Urban soil assessment caused by heavy metals contamination in Yogyakarta City, Indonesia

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Abstract: The effect of urbanization and industrialization in the urban city is soil contamination by heavy metals. This study was conducted to assess Pb, Cu, Zn, and Cd, in the soil of Yogyakarta city and its surrounding, Indonesia. The assessment was done by analyzing 45 surface soil samples in the study area, divided into three-zone. They are divided based on the distance of each zone to the center of the city. The zone III is located in the outermost of the study area, and zone I is inside the city. The results of the study showed that generally, the highest concentration of metals was located in zone I, which is located near or directly situated in a city center. The result indicated that Pb and Cd had the highest pollution index compared to Cu and Zn. The pollution load indeks (PLI) and geoaccumulation indeks (I₇geo) calculations in the whole study area showed that the values demonstrated a moderate class in average. Special attention was needed to be given to the zone I, which has a higher PLI and I₇geo index to reduce the source of emission for Pb and Cd.

1. Introduction
The geochemistry of soil contamination by heavy metals is a common problem that has been investigated in several countries around the world in the last decades [1][2][3][4]. In the urban area, the degradation of soil quality is caused by industrialization and urbanization [5]. Heavy metal contamination in surface soil in the urban area is an important issue since this phenomenon will affect the environment and human health. Several heavy metals have a toxic behavior, such as Pb, Cu, Zn, and Cd, and have a carcinogenic characteristic that seriously affects human health [6]. Today, about 55% of the world population lives in urban areas, and 54% of the world's urban population is located in Asia. The urban area has rapid population growth, especially in developing countries [7]. This growth usually is located in the peripheral area outside of the city or in the area projected for city development [8].

Yogyakarta city is one of the developing cities in Indonesia, and a world tourist destination, hence the urbanization is an important issue in this city today [9]. As the implication of urbanization in Yogyakarta city, soil quality degradation is one of the environmental issues. Several studies on the soil contamination in Yogyakarta city, especially for Pb soil contamination, show that the concentration of Pb in surface soil in Yogyakarta city was exceeding the normal concentration in general [10][11][12]. This study aims to conduct the assessment of Pb, Cu, Zn, and Cd contamination in Yogyakarta city and its surrounding area.
2. Material and methods

2.1 Sampling and analysis

The study area is located in Yogyakarta city, Indonesia, and the location is shown in Figure 1. In this study, Yogyakarta city and its surrounding area were divided into three zones, as shown in Figure 1. This dividing is based on the distance of each zone to the center of the city. Zone III was located in the outside of the study area, and zone I was inside the city.

Soil samples were obtained in each zone (Figure 1), in which 15 samples were taken in each zone for 5 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 about two weeks before sending to the laboratory. Laboratory analysis of heavy metals was conducted by preparing a 1.5-gram soil sample that was crushed by agate mortar and pestle and sieved to obtain grain size for <2 mm. The sample digestion was conducted refer to Bettinelli et al., 2000 [13], by adding with strong acid (HF-HCl and HNO₃ mixture) followed by microwave digestion. The heavy metal concentrations in the solution were analyzed by using inductively coupled plasma - atomic emission spectrometry (ICP-AES).

As a representative of each zone (zone I, II, and III), the samples also were taken to analyze for heavy metal analysis. Organic content analysis that was measured by Walkley and Black (1934) method [14] was included. The pH was also measured referring to Conklin (2013) [15].

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Figure 1. Location of study area and sampling.
2.2 Heavy metal assessment in contaminated soil

In this study, the enrichment of Pb, Cu, Zn, and Cd in contaminated soil samples was assessed by pollution index (PI) calculation and compared to the background concentrations in soil. The background concentration was obtained by the previous study, which studied the level of metals in natural unpolluted soil in the study area (Table 1) [16]. The PI was calculated with the formula below [17]:

\[ PI = \frac{C}{B_n} \]  

where \( C \) is heavy metal concentration in contaminated soils, and \( B_n \) is heavy metal background in uncontaminated soils.

| Parameter | Value (mg/kg) | References |
|-----------|--------------|------------|
| Pb        | 25.5         |            |
| Cu        | 23.6         |            |
| Zn        | 86.1         |            |
| Cd        | 0.10         | Wulaningsih, 2009 [16] |

After calculation, the values of the PI were classified as followed: \( PI < 1 \) for low; \( 1 \leq PI < 3 \) for moderate; \( 3 \leq PI < 6 \) for considerable high, and \( PI \geq 6 \) for very high contamination [18].

A pollution load index (PLI) for each sample was calculated to demonstrate the level of pollution of heavy metals [19]:

\[ PLI = \sqrt[4]{PI_{Cu} \times PI_{Zn} \times PI_{Cd} \times PI_{Pb}} \]  

The rating of pollution was then classified on four levels based on PLI value as followed: no pollution with value \( PLI < 1 \); moderate with value \( 1 \leq PLI < 2 \); high with value \( 2 \leq PLI < 3 \); and very high pollution with \( PLI \geq 3 \) [19].

Geoaccumulation index \( (I_{geo}) \) calculation was also used in this study to measure the degree of pollution. This index was calculated as followed [20]:

\[ I_{geo} = \ln \frac{C_n}{1.5*B_n} \]  

\( C_n \) is the measured concentration in the soil samples, \( B_n \) is the background values, and 1.5 is a constant to allow for natural fluctuations analysis in the environment. In this study, the classes of \( I_{geo} \) were simplified by the classification given by Muller (1969) [20]. The simplified classes were as followed: \(<0-1\) is low class; \( >1-3\) moderate class; \( >3-5\) is moderate high class; and \( >5\) is high class.

3. Result

3.1 Heavy metals in soil

The results of the heavy metal analysis are shown in Table 2. The soil samples taken from the study area had variable values of pH and organic content. The value of pH from 5.5 to 6.5, and this value was slightly lower than normal. This lower value was influenced by organic content in soil samples, ranging from 4.1% to 5.8%. The organic content value had no significant difference in each zone in the study area.

In general, the concentrations of heavy metals (Pb, Cu, Zn, and Cd) showed that the zone I had the highest value which was compared to the other two zones in the study area. Especially for Pb and Cd, the value was higher than the natural background in study area. For Cu and Zn, the value was in the range of the natural background. The results showed that there was no significant enrichment in these two metals. By giving an average value for each zone, it can be easier to determine that the zone with a high value has a high risk and requires special attention for the soil remediation strategy. Then, the
locations that have a maximum heavy metal concentration can be affected by land use. This is because the increase in the number of settlements has an impact on increasing the concentration of heavy metals in the soil.

3.2 Heavy metal pollution index

The results of the calculation of the metals index are shown in Table 3. Generally, the highest PLI value was located in zone I. PLI values for Pb and Cd were higher than Cu and Zn in general. They showed that about 96% of the total samples were included in the considerable high class, and that Pb and Cd occupied 69% of total samples for considerable high class.

The results of the PLI calculation showed that the values ranged from 1.16 to 3.13. Generally, the highest PLI value was located in the zone I; with an average value of PLI of 2.72. Table 3 also showed that the value of Igeo in the zone I was the highest compared to the other two zones. Pb and Cd had higher values than Cu and Zn. It seems that Pb had the highest number in the moderate class of Igeo. However, Cd had 11% of the total sample included in the considerable high class of Igeo. The low values of Igeo for Cu and Zn with minus values were similar to the finding of several studies [21][22][23]. These results indicated that there was very low or no pollution in the soil caused by these metals.

Table 2. Heavy metals and soil properties.

| Zones | I (n=15) | II (n=15) | III (n=15) | All samples n=45 |
|-------|----------|-----------|------------|-----------------|
| Mean±SD | Mean±SD | Mean±SD | Mean±SD | Range |
| Soil heavy metal concentration (mg/kg) | | | | |
| Pb | 105±12.4 | 85±14.4 | 59.8±24.7 | 83.3±6.59 | 22-129 |
| Cu | 46.9±2.83 | 45.4±9.34 | 41.8±3.34 | 44.7±3.62 | 34-67 |
| Zn | 76.3±4.06 | 70.6±10.3 | 75.3±4.95 | 74.1±3.39 | 49-81 |
| Cd | 1.19±0.44 | 0.54±0.31 | 0.19±0.12 | 0.64±0.16 | 0.1-1.9 |
| Soil characteristics | | | | |
| Organic content(%) | 4.8±1.2 | 4.9±1.3 | 4.2±1.1 | 4.6±0.9 | 4.1-5.8 |
| pH | 5.9±0.9 | 6.2±0.8 | 6.1±0.2 | 6.1±0.4 | 5.5-6.5 |

*see Figure 1 for zone I, II and III

Table 3. The value of pollution index (PI), pollution load index (PLI), and geoaccumulation index (Igeo).

| Zones | I (n=15) | II (n=15) | III (n=15) | All samples (n=45) |
|-------|----------|-----------|------------|-------------------|
| Mean±SD | Mean±SD | Mean±SD | Mean±SD | Proportions of total samples (%) | Low | Moderate | Considerable High | High |
| Pollution index (PI) | | | | | 0 | 4 | 96 | 0 |
| Pb | 8.92±1.05 | 7.20±1.22 | 5.07±2.09 | 7.06±2.18 | 1.86-10.9 | 0 | 4 | 96 | 0 |
| Cu | 1.35±0.08 | 1.30±0.27 | 41.8±0.10 | 1.28±0.18 | 0.98-1.93 | 2 | 98 | 0 | 0 |
| Zn | 1.10±0.06 | 1.02±0.15 | 1.09±0.07 | 1.07±0.11 | 0.71-1.7 | 20 | 80 | 0 | 0 |
| Cd | 11.9±4.42 | 5.4±3.09 | 0.83±0.40 | 1.87±1.25 | 1-19 | 0 | 31 | 69 | 0 |
| Pollution load index (PLI) | 2.72±0.23 | 2.14±0.39 | 1.59±0.30 | 21.5±0.56 | 1.16-3.13 | 0 | 44 | 51 | 4 |
| Geoaccumulation index (Igeo) | | | | | 0 | 4 | 93 | 0 | 0 |
| Pb | 2.56±0.17 | 2.24±0.26 | 3.05±2.06 | 1.62±0.68 | 0.31-2.87 | 7 | 93 | 0 | 0 |
| Cu | -0.49±0.08 | -0.23±0.26 | -0.33±0.12 | -0.24±0.18 | -0.62-0.36 | 100 | 0 | 0 | 0 |
| Zn | -0.45±0.08 | -0.57±0.23 | -0.47±0.10 | -0.50±0.16 | -1.09-0.36 | 100 | 0 | 0 | 0 |
| Cd | 2.90±0.51 | 1.60±0.89 | 0.08±0.82 | 1.53±1.38 | -0.58-3.66 | 40 | 27 | 11 | 0 |
4. Discussion

4.1 Heavy metals enrichment in soils

Several studies have been done on the concentration of heavy metals in surface soils in urban cities. The results of this study showed that most Pb had a higher concentration than normal compared to several references. Cd also showed a similar pattern to Pb, but Cu and Zn had a lower than normal values in general. The high Pb and Cd concentration in soil in the study area are suspected from human activities such as vehicle emission and industry. Enrichment of heavy metals in soils may be derived from the host rock. However, the host rock in the study area is a volcanic rock, which has very low contents of the heavy metals. This, the enrichment may be derived from human activities. Another factor influenced may be from pH and organic content in the soil.

Vehicle emission gives the most contributions for Pb concentration in the surface soil in the urban city, as stated by several researchers [24][25]. The vehicle emission is generated by carbon monoxide and the burning of lead in the vehicle engine. The emission comes from engine friction, vehicle rusty, and gas emission due to incomplete combustion. These substances then spread with air and deposit in the surface soil. According to another study, the concentration of heavy metals in the air in several big cities in Indonesia contain Pb (0.2-2664), Zn (2.9-913), Cu (0.05-18.79) in mg/m$^3$ [26].

4.2 Environmental impact assessment

The heavy metals in the soil in urban areas were a serious consideration in several studies [2][3][4]. The result of the calculation of the assessment in this study showed that the heavy metals in the soil in the study area had high Pb and Cd, and the concentration is above normal. The PLI's value in Table 3 showed that the values were on the moderate to high condition, except for Cu and Zn. Generally, the PI value in the soil in the zone I was higher than the other two zones. The $I_{geo}$ values in zone I had the highest value in the study area due to the closest location to the center of the city. Indeed, the location in zone I had the highest risk for the environment.

The calculation of PLI and $I_{geo}$ showed a difference in the prediction of pollution level in the soil in the study area. According to PLI, the soil samples are in the range of moderate to considerable high; however, according to $I_{geo}$ the samples are included in the low to considerable high level. Overall, compared to the natural background level (Table 1), the Pb and Cd had higher values. On the other hand, Cu and Zn had lower values. This difference may be due to the metals' source because the human activities generating Pb and Cd are more intensive in the study area. Another factor that may influence is the soil characteristic in the study area, with strong bonding for Cu and Zn in the soil particle. This phenomenon needs to be investigated more in-depth in the future.

5. Conclusions

The investigation of the study concluded that the concentrations of heavy metals (Pb, Cu, Zn, and Cd) in the soil in Yogyakarta city and its surrounding area varied according to the distance from the city center. The highest concentration of Pb and Cd in the soil in the study area was found in zone I due to the intensive traffic and industrial area. Cu and Zn in the soil in this study showed that the level of pollution in the study area was in a moderate category. There was a different value of pollution index depending on heavy metal and a potential effect on migrating in the soil profile. Generally, the concentration of heavy metals in the soil in the study area were moderate; however, it still needs more attention for zone I especially for human activities in that zone. Several policies need to be made in order to reduce Pb and Cd emission in the area.

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