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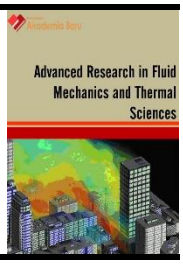

Investigation of Heavy Metals in River Water, Sediments, and Fish in Krueng Geumpang, Pidie Regency, Aceh Province

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Muhammad Nasir[1,2,3,*], Iqbar[1], Munira[4], Zainal Abidin Muchlisin[5], Saiful[6], Suhendrayatna[7], Erdiwansyah[8,3]

[1] Department of Biology, Syiah Kuala University, Banda Aceh 23111, Indonesia
[2] Graduate School of Mathematics and Applied Science, Syiah Kuala University, Banda Aceh 23111, Indonesia
[3] Research Center of Environmental and Natural Resources, Universitas Syiah Kuala, Banda Aceh 23111, Indonesia
[4] Department of Pharmacy, Health Polytechnic Aceh, Banda Aceh 23352, Indonesia
[5] Faculty of Marine and Fisheries, Syiah Kuala University, Banda Aceh 23111, Indonesia
[6] Department of Chemistry, Syiah Kuala University, Banda Aceh 23111, Indonesia
[7] Department of Chemical Engineering, Syiah Kuala University, Banda Aceh 23111, Indonesia
[8] Faculty of Engineering, Universitas Serambi Mekkah, Banda Aceh 23245, Indonesia

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ABSTRACT

The environmental problems throughout the world have become a concern, especially regarding environmental pollution. Environmental pollution can be caused by various industrial and daily human activities. The primary objective of this study is to analyse the level of contamination and accumulation of heavy metals such as Pb, Hg, Cu, and Fe distributed by heavy metals in water, sediments, and fish in Geumpang River. The data samples used such as water, sediment, and fish were collected from Geumpang River, Pidie Regency, Aceh Province. The study conducted several sample tests at the Banda Aceh Unsyiah Laboratory. The sample test results show that heavy metal Fe was the highest for all water samples analysed with a total of 0.244 mg/L. In addition, the most top-heavy metals for sediments were also recorded in Fe heavy metals for the whole areas tested. However, there is almost no Pb metal found in the whole areas. The results show that the highest heavy metal was found fish at 14,018 mg/g, i.e. *Channa striata* species. In addition, rivers experiencing pollution hugely affect living things and biota in water. It is suggested that the government policies and future supervision are needed so that environmental pollution, especially in rivers, can be reduced.

Keywords:
Sediment; Water; Fish Heavy metals; Geumpang river; species

1. Introduction

Heavy metals have caused various environmental pollutions with a level of concern that is quite broad to the rest of the world [1]. In recent years, heavy metal pollution has become one of the quite

* *Corresponding author.*
E-mail address: m_nasir@unsyiah.ac.id

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worrying environmental problems. The increased pollution of ecosystem metals in waters was initially affected by urbanization and industrialization [2–4]. The expanded metal pollution can cause significant health and harm to humans, invertebrates, and fish [5–7]. The impact of various human activities that can accumulate on freshwater ecosystems significantly affects the fish habitat. However, the quality of this fish habitat is very dependent on the density of operation and the population itself [8]. In addition, heavy metals can have harmful or beneficial effects on the lives of humans, plants and animals, but depending on the time of concentration. Metal that enters the environment can be introduced through various routes, for example, smelting, municipal waste, transportation, industrialization, underground pipe corrosion, and fuel combustion [9]. Heavy metals can threaten human health and the ecosystem associated with exposure to chemical pollution in the atmosphere [10-11]. Krueng Geumpang is one of the rivers that has been contaminated by Pb, Hg, Cu and Fe due to the increased pollutant discharge into rivers from various agricultural wastes, fertiliser runoff, and aquaculture pollution in recent years [12-13]. From multiple pollutants found in the atmosphere, heavy metal is one of the pollutants that often accumulates in vital organs in the living things. Therefore, it is hazardous to the circulatory system as well as the central and peripheral nerves [14–16].

Deposits contained in heavy metals are sediments discharged into the environment [17-18]. Sediment can also be a potential source of heavy metals to be released into the waters through natural and anthropogenic processes [19-20]. However, they have a terrible effect on drinking water. Heavy metals contained in sediments have toxins and can be mobilized through biochemical processes, such as causing water pollution, cyanobacterial blooming or can be contaminated by metals accumulating in their tissues so that they can enter the food chain [21-22]. The organism also needs some heavy metals. However, if the concentration is too high, it can cause poisoning. The content of heavy metals on sediment surfaces is more accessible because it can be immobilized through the adsorption process and coagulation [23]. Some studies have shown that sediments can also function as bodies and sources of contaminants in water systems [24–26]. Most of the researches on the heavy metal content associated with suspended particles and sediments distributed by heavy metals into sediments have provided evidence of anthropogenic effects in the screening system [27–29].

Biodiversity in the ecosystem and health status around the waters of Krueng Geumpang in recent years has worsened due to various activities by the community. Krueng Geumpang in Sigli Regency, Aceh Province, is a mountainous area with a denser population. However, people show a lack of awareness of not dumping various wastes into the river. Waste discharged in the river is usually from logging, during the fruit harvest and other industrial processes. River waste disposal can result in a drastic reduction in aquatic fish populations, ecological imbalances, and increase populations so that it can result in the loss of large amounts of fauna and flora on a large scale [30-31]. Krueng Geumpang River can be used as a source of drinking water for several in the coming decades if it is appropriately managed. In addition, rain from Krueng Geumpang has watered hundreds of hectares of residents' rice fields. Krueng Geumpang is not only an agricultural centre, but it can also be used as a tourist destination that can improve the economy. However, the rapid development of agriculture and industry, as well as the increasing population, have caused the occurrence of heavy metals from the disposal of waste into rivers can finally settle and masse [32–36]. Most of the pollutants in the river have flowed from the headwaters to Krueng Geumpang [37]. Unclear industrial waste, exploitation areas, and management have become the main problems of ecosystem destruction.

Aquatic fish is one of the essential parts for human needs; so, it is not surprising that the quality and safety aspects of fish are maintained [38]. High heavy metal content in aquatic ecosystems has given serious concern to the public and even throughout the world. Therefore, their high potential

can accumulate and enter into the food chain with the correlation between exposed heavy metals and cancer in humans [2,39]. The significant attention has paid in recent years to a variety of environmental contamination polluted by various chemicals and heavy metals [40]. Organic and inorganic pollutants that have released into the environment are human activities that have made a fairly serious threat to human life and natural surroundings. The organic contaminants are a series of chemical substances with potential that are continuous and are capable of being contaminated over great distances through water, air, and species migration. Fish found in freshwater are the top consumers. The results of the study reported that elements such as Pb, Hg, Cu and Fe could undergo biomagnification through the food chain in both freshwater and seawater ecosystems [25-26, 41-42]. Several studies discus fish in different environments that trophic position, body size, sex, age, eating behaviour can make the status of metal spawning for the same environment [43-44].

Researches on heavy metal pollution in water and sediments have been widely reported in various literature. Their findings show that the southern and eastern parts of the lake are generally higher than the west, while the most severe pollution is in the northern part of the lake [45-46]. Agricultural activities and some industries around the river have resulted in the accumulation of domestic and unprocessed industrial waste as found in China [47–49]. The presence of heavy metal and sediment toxicity in water has made it a significant source of metal biomagnification for fish, animals, and other aquatic plants. In addition, it can hurt those who consume contaminated fish [50]. Research on the level of heavy metal pollution in marine environments such as water, sediments, oysters and fish have also been reported [51–53].

In the river waters in several regions in Aceh Province and especially in Pidie Regency, no one has carried out multiple investigations or studies regarding heavy metals pollution in water, sediments and fish so far. Therefore, the focus of this research is to analyse the level of contamination and accumulation of heavy metals such as Pb, Hg, Cu and Fe distributed by heavy metals in water, sediments, and fish in Geumpang River. The results of this study are expected to help understand the behaviour of heavy metals in ecosystems in aquatic environments.

2. Methodology

2.1 Study Area

This study was conducted in Geumpang River in Pidie Regency, Aceh Province as shown in Figure 1. Meanwhile, the names, location, and coordinates of the area in the Geumpang River used as locations for sampling are illustrated in Table 1. Six locations were selected for sampling to make the analysis easier. In each area, three different samples such as water, sediment, and fish were taken in this study.

Table 1
Locations of research, GPS coordinates and characteristics of each for sampling

Locations	Coordinates	Characteristics
Lhok Tarok	4°49'40.93"U; 96° 8'10.01"T	bordering to the protected forest
Lhok Kuala	4°49'52.66"U; 96° 8'3.95"T	around the settlements and paddy fields also additional water from a small river
Kuala Keunee	4°51'29.97"U; 96° 6'46.27"T	assembly between Kr. Geumpang with Kr. Keunee
Lhok Tunggok	4°52'51.43"U; 96° 5'43.34"T	Many activities taking Quarry
Alue Landong	4°53'43.19"U; 96° 3'38.98"T	Bordered by natural forest and Alue Landong River
Tanjakan	4°55'12.66"U; 96° 1'18.89"T	River water deeper and larger rivers

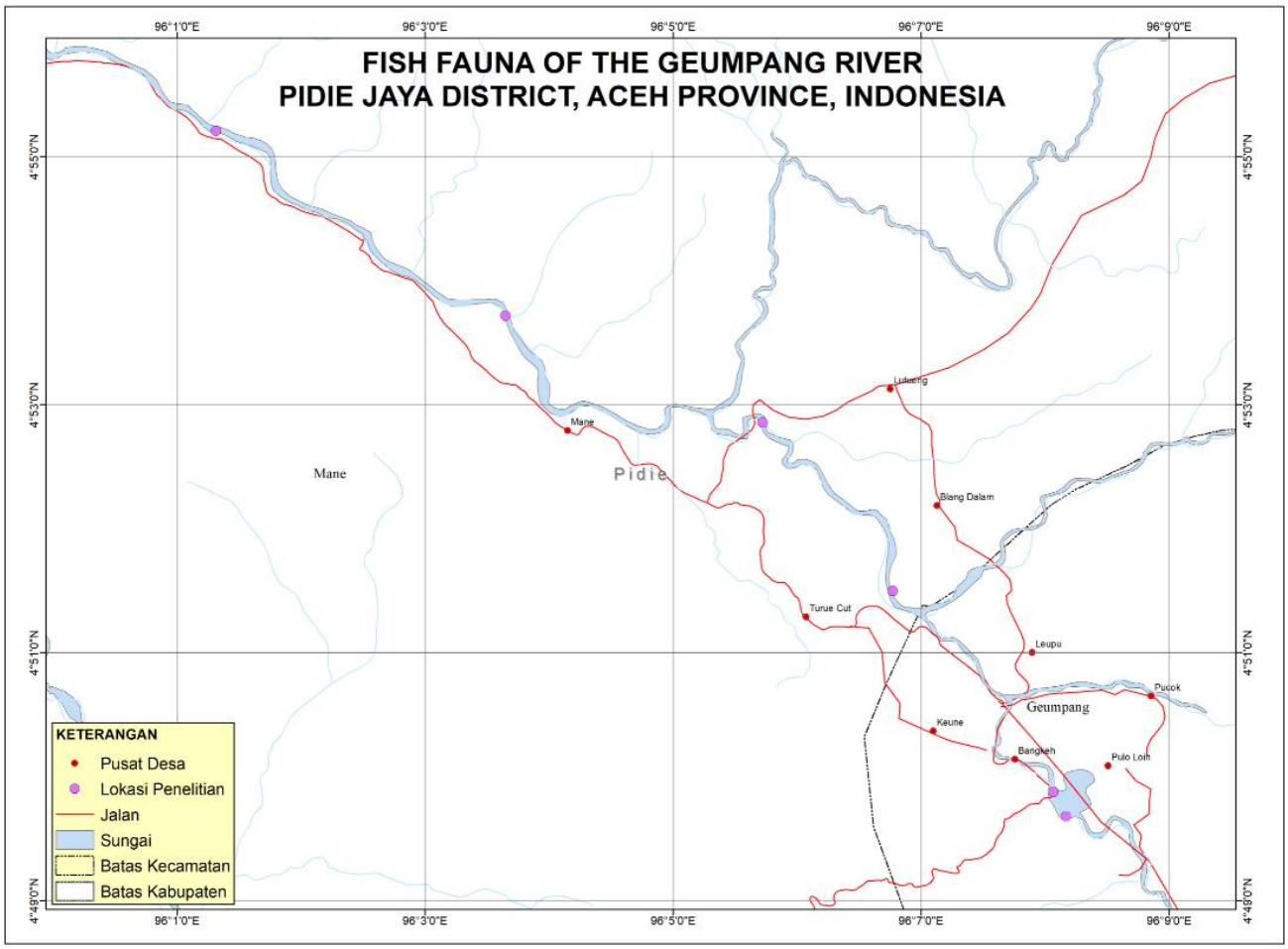

Fig. 1. Map of sampling locations in the Geumpang river area Pidie Regency

2.2 Water

Water samples were taken from six different locations in the Geumpang River and then put in bottles of 1-3 litters per bottle. Every 2 mL concentration of HNO_3 was added to each sample (1 litter) after filtration for further analysis. The liquid extraction techniques were carried out using the method of [54]. Furthermore, the use of condoning was done in the water phase. The use of water and HNO_3 was made with high purity. Organic solvents used in dissolved solvents were evaporated using hot plates at low temperatures and as a final solution carried out using the Atomic Absorption Spectrometry (AAS ZEEnit-700P) [55].

2.3 Sediment

Sampling for sediments was obtained from the same locations in Geumpang River, Pidie Regency, using the Van Veen sampler sample. The sampling standard used the Equal Discharge Increment (EDI) method. Sediment sampling was carried out at the midpoint on the cross-section of the river/channel that has the same amount of discharge. A total of 1 g of dry sediment sample was digested and then mixed with acids containing $HClO_4$ (2 mL), HCl (2 mL) and HF (10 mL) and the residue dissolved with concentrated HCl and the final process diluted as much as 25 mL [56]. Analysis of the sample solution was carried out with AAS flame to be able to estimate metal concentrations (Pb, Hg, Cu, and Fe). After that, the analysis continued with the use of MESS-1 which is a Standard Reference Material (SRM) carried out on all samples tested to check the accuracy of the investigation.

2.4 Fish

Fish sampling was conducted using an electrofishing tool with a power of 24 volts and 18 amperes of current. Electrofishing was operated for 30 minutes on the riverbank. All fish samples were transported to the laboratory on the same day. Furthermore, fish were dissected and stored before analysis. Homogenization of the dry sample was carried out by grinding on a porcelain mortar, then packaged using weighing paper and stored in a desiccator for further analysis. Measurement of heavy metal concentrations carried out by adjusting to the modification method as [57]. The contents of Pb, Hg, Cu and Fe in the tissue were determined by Atomic Absorption Spectrometry (ZEEnit 700P, AAS) and subsequently expressed as mg/g of wet weight in the mass [58]. Calibration using standard chemical solutions was done with chemicals that have been made and are commercially available (Merck, Germany). Empty analytics are carried out in the same process as the concentrations and samples determined using standard solutions prepared on the acid matrix. All reagents used in this analysis were ionized water and analytical grade. After that, all glass and plastic were soaked and washed with nitric acid for about 15 minutes, then rinsed with deionized water so that they were ready to use.

3. Results

3.1 Heavy Metals of Water

The results of total heavy metal concentrations in surface water samples show differences for each study site. The highest amount of heavy metal concentration was Fe type for all sample sites tested. The most top-heavy metal was found in the Lhok Tarok region with a total of 0.244 mg/L, followed with Kuala Keune at 0.187 mg/L, Lhok Kuala at 0.179 mg/L, Ramp at 0.173 mg/L, Lhok Tunggok at 0.170 mg/L and Alue Landong at 0.132 mg/L. Heavy metal with Cu type was the second-highest toxic type compared to Pb and Hg heavy metals. The highest total Cu heavy metal was found in the Ramp region at 0.033 mg/L, while the lowest heavy metal was found in the Lhok Tarok region at 0.23 mg/L as shown in Figure 2. Hg metal showed the lowest level of toxicity for all sample regions tested compared to other kinds of heavy metals. The highest Hg heavy metal reached only 0.002 mg/L, meaning that this value is minimal and can even be said to have no alarming toxicity. Investigations about heavy metals on the surface of the water have also been examined [59–61]. However, the types of heavy metals in their research included As, Cu, Fe, Zn, Hg, Cd and Pb and the samples were taken in winter, spring, summer and autumn. Average concentrations of Pb in water were 5.06, 2.28, 6.00 and 0.26 mg/L during winter, spring, summer and autumn, respectively, as reduced from the standard values. It was established by the WHO for drinking water. While the average concentrations of Cd observed were 0.74, 0.13, 0.24 and 0.14 mg/L during winter, spring, summer and autumn, respectively. Whereas the Cd concentration was the lowest of all observed for the whole sample compared to other heavy metals. Investigations of heavy metals in the water that have an impact on the health of humans and living things have also been carried out by [62]. Water treatment that is contaminated with heavy metals is carried out using palm kernel shells (PKS) applied as bio-adsorbent. The results reported that the use of PKS can remove ions such as Cr6 +, Pb2 +, Cd2 + and Zn2 + which are obtained in water respectively 98.92%, 99.01%, 84.23% and 83.45%.

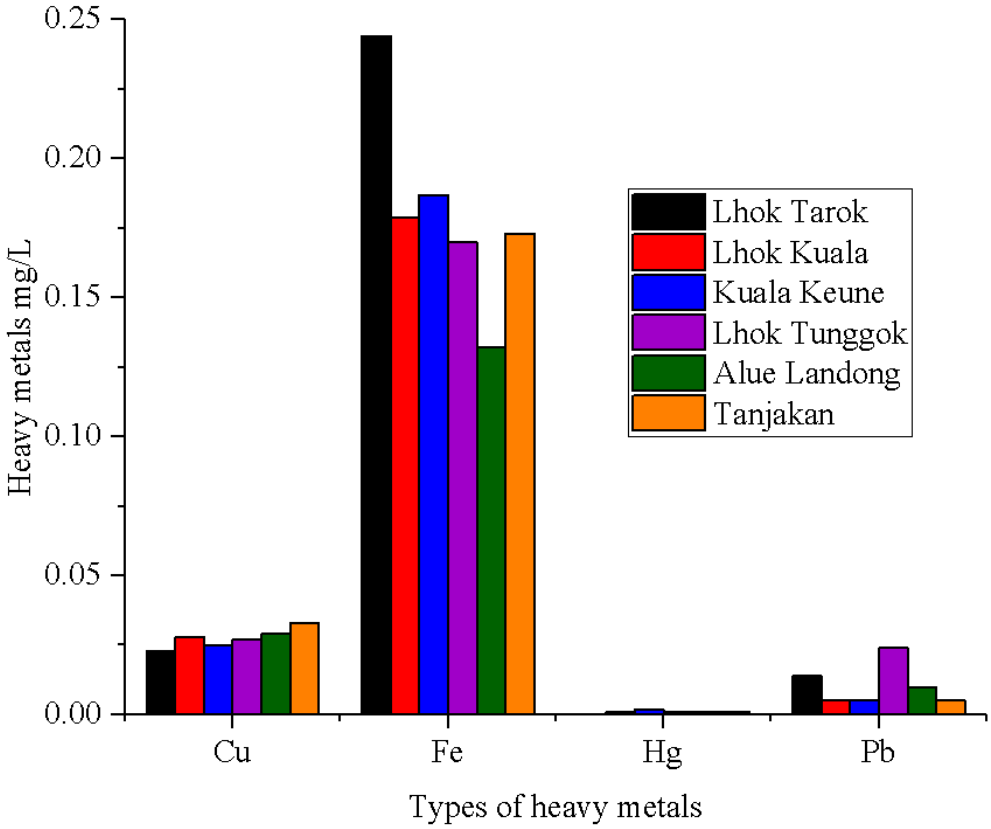

Fig. 2. Heavy metals for water at different river area

3.2 Heavy Metals of Sediment

The analysis aims to determine the level of heavy metals contained in the sample. The heavy metals included Cu, Fe, Hg and Pb. Based on the results of the analysis, as shown in Figure 3, it shows that heavy metal Fe is a toxic level higher than the other three types of heavy metals. The most top Fe heavy metals were recorded in the Lhok Tunggok and Kuala Keune regions with 2800.821 µm and 2779.888 µm, respectively. It was followed by the areas of Lhok Tarok, Alue Landong, Lhok Kuala and Tanjakan with 2660.076 µm, 2534.042 µm, 2489.914 µm, and 2304.8 µm, respectively. While the Pb heavy metal toxic level was almost not seen in five different locations. Pb heavy metal was only found in the Tanjakan area of 1.017 µm, while the other five areas did not show Pb heavy metal, as shown in Figure 3. For the type of heavy metal, Cu showed the second-highest level of toxin compared to the kind of heavy metal Hg. The highest value of Cu heavy metals was recorded in the Lhok Tunggok region of 20.932 µm. Whereas in the Lhok Kuala region, it was the lowest toxicity level of 15.801 µm compared to other places. Meanwhile, the highest type of heavy metal Hg was obtained in the Kuala Keune region of 0.031 µm, higher than 0.002 µm compared to the Lhok Tunggok region. The Tanjakan area was the lowest polluted area with toxins with Hg heavy metal which was equal to 0.024 µm.

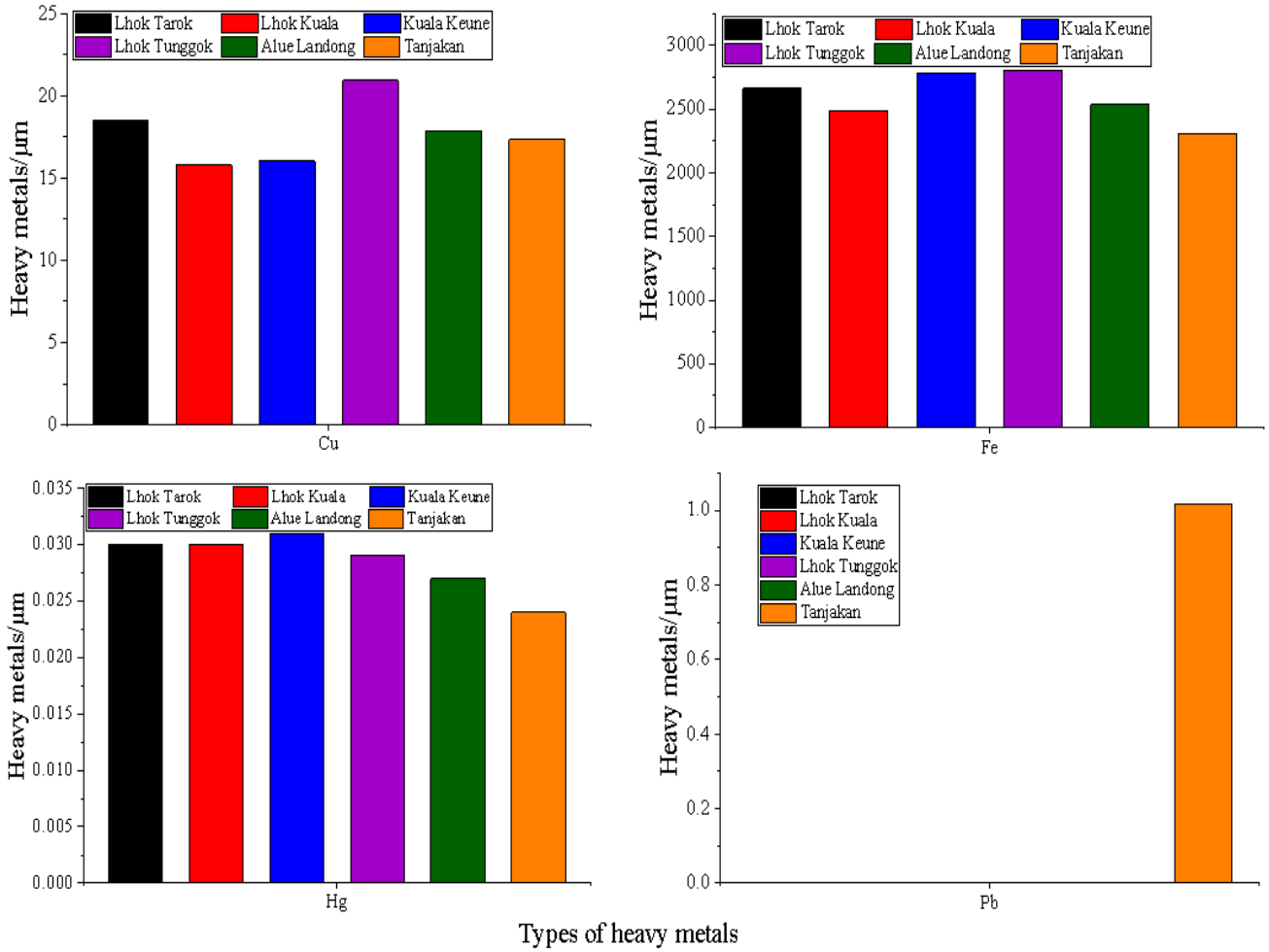

Fig. 3. Heavy metals for sediment at different river area

3.3 Heavy Metals Analysis of Fish

Analysis of fish samples is based on samples taken from the Geumpang River together with water and sediment samples as shown in Table 2. Six types of fish became the primary concern for the analysis of heavy metals Cu, Fe, Hg and Pb. *Channa striata* species have the highest levels of heavy metal Fe compared to other types of fish. The total Fe heavy metal for Channa striata fish reached 14.018 mg/g compared to *Protomyzon griswoldi* which was only 3.205 mg/g. Hg heavy metal is a toxic level lower than Cu and Pb. The type of Puntius brevis heavy metal level contained in it was the smallest and followed by the kind of fish *Neolissochilus thienemanni* for all types of metals tested. Samples of fish used for this analysis were obtained from six different locations as shown in Table 2. The result of the analysis shows that not all regions have the same type of fish, even some areas do not have the type of fish tested. The location that has the most fish is the Lhok Tarok area with a total of 42 fish Followed with Alue Landong area of 15 fish, Kuala Keune of 12 fish, Tanjakan of 12 fish, Lhok Tunggok of 7, and Lhok Kuala of 1 fish. The samples of fish in this study were transported from the Geumpang River to the Laboratory to be analysed.

Investigation of heavy metals found in fish conducted in this study as shown in Figure 4. This analysis uses six different types of fish with four parameters (such as; Cu, Fe, Hg and Pb). The highest heavy metal was found in *Channa striata* fish at 14,018 mg/g and the lowest was *Anematichthys apogon* for Fe. The highest Pb and Hg heavy metals were recorded in the species of *Protomyzon griswoldi* and *Anematichthys apogon* of 1,625 mg/g and 1,112 mg/g, respectively. As for the highest

heavy metal Cu found in fish species *Channa striata, Protomyzon griswoldi* and *Anematichthys apogon* respectively 1,373 mg/g, 1,168 mg/g and 1.57 mg/g as shown in Figure 4. Based on the results of this analysis shows that the fish species found in the Geumpang River have been polluted by heavy metals. Investigation of heavy metals on the environment using phytoremediation techniques has been carried out by [63]. Where the results of the research conducted are considered very cost-effective and also environmentally friendly. Thus, continuous supervision in the future is expected with several policies from the government so that the Geumpang River is no longer polluted in the future.

Table 2
List of *fish* species and total individuals in the Geumpang River, Aceh Province, Indonesia

Family	Species	Local name	Locations						Total	(%)
			A	B	C	D	E	F		
Anabantidae	*Anabas testudineus*	Krup	1	N/A	N/A	N/A	N/A	N/A	1	16,7
Balitoridae	*Protomyzon griswoldi*	Ilee	10	N/A	N/A	N/A	N/A	N/A	10	16,7
Channidae	*Channa striata*	Bace	4	N/A	N/A	N/A	N/A	N/A	4	16,7
Cichlidae	*Oreochromis mossambichus*	Mujair	N/A	N/A	N/A	N/A	1	N/A	1	16,7
	Oreochromis niloticus	Nila	1	N/A	N/A	N/A	1	N/A	2	33,3
Cyprinidae	*Anematichthys apogon*	Ceperas	9	N/A	N/A	N/A	1	N/A	10	33,3
	Hampala macrolepidota	Palung	2	N/A	N/A	N/A		N/A	2	16,7
	*Neolissochilus thienemanni**	Kerling	N/A	1	N/A	N/A	4	11	16	50,0
	Puntius brevis	Groe	3	N/A	N/A	N/A	N/A	N/A	3	16,7
	Rasbora sp.	Depik	6	N/A	N/A	N/A	N/A	N/A	6	16,7
	Tor soro Valenciennes	Naleh	4	N/A	12	7	8	N/A	31	66,7
Eleotridae	*Ophiocarasp*	Ntok	2	N/A	N/A	N/A	N/A	N/A	2	16,7
Number of Individual			42	1	12	7	15	11	88	
Number of species			10	1	1	1	5	1	12	

Note: * IUCN red list, A= Lhok Tarok, B= Lhok Kuala, C= Kuala Keunee, D= Lhok Tunggok, E= Alue Landong, F=Tanjakan

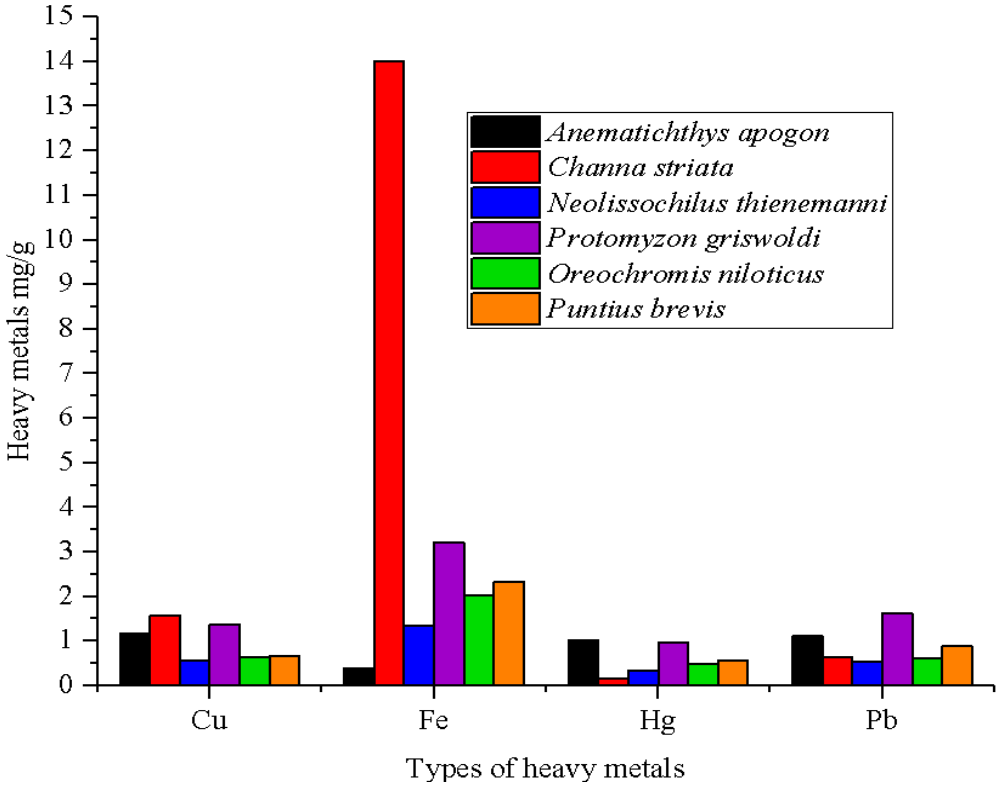

Fig. 4. Total heavy metals for different fish

The overall analysis of the heavy metals found on the surface of the water, sediments and fish has completed. Analysis of the entire sample in this study was conducted at the Unsyiah Chemistry Laboratory and the FMIPA-Unsyiah Laboratory. This research was conducted for almost six months starting from February to August 2019. This study aims to analyse problems related to heavy metals. This research was conducted to investigate the level of pollution in the Geumpang River. The results of this investigation are used as a step for river monitoring in the area. Therefore, it can be concluded that the Geumpang River has been polluted by heavy metals and further supervision is needed to preserve biodiversity and aquatic biota in the future.

4. Conclusions

The heavy metal analysis carried out in this study using samples and data taken directly from the Geumpang River area, Pidie District has completed. So some conclusions that can be conveyed from the results of the analysis are described one by one as follows

I. The highest metals present on the surface of the water recorded in the Fe type for all areas tested. The highest level of heavy metal Fe obtained in the Lhok Taruk area of 0.244 mg/L.
II. The analysis of the sediment tested from the highest different samples obtained in the type of heavy metal Fe for all regions studied while the nature of heavy metal Pb does not have toxins for the three types of heavy metals respectively Cu, Hg and Fe.
III. The highest type of heavy metal (Fe) was found in *Channa Striata* fish at 14,018 mg/g and the lowest was recorded in *Anematichthys apogon* fish type for heavy metal (Fe) at 0.4 mg/g.

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