LEAD CONTAMINATION IN SOIL OF YOGYAKARTA CITY, INDONESIA

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Abstract

This paper investigates on Pb concentrations and mobility in soil of overall Yogyakarta City, Indonesia. The objectives of this study include to investigate Pb concentrations in the surface soil throughout Yogyakarta and to study their possible sources and potential environmental impacts. The soil samples from 168 locations in Yogyakarta was obtained, representing different land use and traffic conditions. Spatial analysis and sequential extraction analysis were performed. Generally, contour pattern of Pb value clearly shows that high value is concentrated in the middle and few north part of the study area, confirmed by traffic condition in the study area. In general, the results of sequential extraction analysis shows that Pb was predominantly associated with exchangeable fraction. The presence of lead in the exchangeable fractions may pose a serious environmental concern since they are highly soluble and potentially bio-available in the soil of study area.

Keywords: Soil, lead contamination, GIS, sequential extraction.

1 Introduction

Anthropogenic activities in urban areas concentrate potential toxic materials, such as lead (Pb) that may be inadvertently or deliberately released into the environment. Human exposure to these toxins may inevitably result in adverse effects on health that may be either accute or chronic. Thus, it becomes imperative that concentrations of Pb and other hazardous materials in the environment must be adequately examined in order to support a development of regulations and standards to minimize the risks associated with these materials in urban areas. Lead emissions from vehicles are the major source of the contamination, and the nature of the source ensures wide dispersion of the pollutant. Since there is a continuous intercompartmental exchange of Pb among air, water, and soil, most airborne Pb is eventually deposited onto some surface, including soil, plants, water bodies, artificial surfaces, and the respiratory tracts of animal and human by dry or wet deposition processes. Pb is a good indicator of contamination in soil because it appears in gasoline, car components, oil lubricants, industrial and incinerator emissions. Pb in soil results mainly from dry and wet deposition of atmospheric Pb, particularly close to emission sources, as well as its natural occurence in soil (Alloway, 1990).

Yogyakarta City is located in the central part of Java Island. In the 1930's, Yogyakarta was just a small town in the interior of Java with a population of approximately 60.000 inhabitants (Baiquni, 2004). In the last two decades, urbanization has transformed the structures of Yogyakarta City and it grows beyond its administrative boundary with about one million inhabitants (Subanu, 2004). Urbanization has transformed rural dwellings to become urban settlements and generated an urban agglomeration area. The Yogyakarta urban agglomeration area consists of the Yogyakarta City municipality and two regencies, i.e., Sleman and Bantul. Percentage of urban population in the Yogyakarta

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Province has significantly increased from 22% in 1980 to 58% in 2000. This increase has simultaneously occurred with the growth of the urban area. The population density in the Yogyakarta City municipality area varies between 10,000 to 30,000 person/km², while the population density in its agglomeration area ranges between 1,000 to 3,000 person/km². Since the 1980s, rapid urbanization has been taking place in the north part with a number of residentials built up in the north and west parts of the traditional agricultural area. The new residentials usually consist of residential and commercial sites. The development of the number of vehicles that create rapid air quality change in the city of Yogyakarta is increased. In addition to cause air pollution, the development of vehicles noise triggers threshold beyond normal level.

Pb has been added to gasoline since the early 1920s as tetraethyl and tetramethyl lead to make combustion more efficient. However, research shows that the Pb emitted through the exhaust system had a detrimental effect on the environment with estimates that over 75% of environmental Pb was derived from this source. The use of lead-containing fuel has been used in all countries and gasoline with Pb additives is still used in many developing countries. The data obtained from the Agency of Environmental Protection of Provincial Government of Yogyakarta throughout 2008 shows that nitrogen dioxide concentration (NO₂) in the air has exceeded the threshold standard of quality. The findings degree of NO₂ in the air was 0.0045-0.0018 ppm, or exceeding the limit of 0.0129 ppm standard quality. In addition to NO₂, Pb concentration in the air has also exceeded standard quality. Currently, the repetitive measurement in the air around 0.3656–75.7 microgram per cubic meter ($\mu g/m^3$), or exceed the maximum threshold in the air of 2.00 μ g/m³.

There were previous studies on the distribution of heavy metals in soil in Yogyakarta (Wardhana and Muryono, 2000; Budianta, 2009a; Budianta 2009b; Budianta, 2010). However, this study focuses only on limited locations, particularly along major traffic roads and there is no extensive survey on general soil conditions in Yogyakarta. Pb concentrations in

the soil of the overall Yogyakarta environment have never been studied systematically, and the extent of soil contamination in the city remains unknown.

The objectives of this study include the investigation of the concentrations of Pb in the surface soil throughout Yogyakarta and to study their possible sources and potential environmental impact. The data arises from this study may provide the needed information that may serve as a springboard for action by private and government environmental managers in the attempt to minimize environmental health problems.

2 Material and Methods

2.1 The study area, sampling method and analysis

The study area is focused in Yogyakarta City and its surroundings which consist of commercial and traditional residential areas that are located in the northern part. The soil samples from 168 locations in Yogyakarta was taken to represent the different land use and traffic conditions. The soil samples was obtained by gridding the study area by 0.5 km \times 0.5 km wide and the samples was obtained in every grid. The area in Yogyakarta City may be classified into two different land use areas, namely urban and suburban areas according to its adjacent land use. Each of the soil samples obtained using a stainless steel hand auger to follow the US EPA standard operation procedure (U.S. Environmental Protection Agency, 2007).

Surface soil from a depth at 0 to 5 cm is sampled from representative areas. Two to three composite samples was collected within each sampling site. The sampling was designed to investigate Pb concentrations in representative soil, and thus soil very close to site-specific pollution sources, e.g. landfills, waste stations, industrial plants, gasoline stations, etc., will be avoided except for a few urban soil near highways (urban soil are commonly distributed near highways in Yogyakarta). The soil samples collected are stored in plastic bags for subsequent sample preparation and analysis. Soil samples collected were air-dried at room temperature for seven days, screened through a 2-mm sieve and then Pb concentrations will be analyzed by a flame absorption spectrophotomer (AAS) (Varian AA-20).

2.2 Spatial analysis and sequential extraction

Analysis Spatial analysis using geostatistics based on Geographic Information System (GIS) was performed. The Pb concentrations are used as the input data for Pb soil contamination maps to study the distribution of Pb in urban soil. The software used for mapping and spatial analysis was Arcview. An interpolation method was determined for the interpolation of the geographical data (Webster and Oliver, 2007; Shi *et al*, 2002)

Sequential extraction analysis was also conducted and used to operationally define heavy metals into different geochemical phases, usually in the order of increasing stability. A fivestep sequential chemical extraction, commonly referred to as Tessier?s method, is probably one of the most widely used sequential chemical extraction methods (Tessier et al, 1979) that defines metals into five fractions with increasing stability through the use of progressively reactive extractants. The extracted metals from these five consecutive steps are operationally defined into five geochemical fractions: (1) readily soluble and exchangeable; (2) carbonate-bound, specifically adsorbed, and weak organic and inorganic complexes; (3) bound to iron and manganese oxides; (4) bound to stable organic and/or sulphide complexes; and (5) residual fractions containing primary and secondary minerals held within their crystal structure.

It is acknowledged that the reactivity and potential bio-availability of heavy metals generally increases with increasing solubility. Thus, the first two forms are usually considered as the two most mobile forms of metals in soil and are potentially bio-available to plants and animals. The last three are relatively immobile and stable, but may sometimes become mobile and bio-available with changes of soil conditions. In each experiment, 10 gram of soil sample was

Table 1: Heavy metals concentration in major rock type.

Element	Geologic rock type ^a				
	Ultra-mafic	Mafic	Sandstone		
Cd	0.12	0.13	0.05		
Со	110	35	0.3		
Cr	2980	200	35		
Cu	42	90	30		
Fe ^b	94000	-	9800		
Mn	1040	1500	460		
Ni	2000	150	9		
Pb 14		3	10		
V 40		250	20		
Zn	58	100	30		
a	1 0 (10(=)	1 5	1 (1 0 = 0)		

Source: ^a Krauskopf (1967) and Rose *et al* (1979); ^bCannon (1978).

used, and the extraction condition for the sequential extraction analysis.

3 Results and Discussion

3.1 Geology of study area

The rocks underlying the study area fall within Holocene age. Volcanic rocks and their derivatives dominate the area, particularly in the Yogyakarta Basin where extensive deposit of alluvium that is derived from Merapi Volcano ejects lies. According to Sir M. MacDonald and Partner (1984), Old Merapi Volcanics consist of strongly fractured basalt and andesite lavas, with indurated breccias, outcrops around upper cone of Volcano Merapi and are the deposit of Upper Pleistocene Merapi volcanism. The buried extension of these deposits does not extend far from south. In Yogyakarta, a deep borehole has penetrated the soil through the post Pleistocene deposits directly into tertiary strata.

Soil background samples gives a Pb concentration of 5 mg/kg. Soil Pb level was also influenced by the composition of parent materials (Table 1). Native concentrations of Pb are relatively high in ultra-mafic usually lower in sandstone and mafic rock. Lead in soil is largely insoluble, low in mobility, and seldom leached from the profile.

Properties	Sample	
Classification	Sandy loam	
Sand-Gravel (%) ¹⁾	77.71	
Silt-Clay (%) ¹⁾	22.9	
pH	7.2	
Organic content (%) $^{2)}$	2.88	
Specific Surface Area (m ²) ³	19.5	
Specific Gravity	2.73	
	Quartz,	
Tatal Minanala sigal	Feldspar,	
Composition ⁴⁾	Albite,	
	Kaolinite,	
	Illite	
Cation Exchange	14	
Capacity (meq/100 gram) ⁵⁾	- ·	

Table 2: Several properties soil sample.

Analyzed by using:

¹⁾ Sieving and hydrometer test

²⁾ Total Organic Carbon (TOC) Analyzer

³⁾ Brunauer, Emmitt and Teller (BET) method

⁴⁾ X-ray Diffraction

⁵⁾ BaCl₂·2H₂O compulsive exchange method

3.2 Soil properties

Several properties of soil samples can be seen in Table 2. The result of grain-size distribution was analyzed by standard methods for soil (ASTM). The average of grain-size distribution indicated that the soil samples contained approximately 10–20% clay-silt size particles and the remaining is sand. The average pH of the soil suspension with a ratio by weight of 1:1 of soil to water was 6.65. Specific surface area was measured using BET method. Determination of the cation exchange capacity (CEC) used $BaCl_2 \cdot 2H_2O$ compulsive exchange method. X-Ray diffraction analysis (powder method) revealed that the samples contained kaolinite, illite, chlorite, quartz and feldspar.

3.3 Distribution patterns of Pb

The Pb content of 168 samples that were selected in urban and suburban areas varied widely, ranging from 25 to 95.5 mg/kg. The mean value was 64 mg/kg, 15 times the background value. In this case, Pb was not associated with lithological factor. It is evident that soil in Yogyakarta city was highly enriched in

Table 3:	Summarized	statistical	obtained	data.
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N	Min	Maks	Mean	Std. Dev
168	16	95.20	65.4	10.18

Pb compared to background levels. The result of laboratory analysis of Pb in soil was summarized (Table 3).

Geostatistical analysis was applied to the Pb lateral distribution. Geostatistics provide an advanced methodology which facilitates quantification of the spatial features of soil parameters and enables spatial interpolation. Generally, contour pattern of Pb value clearly shows that high value is concentrated in the middle and few north part of the study area (Figure 1). The middle part of study area has high traffic density, confirmed by traffic condition in Figure 2. Although we have deliberately avoided sampling close to major roads, it is easily noticed that the highest lead values correspond to the major traffic density, as shown in Figure 2.

During the 1990–2010 period, the number of vehicles (cars and motorbikes) and also the road increased significantly. The main road and traffic activity in study area were shown in Figure 2. Anthropogenic activity in study area results in elevated of lead concentration in top soil.

This must have led to some improvements in the environmental quality of the inner city but may have also led to a deterioration in soil quality in suburban areas. Moreover, the suburban environment was also subject to the effects of emission of vehicles and the development of rural enterprises. Pb in soil usually decreases with increasing distance from a road (Fakayode and Olu-Owolabi, 2003). Lin Jian et al. (2000) described the Pb concentrations in surface soil near the road were greater under heavy than light traffic. This is confirmed by the distribution map of Pb in the atmosphere in study area as shown in Figure 3. Vehicles emission was the primary pollution source.

The occurrence and frequency of higher concentrations of pollutants in the atmosphere primarily depend on the magnitude and distribution of the sources of emissions, local topography, climate conditions and type of pollutant. The level of the pollution is mainly in correla-

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Figure 1: Distribution of Pb in soil in study area.

tion with the degree of the traffic density. However, because of topographical influence, wind speed, wind direction, vehicle speed and traffic flow, the peak of Pb contamination does not always occur near roads, and may have gaps from road location. This may be expressed by the slightly high concentration of Pb in soil the northern part of study area. As a result, Pb contamination occurs mainly in the surface soil due to human factors.

3.4 Lead concentrations in different areas

According to urban planning in Yogyakarta city, there are two area in the city: urban and suburban area. As expected, Pb concentrations in different urban areas differs, yet overall Pb contamination is higher in the urban area. The mean soil Pb concentration was 62 mg/kg, about five times that in the suburban area (14 mg/kg) (Figure 4).

3.5 Lead mobility on soil

As mentioned earlier, the partitioning of soil was performed by the techniques proposed by Tessier et al. (1979). The chemical partition-

Table 4: Summarized statistical Pb soil data in urban and suburban area (mg/kg).

	N	Min	Maks	Mean	Std. Dev
Urban	99	34	95.20	62	7.19
Suburban	69	16	32	14	4.65

ing of Pb in the selected soil at urban and suburban is shown in Figure 5. Despite the differences in Pb concentrations at the areas (urban and suburban), the distribution of Pb in five chemical fractions displayed a similar pattern. In general, the results showed that Pb was predominantly associated with the exchangeable fraction in the study areas, accounting for ~50% of the total Pb in soil. The relatively high concentrations of lead in the exchangeable fractions of the soil may be attributed to pH level which encourages desorption of metals from soil. The presence of lead in the exchangeable fractions may pose a serious environmental concern since they are readily available for uptake by plants.

The second largest fraction of Pb was found in the carbonate fraction (10–25% of the total Pb). Pb in the organic and residual fractions



Figure 2: Traffic condition in study area (Yogyakarta Urban Air Quality Improvement Program-UAQ-i, 2006.

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Figure 3: Distribution of Pb in atmosphere in study area (Yogyakarta Urban Air Quality Improvement Program-UAQ-i, 2006.



Figure 4: The mean of Pb concentration in the urban and suburban area.



Figure 5: Chemical partitioning of Pb in study area.

collectively accounts for approximately 5–10% of the total Pb. Pb in the exchangeable fraction was the highest among the five fractions. Similar observation shows that the high mobility of lead, mainly in the upper soil horizon, may be caused by the enhanced amount of lead by atmospheric fallout (Plant and Raiswell, 1983). The very few proportion of metals found in the residual fraction shows the effect of parent material at soil formation. Metals in the residual fraction are only available after weathering or decomposition.

4 Conclusion

Results obtained in this study generally revealed the presence of Pb soil in Yogyakarta, with the most probable source of Pb is from motor vehicles via atmospheric deposition, although further study is needed to confirm these. The chemical partitioning of the Pb generally showed that the majority of the Pb in the soil was relatively unstable. The comparatively significant association of Pb with the exchangeable fractions indicated that ~50% of the total Pb could be easily soluble and potentially bioavailable in these urban soil.

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