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ASSESSMENT OF HEAVY METAL POLLUTION STATUS IN THE MIDDLE SECTOR OF THE CITARUM RIVER

Eka Wardhani[1], Raden Tina Rosmalina[2], Diana Rahayuning Wulan[3], Fitri Dara[3], Ragil Naga Lanang[4], Athaya Zahrani Irmansyah[5]

[1,4] Environmental Engineering Study Program, Bandung National Institute of Technology, Indonesia;
[2,3] Research Center for Environmental and Clean Technology, National Research and Innovation Agency, Republic of Indonesia;
[5] Civil Engineering Study Program, Bandung National Institute of Technology, Indonesia

* Corresponding Author, Received: 6 June 2023, Revised: 21 July 2023, Accepted: 16 Aug. 2023

ABSTRACT: River quality monitoring in Indonesia is still dominated by checking pollutant concentrations in river water. At the same time, sediment is also essential in providing an overview of the impact of various pollutants entering river water. Thus, it is necessary to review the heavy metal pollution status. Heavy metal pollution in river sediments can be problematic because it negatively impacts living organisms. Conversely, it also reveals the aquatic pollution by its depth layer. The critical parameters used in this study to determine the status of heavy metal pollution are Fe, Cd, Cr, Cu, Hg, Pb, and Zn at 40-60 cm depth. Sediment samples were taken from 3 locations in the Citarum River, Purwakarta Regency segment. The average measured concentration of heavy metals (mg/kg) in October 2022 was Fe: 527.425; Cd: n.d.; Cr: 0.214; Cu: 0.153; Hg: 0.012; Pb: 0.052; Zn: 1.671 and in November 2022 was Fe: 496.161; Cd: n.d.; Cr: 0.214; Cu: 0.164; Hg: 0.009; Pb: n.d.; Zn: 1.442. Contaminant Factor, Pollution Load Index, and Geoaccumulation Index calculations were used to determine the status of heavy metal pollution in water sediment in the Citarum River. The Contaminant Factor indicates that the sediment is low polluted, except for the parameter of Hg, which is categorized as moderately polluted. The Pollution Load Index indicates that heavy metal sediment in the Citarum Watershed is not polluted.

Keywords: Heavy metal, Sediment, CF, PLI, Igeo

1. INTRODUCTION

Citarum River has become the primary water source for West Java and Jakarta Province inhabitants, with 297 km from Situ Cisanti to its downstream at Muara Gembong, across 13 regencies/cities along its waterway. Many domestic, agricultural, and industrial activities around the river catchment area have influenced the Citarum water quality [1].

Heavy metal occurrence in water and sediment endangers the environment significantly when its concentration exceeds the standards [2]. Heavy metal is a pollutant that enters the river body and tends to stay in the sediment. Heavy metal in sediment emerges serious pollution problems due to its adverse effect on aquatic organisms and human health, even in low concentrations [3].

Sediment is a place for various aquatic organisms, which plays a significant environmental component—sediment is also the main accumulator when chemical pollutants enter the environment [4]. Heavy metal would not remain accumulated in the sediment but could also be released through chemical or biological reactions into the water phase. As [5] mentioned, heavy metal accumulation in sediment would be a sensitive indicator for contaminant monitoring in aquatic ecosystems. It also reveals the history of heavy metal sinks in every depth [6].

Studies on heavy metal monitoring in sediment depth describe heavy metal pollution and accumulation during specific periods, such as dry, rainy, or other seasons. However, it needs to be adjusted according to the specific requirements of the research. So, studying heavy metals in different sediment depths becomes essential to give more insight into the long-term pollution and describe the heavy metals accumulated there [7].

This study is focused on sediment from the Citarum River-Purwakarta segment, the middle sector of the Citarum catchment area [8]. It was assumed that this area received more heavy metal from human activity at the upstream area. Previous studies have revealed heavy metal concentration in Citarum water quality [9], but still limited studies on sediment. Heavy metals of iron (Fe), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), and zinc (Zn). The heavy metals Fe, Cd, Cr, Cu, Pb, Hg, and Zn are commonly chosen for sediment river studies due to their environmental persistence and potential toxicity. These metals can harm aquatic life and human health, and their accumulation in sediments can have long-term effects on the ecosystem [10]. This study not solely monitor heavy metal concentration in sediment but also enriches the data about pollution source to support the management effort by related stakeholder.

2. RESEARCH SIGNIFICANCE

Purwakarta Regency is a regency in West Java Province geographically located as in Fig. 1. This study aims to determine the status of heavy metal pollution in sediments of the Citarum River, Purwakarta Regency, West Java Province. The methods used to determine pollution status are the Pollution Load Index (PLI) and Geo-accumulation Index (I_{geo}). This study will identify the pollution level and take necessary actions to maintain and improve the river's water quality.

3. MATERIALS AND METHODS

3.1 Study Site

Purwakarta is one of the regencies in West Java Province, geographically located at 107°30’-107°40’ BT dan 6°25’-6°45’ LS. This study collected the sample from three locations (Table 1), and we can see it geographically in Fig.4 at three different depths, i.e., 40cm, 50cm, and 60cm. The depth was based on one mid-surface segment, assuming the concentration was more contaminated during short-period changes of the season than subsurface [6]. The river flow velocity is obtained using a current meter device with the Flowwatch-03 model calibrated through a flow test, which involves placing the impeller in a flow with a known velocity value. The cross-sectional profiles at each location can be seen based on Fig.1 to Fig.3, obtained using a spatial data approach from Tanah Air Geospatial. Based on geological conditions, the soil type in the study site is categorized as quartz sand [10].

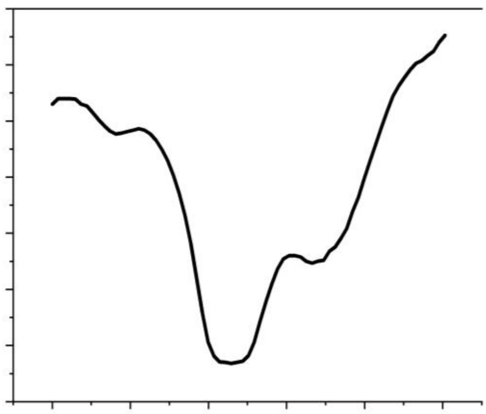

Fig.1 Cross-section profile of river (PWT 1)

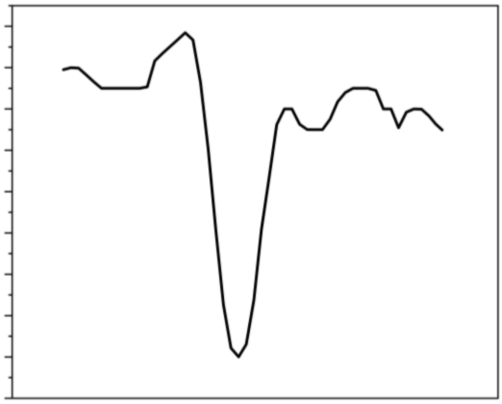

Fig.2 Cross-section profile of river (PWT 2)

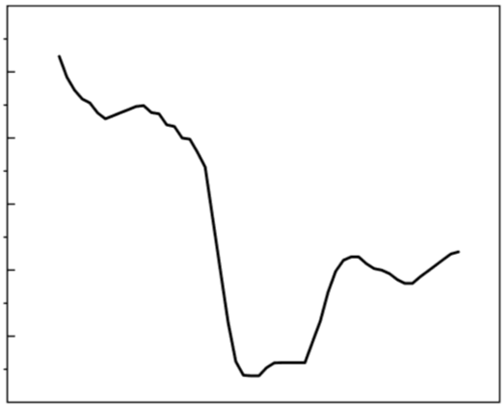

Fig.3 Cross-section profile of river (PWT 3)

Table 1 Sampling location

Sampling Point	Coordinates	Flow Velocity (m/s)	
		October	November
PWT 1	Outlet Jatiluhur: 6°31’07.5”S, 107°23’20.8”E	0.491	0.518
PWT 2	Outlet Domestic area: 6°30’38.83”S, 107°24’43.75”E	0.831	0.866
PWT 3	Downstream textile industry: 6°29’2.28”S, 107°22’54.33”E	0.884	1.101

3.2 Sample Collection and Preparation

Samples were collected from the study site in October 2022 and November 2022. Sediment samples were collected at 40-60 cm depth with a 10 cm interval above the riverbed surface using a hand core sampler and scope, then stored in clean zipped polyethylene plastic bags [11], already appropriately labeled. All samples were preserved in a cooling box.

In the last analysis, collected sediment samples were dried at room temperature to prevent chemical losses. Dried samples were ground using a mortar to simplify the sample sieving process and sieved using

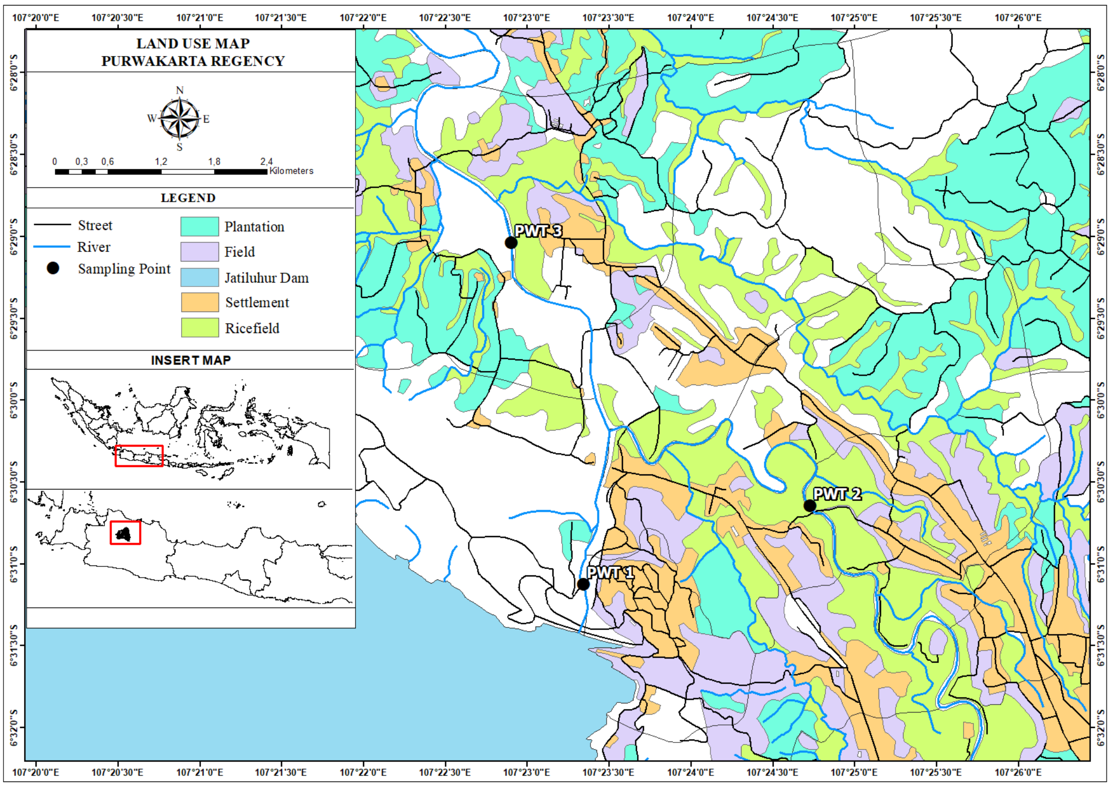

Fig.4 The location of the sampling site and land use in Citarum River

a 200 mesh (aperture 0.0074 mm) shaker sieve to get a homogenous sample. Generally, this soil contains silt and clay, which absorb more heavy metals in sediment [12].

In the last analysis, collected sediment samples were dried at room temperature to prevent chemical losses. Dried samples were ground using a mortar and sieved using a 200 mesh (aperture 0.074 mm) shaker sieve to get a homogenous sample. Generally, this soil contains silt and clay, which absorb more heavy metals in sediment [13].

To digest the heavy metal in samples, a modified EPA 3052 method was applied [14], where 0.45-0.5 g dried sample was placed in a digestion vessel, added 9 ml HNO_3, 2 ml HF, and 2 ml H_2O_2, and digested for 45 min, then allow until room temperature. Afterward, the sample was placed in a 50mL volumetric flask and adjusted with MilliQ water. Soluble heavy metals Fe, Cd, Cr, Cu, Pb, Hg, and Zn were ready to analyze using Agilent Technologies 200 Series AA. The same treatment was applied for blank without sediment samples.

The dried sediment samples were ground with a mortar and pestle and then sieved using a 200 mesh (aperture 0.0074 mm) shaker.

3.3 Pollution Load Index (PLI)

Pollution Load Index (PLI) is one indicator index used to determine heavy metal pollution status in sediment. PLI is calculated by determining each targeted heavy metal's Contamination Factor (CF). CF is the ratio between heavy metal concentration in measured sediment to natural background concentration. This CF value would represent the pollution level of the aquatic ecosystem, even low, moderate, or highly polluted [15]. PLI and CF are calculated using Eq. (1) and Eq. (2).

$$CF = \frac{Mc}{Bc} \quad (1)$$

$$PLI = (CF_1 \times CF_2 \times \ldots \times CF_n)^{1/n} \quad (2)$$

Table 2 Pollution category of CF and PLI value [15]

CF and PLI Value	Pollution Category
CF ≤ 1	Low contamination
1 < CF ≤ 3	Moderate contamination
3 < CF ≤ 6	Considerable contamination
CF > 6	Very high contamination
PLI < 1	Unpolluted by heavy metals combination
PLI > 1	Polluted by heavy metals combination

Where Mc is the measured heavy metal concentration in sediment (mg/kg), Bc is the natural heavy metal concentration (mg/kg), and n is the total measured heavy metal. In this study, the natural background is referred to a previous study [16], where Fe, Cd, Cr, Cu, Hg, Pb, and Zn (mg/kg) are 9,800, 0.3, 35, 45, 0.03, 7, and 16, consecutively. The pollution category is classified in Table 2.

3.4 Geo-accumulation Index

The general classification for evaluating heavy metal pollution levels in sediment is the geoaccumulation index, I_{geo}. This index compares the existing measured concentration to pre-industrial concentration. I_{geo} is calculated using Eq. (3) [17].

$$Igeo = log_2\left(\frac{M_C}{1.5 \times B_C}\right) \quad (3)$$

Where Mc is the measured heavy metal concentration in sediment, Bc is the natural heavy metal concentration, and 1.5 is the correction factor for natural fluctuation.

Pollution level classification represented by I_{geo} is shown in Table 3.

Table 3 Pollution level of I_{geo} index

Range of I_{geo}	Pollution level
$I_{geo} \leq 0$	Practically unpolluted
$0 < I_{geo} \leq 1$	Unpolluted-moderately polluted
$1 < I_{geo} \leq 2$	Moderately polluted
$2 < I_{geo} \leq 3$	Moderately to heavily polluted
$3 < I_{geo} \leq 4$	Heavily polluted
$4 < I_{geo} \leq 5$	Heavily to extremely polluted
$I_{geo} > 5$	Extremely polluted

4. RESULTS AND DISCUSSIONS

4.1 Heavy Metal Concentration

Fig.5 shows the measured heavy metal concentration in each sediment depth. For Fe, the concentration range is between 337.419-759.887 mg/kg dry weight, with an insignificant difference between October and November. The highest concentration resulted at PWT 2-mid with 759.887 mg/kg dry weight in October and the lowest at PWT 1-mid and PWT 1-lower with 337.419-337.924 mg/kg in October. This high concentration is caused by rice field land usage around the sampling point (Fig.1). Domestic activity, such as metal-equipment washing, might also contribute. Although increasing concentration occurs, all Fe sediment is still below the acceptable 20,000 mg/kg level. The concentration of Fe in the literature [9] is used as a reference because it has the same river basin, but Fe is not permitted at this point.

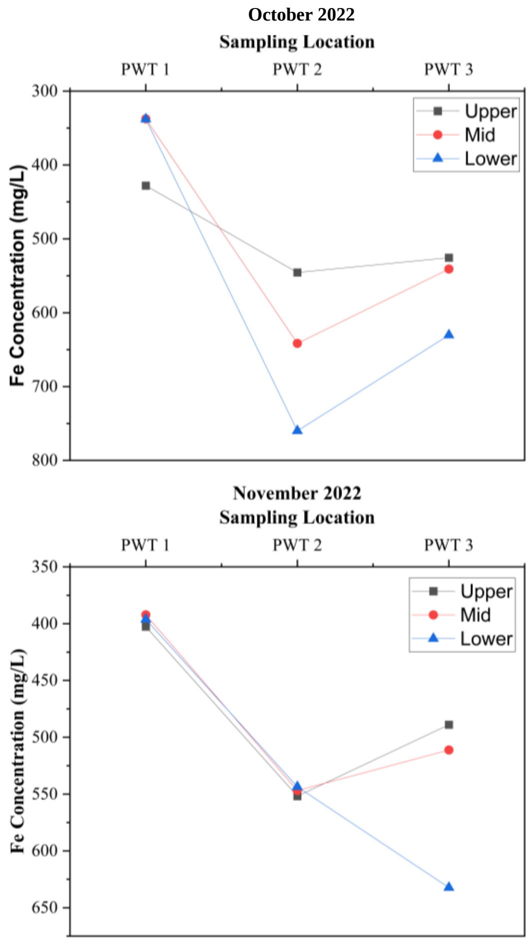

Fig.5 Fe concentration of each sampling point

Cadmium concentration at all sampling points was below the Limit of Detection/LoD 0.002 mg/land and below acceptable sediment standard 1.5 mg/kg. The Cd concentration in the Citarum River (Nanjung) sediment has a Cd value of 13 mg/L as a comparison [9].

Meanwhile, chromium concentrations ranged between 0.114-0.508 mg/kg dry weight. The highest Cr concentration resulted from PWT 1-upper in November with 0.508 mg/kg; the lowest concentration was showed by PWT 3-lower in November with 0.114 mg/kg. In this case, chromium concentration exists mainly by sediment weathering around the study site because no electroplating and leather industry exists. Based on the standard, these Cr concentrations are still below the acceptable 80 mg/kg value. The concentration of Cr in Citarum River (Nanjung) has a value of 13 mg/L as a comparison.

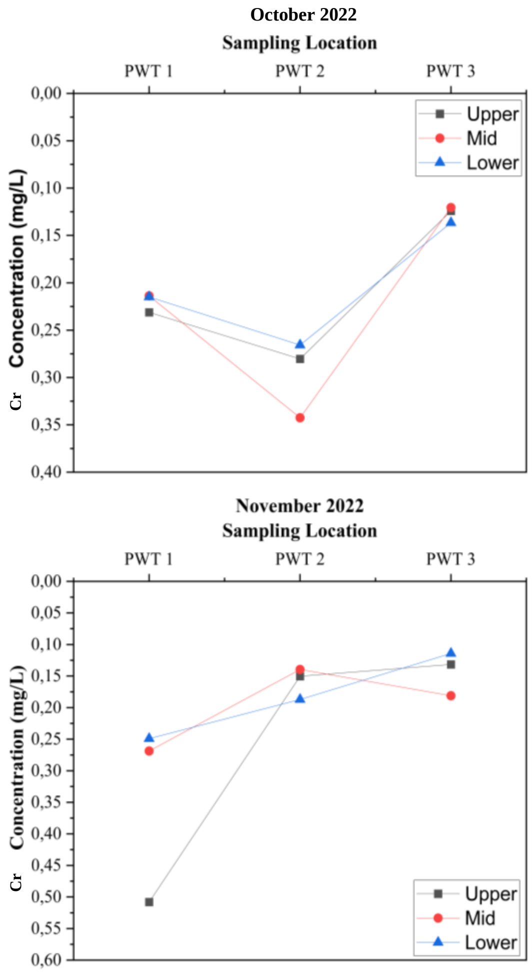

Fig. 6 Cr concentration of each sampling point

The measurement of lead concentration was < 0.03-0.116 mg/kg dry weight. The highest concentration resulted in the PWT 1-lower October sample with 0.116 mg/kg dry weight, even below the Pb standard of 50 mg/kg. Meanwhile, all October samples from PWT 3 concentration were measured below LoD 0.03 mg/L. These low Pb concentration was getting lower in all November samples. Low Pb concentration is caused by precipitation, which affects the sediment deposition rate. Other factors such as volume, time, and rainfall intensity can influence the Pb deposition rate in sediments [18]. The concentration of Pb in the sediment of the Citarum River (Nanjung) has a Pb value of 22,9 mg/L as a comparison [9].

Zinc measurement ranged between 0.861-3.108 mg/kg dry weight. The highest concentration showed a sample from PWT 3-upper in October with 3.108 mg/kg dry weight. Meanwhile, the lowest concentration resulted from the PWT 1-mid sample in November with 0.861 mg/kg dry weight. These Zn concentrations are still below the sediment standard of 200 mg/kg dry weight.

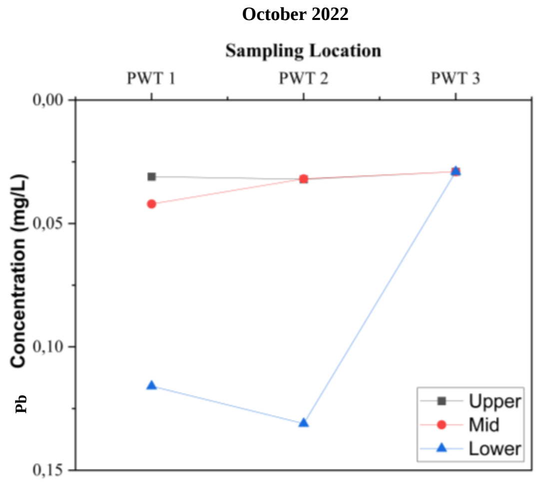

Fig. 7 Pb Concentration of each sampling point

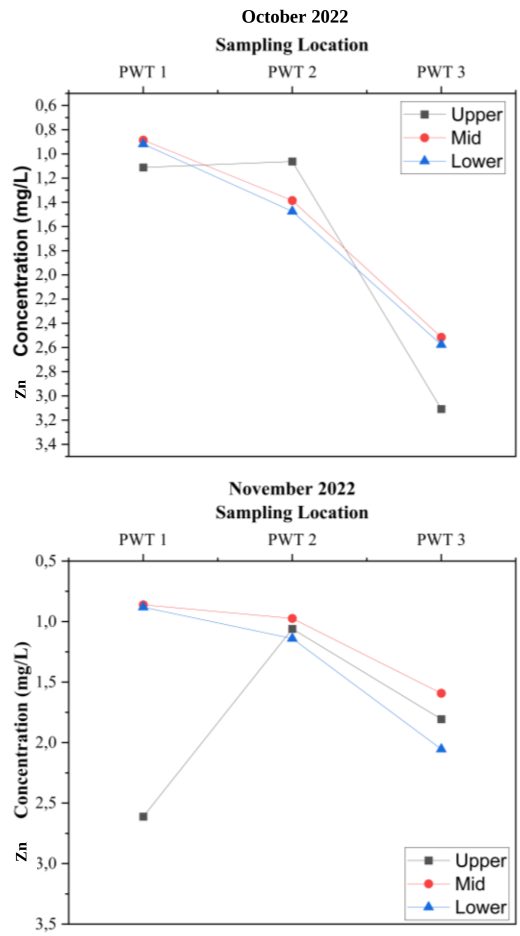

Fig. 8 Zn Concentration of each sampling point

4.2 Contaminant Factor Result

Measured heavy metal concentrations are included in CF calculation for each sampling point (Table 4), which later on will be used for PLI calculation. Based on Table 3, the CF value in October and November ranges between 0.001-1.697. A high value of 1.697 resulted from the

PWT-2 upper October sample by Hg contribution. Since the Hg background concentration of 0.03 mg/kg dry weight is relatively small compared to other heavy metals background concentrations not so commonly found in nature, so when the sample has slightly passed the background concentration, the CF value directly affected and caused the sample into moderate contaminated by Hg category.

Other heavy metal concentrations, such as Fe, Cd, Cr, Cu, Pb, and Zn, were found to be much lower than their background concentration, so their CF is categorized as low contamination by those heavy metals.

4.3 Pollution Load Index Result

Based on CF calculation, PLI was determined to categorize the pollution level at each sampling point [15]. Fig.9 shows the PLI range between 0.012-0.028, which means all samples at all sampling periods have PLI < 1.

It concluded that even the CF on the PWT-2 sample was moderately polluted by Hg, but all sampling points were still categorized unpolluted by the combination of seven targeted heavy metals.

Based on Table 5, it can be seen that the PLI value for each sampling location has a range between 0.012-0.028. The metal concentrations of Fe, Cd, Cr, Cu, Hg, Pb, and Zn measured in PWT 1, PWT 2, and PWT 3 have a PLI value of less than 1 (PLI < 1). The PLI value < 1 indicates that a combination of several metals does not pollute the sediments of the Citarum River Segment of Purwakarta Regency.

4.4 Geo-accumulation Index Result

To determine whether industrial activities caused the existing heavy metals to accumulate, the I_{geo} index was applied. Based on the measured concentration, its natural background concentration, and the correction factor in Eq.(3), I_{geo} was calculated as shown in Table 5.

Table 5 shows that I_{geo} by Fe, Cd, Cr, Cu, Pb, and Zn at all sampling points resulted in < 0, categorized as unpolluted. But, because PWT-2 upper in October has an Hg concentration that exceeded the background concentration, the I_{geo} 0.178 put this sampling point into the unpolluted to moderately polluted category. The possibility of industrial activities on the upper side of PWT 2 should be studied further since there is no mining activity (Fig.4). Based on three indexes CF, PLI, and I_{geo} on seven targeted heavy metals on sediment at three sampling points on the mid-surface level, which are usually contaminated by industrial activities and affected by transitional season, this study revealed that some locations already moderately contaminated by Hg-included activity. Even in the early wet season, when the precipitation mostly washes out some heavy metals from sediment, it was found that some heavy metal remains existed there. Mitigation actions like industrial wastewater treatment regulation should be addressed to maintain safe and healthy water and sediment.

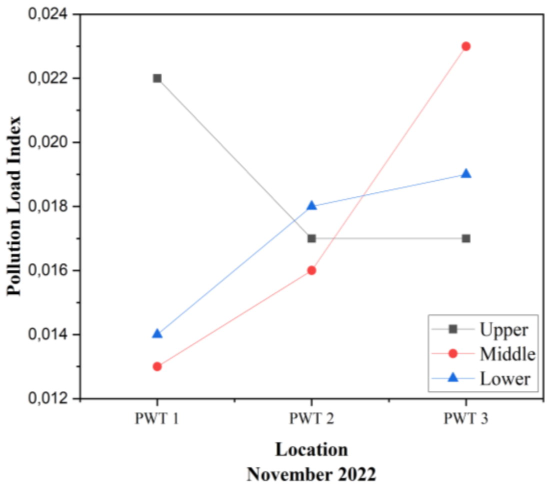

Fig. 9 Pollution Load Index Result

5. CONCLUSION

There are differences in the metal concentrations of Fe, Cd, Cr, Cu, Hg, Pb, and Zn contained in the studied sediments. The heavy metals concentration varies in two sampling periods, caused by anthropogenic and natural activities, e.g., rice fields, domestic activity, and sediment weathering. Based on research using the contamination factor, the sediment is included in the moderately polluted category, except for the Hg parameter, which is included in the moderately polluted category. Overall, the PLI value for each measurement location is less than one, so it is classified that the water sediments in the Citarum River segment of Purwakarta Regency are not polluted by a combination of the seven metals analyzed. The measured I_{geo} value has a value less than 0, which indicates that heavy metal pollution in river sediments is in the unpolluted category, except for Hg metal in PWT 2 in October with an I_{geo} value of 0.178, which indicates that heavy metal Hg has moderately polluted the sediment.

6. ACKNOWLEDGMENTS

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Table 4 Contaminant Factor in each sediment depth at sampling points

Sampling Point		CF on October 2022						
		Fe	Cd	Cr	Cu	Hg	Pb	Zn
PWT 1	Upper	0.044	0.006	0.007	0.001	0.065	0.004	0.070
	Mid	0.034	0.006	0.006	0.001	0.120	0.006	0.055
	Lower	0.034	0.006	0.006	0.001	0.102	0.017	0.057
PWT 2	Upper	0.056	0.006	0.008	0.005	1.697	0.005	0.066
	Mid	0.065	0.006	0.010	0.005	0.506	0.005	0.087
	Lower	0.078	0.006	0.008	0.005	0.491	0.019	0.092
PWT 3	Upper	0.054	0.006	0.004	0.005	0.092	0.004	0.194
	Mid	0.055	0.006	0.003	0.005	0.378	0.004	0.157
	Lower	0.064	0.006	0.004	0.004	0.127	0.004	0.161
Sampling Point		CF on November 2022						
		Fe	Cd	Cr	Cu	Hg	Pb	Zn
PWT 1	Upper	0.041	0.006	0.015	0.002	0.572	0.004	0.163
	Mid	0.040	0.006	0.008	0.001	0.120	0.004	0.054
	Lower	0.040	0.006	0.007	0.001	0.178	0.004	0.055
PWT 2	Upper	0.056	0.006	0.004	0.005	0.178	0.004	0.066
	Mid	0.056	0.006	0.004	0.005	0.190	0.004	0.061
	Lower	0.055	0.006	0.005	0.005	0.224	0.004	0.071
PWT 3	Upper	0.050	0.006	0.004	0.004	0.141	0.004	0.113
	Mid	0.052	0.006	0.005	0.005	0.960	0.004	0.100
	Lower	0.065	0.006	0.003	0.005	0.241	0.004	0.128

Table 5 Geo-accumulation Index at each sampling point

Sampling Point		Sampling period October 2022						
		Fe	Cd	Cr	Cu	Hg	Pb	Zn
PWT 1	Upper	-5.102	-7.888	-7.827	-10.613	-4.531	-8.404	-4.432
	Mid	-5.445	-7.888	-7.940	-10.728	-3.642	-7.962	-4.760
	Lower	-5.443	-7.888	-7.933	-11.376	-3.882	-6.501	-4.708
PWT 2	Upper	-4.752	-7.888	-7.549	-8.315	0.178	-8.354	-4.498
	Mid	-4.518	-7.888	-7.260	-8.214	-1.569	-8.363	-4.115
	Lower	-4.274	-7.888	-7.626	-8.328	-1.612	-6.324	-4.024
PWT 3	Upper	-4.806	-7.888	-8.725	-8.352	-4.030	-8.500	-2.949
	Mid	-4.764	-7.888	-8.767	-8.318	-1.989	-8.500	-3.254
	Lower	-4.544	-7.888	-8.586	-8.413	-3.563	-8.500	-3.220
Sampling Point		Sampling period November 2022						
		Fe	Cd	Cr	Cu	Hg	Pb	Zn
PWT 1	Upper	-5.190	-7.888	-6.691	-9.895	-1.390	-8.500	-3.200
	Mid	-5.228	-7.888	-7.610	-10.036	-3.649	-8.500	-4.801
	Lower	-5.213	-7.888	-7.719	-10.093	-3.076	-8.500	-4.768
PWT 2	Upper	-4.736	-7.888	-8.448	-8.307	-3.076	-8.500	-4.501
	Mid	-4.748	-7.888	-8.556	-8.323	-2.983	-8.500	-4.623

	Lower	-4.757	-7.888	-8.132	-8.274	-2.745	-8.500	-4.396
PWT 3	Upper	-4.910	-7.888	-8.641	-8.393	-3.406	-8.500	-3.732
	Mid	-4.846	-7.888	-8.179	-8.237	-0.644	-8.500	-3.914
	Lower	-4.539	-7.888	-8.846	-8.287	-2.638	-8.500	-3.547

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