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Regional Case Study

Identification of Heavy Metal Concentration in Citarum River Water Using the Heavy Metal Pollution Index Method

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Abstract

The Citarum River is the largest and longest river in West Java Province. The basin in that location has influenced by human activities and the industrial sector. One of the hazardous pollutants contained in industrial wastewater is heavy metals. This study aims to identify the content of dissolved heavy metals (Fe, Cd, Cr, Cu, Hg, Pb, and Zn) in Citarum River water, Purwakarta Regency, at three observation points, namely Jatiluhur Reservoir Outlet, Cilalawak Bridge, and Cilele Bridge. The Atomic Absorption Spectrophotometry Method is used to analyze the dissolved heavy metals. Determination the level of metal contamination using the Heavy Metal Pollution Index (HPI) method. The study results that three metals exceeded the standard based on Government Regulation 22 of 2021 Class II, namely Fe, Hg, and Zn. Fe metal in the range between 0.277-1.179 mg/L, Hg metal between 0.011-0.100 mg/L, and Zn metal between 0.017-0.074 mg/L. HPI analysis showed that at three points in both sampling periods, it indicated that heavy metals highly polluted river water because the results obtained exceeded the HPI pollutant index, which was > 100. The biggest contribution of dissolved heavy metals in the Citarum River water body was dominated by the Hg parameter.

Keywords: Citarum; heavy metal; pollution index

1. Introduction

The Citarum River flows from Mount Wayang, south of Bandung City to the Java Sea in Karawang Regency, which has a length of about 297 km and an area of \pm 11,323 km². The Citarum River Basin crosses 11 regencies/cities, including the regencies of Bandung, West Bandung, Purwakarta, Karawang, Bekasi, Bandung City, Cimahi, some of which are in Sumedang, Cianjur, Bogor, and Garut regencies. Geographically, the Citarum River is located at 106°51'36"-107°51' East Longitude and $7°19'$ -6°24' South Latitude (Juniarti, 2020).

The Citarum River Basin is an object that has an important function for the community in West Java Province, such as the Cirata, Jatiluhur, and Saguling Reservoirs. The Jatiluhur Reservoir is located in Purwakarta Regency which is a vital tourist object that is used as a hydroelectric power plant, a supplier of raw water for drinking water, irrigation, tourism, and fisheries (Sari et al., 2020). The Citarum River Basin is an area with various activities of domestic, agriculture, industry, and mining. Waste from human activities

causes the water quality of the Citarum River to be heavily polluted; some metals do not meet their standard quality (Wardhani et al., 2021). Domestic activities contributed 40% of the Citarum River's waste; the remaining 60% is contributed from industrial wastewater which discharges into water body. One of the hazardous contaminants contained in industrial wastewater is heavy metals. Type of industries that potentially contribute to heavy metal waste are. metal processing, textile, wood processing, leather tanning, fertilizer factory, and so on (Febrita and Roosmini, 2022).

Heavy metals are the most common type of environmental pollutant found in water. Naturally, heavy metal elements are present in waters in small concentration. Heavy metal levels will increase when waste containing heavy metal enters the aquatic environment, making it toxic to aquatic organisms (Rennika et al., 2013). River pollution occurs continuously, such as heavy metals whose presence is very important to study because they are non-degradable, persistent, and can accumulate in the bodies of living things (Wardhani et al., 2016). Heavy metals which bound in organisms body will affect the biological processes of these aquatic organisms, including humans, through the food network pathway (Patty et al., 2018). The Citarum River is the main water source for the Saguling Reservoir in addition to six other rivers, namely the Cipatik, Ciminyak, Cijere, Cijambu, Cimerang, and Cihaur Rivers (Wardhani et al., 2021). The water quality of the Citarum River is categorized as heavily polluted, even several heavy metals were monitored as not meeting quality standards including heavy metals (Wardhani et al., 2023)

Research conducted by Prayoga et al. (2022), regarding the level of contamination in the Jatiluhur Reservoir stated that heavy metals Mercury (Hg), Zinc (Zn), Lead (Pb), Cadmium (Cd), and Copper (Cu) were detected in sediments caused by household and industrial waste discharge. Other research results also stated that heavy metals Chromium (Cr), Iron (Fe), Cu, and Zn were identified in rice and river water samples located in the upper Citarum area (Febrita and Roosmini; Handayani et al., 2022). In order to understand the pollution level of study location and considering that Citarum River water in the Purwakarta Regency sector is needed for daily life, society, and as a habitat for aquatic organisms, this study aims to identify the level of heavy metal contamination, especially for dissolved metal parameters. This study suggests the urgency on evaluating the Citarum River quality monitoring system and the effluent discharged into the water body which can also affect the content of heavy metals in the middle and downstream of the Citarum River. In this study, dissolved heavy metals in middle segment of Citarum River in the Purwakarta Regency is analyzed to determine the heavy metal pollution. This information will be important for the management of the Citarum River.

Heavy metal analysis in this study is used the Atomic Absorption Spectrophotometry (AAS) Method. Khopkar (2003) states this method has high analytical sensitivity and selectivity. In addition, this method is often used in analysis because it is easy to have a small linear range with a large detection limit, and the analysis cost is cheap compared to the Inductively Coupled Plasma (ICP) method (Gracia and Baez, 2021). This method can detect the presence of heavy metals in levels that are difficult to detect with other methods. The working principle of the AAS method is to use the principle of absorption of light wavelengths by atoms. The atoms absorb light at specific wavelengths, depending on the nature of the elements. This AAS method requires a radiation source that emits light at the correct wavelength in absorption (Djunaidi, 2018). The purpose of conducting this research was to identify and analyze the content of dissolved heavy metals consisting of parameters Fe, Cd, Cr, Cu, Hg, Pb, and Zn in Citarum River and to determine the Citarum River water quality index that has been analyzed.

2. Methods

A literature study including land use map evaluation of 2002 Purwakarta Regency Regional Spatial Plan was conducted, also included the characteristics of each heavy metal study and its properties in water. Then, determine the heavy metals using the AAS method and compare with regulations on river water quality standards based on Government Regulation 22 of 2021 concerning the Implementation of Environmental Protection and Management.

Water samples were taken on latest October 2022 (period 1) and mid November 2022 (period 2) at three points of Citarum River in the Purwakarta sector which have been determined, i.e. outlet Jatiluhur Reservoir, Cilalawak New Bridge, and Cilele Bridge, where detail sampling point are showed in Table 1. and Figure 1.

Water sampling refers to Indonesian National Standard (INS) number 6989.57:2008 about surface water sampling methods. Samples were taken at three points each at a distance of $\frac{1}{4}$ the width of the river (L) at a depth of 0.2 times the depth (d) from the sample surface to the bottom evenly. Water sample was collected by composite method. At each station point, one liter of water is taken at a depth of 0.2-0.5 meters using horizontal water samplers. Then, the water sample is collected in a plastic bucket before placing it in a 1L polypropylene (PP) bottle and then preserved to pH<2 by adding concentrated HNO3 solution (Merck, 69%). The sample is filled until full to prevent oxygen from entering during sample transfer from location to laboratory and stored in cool box prior to laboratory. Details of the analytical method for water samples for heavy metals parameters are presented in Table 1. The location of the sampling stations in this study is presented in Table 2.

Table 1. Water sample analysis method for dissolved heavy metal parameters

No	Parameter	Reference	Method
	Fe	INS number 6989.4:2009	AAS
2.	Cd	INS number 6989.16:2009	
3.	Cr	INS number 6989.71:2009	
4.	Cu	INS number 6989.6:2009	
5.	Hg	INS number 6989.78:2019	
6.	Ph	INS number 6968.8:2009	
7.	Zn	INS number 6968.7:2009	

Table 2. Location of heavy metal sampling point

Water velocity data was obtained using the current meter used in each cross-sectional section of the river. Measurements were made at the time of collecting water samples. According to Putra (2016), the velocity of river flow is influenced by many things, including friction with the land, wind, river contours, river location, and disturbances such as weeds, garbage or algae plants that grow in rivers.

Figure 1. Location of sampling points for the Citarum River Sector, Purwakarta Regency

The heavy metals measurement were dissolved metals using the AASWhich was conducted at Advanced Characterization Laboratory of National Research and Innovation Agency, Cisitu, Bandung. Before analysis, a filtering treatment must be carried out using a 0.45 μm filter paper and a vacuum filter device to remove all suspended solid. The sample should free from suspended particles also to prevent interference on the AAS. After filtering the sample, analysis of Fe, Cd, Cr, Cu, Pb, and Zn parameters was conducted directly. Meanwhile for the Hg parameter, the destruction treatment must be carried out before measurement.

Destruction is a sample heating treatment that aims to decompose organic into inorganic metals. The acid destruction method used is the open digestion method, carried out by heating a mixture of sample water and an acid reagent over a water bath (Hasmizal and Bhernama, 2019). Hg metal will not well atomized in AAS-flame, so to analyze Hg, a special method called cold vapor atomization method (AAS-cold vapor) is used. Two preparation processes for making the Hg test solution are acidification and reduction. The acidification process is done by adding strong acids, which maintain Hg metal in a dissolved form. The following process is reduced by adding a reducing agent, namely SnCl₂, to reduce Hg(II) to Hg(I) or Hg(o). Both of these treatments were carried out to measure using AAS-cold vapor (Firdausiyah, 2015). The preparation steps for making the Hg test solution refer to INS number 698.2019.

Calculations were made on the data of seven dissolved heavy metals to determine the pollution status at each sampling point using the HPI method. This method can be calculated by equations 1 to 3 as follows (Appiah-Opong et al., 2021).

$$
HPI = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i}.
$$
 (1)

Wi is the unit weight given for each heavy metal and Qi is the sub-index of the i-th heavy metal. Wi calculated from the equation 2.

$$
W_i = \frac{k}{s_i}.\tag{2}
$$

k is the proportionality constant ($k = 1$) and Si is the standard recommended value for the *i*-th heavy metal. The sub-index value (Qi) is obtained by equation 3 as follows.

$$
Q_i = \left(\frac{M_i}{S_i}\right) \times 100. \tag{3}
$$

Mi is the actual measured value obtained from the laboratory analysis results, S_s is the ideal value, and S_i is the standard value of the i -th heavy metal sourced from water quality standards per applicable regulations. According to Sobhan Ardakani et al. (2016), the HPI risk threshold value is 100, and the pollution level is classified into three levels, i.e. low (HPI<100), at-risk threshold (HPI=100), and critical (HPI>100) presented in Table 3. HPI above 100 can cause greater health damage (Setia et al., 2020).

Table 3. HPI value classification

Source: (Sobhanardakani et al., 2016)

3. Result and Discussion

3.1. Characteristics of the Citarum River Sector in Purwakarta Regency

Based on the 2002 Purwakarta Regency Regional Spatial Plan, there are two watershed systems: the Cikao River Sub-watershed is a system of the Citarum River basin and the Ciheang River Basin is a system of a Cimalaya River basin. The two watershed systems consist of tributaries streams of that have the potential to become the primary network of rainwater drainage systems in urban areas. The water flow pattern of the tributaries around the planning area flows from the Cikao River. It ends in the Citarum River at a position after the Jatiluhur Dam (downstream).

The scope of the study area only focuses on three monitoring station points with a flow direction to the north, i.e. PWT 1 station located in outlet Jatiluhur Reservoir, PWT 2 station at Cilalawak New Bridge, and the last point is PWT 3 station at Cilele Bridge. The location point of the PWT 1 station represents the upstream part of the river flow, the PWT 2 station is the middle part of the river flow, and finally, the point of the PWT 3 station shows the downstream part of the river flow.

The location of first station is located near the Jatiluhur Reservoir outlet in Jatimekar Village, Jatiluhur District. Forest areas and settlements dominates this location. In addition, there are the Perum Jasa Tirta II office of hydroelectric power plant business unit and textile industry which manufactures spinning nylon yarn, textures, and fibers.

The second sampling station was at Cilalawak Bridge. The dominant activities in this area are rice fields and settlements, which water runoff from those locations flows into Citarum River. In addition, several building materials and textiles industries are located around the river.

The third sampling location is at the Cilele Bridge behind the textile industry. Activities in this area are mainly industrial areas. Two major industrial areas in the watershed are the manufacturing wood-based fibers industry and the textile sector of manufacturing synthetic fibers.

The physical condition of the river is measured by the velocity of the river flow, as presented in Table 4; according to Sari and Usman (2012), the current speed is divided into four categories, namely slow currents with speeds in the range of 0-0.25 m/sec, medium currents with speeds in the range of 0.26-0.50 m/second, fast currents with speeds in the range of 0.6 -1.0 m/second, and swift currents with speeds above 1 m/second (Sari and Usman, 2012). Based on the flow velocity measurement results, PWT 1 is included in the moderate flow category, PWT 2 is included in the fast flow category, and PWT 3 period one is included in the fast flow category. PWT 3 period two is included in the high-speed flow; according to Philip et al. (2018), metal levels in water tend to be influenced by patterns of water currents. If the current speed is high, it can result in the spread of dissolved heavy metals in all directions.

Sampling Point	Water Velocity		
	(m/second)		
	Period 1	Period 2	
PWT_1	0.491	0.518	
PWT_2	0.866	0.831	
PWT_3	0.884	1.101	

Table 4. Water velocity

The analysis results of heavy metals in Citarum River water in all periods are presented in Table 5 and Table 6.

Sampling	Heavy Metal Concentration (mg/L)							
Point	Fe	Cd	$\mathbf{C}\mathbf{r}$	Cu	Hg	Pb	Zn	
PWT ₁	0.277	n.d.	n.d.	n.d.	$0.017*$	n.d.	0.018	
PWT ₂	$1.179*$	n.d.	n.d.	n.d.	$0.01*$	n.d.	$0.074*$	
PWT_3	$0.378*$	n.d.	n.d.	n.d.	$0.035*$	n.d.	0.039	
Quality	0.300	0.010	0.050	0.020	0.002	0.030	0.050	
Standard								
\mathbf{LoD}		0.002	0.010	0.005		0.030	$\overline{}$	

Table 5. Heavy metals concentration in Citarum Water River Purwakarta Sector (Period 1)

*Note: n.d. = not detected; *does not meet standard (Government Regulation No. 22 of 2021, Class II, Appendix VI)*

Table 6. Heavy metals concentration in Citarum Water River Purwakarta Sector (Period 2)

Sampling	Heavy Metal Concentration (mg/L)						
Point	Fe	Cd		Cu	Hα	Pb	Zn
PWT_1	$0.352*$	0.002	n.d.	n.d.	$0.062*$	n.d.	$0.112*$
PWT ₂	$1.093*$	n.d.	n.d.	n.d.	$0.035*$	n.d.	0.017

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PWT_3	$0.534*$	n.d.	n.d.	n.d.	$0.100*$	n.d.	$0.055*$
Quality	0.300	0.010	0.050	0.020	0.002	0.030	0.050
Standard LoD	$\overline{}$	0.002	0.010	0.005	$\overline{}$	0.030	$\overline{}$

*Note: n.d. = not detected; *does not meet standard (Government Regulation No. 22 of 2021, Class II, Appendix VI)*

The results of the dissolved metal measurement show that four parameters meet the quality standard with values that are not detected by the AAS or below the LoD value, i.e. parameters Cd, Cr, Cu, and Pb. In contrast, the parameters Fe, Hg, and Zn exceed the quality standards.

The results of the analysis of dissolved Fe measurements carried out in Citarum River water in the Purwakarta Regency sector are presented in Figure 2. Based on Figure 2, only one point does not exceed the quality standard in measuring Fe parameters, namely the PWT 1 station point in period one, and the remain result have exceeded the quality standard based on Government Regulation No. 22 of 2021 class II. Fe values that exceeded the quality standard were PWT 1 station point 2 period 2, PWT 2 period 1 and 2, and PWT 3 period 1 and 2. The highest value of Fe content was 1.179 mg/L at PWT station 2, period 1. Based on research conducted by Kirana et al. (2019), the upstream sector 7 Citarum River water was identified as containing dissolved Fe metal of 0.1 mg/L, which can be originated from the runoff of bedrock of the volcanic system.

Figure 2. Concentration of dissolved Fe in Citarum River Water

The results of the analysis of dissolved mercury in Citarum River water in the Purwakarta Regency sector are presented in Figure 3. Overall, dissolved mercury levels from the three observation points were above the quality standard based on Government Regulation No. 22 of 2021, namely the maximum quality standard value of 0.002 mg/L. According to research by Suteja et al. (2019), in the Banyuasin Estuary, Hg concentrations ranged from 0.001-0.032 mg/L, caused by a large number of agricultural, plantation, mining, and household activities in the upper reaches of the Lalan River. According to Yulis (2018), the high Fe content can be caused by Hg, which naturally comes from volcanic gases. In addition, it can also be caused by anthropogenic activities of gold mining operations using mercury or Hg as a gold-binding medium. Based on data from the Natural Resources and Mining and Energy Service of Bandung Regency (2023), there were illegal gold mining activities in Kotawaringin District. which are spread across 113 locations, namely 85 locations in Kotawaringin Village (74 of which are still operating) and 27 locations are in Cibodas Village. Untreated waste from those illegal activities which usually still contained mercury will be disposed into the water body. Other sources that can cause high levels of Hg in waters are industrial waste such as paper pulp, oil refining, paint, rubber and fertilizer processing, batteries, fabric softeners, fluorescent tubes, highintensity street lamps, peptides, cosmetics, and drugs medicine (Rahayu and Mangkoedihardjo, 2022). Based on study by Prayoga et al.(2022), the sediment concentration around Jatiluhur outlet, which is similar with PWT 1 point in this study, was contained Hg. According to water velocity measurement, PWT 1 was included in the moderate flow category. This flow category and other chemical or physical factors such as temperature or pH, the Hg content in the sediment might be released into the water.

Figure 3. Concentration of dissolved Hg in Citarum River Water

The results of the AAS method analysis of zinc parameters in Citarum River water in the Purwakarta Regency sector are presented in Figure 4. In period 1, station points exceeded the quality standard based on Government Regulation No. 22 of 2021 class II, namely at the PWT 2 station point. In the 2nd period, two observation points exceed the quality standards, namely at the PWT 1 and PWT 3 station points, while the rest still meet the maximum value of the quality standard. The high Zn content in PWT 1 can be assumed to originate from Jatiluhur Reservoir activities resulting from chemical fertilizer (Prayoga et al. (2022), Wulan et al. (2020) or fishing activities. The last activity was confirmed by research by Sari et al. (2020), which states that the decline in water quality is caused by high fish farming activities and exceeding the maximum capacity of the Jatiluhur Reservoir. The results of other dissolved Zn metal studies carried out in the waters of the Saguling Reservoir are researched by Adani et al. (2018). This study identified Zn content ranging from 0.144- 0.598 mg/L, which was thought to originate from the many industrial activities in the area. Zn metal is usually used as a chemical additive in the final refinement process and as a fiber preservative.

Figure 4. Concentration of dissolved Zn in Citarum River Water

3.1.1. HPI

The HPI value at the study site can be determined by entering the average concentration value of each heavy metal parameter generated at each monitoring station. Details of the results of HPI calculations monitored from the three station points in the two sampling periods are presented in Table 7 in period 1 and Table 8 in period 2.

Table 7. HPI calculation results in period 1

Tabel 8. HPI calculation results in Period 2

Heavy Metal	Quality Standard (Si)	Unit Weigh Value (Wi) $(1/Si)$	Monitor Value (Mi)	Sub Index Value (Qi)	Wi.Qi	HPI	Conclusio n
	$(\mu g/L)$	$(\mu g/L)$	$(\mu g/L)$	$(\mu g/L)$	$(\mu g/L)$		
PWT ₁							
Fe	300	0.003	351.8	117.27	0.39	2,154	The HPI
C _d	10	0.100	2.1	21	2.1		value is $>$

Source: (Analysis Results, 2023)

Based on the index analysis results from the three observation stations in the two periods, it can be concluded that all stations have HPI values greater than 100. This indicates that the river water at PWT 1, PWT 2, and PWT 3 stations is highly polluted by heavy metals. Based on the results of the HPI value analysis, it appears that the parameter of heavy metals that contributes significantly to the water body of the Citarum River is dominated by the parameter Hg. Sources of heavy metal Hg come from the agricultural and plantation sectors, such as the result of excessive use of pesticides. The HPI method analysis results are recapitulated in Table 9 and the HPI value graph is in Figure 5. The factor that caused the HPI value for period 2 to be higher when compared to period 1 was due to the contribution of heavy metal concentrations from the test results in period 2 being higher than in period 1. Based on the research of Wulan et al. (2020) and Wardhani et al. (2023), HPI results in the Cipeles River show low upstream pollution levels, moderate to high pollution in the city center, and highly polluted downstream. This is due to the high level of zinc concentrations originating from anthropogenic activities such as industrial discharges, domestic wastewater, or fertilizers carried by runoff water.

Tabel 9. Recapitulation of HPI values

Value HPI Conclusion

Sampling Point	Period 1	Period 2	Pollutant Index Value	
PWT_1	282	2,154	100	High
PWT ₂	2,154	1,225		polluted
PWT_3	1,218	3,447		

Source: (Analysis Results, 2023)

Figure 5. HPI value

Exposure to mercury metal that exceeds the quality standards in the Citarum River can cause adverse effect on human health and other living things, such as fish and plants, in the vicinity. Massing mercury metal can pass through the food chain network between living things. In humans, Hg poisoning will cause symptoms of disorders of the Central Nervous System (CNS) such as personality disorders and tremors, seizures, senile dementia, insomnia, loss of confidence, irritation, pain when chewing, inflammation of the gums, and fear. Organic Hg tends to damage the central nervous system, whereas inorganic Hg is usually more likely to damage the kidneys and cause congenital disabilities (Rahayu and Mangkoedihardjo, 2022).

4. Conclusions

Based on the research results, it can be concluded that there are dissolved heavy metals Fe, Cd, Cr, Cu, Hg, Pb, and Zn in Citarum River water. Dissolved Fe, Hg, Zn concentration is in the range of 0.277 to 1.179 mg/L, 0.011 to 0.1 mg/L, and 0.017 to 0.074 mg/L, respectively. Meanwhile, the concentration of Cd, Cr, Cu, and Pb is ≤ 0.002 mg/L, ≤ 0.01 mg/L, ≤ 0.005 mg/L, ≤ 0.03 mg/L, respectively. In comparison with Government Regulation no. 22 of 2021 class II, concentration of dissolved heavy metals that exceed the quality limits are Fe (0.3 mg/L), Hg (0.002 mg/L), and Zn (0.05 mg/L). Based on the HPI analysis results from each three sampling points in both periods, it can be concluded that the status of heavy metal pollution in the Citarum River sector of Purwakarta Regency from upstream to downstream is increasing at the three observation points. In both periods, at PWT 1 to PWT 3, heavy metals highly pollute the river water. The most significant contribution of dissolved heavy metals index in Citarum River is dominated by the Hg parameter. It indicates the immediate response should be implemented to minimize the adverse effect on human health.

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