and Effendi, 2022), aligning with previous study (Fatoki and Mathabatha, 2001) that identified sediment as a reservoir for metals capable of leaching into water through natural and anthropogenic processes. Studies by Permanawati *et al.* (2013) have indicated that heavy metals in sediments can degrade water quality and transfer harmful substances to aquatic organisms. Moreover, water contaminated with heavy metals poses risk to human health (Prayoga, Utomo and Effendi, 2022). The presence of heavy metals in water and sediments is a primary driver of metal biomagnification in fish, aquatic plants, and animals, thereby posing health hazards to consumers (Rajeshkumar *et al.*, 2018). Despite these concerns, our study suggests that freshwater aquaculture activities for catfish and tilapia, as well as marine aquaculture for shrimp, grouper, and seaweed commodities, remain viable in East Java due to the current safety of its waters.

Based on *PERI* calculations (Akarsu, Sönme and Sivri, 2023), it was shown that only the accumulation of Cd and Pb could be assessed due to the absence of detectable Hg in sediments and water samples. The *PERI* for Cd contamination in water indicates a relatively low index value compared to the threshold of heavy metal toxicity, which is 30. Similarly, the *PERI* value for Pb accumulation remains below the toxic threshold, which is 5 points (Fig. 2).

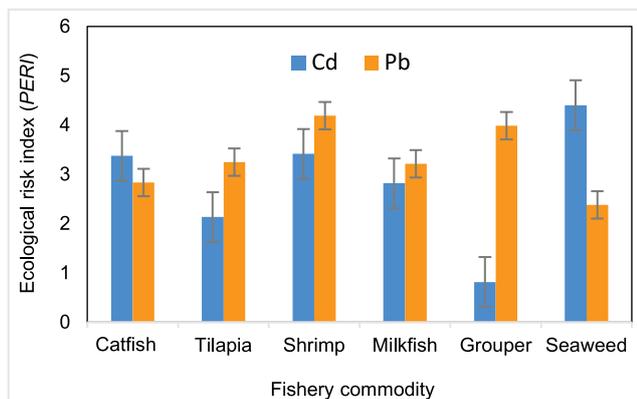


Fig. 2. Ecological risk index (*PERI*) from several fishery commodities environment in East Java; source: own study

The analysis of heavy metal content, including Pb, Cd, and Hg, in various fishery commodities in East Java, revealed that nearly all fishery commodities exhibited heavy metal accumulation. This trend was observed in freshwater fish, marine fish, shrimp, and seaweed (Fig. 3), albeit at varying concentrations. Particularly high levels of Hg were detected in shrimp products ( $0.19757 \text{ mg}\cdot\text{kg}^{-1}$ ), whereas grouper exhibited a higher accumulation of Pb ( $0.13622 \text{ mg}\cdot\text{kg}^{-1}$ ) compared to other types of fishery commodities. The distribution of metal concentrations is illustrated in Figure 4.

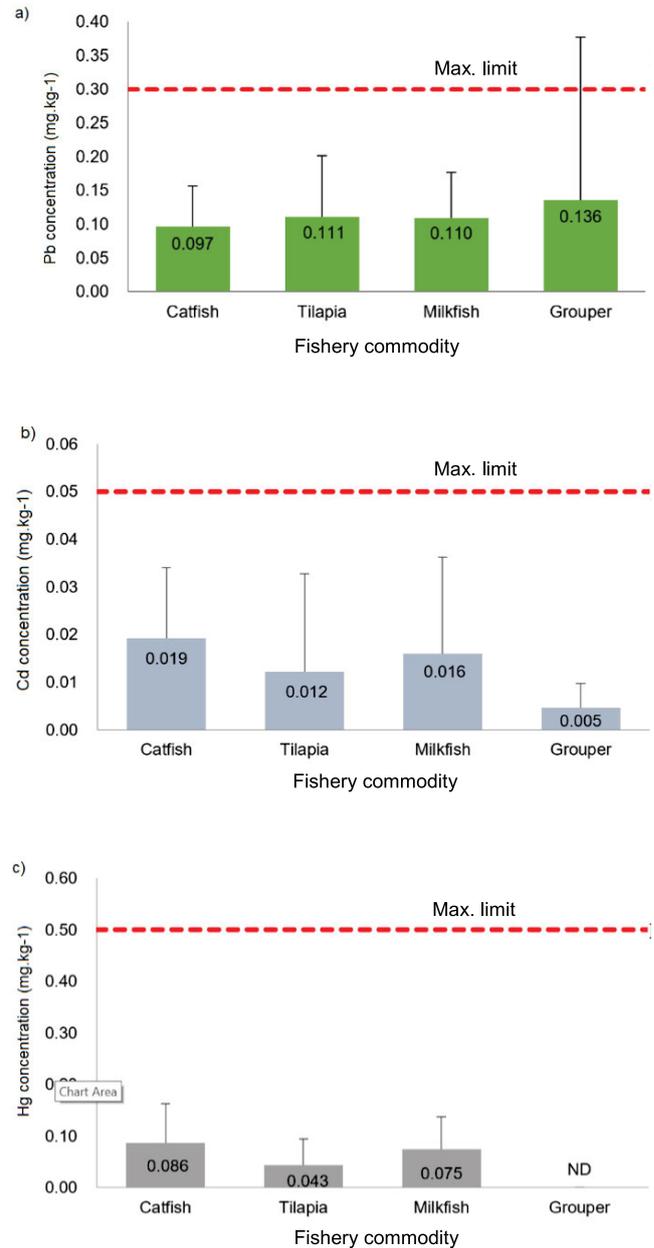


Fig. 3. Heavy metal concentration on fishery commodities in East Java: a) Pb, b) Cd, c) Hg; source: own study

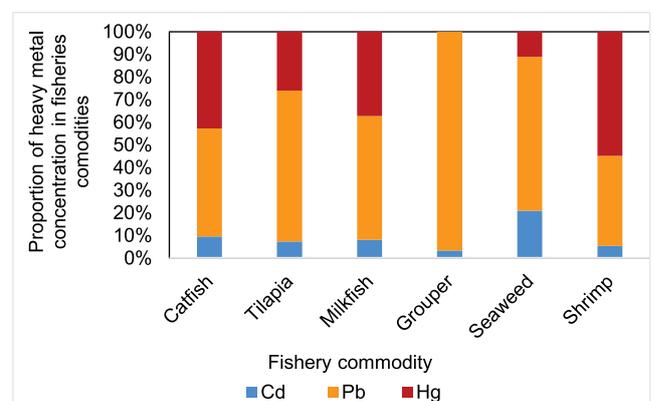


Fig. 4. Proportion of heavy metal concentration on fishery commodities in East Java; source: own study

The observed patterns of metal accumulation in fishery commodities followed a consistent decreasing order, with Pb exhibiting the highest concentration and Cd the lowest. Overall, the descending order of metal concentrations in this study was as follows:

- catfish – Pb > Hg > Cd,
- tilapia – Pb > Hg > Cd,
- milkfish – Pb > Hg > Cd,
- grouper – Pb > Cd,
- seaweed – Pb > Cd > Hg,
- shrimp – Hg > Pb > Cd.

Heavy metal concentrations in aquatic organisms have been extensively studied over the past few decades. The amount of heavy metals deposited in aquatic organisms varies depending on the tissue, type of metal, and the organism itself (Susanto, Hidayati and Syakti, 2014; Hidayati *et al.*, 2020; Hossain *et al.*, 2022; Zaghoul *et al.*, 2022). In this study, we found concentrations of Pb, Cd, and Hg (in mg·kg<sup>-1</sup>) ranging from (0.005 ± 0.005) to (0.198 ± 0.19). Figure 3 shows a comparison of metal accumulation levels based on the organism. Compared to fish and seaweed, shrimp may be considered as having the highest accumulation of metals. Shrimps are typical benthic organisms (Martínez-Guerrero and López-Pérez, 2018; Zhang Xiaojun *et al.*, 2019) and might also serve as good indicators reflecting contaminant levels in surface sediment (Dauvin, 2008).

Numerous studies have assessed heavy metal contamination in shrimps (Hidayati *et al.*, 2020; Groffen *et al.*, 2021; Arisekar *et al.*, 2022; Acharya, Muduli and Das, 2023). Although we did not find heavy metals in the water, they were present in sediment as well as in aquatic organisms. Previous studies have reported that heavy metals can enter shrimps through feed (Islam *et al.*, 2017). Additionally, sediment-associated metals may serve as a long-term source of contamination, as they can be released into the water (Luoma, 1983). While metals entering the aquatic environment may not directly affect species, they can accumulate in aquatic organisms through processes such as bioconcentration, bioaccumulation, and the food chain (Murtic *et al.*, 2020). In the aquaculture areas, heavy metals are also found in algacide, feed additives, fertilisers, and chemicals such as medications, disinfectants, and pH correctors used in farming (Lyle-Fritch, Romero-Beltrán and Páez-Osuna, 2006; Hidayati *et al.*, 2020).

Heavy metal accumulation order in tissues was nearly consistent across different organisms, as shown in Figure 3. Lead (Pb) exhibited the highest accumulation levels in all tissues of the targeted organisms, except for shrimp. Conversely, Cd accumulated at the lowest levels in all fishery commodities, except for seaweed. Several factors, including ecological demands, swimming patterns, metabolic activities, and living conditions, have been shown to influence the ability of fish to accumulate metals (Canli and Atli, 2003).

Fish, shrimp, and seaweed are essential components of a healthy human diet because of their high nutritional quality and status as significant sources of easily digestible protein-rich food. However, they also possess the capacity to accumulate heavy metals in their tissues (Hidayati *et al.*, 2020; Łuczyńska *et al.*, 2022). In this study, we found Pb, Cd, and Hg in these fishery commodities within the concentration range of (0.005 ± 0.005)–(0.198 ± 0.19) mg·kg<sup>-1</sup> (Fig. 3). A comparison of metal accumulation with standard quality guidelines is shown in Figure 3.

According to the previous report, the max. limits based on FAO-WHO (2022) vary for several heavy metals: Pb (2.0 mg·kg<sup>-1</sup>), Cd (1.0 mg·kg<sup>-1</sup>), and Hg (1.0 mg·kg<sup>-1</sup>). Additionally, Indonesian regulations (Peraturan Pemerintah, 2021) stipulate lower max. limits compared to FAO-WHO (2022) for Pb (0.3 mg·kg<sup>-1</sup>), Cd (0.05 mg·kg<sup>-1</sup>), and Hg (0.5 mg·kg<sup>-1</sup>).

Fish occupy the top position in the food chain, offering a balanced nutritional profile characterised by low-calorie, high-quality protein, vital nutrients, omega-3 fatty acids, and low levels of saturated fat. Shrimp is the most popular seafood commodity consumed globally, boasting a rich content of high-quality protein, fatty acids, vitamins, and minerals (Baboli, Velayatzadeh and Branch, 2013). However, contaminants have the potential to accumulate in the tissue of fish and shrimp, posing risk to human health upon consumption (Yu *et al.*, 2020). In our study, we determined the content of several heavy metals (Pb, Cd, Hg), indicating their accumulation in various fishery products such as fish, shrimp and seaweed. These fishery products are highly regarded for their superior quality and represent major sources of regional income. To mitigate the risk of heavy metal toxicity to human health, regulatory authorities have established optimal range values for every heavy metal.

Heavy metal concentrations (mg·kg<sup>-1</sup>) in fish, shrimp, and seaweed ranged from (0.005 ± 0.005) to (0.136 ± 0.24), (0.019 ± 0.034) to (1.198 ± 0.19), and (0.013 ± 0.008) to (0.081 ± 0.038) respectively, for all metals studied. Lead emerged as the predominant metal in all fishery commodities, although Pb concentrations in fish and shrimp remained below maximum residue limit in all samples. However, the presence of mercury (Hg) in fish, shrimp, as well as in seaweed poses health risks to the aquatic organisms and their consumers, including humans (Hidayati *et al.*, 2020). Fish with Hg levels exceeding 5 mg·kg<sup>-1</sup> may experience adverse effects such as loss of appetite, reduced coordination, and even mortality (Moallem *et al.*, 2010). In the present study, the maximum Hg level in fish samples was below 0.089 mg·kg<sup>-1</sup>, indicating that Hg does not pose a direct threat to the fish themselves. In addition, these levels fell below the guideline thresholds established by the Ministry of Marine Affairs and Fisheries Republic of Indonesia No. 37/PERMEN-KP/2019 as well as the Joint FAO-WHO Expert Committee on Food Additives (JECFA), suggesting that toxicity of Hg through fish consumption does not pose a significant risk to human health (FAO-WHO, 2022). Overall, our results indicate that heavy metal concentrations in the organism studied are well within the maximum levels set by the law, as shown in Figure 3. These concentrations comply with guidelines set by the WHO, FAO/WHO, EU, and Indonesian regulatory bodies, suggesting that fishery products are safe for consumption.

The estimated daily intake of metals (*EDI*) corresponds to the individual daily load of heavy metals: (0.04 ± 0.03)–(0.36 ± 0.56) µg·kg<sup>-1</sup>·d<sup>-1</sup> for Cd, (0.2 ± 0.12)–(1.27 ± 1.93) µg·kg<sup>-1</sup>·d<sup>-1</sup> for Pb, and (0.11 ± 0.05)–(0.21 ± 0.08) µg·kg<sup>-1</sup>·d<sup>-1</sup> for Hg (Tab. 3).

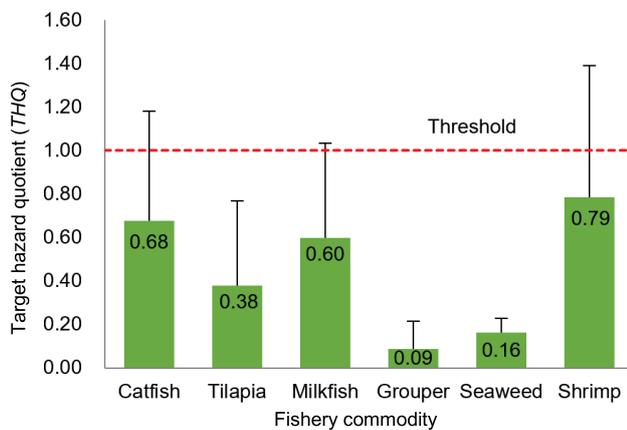
The current study focuses on the safety of popular fisheries products for consumers, assessed through the *THQ* and *HI* values. When the *THQ* < 1, consumption of fish, shrimp and seaweed is deemed to offer health benefits. Conversely, the *THQ* > 1 suggests a high probability of adverse risk to human health (Ahmed *et al.*, 2016). Similarly, the *HI* < 1 suggests no risks from non-carcinogenic effects, whereas the *HI* > 1 indicates potential

**Table 3.** Estimated daily intake of metals (*EDI*) per several fishery commodities

Fishery commodity	<i>EDI</i> ( $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ )		
	Cd	Pb	Hg
Catfish	0.04 $\pm$ 0.03	0.20 $\pm$ 0.12	0.17 $\pm$ 0.15
Tilapia	0.36 $\pm$ 0.56	1.27 $\pm$ 1.93	0.21 $\pm$ 0.08
Milkfish	0.25 $\pm$ 0.26	0.88 $\pm$ 0.88	0.15 $\pm$ 0.05
Grouper	0.36 $\pm$ 0.34	1.25 $\pm$ 1.17	0.16 $\pm$ 0.08
Seaweed	0.31 $\pm$ 0.16	1.07 $\pm$ 0.55	0.13 $\pm$ 0.06
Shrimp	0.28 $\pm$ 0.07	0.97 $\pm$ 0.25	0.11 $\pm$ 0.05

Source: own study.

adverse health effects, with the probability of effects increasing with higher *HI* values. Figure 5 shows the estimated *THQ* values for various fishery commodities.

**Fig. 5.** Estimation of the target hazard quotient (*THQ*) for various fishery commodities; source: own study

In the present study, the target cancer risk (*TR*) value is shown in the Table 4.

According to the New York State Department of Health (McKelvey *et al.*, 2007), the *TR* categories are described as follows:  $TR \leq 10^{-6}$  = low; from  $10^{-4}$  to  $10^{-3}$  = moderate risk; from  $10^{-3}$  to  $10^{-1}$  = high risk;  $\geq 10^{-1}$  = very high risk.

**Table 4.** Target cancer risk (*TR*) values for the consumption of fishery commodities

Fishery commodity	<i>TR</i> of metal	
	Cd ( $\cdot 10^4$ )	Pb ( $\cdot 10^6$ )
Catfish	5.81	1.6597
Tilapia	3.67	1.9026
Shrimp	5.93	2.3245
Milkfish	4.85	1.8816
Grouper	0.97	2.5723
Seaweed	7.58	1.3918

Source: own study.

A comparison was done with the tolerable intake levels to assess whether the metal levels found in the fishery commodity samples are safe for consumers. The *EDI* was highest for lead and lowest for cadmium throughout the year for adults (Tab. 3). The estimated daily intake of the metals through the consumption of each fishery commodity was found to be below the acceptable daily intake recommended by the joint FAO/WHO Expert Committee on Food Additive and ATSDR for all samples (FAO-WHO 2022).

The United States Environmental Protection Agency (USEPA) developed quantitative frameworks for assessing target hazard quotient (*THQ*), hazard index (*HI*) and target cancer risk (*TR*) of heavy metals, addressing both carcinogenic and non-carcinogenic threats to human health (Krishna *et al.*, 2014). The *HI* values derived from Pb, Cd, and Hg for all studied aquatic organisms were consistently below 1.0. Specifically, for a 70-kg adult consuming  $116.3 \text{ g}\cdot\text{d}^{-1}$  of the fisheries products, the *HI* values were as follows:  $0.67 \pm 0.5$  for catfish,  $0.38 \pm 0.39$  for tilapia,  $0.60 \pm 0.44$  for milkfish,  $0.09 \pm 0.13$  for grouper,  $0.16 \pm 0.07$  for seaweed, and  $0.79 \pm 0.61$  for shrimp. The hazard index reflects the cumulative non-carcinogenic risk, with the highest values observed for Pb, Cd, and Hg in shrimp for adults ( $0.79 \pm 0.61$ ). As shown in Figure 3, no *HI* value exceeded 1.0 through the consumption of fisheries product in East Java, indicating significant health risks associated with heavy metals exposure. However, when calculating the cumulative *HI* for all fisheries product across the three studied toxic metals, the resulting values exceeded 1.0, suggesting possible adverse health effects. Regarding carcinogenic risk, Pb and Cd were evaluated, and their sum expressed as the target cancer risk (*TR*) (Tab. 4). The range of moderate risk (from  $1.0 \cdot 10^{-6}$  to  $1.0 \cdot 10^{-4}$ ) was observed in the case of Cd for adults across almost all fishery commodities, while a low risk was determined for Pb.

## CONCLUSIONS

This study represents the first comprehensive examination of heavy metal content in fish, shrimp, and seaweed in East Java, shedding light on potential ecological and human health risks. The concentration of Pb in all fishery commodities ranged from 0.097 to 0.136  $\text{mg}\cdot\text{kg}^{-1}$ , with the highest levels detected in grouper. Cd levels varied between 0.005 and 0.019  $\text{mg}\cdot\text{kg}^{-1}$ , with catfish exhibiting the highest concentration. The highest accumulation of Hg was observed in catfish (0.086  $\text{mg}\cdot\text{kg}^{-1}$ ). Importantly, heavy metals concentrations in fish, shrimps, and seaweed fell within the environmental and food safety guidelines established by FAO and USEPA, indicating that they are safe for human consumption. Furthermore, concerning potential human risks, this study concludes that fish and shrimp consumption from East Java poses no significant threat to human health. However, it is vital for customers, particularly the younger generation, to be aware of the potential adverse effects of excessive fish consumption, which may lead to health hazards. Therefore, regular and comprehensive monitoring of water quality and coastal pollution is essential. Such monitoring efforts should be undertaken collaboratively, involving stakeholders from government agencies, academia, local communities, and industrial sector. This multi-stakeholder approach ensures a holistic understanding of the

environmental and health implications and facilitates effective management strategies to safeguard both ecosystems and human well-being.

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## CONFLICT OF INTERESTS

The authors declare no conflicts of interest.

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