



Distribution pattern of Pb and Zn contamination in rivers near industrial zone in Aceh, Indonesia, revealed by principal component analysis (PCA)

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Article History

Received: 14 September 2021; Received in Revision: 22 October 2021; Accepted: 3 November 2021

Abstract

The assessment of the heavy metal contamination in aquatic environment especially in rivers near industrial zone is critically needed. Therefore, the objective of this research was to evaluate the pollution of Pb and Zn in water, sediment, and *Faunus ater* samples collected from Krueng Balee (KB) and Krueng Reuleung (KR) Rivers, Aceh, Indonesia. The samples were collected at the upstream, midstream, and downstream of each river and analyzed using Atomic Absorption Spectrophotometry (AAS). The distribution pattern of Pb and Zn pollution was analyzed using Principal Component Analysis (PCA). The result of the investigation revealed that the presence of Pb and Zn in water was about 0.0087 and 74.79 mg/kg, respectively. This level fell below the maximum threshold set by Indonesian Presidential Decree No. 22 Year 2021 (0.03 and 0.05 mg/L for Pb and Zn, respectively). Similarly, the highest concentrations of Pb and Zn in the sediment were 74.79 and 148.69 mg/kg, respectively, where according to the Ontario Sediments Quality Guidelines the maximum thresholds for Pb and Zn are 250 and 820 mg/kg, respectively. Nevertheless, Pb and Zn contaminations found in *F. ater* samples had exceeded the maximum thresholds set by the World Health Organization (0.5 and 50 mg/kg, respectively), in which the highest concentrations of Pb and Zn reached 8.13 and 98.57 mg/kg, respectively. PCA analysis revealed correlations between samples suggesting the roles of physical and chemical properties of the river in the pollutant retainment. The analysis also revealed the possible antagonism between Pb and Zn accumulation in *F. ater* which is a novel finding. We suggest routine monitoring of Pb and Zn concentrations. The role of the surrounding industry in the Pb and Zn pollution should be further studied.

Keywords: Accumulation, bioindicator, heavy metal, mollusk, PCA, pollution

1. Introduction

Anthropological activities in various industries (including processing industry, animal husbandry, agriculture, and fisheries) has led to the increase of chemical discharge causing pollutions in aquatic ecosystems such as rivers, lakes, and oceans (Li *et al.*, 2018; Song *et al.*, 2018; Wong *et al.*, 2017; Zhang *et al.*, 2017). In general, the wastewater effluent released to an aquatic ecosystem is divided into two; non-biodegradable inorganic and biodegradable organic waste.

Heavy metals are non-biodegradable substances, capable of rapidly assimilated into the environment, and acutely toxic (Zhang *et al.*, 2017), and their presence is common in industrial waste (Vardhan *et al.*, 2019). In

high concentrations, these pollutants are hazardous to aquatic biota (Jafarzadeh *et al.*, 2020). Researches have been conducted to counter the environmental pollution such as phytoremediation (Wei *et al.*, 2021), flocculation (Sun *et al.*, 2020), and adsorption (Rahmi *et al.*, 2021; Rahmi *et al.*, 2021), yet the problem still persists.

The bioaccumulation of hazardous heavy metals could threaten human lives through food chain exposure (Goretti *et al.*, 2020). Several examples of industrial waste-derived heavy metals are Hg, Cd, Ni, Cu, Pb, and Zn (Mohiuddin *et al.*, 2011; Viczek *et al.*, 2020). In aquatic environment, heavy metals could be accumulated in sediments and biotas.

Heavy metal exposure to living organism could occur through respiratory tract, food, and skin absorption (He et al., 2020). The toxicity itself depends on the medium carrying the heavy metals (sediment, water, or living tissue) (Keser et al., 2020). To evaluate heavy metal pollution in an aquatic environment, organisms could be used as bioindicators including gastropods which have been known as heavy metal accumulator (Corrias et al., 2020). The ability of gastropods to highly accumulate not only heavy metals but also other pollutants (i.e. hydrocarbons and pesticides) is attributed to their slow locomotion and filtration-dependent feeding (Radwan et al., 2020).

Herein, we analyzed the heavy metal contents in water, sediments, and shells (*Faunus ater*) to determine the aquatic environment quality collected from Krueng Balee (KB) and Krueng Reuleung (KR) Rivers. The strategy of analyzing the environment quality using the aforementioned samples has also been suggested by previous research (Wong et al., 2017). An investigation of aquatic heavy metal pollution has been conducted in the East Black Sea coast region, Turkey, by using water, sediments, and bioindicators (Baltas et al., 2017). The stated study suggested higher accumulation of heavy metals (Cu, Zn, and Pb) in the living tissues of seashells and sea snails in comparison with that in the sediments and water, in which the results were consistent in all sampling points (Baltas et al., 2017). Another study at Douglas Creek, the Qua Iboe Estuary, Nigeria, showed that the heavy metal pollution was dominated by Zn among all tested heavy metals (Benson et al., 2018). To understand the characteristics of the heavy metal contamination, Principal Component Analysis (PCA) could be used. In a previous study, PCA analysis had been used to evaluate the Hg distribution pattern around gold mining area in Aceh Jaya District, Aceh (Wahidah et al., 2019).

In Aceh itself, the heavy metal analysis in water, sediments, and fish from aquatic environment have been reported (Ukhty et al., 2020; Rizkiana et al., 2017; Supriatno & Lelifajri, 2009). Nonetheless, another study conducted in Riau Islands Province, Indonesia, reported significant contamination of Pb in *Nerita lineata*, with the minor presence of other heavy metals (Cu and Zn) (Sari et al., 2016). The same study also suggested that the accumulation was significantly higher in the shell than in the flesh of *N. lineata* (Sari et al., 2016). The inconclusive results from the stated studies could be associated with different sampling locations; meaning

different environment characteristics as well as surrounding anthropogenic activities.

In present study, we aimed to investigate the role of industrial activities on the Pb and Zn pollution in nearby river. KB was selected as the sampling point because it is the closest one to the cement industry. During the manufacturing, Pb and Zn are used as cement clinkers (Nouairi et al., 2018), leading to our suspicion on the pollution near the area. For comparison, sampling point KR was selected; located distant away from the cement industry. Previously, reports pertaining to the characteristics of Pb and Zn contamination in the river near the cement industry, in Aceh – Indonesia, are scarce. Thus, this paper is significant for tackling the heavy metal pollution problem.

2. Methodology

2.1. Materials and equipment

Chemicals used in the study were analytical grade H₂O₂ and HNO₃ (Merck, Selangor, Malaysia). Heavy metal analysis was carried out using Absorption Spectroscopy (AAS) (Shimadzu AA 6300, Kyoto, Japan). Statistical analysis was carried out on XLSTAT 5.0 software (Addinsoft, Paris, France).

2.2. Sample collection

Water, sediment, and *F. ater* samples were collected from KB and KR, Aceh Besar District, Aceh, Indonesia within June-September 2020. The sampling locations in KB were divided into three stations. The first station was the upstream located at coordinates 5°27'08.2 "LU-95°14'40.4" east longitude and at coordinates 5°27'01.4 "North Latitude-95°14'39.2 "East Longitude. The second station was the midstream located at coordinates 5°27'06.5" North Latitude - 95°14'38.6 "East Longitude. The last station was the downstream located at coordinates 5°27'0.7.3" North Latitude – 95° 14'37.7" East Longitude.

As for the KR, samples were collected from: the upstream - 5°23'05.0" North Latitude and 95°15'51.3" East Longitude; the midstream - 5°23'02.2" North Latitude and 95°15'36.9" East Longitude; and the downstream - 5°23'14.3" North Latitude and 95°15'18.2" East Longitude. Samples were labeled according to sampling locations and date. For example, a sample from KB collected on the first month of the study (June) was labeled as KB 1. During sampling and analysis, H₂O₂ and HNO₃, were used.

2.3. Sample preparation and analysis

2.3.1. Water sample

The procedure of heavy metal content analysis in water samples was based on the previous report (Clesceri, 1998). Briefly, 50 mL water was evaporated to reach 15 mL using a water bath. Afterward, 5 mL $\text{HNO}_3(\text{concentrated})$ was added into sample and heated again for 15 minutes, followed by another addition of 5 mL $\text{HNO}_3(\text{concentrated})$ and heated for another 15 minutes. The prepared samples were then analyzed using Atomic Absorption Spectroscopy (AAS) (Shimadzu AA 6300, Kyoto, Japan).

2.3.2. Sediment Sample

Sediment sample preparation also followed the previous report (Clesceri, 1998). Sediment samples were weighed 5 g and oven-dried at 60°C . The heavy metal in the sediment was extracted by an addition of 25 mL $\text{HNO}_3(\text{concentrated})$ solution, followed by heating process where the sample container was covered using a watch glass to prevent evaporation. The sample was filtered in a volumetric flask and added with double distilled water to reach 50 mL prior to AAS analysis.

2.3.3. Mollusk Sample

The *F. ater* sample was crushed and weighed 20 grams, then oven-dried at 105°C for 12 hours. Heavy metals extraction from the *F. Ater* was carried out with the addition of 35 mL $\text{HNO}_3(\text{concentrated})$ and heating, similar to the procedure for sediment samples. Afterward, 5 mL H_2O_2 solution was added to dissolve the existing fat and protein. Samples were filtered in a 50 mL volumetric flask and added with double distilled water to reach 50 mL before analyzed using AAS instruments as suggested previously (Agustina et al., 2019).

2.4. Data Analysis

To evaluate the spatial distribution pattern of Pb or Zn contamination in the two different rivers, ANOVA and Principal Component Analysis (PCA) were used, as suggested by previous report (Wahidah et al., 2019). The analyses were carried out in XLSTAT 5.0 software (Addinsoft, Paris, France). The information obtained from the PCA includes the relative similarity between the objects of observation, where points nearby indicate objects sharing similar characteristics. In this analysis, positive correlation between

variables is indicated by a value that is close to 1.

3. Results and discussion

3.1. Pb contents in water, sediment and *F. ater*

The results of Pb analysis in water, sediment and *F. ater* samples are showed in Table 1. Our investigation concluded that Pb concentrations have statistical significance with respect to the sampling location and date based on the ANOVA analysis (Table 2), where the $F_{\text{experimental}} > F_{\text{theoretical}}$ ($61.80 > 3.93$). The Pb concentrations at all sampling points in KB were found higher than that in KR. The heavy metals could be suspected to be originated from industrial activity and motorized fumes that eventually released to the river through wind and rain (Huang et al., 2021).

The source could be either from the manufacturing process or material transportation involving shipping process. Previously, Pb has been detected high in industrial zone associated with intense anthropological activities including that of Pb-containing fuels combustion (Li et al., 2018). It is further strengthened by the fact that KB is the closest one to the highly traffic road. Both Pb contamination in water collected from KB and KR has not exceeded the maximum threshold set by Indonesian Presidential Decree No. 22 Year 2021 (0.03 mg/L).

In case of sediment sample, Pb concentration in KR is higher compared to KB, which could be associated to the slow movement of the river current in KR. The slow current promotes heavy metals deposition in the sediments, as suggested by other studies (Qiao et al., 2020). Pb concentration in the sediment in KR has not preceeded the maximum thresholds from the Ontario Sediments Quality Guidelines (250 mg/kg). Our study also found that Pb contents in *F. ater* collected from KB was relatively higher than that of KR. It is in agreement with the previous study where heavy metal accumulation in living organism is affected by the amount of waste released to the aquatic habitat (Zhou et al., 2020). Some parameters determining the concentration of Pb contamination in the *F. ater* tissue include heavy metal concentration and exposure duration, and other environmental factors. Our present study suggested that the *F. ater* samples were contaminated by Pb with concentration exceeding maximum threshold from the World Health Organization (0.5 mg/kg) (Li et al., 2020).

Table 1. Obtained Pb concentrations in water, sediment, and *F. ater* samples collected from KB and KR based on AAS analysis.

Labels	Pb concentrations			Zn concentrations		
	Water (mg/L)	Sediment (mg/kg)	<i>F. ater</i> (mg/kg)	Water (mg/L)	Sediment (mg/kg)	<i>F. ater</i> (mg/kg)
KB 1	0.0058	44.91	5.41	0.0041	48.22	91.38
KB 2	0.0060	48.22	4.19	0.0048	78.06	35.24
KB 3	0.0063	47.93	8.13	0.0066	55.92	21.38
KB 4	0.0059	28.60	2.09	0.0057	47.33	10.92
KB 5	0.0065	27.85	2.51	0.0045	56.05	15.83
KB 6	0.0087	23.74	3.94	0.0048	33.33	18.45
KR 1	0.0051	74.79	2.63	0.0029	75.98	98.57
KR 2	0.0056	70.50	2.86	0.0031	148.69	69.94
KR 3	0.0059	57.25	6.15	0.0051	116.33	26.96
KR 4	0.0053	20.89	1.48	0.0048	123.04	18.06
KR 5	0.0061	34.48	1.71	0.0032	107.87	13.98
KR 6	0.0084	32.98	1.75	0.0038	46.18	16.07

Bold number is the highest in the same column

Table 2. Statistical tests comparing the averaged concentrations of Pb and Zn from different sampling locations and dates.

Heavy metal	Source of variation	df	SS	MS	F _{experimental}	F _{theoretical (95 %)}
Pb	Group	11	1256.74	114.25	1.05	2.27
	Treatment	2	13432.72	6716.36	61.80	3.93
	Error	21	2282.22	108.67		
	Total	34	16971.69			
Zn	Group	11	9893.47	899.40	1.16	2.27
	Treatment	2	36701.65	18350.80	23.69	3.93
	Error	21	16265.99	774.57		
	Total	34	62861.12			

df = degree of freedom; SS = sum of squares; MS = middle-square

3.2. Zn contents in water, sediment, and *F. ater*

The Zn concentrations in water, sediments, and *F. ater* collected from KB and KR could be seen in Table 1. Similar to Pb concentrations, Zn concentrations in KB were significantly higher than in KR. It is corroborated by the data shown in Table 2, with experimental *F* value is higher than that of theoretical ($23.69 > 3.93$). The presence of Zn in the water from both KB and KR is still below the maximum threshold under Indonesian Presidential Decree No. 22 Year 2021 (0.05 mg/L). Other than the foregoing anthropogenic sources, Zn could be originated from natural sources including the erosion of Zn-containing rocks around the river (Zhang & Wang, 2020).

This research revealed the Zn contents in *F. ater* is very concerning. For consumption, Zn concentration should be below 50 mg/kg in *F. ater*, where the concentration obtained from this study reached more than 90 mg/kg. The average concentrations of Zn in *F. ater* collected from KR were higher than that of KB. It could be associated to high Zn concentration

in the sediment, in which it is higher in KR than in KB. The presence of Zn in the sediment allows higher Zn uptake to the *F. ater* via filtration and absorption. Zn content in sediment found in this study is still tolerable according to Ontario Sediments Quality Guidelines (820 mg/kg). Our findings are consistent to previous reports (Custodio *et al.*, 2020; Zhou *et al.*, 2020), where heavy metal concentration is enriched in the living tissue of aquatic biota. The retained Zn in the sediment is possibly ascribed to the different soil types and water pH from the two rivers, although further investigation needs carried out. Previous report suggests the mobility of Zn and Pb are sensitive to pH changes and soil types (Mariussen *et al.*, 2017).

3.3. PCA analysis

The PCA results on the distribution pattern of Pb concentrations in water, sediment, and *F. ater* collected during the research timeframe (six months) showed the relationship between the tested variables. Significance of the correlation increases depending the position of the variable to the main component's axis.

Position or coordinate describes the direction of correlation. If the position is close to the main component (angle $\leq 45^\circ$), the variable has a positive correlation. Correlation formed between variables obtained from different sampling time (for example KR 1 and KR 2) were excluded because of the weather changes.

The correlation between the Pb concentration in water at KB 6 and KR 6 is indicated strong and positive (Figure 1a). It suggests that higher aqueous Pb concentration at KB 6 increases that at KR 6. If we use the previous assumption that the contamination is from the cement industry, KR should not be positively correlated as it is far from KB and the industry. In this sense, there might be similar factors affecting the the distribution of Pb in both rivers. Meanwhile, the correlation for Zn contaminations in water, sediment, and *F. ater* are negative; meaning the three variables do not affect one another (Figure 1b). Pb contents in sediment and *F. ater* are negatively correlated, suggesting that the Pb uptake by *F. ater* is not mainly influenced by Pb concentration in the sediment. The PCA also revealed that the Pb contents in sediment and in water are negatively correlated. It could be ascribed to the likelihood of Pb to remain deposited in the soil which could be affected by the physical (i.e. current and temperature) and chemical (i.e. pH, dissolved oxygen, and concentrations of phosphate, nitrite, nitrate, and sulphate) properties of the river water (Lee et al., 2017). Pb concentrations in *F. ater* are the highest at KB 3 and KR 3 sampling points due to the proximity of variables (in accordance to the data in Table 1).

Similarly to Zn concentrations in KB 1 and KR 1 *F. aters* that are found to be the highest with postive correlation. To observe the interaction beteen Pb and Zn and its effect againts their pollution and characteristics, PCA plot was constructed based on Pb and Zn contents in water, sediments, and *F. ater* samples from both sampling locations (Figure 1c). The results suggest that Pb and Zn concentrations in sediment samples collected from KR 1 have positive correlation, indicating variables are influenced one another. This finding could be explained by the fact that the mobilities of deposited Pb and Zn are influenced by the same factor (Mariussen et al., 2017).

Furthermore, Zn concentrations found in *F. ater* and sediment samples collected from KR1

also possess a positive correlation; suggesting that Zn uptake by *F. ater* is mainly from the sediment. This finding could explain the previously negative correlation between Pb in sediment and *F. ater*, where Zn absorption acts antagonistically to Pb absorption. It is further supported by the fact that Zn concentration in *F. ater* is dramatically higher than of Pb concentration. Antagonistic interaction between Zn and Pb absorptions have been recorded in plants (Musielńska et al., 2016; Ongh et al., 2013). This is the first study that suggests the antagonism between Zn and Pb absorptions in *F. ater*.

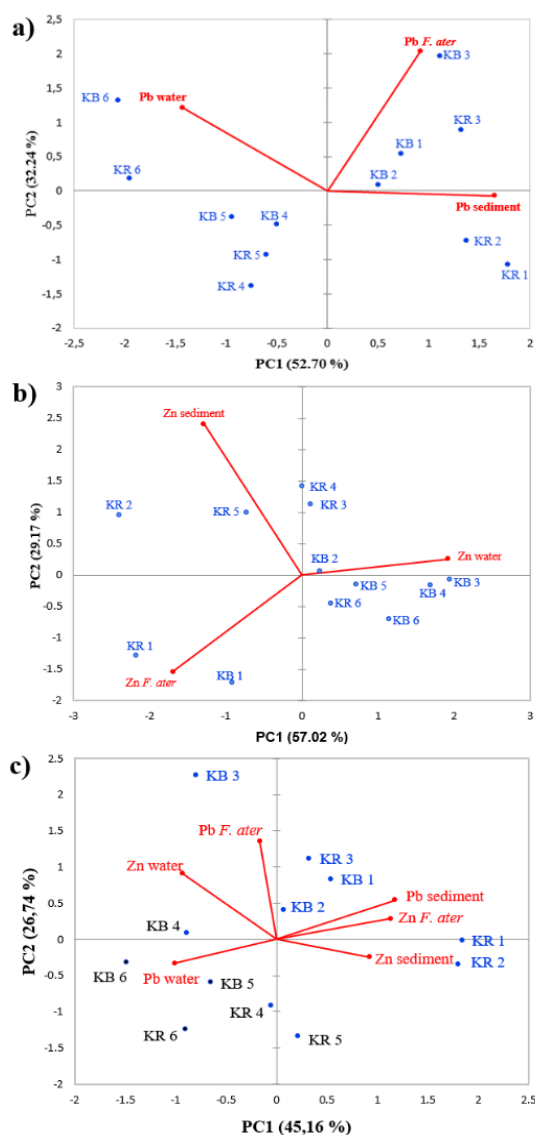


Figure 1. PCA results for Pb (a), Zn (b), and Zn and Pb (c) contents in samples collected from KB and KR.

As a limitation, we did not determine the correlation of Pb and Zn with other heavy or

light metals. The inclusion of other elements could help to understand the profile of Pb and Zn pollution in KB and KR. The use of multielemental analysis such as Laser-Induced Breakdown Spectroscopy (LIBS) could be a solution (Iqhrammullah et al., 2021; Iqhrammullah et al., 2021). Moreover, an investigation correlating the weather and other relevant properties (such as pH, current speed and so forth) needs carried out in the future.

4. Conclusions

The concentrations of Pb and Zn in water, sediment, and *F. ater* were dependent to the sampling location and date. In water sample, the concentrations of Pb and Zn were found higher in the river near the industrial area (KB) than in KR. However, the heavy metal concentrations in sediment sample were the otherwise. As for *F. ater*, Pb concentration was significantly higher in samples collected from KB than that of KR. On contrary, Zn in *F. ater* from KB was significantly lower compared with that of KR. These differences might be attributed to the physical (*i.e.* current and temperature) and chemical (*i.e.* pH, dissolved oxygen, and concentrations of phosphate, nitrite, nitrate, and sulphate) properties of the river; confirmed by the multiple correlations generated by PCA. The correlation also reveals possible antagonistic accumulation between Pb and Zn in *F. ater*. Additionally, nearby industrial activities are not the sole contributor influencing the distribution of Pb in the water. Our study is limited in identifying what factors involved in the distribution of Pb and Zn pollution. Hence, more investigation is required focusing on the identifications of the possible involved factors.

Acknowledgements

Authors acknowledge all contributors who have made their contribution during the research, especially those who have provided some assistance during the data analysis.

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