Heavy Metals in the Water, Sediment, and Fish Harvested from the Krueng Sabee River Aceh Province, Indonesia

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ABSTRACT

The pollution of rivers from human activities such as housing, markets, workshops, transportation, land cultivation, and industry has become an issue. The increasing contribution of heavy metals to pollution in rivers has a direct effect on the metal bioaccumulation in fish. Therefore, this study aimed to examine the concentrations of Cd, Cu, and Pb in the waters, sediment, and fish harvested from the Krueng Sabee River, Aceh Province, Indonesia. Furthermore, the water, sediment, and fish samples were collected from six locations representing the upstream and downstream regions of the river. Cd, Cu, and Pb were analyzed using Atomic Absorption Spectrophotometry (AAS). The results showed that Cd, Cu, and Pb were not detected in the water, while the concentration of Cd in the sediments ranges between 0.0544 to 0.2683 mg kg^{-1}, Cu ranges between 4.4149 to 14.8160 mg kg^{-1}, and Pb ranges between 0.9186– 15.4954 mg kg^{-1}. Therefore, Cd, Cu, and Pb in the water and sediment were below the quality standard, but these heavy metals in the fish sample met the threshold limit. The highest Cd concentration was reported in *Tor soro* (5.5591 mg kg^{-1}), and the highest concentration of Cu was reported in *Mugil cephalus* (6.7021 mg kg^{-1}), while the higher Pb concentration was reported in *Cyclocheilichthys Apogon* (0.0279 mg kg^{-1}).

Keywords: water quality, sediment, gold mine, fish, heavy metals, *Krueng Sabee*.

INTRODUCTION

Currently, river pollution is becoming an interesting and popular issue [Lipy et al., 2021]. Pollutants are generally introduced into the environment as a result of human activities such as housing, tourism, agriculture, aquaculture, and industries [Lu et al., 2021]. Meanwhile, the addition of dissolved matter and the increase in the retention time has a direct effect on aquatic organisms by changes in their physical, chemical, and biological characteristics. [Seo 2008; Rubalingeswari et al., 2021]. One of the major issues in river pollution is the heavy metal contamination. Subsequently, out of all the heavy metals, cadmium (Cd), copper (Cu), and lead (Pb) occurred in the river most frequently [Alengebawy et al., 2021]. These metals are toxic to human and aquatic organisms [Rubalingeswari et al., 2021]. The high accumulation of heavy metals in fish inhibits the growth rate by hampering the activity of metabolic enzymes [Elwasify et al., 2021]. According to Edogbo et al., [2020], the consumption of fish contaminated by Cd has the potential to pose a risk

to human health [Edogbo et al., 2020]. The study showed that Pb reduced the growth rate of tilapia *Oreochromis mossambicus* [Yulaipi and Aunurohim, 2013], and this metal was mainly accumulated in the liver of the fish [Sarong et al., 2013].

The Krueng Sabee River is located in the Aceh Province, Indonesia. It is one of the large rivers, which plays an important role for the local community as a source of water for the agricultural, fishery, and domestic purposes. Furthermore, the river is also an important fishing area for local fishermen [Timorya et al., 2018]. The watershed of the Krueng Sabee River is made up of protected forests, production forests, agriculture, plantations, and human settlements. In the 1980s, the Krueng Sabee Forest was owned by the Aceh Wood Company. Currently, the status has changed in the oil palm plantations managed by the Boswa Company and most of the forests have been converted to oil palm plantations. In addition, since 2008, some upstream areas of the river have also been dredged for illegal gold mining.

The previous studies showed that this river was contaminated by mercury that exceeded the maximum threshold [Purnawan et al., 2017; Suhendrayatna et al., 2011; Wahidah et al., 2019]. However, potential contaminations by other heavy metals such as Pb, Cd, Cu were not assessed. The potential for the Cd, Cu, and Pb pollution results from plantations, agriculture, and fisheries activities along the watershed of this river. Therefore, this study aimed to examine the Pb, Cd, and Cu concentrations in the water, sediments, and the fish harvested from the Krueng Sabee River, Aceh Jaya District, Indonesia.

METHODS

Location and time

This study was conducted at three sampling locations along the Krueng Sabee River, Aceh Jaya Regency, Aceh Province (Figure 1) from 2018 to 2019. The sampling locations represent the condition of the river, upstream to downstream. A total of six sampling sites were determined purposively (two sites upstream, two sites downstream, and two sites at the middle stream). The characteristic of the sampling location is shown in Table 1.

Samples collection

The water samples were collected at three points using the bottled sampler, and the samples from three points were pooled in one plastic container. The samples were kept in an icebox, then 2 mL of HNO_3 was added to each sample for preservation. Furthermore, the analysis was carried out at the Laboratory of Chemistry Department, Faculty of Mathematics and Natural Sciences, Universitas Syiah Kuala. Meanwhile, the sediments were collected from the same sites using Ekman Grab and then placed into a plastic bag and kept in the icebox. In the laboratory, the sediment was dried in an oven at 105 °C for 24 hours. A total of 5 g dried sediment was mixed with 2 ml $HClO_4$ and 2 ml HCl, followed by deionized water to reach 100 ml with

Table 1. Research location, coordinate, characteristics for the sampling process from upstream to downstream of *Krueng Sabee* River, Aceh Jaya Regency

| Location	Coordinate	Characteristics
Panggong	04° 46' 32.8"N, 95° 47' 09.6" E	The area located in the upper reaches of the river is bordered by protected forests. There are few settlements
Buntha	04° 41' 50.5"N, 95° 43' 11.1" E	Areas bordering community agricultural land and residential areas
Ranto Panyang	04° 38' 20.4"N, 95° 40' 17.0" E	Agriculture plantation and settlement Areas located in residential areas
Blang Phong	04° 37' 17.0"N, 95° 39' 02.9" E	Agriculture plantation, paddy field, and settlement
Mon Mata	04° 36' 56.7"N, 95° 38' 43.2" E	Agriculture plantation, paddy field, and settlement. Mining mineral processing area. The area is located in the lower reaches of the river and is bordered by a swamp
Keude Kr. Sabee	04° 35' 58.5"N, 95° 38' 23.3" E	Trade areas, markets, government centers, and dense settlements. The estuary area is located in the lower reaches of the river and borders the sea
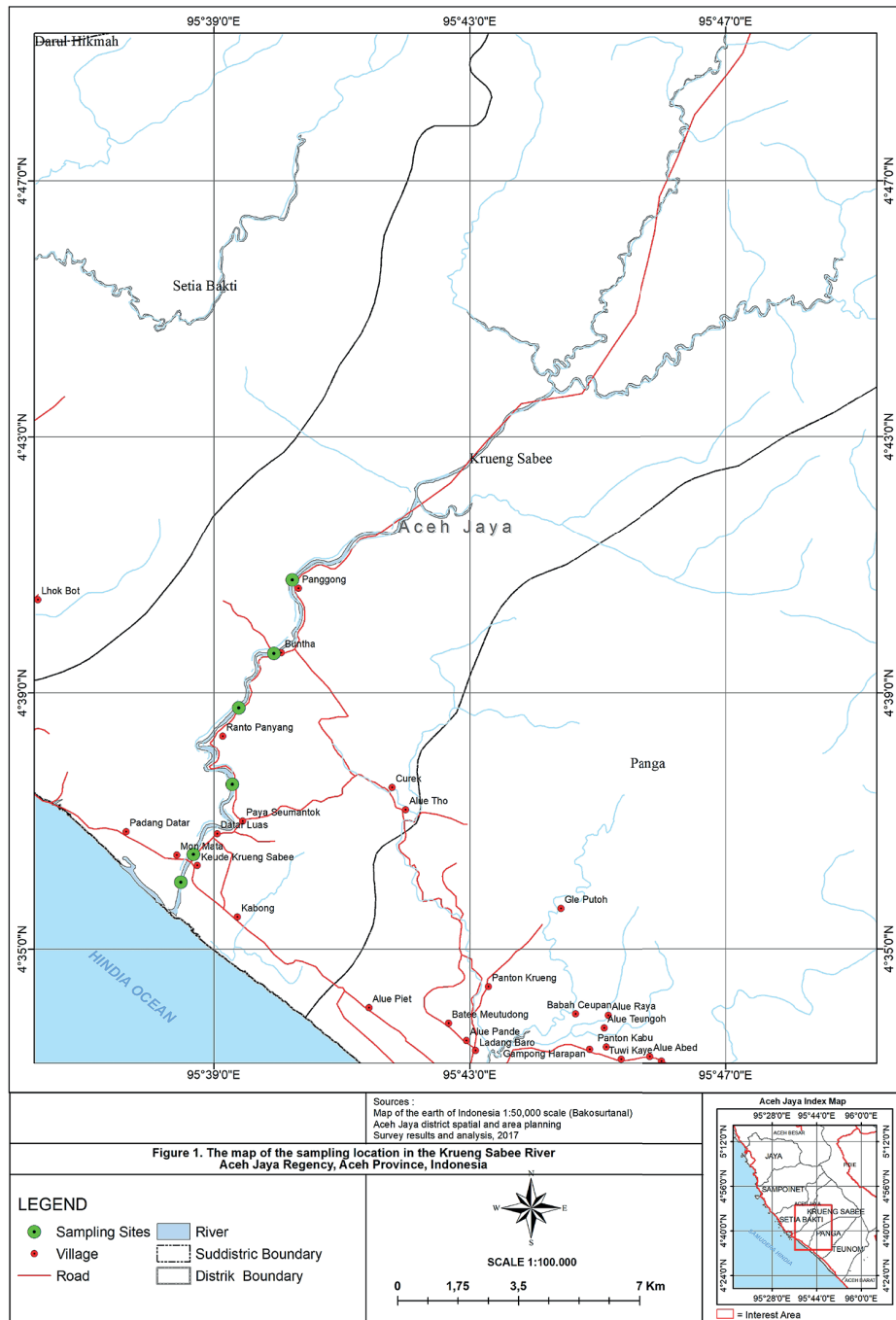
Figure 1. The map of the sampling location in the *Krueng Sabee* River,
Aceh Jaya Regency, Aceh Province, Indonesia

deionized water [Loring and Rantala, 1992]. The Cd, Cu, and Pb concentrations were examined using Atomic Absorption Spectrophotometry (AAS). Meanwhile, the fish samples were caught using casting nets and gillnets. The sampled fish were identified to the species level based on Kottelat et al. [1993] and Muchlisin and Siti-Azizah [2009], then preserved in an icebox (4 °C), and transported to the laboratory for further analysis.

Sample preparation

The sample of sediment and fish muscle was weighed approximately 500 g (wet) and washed with distilled water, then dried in a furnace at 105 °C for 24 h. The samples were cooled in a desiccator and crushed using a portal grinding machine [Khaled et al. 2014]. Furthermore, the sample powder was weighed for 5 mg. Then, 1.5 ml of $HClO_4$ and 3.5 ml of HNO_3 were mixed into the sample and kept for 24 h. The obtained dissolved

solid solution was then heated at 60–70 °C for 3 h to obtain a clear and colorless solution. The samples were analyzed using AAS with air-acetylene flame, and the metal concentrations were detected using a spectrophotometer [Topcuoğlu et al., 2003].

Data analysis

The data were presented in the tables and analyzed descriptively by comparing the national and international thresholds regulations, for example, the Indonesian Government Regulation Number 82 of 2001. The quality standards from the National Food and Drug Agency Republic of Indonesia [BPOM RI 1989 and 2017] regarding the maximum threshold of heavy metal contamination in foodstuffs, and the quality standard of the Canadian Freshwater Sediments Guidelines [Burton, 2002].

RESULTS

Metals in the water and sediment

The contents of heavy metal cadmium (Cd), copper (Cu), and lead (Pb) in the water of the Krueng Sabee River were very low. Therefore, the concentrations of Cd, Cu, and Pb in all sampling locations were under the limit of detection (LOD) of the AAS. The results showed that Cd in the sediment ranges from 0.0544 to 0.2683 mg kg⁻¹ with an average of 0.09 mgkg⁻¹. The highest Cd concentration was recorded at Keude Kreung Sabee (station 6). In turn, Cd was not detected in two locations; Buntha (Station 2), and Ranto Panyang (station 3). However, in general, the Cd concentration in all sampling locations was still under the maximum threshold (Table 2). Furthermore, Cu in the sediment

ranges from 4.4149 to 14.8160 mg kg⁻¹ with an average of 9.6002 mg kg⁻¹. The highest Cu was also observed at Keude Kreung Sabee. However, the levels of Cu in every sampling location were still under the maximum threshold (Table 2). In addition, the Pb concentrations in the sediment range from 0.9186–15.4954 mg kg⁻¹ with an averaged value of 3.09 mgkg⁻¹. The highest concentration of Pb was detected at Mon Mata (station 5). However, the concentration of Pb in the sediment at every sampling location did not exceed the maximum threshold.

Metals in fish

The concentration of Cd in the fish ranges from 0.1922 to 5.5591 mgkg⁻¹ with an average value of 1.79 mg kg⁻¹. The highest concentration of Cd was reported in *Tor soro* (5.56 mg kg⁻¹), followed by *Rasbora sumatrana* (1.9572 mgkg⁻¹), and the lowest Cd was reported in the *Neolissochillus thienemanni* (0.1922 mg kg⁻¹) (Table 3). Therefore, according to BPOM RI - 2017 regulations, the concentration of CD in these five species of fish caught in the Krueng Sabee River has exceeded the maximum threshold.

The concentration of Cu in fish muscles ranges from 2.34 to 6.70 mg kg⁻¹ with an average level of 4.36 mg kg⁻¹. While the highest concentration of Cu was reported in *M. cephalus* (6.7021 mg kg⁻¹) followed by *R. sumatrana* (5.7384 mg kg⁻¹) and the lowest was reported in *N. thienemanni* (2.3370 mg kg⁻¹) (Table 3), and therefore, the concentration of Cu in *M. cephalus* and *R. sumatrana* exceeded the threshold limit. Meanwhile, the concentration of Pb was not reported in almost all fish except in *C. apagon* with a concentration of 0.028 mg kg-1 which exceeded the maximum threshold limit (Tabel 3).

Table 2. Levels of Cd, Cu, and Pb in the sediment at six sampling locations of the Krueng Sabee River, Aceh Jaya, Indonesia

Metal	Station						Maximum Threshold[a]
	1	2	3	4	5	6	
Cd (mg kg⁻¹)	0.1104	ND	ND	0.0544	0.1096	0.2683	0.6
Cu (mg kg⁻¹)	10.3073	10.5852	6.9325	10.5443	4.4149	14.8166	37.5
Pb (mg kg⁻¹)	ND	0.9186	ND	1.1658	15.3954	1.0336	35

ST1: Panggong; ST2: Buntha; ST3: Ranto Panyang; ST4: Blang Phong; ST5: Mon Mata; ST6: Keude Krueng Sabee; ND: Not detected with the LOD = 0.000001 mg kg⁻¹;
[a]According to the Canadian Freshwater Sediments Guidelines [Burton, 2002];
*exceeding the maximum threshold

Table 3. The concentration of Cd, Cu, and Pb in five dominant species of fish in the Krueng Sabee river, Aceh Jaya, Indonesia

Metal	Fish species					
	Mugil cephalus	*Neolissochillus thienemanni*	*Rasbora sumatrana*	*Cyclocheilichthys apogon*	*Tor soro*	Maximum threshold[a]
Cd (mg kg^{-1})	0.8854*	0.1922*	1.9572*	0.3359*	5.5591*	0.05
Cu (mg kg^{-1})	6.7021*	2.3370	5.7384*	3.9576	3.0701	5.0
Pb (mg kg^{-1})	ND	ND	ND	0.0279*	ND	0.02

ND: not detected with the LOD = 0.000001 mg kg^{-1};
[a]According to the BPOM RI 2017 [BPOM, 2017];
*exceeding the maximum threshold

DISCUSSION

Cadmium, copper, and lead in the water of the Krueng Sabee River were not detected or were below the limit of detection (LOD) of the AAS analytical instrument. However, these heavy metals have been detected in sediments at various locations with low concentrations or below the threshold limit. In contrast, almost all fish species examined were contaminated with a high concentration of Cd and Cu, while Pb contaminated only one species of *C. Apogon*, and the concentration of which exceeded the threshold limit. A similar finding was also been reported by several researchers, in which heavy metal content was not detected in waters, but it was detected in sediments, for example; Wahyuni et al., [2013] studied the content of Pb and Cu in the waters of mining areas in the Batu Behole Village, Central Bangka Regency that most of the locations were not detected, but these metals were detected in the sediment at low concentration. Furthermore, [Priyanto et al., 2008] reported that Pb was not detected in water in the Cirata Reservoir, West Java, but Hg, Cd, and Cu were detected and some exceeded the quality standard. According to Wahyuni et al., [2013] the heavy metal content can be in low concentrations because most of the metal ions are absorbed by suspended solids and also planktons. In addition, the concentration of heavy metals in the Lotic ecosystem may be lower than that in the Lentic ecosystem, as reported in this study.

The study showed that the heavy metal contents in the sediment were higher downstream (Location 5 and 6), this may be due to the flow of water carrying heavy metal material from the watershed and settled downstream, where the current velocity is low. In addition, the downstream area is a densely populated area with housing and community activities such as markets and fishing ports, and workshops that have the potential to produce heavy metal pollutants.

Cd, Cu, and Pd can interact with organic matters in a soluble phase and then precipitate, which leads to high concentrations in the sediment [Rahman et al., 2019]. At low water velocity, sediments settle more easily, especially in clay and silt [Johnson et al. 2019]. In general, the concentration of heavy metals in sediments is below the threshold, but over a long time there will be accumulation and therefore the concentration will continue to increase in the sediment [Rahman et al., 2019]. The results showed that the concentration of heavy metals, especially Cd in Tor soro and *R. sumatrana* fish were higher than the other three species, this was believed to be related to the feeding habit of the two fish, both species are omnivorous [Muchlisin et al., 2015; Choy et al., 1996], and often feed at the bottom of the water [Johnson 1967]. In addition, *R. sumatrana* also contained high Cu, and it has exceeded the threshold limit. The high Cu content was also reported in *Mugil cephalus*, probably because the natural habitat of the fish is the area downstream of the river [Wairara and Elviana 2021; Mardani et al., 2018], which is still affected by the tides in this area, the metal content in the sediment is higher than in other areas. The concentration of Pb was only detected in *C. apogon,* which exceeded the threshold limit, this may also be related to the feeding habit and habitat of this fish, which is omnivorous and only reported in the downstream area of the river.

One of the causes of the heavy metal contamination in fish can be due to the direct contact between fish and water that contains metal compounds, resulting in the transfer of the metal substances from the waters into the fish's body through the body surface or gills [Olayinka-Olagunju et al., 2021]. This is also in concordance with the statement by Affandi and Ishak [2019] that heavy metals can accumulate in the body of fish through the respiration process, feeding

activity, (biomagnification), and through the skin (diffusion). Furthermore, metals are absorbed in fish flesh by the blood, which then binds to blood proteins and is distributed throughout the body tissues. The highest metal accumulation is usually reported in the liver and kidneys [Hazrat et al., 2020; Karadede and Ünlü, 2000].

Cadmium is a non-corrosive material and is therefore often used for coating the materials made of iron and steel [Mahmood et al., 2019]. Furthermore, it is also often used in the batteries industry, paints, color pigments, and helium-cadmium lasers [Hayat et al., 2019]. Copper naturally comes from forest fires, and volcanic ash, penetrating the water during the rainy season [Barceloux and Barceloux, 1999]. It is widely used in the agricultural industry, for example, fertilizers and pesticides [Bui et al., 2016]. In addition, Cu is also often used in the electrical industry and industrial machinery as an alloy of bronze and brass [Barceloux and Barceloux, 1999].

It was suspected that the main source of cadmium and lead at the study site is from the waste of used batteries, electronic equipment, and paint used by fishing boats. Oil spills or washing of diesel oil from fishing boats are also potential sources of pollutants. In addition, there are also several vehicle washing services located on the banks through which the sewage vehicle with oil and diesel can penetrate the river. It was also believed that the most likely source of the Cu pollutant in the research location is from agricultural areas adjacent to the river which intensive uses of chemical fertilizers and pesticides are carried out, where the contaminated water from agricultural areas penetrates the river during the rainy season, settles at the bottom and then contaminates fish that live in this area.

The results of the analysis carried out showed that to maintain the environmental biodiversity and fish preservation in the Krueng Sabee River, better supervision is needed. Therefore, the monitoring should be carried out in stages and periodically tested for metal accumulation in river fish, as well as public education about the dangers of metals and pesticides to health.

CONCLUSIONS

The metal content of Cd, Cu, and Pb in the water was not detected, but these metals were present in the sediment at low concentrations.

However, the concentration of Cd, Cu, and Pb in the fish muscle exceeded the threshold limit and is not safe for consumption.

Acknowledgements

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