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Heavy metals concentration in the sediment of the aquatic environment caused by the leachate discharge from a landfill

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ABSTRACT

BACKGROUND AND OBJECTIVES: Heavy metals are categorized as hazardous pollutants due to their incapability in decomposing and undergoing bioaccumulation and biomagnification. Heavy metal pollution is a global issue, particularly in emerging nations such as Indonesia. In this case, sediments contribute to pollution dispersion because they can transport, mobilize, and redistribute toxic compounds. The Cisadane river is one of 15 watersheds in Indonesia with the highest restoration priority. Therefore, it is essential to conduct study on the sediment quality of this river. This investigation aimed to evaluate the levels of cadmium, chromium, and lead in the sediments to assess the conditions of the Cisadane River.

METHODS: At eight stations (representing the midstream and downstream region), surface sediment samples were collected using a van Veen sediment grab based on the hypothesis that heavy metal pollution originated from land-based activities and migrated down river estuaries. The Thermo Scientific iCAP 7400 was utilized to assess heavy metals (cadmium, chromium, and lead) by adopting prior research methodologies and method guidelines.

FINDINGS: Except for lead, which surpassed the interim sediment quality standard, the levels of heavy metals observed in the midstream and downstream sections of the Cisadane River were found to be well below the guideline level. In this case, lead was the metal with the highest concentration in the sediments of the Cisadane River, followed by chromium and cadmium. The enrichment of heavy metals in river sediments was most likely caused by soil leaching, municipal and industrial sewage, as well as land waste disposal. After the landfill area, there were two areas with the highest concentration. Therefore, this investigation indicated the existence of landfills as point sources of heavy metals. Regarding specifics, two sites following the landfill constitute the apex of heavy metal amplification.

CONCLUSION: This analysis shows that the sediment's cadmium, chromium, and lead contents are below the standards' threshold and safe for the habitat. Cadmium, chromium and lead exceed sediment quality requirements in sample sites after landfills, assumed to be due to leachate discharge and landfill activities. This study further also reveals that landfills are point sources of heavy metals. In this case, the heavy metals are two to four times higher in one kilometer from the landfill's leachate discharge. Therefore, the Enforcement of the Indonesia Waste Law Number 18 Year 2008 would have replaced unsanitary dumping including implementation of physicochemical, biological, and combination remediation techniques, with a vastly superior waste management system.

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INTRODUCTION

The persistence and toxicity of heavy metals in waterways have sparked global alarm (Islam et al., 2015). The rapid global population growth, which has led to a rise in agricultural and industrial activity, is regarded as a significant source of metal pollution in aquatic environment, which may offer deadly health dangers to humans and wildlife (Riani et al., 2014, 2018; Xie et al., 2022). The improper management of industrial and domestic waste contributes to the environmental problem of heavy metals in emerging nations, including Indonesia. Heavy metals are hazardous pollutants because such pollutant cannot be decomposed, accumulates in the body (bioaccumulation), and gradually moves up the food chain to higher concentrations (biomagnification) (Ali and Khan, 2019; Saher and Siddiqui, 2019; Vandecasteele et al., 2004). Environmental pollution occurs when a contaminant, including heavy metals, enters the environment and surpasses the tolerance level to be disruptive to the environment and dangerous to living beings (Mohammed et al., 2011; Rosado et al., 2016). Among the various types of heavy metals, some are toxic, including cadmium (Cd), chromium (Cr) and lead (Pb), which are also classified as harmful heavy metals by the Agency for Toxic Substances and Disease Registry (ATSDR, 2012). People often use Cd, Cr, and Pb as raw materials for making furniture, as coating materials for toolkits, as mixtures of metallic ingredients and antifouling colorants, as well as in agriculture (Ayangbenro and Babalola, 2017; Burton et al., 2005a; Burton et al., 2005b; Li and Ren, 2011; Liu et al., 2016). By measuring the heavy metal concentrations directly and estimating what has accumulated in the environment, specifically in sediments, therefore, it is possible to determine the degree of heavy metal contamination (Kaewtubtim et al., 2016). Sediments have a role in dispersing pollution because they can carry, mobilize, and redistribute harmful substances (Miranda et al., 2021). Although heavy metals can be deposited in sediments after being absorbed by suspended materials and amassing to high concentrations, this retention is not permanent, and the metals may be released if the surrounding environment changes (Custodio et al., 2020). United States Environmental Protection Agency (USEPA, 2005) stated heavy metals' sorption is influenced by several conditions, including pH, alkalinity, and fractions of clay-silicate and exchangeable carbonate. Frequently, heavy

metal contamination in the sediments is evaluated by calculating their total metal content; where differences in metal levels along the watershed may indicate distinct metal input sources (Pagnanelli et al., 2004). Measuring the overall concentration of metals in sediments is beneficial for detecting anomalies in the watercourse caused by several conceivable events, exempli gratia (e.g.) leaching from or to groundwater, erosion, sedimentation event; however, it does not reveal the chemical pattern of metals in sediments (Pagnanelli et al., 2004). Understanding heavy metals partitioning among different geochemical phases is crucial to evaluate the potential of bioavailable materials and related ecotoxicity hazards (Dixit et al., 2015; Kalender and Çiçek Uçar, 2013). Previous research has revealed that non-point source pollution (such as scattered residential and industrial activities) and point source pollution in the study area are responsible for various geographical and temporal distributions of hazardous pollutants in sediments (such as landfills in the Cisadane watershed). A landfill is a place where hazardous waste, such as exhaust gas emissions and liquid waste through leachate, is deposited (Roudi et al., 2020). Waste and pollution from landfills can infiltrate the aquatic environment because landfills are typically situated on the banks of river regions. Due to a lack of technological advances and infrastructure, waste disposal and treatment are especially problematic in developing nations (Essien et al., 2022). In Indonesia, the waste management system is poor, and there are almost no landfills with proper liner bottoms. Toxic leachates could contaminate soil and groundwater through non-sanitary landfills, which are the norm in Indonesia (Meidiana and Gamse, 2010; Munawar et al., 2018). Consequently, detecting and characterizing the ecotoxicological profile of heavy metals in the environment affected by landfill leachate and the accompanying health hazards posed by municipal dumpsites is of utmost importance. Rivers are essential to life, serving as potable water sources, transportation routes, recreational spaces, and agricultural irrigation water. Introducing pollutants such as heavy metals into rivers will disturb the river's ecosystem. Indonesia has about 5,500 major rivers and over 65,000 tributaries; however, over 50% of the rivers are contaminated (Statistic Indonesia, 2021). The Cisadane River is one of 15 Indonesian watersheds with the highest restoration priority. Same as other rivers, the Cisadane river provides raw water for drinking water,

food production, sanitation, purification, and coastal stabilization, among other essential uses. Research on heavy metals such as Cd, Cr and Pb in the Cisadane River is crucial, given that the river serves as a source of raw water for the potable water delivery systems in the midstream (South Tangerang, Tangerang) and downstream (Tangerang, and Tangerang Regency) areas. In addition to being surrounded by various anthropogenic activities (settlement, agriculture, and industry), the Cisadane River is a home to three final disposal sites for the inhabitants of this region. Even as recently as May 2020, one of the landfill walls collapsed, resulting in the release of approximately 100 tons of waste into the Cisadane River ([The Jakarta Post, 2020](#)). It resulted in an immediate release of waste material, including heavy metals, which ultimately deposited in the sediment of the Cisadane River. Population growth and human activity along the river's flow made it possible for metal pollutants to get into the river. These pollutants then followed the river's flow downstream and into the sea. Therefore, this study was conducted based on the hypothesis that the increase in toxic metals in the Cisadane River was linked to anthropogenic activities in the area, namely non-point sources (scattered household, commercial, and industrial activities) and point sources (two landfills on the edge of Cisadane River). The hypothesis provided was that there was a significant difference between the two types of sources, which made point sources more likely to influence the accumulation of toxic heavy metals in river sediments. The above background shows that enriching the comprehension of toxic heavy metals (Cd, Cr, and Pb) contamination in urban river sediment to build realistic ways and strategies for mitigating the conflicting effects of heavy metals contamination on the aquatic ecosystem is necessary. In this case, the current study aimed to measure Cd, Cr, and Pb levels in the riverine sediments and to evaluate the river conditions based on specific heavy metal concentrations. Furthermore, this study was conducted on the Cisadane River in Indonesia during the dry season (April-May) of 2022.

MATERIALS AND METHODS

Study area

The Cisadane River is 138 miles long and covers an area of 154,652 hectare (ha). Due to its high pace of land change, the Cisadane watershed is among Indonesia's 15 priority watersheds. Between

1995 and 2012, the land cover changed by 63.05%. The most changes happened in bushland, forests, and rice fields by 29.65%, 27.87%, and 13.44%, respectively. The distance between the river and the city's downtown area is primarily responsible for this disparity. The city downtown offers convenient and essential services, enabling the expansion of settlements and the fulfillment of other community demands. The Cisadane River receives water from the Gede-Pangrango and Halimun-Salak mountains upstream and empties into the Teluk Naga coast in the Java Sea. Furthermore, the Cisadane River passes through 44 sub-districts in five cities. These cities are Bogor Regency and Bogor City in the upstream area, South Tangerang and Tangerang City in the midstream, and Tangerang Regency in the downstream area. In this case, more than 15 million people reside in the Cisadane watershed, contributing over 8,400 tons of solid waste daily. In this watershed, however, three landfills are located directly on the Cisadane River: the Galuga Landfill in Bogor Regency, the Cipeucang Landfill in South Tangerang, and the Rawa Kucing Landfill in Tangerang City. The leachate water from the three landfills drains to the Cisadane River via a canal. In addition, the Cisadane River supplies tap water sources for the midstream and downstream regions.

Field sampling

At eight stations sampling (ST), a van Veen sediment grab was used to acquire samples of surface sediment under the assumption that heavy metal pollution originated from land-based activities and migrated down river estuaries into the ocean, such as industry, mining, and agriculture ([Koesmawati et al., 2018](#); [Lestari et al., 2018](#); [Riani et al., 2014](#)). The first four stations (ST01-4) represent the midstream region, whereas the subsequent four stations (ST05-ST08) represent the downstream region ([Fig. 1](#)). The two stations were situated 1 kilometer (km) before and after the two landfill sites (Cipeucang and Rawa Kucing Landfill) and the leachate disposal sites from the two landfills. The samples were collected from three spots on the river's right, middle, and left sides. Using a clean plastic spoon, the top layer of sediment; ~10 centimeter (cm) from each sediment grab sample was collected, and the sediments were mixed to generate a composite sample. In stainless steel canisters, triplicate sediment samples were

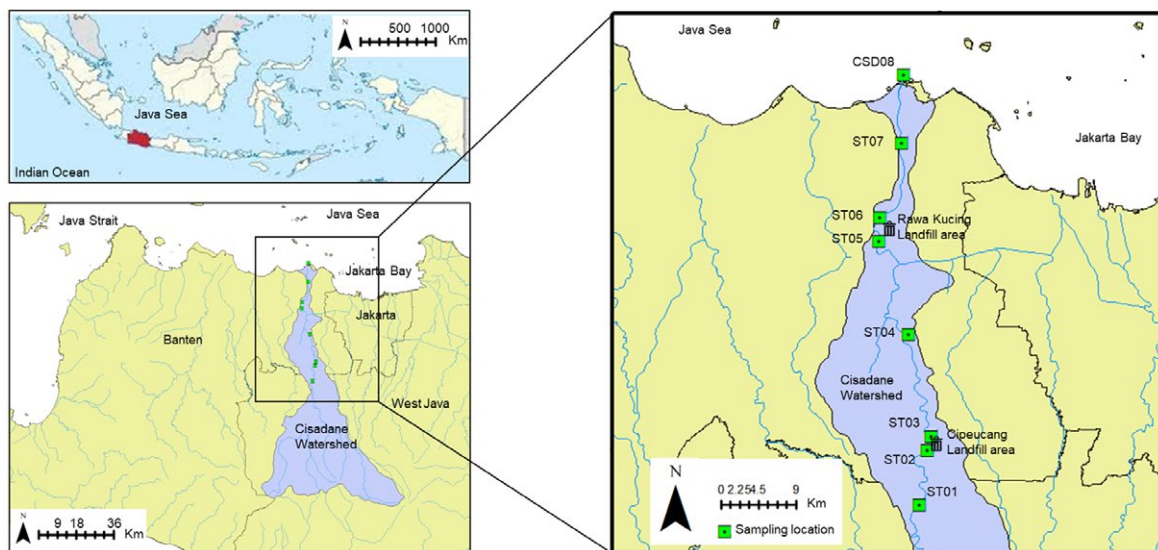


Fig. 1: Geographic location of the study area along with sampling locations at Cisadane River, Indonesia

homogenized. The samples were then scooped using a rinsed plastic scoop and transferred to a 500 milliliter (mL) cleaned and rinsed glass canister. Furthermore, all samples were preserved at 4 ± 2 degree Celsius ($^{\circ}\text{C}$) in sealed plastic containers to prevent contamination.

Laboratory analysis and quality assurance/quality control

The Thermo Scientific iCAP 7400 Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) was used to analyze heavy metals by accommodating the USEPA (2007) method 3051a from prior research (Cordova *et al.*, 2017; da Silva *et al.*, 2013; Harmesa and Cordova, 2020; Puspitasari *et al.*, 2020; Puspitasari and Lestari, 2018; Riani *et al.*, 2018). Briefly, the river surface sediment samples were dried in an oven at 105°C for twenty-four hours. The objective of the drying process was to eliminate the moisture. A mortar was further used to grind the dried samples. The grinded samples (range 0.49-0.51 gram) were then mixed with 9 mL of nitric acid (HNO_3 or Hydrogen nitrate) and 3 mL of Hydrochloric acid (HCl or Hydrogen chloride). Complex organometallic compounds were converted to inorganic compounds through the treatment of HNO_3 and HCl. The sample was then being placed in a the CEM MARS5 Xpress microwave digestive reactor for 15 minutes at 185°C and kept for 30 minutes. The samples were filtered using Whatman filter paper No. 41 and diluted

to 25 mL with DDDW (Double Distilled Deionized Water). The sample was subsequently introduced to the ICP-OES to determine the concentrations of selected heavy metals. The National Research Council of Canada's Certified Reference Material (CRM) PACS-3 for sediments was utilized to verify that the instruments and procedures were reliable and controlled. The sample analysis of CRM was still within the standard range, indicating that the method and ICP-OES utilized in this study were valid and regulated.

Data analysis

The statistical test was conducted using PAST Software Version 4.03, which included examining univariate statistics and assessing statistical significance differences between Cd, Cr, and Pb for each sampling region via the Kruskal-Wallis test for equal medians and Mann-Whitney pairwise post hoc test. Due to the absence of sediment quality guidelines in Indonesia, a descriptive analysis of the mean and standard deviation of Cd, Cr, and Pb concentrations in riverine sediments was performed and compared to sediment quality guidelines from the Australia New Zealand Environment and Conservation Council (ANZECC) and Agriculture and Resource Management Council of Australia and New Zealand (ARMCANZ), as well as Canadian Council of Ministers of the Environment (CCME). Using the

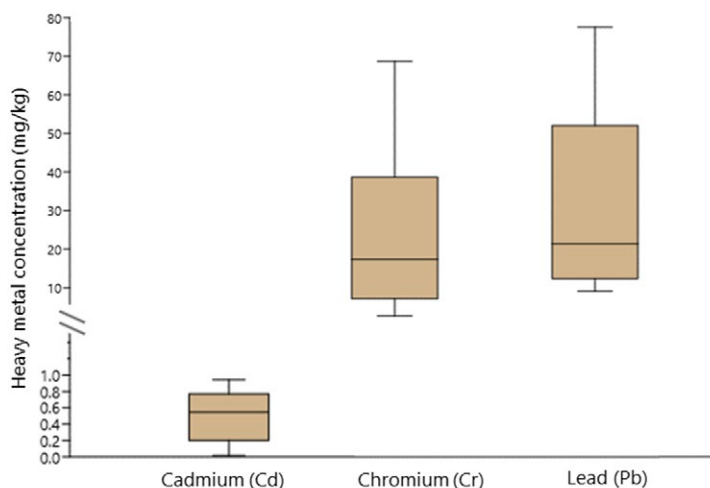


Fig. 2: The concentrations of selected heavy metals in the sediments of Cisadane River.

same software, a Pearson correlation test was done to comprehend the metal's interaction with the sediment. With a higher coefficient value, it was considered that the metals originate from the same place, were interdependent and had an identical flow behavior (Harmesa and Cordova, 2020).

RESULTS AND DISCUSSION

Selected heavy metal concentrations in the Cisadane River sediments

Fig. 2 depicts the distribution of Cd, Cr, and Pb concentrations in riverine sediments. With an average value of 30.3053 ± 23.4339 milligram per kilogram dry weight (mg/kg), Pb had the highest concentration in the Riverine Sediment of the Cisadane River, followed by Cr (23.4672 ± 20.1228 mg/kg). Compared to Pb and Cr, Cd was found to have the lowest concentration (0.5140 ± 0.2983 mg/kg). Furthermore, the Kruskal-Wallis and Mann-Whitney tests revealed a statistically significant difference ($p < 0.05$) between Cd - Cr and Cd - Pb. Table 1 further illustrates the spatial distribution of the mean and standard deviation of the Cd, Cr, and Pb concentrations at each location, as well as their comparison to sediment quality standards. On the basis of sediment quality guidelines from ANZECC and ARMCANZ (2000) and CCME (2001), selected heavy metals observed (Table 1) in midstream and downstream Cisadane River were moderately lower than the guideline level, with the exception of Pb, which exceeded the ISQG (Interim Sediment Quality

Guideline) from CCME (2001). The selected heavy metal concentrations in this study were below or comparable to those in other well-known major rivers (Gaillardet *et al.*, 2013; Nasrabadi *et al.*, 2018). Selected heavy metals in the Cisadane River sediments were relatively lower than those in Yellow River (W. Li *et al.*, 2022), Han River (X. Li *et al.*, 2022), Nanfei River (Fang *et al.*, 2022) in China, and the Brahmaputra River in India (Saikia *et al.*, 2022); on the same magnitude compared to the Teesta River, India (Chettri *et al.*, 2022) and the Oder river and Vistula river in Poland (Jaskuła and Sojka, 2022). However, heavy metals in the Cisadane River sediments were higher than the Kafue River and Zambezi River in Zambia (Nakayama *et al.*, 2010). Compared to dissolved concentrations discovered in urban runoff, the results of this study showed a stronger link (Pinedo-Gonzalez *et al.*, 2017). Greater urban land use resulted in higher quantities of dissolved metals in watersheds compared to those found in natural areas (Gardner and Carey, 2004; Pinedo-Gonzalez *et al.*, 2017; Yoon and Stein, 2008). In general, heavy metal pollution levels in the Cisadane River sediments were still below the threshold at which they could cause harm to the aquatic organism. Albeit the selected heavy metals concentration in this study was moderately modest compared to the guidelines (Table 1), it must nevertheless be taken into account because Cd and Pb surpass their natural values in the upper continental crust. Wedepohl (1994) specifically

Table 1: The concentrations of selected heavy metals in each sampling station in the Cisadane River

| Location | Station code number | Concentration (mg/kg) | | |
|-------------------------------|---------------------|-----------------------|------------------|-------------------|
| | | Cd | Cr | Pb |
| Midstream (ST01-ST04) | ST01 | 0.0628 ± 0.0614 | 5.3425 ± 0.7623 | 9.8746 ± 1.4921 |
| | ST02 | 0.1825 ± 0.0139 | 4.2874 ± 2.1266 | 9.1596 ± 0.3991 |
| | ST03 | 0.8000 ± 0.0295 | 52.1811 ± 8.2727 | 63.6332 ± 6.3275 |
| | ST04 | 0.6854 ± 0.0573 | 15.8714 ± 6.2011 | 26.4032 ± 8.9145 |
| Downstream (ST05-ST08) | ST05 | 0.3214 ± 0.1143 | 13.0176 ± 2.6695 | 20.5500 ± 10.2734 |
| | ST06 | 0.9278 ± 0.0126 | 57.9242 ± 9.3170 | 72.0347 ± 7.0349 |
| | ST07 | 0.6196 ± 0.1644 | 15.7160 ± 6.0428 | 20.2391 ± 1.3909 |
| | ST08 | 0.5127 ± 0.0941 | 23.3976 ± 3.3947 | 20.5476 ± 0.9902 |
| ANZECC and ARMCANZ Guidelines | Low | 1.5 | 80 | 50 |
| | High | 10 | 370 | 220 |
| CCME Guidelines | ISQG | 0.7 | 52.3 | 30.2 |
| | PEL | 4.2 | 160 | 112 |

ISQG: interim sediment quality guidelines; PEL: probable effect levels

stated that the natural concentrations of Cd, Cr, and Pb were 0.102 mg/kg, 35 mg/kg, and 17 mg/kg, respectively.

The largest concentration of heavy metals in the Cisadane River sediments was Pb, followed by Cr and Cd. The enrichment of heavy metals in riverine sediments is caused by natural sources, including volcanic eruption and weathering from soil or rocks, which causes leaching (Fang et al., 2016). In addition to natural sources, anthropogenic activities waste, such as unmanaged waste, is a source of heavy metal pollution (Fang et al., 2016). Pb is emitted through urban runoff, atmospheric deposition, combustion engine automobile and industrial emissions, and craft maintenance activities (Burton et al., 2005a; Hossain et al., 2019; Liu et al., 2016; Sakawi et al., 2013). Meanwhile, Cd pollution is linked to farming activities such as using many phosphate fertilizers with Cd impurities (Liu et al., 2016). Moreover, Cd usually comes from activities that make farming more productive, such as using a lot of fertilizers and pesticides and mining (Ayangbenro and Babalola, 2017; Tang et al., 2010). Furthermore, Cr is often found in antifouling paints used in the screen-printing and textile industries (Costa-Böddeker et al., 2017; Duodu et al., 2017).

The linear correlation matrix of Pearson's correlation coefficients between Cd, Cr, and Pb

in Cisadane river sediments is shown in Table 2. According to the Pearson correlation value, the statistical output between Cd - Cr and between Cr - Pb suggested a moderately positive relationship (r between 0.6 and 0.8), although the correlation between Cr - Pb indicated a fairly strong positive relationship (r between 0.8-0.99). In this case, the three correlations had a 99% confidence interval (Table 2). Based on the discovered Pearson's correlation coefficient (Table 2), Cd - Cr and Cr - Pb were shown to have a moderately positive connection. Moreover, the correlation between Cr and Pb was significantly positive. This linkage demonstrated that heavy metals were interdependent, exhibited the same transit behavior, and most likely originated from the same origin, whether natural or manmade (Harmesa et al., 2020; Liu et al., 2016; Suresh et al., 2012).

Spatial distribution of heavy metals in Cisadane River

Table 1 demonstrates that all sampling locations detected heavy metals (Cd, Cr, and Pb), showing that the trend of heavy metals pollution is increasing to the Cisadane River's downstream section. In this investigation, the lowest concentrations of heavy metals were identified at ST01 and ST02, the first and second midstream sites. Nonpoint and point sources were the sources of heavy metals in the sediments of the Cisadane River. Dispersed throughout the Cisadane

Table 2: Pearson's correlation coefficient (r) for selected heavy metals concentration in Cisadane River sediments

| Heavy metals | Cd | Cr | Pb |
|--------------|---------|---------|----|
| Cd | 1 | | |
| Cr | 0.7916* | 1 | |
| Pb | 0.7877* | 0.9254* | 1 |

watershed were nonpoint sources in the form of effluent from community activities. The household and commercial areas were the densest in the mid-midstream and downstream regions. Similar to households, the majority of industries in the Cisadane watershed were relatively small (those located in the upstream and early midstream), with 143 medium and heavy industries dispersed throughout the mid-midstream and downstream regions (Kementerian Perindustrian, 2020). The landfills on the river's banks were considered to be the point sources of heavy metals in the sediments of the Cisadane River. After landfill leachate disposal, the mean and standard deviation of the examined heavy metals were higher than the data without area (ST03 and ST06). ST03 and ST06, both of which are located after the landfill area, contained the highest concentration. Detail-wise, ST03 and ST06 represented the pinnacle of heavy metal amplification (Table 1). Moreover, at these two stations, Cd, Cr, and Pb concentrations exceeded the ISQG guidelines from CCME (2001). Furthermore, Pb in ST03 and ST06 surpassed the lower limit specified by ANZECC and ARMCANZ (2000). Heavy metals data's mean and standard deviation without a sample point before a landfill were two to four times lower. In addition, a statistically significant difference occurred between the regions before and after landfilling for all examined heavy metals ($p < 0.01$). Depending on the type of heavy metal, the increase occurred between 2.89 and 12.17 times. At ST03 and ST06, Cd levels were 4.38 and 2.89 times higher, Cr levels were 12.17 and 4.45 times higher, and Pb levels were 6.95 and 3.50 times higher, respectively. There was a presumption that there was a correlation between the landfill location and landfill leachate and the concentration of heavy metals in the sediments of the Cisadane River. The limitation of this study was that it did not investigate landfill leachate in two dump regions located along the Cisadane River. If a comparative study is undertaken, the heavy metals level in landfill leachate must be analyzed in greater detail. The

results of this investigation indicated the existence of landfills as probable point origins of harmful heavy metals (Deng *et al.*, 2018; Houessionon *et al.*, 2021; Hussein *et al.*, 2021; Robinson, 2017). Leachate treatment facilities in Indonesian landfills typically consist of ponds for collection and treatment. Some of the leachate in the pond is disposed of as a result of microbial fermentation and gravity effects (Xu *et al.*, 2018); however, because the leachate treatment is not comparable to a sanitary landfill, the leachate produced and discharged into the watershed has a complex composition and contains toxic heavy metals (Hou *et al.*, 2019; Singh *et al.*, 2015). The leachate carried into the watershed will negatively impact the river's environment. In turn, this will increase the expense of environmental management and may adversely affect human health (Hussein *et al.*, 2019; Ishak *et al.*, 2016). Along the Cisadane River, three unsanitary landfills have direct leachate discharge channels into the river (Nurhasanah *et al.*, 2021; Sulistyowati *et al.*, 2022). After one kilometer from the landfill leachate outlet, the concentration of heavy metals at the sampling station was two to four times greater. In landfill leachate, critical heavy metals are frequently abundant, physiologically complex, and bioaccumulative, rendering them highly accessible to trophic food systems (Atta *et al.*, 2015; Kaschl *et al.*, 2002; Sánchez-Chardi and Nadal, 2007). A transition from an unsanitary (open) dumping system to a properly sanitary landfill system, is essential for improving this situation since it can raise the heavy metals removal rate (Deng *et al.*, 2018; Jaradat *et al.*, 2021; Robinson, 2017). Some levels of heavy metals in natural soil were found to exceed predetermined background values. Despite the fact that the metal content of these soils is considerable, it is significantly lower than that of waste/leachate-affected soils (Hussein *et al.*, 2021). Notably, heavy metals are also natural trace elements that are infrequently harmful (Wuana and Okieimen, 2011). Under typical conditions, it is unlikely that metals are dissolved,

Heavy metals in the aquatic environment

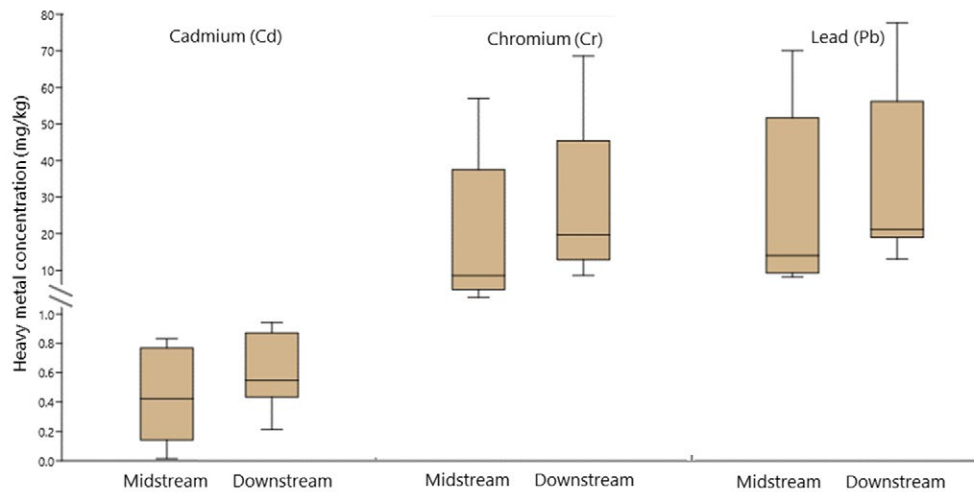


Fig. 3: Comparison of selected heavy metals concentration in midstream and downstream of Cisadane River

leached, and transportable in the environment. However, it can be induced by successive reductive circumstances and anaerobic bio-decompositions (Hussein *et al.*, 2021; Ishchenko, 2019; Li *et al.*, 2009; Thongyuan *et al.*, 2021). These circumstances may result in the leaching of toxic metals deposited in the riverine sediment (Demirbilek *et al.*, 2013). Due to the decrease in oxygen content, methanogenic conditions have led to the dissolution of a reductive metal in the leachate (Demirbilek *et al.*, 2013). Additionally, metal leaching occurs in soil affected by leachate with high organic content (DeLemos *et al.*, 2006; Ford *et al.*, 2011).

Fig. 3 depicts the calculated average concentrations and fluctuations of Cd, Cr, Pb in the midstream and downstream sediments of the Cisadane River. Along the Cisadane Yellow River, all investigated heavy metals in sediments demonstrated an upward trend, with greater average concentrations in downstream than in the midstream. Cr had the most significant increase in the mean concentration of selected heavy metals from midstream to downstream, followed by Cd, while Pb had the smallest increase (Fig. 3). The average concentration of Cr in the midstream (19.4206 ± 20.8090 mg/kg) increased by 41.67% in the downstream (27.5138 ± 19.4402 mg/kg). The mean Cd concentration in the midstream (0.4327 ± 0.3318 mg/kg) increased by 37.60% in the downstream (0.5953 ± 0.2481 mg/kg). The average Pb content in the midstream increased by 22.28% in the

downstream (27.2677 ± 23.5577 mg/kg to 33.3429 ± 23.9400 mg/kg). Our findings are consistent with the previous studies on heavy metal contamination of soils in the middle and lower reaches of China's Xiangjiang River (Wang *et al.*, 2008). Heavy metals originating from improperly managed pollution in the Cisadane watershed reached the aquatic ecosystem and were further carried by the water, and were deposited in river sediments. Moreover, the water carried pollutants from the land to the aquatic environment. Although many heavy metals might be predominantly associated with organic material particles or suspended sediment, heavy metals can be shifted into a more bioavailable dissolved form (Bergamaschi *et al.*, 2012; Roussiez *et al.*, 2011). Various hydrological regimes can impact heavy metal mobility. For instance, during a precipitation event following a prolonged period of drought, overland water runoff is frequently the primary transport mechanism for anthropogenic toxic metals (Wijesiri *et al.*, 2018; Yong and Chen, 2002). In this study, landfills were also considered point sources of toxic metals in addition to non-point sources of heavy metals throughout the Cisadane river. Widespread usage of non-engineered and unmanaged landfills without suitable bottom liners, leachate collection, or treatment systems prevented better management of solid waste from municipalities (Ishchenko, 2019). This has resulted in the production of leachates from landfills that include substantial amounts of

organic and inorganic pollutants and are particularly dangerous to the environment (Naveen *et al.*, 2017; Öman and Junestedt, 2008). In this case, 84% of waste management in Indonesia was accomplished through waste disposal sites with open dumping landfill system (Munawar *et al.*, 2018). However, the proportion of landfills with improved systems has not yet met the requirements for a sanitary landfill system (Meidiana and Gamse, 2010), meaning that only a portion of the fundamental conditions for controlled landfills was achieved. This leads in the release of leachates containing harmful substances including toxic heavy metals into the aquatic environment (Hussein *et al.*, 2021). In some landfills, leachate consists of fluids that have reached the open landfill from a variety of external sources, such as wastewater, groundwater, soil erosion, and precipitation generated from the breakdown of organic waste (Ghosh *et al.*, 2017). In certain instances, groundwater flow following flood water subsidence can contribute significantly to metal loading in downstream waterways (Santos *et al.*, 2011). Heavy metal emissions could be deposited within riverine to estuarine sediments and transferred to the marine system (Fernández-Cadena *et al.*, 2014; González-Ortegón *et al.*, 2019). The Indonesian government might have helped alleviate this problem by enforcing the Waste Law Number 18 Year 2008, which mandates the replacement of all open dumping with more regulated landfills or sanitary landfills. The elimination of heavy metals from non-point sources, such as leachate landfills, can be accomplished through various methods, including physicochemical (coagulation/flocculation treatment, membrane application, and adsorption treatment), biological (phytoremediation, bioremediation, and arrangement of aerobic and anaerobic bioreactors), and combination (physicochemical and biological) techniques. The elimination of heavy metals in aquatic environments directly results from the transition to sanitary landfills (Mojiri *et al.*, 2011). Changes should also be made at the regional level, considering available infrastructure, human capacity, and funding.

CONCLUSION

The purpose of this study is to characterize heavy metals in Cisadane River sediments. This investigation leads us to conclude that the Cd, Cr, and Pb concentrations in the sediment are still below

the threshold established by the standards and are, therefore, safe for the habitat inside. In this study, concentrations of Cd, Cr, and Pb were found to be lower than or comparable to those in other well-known large rivers. Although the concentrations of heavy metals in this study are relatively modest in comparison to the guidelines, they must still be taken into account because Cd and Pb are higher than they should be in the upper continental crust. However, the concentration of Cd, Cr, and Pb exceeds the sediment quality guidelines in the sampling sites after landfills, which is believed to be the result of leachate discharge and landfill activities. Therefore, it requires special consideration including the implementation of physicochemical, biological, and combination remediation techniques. This study discovers that heavy metals are interdependent, have the same transport patterns, and most likely originate from the same sources. In the Riverine Sediment of the Cisadane River, Pb has the highest concentration, followed by Cr, while Cd that has the lowest concentration. Sources of heavy metals in the Cisadane River sediments comprise nonpoint and point sources. The nonpoint sources in the form of wastewater from community activities are dispersed throughout the Cisadane watershed. In comparison, two riverbank landfills are recognized as the point sources of heavy metals in the sediments of the Cisadane River. Two landfills along the Cisadane River discharge their leachate directly into the waterway. The concentration of heavy metals at the test location is two to four times higher one kilometer from the landfill's leachate discharge. In this case, investigation indicates that heavy metal pollution in the Cisadane River's downstream segment is escalating. Therefore, through the enforcement of the Indonesian Waste Law Number 18 Year 2008 which would have replaced all open dumping with better-controlled landfills or sanitary landfills, would have helped the Indonesian government address this problem. Furthermore, further research is required to determine the levels of harmful heavy metals in leachate landfills, drain sediment in leachate ponds, and groundwater, which can also be contaminated by suboptimal management of leachate landfills.

AUTHOR CONTRIBUTIONS

L. Sulistyowati, the corresponding author, has contributed in interpreted the results, and preparing

the manuscript. Nurhasanah performed data analysis. E. Riani designed the field sampling contributed to the data analysis and interpretation of the results. M.R. Cordova contributed to supervision, study design, laboratory analysis, data analysis and writing with the assistance of L. Sulistyowati, Nurhasanah and E. Riani. M.R. Cordova is the main contributor to this manuscript.

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CONFLICT OF INTEREST

The author declares that there is no conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy have been completely observed by the authors.

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ABBREVIATIONS

| | |
|------------------------|--|
| <i>ANZECC</i> | Australia New Zealand Environment and Conservation Council |
| <i>ARMCANZ</i> | Agriculture and Resource Management Council of Australia and New Zealand |
| <i>ATSDR</i> | Agency for Toxic Substances and Disease Registry |
| °C | degree Celsius |
| <i>CCME</i> | Canadian Council of Ministers of the Environment |
| <i>Cd</i> | Cadmium |
| <i>cm</i> | Centimeter |
| <i>Cr</i> | Chrome |
| <i>CRM</i> | Certified reference material |
| <i>DDDW</i> | Double distilled deionized water |
| <i>e.g.</i> | Exempli gratia (for example) |
| <i>ha</i> | hectare |
| <i>HCl</i> | Hydrogen chloride (Hydrochloric acid) |
| <i>HNO₃</i> | Hydrogen nitrate (nitric acid) |
| <i>ICP-OES</i> | Inductively coupled plasma – optical emission spectrometry |
| <i>ISQG</i> | Interim sediment quality guidelines |
| <i>kg</i> | Kilogram |
| <i>km</i> | Kilometer |
| <i>mg/kg</i> | Milligram per kilogram dry weight |
| <i>mL</i> | Milliliter |
| <i>PEL</i> | Probable effect levels |
| <i>Pb</i> | Lead |
| <i>r</i> | Pearson's correlation coefficient |
| <i>ST</i> | Station sampling |
| <i>USEPA</i> | United States Environmental Protection Agency |

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