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Assessment of Heavy Metal Contamination in Soil around Piyungan Landfill, Yogyakarta, Indonesia

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ABSTRACT. One of the negative impacts of the landfill as solid waste disposal is soil contamination by heavy metals. This study assessed heavy metals impact, especially Pb, Cu, Zn, and Cd, in the soil in Piyungan landfill, Bantul, Yogyakarta, Indonesia. The assessment was conducted by analyzing 15 soil samples from 25 cm depth in the study area, divided into three zones. The study results showed that generally, the highest content of metals was found in zone II, which is located near or directly situated in a landfill site. The pollution index (*PI*) calculated showed in order Cd > Cu > Pb > Zn. The result also indicates that Cd has the highest pollution index and risk compared to Pb, Cu, and Zn. The eco-risk index (*RI*) calculation showed that the value was 29 to 70 demonstrating a low class. The result also indicates that the accumulation of heavy metals investigated in this study was normal and that the ecological risk was relatively low.

Keywords: Assessment · Soil · Contamination · Metals · Landfill.

1 INTRODUCTION

One of the solid waste disposal methods for municipal solid waste is the landfill method (Tchobanoglous *et al.*, 1993). This method is preferably used to reduce solid waste in the city and municipality and usually has a negative impact on the environment (Vaverková, 2019; Gworek *et al.*, 2016). The presence of the Piyungan landfill is indicated to impact the surrounding environment, including pollution to the soil due to the presence of hazardous metal content in waste such as Pb, Cu, Zn, and Cd. The waste disposed to the landfill generates leachate due to waste decomposition (Fagbenro, 2016).

The leachate's metal ion will distribute to the surrounding through lateral and vertical movement to the soil profile. This phenomenon occurs in the landfill with no liner material or leakage in the landfill liner during the landfill operation (Agamuthu, 2001). Several studies for contamination of heavy metals in the soil in the landfill site (Bahaa-Eldin *et al.*, 2008; Azeez *et al.*, 2011; Liu *et al.*, 2013).

Piyungan landfill in Bantul district, Yogyakarta Province, is operated by a controlled landfill method to develop the open dumping method (Ariyani *et al.*, 2019). However, the Piyungan landfill in the study area will affect the environment due to heavy metals in the waste. Several studies have been done in the Piyungan landfill and indicate heavy metals in groundwater (Harjito *et al.*, 2018; Sartohadi *et al.*, 2017; Parhusip *et al.*, 2017; Phonhalath, 2012; Putra, 2001).

This study aimed to assess the impact of soil contamination by heavy metals, especially Pb, Cu, Zn, and Cd, in the Piyungan landfill. The location of the study area is shown in Figure 1.

1.1 Regional geology

The research area was regionally included in the Semilir Formation and the Young Merapi Volcanic Deposit. The Semilir Formation consisted of tuff breccias, pumice breccias, dacite tuff, andesite tuff, and tuff claystone. In contrast, the Young Merapi Volcanic Sediment was

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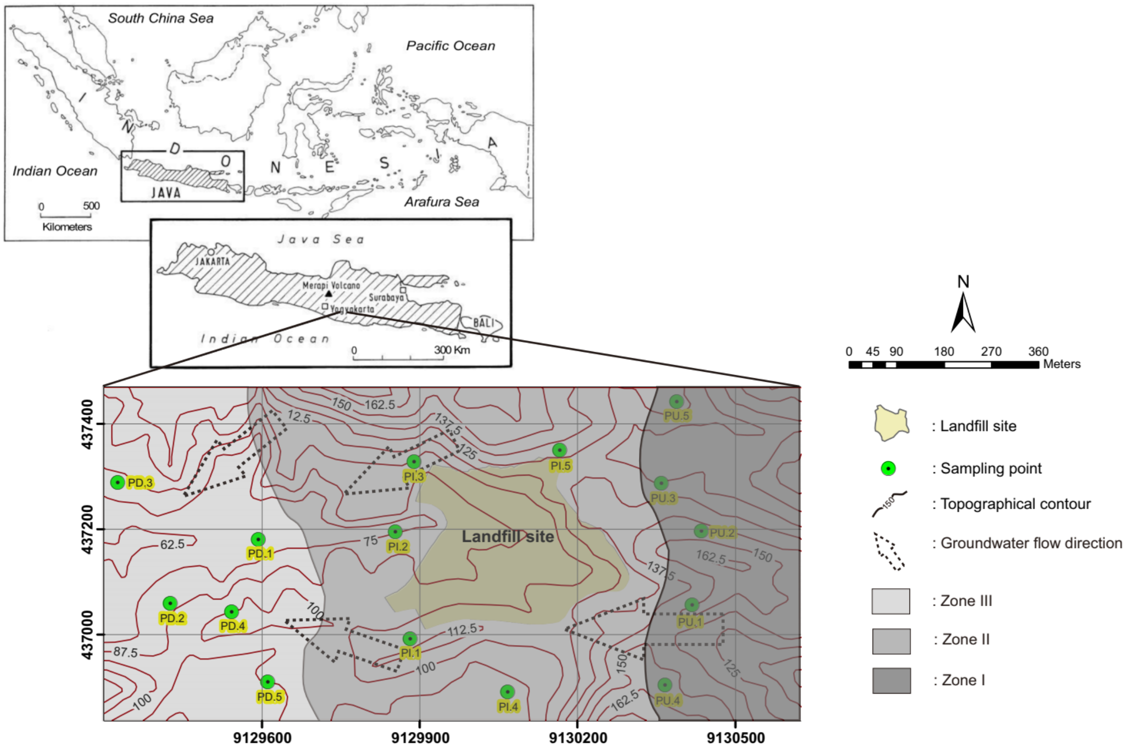

FIGURE 1. Location of the study area and sampling point.

composed of tuff, ash, breccias, agglomerates, and lava. The Young Merapi Volcanic Sediment was a quaternary deposition (Rahardjo *et al.*, 1995).

The lithology in the Piyungan landfill area and its surroundings was divided into five lithology units. From old to young, the lithological unit was composed of tuffaceous claystone - tuffaceous sandstone, andesite breccia, and pumice breccia, which were tertiary depositional products. Furthermore, on top of these units, the alluvial sediment unit and the Kali Opak fluvial deposit unit, which were the products of the quarternary deposition, were deposited. Tuffaceous claystone–tuffaceous sandstone units were most widely exposed in the study area. The pumice breccia unit inserted the tuffaceous claystone–tuffaceous sandstone unit, and it was an autoclastic breccia from the lava flow. The fragments and matrices of these andesite breccias had the same composition, blackish gray, porphyritic, massive, and hard. The cover soil of this andesite breccia was clay in size, reddish-brown, plastic, and soft. There were fragments of pumice and tuffaceous sandstones. At the top, this pumice breccia unit was an intersection of pumice breccia and tuffaceous sandstone with pumice breccia's dominance, and the lower part was tuffaceous sandstone. Alluvial sediment units occupied the morphology of the valley and land. This unit was the result of deposition from rock erosion from the hill morphology. Macro characteristics and descriptions of these alluvial deposits were grayish browns in color, sand size, and poorly sorting. There were rock fragments, not compacted and easy to be crushed. This unit occupied valleys and hills with more than 3 meters of thickness. The Kali Opak fluvial deposit settled the morphology of the river steps. This unit is the result of the fluvial deposition of the Opak River, and this unit was found along the cliffs of the Opak River. Macro characteristics and descriptions of this unit were grayish brown, sand size, good sorting, and the layer had unclear changes. The thickness of this unit is approximately 12–15 meters (Putra, 2001).

1.2 Regional hydrogeology

The leachate flow pattern influenced soil contamination at Piyungan Landfill. Leachate flow was controlled by groundwater networks (flow-net). The direction of groundwater flow movement was also controlled by topographic conditions that affect the groundwater level (hydraulic head). The difference between the two points can also be called the hydraulic slope. So, it was known that the West-Northwest area tends to have a lower groundwater level than the landfill area. The groundwater flows towards the West-Northwest (Figure 1) also showed that the area tends to have shallow groundwater levels (Ramadhan *et al.*, 2019).

2 MATERIAL AND METHODS

2.1 Sampling and analysis

Based on the morphological condition, the Piyungan landfill and its surrounding area are divided into three-zone, as shown in Figure 1. This dividing is based on the elevation and position of each zone to the landfill site. Zone III is located in the area's downslope, and zone I was on the upper slope. Zone II is situated around or very close to the landfill site. The information of groundwater flow direction was obtained from the previous researcher as an essential parameter in assessing the impact of soil heavy metal contamination in the location of the Piyungan landfill.

A sampling of the soil was conducted in each zone (Figure 1), in which five samples were taken in the 25 cm depth from the surface by using a hand auger. About 0.5 kg of soil samples were obtained at each sampling point and put in a plastic bag for laboratory analysis. The samples were dried at room temperature for one week before sending to a laboratory for heavy metal analysis. For laboratory analysis of heavy metals, a 1.5-gram soil sample was crushed by an agate mortar and pestle and sieved to obtain a grain size of < 2mm. The sample digestion was conducted by adding HF–HCl and HNO_3 mixture assisted by microwave digestion described by Bettinelli *et al.* (2000). After being filtered by a 0.45 µm membrane filter, the filtrate was analyzed for the heavy metal concentrations using ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectrometry). The heavy metals analysis in the soil samples was conducted in Hinode Laboratory, Department of International Development Engineering, Tokyo Institute of Technology, Japan. As a representative of each zone, the samples also were taken to analyze physical and chemical properties. Grain size analysis was done by the sieving and hydrometer method (ASTM, 2002). The organic content was measured by Walkley and Black (1934) method. The pH was measured, referring to Conklin (2013). The result of the physical and chemical analyses can be seen in Table 2.

2.2 Heavy metal soil contamination assessment

In this study, the Pb, Cu, Zn, and Cd enrichment in contaminated soil samples was measured by pollution index (*PI*) value and compared with the background concentration of soil. The *PI* is calculated with the Equation 1 (Hakanson, 1980).

$$PI = \frac{C_s^i}{C_n^i} \qquad (1)$$

C_s^i is heavy metal I content in polluted soils, and C_n^i is the heavy background metal I content in unpolluted soils.

After calculation, the values of the *PI* are interpreted by classification as low contamination ($PI < 1$), moderate contamination ($1 \leq PI < 3$), considerable contamination ($3 \leq PI < 6$), and very high contamination ($PI \geq 6$) (Loska *et al.*, 1997).

A pollution load index (*PLI*) was measured in each sampling point to demonstrate the level of heavy metal contamination (Hakanson, 1980). The rating of pollution was classed on four levels based on *PLI* value as follows: $PLI < 1$ no pollution; $1 \leq PLI < 2$ moderate pollution; $2 \leq PLI < 3$ high pollutions, and $PLI \geq 3$ very high pollution (Wang *et al.*, 2010).

$$PLI = \sqrt[4]{PI_{Cu} \times PI_{Zn} \times PI_{Cd} \times PI_{Pb}} \qquad (2)$$

Hakanson (1980) and Deng *et al.* (2010) used the index for the potential soil pollution by eco-risk index (*RI*) and calculated by the Equation 3.

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \cdot PI_i \qquad (3)$$

where *RI* is the index of potential ecological risk and E_r^i is the total calculation for individual heavy metal; each heavy metal toxic response factor i represented by T_r^i; ecological risk represented by subscript r. The background value of the soil used in the assessment was adopted from other studies Wang and Wang (1992) and Hakanson (1980) since there is no available information on the background value of soil and rock in the study area. The T_r^i, C_n^i and RI values are shown in Table 1.

3 RESULTS

3.1 Heavy metals distribution

The soil sample obtained in the study area from three zones had a variation in pH, grain size, and organic content, as shown in Table 2. The pH value had a range of 5.4 to 6.1, and this value appears lower than the normal pH value and is suspected influenced by organic content. This value is confirmed by organic content in soil samples, ranging from 6.8–7.2 %, with no significant difference in the three zones. The grain size analysis showed that the percentage of fine fraction (silt-clay) ranged from 43–48 %, meaning that the samples were loamy sand.

Generally, the heavy metal contents investigated in this study (Pb, Cu, Zn, and Cd) showed that zone II was higher than those in the other two zones. Pb in zone II was higher than the contents in the normal value, but in the zone I and III, the values were normal under normal or natural conditions. Cu and Zn contents in all zone were normal values. However, Cd had a similar pattern as Pb, which only had a value higher than normal in zone II.

3.2 Heavy metal pollution index

Table 3 shows the pollution index's value (PI) in the study area. Generally, Cd showed the highest PI value compared to Pb, Cu, and Zn, with the value of 0.83 to 1.98, and the average was 1.26. Cu had the lowest value compared to Pb, Zn, and Cd and less than one. Zn also had a low PI value, and the highest value was located in zone II. The proportions of total samples of PI value had 60 % in low and 40 % in moderate classification. Generally, the PI value in zone II had a higher value compared to the other two zones. This phenomenon occurred because zone II is located near the landfill site or directly connected to the landfill location.

The PLI value obtained from the calculation, as shown in Table 3, indicates that the PLI value ranged from 0.35 to 1.02. The average PLI value for all zones is 0.61. The proportions of total samples of PLI value had 80% in the low category and 20% in the moderate category. Generally, the highest value of PLI was located in zone II which had a value higher than one, and PLI value in the zone I and III had a value less than one.

The value of E showed that Cd had the highest value compared to the other metals investigated, ranging from 24.92 to 59.54, with an average of 37.69. Cd with many high classifications occupied 20 % sample, 27 % moderate, and 53 % low classification. However, three other metals investigated in this study were included in the low category.

The value of RI for heavy metals in this study had a range of 29.51 to 70.25, showing that heavy metals had a low risk since the proportions of total samples had 100 % in low classification. Generally, the value of RI in zone II was higher than those in the zone I and III.

4 DISCUSSION

4.1 Heavy metal enrichment

Several studies indicated that the presence of landfills had a negative effect on the environment (Vaverková, 2019; Gworek *et al.*, 2016; Fagbenro, 2016). In this study, the value of heavy metal content was normal, except for Cd, which showed significant enrichment. For example, in zone II, the Cd content ranged from 0.05 to 0.29 mg/kg, and this value was higher than in another study (Wuana and Okieimen, 2011). For Pb, Cu, Zn, and Cd, the value was normal, referring to several references (Alfaro *et al.*, 2015; Chernova and Bezuglova, 2019; Santos-Francés *et al.*, 2017). Generally, the enrichment of Cd is due to metals containing in waste in the landfill.

The enrichment of heavy metals in the study area may derive from natural sources in the host rock. However, the rock underlying study area is volcanic rock, which has lower Cd content (Chernova and Bezuglova, 2019; Santos-Francés *et al.*, 2017). The other possibility of the enrichment is leachate containing Cd generated by landfills, which may come from heavy metals waste. According to the previous study, heavy metals were found in the waste in the

TABLE 1. Value of T_r^i, C_n^i, E_r^i and RI for assessment.

Parameter	Value				References
	Pb	Cu	Zn	Cd	
C_n^i	25.5	23.6	86.1	0.13	Wang and Wang (1992)
T_r^i	5	5	1	30	Hakanson (1980)
	Potential ecological risk level				
	Low level	Moderate level	Considerable level	High level	Significantly high level
E_r^i	<30	30 – 60	60 – 120	120 – 240	>240
RI	<110	110 – 220		220 – 440	>440

TABLE 2. Heavy metals and soil properties.

Zones[a]	I (n = 5) Mean ± SD	II (n = 5) Mean ± SD	III (n = 5) Mean ± SD	Total samples n = 15 Mean ± SD	Total samples n = 15 Range
Soil heavy metal content (mg/kg)					
Pb	10.26 ± 1.12	40.38 ± 9.20	15.58 ± 10.49	22.07 ± 16.08	8.90 – 54.20
Cu	2.32 ± 0.46	5.32 ± 1.22	2.58 ± 1.24	3.41 ± 1.66	0.90 – 7.20
Zn	37.96 ± 9.70	143.60 ± 34.99	84.76 ± 11.95	88.77 ± 52.93	23.30 – 187.20
Cd	0.12 ± 0.04	0.26 ± 0.06	0.11 ± 0.05	0.16 ± 0.08	0.05 – 0.32
Soil characteristics					
Organic content (%)	6.8 ± 1.2	6.9 ± 1.3	7.2 ± 1.1	6 ± 0.9	5.8 – 7.8
pH	5.2 ± 0.9	5.7 ± 0.8	6.7 ± 0.2	5.8 ± 0.4	5.2 – 6.4
% silt-clay	45 ± 1.2	46 ± 1.5	48 ± 2.1	43 ± 3.2	43 – 48

[a]zone I, II, and III refer to Figure 1.

Table 3. Summary of the index of pollution (*PI*); load index of pollution (*PLI*); risk factor of potential ecological effect (*E*); risk index of potential ecological effect (*RI*).

Zones	I (n = 5)	II (n = 5)	III (n = 5)	All samples (n = 15)		Proportions of total samples (%)		
	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Range	Low	Moderate	Considerable High
Pollution index (PI)								
Pb	0.40 ± 0.04	1.58 ± 0.36	0.61 ± 0.41	0.87 ± 0.61	0.35 – 2.13	60	40	0
Cu	0.10 ± 0.02	0.23 ± 0.05	0.11 ± 0.05	0.14 ± 0.07	0.04 – 0.31	100	0	0
Zn	0.44 ± 0.11	1.67 ± 0.41	0.98 ± 0.14	1.03 ± 0.57	0.27 – 2.17	47	53	0
Cd	0.95 ± 0.30	1.98 ± 0.43	0.83 ± 0.40	1.26 ± 0.64	0.38 – 2.46	53	47	0
The pollution load index (PLI)								
	0.35 ± 0.05	1.02 ± 0.10	0.45 ± 0.07	0.61 ± 0.32	0.28 – 1.20	80	20	0
Potential ecological risk factor (E)								
Pb	2.01 ± 0.22	7.92 ± 1.80	3.05 ± 2.06	4.33 ± 3.04	1.75 – 10.63	100	0	0
Cu	0.49 ± 0.10	1.13 ± 0.26	0.55 ± 0.26	0.72 ± 0.36	0.19 – 1.53	100	0	0
Zn	0.44 ± 0.11	1.67 ± 0.41	0.98 ± 0.14	1.03 ± 0.57	0.27 – 2.17	100	0	0
Cd	28.62 ± 8.88	59.54 ± 12.79	24.92 ± 12.04	37.69 ± 19.20	11.54 – 73.85	53	27	20
Potential ecological risk index (RI)								
	31.56 ± 8.75	70.25 ± 11.24	29.51 ± 10.76	43.77 ± 21.62	19.94 – 82.95	100	0	0

Piyungan landfill (Sihombing and Darmawan, 2020). Other factors that may influence the enrichment of the metal accumulation in the soil profile were the soil characteristics such as pH, clay, and organic content.

The solubility and transport of most heavy metals in soils, such as Cd and Pb, are likely to be enhanced at low pH (Schubauer-Berigan *et al.*, 1993). Clay content can increase the ability of the soil to absorb heavy metal elements because clay has a large surface area, so that the reaction to heavy metals is getting bigger. At the same time, the presence of organic content in the soil will increase the cation exchange capacity. The high cation exchange capacity will make the soil more reactive and easier to adsorb cations in heavy metals.

Another aspect influencing the accumulation of metals in the soil was rain infiltration, which caused the heavy metal migration down to the soil profile easier. However, the metal accumulation in the soil was not established during the study. Additionally, the extent of migration and enrichment of heavy metals was also influenced by climate conditions, e.g., temperature and precipitation (Clark *et al.*, 1998).

4.2 Ecological impact assessment

The environmental problem near landfills was a severe concern in several studies (Vaverková, 2019; Gworek *et al.*, 2016; Fagbenro, 2016). The problem occurs because of the leachate generation, which affects the soil and groundwater quality.

The assessment result in this study showed that the content of heavy metals in the soil in the study area was generally in the normal values; however, especially for Cd, the content was above the normal. The PI's value in Table 3 showed the low to moderate condition, indicating those metals moderately contaminated soils close to the landfill. Generally, the *PI* value in the soil in zone II was higher than the other two zones. The *PI* value in zone II was higher than the other two zones, indicating that zone II had more heavy metal content due to the waste pile above it. The pile of waste generated leachate that contained large amounts of heavy metal elements. *RI* value also had a similar order in which the location in zone II needs serious attention for agricultural purposes.

The calculation of *PLI* and *RI* showed no significant ecological effect related to soil contamination by heavy metals in the study area. The value of *PLI* in all zone had a value less than two, indicating moderate pollution. Cd's value showed that 27 % sample had a moderate risk and 20 % considerable high risk. This fact demonstrates why Cd's response factor is higher than other metals investigated in this study, and the value of PI for Cd was also higher. The value of RI in the soil in the study area was incorporated to low class, less than 110, which indicates that the ecological risk for four heavy metals in this study covers a low level. This indication may suggest that soon, the effect of heavy metals in the soil in the Piyungan landfill is not significant; however, it will be better to avoid it for agricultural purposes in zone II.

5 CONCLUSION

The study concludes that the heavy metal content investigated in the soil in the Piyungan landfill was varied according to the distance from the Piyungan landfill site. The highest content of heavy soil metals in the study area was found in zone II. Except for Cd, the heavy metals investigated in this study showed that the study area's pollution level was in a low category. There was a different value for Cd than Pb, Cu, and Zn, and it had a potential effect on migrating in the soil profile and needed severe attention. Generally, the content of heavy metals in the soil Piyungan landfill and its surroundings had a low risk to the ecological environment; however, it needs more attention for zone II in agricultural purposes.

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