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Distribution and Contamination Level of Heavy Metal Iron (Fe) in Benanga Reservoir Sediment, Samarinda City, East Kalimantan

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Abstract

This study was conducted on the sediments of Benanga Reservoir, Samarinda, East Kalimantan. The purpose of the study was to determine the content and distribution pattern of heavy metal iron (Fe) in the sediments of Benanga Reservoir, as well as its potential as a contaminant that has an impact on the environment. Sampling was carried out by core drilling and determination of Fe content in sediment through laboratory analysis using the XRF method. The results of the analysis showed that the average content of Fe metal in the sediment deposits of Benanga Reservoir increased from the bottom layer (A) to the middle layer (B) and continued to the top layer (C). The Fe content in the sediments of Benanga Reservoir far exceeded the toxicity classification standards for heavy metals in sediments according to the USEPA standard (1977), so the distribution of "heavily contaminated" Fe contamination was spread throughout the sediment deposits of Benanga Reservoir. The controlling factors for the amount of Fe content in the sediment deposits of Benanga Reservoir are current dynamics, sediment texture, organic matter content, and sediment deposition velocity.

Keywords

Bottom Sediment, Iron, Reservoir, Contamination Index

1. Introduction

Reservoir sedimentation is the process of sediment accumulation from rivers

that enter reservoirs gradually. The intensity of sedimentation in reservoirs depends on land erosion in the reservoir catchment area, while the intensity of soil erosion is the result of a combination of natural factors and various human activities [1]. Sediment is an important environmental medium that functions as a reservoir and secondary source of pollutants [2]. These sediments contain organic and inorganic materials, both from rock weathering and from anthropogenic waste, which flow with water. In stable conditions, the accumulation of pollutants in sediments does not pose a threat to the aquatic environment, but pollutants can be released into the water during floods or other work activities such as reservoir dredging that cause the unloading of bottom sediments, as happened in the Benanga Reservoir in 2021 to 2022 [3] (**Figure 1**). Thus, reservoir sedimentation can be a serious problem that impacts water management, flood control, and energy production [4]. Contaminants are not all fixed in the crystal structure of the sediment, and their mobilisation to the water phase can occur under physicochemical changes of the surroundings [5]. Metals are partitioned into several chemical forms, where some are more labile during physical and chemical fluctuations. Variations of the aquatic system are caused by natural and human activities like dredging and use of engines, or under environmental changes such as fluctuation of salinity, pH, redox potential and ionic strength [5].

Figure 1. Benanga reservoir dredging project in 2021 - 2022 (Historical Imagery Google Earth, 2024).

One of the most dangerous types of pollution in aquatic systems is the discharge of heavy metals. The increasing use of heavy metals in industry and other activities considered essential for modern human life has resulted in modifications in the natural geochemical cycles of these elements, which have caused several environmental problems. Determination of heavy and toxic elements allows the study of their distribution, pollution levels, and risk assessment in the studied ecosystem [6].

Heavy metals tend to signal potentially hazardous conditions for organisms, due to their persistence in the environment and toxicity above threshold levels. Heavy metals can be divided into two categories: (i) transition metals (e.g., cobalt, copper, iron, manganese, etc.) that are essential for metabolism at low concentrations but can be toxic at high concentrations and (ii) metalloids (e.g., arsenic, cadmium, lead, mercury, selenium, tin) that are generally not required for metabolic

functions but are toxic at low concentrations. The spatial distribution of heavy metals in sediments is of great importance in explaining the pollution history of aquatic systems. These processes are influenced by natural and anthropogenic factors, such as climate, weathering of parent rocks, industrial and domestic wastewater, aquaculture, and agriculture. It is important to distinguish between natural and human impacts on heavy metals in marine sediments. Finally, geoaccumulation indices, enrichment factors, and other contamination factor analyses have been applied to indicate the degree of contamination by heavy metals from lithogenic and anthropogenic sources [7].

Among the various types of heavy metals that are often found in reservoir sediment deposits is iron (Fe). Iron has received much attention because of its wide distribution and is one of the most common elements in the earth's crust [5]. Fe metal is an essential metal whose presence in certain amounts is very much needed by living organisms, but in excessive amounts it can cause toxic effects. Anthropogenic waste containing Fe heavy metal compounds is not only toxic to plants but also to animals and humans. This is related to the properties of heavy metals that are difficult to degrade, so they easily accumulate in the aquatic environment and their presence is naturally difficult to remove, can accumulate in aquatic biota including shellfish, fish and sediments, have a high half-life in the bodies of marine biota and have a large concentration factor value in the body of organisms. High Fe metal content will have an impact on human health, including causing poisoning (vomiting), intestinal damage, premature aging to sudden death, arthritis, birth defects, bleeding gums, cancer, kidney cirrhosis, constipation, diabetes, diarrhea, dizziness, fatigue, hepatitis, hypertension, insomnia. Most metals such as Fe, Pb, Zn, Al & Cu are easily dissolved and very mobile at pH < 5 and at low pH, elements such as Al, Mn and Fe will be toxic [8].

The aim of this study was to identify the content and distribution pattern of heavy metal Fe in Benanga Reservoir sediments, and to determine the potential of Fe as a contaminant that is dangerous to living things.

2. Experimental (Materials and Methods)

2.1. Research Area

The research was conducted in the period between June 2023 - April 2024 in the Benanga Reservoir (Karang Mumus Watershed) located in North Samarinda District, Samarinda City, East Kalimantan, with coordinates 0˚24'41.40"S - 0˚24'04.60"S and 117˚11'29.40"E - 117˚12'50.40"E. Physiographically, the Benanga Reservoir is the estuary of three rivers, namely the Pampang Kiri River, the Selindung River, and the Binanga River (**Figure 2**). The Pampang Kiri River is the main source of water supply entering the reservoir, because the Pampang Kiri River is permanent throughout the year, while the other two rivers are intermittent (episodic rivers). The results of bathymetric mapping in 2008 [9] and physiographic imagery of Benanga Reservoir from Google Earth in the 2010 - 2024 time span also show that the dominant river flow recorded at the bottom

of the reservoir comes from the Pampang Kiri River. One of the functions of Benanga Reservoir is as a flood controller because the water flow from Benanga Reservoir crosses the central area of Samarinda City via the Karang Mumus River.

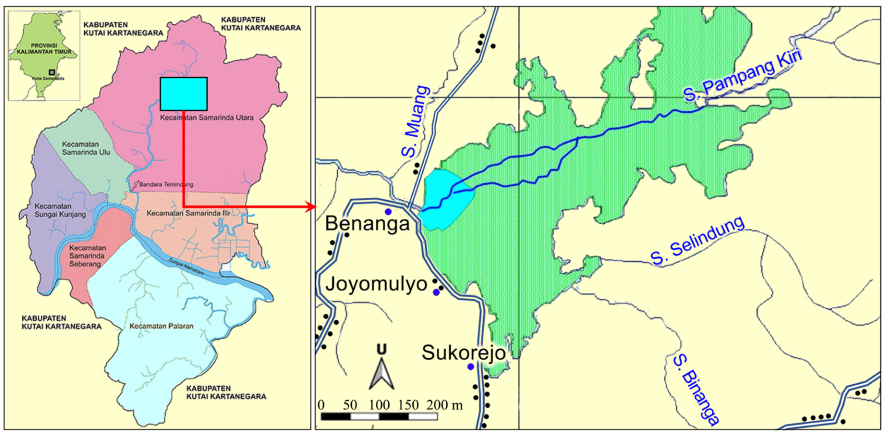

Figure 2. Benanga dam reservoir research location (Karang Mumus Watershed).

2.2. Materials

The research materials consisted of sediment samples collected from the bottom of Benanga Reservoir and Pampang Kiri River, Samarinda, Indonesia. Sediment sampling was carried out by core drilling from 11 shallow drilling points along the planned trajectory. Sampling points were generally divided into three groups, namely (i) three points in the upstream Pampang Kiri River to determine the background Fe content in the original sediment which then accumulated in the reservoir, (ii) three points in the downstream or estuary of the Pampang Kiri River to determine the Fe content before the sediment entered the Benanga Reservoir, and (iii) five points in the reservoir were made in a straight line perpendicular to the Pampang Kiri River as the main river to determine the distribution pattern of Fe in the reservoir vertically and laterally (**Figure 3**).

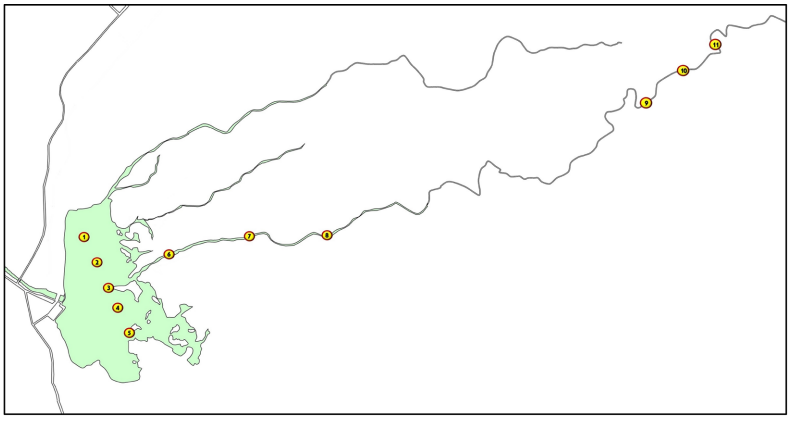

Figure 3. Schematic of data collection location points in Benanga Reservoir and background sediment deposits.

Sediment samples in the form of cores were taken from the surface of the reservoir bottom to the basement. Samples per layer from the drill core were prepared for analysis of Fe heavy metal content in the sediment. The scheme of data collection points in Benanga Reservoir and the background sediment deposits are presented in **Figure 3**.

2.3. Analysis Method

The surface layer of bottom sediment (up to 2 m depth) was collected using a specially designed drill bit to collect and hold the sediment core in the drill pipe. The sediment sample was removed and dried at room temperature until air dry. The sample was then prepared for XRF test which is one of the atomic spectrometry techniques with the ability to identify and quantify complex sample constituents such as those related to environmental problems, and is also able to provide high analysis speed answers. The basic principle of XRF is to excite the sample with electromagnetic rays (X-rays or gamma rays) or charged particles and directly measure the energy of the characteristic X-rays emitted by the sample in a detector. It is indicated for the identification and quantification of inorganic chemical elements in various types of samples and for various atomic numbers and concentrations [6].

Sample preparation was carried out by grinding and sieving through a 2 mm diameter sieve, then weighing 1 gram and placing it in disposable plastic cells equipped with mylar as a base, then pressing until the surface is homogeneous, then the sample was placed in the sample holder on the XRF Minipal 4 tool. The reading of the XRF analysis results graph is indicated by peak energy to identify an element, while the peak height/intensity indicates its concentration.

Table 1. Guidelines for iron (Fe) content in several sediment quality standards [10]-[13].

	Concentration (µg/g or ppm dry weight)		
	NEL	LEL	SEL
Ontario Sediment Quality Guidelines (%)	-	2	4
Sediment Quality Standard Values (Hayton, 1993)	-	20.000	40.000
	Non-Polluted	Moderately Polluted	Heavily Polluted
EPA (1977)	**<17.000**	**17 - 25.000**	**>25.000**
Ontario (MOE)	10.000		
Screening Level	20.000		

Keterangan: NEL (no effect level), LEL (lowest effect level), SEL (severe effect level).

The results of the analysis in the form of iron (Fe) content data in each sample were then processed to determine the vertical and lateral distribution patterns of

iron content in the bottom sediment of Benanga Reservoir. This distribution pattern will show the distribution of sediment with potentially dangerous Fe heavy metal contamination. Of several guidelines for iron content quality standards in sediment (Table 1), the EPA (1977) guideline was used because it covers the most types of heavy metals and the guideline is still used today. Thus, this guideline can be used when conducting research with more different heavy metals. As a comparison to the EPA (1977) guideline, Fe content data was also processed to obtain the Contamination Factor (CF), Degree of Contamination (DC), Metal Enrichment Factor (EF), and Geoaccumulation index (I_{geo}) using the classification presented in Table 2, Table 3, and Table 4.

Table 2. Classification of Contamination Factors (CF) and Contamination Degree (DC), [14]-[16].

Contamination Factors (CF)	Contamination Level	Contamination Degree (DC)	Contamination Level
$CF < 1$	Low	$DC < 6$	Low
$1 \leq CF < 3$	Medium	$6 < DC < 12$	Medium
$3 \leq CF < 6$	Significant	$12 < DC < 24$	Significant
$CF \geq 6$	Very high	$DC > 24$	Very high

Table 3. Classification of Enrichment factor (EF) [14]-[16].

Enrichment factor (EF)	Degree of enrichment
$EF < 2$	Depletion to mineral enrichment
$2 \leq EF < 5$	Moderate enrichment
$5 \leq EF < 20$	Significant enrichment
$20 \leq EF < 40$	Very high enrichment
$EF > 40$	Extremely high enrichment

Table 4. Classification of Geoaccumulation index (I_{geo}) [14] [15].

(I_{geo})	Class	Contamination Level
$I_{geo} \leq 0$	0	Uncontaminated
$0 \leq I_{geo} \leq 1$	1	Uncontaminated to moderately contaminated
$1 \leq I_{geo} \leq 2$	2	Moderately contaminated
$2 \leq I_{geo} \leq 3$	3	Moderately to heavily contaminated
$3 \leq I_{geo} \leq 4$	4	Heavily contaminated
$4 \leq I_{geo} \leq 5$	5	Heavily to very heavily contaminated
$I_{geo} > 6$	6	Very heavily contaminated

The calculation of the Contamination Factor (CF), Degree of Contamination (DC), Metal Enrichment Factor (EF), and Geoaccumulation index (I_{geo}) was carried out using the following method:

-Contamination factors by heavy metals (CF) [14]:

$$CF = C/Co \tag{1}$$

where:

CF: contamination factor by aluminum and heavy metals;

C: average concentration of aluminum in sediment;

Co: geochemical background.

-Degree of contamination (DC) [14]:

$$DC = \sum Cf \tag{2}$$

where:

CF: contamination factor.

-Metal Enrichment Factor (EF) [14] [15] [17]:

$$\mathrm{EF} = \frac{\left(\frac{\mathrm{M}}{\mathrm{M_{bg}}}\right)}{\left(\frac{\mathrm{R}}{\mathrm{R_{bg}}}\right)} = \frac{\left(\frac{\mathrm{M}}{\mathrm{R}}\right)}{\left(\frac{\mathrm{M_{bg}}}{\mathrm{R_{bg}}}\right)} = \frac{\mathrm{M}}{\mathrm{M_{bg}}} \times \frac{\mathrm{R_{bg}}}{\mathrm{R}} = \frac{\mathrm{M}}{\mathrm{R}} \times \frac{\mathrm{R_{bg}}}{\mathrm{M_{bg}}} = \frac{\left(\mathrm{M} \times \mathrm{R_{bg}}\right)}{\left(\mathrm{R} \times \mathrm{M_{bg}}\right)} \tag{3}$$

where:

M: metal concentration in sediment at sampling point;

M_{bg}: background metal concentration;

R: reference metal concentration in sediment at sampling point;

R_{bg}: background metal concentration.

-Geoaccumulation index (I_{geo}) [14] [15] [17]:

$$I_{geo} = log_2\left[\frac{M_0}{\left(1.5M_{bg}\right)}\right] \tag{4}$$

where:

M_0: metal concentration in sediment at sampling point;

M_{bg}: background metal concentration.

The calculation of EF and I_{geo} contamination indices uses Al for normalization because this element is evenly distributed in the natural environment and is a major element in the earth's crust.

3. Results and Discussion

3.1. Distribution of Heavy Metal Iron (Fe) in Benanga Reservoir Sediment

The average Fe metal content in Benanga Reservoir sediment deposits vertically shows an increasing trend from the bottom layer (A) to the middle layer (B) and continues to the top layer (C). The average Fe content in layer A is 145,185 ppm, layer B is 159,254 ppm, and layer C is 174,556 ppm. The same increasing trend pattern is also shown by the average Fe content in background samples taken in the upstream of the Pampang Kiri River, where the average Fe content in layer A is 81.714 ppm, layer B is 128.733 ppm, and layer C is 120.247 ppm (**Figure 4**).

The Fe content in the Benanga Reservoir sediment shows a buildup of Fe concentration of 204%, greater than the background. Thus, there is a buildup of Fe in the reservoir more than twice the Fe content from the upstream of the Pampang Kiri River. The buildup of heavy metal content occurs due to reduced energy of the transport media after the sediment enters the reservoir which is caused by the increasing width of the channel cross-section which causes the sediment deposition process to be faster with a larger volume.

The distribution of the highest levels of heavy metal Fe was found at points 3 and 4 which shows that heavy metal deposition effectively occurs at the mouth of the main river, namely the Pampang Kiri River. The reduced current speed after entering the reservoir causes heavy metals to settle more easily to the bottom of the water and bind to the sediment. This is in accordance with the statement of Maslukah *et al.* (2017) which states that calm currents cause heavy metals to settle at the bottom of the water [17]. In addition to current dynamics, the ability of sediment to accommodate heavy metals also depends on the texture and organic material content [18].

In the bottom sediment deposits of Benanga Reservoir, other factors that control the distribution of selected heavy metal levels are the sedimentation rate and the subsidence of the basin floor. This can be seen at point 2 which has a greater sediment thickness compared to other points, has a lower vertical Fe heavy metal content. This shows that the rate and capacity of Fe heavy metal deposition are inversely proportional to the sediment deposition rate in Benanga Reservoir. Lake or reservoir sediments are characterized by rapid sedimentation rates and temporal continuity, which can provide a continuous record of climate, environment, and human activities in the lake and upstream watershed [2]. The difference in sediment deposition velocity seen from the sediment thickness at each sampling point controls the difference in Fe content in the sediment.

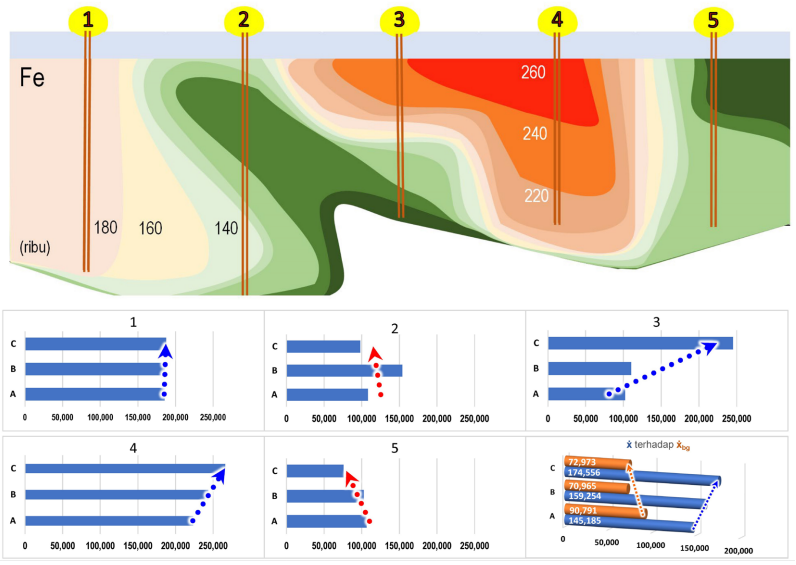

Figure 4. (Top) Distribution of Fe content and (bottom) vertical trend of iron (Fe) content in Benanga Reservoir sediment deposits.

For each point, the increasing trend of Fe metal content is seen at points 1, 3, and 4, while the decreasing trend of Fe content in the Benanga Reservoir sediment is seen at points 2 and 5 (Figure 4).

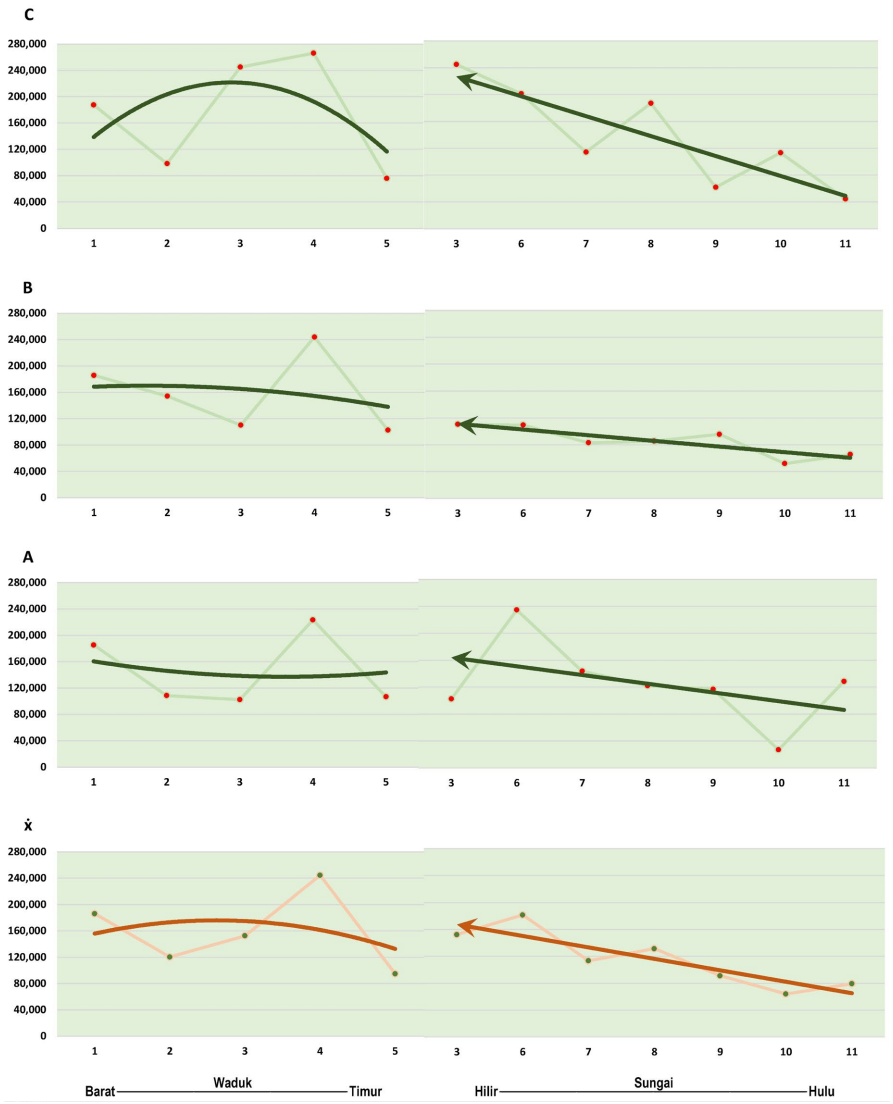

Figure 5. Lateral trend of heavy metal content of Iron (Fe) in sediment deposits of Benanga Reservoir (points 1 - 5) and Pampang Kiri River (points 6 - 11).

The average content of Fe metal in the sediment deposits of Benanga Reservoir and Pampang Kiri River laterally has a value ranging from 64,011 - 244,392 ppm. The highest average content of heavy metal Fe was found at point 4 located in Benanga Reservoir and the lowest average content was found at point 10 in the upstream part of Pampang Kiri River. From the upstream part of Pampang Kiri River to the downstream, the trend of the average Fe content increased, while the trend of the average Fe content in Benanga Reservoir showed a higher concentration in the middle part of the reservoir compared to the east and west sides of the reservoir as shown in Figure 5($\bar{x}$).

The Fe content per layer of sediment deposits of Pampang Kiri River and Benanga Reservoir laterally is:

-Layer C; Fe metal content laterally has a value ranging from 44,185 - 266,148 ppm. The lowest value of Fe heavy metal content is at point 11 in the upstream part of the Pampang Kiri River and the highest value is at point 4 located in the Benanga Reservoir. From the upstream part of the Pampang Kiri River to the downstream, the average Fe content trend has increased quite sharply, while the average Fe content trend in the Benanga Reservoir shows a higher concentration in the middle part of the reservoir compared to the east and west sides of the reservoir as shown in Figure 5(C).

-Layer B; Fe content laterally has a value ranging from 51,932 - 243,667 ppm. The lowest value of Fe heavy metal content is at point 10 in the upstream part of the Pampang Kiri River and the highest value is at point 4 located in the Benanga Reservoir. From the upstream of the Pampang Kiri River to the downstream, the average Fe content trend experienced a relatively gentle increase, while the average Fe content trend in the Benanga Reservoir showed a fairly even distribution from the east side to the west side of the reservoir as shown in Figure 5(B).

-Layer A; Fe content laterally has a value ranging from 27,056 - 234,602 ppm. The lowest value of Fe heavy metal content is at point 10 in the upstream of the Pampang Kiri River and the highest value is at point 6 located at the mouth of the Benanga Reservoir. From the upstream of the Pampang Kiri River to the downstream, the average Fe content trend experienced an increase, while the average Fe content trend in the Benanga Reservoir shows a fairly even distribution from the east side to the west side of the reservoir as shown in Figure 5(A).

3.2. Heavy Metal Iron (Fe) as a Contaminant in Benanga Reservoir Sediment

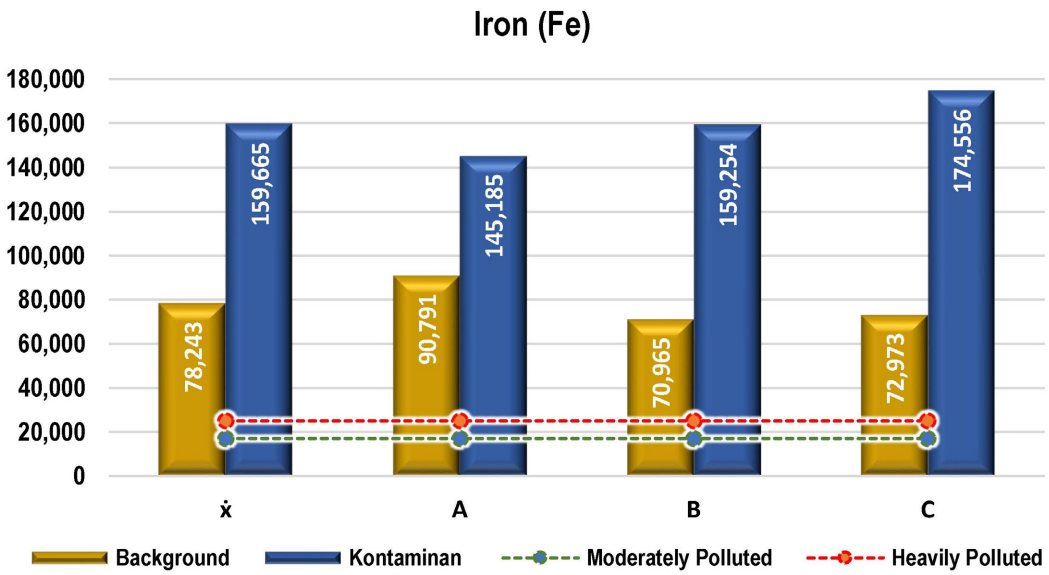

Figure 6. Contamination level of heavy metal Iron (Fe) in sediment deposits of Benanga Reservoir.

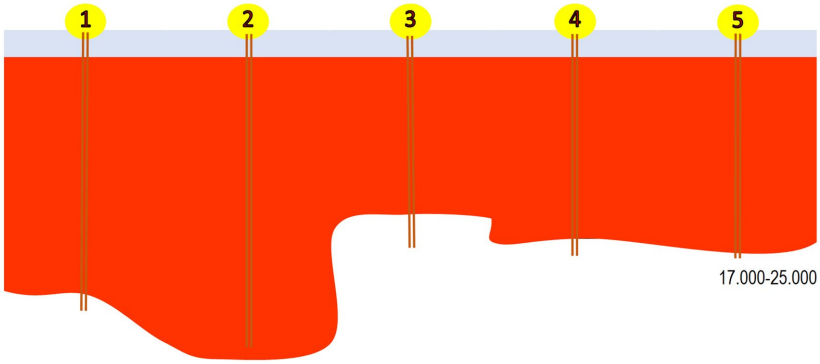

Figure 7. Distribution of heavy metal contamination of Iron (Fe) in Benanga Reservoir sediment deposits based on USEPA Standards, 1977 [10].

The Fe content in Benanga Reservoir sediments far exceeds the heavy metal toxicity classification standards in sediments according to the USEPA (1977) standards, both in background samples and in Benanga Reservoir sediment samples. The Fe content in Benanga Reservoir sediments ranges from 145,185 - 174,556 ppm, far exceeding the highest content limit for sediments that experience very heavy pollution for Fe (>25,000 ppm). The large Fe content in the bottom sediments of Benanga Reservoir is caused by the content in the background sediments which also exceed the standards in the range of 70,965 - 90,791 ppm (**Figure 6**). Thus, the distribution of "heavily polluted" Fe contamination is spread throughout the Benanga Reservoir sediment deposits (**Figure 7**).

As a comparison of Fe contamination analysis based on the USEPA (1977) guidelines, the following are calculations of several Fe heavy metal pollution indices in sediments which show that:

- The contamination factor (CF) of Fe heavy metal in Benanga Reservoir sediment is 2.041 (medium contamination level), with the lowest value found in the bottom layer (A) is 1.599 (medium contamination level), followed by the middle layer (B) 2.244 (medium contamination level), and the highest in the top layer (C) 2.392 (significant contamination level). The contamination level (DC) which is the accumulation of Cf is 6.235 indicating a medium contamination level.
- The enrichment factor (EF) of Fe heavy metal in Benanga Reservoir sediment of 1.719 indicates that there is no effect of anthropogenic pollution on the metal content in the bottom sediment of Benanga Reservoir.
- Calculation of the Geoaccumulation Index (I_{geo}) shows that the sediment from Benanga Reservoir is not contaminated to moderately contaminated (class I = 0.444).

The difference in Fe contamination levels using the USEPA (1977) guidelines and the calculation of the contamination index occurs because both have different approaches. The USEPA (1977) guidelines use a standard contamination threshold level without considering the initial/local level conditions, while the calculation of the contamination index uses a comparison between the Fe content in reservoir sediments and the Fe content of upstream river sediments originating from

source rock. The results of the analysis show that the Fe levels in both upstream and reservoir sediments have exceeded the USEPA (1977) standards, but the high accumulation of Fe in reservoir sediments is not influenced by anthropogenic activities. The high Fe content in sediments from the upstream Pampang Kiri River and in Benanga Reservoir sediments that exceed the USEPA (1977) guidelines is because the Fe content in source rocks in the form of sedimentary rocks from the Kutai Basin is indeed high, as seen from the quality of surface water and groundwater which have high iron oxide deposits after being left for some time.

The decrease in Fe levels at points 2 and 5 is influenced by sedimentation dynamics. Point 2 has a greater sediment thickness than other points indicating a faster sedimentation process so that Fe binding does not run effectively. While at point 5 which is relatively further from the river mouth, it has a relatively calmer current condition which also tends to cause Fe binding to run ineffectively.

Metal-contaminated environments can release metals into the surrounding waters in three ways: (a) through desorption from suspended particles after contact with seawater, (b) through desorption from bottom sediments and (c) through diffusion from interstitial water after diagenetic alteration of sediments. However, if the balance between the (marine) sediment and the overlying water body is disturbed, the sediment will transfer most of the pollutants to the seawater. When the concentration of metals in the sediment increases, more heavy metals will return to the water body through chemical and biological processes [7].

Sediments with very high Fe content in reservoirs used for agriculture and drinking water can pose several environmental and health risks. Fe is stable in the form of oxides, and the impact is more on aesthetics (color, odor, and sediment), but can still be managed by filtration. While Fe in dissolved form (Fe^{2+}) or associated with sulfide minerals can pose serious risks to water quality, agriculture, and reservoir ecosystems.

High Fe content can be directly toxic to humans [8]. Fe in the form of Fe^{2+} (soluble) in high concentrations can stimulate the growth of iron bacteria such as Gallionella spp. and Leptothrix spp., which can cause biofilms and unpleasant odors [19]. In the agricultural sector, high Fe can cause iron poisoning in plants [20], especially in some plants, especially rice, where high Fe can inhibit root growth and reduce crop yields. High Fe in water can interfere with the absorption of phosphorus (P), zinc (Zn), and manganese (Mn) by plants, causing deficiencies of nutrients that are important for growth. Meanwhile, in reservoir ecosystems, high Fe content can reduce oxygen levels which are dangerous for fish and aquatic organisms and can cover the bottom of the reservoir, thus inhibiting the growth of phytoplankton and basal organisms.

Risk management of high Fe content in the bottom sediment deposits of Benanga Reservoir can be done by reducing the erosion rate in the catchment area, minimizing the sediment load entering the reservoir, minimizing the amount of sediment that settles in the reservoir, removing sediment deposits in the reservoir, and carrying out vegetative and social management. The most important ap-

proach currently being taken is through limiting deforestation and mining activities in the upstream Pampang Kiri River which are still occurring massively. Meanwhile, for the management of Benanga Reservoir, sediment handling, especially for dredging activities, needs to pay attention to the distribution pattern of Fe content in Benanga Reservoir sediment.

4. Conclusion

The Fe metal content in the Benanga Reservoir sediment deposits shows an increasing trend from the bottom layer (A) to the middle layer (B) and continues to the top layer (C). The Fe content in the Benanga Reservoir sediments far exceeds the quality standards for the classification of heavy metal toxicity in sediments according to the USEPA (1977) guidelines, so the distribution of "heavily contaminated" Fe contamination is spread throughout the Benanga Reservoir sediment deposits. The high levels of Fe in both upstream and reservoir sediments are not influenced by anthropogenic activities, but come from source rocks that have high Fe content. The controlling factors for the amount of Fe content in the Benanga Reservoir sediment deposits are current dynamics, sediment texture, organic matter content, and sediment deposition velocity. To reduce the risk of Fe contamination in Benanga Reservoir sediments, it is necessary to limit deforestation and mining activities in the upstream Pampang Kiri River, while for the management of Benanga Reservoir, sediment handling needs to pay attention to the distribution pattern of Fe content in Benanga Reservoir sediments.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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