Distribution and Contamination Level of Cuprum (Cu) and Plumbum (Pb) in Bulk Sediments of the Bangka Island

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Abstract

The distribution and enrichment of heavy metals in sediments will affect the life of the organisms that lives in it. The purpose of this study is to explain the enrichment and contamination levels of heavy metals Cu and Pb in bulk sediments in the northern and southern parts of Bangka Island. This research was conducted in August - September 2019, using a purposive sampling method. Heavy metal analysis using the Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES) instrument. The results showed that the concentration of the heavy metal Pb in the sediment bulk of northern Bangka Island (Kelabat Bay) was higher than that of South Bangka Island. The concentration of heavy metal Cu at 8 (eight) sites is still below the quality standard (18.7 mg.kg^{-1}) even if the heavy metal Pb at stations 3, 4, and 5 in the waters of the northern half of Bangka Island (Kelabat Bay) is reported to have exceeded the quality standard limit (30.2 mg.kg^{-1}). The highest level of enrichment of heavy metal Pb occurs at station 4 with a value of EF_62.88, and is categorized as very high enrichment and a contamination factor (CF) value of 2.24 (medium category). This condition is due to station 4 being located in Kelabat Bay (semi-enclosed area) with unstable water conditions due to the influence of many activities. The results of this study can be used as input for local governments for the management of water areas on Bangka Island.

Keywords: *Contamination, Enrichment, Heavy Metals, ICP-OES, Sediments*

Introduction

Bangka Island is an archipelago known for abundant tin (Sn) minerals and rich marine resources. In the northern (Kelabat Bay) and the southern (Tukak Beach, Lepar Island, Anak Air Island) parts, it is possible to discover an abundance of bivalves and gastropods. Unconventional Sn mining activities in Kelabat Bay introduced heavy metals to bulk sediments (Komalasari *et al.*, 2019; Tawa *et al.,* 2019; Nugraha *et al.,* 2022; Umroh *et al.,* 2022). Moreover, these mining activities elevated water turbidity and the presence of suspended particles (Adi *et al*., 2016; Nurtjahya *et al*., 2017). Bulk sediments composition resulting from mining primarily consists of muddy sand (Umroh *et al*., 2022). Mud, characterized by fine texture and a substantial particle surface, possesses heightened density and metal-binding capabilities (Maslukah 2013), leading to increased concentrations of heavy metals in water. These heavy metals in bulk sediments can disperse due to natural processes such as currents, tides, and waves. Water surrounding Bangka Island is situated in the monsoon wind flow area, positioned between the Java Sea and the Natuna Sea. The monsoon wind influences current directions (Pamungkas, 2018), subsequently impacting the distribution and accumulation of heavy metals in water sediments.

The increased presence of heavy metals in sediments causes a significant threat to organisms, particularly those belonging to the highly toxic class B heavy metals, including Hg, lead (Pb), Sn, and cuprum (Cu) (Budiastuti *et al*., 2016; Pertiwi *et al*., 2021). Elevated levels of Cu and Pb in sediments can disrupt

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https://ejournal.undip.ac.id/index.php/ijms
DOI: 10.14710/ik.ijms.28.3.278-288
Received : 10-01-2023
Accepted : 23-08-2023

the lives of benthic organisms when these heavy metals surpass established quality standards. These conditions can likely lead to larval mortality and negatively affect the growth of mollusk shells (Umroh *et al*., 2021; Nugraha *et al*., 2017; Riani *et al*., 2017). It was also observed that tailings, a byproduct of Sn mining containing minimal organic carbon (C), nutrients, and microorganisms (Nurtjahya *et al*., 2017; Sukarman and Gani 2017), impact the quality of the food source for macrobenthos habitat substrates.

Numerous studies showed the adverse impact of mining activity waste on water quality (Muslih *et al*., 2014; Komalasari *et al*., 2019; Adibrata *et al*., 2021). High turbidity reduces dissolved oxygen (DO) levels and water clarity, thereby slowing down photosynthesis in water. Physico-chemical factors in water, such as current velocity, salinity, pH, DO, and metal sources, play a crucial role in influencing the distribution of heavy metals in water (Liu *et al*., 2016), thereby leading to a reduction in water quality.

Previous studies examined the presence of heavy metals in water of Bangka Island. In general, water and sediment samples have higher levels of heavy metals, including iron (Fe), Cu, Pb, and cadmium (Cd). It is crucial to acknowledge that the concentration of heavy metals in water exceeds typical levels. On the other hand, sediments are primarily composed of Fe, followed by zinc (Zn), chromium (Cr), Cu, Pb, and arsenic (As) in descending order of abundance.

Sediment samples from the discharge area of Kolong Retention Kacang Pedang contain no Cu or Zn (Irvani and Pitulima, 2016). According to the Risk Assessment Code (RAC), Zn has no significant risk at low levels, while Pb carries a low to high risk in the environment. Cu, on the other hand, presents no environmental risk, which raises concerns about the potential bioavailability of Pb and Zn in the inner bay water around Kalabat Bay (Nugraha *et al*., 2019). The accumulation of Pb in biota, such as shrimp, is closely linked to Pb content in water. The hepatopancreas, gills, and flesh of *Penaeus merguiensis* show the highest average Pb values, ranging from 0.1897 to 0.4064 mg.kg^{-1}, 0.2424 to 0.4780 mg.kg^{-1}, and 0.1348 to 0.1636 mg.kg^{-1}, respectively. Furthermore, average Pb concentration in the air and soil are recorded at 0.2624-0.5713 mg.L^{-1} and 0.2783-0.9760 mg.kg^{-1}, respectively. *Penaeus merguensis* shows a limited capacity for Pb accumulation, with a weekly intake ranging from 9.760 to 11.128 kg.day^{-1} (Tawa *et al*., 2019).

The potential for metal components from Sn mining regions to contaminate the food chain, affecting farmers and consumers, is a matter of concern. Various physical and chemical (physicochemical) methods, such as anion precipitation, electro-winning, electro-coagulation, cementation, reverse osmosis, and electro-dialysis, can be used to remove metal contamination. Biological methods, including phytoremediation, mycoremediation, and bacteria bioremediation, also offer solutions (Kurniawan and Mustikasari, 2019). Explorations of the Enrichment Factor (EF) across different regions in the Indonesian archipelago show that the concentration of heavy metals in sediments around Bangka Island are decreasing in correlation with the reduced water quality. These contaminants originate from a combination of anthropogenic and natural sources. Anthropogenic sources in water of Bangka Island include Sn mining tailings, emissions from Pb mining machinery and paint fumes. The results show the increasing concentration of Zn, Cu, Pb, Cd, and Sn due to ongoing natural resource activities (anthropogenic) in local communities and the interactions (Umroh *et al*., 2022; Nugraha *et al*., 2022).

Spatial distribution patterns indicate similarities in the levels of heavy metals in both seawater and sediment. In general, high metal concentration is observed in stations close to the soil surface, suggesting that human activities in the soil contribute to heavy metals enrichment (Harmesa *et al*., 2020). Surface marine sediments in Java show enrichment in Zn, nickel (Ni), Cu, As, and Cr, with moderate Zn contamination across the board (Pugung *et al*., 2018). The examination of the distribution of Cu, Fe, and Zn in Ketapang Harbour, shows varying concentration levels. In particular, Fe concentration is known to be high in most study areas. The EF index indicates an EF value >50 for Cu and Zn, mainly due to the high background value of Fe used in the absence of a reference for normal Fe concentration (Yona *et al*. 2018). In Lampung Bay Industrial Area, Cu and Pb concentration in water remains below established quality standards. The results suggest that the enrichment of Pb and Cu in water primarily comes from natural processes, meeting the minimum required enrichment class (Permata *et al*., 2018).

Heavy metals (Fe, manganese (Mn), Cr, Zn, Ni, Pb, Cu, Co, and Cd) in the surface sediments of the Kochi estuary on the Southwest coast of India have both natural and anthropogenic origins. While Cu and Ni originate from natural weathering processes, Cd, Cr, Zn, and Pb result from anthropogenic activities. The relationship between organic carbon and metallic silts and clays shows the significance of fine sediments and organic matter as carriers of these metals (Salas *et al*. 2017).

Previous studies did not extensively focus on establishing criteria for assessing Cu and Pb contamination levels in bulk sediments due to the influence of heavy metals inputs from both mining and non-mining sources. Therefore, this current study aims to determine the enrichment and contamination levels of Cu and Pb in bulk sediments of the northern and southern parts of Bangka Island. The obtained results can provide valuable insights to local governments for formulating effective management policies.

Materials and Methods

This study was conducted between August and September 2019 at nine stations in water of Bangka Island. Bulk sediments sampling was categorized into two clusters, namely the northern part, with active mining activities, and the southern part, where there were no mining activities. In the northern part, sediment samples were collected at five stations, which included Station 1 (mouth of Kelabat Bay), Station 2 (Romodong Beach), Stations 3 and 4 (Cupat Beach), known for unconventional Sn mining activities, and Station 5 (Semulut Beach). In the southern part, sediment collection was carried out at four stations, consisting of Stations 6 (Anak Air Island), 7, 8 (Tukak Beach), and 9 (Anak Air Island), as shown in Figure 1.

Sediment texture analysis and measurement of heavy metals concentration in bulk sediments

Bulk sediments grain size was analyzed using a sieve shaker, and the percentage of each fraction was determined and categorized according to Wentworth's classification (Romano *et al.*, 2017). Pb and Cu concentration were measured in dry bulk sediments samples and was analyzed in accordance with established procedures (APHA, 2012a; Suteja *et al.*, 2020). The dry sediment samples were pulverized and homogenized, and then 0.5 g sediment sample was placed in a vessel tube where nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) were added. The concentration of heavy metals in the prepared solution of dry bulk sediments samples was measured using an Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES) instrument.

The process of measuring heavy metals concentration comprised washing and heating the ICP-OES instrument for 2 hours by flowing plasma from ionized argon (Ar) gas. After the instrument was ready for measurement, the standard series solution was mixed with bulk sediments sample and placed in the tube, which was then positioned on the autosampler rack in a clockwise direction. The standard solution and sample solution were absorbed alternately (sequentially), with each injection absorbing 0.5 ml, which was subsequently flowed through the capillary tube to the nebulizer. In the nebulizer, the sample was converted into an aerosol, and atomic excitation occurred. The constituent atomic counting took place at a temperature between 6000-8000ºC (Skoog *et al.,* 1998).

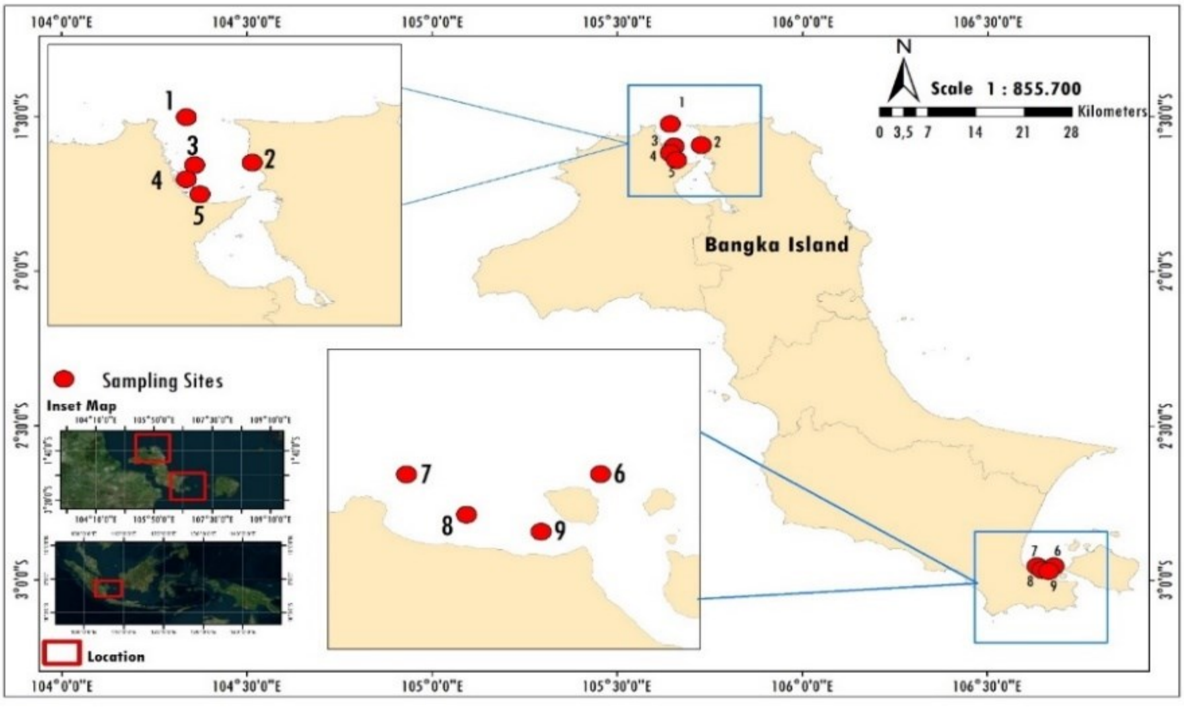


Figure 1. Sediment collection stations in the northern and southern of Bangka Island water

Analysis of Total Suspended Solids (TSS) in water

Analysis of total suspended solids (TSS), according to APHA (2012b) guidelines, included filtering a volume of 1000 ml seawater through a 0.45 μm filter paper (cellulose filter) with a known initial dry weight. The filtered cellulose was then dried for 1 hour in an oven set at 103-105°C, cooled in a desiccator, and weighed to obtain the final weight.

Distribution map analysis of Cu and Pb concentrations in sediments

The distribution map of Cu and Pb concentration in bulk sediments was processed using ArcGIS 10.4.1 and interpreted with different color scales. These color scales were in the form of displays and the results were analyzed descriptively (Petus *et al.*, 2019; Akhrianti *et al.,* 2023).

Analysis of the Enrichment Factor (EF) and the level of heavy metals contamination in bulk sediments

EF was used to determine the origin of heavy metals, whether from anthropogenic (human-made) or natural sources. The reference metal from the Earth's crust for calculating EF values was Mn. The Earth's crust values for Cu= 55, Pb= 12.5, and Mn= 950, as established by Taylor in 1964, were used. Based on Barbieri's work in 2016, EF was calculated using the following equation:

$$\text{EF} = \frac{X_{(s)}/Mn_{(s)}}{X_{(ec)}/Mn_{(ec)}}$$

Note: X(s) = Cu, Pb, and Mn (s) = Mn levels in sediments measured during the study, X(ec), Mn(ec) = concentration of Cu, Pb, Mn in earth crust. The EF criteria, as outlined by Barbieri (2016), are as follows: EF value between 0.5 and 1.5 indicates heavy metals are naturally occurring. EF ≥ 1.5 suggests that some or all heavy metals come from human-made sources. EF criteria <2 indicate minimal enrichment. EF criteria between 2 and 5 indicate moderate enrichment. EF criteria between 5 and 20 indicate quite high enrichment. EF criteria between 20 and 40 indicate very high enrichment.

The contamination level described the impact of anthropogenic contaminants on the environment using the contamination factor (CF). The calculation of CF for metals was performed based on the formula below (Ahmed *et al.,* 2018):

$$\text{CF} = \frac{X_{(s)}}{X_{(ec)}}$$

Note: X(s) = Cu, Pb metals in bulk sediments measured during the study; X(ec) = concentration of Cu, Pb in earth crust. The criteria for contamination level are defined as low (CF<1), moderate (1≤CF<3), high enough (3≤CF<6) and very high (CF≥6).

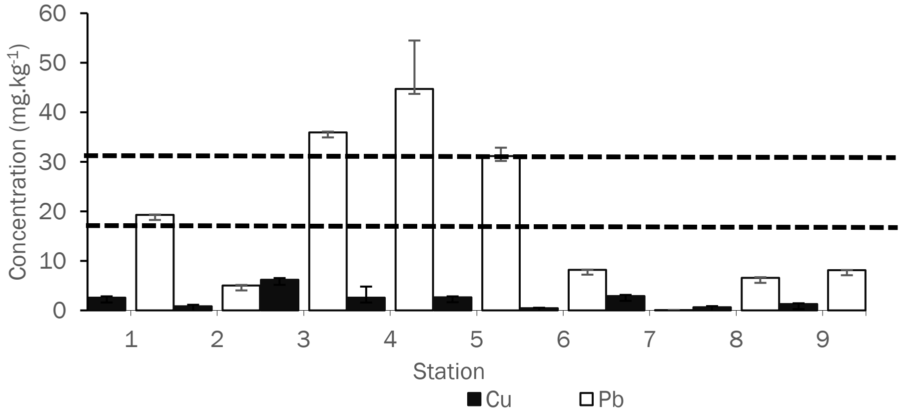
Result and Discussion

In the northern part of Bangka Island, which excluded Kelabat Bay, Stations 1, 2, 3, 4, and 5 exhibited higher concentrations of Pb in bulk sediments compared to Stations 6, 7, 8, and 9 located in the southern Bangka Island. One station in the northern part slightly exceeded the CCME quality standard (CCME, 1991). However, according to the contamination level criteria, the concentration still fell in the relatively low contamination range, as shwon in Table 2, making it suitable for sustaining biota life. The concentration of Cu at eight stations was below the quality standard shown in Figure 2. Large populations of the Dog conch (*Laevistrombus canarium*) were observed from May to July (Umroh *et al*., 2021), and Anadara granosa was still present in water of the northern part (Kelabat Bay) (Selpiani *et al*., 2015). This suggested that endemic macrobenthos could still thrive and reproduce, even though the growth rate might have been lower than macrobenthos in the southern part, such as the growth of the Dog conch (Supratman *et al*., 2019; Umroh *et al*., 2021). Pb concentration at Station 4 was higher than in previous studies, even though it was not measured in bulk sediments, as shown in Table 1. This indicated that when measurements had not been taken in bulk sediments, the high concentration of Pb at Stations 3, 4, and 5 would likely have been even more significant.

Stations 3 and 5 with a dominant sand texture of 70.61% as shown in Figure 6, had a limited capacity to retain heavy metals. These stations, particularly 3, 4, and 5, experienced higher levels of community activities, including mining and non-mining, when compared to other stations. As a result, the activities led to increased levels of Pb in bulk sediments. According to the EF value in Table 2, Pb concentration at Stations 3, 4, and 5 indicated an extremely high enrichment, strongly associated with anthropogenic sources in Kelabat Bay (Umroh *et al.,* 2022). Communities in the coastal area were known to use materials containing Pb, found in gasoline or diesel fuel, which contained tetraethyl-Pb and tetramethyl-Pb (Stancheva *et al*., 2013). Smoke generated by combustion in both mining and non-mining machinery, which contained Pb, would bind to atmospheric particles and, when combined with rain, settle into water (Takarina *et al.,* 2013**;** Umroh *et al.,* 2022). Heavy metals from the smoke descended into water, formed aggregates, and settled at the bottom, thereby increasing Pb concentration in bulk sediments.

Station 4 had the highest Pb enrichment level with EF and CF values of 62.88 and 2.24, respectively, as shown in Table 2. Heavy metals found in Kelabat Bay, which is a semi-enclosed area with unstable water conditions and influenced by numerous community Sn mining activities, could easily spread due to currents and waves. This led to an increase in Pb concentration at Stations 3 and 5, as shown in Figure 4.

The distribution of heavy metals from Station 4 was believed to be responsible for the enrichment observed at Stations 3 and 5. Pb enrichment values at Stations 3 and 5 were 35.52 and 27.93, respectively, both categorized as very high enrichment. However, Cu enrichment was minimal, with CF < 1 (Table 2.), indicating low contamination of Cu in bulk sediments of the northern and southern parts. Pb contamination factor in bulk sediments in Kelabat Bay ranged from 0.25 to 2.24, classifying it as a low to moderate level. Based on the contamination factor values, organisms could live normally, but prolonged exposure might not result in optimal growth (Umroh *et al*., 2022). This observation



Pb : 30.2 mg.kg-1 (ISQG) CCME (2002); Cu : 18.7 mg.kg-1 (ISQG) CCME (2002)

Figure 2. Concentration of heavy metals Cu and Pb in the bulk sediments of Bangka Island waters

Table 1. Differences in heavy metals concentrations between bulk and non-bulk sediments in the northern and southern parts of Bangka Island

Location	Time	Cu (mg.kg-1)	Pb (mg.kg-1)	Reference
Northern and Southern Bangka water	August 2019	0.407-6.157	0.06-44.71	This study
Matras seawater	March 2021	0.01 (0.01)	0.12-0.18 (0.15)	Nugraha *et al.* (2022)
Inner Kelabat Bay	March 2018	0,00160-0,00219	7.15–7.73	Komalasari *et al*. (2019)
Cimanuk sea water	Mei 2017	12.36-54.0 (28.75)	6.43-15.72 (12.24)	Harmesa *et al.* (2020)
Lampung Bay	Mei 2016	1.02 -38.75	15.41-32.66	Permata *et al.* (2018)
Ambon Bay	Mei 2016	-	0.13-0.75	Sukaryono dan Dewa (2018)
Kelabat Bay	April 2018	0.16-9.54 (4.39)	8.86-29.21 (16.85)	Nugraha *et al.* (2019)
Jakarta Bay	September 2014	11.42-67 (33,13)	24.86-59.32 (38,53)	Kusuma *et al*. (2015)
Gresik, East Java	February 2012	23.7-234 (85.5)	1.7-12.7 (4.29)	Lestari dan Budiyanto (2013)
SQGs				
CCME (2002)	ISQG	18.7	30.2	
	PEL	108	112	
ANZECC/ARMCANZ Guidelines (2000)	Low	65	50	
	High	270	220	

- = The study were not measured in specific parameter, SQGs = Sediment Quality Guidelines, ISQG = Interim Sediment Quality Guidelines, PEL = Probable Effect Level was due to the toxic nature of Pb, which could likely interfere with enzyme function and growth (Riani *et al.,* 2018; Umroh *et al.*, 2021).

The EF_Pb enrichment level in the northern part at Stations 3, 4, and 5 met the criteria for very high enrichment in Table 2, primarily due to a combination of anthropogenic inputs from various activities on the island. The anthropogenic sources included ballast water disposal activities, diesel fuel spills, gasoline from fishing boat engines, and community Sn mining pontoon engines. Additionally, the enrichment of Pb from natural sources occurred due to drilling in the earth. Kelabat Bay, being the center of ongoing community mining activities, contributed to this natural enrichment of Pb (Umroh *et al*., 2022).

Geologically, there is a mineral called galena (PbS) that forms alongside cassiterite (SnO_2), although in relatively small quantities. The process of weathering and sedimentation led to the precipitation of PbS, which was then mixed with secondary Sn in ancient river channels. It was physically enriched due to its specific gravity (Powell *et al.,* 2021). Mining activities, comprising the excavation of earth layers, exposed the galena content in secondary Sn deposits, which reacted in various locations. Pb was resistant to dissolution and tended to bind to particles, sinking and accumulating at the bottom, consequently increasing the enrichment in bulk sediments of Kelabat Bay.

The presence of numerous mining activities in water, together with the disposal of sludge waste and the specific current conditions in semi-enclosed areas, led to increased turbulence, resulting in elevated TSS levels, as shown in Table 3. These TSS concentrations exceeded the standards set by government regulation number 22/2021, which was designed to standardize environmental protection and management, including the prevention of harm to the sea environment and the biota inhabiting the place. This situation impacted marine life because suspended particles bound with heavy metals, thereby forming aggregates that settled and increased the heavy metals content in sediments. Additionally, at Station 4, the predominant substrate texture was 84.63%, as shown in Figure 6. This texture was characterized by fine particles with a broad surface area, high density, and a strong ability to bind with metals, resulting in higher heavy metal content in the mud texture.

Various factors including temperature, salinity, pH, and DO influenced the solubility of heavy metals in water. The recorded water temperature averaged between 29 and 31°C, salinity ranged from 30-34‰, and DO levels were measured between 3.45 and 6.20 mg.L^{-1}. These conditions were still considered in the normal range, as defined by government regulation number 22/2021. However, rising temperatures and decreasing salinity in water led to increased solubility of Cu and Pb metals, resulting in higher concentrations of these metals in water. Lewis *et al*. (2016) added that the toxicity of Cu metal increased with every 10°C rise in temperature.

Table 2. Enrichment factor (EF) and contamination factor (CF) of Cu and Pb in bulk sediments in the northern and southern parts

Station	Enrichment Factor (EF)				Contamination Factor (CF)			
	Cu	Category	Pb	Category	Cu	Contamination level	Pb	Contamination level
1	0.35	Minimum enrichment	5.82	Moderately high enrichment	0.06	Low (CF<1)	0.96	Low (CF<1)
2	0.25	Minimum enrichment	3.49	Moderate enrichment	0.02	Low (CF<1)	0.25	Low (CF<1)
3	8.96	Moderately high enrichment	35.52	Very high enrichment	0.14	Low (CF<1)	1.80	Moderate (1≤CF<3)
4	1.62	Minimum enrichment	62.88	Extremely high enrichment	0.06	Low (CF<1)	2.24	Moderate (1≤CF<3)
5	1.05	Minimum enrichment	27.93	Very high enrichment	0.06	Low (CF<1)	1.56	Moderate (1≤CF<3)
6	0.17	Minimum enrichment	7.54	Moderately high enrichment	0.01	Low (CF<1)	0.41	Low (CF<1)
7	1.90	Minimum enrichment	0.07	Minimum enrichment	0.06	Low (CF<1)	0.00	Low (CF<1)
8	0.39	Minimum enrichment	9.39	Moderately high enrichment	0.01	Low (CF<1)	0.33	Low (CF<1)
9	0.51	Minimum enrichment	7.43	Moderately high enrichment	0.03	Low (CF<1)	0.40	Low (CF<1)

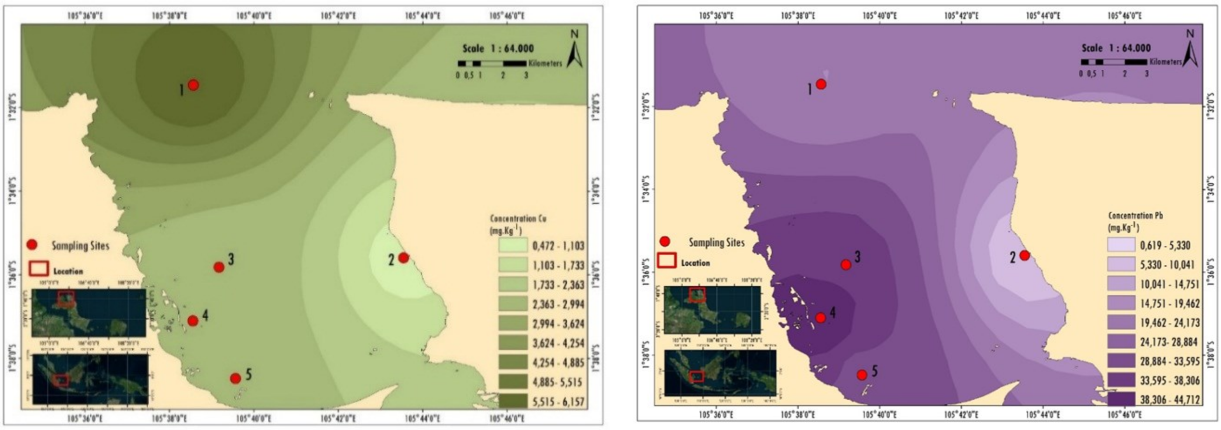


Figure 4. Concentration of Cu (left) and Pb (right) in bulk sediments in the northern part of Bangka Island (Kelabat Bay)

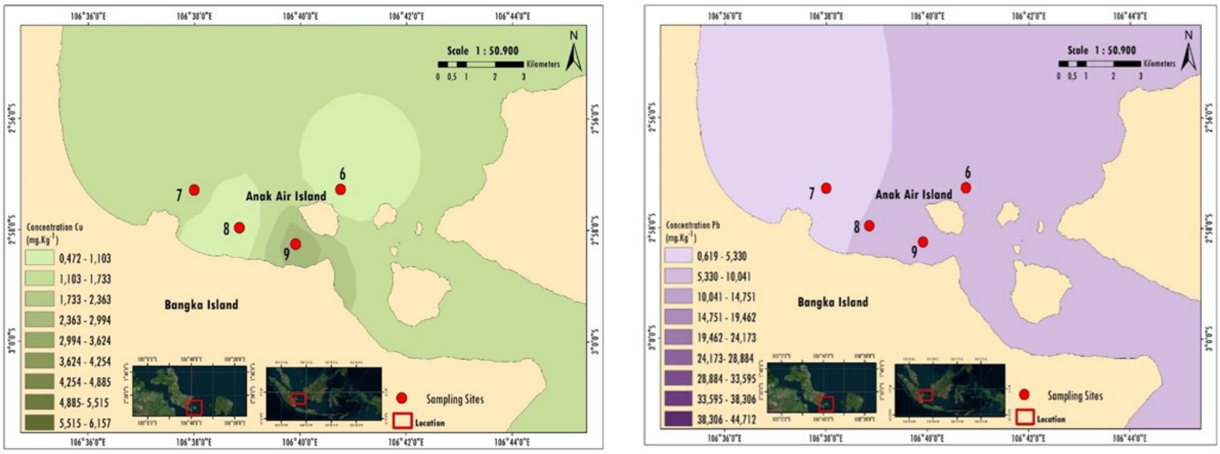


Figure 5. Concentration of Cu (left) and Pb (right) in bulk sediments in the southern part of Bangka Island

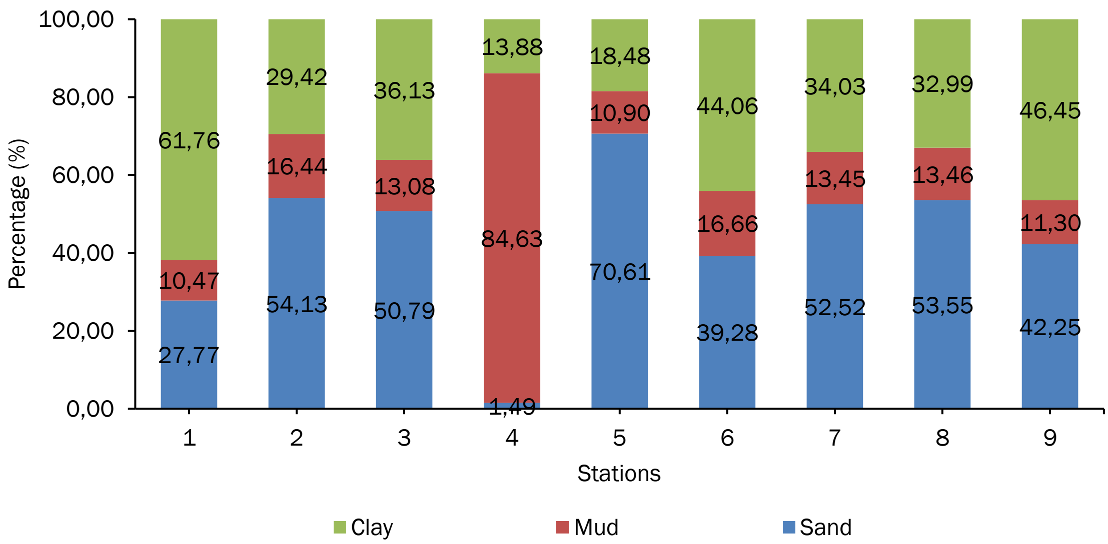


Figure 6. Sediment texture with sediment fraction sizes: clay (<0.002 mm), silt (0.06-0.002 mm), and sand (2-0.06 mm) in Bangka Island water

Table 3. Measurement of physical-chemical parameters of the northern and southern parts

Stations	Temperature (°C)	Depth (m)	Current speed (m.s^{-1})	TSS (mg.L^{-1})	pH	Salinity	DO (mg.L^{-1})
1	30-31°C	10-80	0.45-0.5	28.5-30.5	7.8-7.82	31	4.50-4.62
2	29-30°C	0.9 - 1	0.3-0.45	27.2-34	7.8-7.94	31	4.50-4.52
3	30-31°C	1.8-3	0.4-0.5	28-30	7.10-7.20	30	3.45-4.20
4	30-31°C	1.5-1.8	0.35-0.45	28.0-28.8	7.10-7.20	30	4.10-4.20
5	30-31	2.5-3	0.4-0.45	36.5-50-5	7.10-7.10	30-31	4.32-4.50
6	30-31	1.5-1.7	0.4-0.45	26.8-32.5	7.69-7.82	34	6.10-6.20
7	30	1.6-2.2	0.45-0.6	26.6-29.2	7.62-7.73	34	6.10-6.20
8	29-30	0.6-1.1	0.8-1.2	26.6-29.2	7.62-7.73	34	5.20-6.00
9	30-31	1.5-1.7	0.4-0.5	26-30.5	7.60-7.80	34	5.60-6.10

*Indonesian Government Regulation Number 22/2021

Several studies showed an increase in Cu values attributed to temperature elevation and decreased water salinity (Sari *et al*., 2017; Setyaningrum *et al*., 2019). Moreover, the observed pH levels fell in the neutral range (pH 7–7.9), which stabilized metal solubility and led to the formation of organometallic complexes. It should be acknowledged that these complexes eventually precipitated at the bottom of water. In environments with slightly alkaline pH (7.5-8.5), most metals namely Pb were found in the dissolved Pb (OH)+ form, rather than as $PbCl_2$ or $PbCO_3$. With increased water hardness, heavy metals tended to form complex compounds and precipitate on substrates, thereby reducing their toxicity (Neff 2002; Sanusi 2006). These conditions maintained the aquatic environment as a suitable habitat for various organisms.

Conclusion

In conclusion, Cu enrichment level in water of the northern part fell in the minimum to quite high category, while Pb enrichment level was very high, accompanied by a low to moderate level of contamination. In the southern part, Cu enrichment was in the minimum category, and Pb enrichment was in the minimum to quite high category, with a low level of contamination.

Acknowledgment

The authors are grateful to the University of Bangka Belitung, the Institute for Research and Community Service (LPPM), the Department of Marine Science, and the lecturers of Marine Sciences and Chemistry for their valuable cooperation and assistance during the investigation. This study was funded by the Lecturer Research Program (PDTU) at Bangka Belitung University, under contract number 189.D/UN50.3.1/PP/2019.

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