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Heavy Metals Pollution in the Bed Load Sediments of the Brantas River in Batu City

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Abstract

Aim to Evaluation the contamination levels of heavy metals As, Cr, Zn, and Pb in upstream of Brantas River using IP (Inductively Coupled Plasma) to measure heavy metal and identify the use of synthetic pesticide and fertilizer and Recommend strategies reduce contamination of heavy metal, Where were collected16 samples sediment from two different branches river, Each Location has different grains size 0.063 µm and 0.200 µm. The increasing trend of metals was observed in sediments of the Grain size 0.200 µm As < Pb < Zn < Cr respectively and in the Grain size 0.063 µm As< Cr <Zn <Pb in the river 1 and for the river 2 in the Grain size 0.200 µm The increasing trend of metals was observed in sediments Pb< As < Cr < Zn and in the Grain size 0.063 As <Zn< Pb< Cr which means the concentration levels in sediment all the metals in the Grain size 0.200 µm in river 1 and river 2 higher than safe values and in grain size 0.063 µm lower than safe values. This is what was shows it in Assessment of the degree of pollution in sediments using (EF), (PLI) , (Igeo) and (CF).

Keywords: heavy metal, sediment, evaluate contamination

INTRODUCTION

Large amounts of risky chemical compounds, particularly significant metals, have been launched in rivers around the world because of fast and intensive native activities, furthermore because of the enlargement of commercial and agricultural production [1]–[3] Heavy metals discharged into a river system are distributed by natural or anthropogenic sources while transported between the water phase and the sediments of the stratum[4] Heavy metals can come from pesticides, fertilizers, industrial waste, sewage, workshop waste, runoff, hospital waste, waste market [5].

The chemical washing of sidewalks, Sinks, and runoff from banks is the primary source of heavy metals [6]. Due to adsorption, hydrolysis, and co-precipitation, only a small fraction of the free metal ions remain dissolved in the water, and a large amount is deposited in sediment [7]. Heavy metals may accumulate in aquatic plants, fish, and oysters and can be transported to humans through the food chain.

This leads to developmental and behavioral changes, psychological and cognitive exhaustion in the exposed person and sometimes death [8].

The Brantas River is one of the longest rivers in East Java, after the Begawan Solo River. The watershed of the Brantas River helps control hydro disc line, irrigation, hydropower station (PLTA), drinking water, fisheries, and recreation [9]. The increase in the population of the Brantas River and waste from activity contributes to the high level of pollution [10]. Analysis of dissolved minerals in water is a practical A valuable tool for assessing condition pollution in a given ecosystem, reflecting its degree of contamination [11] [12] [2] [13].

Objectives of the study

1. To calculate the concentration levels of arsenic (As), chromium (Cr), zinc (Zn) and lead (Pb) in the sediment upstream Brantas River in Batu area using two different grain sizes 0 .200µm and 0.063µm.
2. To identify the use of synthetic pesticide and fertilizer.
3. To recommend strategies that can reduce the heavy metal contamination in up stream of Brantas River.

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MATERIAL AND METHOD

Describe

This research was conducted in the Brantas River in Batu, with 2 study sites and 4 Stations in each study site (Figure 1). The Brantas River is one of the longest rivers in East Java, after the Begawan Solo River. The watersheds of the Brantas River are helpful for water, irrigation, hydropower, drinking water, fisheries, and recreation. But the most significant proportion of agricultural pollutants is due to the use of pesticides and fertilizers in agriculture, which contributes to the high pollution level of river deposits with heavy metals.

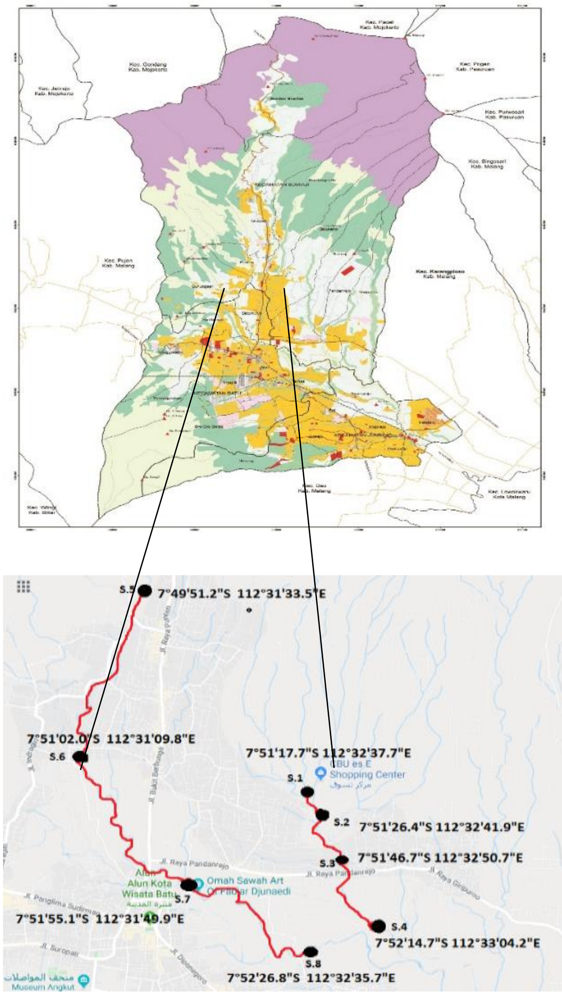

Figure 1. Map of Batu Area

Sampling Collection and Analysis

Some sediment samples were collected from two different branches of the Brantas River in Batu region in September and October 2019. 16 samples were collected from 8 sites. The samples were divided into two branches of the river for each branch 8 samples. Each site has two different samples of sediment grains 0.063 µm and 0.200 µm of different points. The locations were determined by manual GPS. Ekman Grab Sampler was used to sample sediments. After collection, sediment samples (stored in compressed bags of clean polyethylene) were transferred to the laboratory immediately. In case of freezing (at 4 ° C). After that at 104 ° C were dried for forty-eight hours. then stored in a PE container vessel under routine high pressure temperature became routine and a routine temperature to avoids external contamination and need smaller and shorter time amounts of acids, thereby improving detection limits and the overall total of the method analytical [14]. 10 grams of sediment sample was placed in the reference vessel. Then 20 ml of HNO3 were added to reaction vessel, and boiled until the solution clear, if not clear added more HNO3, boild again until the solution clear. The digestion solution was cooled and filtered the filtered sample was then manufactured to 100 ml in volumetric flash with distilled water. Use IP (Inductively Coupled Plasma) to measure and detect the heavy metal in sediment samples. [15]

Assessment of the degree of pollution in sediments

Several researchers have used the average shale values or the average crustal abundance data as reference baselines[16]–[19] Assessment of the degree of pollution in sediments , we are using 4 parameters: Enrichment Factor (EF), Contamination Factor (CF), Pollution Load Index (PLI) and Geo-accumulation Index (I_{geo}).

Enrichment Factor (EF)

Enrichment factors were considered (EF) to estimate the abundance of metals in sediment samples indicator reflecting the status and degree contamination of the environmental [20]. EF was calculated by a comparison of metal concentration with that of a reference metal That were chosen from conditions from regional or global average composition [21], [22] . was calculated The EF using the methods [23] as follows:

$$EF = (Me/Fe)_{sample} / (Me/Fe)_{background} \quad (1)$$

Where (Me / Fe) of the sample is the ratio of minerals to Fe in the interest sample; (Me / Fe) the value of background is the natural background of minerals to Fe ratio. Since there are no mineral values of background with our study area, we are used values from surface world rocks[24]. Iron was chosen as a normalizing component because natural sources (1.5%) largely controlled its inputs [25]. The categories of enrichment factors are listed in Table 1

Table 1. Enrichment factor (EF) categories[26]

Enrichment factor (EF)	Enrichment factor (EF) Categories
EF < 2	Deficiency to minimal enrichment
2 ≤ EF< 5	Moderate enrichment
5 ≤ EF < 20	Significant enrichment
20 ≤ EF < 40	Very high enrichment
EF ≥ 40	Extremely high enrichment

Contamination Factor (CF)

Level of sediment contamination with minerals Expressed by the contamination factor (CF) calculated as follows:

$C F= C_m \text{ Sample} / C_m \text{ background}$ (2)

where, C_m is the concentration of Sample of a particular mineral in river sediments, and the background of C_m is the value of the mineral that is equal to the global surface rock average given by [24] .CF values to describe the level of pollution appear in Table 2.

Table 2. Contamination factor (CF) and contamination level

Contamination Factor (CF)	Contamination Level
CF<1	Low contamination
1 ≤ CF < 3	Moderate contamination
3 ≤ CF< 6	Considerable contamination
CF > 6	Very high contamination

Pollution Load Index (PLI)

Pollution load index (PLI), For a specific site, it was evaluated using the method proposed by [27].

Expressed this is parameter as:

$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots\ldots\ldots CF_N)^{1/N}$ (3)

Where, is the number of metals.

Geographic accumulation Index (I_{geo})

The mineral concentration enrichment was calculated using the method proposed by [28], [29] termed the geo-accumulation index (I_{geo}). :Geo- accumulation index is expressed as follows

$L_{geo} = Log_2 (C_m \text{ Sample}/ (1.5\times B_m \text{ Background}))$ (4)

where C_m is the measured concentration of element in the sediment sample and Bm is the Background value of the geochemical background (rocky mean of the surface of the world given by [24]. Factor 1.5 is combined to include a possible variation in the background values Because the lithogenic effect Muller suggested [30] seven degrees or Categories of the geographical accumulation index, and these categories are presented in Table 3.

Table 3. Classification of Muller for geo-accumulation index (I_{geo}) [31], [32]

level	I_{geo} Value	Quality assessment of sediments
0	< 0	Unpolluted
1	0 -1	From unpolluted to moderately polluted
2	1-2	moderately polluted
3	2-3	From moderately to strongly polluted
4	3-4	Strongly polluted
5	4-5	From strongly to extremely polluted
6	>6	Extremely

RESULTS AND DISCUSSION

The heavy metals concentration of (As, Cr, Zn, and Pb) in sediment samples are presented in Table 4. While the contamination factor (CF), Geographic accumulation Index (I_{geo}), Enrichment factor (EF) and pollution load index (PLI) calculated in each of the heavy metals are presented in Table 5, 6 and 7 respectively. Table 8 represent the comparison of heavy metals concentration studied with results of other works.

Metal Concentration in Sediment

The heavy metals concentrations by (mg/kg) of (As, Cr, Zn, and Pb) in sediments are presented in Table 4. It is clear from the Table that highest concentration value heavy metal (As) in the Grain size 0.200 µm at river 2 are much higher than the river 1 (106.9mg/kg>14.5mg/kg) and the highest concentration value of heavy metal (As) in the Grain size 0.063 µm in the river 2 are much higher than the river 1 (10.8 mg/kg >2.3 mg/kg). The comparison between highest value concentrating of heavy metal (As) in the Grain sizes of 0.200 µm and 0.063 µm for river 1, are the highest for the Grain size 0.200µm (14.5 mg/kg) are higher than the Grain size 0.063µm (2.3 mg/kg). While the highest value of concentration for heavy metal (As) in the river 2 is in the grain size of 0.200 µm (106.9mg/kg) are higher than the Grain size 0.063 µm (10.8 mg/kg).

It is clear from the Table 4 that highest concentration value of heavy metal (Cr) in the Grain size 0.200 µm at river 1 are much higher than the river 2 (270 mg/kg >177.9mg/kg). While highest concentration value of heavy metal (Cr)of the Grain size 0.063 µm in the river 2 are much higher than the river 1 (59.7 mg/kg >36.93 mg/kg). The comparison of concentration value of heavy metal (Cr) in the Grain sizes of 0.200 µm and 0.063 µm for the river 1 are the highest in the Grain size 0.200µm (270 mg/kg) and in the Grain size 0.063 is (36.93 mg/kg). While highest value of heavy metal concentration in the river 2 was in the grain size of 0.200µm (177.9 mg/kg) and in the Grain size 0.063 are (59.7 mg/kg). It is clear highest concentration value for heavy metal (Cr) are for the river 1 in The Grain size 0.200µm (270 mg/kg).

It is clear from the Table 4 the highest concentration value of heavy metal (Zn) in the grain size 0.200 µm at river 1 are higher than the river 2 (207.7mg/kg >173.4mg/kg). While the concentrating value of heavy metal (Zn) in the Grain size 0.063 µm in the river 1 are higher than the river 2 (105.4 mg/kg >30 mg/kg). While the comparison of concentration value of heavy metal (Zn) in the Grain sizes of 0.200 µm and 0.063 µm for the river 1, in the Grain size 0.200µm (207.7mg/kg) are higher than the Grain size 0.063µm (105.4 mg/kg). While highest concentration value of heavy metal (Zn) in the river 2 for the grain size of 0.200µm (173.4 mg/kg) are higher than the Grain size 0.063µm (30 mg/kg). It is clear the concentration value heavy metal (Zn) in the river 1 are the highest in the grain size 0.200µm (207.7 mg/kg).

It is clear from the Table 4 that highest concentration value of heavy metal (Pb) in the Grain size 0.200 µm at river 1 are much higher than the river 2 (364.1mg/kg >90.8mg/kg), While highest concentration value of heavy metal (Pb) in the Grain size 0.063 µm in the river 1 are much higher than the river 2 (80.7mg/kg >72.2 mg/kg).

The comparison of concentration value of heavy metal (Pb) in the Grain sizes of 0.200 µm and 0.063 µm for the river 1 are the highest for the Grain size 0.200µm (364.1mg/kg) are higher than the Grain size 0.063µm (80.7 mg/kg). While highest concentration value of heavy metal (Pb) in the river 2 in the grain size 0.200 µm (90.8 mg/kg) are higher than the Grain size0.063µm (72.2 mg/kg). For all heavy metals in the grain size 0.200µm in the two rivers were compared with rock average surface of the world And they are higher than safe values.

Table 4. Concentration of heavy metals in up stream of Brantas River sediments and their world surface rock average

S. S		As mg/kg		Cr mg/kg		Zn mg/kg		Pb mg/kg		Fe mg/kg	
		Grain size		Grain size		Grain size		Grain size		Grain size	
		0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm
RIVER 1	**S1**	0.3	0.1	66.4	35.4	70.7	28.3	56.8	49.6	129800	45610
	S2	0.3	0.2	270	36.9	156.8	7.93	364.1	80.7	373900	166200
	S3	14.5	2.3	90.9	34.5	128.5	105.4	68.5	38	366100	124400
	S4	7.7	<0.005	267.7	36.7	207.7	51.3	65.1	62.9	430900	70140
	AVG	5.7	0.867	173.8	35.9	140.9	48.23	138.6	57.8	325175	101588
RIVER 2	**S5**	<0.005	<0.005	150.8	31.1	173.4	17.4	43.2	14.4	319200	7157
	S6	0.6	0.1	127.8	14.8	149.6	30	46.8	13.9	320500	1631
	S7	93.9	9.4	177.9	19.8	148.8	22	44.6	10.9	347300	3894
	S8	106.9	10.8	129.5	59.7	146.4	23.5	90.8	72.2	250900	13720
	AVG	67.13	6.767	146.5	31.4	154.6	23.23	56.35	27.85	309475	6600.5
WORLD SURFACE ROCK AVERAGE [33]		7.9		71		127		16		35900	

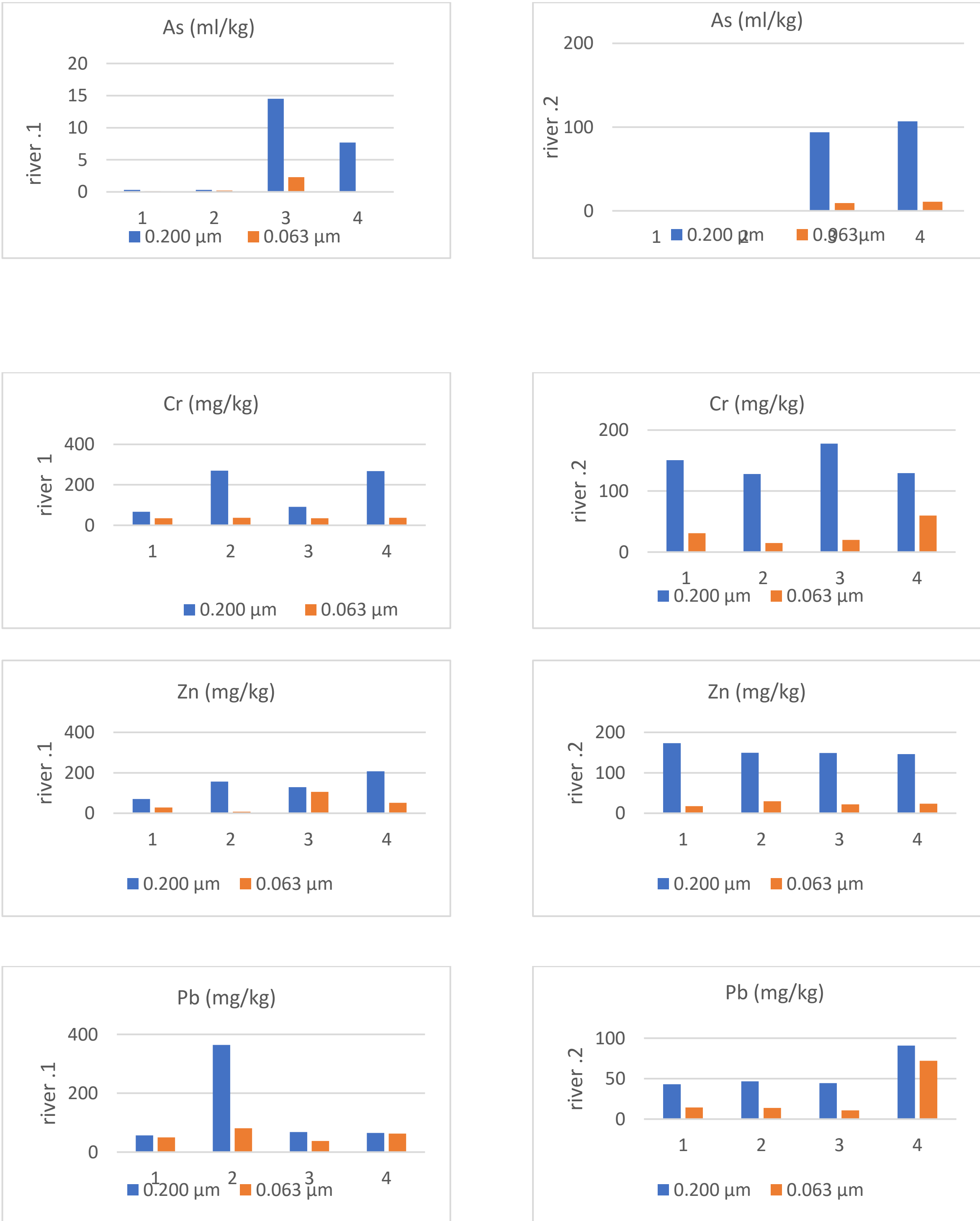

Figure 2. Heavy metal concentrations of different Grain size 0.200 µm and 0.063 µm

Contamination Factor (CF) and Pollution Load Index (PLI)

The average CF values in the river 1 in the Grain size 0.200 µm for heavy metals (As) the CF valuewas lower than CF<1 so that river is Low contamination, while the average CF values different heavy metals (Cr and Zn) in the Grain size 0.200 µm For Some sites along the river 1, the

CF value is from 1 < CF < 3, so that river is Moderate contamination, while the average CF values for Pb. Is higher than CF > 6 so that river is very high contamination.

In the Grain size 0.063 µm in the river 1The average CF values for different heavy metals (As, Cr and Zn) the CF value was lower than CF<1 so that river is Low contamination while The average CF values for (Pb) is from 3 < CF < 6 so that river is Considerable contamination, while the average CF values for river 2 in (As) is higher than CF > 6 so that river is Very high contamination and The average CF values for different heavy metals (Cr and Zn) in the Grain size 0.200 µm for Some sites along the river 2, the CF value is from 1 < CF < 3, so that river is Moderate contamination while the average CF values for (Pb) is from 3 < CF < 6 so that river is Considerable contamination.

In the Grain size 0.063 µm the average CF values for different heavy metals (As, Cr and Zn) in the river 2 the CF value was lower than CF <1 so that river is Low contamination. While the average CF values for Pb is from 1 < CF < 3 so that river is Moderate contamination as shown in the (table 2).

Pollution Load Index (PLI):

The average value of PLI in the Grain size 0.200 µm for the sites along the river 1 it is (1.486) and in the river 2 it is (1.950) so that rivers are Moderate contamination. While in the Grain size 0.063 µm in the river 1 it is (0.347) and in the river 2 it is (0.429) so that rivers are Moderate Low contamination. As shown in the (table 2)

Table 5. Contamination Factor (CF) AND Pollution Load Index (PLI) values of heavy metals in upstream of Brantas River sediment

Sampling site		As mg/kg		Cr mg/kg		Zn mg/kg		Pb mg/kg		**PLI**	
		Grain size		Grain size		Grain size		Grain size		Grain size	
		0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm
River 1	**S1**	0.038	0.013	0.935	0.499	0.557	0.223	3.55	3.1	0.515	0.257
	S2	0.038	0.025	3.803	0.52	1.235	0.062	22.756	5.044	1.419	0.254
	S3	1.835	0.291	1.28	0.486	1.012	0.83	4.281	2.375	1.786	0.727
	S4	0.975	0.0006	3.77	0.517	1.635	0.404	4.069	3.931	2.224	0.151
	AVERAGE	0.722	0.082	2.447	0.506	1.11	0.38	8.664	3.613	1.486	0.347
River 2	**S5**	0.0006	0.0006	2.124	0.438	1.365	0.137	2.7	0.9	0.265	0.128
	S6	0.076	0.0127	1.8	0.208	1.178	0.236	2.925	0.869	0.828	0.153
	S7	11.886	1.19	2.506	0.279	1.172	0.173	2.788	0.681	3.14	0.445
	S8	13.532	1.367	1.824	0.841	1.153	0.185	5.675	4.513	3.565	0.99
	AVERAGE	6.374	0.643	2.064	0.442	1.217	0.183	3.522	1.741	1.95	0.429

Geographic Accumulation Index (I_{geo})

The highest values for the I_{geo} in heavy metal (As) in the Grain size 0.200 µm in the river1 was from 0-1 so that river is from unpolluted to moderately polluted (Table 3) .While The highest values for the I_{geo} in heavy metal (As) in the Grain size 0.200 µm in the river 2 was from 3-4 so that river is Strongly polluted (Table 3), the highest values for the I_{geo} in heavy metal (As) in the Grain size 0.063 µm in the river1 and river 2 was lower than >0 so that river is unpolluted (Table 3).

The highest values for the I_{geo} of heavy metal (Cr)in the Grain size 0.200 µm in the river1 was from 1-2 so that river is moderately polluted and in the river 2 was from 0-1 so that river is unpolluted to moderately polluted. While the highest values for the I_{geo} in heavy metal (Cr) in the Grain size 0.063 µm in the river 1 and river 2 were lower than <0 so that rivers is unpolluted.

The highest values for the I_{geo} in heavy metal (Zn) in the Grain size 0.200 river 1 was from 0-1 so that river is from unpolluted to moderately polluted (Table 3) and in the river 1 lower than < 0 so that river is unpolluted. While The values for the I_{geo} in heavy metal (Zn) in the grain size 0.063 µm in the river 1 and river 2 were lower than < 0 so that rivers is unpolluted.

The highest values for the I_{geo} in heavy metal (Pb) in the Grain size 0.200 µm in the river1 was from 3-4 so that river is Strongly polluted and in the river 2 was from 1-2 so that rivers is moderately polluted. While the highest values for the I_{geo} in heavy metal (Pb) in the Grain size 0.063 µm in the river1and the river 2 were from 1-2 so that rivers are moderately polluted.

Table 6. Geo-accumulation Index (I_{geo}) values of heavy metals in upstream of Brantas River sedime

Sampling site		As mg/kg		Cr mg/kg		Zn mg/kg		Pb mg/kg	
		Grain size		Grain size		Grain size		Grain size	
		0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm
River1	**S1**	-5.304	-6.889	-0.682	-1.589	-1.43	-2.751	-0.134	1.047
	S2	-5.304	-5.889	1.34	-1.528	-0.281	-4.586	3.923	1.75
	S3	0.291	-2.365	-0.229	-1.626	-0.568	-0.854	1.513	0.663
	S4	-0.622	-11.211	1.33	-1.537	0.125	-1.893	1.44	1.39
River2	**S5**	-11.211	-11.211	0.502	-1.776	-0.136	-3.453	0.848	-0.737
	S6	-4.304	-6.889	0.263	-2.847	-0.349	-2.667	0.963	-0.788
	S7	2.986	-0.334	0.74	-2.427	-0.356	-3.114	0.894	-1.139
	S8	3.173	-0.134	0.282	-0.835	-0.38	-3.019	1.92	1.589

Enrichment Factor (EF)

The highest Enrichment Factor (EF) heavy metals (As, Cr and Zn) of river 1 in the Grain size 0.200 µm and 0.063 µm was lower than EF < 2 so that is river is deficiency to minimal enrichment. and the highest values for heavy metals (Pb) in the Grain size 0.200 µm and 0.063 µm was from 2 < EF < 5 so that is river is moderate enrichment. (**table 1**). The highest Enrichment Factor (EF) for heavy metals (As, Cr, Zn and Pb) of river 2 in the grain size 0.200 µm were lower than EF < 2 so that is river is Deficiency to minimal enrichment and in the Grain size 0.063 µm for heavy metals (As ,Zn and Pb) were 5 < EF < 20 so that is river is Significant enrichment and in heavy metal (Cr) was 2 < EF < 5 so that is river is Moderate enrichment (**table 1**)

Table 7. Enrichment Factor (EF) values of heavy metals in upstream of Brantas River sediment

Sampling site		As mg/kg		Cr mg/kg		Zn mg/kg		Pb mg/kg	
		Grain size		Grain size		Grain size		Grain size	
		0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm	0.200 µm	0.063 µm
River1	**S1**	0.011	0.01	0.258	0.392	0.154	0.175	0.982	2.44
	S2	0.004	0.005	0.365	0.112	0.119	0.013	2.185	1.089
	S3	0.18	0.084	0.126	0.14	0.099	0.24	0.42	0.685
	S4	0.081	0.0003	0.314	0.265	0.136	0.207	0.339	2.012
	AVERAGE	0.069	0.025	0.266	0.227	0.127	0.159	0.982	1.557
River2	**S5**	**7.12*10^-5**	0.003	0.239	2.197	0.154	0.6877	0.304	4.514
	S6	0.009	0.279	0.202	4.588	0.132	5.1997	0.328	19.122
	S7	1.229	10.97	0.259	2.571	0.121	1.597	0.288	6.281
	S8	1.936	**3.577**	0.261	2.2	0.165	0.484	0.812	11.807
	AVERAGE	1.058	3.707	0.24	2.889	0.143	1.992	0.433	10.431

Comparison of Average Heavy Metals Concentration in Sediment of Brantas River with Other Studies

In comparison of the average heavy metals concentration in the sediment of Brantas River with other studies, I find the Concentration of heavy metal (Cr) in this study is 146.5, and in other rivers, the observed increasing trend of heavy metals (Cr) respectively were, river 3 < river 7 < river 2 < river 6 < river 5 < river 8 the value of the concentrations are 38.9 < 58.4 < 69.9 < 101.2 < 108 < 126 The highest Concentration of heavy metal in this study. While the Concentration of heavy metal (Zn) in this study is 154.55, and in other rivers, the observed increasing trend of heavy metals (Zn) respectively were river 4 < river 7 < river 2 < river 3 < river 6 < river the value of the concentrations are 1.2–6.1 < 48 < 67.8 < 93.1 < 303 < 502.3 in this is study the Concentration was moderate from Among the studies other studies. While the Concentration of heavy metal (Pb) in this study is 56.35, and in other rivers, the observed increasing trend of heavy metals (Pb) respectively was river 3 < river 7 < river 2 < river 5 < river 6 < river 8 the value of the Concentration are 4.3 < 22.6 < 26.7 < 49.2 < 79.8 < 230.8 in this is to study the Concentration was moderate.

Table 8. Comparison of average heavy metals concentration in sediment of Brantas River with other studies

NO	River	Concentration (mg/kg)				Ref
		As	Cr	Zn	Pb	
1	**Brantas River**	67.133	146.5	154.55	56.35	This study
2	**Ganga River**	-	69.9	67.8	26.7	[34]
3	**Cauvery, India**	-	38.9	93.1	4.3	[6]
4	**Tapti, India**	-	-	1.2–6.1	-	[35]
5	**Yangtze, China**	-	108	230.4	49.2	[36]
6	**Buriganga, Bangladesh**		101.2	502.3	79.8	[37]
7	**Euphrates, Iraq**	-	58.4	48	22.6	[32]
8	**World average**	-	126	303	230.8	[24]

CONCLUSION

The purpose this study was to determine Sources and concentration levels of heavy metals of Brantas River sediments for the purpose of prioritizing future river remediation efforts, In general, heavy metal levels in the river 1 were exceedingly higher than the river 2, The analysis showed that the concentration value in the size of granules is 0.200 µm higher than the size of granules 0.063 µm due to the large surface of the granules. Where it was used Geographic accumulation index, Contamination factor and degree of contamination, Metal pollution index and Enrichment factor to assess heavy metal pollution of Brantas River sediments.

Recommendation

It should be given attention to strategies reduce pollution and remediation efforts:

- Full awareness using pesticides and organic fertilizers instead of chemical.
- Use alternative means of pesticides by planting a type of insect repellent plant.

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