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Distribution of heavy metal (Pb, Cd and Hg) concentrations in sediment of Bone River, Gorontalo

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ABSTRACT

The concentration of three heavy metals Pb, Cd and Hg were measured in Bone River, Gorontalo sediment. The heavy metal pollution on sediment in Bone River have not been explored longitudinally. The present study aimed to analyze the concentration of heavy metals Pb, Cd, and Hg in the sediments of the Bone River based on Contamination factor (CF) and geoaccumulation index (Igeo). The samples were taken in July 2021 at 8 stations. Heavy metal concentration measured using AAS method. The trend of metals were observed in sediment as Pb>Cd>Hg. The level of studied metals Cd and Hg did not exceed the safe limit by ANZECC, OSQG LEL, and CCME TEL; meanwhile, Pb met the limit by ANZECC. However, the investigated showed fluctuations due to differences in the characteristics of each location. CF and Igeo demonstrated that the sediment samples were low contamination and lightly polluted. Contamination by heavy metals in the Bone River implies that the conditions are much frightening for the biota and residents around the river.

Introduction

The quantity and quality of watershed water are influenced by the nature and conditions of nature and human activities based on place and time. Rivers have a crucial role in society. Apart from being a place for the ecosystem, it is also a source of life for the surrounding community. Various human activities, such as industrial and household waste disposal, have caused a decline in river water quality. Anthropogenic activities can change the input of organic matter, nutrients, and heavy metals into river ecosystems through changes in land use and urbanization. Anthropogenic modifications have disrupted aquatic ecosystems by modifying river flow and sediment transport.

Water as an integrator in a watershed will reflect all qualitative and quantitative anthropogenic pressures. Heavy metal pollution from anthropogenic activities can cause changes in the quality of the river water environment (Islam *et al.*, 2015) which also affects the life of the biota in it (Kurz *et al.*, 2013; Fuoco and Giannarelli, 2019; Barbieri *et al.*, 2020). Continuous pollution can impact biota and even cause death, affecting the population (Kadim and Arfiati, 2022).

Heavy metals are the most dangerous pollutants because they have toxic, carcinogenic, biomagnifying and bioaccumulative properties. Moreover, self-purification cannot remove heavy metals from water but accumulate in suspended particulates and sediments. As a result, higher consumers will absorb the food web process. Therefore, metal pollution in waters, especially rivers, is an essential issue in many countries, including Indonesia. Metal contamination in waters has been reported in several areas in Indonesia, such as the Lamat River (Arkianti *et al.*, 2019), Ciliwung River (Sudarso *et al.*, 2013), Kaligarang River (Dewi *et al.*, 2014), Gempol River (Widiastuti *et al.*, 2018), the Bonded River (Wahyuningsih *et al.*, 2021), Wobatu Bay (Edward, 2014) including the eastern coast of Sumatra Island, North Coast of Java,

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Kalimantan (Arifin *et al.*, 2012; Budiyanto and Lestari, 2017; Yona *et al.*, 2018).

One of the largest rivers in Gorontalo Province is the Bone River. The Bone River Corridor is a water conservation facility for the surrounding area because it provides various water needs (managed by PDAM), ranging from clean water and water for agriculture to tourism. The Bone River is also part of the Bogani Nani Wartabone National Park area. In addition, the study's results (Sahami and Habibie, 2020) found that the Bone River is the habitat of Nike brood fish, which are economically important fish for the people of Gorontalo. Balihristi data for 2008-2011 (Biki *et al.*, 2012) shows that several land use activities around the river potentially threaten the Bone River ecosystem; these activities include agriculture, settlements, gold mining, sand mining, industry, and illegal logging.

Lihawa and Mahmud (2012) reported that high mercury levels in water and sediment floated in rivers in Tulabolo Village, where there was gold tailings activity, and the flow of these rivers would enter the Bone River. In addition, Wahyuningsih *et al.* (2021) stated that heavy metals can form complex bonds with particles at the bottom of the water so that they settle and accumulate in sediments; this condition causes the concentration of heavy metals in sediments to tend to be higher than those bound to water. Therefore, activities along the Bone River segment can disrupt the ecological balance of the aquatic organisms and interfere with human health. Therefore, studying the distribution of heavy metals in the Bone River is necessary. Therefore, this study aims to analyze the concentration of heavy metals Pb, Cd, and Hg in the sediments of the Bone River. Finally, our research should allow us to answer whether analyzing heavy metal concentrations in sediments based on an index analysis approach can provide information on the quality of the Bone River sediments.

Materials and Methods

Location and time of research

The research was carried out in the Bone River, Gorontalo Province. Sampling was carried out in July 2021 at eight observation stations. These were determined purposively based on ecological conditions and anthropological factors. Preferably rifle area, considering land use activities that may be a source of metal contamination such as agriculture, settlement, and mining (Figure 1). The material studied is the concentration of metals (Pb, Cd, and Hg) in river sediment samples.

Data collection procedure

Sediment sampling was carried out at a depth of ±50 cm at each station using a modified soil sample ring from the Parallon pipe; Sediment samples taken as much as 250 g (Widiastuti *et al.*, 2019). The prepared sediment sample was then stored in a cool box for analysis. The concentrations of Pb, Cd and Hg in the sediment were observed using the Atomic Absorption Spectrophotometer (AAS) method at the Surabaya Health Laboratory Centre, and the procedure refers to APHA, 23rd Edition, 3111-B, 3030-H, 2017. The number read on the spectrophotometer is expressed as the values of the heavy metal concentration of the sample.

Data analysis

The metal concentrations of Pb, Hg, and Cd from each station were compared with sediment quality standards from other countries (Table 2) and also analyzed using the contamination factor (CF), Geoaccumulation Index (Igeo) to determine the level of heavy metal contamination in sediments, referring to research conducted by several previous researchers, including Edward (2020), Wahyuningsih *et al.* (2021) and Liao *et al.* (2022). Tables of natural levels and normal levels of metals measured in this study are presented in Table 1.

Table 1. Natural levels and normal levels of metals Pb, Cd, and Hg.

Parameters	Natural levels* (ppm)	Normal levels** (ppm)
Pb	41,73	20
Cd	0,5	0,3
Hg	0,528	0,4
Reference	Edward (2020)	

Note: *Natural content of metals in nature (earth's crust); **Normal levels of metals in nature

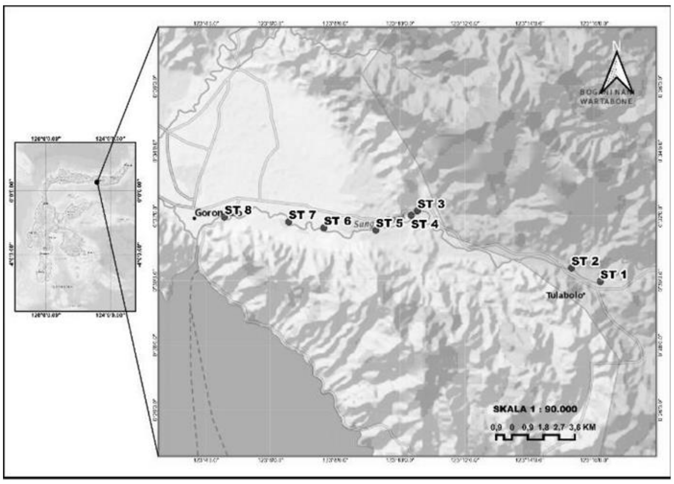

Figure 1. Maps showing the location of the sampling station in Bone River (Maps created using QGIS software).

Results

The concentrations of heavy metals Pb, Cd, and Hg measured at each observation station varied. This difference is thought to be influenced by differences in the characteristics of both physical and chemical properties of the waters of each observation station (Dewi *et al*., 2014; Edward, 2020; Takarina, 2014; Yang *et al*., 2020). The measurement results are presented in Table 2.

Table 2. Measurable concentrations (ppm) of Pb, Cd, and Hg metals in Bone River sediments.

Station	Pb	Cd	Hg
1	28.75	0.012	0.008
2	32.09	0.008	0.003
3	26.08	0.011	0.002
4	39.76	0.006	0.003
5	27.1	0.004	0.001
6	30.84	0.014	0.007
7	34.1	0.018	0.009
8	31.73	0.009	0.004
Average	31.306	0.010	0.005
Limits (ppm)*			
ANZECC/ARMCANZ	50	1,5	0,15
OSQG LEL	31	0,6	0,2
CCME TEL	30,2	0,7	0,13

*ANZECC (*Australian and New Zealand Environment and Conservation Council*) (*Simpson et al*., 2013); OSQG LEL (*Ontario Sediment Quality Guidelines-Lowest Effect Level*); CCME TEL (*Canadian Council of Ministers of the Environment- Threshold Effect Level*) (Edward, 2020)

Based on the data presented in Table 2, it can be seen that the highest Pb concentration was at St4, then St7, St2, St8, and St6 (concentration >30 ppm), while the lowest was at St3. The highest Cd concentration was found at St7, followed by St6, St1, and St3 (concentration >0.01 ppm), and the lowest concentration was at St5. Mercury (Hg) became a heavy metal with the lowest average concentration during the sampling period compared to Pb and Cd. The highest Hg concentration was at St7, then St1 and St6 (concentration >0.005 ppm), while the lowest was found at St5. If mapped, then St7 and St6 are observation stations with high Pb, Cd, and Hg metal concentrations in the Bone River. Specifically, for Pb, the data in Table 2 shows that St4 is the location with the highest concentration. This indicates that these stations receive much input, producing Pb, Cd, and Hg waste. Meanwhile, St5 (for Cd and Hg) and St3 (for Pb) became stations with low concentrations compared to other observation stations.

Discussion

Pb generally has the highest average concentration value of 31.3 ppm, which is very different from the concentrations of Cd (0.01 ppm) and Hg (0.005 ppm). Therefore, the average concentration of Pb in this study was higher than the standard value of OSQG LEL and CCME TEL but still did not exceed the standard value set by ANZECC.

The high concentration of Pb was due to the input of waste from dense residential and agricultural activities (St2, St4, and St8) as well as heavy equipment and loading and unloading vehicles for sand mining (St6 and St7), causing many fuel spills and spills around the waters. On the other hand, the low concentration of Pb in St3 is because it is an area or station with low activity; there are no settlements and other activities. According to Wahyuningsih *et al.* (2021), the high and low concentrations of Pb in the waters are influenced by the intensity of human activities in the vicinity, both from agricultural, household, and industrial activities, as Ahmed *et al.* (2017) stated that Pb is a typical anthropogenic metal where its presence in waters can be influenced by surface runoff and input of organic waste from municipal waste or solid waste. In addition, the high and low concentrations of Pb in the waters are caused by anthropogenic sources of metals such as spilled fuel containing lead from heavy equipment (machinery), industrial activities, or transportation.

The low concentrations of Cd and Hg were caused by several factors, such as the low anthropogenic activity of these metals or the nature of the metals. The low metal concentration in the waters also indicates the low level of metal accumulation in the sediments due to the low sediment capacity in metal particle deposition. Zhang *et al.* (2014) explained that the concentration of heavy metals in sediments is influenced by the ability of the sediment to adsorb surface particles, ion exchange, deposition, and complexation with organic matter.

According to Wang *et al.* (2019), Cd is a heavy metal that sediment surfaces can absorb. However, the concentration and properties of Cd metal in waters can change over time due to pollution and oxidation. Meanwhile, Mercury that enters public waters can be converted from one form to another in the ecosystem to form a complex cycle, then reaches sediments by deposition of particles and then released by simple diffusion or resuspension (De *et al*., 2014). According to Lihawa and Mahmud (2012), microorganisms can convert Hg into CH_3-Hg (methyl mercury). Methyl mercury has the most toxic properties with sturdy binding and high solubility, especially in the body of aquatic biota. Mercury, lead, and cadmium are non-essential heavy metals that are the most toxic than other heavy metals and are difficult to degrade naturally (Dewi *et al.*, 2015; Erasmus *et al.*, 2020). Therefore, lead (Pb) can be used as an indicator of pollution, especially from fuel use (Mohiuddin *et al.*, 2010). In this study, we analyzed the measured heavy metal concentrations using an analytical approach, namely contamination factor and geoaccumulation index. Contamination factors widely used to indicate the degree of contamination with a single metal and the ratio of the content of each metal to its background value. Meanwhile, the geoaccumulation index (Igeo) is often used for the assessment of heavy metal pollution and reveals the relationship between heavy metals in sediments and the value of the geochemical background (Liao *et al.*, 2022). Figure 2 below presents the value of the contamination factor, geoaccumulation index and pollution load index of Pb, Cd, and Hg.

The data in Figure 2 shows that distance does not affect fluctuations in the concentration of Pb, Cd, and Hg in this study. The opposite was found by Lihawa and Mahmud (2012) in a study in the Tulabolo River, whose flow will empty into the Bone River (St2 in this study) on Hg concentrations, where distance has a significant effect on mercury concentrations in bottom sediments, the farther from the source pollution levels are lower. The existence of fluctuations in these levels can be caused by differences in the characteristics of each location or station, such as the physical and chemical properties of the waters. The level of acidity (pH) (Yona *et al.*, 2018), temperature, dissolved oxygen (DO) (Mohiuddin *et al.*, 2010), and current velocity (Li *et al.*, 2012) affect fluctuations in heavy metal levels in sediments. Metals deposited or dissolved in sediments are influenced by water's physical and chemical properties, such as pH and organic matter (Yang *et al.*, 2020; Zhang *et al.*, 2014).

The average value of contamination factor (CF) for Pb is 0.75; Cd is 0.021, and Hg is 0.009. The interpretation of the CF value refers to Hakanson (1980), where if the CF value <1 means the level of contamination is low; if the value of 1≤CF<3 means the level of contamination is moderate; if the value of 3≤CF<6 means the level of contamination is high; and the value of CF> 6 means the level of contamination is very high. The mean value of CF Pb, Cd, and Hg shows a number smaller than 1 (CF<1). This means that the sediment at all observation stations is in the category with a low level of contamination.

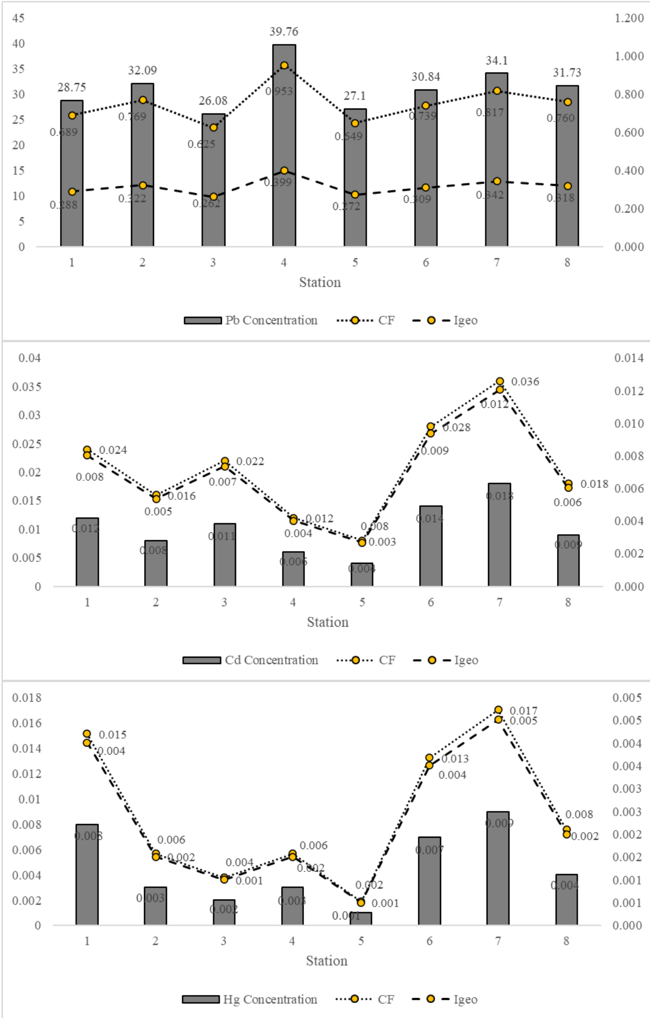

Figure 2. The value of the contamination factor and geoaccumulation index of Pb, Cd, and Hg.

The mean geoaccumulation index (I_geo) of Pb is 0.314, while for Cd, it is 0.007, and Hg is 0.002. The interpretation of the I_geo value refers to 7 classes (Muller (1979) in Edward (2020)) namely class 0 (not polluted) the value of I_geo<0; class 1 (not contaminated to moderately contaminated) value 0<I_geo<1; class 2 (medium polluted) value 1<I_geo<2; class 3 (moderate to heavily polluted) value 2<I_geo<3; class 4 (sufficiently polluted) value 3<I_geo<4; class 5 (severely polluted) value 4<I_geo<5; and grade 6 (extremely badly polluted) value I_geo>5. The mean value of I_geo Pb, Cd and Hg based on the calculation results is greater than 0 but less than 1 (0<I_geo<1) or belongs to class 1, meaning that the sediment at all observation stations is categorized as lightly polluted.

According to statistics, more than 99% of heavy metals that enter the waters can be stored in sediments (Salomons, 1995). River sediments are carriers of heavy metals and thus play an essential role in assessing and tracking sources of contamination. Heavy metals can cause solemn environmental damage in the vicinity due to their toxicity, persistence, and bioaccumulation (Shen *et al*., 2019). Anthropogenic activities such as industrial waste, vehicle exhaust, mining operations, agriculture, and rainfall are the leading causes of heavy metal pollution in waters (Liu *et al*., 2018; Wang *et al*., 2007). Heavy metals enter through several biogeochemical cycles and eventually enter the human food chain, causing bioaccumulation and biomagnification (Doyi *et al*., 2018).

Conclusion

Pb has a much higher average concentration than Cd and Hg. The Pb concentration is still under ANZECC but has exceeded the quality standard OSQG LEL and CCME TEL set. Based on the CF value, the sediment in the Bone River is in the category of low contamination, while the Igeo value of the sediment in this river is in the lightly polluted category.

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