

Lead concentration and factors affecting its fractions in soil around a selected industrial estate

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Abstract

Related activities conducted in lead industries could bring about various negative impact on the environment due to the contamination emitted by the waste, which can transfer to environmental compartments due to the fate of the substance in soil. This study conducted a field survey and laboratory analysis of soil samples collected from an industrial estate zone. It was found that the maximum level of total lead was 3.11 mg/kg dry weight. Lead fractions for 5 forms were analyzed using a sequential extraction method. The results showed that the largest contribution of lead contamination was presented in bound to organic matter fraction (F4) while the smallest contribution was exchangeable fraction (F1) and bound to carbonate fraction (F2). It was found that pH tends to be an important factor on the availability of lead fractions. Additionally, it was observed that bioavailable fractions of lead tend to increase at lower level of pH. In other words, in highly acidic conditions, bioavailable fractions of lead presented the largest fraction whereas the fractions tend to decrease in lower acidic and neutral conditions.

Keywords: Lead fraction; Sequential Extraction; Exchangeable fraction; Soil pH.

1. Introduction

Lead contamination in soil across industrial zone could occur due to activities such as mining industrial waste dumping, and the use of lead as a raw material in an industrial process (e.g., that in a battery factory and a recycled battery factory). Lead contamination in the environment may be caused by direct transmission in soil or lead fume dispersion in ambient air. In the latter form, lead fume can be possibly washed and removed by the precipitation from the ambient air. However, the washout can be transferred to soil in the catchment zone. In this case, the study of lead fraction and factors related to its transmission should be conducted so that a solution to lead contamination in soil can be suggested.

There are several studies on lead contamination in environment. One example is the study of lead contamination in sediment of Klong Bangyai, Phuket [1]. However, most of the studies have focused on total lead contamination in the environment without investigating lead fractions in the environment. Meanwhile, it has been proven that lead can be transformed in environment by the nature of its form or fraction. The study by Supamas [2] reported that, if heavy metal is accumulated in soil or sediment for a long time, it can bind with neighboring stable elements to form silicate compound and persist in the environment. Furthermore, the study of Tang, et al. [3] found that long-term lead-spiked soil may have cumulative lead

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contamination from residue fraction of lead while lead tended to have high reactivity to increase the rate of transmission under a lower pH (in acidic soil). Moreover, lead in mobile form (such as methyl lead) can bind with plants and other living organisms and then precipitate on a deeper soil layer otherwise become widely transmitted in the environment, a process that can contaminate the food chain and thus cause harm to human [4][5]. Therefore, researchers in this field should study the fractions of lead in contaminated soil and the factors controlling lead remobilization in soil. The findings from lead fraction remobilization studies can be useful in controlling lead contamination in soil as well as reducing hazards of lead accumulation in living organisms.

2. Methodology

2.1 Soil sampling and analysis

Soil sampling was conducted across a 5 km radius area surrounding industrial estate. There were 10 sampling stations, and each sampling point was collected at the depth of 10-30 cm from soil surface [6] with sample weight of 500 g per station. The samples were analyzed for soil properties in terms of size distribution analyses, cation exchange capacity (CEC), pH, and metal content.

2.2 Total lead and lead fractions in soil analysis

The procedure involved sieving soil samples to achieve a fine size of less than 2 mm. Then, the samples were dried at 80-100 °C until the sample weights were stable (for approx. 2-3 day). Each sample weighting 1 gram was analyzed for total lead concentration with acid digestion by using concentrated nitric acid (HNO_3). The digested samples were measured for lead concentrations using Thermo Fisher Scientific iCE 3000 series AA Atomic absorption (AA) spectrophotometer, with a limit of 0.03 µg/L. Quality control for each batch of samples processed was conducted by providing a method blank throughout the entire sample preparation and analytical process [7], five

lead fractions were analyzed by using a sequential extraction method [8]. The five fractions are exchangeable (F1), bound to carbonate (F2), bound to Fe/Mn oxide (F3), bound to organic matter (F4), and residual (F5) fractions.

2.3 Factors controlling lead form in soil

Our methodology for studying factors controlling lead forms in soil was to take soil samples from the industrial estate's surroundings, as previously described (step 2.1), and to adjust the pH by using 1 M HCl (10-200 mL/kg soil) and CaCO_3 (0.25-10 g/kg soil) [9][10]. Five ranges of pH adjustment were used: strongly acidic (pH 2-4), weakly acidic (pH 5-6.5), neutral (pH 7.0-7.5), weakly basic (pH 8-9), and strongly basic (pH > 9). Then, we left the treated samples for 1 month then added lead nitrate ($\text{Pb}(\text{NO}_3)_2$) in the concentrations of 100 and 1,000 mg/kg, respectively. Later on, we left the lead spiked samples for 1 month before analyzing lead fractions using the procedures described in step 2.2. Triplicated analysis of the samples was conducted.

3. Results

3.1 Soil composition and other metal content

From soil composition analysis, the bulk of the sample was sand (62.38%) while the rest comprises loam soil (28.28%), and clay (9.34%), respectively. The Cation Exchange Capacity (CEC) was 8.80 cmol/kg, and the soil samples were slightly weakly acidic (pH around 5.21-6.70). In term of heavy metal content in soil samples, we found that the concentration of copper (Cu) was approx. 4.02-186.41 mg/kg dry weight, that of chromium (Cr) was approx. 1.36-14.05 mg/kg dry weight, that of iron (Fe) was in the range of 2,139.67-4,824.60 mg/kg dry weight, and that of manganese (Mn) was approx. 22.55-324.20 mg/kg dry weight. Most of the heavy metals (Cr, Fe, and Mn) met soil quality standard for residential and agricultural area

according to the Notification of National Environmental Board Issued 25th B.E. 2547. Meanwhile, the concentration of cadmium (Cd), nikle (Ni), and zinc (Zn) were found to

be lower than the detectable limit. The results of soil composition analysis and heavy metal concentrations are shown in Table 1.

Table1. Soil properties.

Soil property and composition	Unit	Result	Standard*
Soil composition (Particle size distribution)	Percentage		
- Sand		62.38	
- Silt		28.28	
- Clay		9.34	
Organic matter	Percentage	2.08	
CEC	cmol/kg	8.80	
pH	-	5.21-6.70	
Cd	mg/kg	ND	37
Cu	mg/kg	4.02-186.41	-
Cr	mg/kg	1.36-14.05	300
Fe	mg/kg	1,976.32-4,824.60	-
Mn	mg/kg	22.55-324.20	1,800
Ni	mg/kg	ND	1,600
Zn	mg/kg	ND	-

Remark * soil quality standard for residential and agriculture B.E. 2547 according to the Notification of National Environmental Board Issued on 25th B.E. 2547

- No limitation ND : not-detectable

3.2 Total lead and lead fractions in soil

It was found that 3 stations had the highest total lead which as 3.11 mg/kg dry weight. The 3 stations are located in an area adjacent to a drainage canal of wastewater treatment plant in the industrial estate. This may cause lead contamination due to residual lead in wastewater. However, comparing of lead contamination in soil samples to the soil quality standard for residential and agricultural area according to the Notification of the National Environmental Board Issued on 25th B.E. 2547 under the environmental quality protection and promotion Act. B.E. 2535 on soil quality threshold we found that levels of lead in the soil samples did not exceed the national soil quality standard (standard of Pb in soil samples was lower than 400 mg/kg dry weight). The analysis of 5 lead

fractions by using sequential extraction found that most of the lead contamination was presented in bound to organic matter fraction (F4) followed by the form of residual fraction (F5), as shown in Table 2. The bound to organic matter fraction was the most important fraction because this fraction causes organometallic complex to accumulate and to be released in the environment. For residual fraction, it is present in the form of primary mineral crystal, which is low mobility. Therefore, this fraction tends to cause less impact to the environment. While, some fractions of lead founded in exchangeable fraction (F1) and bound to carbonate fraction (F2). These fractions are easily transformed and absorbed through a biological process, and they could accumulate in living organisms or the environment [4],[5].

Table 2. Amounts and fractions of lead in soil.

Sampling station	pH*	Total lead (mg/kg dry weight)	Lead fraction (mg/kg dry weight)				
			F1	F2	F3	F4	F5
Station 1	6.70	1.709±0.101	0.028±0.005	0.026±0.005	0.172±0.014	0.902±0.211	0.500±0.035
Station 2	6.45	3.106±0.097	0.029±0.003	0.116±0.014	0.678±0.022	1.028±0.005	1.023±0.241
Station 3	5.54	3.109±0.032	0.025±0.002	0.109±0.024	0.266±0.019	1.053±0.016	1.277±0.133
Station 4	5.87	1.260±0.034	0.019±0.000	0.033±0.001	0.116±0.008	1.040±0.012	0.339±0.005
Station 5	6.25	1.716±0.111	0.023±0.004	0.028±0.001	0.287±0.027	1.064±0.019	0.662±0.038
Station 6	5.45	1.808±0.183	0.028±0.016	0.014±0.001	0.174±0.053	1.053±0.021	0.555±0.097
Station 7	5.49	1.422±0.002	0.030±0.023	0.026±0.003	0.219±0.006	0.889±0.172	0.614±0.160
Station 8	5.21	1.210±0.170	0.032±0.018	0.016±0.001	0.138±0.009	0.853±0.003	0.586±0.058
Station 9	5.53	3.244±0.217	0.055±0.031	0.057±0.002	0.805±0.030	1.478±0.001	0.608±0.030
Station 10	5.40	1.741±0.304	0.046±0.012	0.020±0.000	0.111±0.002	1.041±0.003	0.600±0.061

Remark: F1 (Exchangeable fraction), F2 (Bound to Carbonate fraction), F3 (Bound to Fe/Mn oxide fraction), F4 (Bound to Organic matter fraction), F5 (Residual fraction)

* pH of original soil sample

3.3 Factors affecting lead fraction in soil

3.3.1 pH

Soil samples that had been treated with 5 ranges of pH adjustment, namely were left for 1 month prior to the measurement of lead fractions. The results of lead fraction detected as percentages of lead fractions were varied as shown in Figure 1. Lead fraction analysis indicated that a lower pH condition could increase exchangeable fraction which could easily be transformed and absorbed through a biological process, but F2 and F3 were found at high levels in the acidic condition, which might cause the Fe/Mn or carbonate content to be soluble, possibly leading to lead fraction. In addition, the level of lead in each fraction was found not to vary significantly. This observation suggests that the original samples may already contain low level of Pb. It appeared that lead could easily dissolve in acidic water than in base water. Therefore, increasing the pH of water tends to make lead precipitate in the form of hydroxide at a pH higher than 6.5 [11].

3.3.2 Amount of lead in soil

Soil samples were spiked with lead at 100 and 1,000 mg/kg, and their pH was adjusted into 5 ranges (strongly acidic, weakly acidic, neutral, weakly basic, and

strongly basic) prior being left for 1 month. It was found that the percentages of lead fraction were different as shown in Figure 2 and Figure 3. The exchangeable fraction (F1) was the largest contribution in strongly acidic environment (compared to other fractions). The lead contamination soil would be harmful to living organisms through an absorption process if the environment is strongly acidic. In contrast, the amount of lead may not affect lead accumulation in living organisms if the environment is neutral or basic. A crucial point of this finding is supported by lead remobilization trends as shown in Figure 3. The experiment indicated that if the level of lead spike was increased 10 folds, however, the lead remobilization in strongly acidic condition was still not different. Therefore, pH might be more important to control lead remobilization in soil because most trace metals become more mobile in increasingly acidic conditions [12], and pH is the principal factor governing concentrations of solubles and plant available metals [15]. In addition, it is noted that there are many factors that relate to metal fractions, especially physico-chemical properties in soil such as redox potential and organic matter content [13], [14].

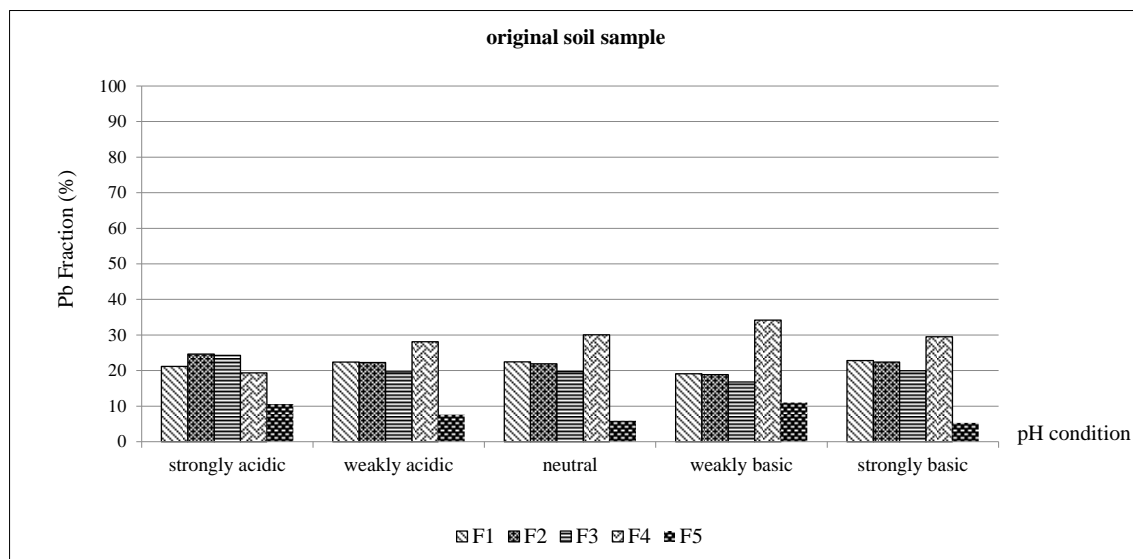


Fig.1. Percentages of lead fractions in soil after pH adjustment without lead-spikes.

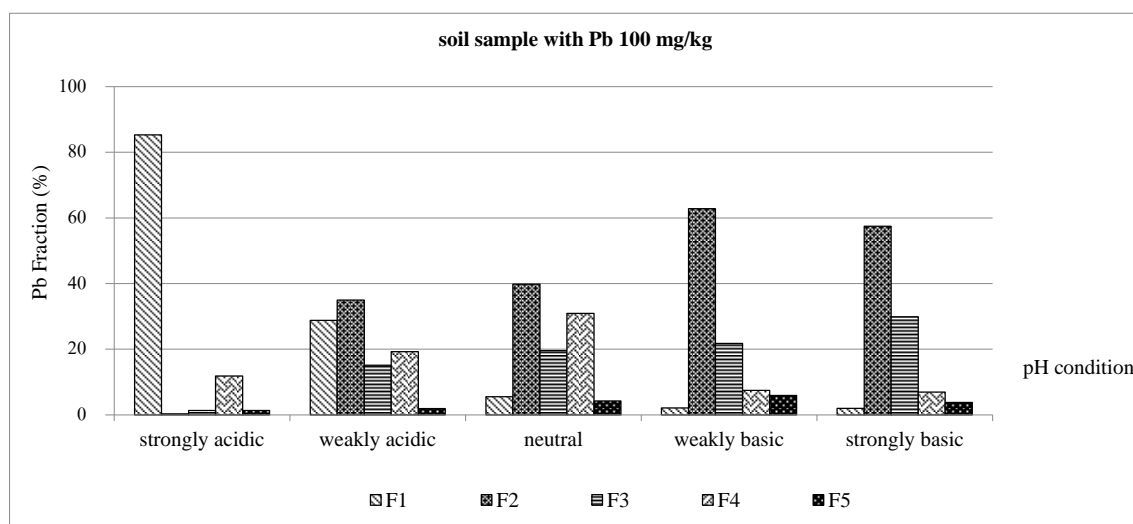


Fig.2. Percentages of lead fractions in soil after pH adjustment with 100 mg/kg lead-spikes.

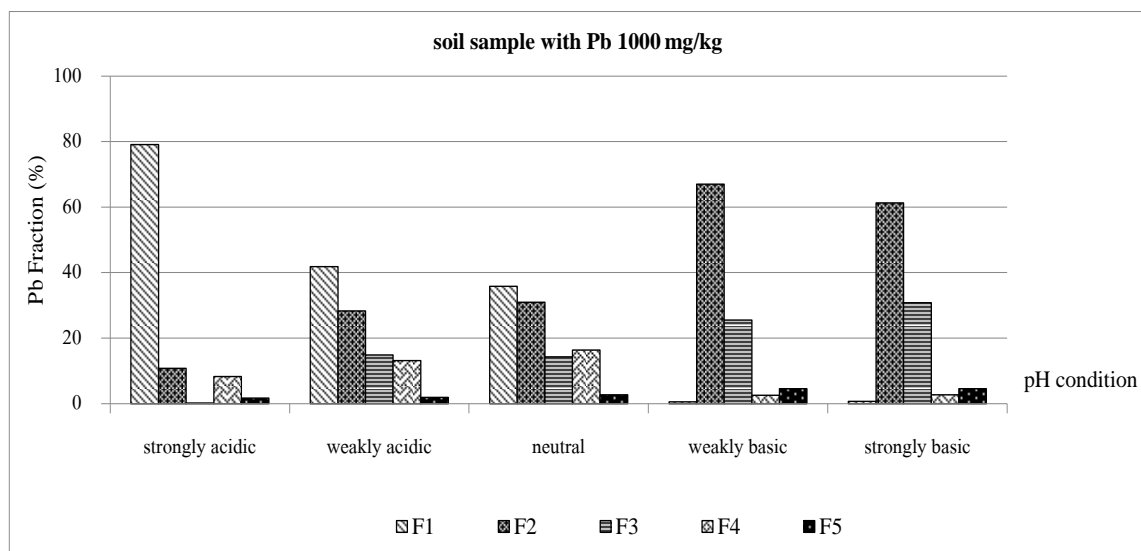


Fig.3. Percentages of lead fractions in soil after pH adjustment with 1,000 mg/kg lead-spike

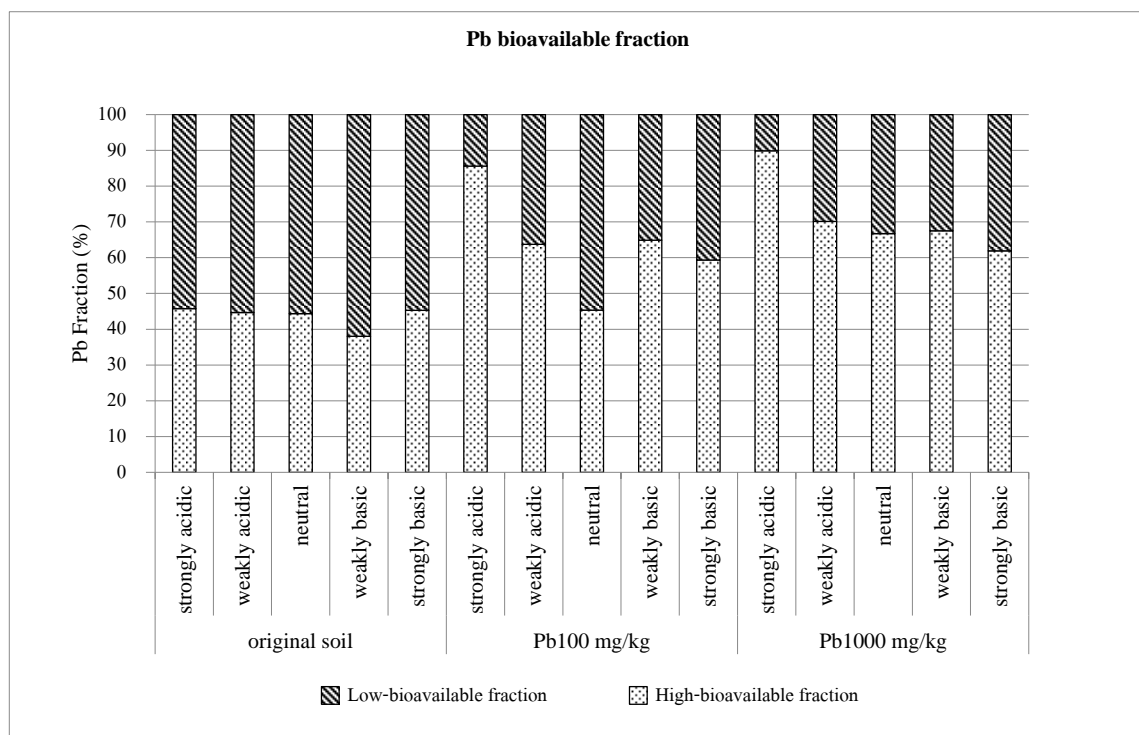


Fig.4. Percentages of lead fractions in soil that are easily absorbable by living organisms and that are difficult to be absorbed by living organisms under treated soil with pH adjustment.

According to Figure 4, if 5 fractions of Lead were considered as high bioavailable fraction, which is exchangeable fraction (F1), bound to carbonate (F2) and low bioavailable fraction [15], the remaining fractions (F3, F4 and F5) yielded the results that the total lead in the original soil samples was around 1.2-3.2 mg/kg dry weight (Table 2), lead fraction in original soil is mostly presented in bound to organic matter fraction (F4). This fraction (F4) is difficult to be absorbed by living organisms. After pH adjustment, lead transformation did not change significantly in each pH level. However, the lead-spiked and pH adjusted soil showed different fractions of those 2 forms (bioavailable and low bioavailable fractions), especially in the strongly acidic soil. Lead contamination in strongly acidic soil increased by 90% for bioavailable fraction as shown in Figures 1 to 3 which were indicated that most of the bioavailable fractions were presented in exchangeable form (F1). This observation revealed that lead contamination in soil is easy to be absorbed in plants or living organisms if the contaminated soil is under a strongly acidic condition. However, contaminated soil under a strongly acidic condition is not suitable for growing many plants.

4. Conclusion

Total lead and lead fractions in soil samples around an industrial estate area were extracted by using sequential extraction for 5 fractions. It was found that the majority fraction contaminated in soil was the bound to organic matter (F4). The pH was an important factor affecting the availability of lead fraction in soil.

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6. References

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