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Assessment of heavy metals pollution in sediment of Citarum River, Indonesia

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Abstract. *Heavy metals have been reported to accumulate in the sediment of Citarum River. The measurement of total heavy metals may not be able to provide information about the exact dimension of pollution, thus the determination of different fractions assumed great importance. This study was performed to determine chemical fractions of heavy metals (Cu, Ni, Cr, Pb, and Cd) in sediment collected at 8 locations from Citarum River. The sequential extraction procedure was used to extract heavy metals in water-soluble, acid-soluble, MnO occluded, organically bound, FeO occluded, and residual fraction in sediment. Bioavailability and potential ecological risk level of heavy metals were evaluated based on bioavailability factor (BF) and risk assessment code (RAC) method. The results showed that Cu, Ni, Cr were mostly in residual form, indicate those from geological sources. Cu had low bioavailability and no risk in all sediment samples of Citarum River. Ni and Cr each were found to have risk at 2 locations. Pb and Cd were found dominantly in non-residual fractions, suggest those from anthropogenic sources. BF and RAC analysis of Pb and Cd suggest that there is a potential risk to the aquatic environment.*

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INTRODUCTION

Citarum watershed is located in western part of Java island, and its basin covers 5 960 km^2. The total length of the main river is about 315 km and flows from its water sources from Mount Wayang to the Java Sea in the north. The greater part of the basin is highly productive land. More than 56% of the area is used for agricultural purposes, while the rest of the area is covered by forest (27.4%), industry (7.9%), settlements (7.4%), fishpond (1.3%) and other miscellaneous uses (5.7%) (Bukit, 1995).

Besides giving great benefit, Citarum River has an equally great tendency for pollution. High pollution makes Citarum River has been declared as the most polluted river in the world by World Bank version, due to toxic waste that enters Citarum River (IDN Times, 2018). To illustrate how dirty the Citarum River is, at some places we cannot even see the water. Its surface is completely covered by high amount of waste, trash, and dead animals floating on it. Toxic metals have been reported to accumulate in the both water and aquatic sediment of Citarum River. In water (midle segment of the Citarum River), several heavy metals were found such as Cd 0.01 mg/kg, Cr 0.107 mg/kg, Zn 0.109 mg/kg, Hg 0.00001 mg/kg, Cu 0.024 mg/kg, Pb 0.07 mg/kg and As 0.00079 mg/kg (Septiono *et al.*, 2016). According to Muhajir *et al.* (2004) accumulation of heavy metals in sediment in the downstream of Citarum River, Jakarta bay, consists of Pb 14.7 mg/kg, Cd 0.041 mg/kg, Cu 16.22 mg/kg, Zn 90.52 mg/kg, and Ni 10.21 mg/kg.

Sediment is crucial components of the water environment which acts as pollutant sinks, therefore investigating contamination in sediment is very important. There have been many studies about heavy metals in sediment (Paundanan *et al.,* 2015; Syahminan *et al.,* 2015). In sediment, heavy metals can be present in various chemical forms. Jain (2004) noted that each form of heavy metals have different bioavailability and toxicity as the environmentalists are rightly concerned about the exact forms of metal present in the aquatic environment. There have been many studies on the fractionation and bioavailability of heavy metals in sediment. There is no recent information about heavy metal pollution level and fractionation on sediment from Citarum River, Indonesia.

The objectives of this research were: (1) to quantify and assess spatial variations of studied elements copper (Cu), chromium (Cr), nickel (Ni), lead (Pb), and cadmium (Cd) in sediment of Citarum River and (2) to determine the bioavailability and the potential ecological risk level based on the bioavailability factor (BF) and risk assessment code (RAC) method to support the management of heavy metals contamination in Citarum River. Sequential extraction was used for evaluating particular metal-sediment phase associated.

MATERIALS AND METHODS

Time and Location

Sediment samples were collected only in one period that was on May 14th to 18th, 2018 at 8 sampling locations differentiated as upstream (Cisanti and Wangisara), middle (Koyod, Cisurug, and Nanjung) and downstream (Jatiluhur, Tunggak Jati, and Walahar) area of Citarum River as shown in Figure 1. Laboratory analysis was conducted at soil chemistry laboratory, IPB University and Laboratory of soil environmental science, Kochi University, Japan.

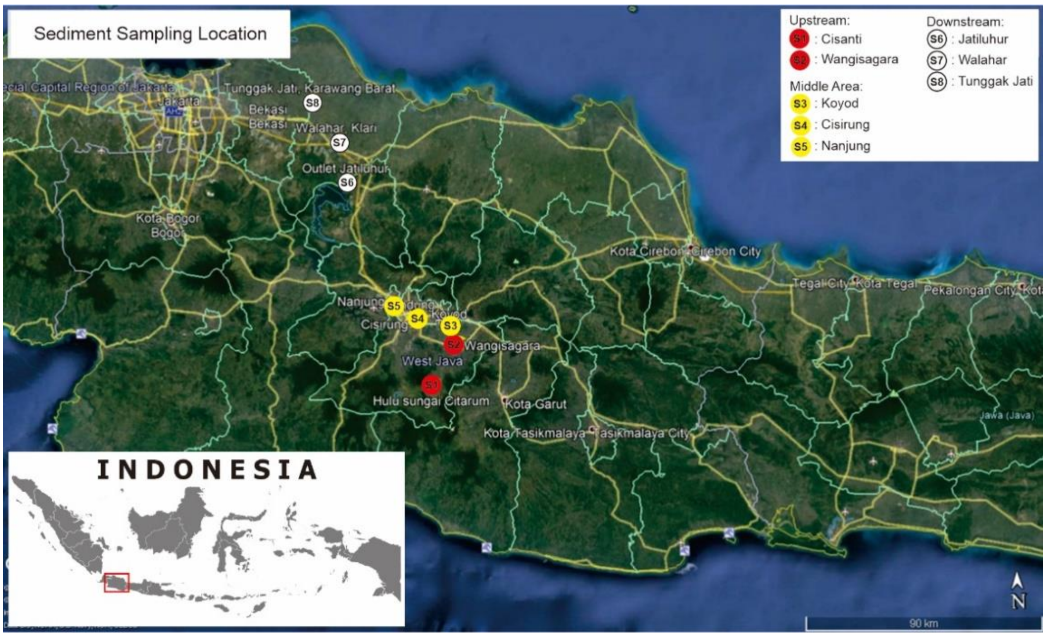

Figure 1 Distribution of sediment sampling point

Sediment Sampling

At each location, sediment sample was taken in the composite manner from three sampling points to a depth of 60 cm from the sediment surface. The composite samples were air dried, sieved to pass 2 mm and stored for analyses.

Physico-chemical Properties Analyses

The physico-chemical properties of the studied sediment analyzed namely pH, Cation Exchange Capacity (CEC), total carbon (TC), and texture. Potentiometric method using a glass electrode was used to measure the sediment pH. CEC was estimated using 1 M Ammonium acetate (NH_4OAc) solution at pH 7.0. Total carbon and texture of the studied sediment were analyzed using N-C Analyzer and pipette method, respectively.

Heavy Metal Fractionation Analysis

Chemical forms of heavy metals were estimated using sequential extraction method reported by Iwasaki *et al.* (1997) with some modification (Phuong *et al.*, 2010) as shown in Table 1. The supernatant was analyzed using AAS Shimadzu AA-6800.

Table 1 Sequential extraction procedure employed for the fractionation of heavy metals

Fractions	Reagents	Sample/Solution Ratio	Condition
Water-soluble (Ws)	Deionized water	1: 5	Shake 2 hours
Exchangeable (Exc)	1.0 mol/L CH_3COONH_4 (pH 7.0)	1: 10	Shake 2 hours
Acid soluble (Aci)	25 g/L CH_3COOH (pH 2.6)	1: 10	Shake 6 hours
Mn oxide occluded (MnO)	0.1 mol/L $NH_2OH.HCl$ (pH 2.0)	1: 10	Shake 0.5 hour
Organically bound (OM)	0.1 mol/L $Na_4P_2O_7$ (pH 10)	1: 50	Shake 24 hours
Fe oxide occluded (FeO)	0.175 mol/L $(NH_4)_2C_2O_4$, 0.1 mol/L $H_2C_2O_4$, 0.1 mol/L ascorbic acid (pH 3.1)	1: 50	Shake 4 hours, then stir occasionally in boiling water for 0.5 hour
Residual (Res)	H_2SO_4: HNO_3: $HClO_4$ (1:5:20)		

Data Analysis

Total Content of Heavy Metals

Total content of heavy metals was calculated by the total sum of the fractions. To assess the environmental pollution, total content of heavy metals was compared with freshwater sediment quality guidelines for heavy metals.

Fractionation Analysis to Identify Potential Source of Heavy Metals

Fractionation of heavy metals were demonstrated in relative percentage. The equation is shown below:

$$C\ (\%) = \frac{C_{fraction}}{C_{total}}\ x\ 100$$

Were $C_{fraction}$ and C_{total} are the concentration of heavy metals in each fraction and the total concentration of heavy metals in sediment.

Fractionation is used to estimate the sediment metal source contributed by anthropogenic activities or by geochemical sources. Anthropogenic inputs were important sources of labile fractions (non-residual) of heavy metals in sediments, while residual fraction was mainly controlled by natural source (Liang *et al*., 2018).

Bioavailability Factor (BF) and Risk Assessment Code (RAC)

Heavy metal bioavailability depends more on the proportions of the different fractions than on the total concentration (Yang *et al.*, 2017). Bioavailability factor was described by Bielicka-giełdoń *et al.* (2013). Bioavailability factor allows for the determination of possibly toxic elements in sediment. BF was calculated by following formula:

$$BF = \frac{C_{bio}}{C_{total}}$$

Where C_{bio} and C_{total} are the concentration of bioavailable heavy metals and the total concentration of heavy metals in sediment. Bioavailable heavy metals are those extracted in Ws, Exc, Aci, MnO, OM, and FeO fraction (non-residual fraction).

Risk assessment code (RAC) can be used to assess the risks and mobility of the non-stable chemical fractions of heavy metals in the sediment. In this study, risk assessment code based on the values of heavy metals in water soluble and exchangeable fraction. According to Basta *et al.* (2005) cationic heavy metals in water soluble and ion exchangeable forms are available to plants.

RESULTS AND DISCUSSION

Physico-chemical Properties of Sediment

Sediment physico-chemical properties such as pH, CEC, total carbon (TC), and textural properties is shown in Table 2. Sediment characterized by pH varied from slightly acidic to alkaline (5.73-7.45), with the highest value at Jatiluhur. The acidity (pH) distribution among the studied sediment was influenced by total carbon content.

Table 2 Physico-chemical properties of sediment

Locations	pH	CEC (cmol(+)/kg)	TC (g/kg)	Texture
Cisanti	5.7	38.5	28.0	Clay loam
Wangisagara	6.9	23.8	22.1	Sandy clay loam
Koyod	6.5	19.3	14.1	Sandy clay loam
Cisirung	6.8	27.0	15.0	Sandy clay loam
Nanjung	6.7	28.0	21.9	Sandy clay loam
Jatiluhur	7.6	13.5	5.0	Sandy clay loam
Walahar	7.3	28.6	8.1	Clay
Tunggak Jati	7.4	31.2	10.3	Clay

All sediment varied in total carbon from 5.04 g/kg to 27.97 g/kg. Cisanti, upstream area, has the highest total carbon. Wetland and riparian environments are characterized by high organic carbon concentrations in the form of animal and plant detritus. A high percentage of clay in texture analysis was usually observed at most of sampling sites. The distribution pattern of sediment physico-chemical properties was a combined result of sediment input, depositional process and hydrodynamic conditions (Astuti and Rahmanto, 2015). In this study, physico-chemical properties analysis shows the heterogeneity of the studied sediment.

Distribution of Total Heavy Metals

Total content of heavy metals in sediment of Citarum River displayed different ranges among elements as shown in Table 3. Total content of heavy metals in sediment of Citarum River were compared with freshwater sediment quality guidelines for heavy metals.

Table 3 Total content of heavy metals in the studied sediment.

Locations	Total Content (mg/kg)				
	Cu	Cr	Ni	Pb	Cd
Cisanti	47.5	8.8	10.9	16.3	0.400
Wangisagara	49.8	7.5	14.2	12.3	0.196
Koyod	44.5	12.9	15.2	11.7	0.325
Cisirung	54.5	9.1	13.0	17.0	0.418
Nanjung	59.5	13.0	13.5	22.9	0.464
Jatiluhur	11.2	18.2	14.2	26.8	0.426
Walahar	18.1	10.1	13.3	29.2	0.377
Tunggak Jati	26.6	10.9	12.7	20.9	0.583
Threshold (Environment Canada 1994)	35.7	37.3	18.0	35.0	0.596
Default guideline value (Australian Government Initiative 2018)	65	80	21	50	1.5

Copper was found in higher content in upstream and middle area, particularly in Cisanti which described unpolluted area of Citarum River. Total content of Cu in Cisanti, Wangisagara, Koyod, Cisurung, and Nanjung were above threshold effect levels for freshwater sediment. However, the total concentration of Cu in these locations did not exceed value according to default guideline value for toxicant in sediment. Cr, Ni, Pb, and Cd content in sediment of Citarum River did not exceeded the permissible limits of threshold effect levels and default guideline value for toxicant in sediment.

Enrichment of heavy metals over and above to their respective background values indicates their accumulation either by lithogenic and/or anthropogenic sources. Sediment with high total heavy metal concentration may be relatively harmless to organism if conditions are such that desorption or dissolution of metals from sediment is restricted. Conversely, sediment with lower total heavy metal concentrations may affect organism to a great extent if sediment conditions are optimal for dissolution or desorption.

Heavy Metal Fractionation

Heavy metal fractionation analysis is shown in Figure 2. The percentage of Cu, Cr, Ni, Pb, and Cd in water soluble and exchangeable fraction occurred only in trace amounts among the sediment of Citarum River. Cu, Cr, and Ni were mainly found in residual form. Singh *et al.* (2005) showed that the greater the percentage of heavy metal present in residual form, the smaller the pollution of the zone.

The concentration of Cu, Cr, and Ni in this study mainly depends on the parent material from which the soil and sediment was formed. Though total content of Cu in upstream and middle area were above the acceptable limit, it was in the most stable fraction. This study confirmed that total content of heavy metal alone is not able to evaluate the ecological risk of heavy metals.

Alloway (2012) found that possible Cu natural origin is Cu from the basalt parent material which contain Cu 90 mg/kg. According to Ratman and Gafoer (1998), geological properties in upstream area of Citarum River is characterized by andesite-basalt material. This suggests the source of high amount of Cu in upstream area of the river. Cu was also found in organic matter occluded fraction. Chakraborty *et al.* (2016)

also reported that the non-residual Cu was more associated with organic matter in the sediment of Western Continental Margin of India. Cu is known to form strong bonds with humic substances (Stevenson, 1994).

Other study carried out in Spain and China (Morillo *et al.*, 2004; Yuan *et al.*, 2004) also showed that Cr was found mainly in the residual fraction in all sediment samples. On the other hand, in non-residual fraction, Cr mainly associated with Fe oxides. Chromium in household and textile industry waste (Das *et al.*, 2015; Alam *et al.*, 2018) in Citarum River may influence Cr content in non-residual form. Figure 2 shows Ni in non-residual form increased from upstream to the downstream area, though there was no difference in the total content of Ni. This indicate that Ni content in upstream and middle area (from Cisanti to Nanjung) are natural input. Conversely, Ni in downstream area (from Jatiluhur to Tunggak Jati) are mostly from anthropogenic sources and enrichment factor.

In contrast, sediment Pb and Cd concentrations in Citarum River display a much different pattern from those of other metals. According to Figure 2 measurement of the proportions of Pb and Cd in the seven fractions showed that non-residual form was dominant. This suggests that Pb and Cd are linked with human activities in Citarum River. Cadmium contamination linked to anthropogenic activities was found in all segment of Citarum River, including Cisanti as upstream area. It is known that there have been agricultural activities in upstream area of Citarum River. Besides industrial waste, commercial inorganic P fertilizers contribute to heavy metals contamination to the environment including river sediment (Mary *et al.*, 2018). Phosphate fertilizer contains some heavy metals at various amount such as Cd (0.1-170 mg/kg) and Pb (40-2 000 mg/kg) (Setyorini, 2003). Thus, Cisanti as upstream area cannot be used as a benchmark of unpolluted area of Citarum River.

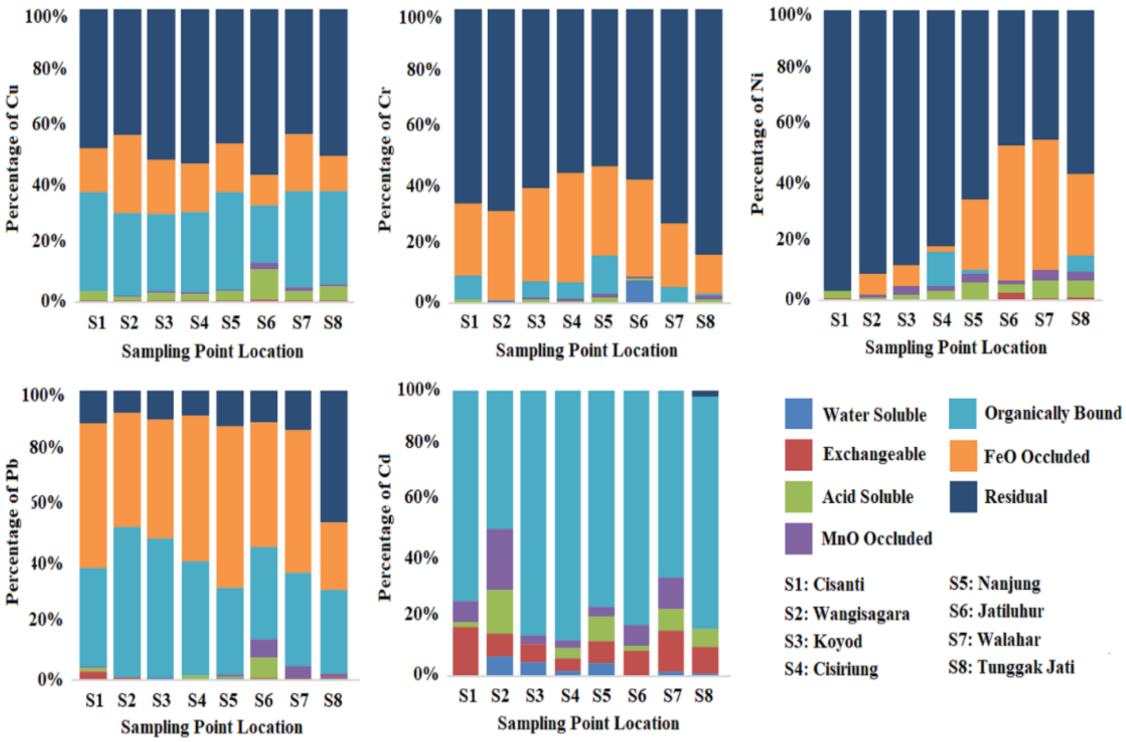

Figure 2 Percentage of Cu, Cr, Ni, Pb, and Cd in each fraction

Pb was found dominantly in FeO fraction except in Tunggak Jati. Chakraborty *et al.* (2017) showed that the positive correlation between Pb and Fe denotes that, the Fe oxy hydroxide is primary hosting phase for non-residual fraction of Pb in sediment. Heavy metals in FeO fraction are those bound to hydrous oxides of iron, Fe oxides scavenge transition and heavy metals in soil, sediment, and wastes. Organically bound fraction ranked second to the FeO fraction. Organic materials appear to reduce the availability of Pb. The Fe-Mn oxides and organic matters have scavenging effect for heavy metals, especially Pb (Duan *et al.*, 2010).

Cadmium was mostly occluded by organic matter. Soil organic matter influences Cd bioavailability. In spite of soil with higher organic matter have higher cation exchange capacity, heavy metals are probably translocated as metal-organic complexes (Egli *et al.,* 2010). Heavy metal associated with organic compounds can remain in the sediment long time until they are released by decomposition processes or oxidizing agents, leading to their complexation and bioaccumulation (Kennedy *et al.,* 1997).

Bioavailability Factor (BF)

Bioavailability factor of heavy metals in the studied sediment is described in Table 4. BF of Cu was found from 0.43 to 0.57, indicating the mobilization of approximately 50% Cu in the studied sediment. The possibly mobility Cr was recorded ranging from 0.16 to 0.47. This reflects the lower bioavailability of Cr. Bioavailability of Ni varied from one location to another. Ni in sediment from upstream and middle area had low mobility (>20%), whereas more than 40% of Ni were in bioavailable form at downstream area of Citarum River.

Table 4 Bioavailability factor (BF) of heavy metals in sediment from Citarum River

Locations	Bioavailability Factor (BF)				
	Cu	Cr	Ni	Pb	Cd
Cisanti	0.53	0.34	0.04	0.89	1.00
Wangisagara	0.57	0.32	0.09	0.93	1.00
Koyod	0.49	0.39	0.12	0.91	1.00
Cisirung	0.48	0.44	0.19	0.92	1.00
Nanjung	0.54	0.47	0.35	0.89	1.00
Jatiluhur	0.43	0.42	0.54	0.90	1.00
Walahar	0.57	0.27	0.56	0.87	1.00
Tunggak Jati	0.50	0.16	0.44	0.55	0.98

The BF of Pb showed relatively higher compare to Cu, Cr, and Ni in all locations. Al-mur (2020) stated that high levels of heavy metals bioavailability factor indicate their potential toxicity and that they could be easily released into the environment and ingested by organisms, thereby entering the food chain. The BF of Cd was the highest among other heavy metals. Generally, this result illustrated approximately 100% of Cd exist in bioavailable form and demonstrates a potential risk to aquatic life. Bioavailability of heavy metals could be mitigated, due to several processes that deactivation of heavy metals (Chen *et al.,* 2019). Heavy metals in exchangeable fraction can be released into the environment when conditions become more acidic. Heavy metals in FeO fraction also can be released into the environment when a reducing condition develops, promoting a dangerous effect to the environment (Al-mur, 2020).

Risk Assessment Code (RAC)

From ecological point of view, water soluble and exchangeable fraction may easily be leached in neutral or slightly acidic waters and they may be accumulated by living organisms (Wang *et al.,* 2017). Thus, the fractions may be used for evaluating the potential adverse effects on the organisms. Risk evaluation and the RAC classification can be seen in Table 5. The result of speciation shows that Cu are not at risk, which means the release of Cu into solution is undetectable and safe to the environment. Cr at Wangisagara and Jatiluhur shows low risk, while other locations are not at risk with regard to RAC. Ni falls in the no risk to low-risk criteria. The levels of Ni do not suggest that there is any serious hazard to the environment. Risk evaluation of Pb namely no risk to low risk, Cd are low to medium risk. These conditions indicate that Pb and Cd are likely to be harmful to the environment. Risk of heavy metals in sediment of Citarum River are Cd>Pb>Cr=Ni>Cu.

Table 5 Risk assessment code (RAC) of heavy metals in sediment from Citarum River

Locations	Risk Assessment Code (RAC)					Criteria (%) (Singh *et al.*, 2005)
	Cu	Cr	Ni	Pb	Cd	
Cisanti	0.6	0.0	0.9	3.0	17.4	No risk <1
Wangisagara	0.5	1.0	0.0	1.1	15.1	Low risk 1-10
Koyod	0.5	0.0	0.0	1.1	11.2	Medium risk 11-30
Cisirung	0.6	0.0	0.0	0.4	6.2	High risk 31-50
Nanjung	0.7	0.0	0.0	0.8	12.3	Very high risk >50
Jatiluhur	0.9	7.9	3.0	0.8	8.9	
Walahar	0.5	0.0	0.8	0.5	15.9	
Tunggak Jati	0.5	0.0	1.2	1.3	10.2	

CONCLUSION

Cu was found exceed the permissible limit in upstream and middle area of Citarum River sediment. Whereas, presence of Cr, Ni, Pb, and Cd in sediment was relatively low compared to the standard. Fractionation analysis showed that Cu, Cr, and Ni were mostly in residual form, while Pb and Cd were mainly in non-residual form. This results in high bioavailability of Pb and Cd. Cu was not at risk at all locations. Ni displayed a low risk at Jatiluhur and Tunggak Jati, Cr was at low risk in Wangisagara and Jatiluhur, while other locations were not at risk. Risk analysis of Pb and Cd in the studied sediment suggested that there was a potential risk to the environment.

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