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WORD COUNT

5461

TIME SUBMITTED

08-APR-2020 04:53PM

PAPER ID

57311320

Effect of Organic Matter Amendment on Lead Contamination in Roadside Soil and Plant

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Received 26 November 2009 / accepted 4 January 2010

ABSTRACT

Lead Contamination in Roadside Soil and Plant and Effect of Organic Matter Amendment (Sabaruddin, D Budianta and Mardia): Roadside soils and plants may be the most important sink of lead (Pb). It has been widely known that soil organic matter (SOM) plays important roles in determining concentrations of metals in soil solution and their extractability from the soil. To investigate Pb contamination in the roadside soils and plants, as well as the effect of organic matter (OM) on the soluble Pb in the roadside soils, surface soils (0 to 20 cm) were collected from a busy road. The soils were incubated for 4 weeks under room temperature after being treated with 0, 30, 60 and 90 Mg ha⁻¹ of OM. Leaves of oil palms (*Elaeis guineensis*) planted on the roadside were also analyzed for Pb content. Current study revealed that Pb content in roadside soils and leaves of oil palm was 1.5 and 5.5 times higher than the safe level of Pb in soil and plant. It confirms that both soil and plant at the study site were contaminated by Pb. Current study also showed that SOM amendment significantly (P<0.01) affected soluble Pb content in the soils. Adding OM to the soil at 30 Mg ha⁻¹ to correct the level of SOC from very low to low was sufficient to significantly reduce soluble Pb in the soils. However, the application of 60 Mg ha⁻¹ of OM triggered the increases in soluble Pb in the soils. Further increases in OM application to 90 Mg ha⁻¹ resulted in significant increases in soluble Pb as compared with that in the soil receiving 30 Mg ha⁻¹ of OM. In spite of the increases, the level of soluble Pb in the soils receiving 60 and 90 Mg ha⁻¹ of OM was still much below the safe level of Pb in soil.

Keywords: Organic matter, Pb, roadside soil

INTRODUCTION

Heavy metal contamination in the environment is of major concern because the majority of the heavy metals are toxic to the living organisms and the environment, even those considered as essential can be toxic if present in excess (Chirenje *et al.* 2003; Dolan *et al.* 2006; Jarup 2003; Ma and Rao 1997; Silva *et al.* 2005). Heavy metals may come from many different sources, including vehicle emissions, industrial discharge and other activities (Gibson and Farmer 1986; Harrison *et al.* 1981; Li *et al.* 2001; Thornton 1991; Wu *et al.* 2006). However, the extensive use of automobile is one of the most important sources of heavy metal contamination in the roadside environment (Garcia and Millán 1994). Worldwide, leaded petrol has been reported to have

caused more exposure to lead pollution in human beings than any other single source. Therefore, the USA, all European countries, and many developing countries have outlawed or strictly regulated the use of leaded petrol.

Research into ecological effects of road traffic has greatly increased. The impact of traffic on the surrounding environment is complex, including the disturbance during construction and the pollution from both road material itself and from the traffic once the road is built (Gibson and Farmer, 1986; Harrison *et al.* 1981; Li *et al.* 2001; Thornton 1991; Wu *et al.* 2006). Motor vehicles introduce a number of pollutants into the environment. In the vicinity of roads, dust and the other traffic-related pollutants such as heavy metals, polycyclic aromatic hydrocarbon particles (PAH) and volatile organic compounds

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(VOCs) exert influence over plant growth and soil. A variety of heavy metals derived from gasoline additives settle into the roadside environment (Gibson and Farmer 1986; Harrison *et al.* 1981; Li *et al.* 2001; Sujetovienė and Griauslytė 2008; Thornton 1991; Wu *et al.* 2006).

Roadside soils and plants may be the most important sink of metals and other pollutants. A variety of heavy metals derived from petrol additives settle into the roadside soils and plants. Hence both roadside soils and plants may contain elevated concentrations of metals which may have detrimental and toxicological effects on both environmental quality and human health (Sinha *et al.* 1997). The most widely documented metal is lead, but the others are aluminum, iron, cadmium, copper, manganese, titanium, nickel, zinc and boron (Spencer and Port 1988; Sujetovienė and Griaušytė 2008).

More recently Nabuloa *et al.* (2006) and Viard *et al.* (2004) reported that the deposition of metals in surface soils decreased with increasing distances from the highway. Although the concentrations of metals in the roadside soil were influenced by meteorological conditions (Othman *et al.* 1997; Sezgin *et al.* 2003), traffic density (Garcia and Millán 1994; Nabuloa *et al.* 2006) and the kind of vehicle in traffic (Nabuloa *et al.* 2006; Sezgin *et al.* 2003), and soil parameters (Viard *et al.* 2004). However, this elevated level of heavy metals is frequently found 200 m or more from the road (Nurjaya and Saeni 2006).

Heavy metal mobility and solubility in soils are closely related to metal solubility, which is further regulated by adsorption, precipitation and ion exchange reactions in soils. Since Pb in soils is elemental, it is not subject to chemical degradation. Formation of insoluble heavy metal compounds immobilizes the metal and reduces their bioavailability (Cotter-Howells and Caporn 1996). Chemical conditions in soil are important determinants of heavy metal transport and fate. The importance of interactions between metals and solid phases of soils, soil water, and air within and above soil depends on a variety of chemical factors. Absorption of metals from soil water to soil particles is the most important chemical determinant that limits mobility in soils (Alloway 1995). One of chemical determinants is soil organic matter (SOM). SOM (e.g., humus) adsorbs metals by forming chelate complexes, with carboxyl groups playing a predominant role and has the greatest capacity and strength of bonding with most metals of any soil

component. This is illustrated by the commonly observed positive association between solubility of metals (for example Cu and Cd) and SOM content as well as the greater concentration of trace metals in organic rich soils compared with organic-poor soils.

In a study by McGrath *et al.* (1988) the effect of SOM was determined on concentrations of metals in solution and their extractability from the soil. They found that increased concentration of SOM depressed the concentration of cupric ions in soil solution as well as the extractability into acetic acid and CaCl₂ of both native and added Cu; effects on the extractability of Zn and Mn, in the other hand, were much smaller. Cu was observed to be more strongly adsorbed into acid-washed peat, peat and solid humic acids compared with clay minerals or iron oxides.

Although there have been a considerable number of studies on the concentrations of heavy metals in roadside soil and plants, the vast majority have been carried out in developed countries (Chen *et al.* 2003; Dolan *et al.* 2006; Jarup 2003; Li *et al.* 2001; Ma and Rao 1997; Silva *et al.* 2005; Sujetovienė and Griauslytė 2008; Wu *et al.* 2006). Very few studies have been carried out in developing countries such as Indonesia (Nurjaya and Saeni 2006). Therefore, the present research was undertaken to study lead contamination in the roadside soils and plants in relation to their safe limit. In addition, the effect of organic matter on the solubility of lead in the roadside soils was also investigated.

MATERIALS AND METHODS

Site Description

One of major roads in Palembang City was chosen for the study site. The roadsides were planted with oil palms (*Elaeis guineensis*). There were 350 plants on one side and 317 plants on the other side of the road. The fruit bunches of the oil palms are regularly harvested. The surrounding areas on the right and left of the road were rain-fed paddy field. The average traffic load from 07.00 AM to 05.00 PM was 12,000 vehicles per day. The soil belongs to Ultisols (Soil Survey Staff 1992) or Acrisols (FAO/UNESCO 1974).

Sampling and Analyses

For sampling purposes, 10% of the total plants (35 stands and 31 stands on each site) were chosen. The leaves of the oil palms were collected from each

of the selected plants. Top soils (0-20 cm) underneath the canopy of each of the selected plants were also sampled. All the roadside soil samples were air dried at room temperature and sieved through a 2-mm nylon sieve to remove coarse debris. All air-dried soil samples were then ground with a pestle and mortar until all particles passed a 100-mesh nylon sieve and characterized. Soil pH was measured potentiometrically in a soil- to water- ratio of 5 g to 25 mL (designated as pH-H₂O). Exchangeable cations (Ca, Mg, K, and Na) were extracted with 1M NH₄-OAc (pH 7.0) with titration method. Then, the filtrate was washed with deionized water once and twice with 99% EtOH to remove the salt excess. The ammonium was extracted with 10%-NaCl solution twice, followed by reciprocal shaking for 1 h and centrifugation for 10 min at 1,000 rpm. The ammonium ion content was determined as cation exchange capacity (CEC) by Kjeldahl distillation and titration method. Organic Carbon was measured by the Walkley-Black titration method (Walkley and Black 1934), and plant available Pb was extracted according to EDTA method by Miller *et al.* (1975) and measured using atomic absorption spectrophotometry. Particle size distribution was analyzed following the hydrometer method (Bouyoucos 1962).

The soil used in the current experiment was highly acidic (pH_{H₂O} = 4.12), with low base content (K-dd = 0.19 cmol(+) kg⁻¹, Na-dd = 0.05 cmol(+) kg⁻¹, Ca-dd = 0.85 cmol(+) kg⁻¹, and Mg-dd = 0.45 cmol(+) kg⁻¹), very low Organic C content (5.70 g kg⁻¹), low CEC (15.23 cmol(+) kg⁻¹), dominated by sand (43%) and clay fraction (40%) and had initial content of Pb of 20.01 mg kg⁻¹.

To evaluate the plant and soil contamination level, current findings were compared to the background levels of Pb in plant and soil, which were 2.0 mg kg⁻¹ and 12.75 mg kg⁻¹ for plant and soil, consecutively (Balai Penelitian Tanah Indonesia 2002).

Pb Emission Potentials

Pb Emission Potentials was calculated using the following formula:

$$E_{pb} = A \times B \times C \times D$$

where:

E_{pb}: Pb Emission Potentials

A: Proportion of vehicle using premium and solar (%)

B: Total vehicle passing through the road (unit day⁻¹)

C: Fuel consumption for the 3.5 km of road (L), assuming 1 L per 10 km and per 1 L per 15 km for gasoline and diesel, consecutively.

D: Pb content in petrol and diesel in Palembang is 0.999 g L⁻¹ and 0.149 g L⁻¹, consecutively (Suyasning 2008).

Soil Organic Matter Amendment

As mentioned above that the soils used in current experiment had very low organic C content (5.70 g kg⁻¹); whereas SOM plays an important role in determining concentrations of metals in soil solution and their extractability from the soil (McGrath *et al.* 1988). To investigate effects of SOM on the solubility of Pb, the contaminated soils were amended with 30, 60 and 90 Mg ha⁻¹ of compost to attain organic C of 10.00, 20.10 and 30.10 g kg⁻¹, equivalent to low, moderate and high content of organic C, respectively. Untreated soil (0.00 Mg ha⁻¹ of compost) with inherent level of Pb (5.70 g kg⁻¹) was used as a control. The compost contained 20 to 25 mg kg⁻¹ of total N, 5 to 7 mg kg⁻¹ of available P, 30 to 50 mg kg⁻¹ of K₂O, and had C/N ratio of 15. More importantly that the inherent Pb content of the compost (3.86 mg kg⁻¹) is much below standard according to SNI 19-7030-2004 (<150 mg kg⁻¹). The compost was added to 1 kg of soil, thoroughly mixed, potted, then the mixture incubated in room temperature for 30 days. Water level was maintained at field capacity during incubation.

Data Analysis

Data on Pb content of incubated soils were expressed as an average of six replicates and treatment effects were determined by analysis of variance. Differences among treatment means were compared by least significant difference (LSD) at *P*<0.05.

RESULTS AND DISCUSSION

Pb Accumulation in Soil

Current study revealed that Pb content in roadside soils at the study site was 20.01 mg kg⁻¹. This level was 1.5 times higher than the safe level of Pb in soil (12.75 mg kg⁻¹) as regulated by Balai Penelitian Tanah Indonesia (2002) and it reveals that the roadside soils at the study site are polluted by Pb. The solubility of metals in soil is predominantly controlled by pH (Henry 2000), cation exchange capacity (Martinez and Motto 2000), organic carbon

1 content (Elliot *et al.* 1986), the oxidation state of the mineral components, and the redox potential of the system (Connell and Miller 1984). The first three factors appear to be predominant contributing factors to the excessive level of Pb content in the soils of current study.

Soil pH and CEC levels had a very high significant effect on 3 the amount of Pb content in the soil samples (Shaikh *et al.* 2006). In general, soil pH seems to have the greatest effect of any single factor on the solubility or retention of metals in soils, with a greater retention and lower solubility of metal cations occurring at high soil pH (Basta *et al.* 1993). Similar studies elsewhere (Adriano 2001; McLaughlin and Singh 1999) showed that high levels of pH and CEC tended to be negatively associated with concentrations of heavy metals in soil. However, the soil at the study site was highly acidic (pH of 4.12), low CEC (15.23 cmol(+) kg⁻¹) and very low organic C content (5.70 g kg⁻¹).

In addition to the above factors, the content of heavy metals in roadside soils is also predetermined by traffic intensity on the road (number of vehicles per day), predominant wind direction, soil characteristics and vegetation (Zinkutė *et al.* 2007). In relation to the current study, number of vehicles, soil characteristics and vegetation seem to be important causes of significantly elevating Pb content in soil because total vehicles passing through the road were 12,000 vehicles per day (from 07.00 am to 05.00 pm).

Assuming that 75% of total vehicles (12,000 units per day) passing through the study site was gasoline-based vehicles and the rest (25%) was diesel-based vehicles, daily emission of Pb was 731.14 g d⁻¹ (626.85 g d⁻¹ from gasoline-based vehicles and 104.29 g d⁻¹ from diesel-based vehicles, respectively) (Tabel 1).

Heavy metal concentrations in roadside soils could also be impacted by longer transportation period (e.g. 20 years), since soil tended to accumulate and persist metals on a relatively long-term period (Garcia and Millán 1994; Kelly *et al.* 1996). The road in the study site has been operated for almost 20 years.

Therefore, combination of long-term period and heavy traffic density are important contributing factors to the accumulation of Pb in the soil of the study site. More importantly that the results of the current study confirm that there is a gradual build up of Pb and probably the other heavy metals in the soil at the study site. Therefore, a constant monitoring of the levels of these metals is required to safeguard farmlands, fish pound and water bodies around this area.

Pb Accumulation in Oil Palm Leaves

Bioavailability of Pb at the study site was investigated by measuring Pb content in leaves of oil palm as a test plant. Similar to Pb content in the soil at the study site, Pb content in leaves of oil palm planted along the roadside site was 11.05 mg kg⁻¹. This level was 5.5 times higher than the safe level of Pb in plant (2.00 mg kg⁻¹) as regulated by Balai Penelitian Tanah Indonesia (2002). Although current data was too small to perform statistical calculations on the dependencies of the two parameters, current result confirms that the high level of Pb in test plant is proportional to the content of Pb in soil.

The transfer of Pb from soils to plants is dependent on three factors; total amount of potentially available Pb (quantity factor), the activity as well as the ionic ratios of Pb in the soil solution (intensity factor), and the rate of element transfer from solid to liquid phases and to plant roots (Brummer *et al.* 1986). These three factors were applicable to the condition of soils at the study site. As explained above that the content of Pb in soils at the study site was 1.5 time higher than the safe level. Much higher level of Pb in the leaves of the palm oil was due to the fact that the absorption of Pb by the test plant occurred not only by roots but also by stomata because diameter of elemental Pb (< 4µm) is smaller than the size of the stomata (length 2µm and width 7 µm). More importantly that there is a possibility that high level of Pb both in soils and leaves of oil palm at the study site can also be detected in oil palm fruits. However, this hypothesis still requires further investigation.

Table 1. Daily number of vehicles and estimated Pb emmision.

	Gasoline-based *	Diesel-based**	Total
Number of vehicles (unit day ⁻¹)	9,000	3,000	12,000
Estimated Pb emmision (g day ⁻¹)	626.85	104.29	731.14

Note: *Assumed to be 75% of the total number of vehicles, ** Assumed to be 25% of the total number of vehicles.

Table 2. Effect of SOM amendment on soluble Pb in soils.

SOM (Mg ha ⁻¹)	Soluble Pb at			
	2 WAI *	Percentage of Reduction	4 WAI	Percentage of Reduction
	--- mg kg ⁻¹ ---	--- % ---	--- mg kg ⁻¹ ---	----- % -----
Control	11.15 ± 2.31 b	-44.28 **	7.43 ± 1.26 b	-62.87** (-33.36****)
30	6.82 ± 1.89 a	-38.83 ***	3.77 ± 0.74 a	-49.26 *** (-44.72****)
60	7.19 ± 2.67a	-35.52 ***	5.34 ± 2.46 ab	-28.13 *** (-25.73****)
90	10.19 ± 2.54b	-8.61 ***	6.79 ± 0.35 b	-8.61 ** (-33.36****)
LSD _{0.05}	1.67		2.86	

Note: * WAI = week after incubation, ** as compared with inherent content of Pb (20.01 mg kg⁻¹), *** as compared with control, **** as compared with 2 WAI, values are presented as mean ± standard deviation (n=6), values with different letters along a vertical column are statistically different.

Organic Matter to Reduce Pb Bioavailability

Much higher level of Pb in the soils and the leaves of the palm oil at the study site indicate high bioavailability of Pb in the soil. Hence, it is necessary to reduce plant-available Pb concentration in the soils, one of which is through site stabilization. The aim of site stabilization is to rapidly reduce Pb availability to plants. This can be achieved by adding various agents to the soil, one of which is OM. Changes in Pb content with SOM amendment and changes in Pb with incubation are described in 7s section.

SOM amendment through the addition of compost significantly ($P < 0.01$) affected soluble Pb content in the soils of current study. Although the effects of the addition of OM to heavy metals are frequently varied (Gorlach and Gambus 1992; Narwal and Singh 1998; Schmidt 2003), the addition of organic matter at 30, 60 and 90 Mg ha⁻¹ significantly reduced soluble Pb in soils as compared with the control in the current study (Table 2). The disagreed findings between the current study and the previous studies are reasonable because OM is a quite heterogeneous material that can concurrently exert mobilizing and stabilizing effects. Small structural and chemical differences can favor one or the other effect. On the one hand, organic acids released through the decomposition of both the natural SOM and the added OM reduce soil pH, which then increases the concentration of soluble heavy metals in soils. On the other hand, it increases the CEC of soil, providing sorption sites and reducing heavy metals mobility.

In relation to the results obtained in the current study, both argument described above are applicable.

The fact that the addition of OM significantly reduced the soluble Pb in the soils of the current study (Table 2) implies that the addition of OM provides sorption sites and reduces Pb mobility. The addition of OM at 30 Mg ha⁻¹ to correct the level of SOC from very low to low was sufficient to significantly reduce Pb solubility in the soils of current study. It resulted in a decrease of soluble Pb with a magnitude of 39% at 2 weeks after planting (WAI) and 49% at 4 weeks after planting (WAI) as compared with the control soils (Table 2).

Sorption reactions involving SOM are particularly important with respect to ameliorating the effect of metal contaminants. As widely known that metals including Pb have a high affinity for humic acids, organo clays, and oxides coated with organic matter (Berti *et al.* 1996; Elliot *et al.* 1986). The most common form of interaction between SOM and positively charged ions is via cation exchange reactions (e.g. between negatively charged carboxyl groups and positively charged cations) and is associated with proton replacement from acid functional groups (ligands).

However, further increases in OM rate to 60 and 90 Mg ha⁻¹ were followed by increases in Pb level in the soils (Table 2). An increase in OM application to 60 Mg ha⁻¹ triggered a small but not significant increase in soluble Pb as compared with soluble Pb in soils receiving 30 Mg ha⁻¹ of OM. Nevertheless, the application of OM up to 90 Mg ha⁻¹ significantly increased the soluble Pb as compared with those in the soil receiving 30 Mg ha⁻¹ of OM (Table 2). It indicates that the effort to increase SOC of the soils

from very low to moderate and high in the current study triggered the increases in the soluble Pb in the soils through mechanisms described before. Organic acids released through the decomposition of SOM increased soil acidity or at least maintained soil acidity of the current study at pH of around 4.00 during the incubation, increasing the concentration of soluble Pb in soils. As explained by Andriano *et al.* (2002) that the effect of pH value < 6 in increasing metal ion activities in soil can be attributed to the decrease in pH-dependent surface charge and chelating by organics of metal hydroxide. Besides continuous loading of Pb from vehicle emission, the increases in solubility of Pb as a results of acidification by organic acids possibly facilitates side effects of the higher reported accumulation of Pb in the leaves of oil palm at the study side. In spite of the increases, the level of soluble Pb in the soils receiving 60 and 90 Mg ha⁻¹ of OM was still much below the safe level of Pb in soil (12.75 mg kg⁻¹) as regulated by Balai Penelitian Tanah Indonesia (2002).

Although Ma and Dong (2004) described that reported data on the solubility of trace metals in relation to incubation time were often contradictory, current study showed that soluble Pb in soils decreased with incubation time (Table 2). The levels of soluble Pb at 4 WAI at all OM levels decreased with magnitude of ranging from 26% to 33% as compared with those at 2 WAI (Table 2). Such reduction occurred not only due to the discontinuation of Pb loading because the soils were incubated in the laboratory but more importantly also due to the formation of organic acids-Pb complexes as a result of the decomposition of soil SOM.

Significance of SOM in reducing the soluble Pb is also evident in control soil. Although the initial content of SOC of the soils used in the current study was very low (5.70 g kg⁻¹), incubation under control environment had created a favorable condition for SOM decomposition. The decreases in the soluble Pb in the control soil from 20.01 mg kg⁻¹ at pre-incubation time to 11.15 mg kg⁻¹ at 2 WAI and 7.43 mg kg⁻¹ at 4 WAI reflected inherent self-remediation capability of the soils. In spite of the increases, the level of soluble Pb in the soils receiving 60 and 90 Mg ha⁻¹ of OM was still much below the safe level of Pb in soil.

CONCLUSIONS

Based on the results obtained it can be concluded that both roadside soil and oil palm at the study were contaminated by Pb. Adding OM significantly (P<0.01) reduced soluble Pb in the soils. The addition of OM at 30 Mg ha⁻¹ to correct the level of SOC from very low to low was sufficient to significantly reduce the soluble Pb in the soils of current study. However, the application of 60 Mg ha⁻¹ of OM triggered the increases in Pb solubility in the soils and further increases in OM application to 60 Mg ha⁻¹ resulted in significant increases in soluble Pb as compared with that in the soil receiving 30 Mg of OM ha⁻¹.

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