INTRODUCTION

Lead (Pb) pollution in the soil is a serious global problem due to the potential risk of contamination on food chain and groundwater (Lim et al., 2013). Pb concentration based on toxicity characteristic leaching procedure (TCLP) test should be less than nonhazardous regulatory limit of 5 ppm (Huang, Su, Rizwan, Zhu, & Hu, 2016). Although Indonesia does not have soil quality standards, the government has released Government Regulation No. 101-2014 about the management of hazardous and toxic waste (B3). Economic activities such as mineral production and processing, agriculture, and industries have impacted in accumulation of Pb in soil. Heavy metal cannot be decomposed by microorganisms and persists for hundreds of years in soils environment (Mahar, Wang, Li, & Zhang, 2015) so that immediate and effective remediation is necessary (Khalid et al., 2017).

Traditional remediation methods for contaminated soil such as solidification, soil washing, and soil replacement are considered costly and most of cases, they were incompatible with agricultural perspective, especially in developing countries (Khalid et al., 2017). The sustainable method that is considered cheaper, able to upgrade soil fertility, and gives economic benefits is more preferable (O’Connor et al., 2018). Phytoextraction and immobilization are considered to be the most effective in situ remediation methods, yet contradictory in resolving lead-contaminated farmland soil in developing countries. Phytoextraction relies on improving the ability of
plants in absorbing Pb so that efforts in enhancing Pb solubility in soil solutions are essential. On the other hands, immobilization (chemical fixation) is focused to diminish lead mobility in contaminated soil to prevent plant uptake and groundwater contamination.

Although effective, ligand assisted phytoextraction with synthesized chelators such as ethylene diaminetetra acetic acid (EDTA) is not favorable because of its high cost, persistence, phytotoxicity, and metal-chelates leaching risk (Kim & Lee, 2010; Krueger et al., 2013; Wuana & Okieimen, 2011). As a result, cheaper and more biodegradable chelator such as citric acid, succinic acid, glutaric acid, and amino acids are employed in the phytoremediation of heavy metal contaminated soil (Hou et al., 2015; Jiang, Li, Han, Yang, & He, 2012; Mao et al., 2015). These acids are not as effective as EDTA but more environmental friendly because they naturally exist in soil as root exudates, and also as microbial and decomposer products. Organic acids are also contained in many naturals produces such as lemon, tamarind, tomato, orange and therefore can be exploited as natural, low-cost amendments.

Chemical stabilization/immobilization of lead (metals) with low-cost materials is another side of soil remediation. Biochar, a carbon-rich porous material made from the heating of agriculture by products, has been widely exploited because of its basic and excellent absorptive properties for various soil contaminants (Ahmad et al., 2017; Puga, Abreu, Melo, & Beesley, 2015). Digestate or bioslurry, an alkaline and nutrient-rich biogas slurry has been applied to mitigate heavy metals impact from contaminated soils. The reduction of Pb levels in soil extracts and test plant seeds is proved by the application of these substance (García-Sánchez, García-Romera, Száková, Kaplan, & Tlustoš, 2015). Bioslurry also reduces the toxicity of Hg on soil microorganisms which is proved by the increase of weight and activity of microorganism biomass in amended soil (García-Sánchez, Klouza, Holečková, Tlustoš, & Száková, 2016). Utilization of natural lime-based materials showed that the addition of 1% eggshell and oyster shell can reduce 26.60% Pb in TCLP leachate (Lim et al., 2013). Similarly, incubation soil with 5% of calcined cockle shells showed the increase of pH up to 12.2 and reduction of 85% lead in 0.1 M HCl leachate (Islam et al., 2017).

Sequential extraction procedure (SEP) is a widely accepted method in assessing bioavailability and ecological risks of heavy metals in soil or sediment. A simple, three steps, SEP method suggested by the Community Bureau of Reference (BCR) was performed in this study (Lu et al., 2017; Nemati, Bakar, Abas, & Sobhanzadeh, 2011). The SEP distinguishes the most soluble metal fraction from other less soluble fractions by several extraction steps. BCR is the simplest SEP which classifies metal in soils into four geochemical fractions/phases. The BCR fractions are acid extractable metals (F1), associated with Fe/Mn oxides (F2), oxidizable or organic fraction (F3), and the residual fraction (F4). The most soluble metal fraction (F1) is crucial, since it determines the contamination degree and ecological risks of the metal contaminated soil (Nemati, Bakar, Abas, & Sobhanzadeh, 2011; Tytla, 2019; Yang et al., 2018; Zhang et al., 2015).

The chemical composition, surface properties, and acidity of the amendment are often contributed to the ability of the metal absorption and immobilization capacity (Suárez-Hernández, Ardila-A., & Barrera-Zapata, 2017; Zhang et al., 2013). Even so, there is still limited information on the characteristics of the amendment which influence the efficacy of Pb [im]mobilization due to Pb level and soil properties (Nartey & Zhao, 2014). The properties of amendment material were characterized by electron microscope (SEM) scanning, fourier transform infrared spectroscopy (FTIR), and x-ray fluorescence spectrocope (XRF). Contaminated soil was incubated with amendment materials and underwent three-step extraction method according to the modified Community Bureau of Reference (BCR) protocol. Quantification of Pb levels in BCR extracts was determined using atomic absorption spectrophotometer (AAS). The main goals of this study are (i) to explore the effectiveness of low-cost amendments in altering solubility of Pb in soil, (ii) assess the potential ecological risks of Pb after [im] mobilization, and (iii) evaluate the most important properties of amendment in affecting Pb mobility in soil.

MATERIALS AND METHODS

All material used to prepare soil amendments was collected from traditional farmers around Surakarta, Central Java, Indonesia. Lead...
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Contaminated soil due to battery recycling was taken from Cinangka in Bogor, West Java, Indonesia. Research work was conducted from March to May 2019. As many as 13 amendments were prepared including chars, bioslurry, rice husk, gypsum waste, lime-based materials, and natural acidic crops. Biochars were produced from chicken bone (CB), chicken manure (CM), and farmyard (goat) manure (FM) by heating in a limited oxygen environment. The process is conducted at 450°C for about 90 minutes in an electric laboratory muffle furnace. Lime-based materials were made from eggshell and snail shells by calcination at 860°C. Selected solid amendments were grounded in a wooden mortar and pestle, sieved through a 0.5 mm plastic filter, and characterized. The FTIR and XRF were employed to identify functional groups and analyze the elemental composition of solid amendments. Before the amendment process, the surface morphology and pore properties of the solids were examined with SEM. Amended soil incubation was conducted according to Huang, Su, Rizwan, Zhu, & Hu (2016). Organic content in the soil was analyzed with loss on ignition (LOI) and pH measurement (in water 1:2.5) was also carried out. The soil was mixed homogenously with 10% amendments and throughout the incubation period, soil was examined and moistened regularly.

The SEP method of modified BCR (Lu et al., 2017; Nemati, Bakar, Abas, & Sobhanzadeh, 2011) was conducted to assess the efficiency of immobilization and distribution of Pb geochemical fractions. This method differentiates soil-Pb into 4 fractions (F1-F4) in the order of declining solubility. Extractant solutions used for three-stage SEP were: 0.11 M acetic acid; 0.5 M NH₄OH.HCl pH 1.5; and twice of 8.8 M H₂O₂ then 1 M NH₄OAc at pH 2. In the last, aqua regia was employed to extract Pb form residual fraction (Lim et al., 2013; Mahar, Wang, Li, & Zhang, 2015; Rodríguez, Gómez, Sánchez, & Alonso-Azcárate, 201). Solid separation and washing were enhanced by centrifugation at 2500 rpm. Lead content in the solutions of each fraction was measured with atomic absorption spectrophotometer (AAS) Shimadzu AA 6500.

The values of individual contamination factor (CF) and Risk Assessment Code (RAC) were dedicated to assess the ecological impact of the amendment. CF was the measure of retention time of metal before reaching the environment and calculated as the comparison of non-residual fractions and the most stable fraction (F4) (Nemati, Bakar, Abas, & Sobhanzadeh, 2011; Tytła, 2019) and also referred as the ratio of secondary to primary phase, RSP (Yang, Hu, Yu, He, & Lin, 2016). The CF or RSP values can distinguish anthropogenic sources of heavy metal from natural ones and express the pollution degree of the studied soil. The efficiency of metal immobilization (E) was exploited to assess the efficacy of the immobilization methods (Wuana & Okieimen, 2011) according to the following formula.

\[ E = 100 \left( \frac{M_o - M_e}{M_o} \right) \]

Where: \( M_o = F1 \) concentration of Pb in control soil, \( M_e = F1 \) concentration of Pb in amended soil.

Simple linear regression analysis is applied to determine the relationship between the amendment properties and the efficiency of immobilization and to evaluate the important properties. Due to the limitation of this research, data pH and LOI is available for 14 amendments whereas elemental composition is available only for 5 amendments.

RESULTS AND DISCUSSION

Characteristics of Soil and Amendments

The important physical-chemical properties of soil samples are summarized in Table 1. The studied soil was loam according to USDA, quite acidic, and the pH was 4.29 ppm and considered high for arable soil compared to those in other countries (Yang et al., 2018). In China, the maximum Pb level in grade C soil quality standard is 500 ppm. This standard includes upper Pb limit level to guarantee the safety of plant growth and agricultural production. The Pb level of the studied site, thus exceeded 8 times the grade C of China standard and 11 times the grade C of China standard and 11 times the US EPA standard (400 ppm). Similarly, compared to health investigation level (HIL) A and C standard in Australia for residential with garden/accessible soil and open space (300 and 600 ppm), the level is about 15 times and 8 times respectively. According to Government Regulation of Indonesia (PP No. 101-2014), the soil has to be managed as hazardous and toxic waste category 2. Other metal contents of the studied site such as Fe and Mn were also high. Fe and Mn contents were observed 16.15 and 0.47, respectively and considered abnormal for uncontaminated soil. Thus, this soil might be suffered from multi-metal contamination.
The spectra of FTIR (Fig. 1) showed that biochar and bioslurry possessed similar spectra indicating the presence of O-H, C=O, carbonate, phosphate, C=C, and C-O-C groups in the chars and bioslurry (Cao, Ma, Liang, Gao, & Harris, 2011). The peak of the hydroxyl group appeared as broadband near 3500/cm affiliated to OH bond stretching vibration indicated strong hydrogen-bonding. Aromatic C=C and C=O bands were observed at 1613/cm (Suárez-Hernández, Ardila-A., & Barrera-Zapata, 2017). The bands showed near 1430/cm and 1040/cm suggested that biochar also possessed an abundance of CO$_3$$^{2-}$ and PO$_4$$^{3-}$ groups. The hydroxides, carbonates, and phosphates can precipitate Pb and act as effective sorption sites in the soil. Carbonate and phosphate compounds of Pb mostly form precipitates. For example, the formation of PbCO$_3$, Pb$_2$(CO$_3$)$_2$(OH)$_2$, and hydroxyphosphomorphite, Pb$_{10}$(PO$_4$)$_6$(OH)$_2$, $K_{sp} = \sim 10^{-70}$ will significantly lower lead level in soil solution.

Photo microscopic of SEM in Fig. 2 showed a porous structure of the char having pores dimension between 1.95 and 3.76 µm. Compared to other biochars derived from hardwood feedstock (Acacia mangium, Eucalyptus grandis, Gmelina arborea), these evaluated biochars (CB, CM, and FM) possessed less regular pore dimension and shape (Suárez-Hernández, Ardila-A., & Barrera-Zapata, 2017). It is caused by the nature of the raw material to produce biochar. Biochar materials used in this study (chicken manure, goat manure) are derived from a biomass mixture with structure irregularities. The lower temperature (450°C) and particularly short pyrolysis time (90 minutes) may cause an incomplete carbonization process. Higher pyrolysis temperature will result in better pore size and more alkaline biochar. The irregular pore properties resulted from lower temperature pyrolysis is compensated with higher available P, K, and Ca amendment which is important in effective Pb immobilization (Bolan et al., 2014; Rajapaksha et al., 2015).

Table 1. The basics important properties of contaminated farmland soil

| pH | % Particle size (mm) | Texture | Pb content (mg/kg) | Fe (%) | Mn (%) | EC (mS/cm) | CEC (me/100 g) | LOI (%) |
|----|---------------------|---------|-------------------|-------|--------|------------|----------------|--------|
| 5.5| 44.83               | Loam    | 4296              | 16.15 | 0.47   | 0.16       | 14             | 8.78   |

Fig. 1. The infrared spectra of chicken manure derived biochar (CM) and solid biogas slurry (BS)
Elemental composition data from XRF (Table 2) showed that solid amendments possessed significantly high mineral content. Essential minerals content in amendment material can ameliorate soil and provide essential nutrients for plants and microorganisms. The CB and CM showed high calcium and phosphorus (Ca and P) contents, which were 34.16, 10.4% for CB, and 17.91, 7.08% for CM. The highest K content as much as 18.86% was possessed by CM, biochar made from chicken manure. The FTIR spectra data for CB and CM at 1046 /cm indicated phosphate functional group and referred to the high content of P in the biochars. The main phosphorus compounds in the chicken litter were orthophosphate, orthophosphate esters, and phytate (Uchimiya et al., 2010).
Efficacy of Low-cost Amendments in [Im] mobilization of Soil-Pb

The amendments had changed soil properties, Pb-fraction redistribution, the efficiency of immobilization, and ecological risk (Table 3). Amendments altered soil pH (5.0 at 2.9 – 12.6), F1-lead (17.1 at 1.54 – 32.7 ppm), and induced immobilization efficiency (% E) between -91.2 (highly mobilize) and 91.0 (highly immobilize). Reducible fraction (F2) and the most stable residual fraction (F4) were also changed significantly but organic fraction Pb (F3) was relatively constant in amended soil. Reducible fraction F2 was changed between -45.1 and 1.3 ppm (mostly decreased) but residual fraction (F4) was changed between -2.5 and 52.8 ppm (mostly increased).

Seven amendments mobilized Pb in the order of CA > BO > SS > ES > TA > LJ > TO whereas the immobilization order was CB > CM > FM > GW > BS > RH. Remediation of Pb-polluted soil with CA, BO, TA, and LJ can mobilize Pb to be uptaken by plant in a green, sustainable chemical assisted phytoextraction. CA, BO, TA, TO, and LJ immobilized Pb because of organic acid content which acted as a chelator for Pb. The formed Pb chelates with these organic acids possessed high solubility; hence increasing lead mobility. The citric acid (CA); 2 hydroxy-propane tricarboxylic acid, was the most effective organic acid chelator in mobilizing Pb and also worked well for Cd and Cu (Kim & Lee, 2010). For Cd, EDTA is a bit better (20%) than CA but CA possessed many advantages in cost, toxicity, biodegradability, and ground-water leaching risk. The logarithm of stability constant (log K) of the Pb-CA complex was 5.9-6.1 (Meers et al., 2008) and much lower than that of Pb-EDTA (~18). On the contrary, the low-cost immobilizing amendments such as CB, CM, FM, GW, BS, and RH can be used, either individually or in combination, in phytostabilization remediation. The use of combination amendments enabled multi-benefits for plants such as reducing Pb solubility and toxicity, increasing soil nutrients, and upgrading physical and biological properties of the soil (Herath, Kumarathilaka, Navaratne, Rajakaruna, & Vithanage, 2015; Kabas et al., 2014; Yang et al., 2016).

Biochars (CB, CM, and FM) raised soil pH, immobilized Pb, as also confirmed by several reports (Lu et al., 2017; Rajapaksha et al., 2015; Uchimiya et al., 2010; Yang et al., 2016). Bioslurry was less effective than chars because of the difference in its alkalinity and phosphorus content. Uchimiya et al. (2010) proposed three possible mechanisms of metal retention by chars, i.e. precipitate formation and or surfaces activation by pH rise, interaction with aromatic phi-electron, and complexation with functional groups. The mobilizing effect of lime material (SS and ES) is not in line with Islam et al. (2017). The very alkaline pH (11.6 and 12.6) resulted from the addition of ES and SS, has induced unexpected soil-Pb mobilization (%E - 53.2 and - 57.3). This is might be due to the formation of a stable polyhydroxy complex of Pb; Pb(OH)$_3^-$ and Pb(OH)$_4^{2-}$; at pH above 11 and 12 which increases Pb solubility and mobility (Heffron, 2015). Previous studies also showed that amendment of lime-based material without calcination (calcium carbonate, marble waste, lime, eggshell, oyster shell; pH 7.93-8.37) effectively immobilized/stabilized Pb (Ashrafi, Mohamad, Yusoff, & Hamid, 2015; Huang, Su, Rizwan, Zhu, & Hu, 2016; Kabas et al., 2014; Lim et al., 2013). For example, the addition of 5% eggshell to multi-contaminated soil (Pb, Zn, Cd) decreased leachable Pb by 28% at the end of 12 weeks experiment (Ashrafi, Mohamad, Yusoff, & Hamid, 2015). Similarly, Lim et al. (2013) remediated Cd,
Pb-contaminated soil with 5% eggshell and oyster shell and measured metals concentration in the TCLP-leachate. Results revealed that the lime-based amendments reduced TCLP-Cd by 30.13 and 57.66% and TCLP-Pb by 67.77 and 99.42%. Amendments containing CaCO$_3$ also increased pH value from 6.74 to 7.8-8.13 and these was the reaction of CO$_3^{2-}$ with water releasing OH$^-$ after CaCO$_3$ dissociation to Ca$^{2+}$ and CO$_3^{2-}$.

**Risk Assessment of Pb-contaminated Soil After [Im]Mobilization**

Risk assessment is conducted based on lead concentration in fractions of SEP. Assessment is based on the value of Contamination Factor (CF) and Risk Assessment Code (RAC). The relationships between CF and contamination degree were dedicated as CF < 1 (low contamination); 1 ≤ CF < 3 (moderate contamination); 3 ≤ CF ≤ 6 (considerable contamination); and CF > 6 (very high contamination) (Tytła, 2019). The RAC value expresses the ecological risk level of the metal. The criteria to assess ecological risk were categorize as RAC < 1% (no risk); 1% ≤ RAC < 10% (low risk); 10% ≤ RAC < 30% (moderate risk); 31 ≤ RAC ≤ 50 (high risk); and % RAC > 50 (very high risk).

Table 3 points out that contaminated soil has CF and RAC values of 7.1 and 15.39 respectively. It indicates that the soil possesses a very high Pb-contamination level with medium ecological risk (Tytła, 2019). In our work, CF values of amended soil varied from 0.52 to 8.34, and RAC values varied from 1.52 to 31.26%. The CB (CF 0.52; RAC 1.52) and CM (CF 0.76; RAC 2.98) are the two most effective immobilizing agents which improved soil into low contamination - low environmental risk status. On the other hands, effective mobilizing amendment CA (CF 8.34; RAC 31.26) changed soil status into very high contamination - high environmental risk. The effect of amendments on immobilization efficiency (E), contamination factor (CF), and the value of risk assessment code (RAC) is depicted in Fig. 3. Generally speaking, the more efficient amendment (higher E) will contribute to lower CF, RAC and the risk to the environment.

Table 3. Soil properties, BCR fractions Pb, immobilization efficiency (%) E, and risk assessment after incubation with amendments

| Amd | pH  | LOI  | F1  | F2  | F3  | F4  | (%) E | RSP; CF | RAC (%) and Risk |
|-----|-----|------|-----|-----|-----|-----|-------|---------|------------------|
| C   | 5.5 | 8.78 | 17.1| 69.3| 11.0| 13.7| 0.0   | 7.1     | 15.39 Moderate    |
| CM  | 8.3 | 10.92| 3.01| 32.9| 8.6 | 57.5| 82.4  | 0.76    | 2.98 Low          |
| CB  | 8.6 | 10.01| 1.54| 24.2| 9.0 | 66.5| 91.0  | 0.52    | 1.52 Low          |
| FM  | 8.6 | 10.55| 7.90| 59.9| 15.2| 19.6| 53.8  | 4.23    | 7.70 Low          |
| BS  | 6.8 | 11.95| 12.6| 70.6| 14.1| 12.1| 26.3  | 8.04    | 11.52 Moderate    |
| ES  | 11.6| 8.82 | 26.2| 47.8| 13.1| 16.0| -53.2 | 5.44    | 25.41 Moderate    |
| SS  | 12.6| 8.76 | 26.9| 50.8| 12.8| 17.2| -57.3 | 5.26    | 24.98 Moderate    |
| GW  | 7.8 | 8.76 | 9.2 | 64.8| 9.1 | 19.2| 46.2  | 4.33    | 8.99 Low          |
| RH  | 6.8 | 9.97 | 15.4| 64.5| 14.4| 17.1| 9.9   | 5.51    | 13.82 Moderate    |
| LJ  | 5.4 | 9.79 | 20.51|61.5| 16.4| 14.0| -19.9 | 7.03    | 18.23 Moderate    |
| TA  | 4.4 | 9.50 | 22.3| 56.4| 17.0| 16.9| -30.4 | 5.66    | 19.80 Moderate    |
| BO  | 3.5 | 9.88 | 29.2| 49.7| 12.6| 14.0| -70.8 | 6.54    | 27.68 Moderate    |
| TO  | 5.2 | 9.72 | 18.8| 67.2| 14.8| 13.1| -9.9  | 7.69    | 16.51 Moderate    |
| CA  | 2.9 | 15.04| 32.7| 44.8| 15.9| 11.2| -91.2 |8.34    | 31.26 High        |

Remak: Amd: Amendent; LOI: loss on ignition; F1: Pb-acid fraction; F2: Pb-Mn/Fe oxides fraction; F3: Pb-organics fraction; F4: Pb-residual fraction; C: control soil; CM: biochar from chicken manure; CB: biochar from chicken bone; FM: biochar from farmyard manure; BS: solid biogas slurry; ES: calcined eggshell; SS: calcined snail shell; GW: Gypsum waste; RH: rice husk; LJ: lemon juice; TA: tamarind; BO: baby orange juice; TO: sour tomato; CA: commercial citric acid
Effective immobilization amendments like CB and CM can be employed at the remediation of moderately lead-contaminated agricultural land to reduce the mobility of lead. At the same time, CB and CM may improve the physical, chemical, and biological qualities of the soil. On the contrary, the application of effective mobilization amendments (CA and BO) should be accompanied by the cultivation of plants that have strong roots to uptake lead as well as protect topsoil from erosion. By this means, the ecological risk of Pb contamination can be minimized.

The alteration of environmental risk may be influenced by redistribution of lead geochemical phases during soil incubation with amendments (Fig. 4). The control soil (before incubation) was dominated by a reducible fraction (F2) (69.3 ppm). F2 is a fraction of Pb bound to oxides or hydroxides of Fe and Mn. The dominant F2 fraction in this contaminated soil (C) may be correlated with the high Fe and Mn content of this soil (16.15 and 0.47%) (Table 1). Effective immobilizing amendments (CB & CM) significantly decrease F2, F1, and increase residual fraction (F4). The decrease of F2 is 45.1 and 36.4 ppm while the decrease of F1 is 15.56 and 14.09 ppm for CB & CM respectively. Meanwhile, the increase of F4 was 52.8 and 43.8 ppm. On the contrary, effective mobilizing amendments, such as CA & BO, mainly decrease F2 and increase F1 which results in the increase of environmental risk. CA & BO decreased F2 by 24.5 and 19.5 ppm and increased F1 by 15.6 and 12.1 ppm respectively.
Remarks: CA: commercial citric acid; BO: baby orange juice; SS: calcined snail shell; C: control soil; FM: biochar from farmyard manure; CM: biochar from chicken manure; CB: biochar from chicken bone

**Fig. 4.** The effect of amendments on the distribution of Pb-soil fractions

**Table 4.** Correlation and linear regression analysis of E with selected properties of amendment

| Properties    | Linear equation of E    | R     | R²      | N  |
|---------------|-------------------------|-------|---------|----|
| pH            | E = -39.016 pH + 5.397  | -0.266| 0.071   | 14 |
| pH (< 8.6)    | E = 28.217 pH – 0.166   | 0.948 | 0.899   | 12 |
| LOI           | E = -5.151 LOI + 51.174 | -0.150| 0.0225  | 12 |
| P             | E = 10.882 P + 8.812    | 0.907 | 0.823   | 14 |
| S             | E = 40.666 S + 7.683    | 0.222 | 0.049   | 5  |
| Ca            | E = 16.034 Ca + 2.595   | 0.839 | 0.704   | 5  |
| P+Ca          | E = 14.617 (P+Ca) + 2.018| 0.858| 0.736   | 5  |

**Important Properties of Amendment Affecting Pb Mobility**

The elemental composition of amendments in Table 2, pH, and LOI are correlated with immobilization efficiency (Table 3) and dedicated to evaluate important properties that determine amendment efficacy for remediating Pb-contaminated soil. The result is summarized in Table 4. By omitting extreme data of ES and SS, immobilization efficiency (E) was well correlated positively ($r = 0.948$) with pH up to 8.6. The lead was immobilized better in higher pH conditions. The abundant precipitating agents such as hydroxide and carbonate in alkaline conditions increased precipitation of lead hydroxide and lead carbonate. Secondly, alkaline pH also increased the negative charge of soil particles thus, to be effective sorption of Pb cation. This result is strongly supported by previous studies (Rajapaksha et al., 2015; Yang et al., 2016; Zhang et al., 2013). The exception result observed at extremely high pH on the addition of the lime-based amendment (ES and SS). The major
content of lime was calcium oxide (CaO) which increase hydroxide ion concentration in soil solution up to 0.01 M (pH 12). This high hydroxide ion tended to react with insoluble Pb(OH)\(_2\) formed soluble lead hydroxide complex, Pb(OH)\(_3^{-}\) and Pb(OH)\(_4^{2-}\). As a result, soil Pb was significantly solubilized. The study of Pb removal by electrocoagulation also revealed that removal efficiency decreased at pH higher than 11 (Heffron, 2015).

Good correlation with E was also observed at P, Ca, and the sum of phosphorus and calcium content, (P+Ca) (r = 0.907; 0.839; and 0.858) which suggested the formation of highly insoluble Pb phosphates during incubation. Mahar, Wang, Li, & Zhang (2015) stated that formation of hydroxypyromorphite, Pb\(_2\)(PO\(_4\))\(_3\)(OH)\(_2\) from Pb\(_2^+\) with hydroxyapatite, Ca\(_{10}\)(PO\(_4\))\(_6\)(OH)\(_2\) was putative mechanism in phosphate material remediation. This finding was supported by Shaheen & Rinklebe (2015) and Rizwan et al. (2016). In studying Pb immobilization by biochar, Cao, Ma, Liang, Gao, & Harris (2011) even proved the formation of hydroxypyromorphite (K\(_{sp}\) ~ 10\(^{-78}\)) from less stable cerussite, PbCO\(_3\) (K\(_{sp}\) ~ 10\(^{-12}\)), and hydrocerussite Pb\(_2\)(CO\(_3\))\(_3\)(OH)\(_2\) (K\(_{sp}\) ~ 10\(^{-18}\)) with XRD spectra at two-theta angles of 22 and 30\(^\circ\). Precipitation and coprecipitation mechanism contributed up to 84% of the Pb, which was much higher than surface sorption immobilization. In our result, the suggestion of hydroxypyromorphite formation is supported by the data in Table 3, that is, the increase of Pb residual fraction (F4) after incubation with CB and CM was as high as 52.8 and 44.2 ppm.

**CONCLUSION**

Chicken manure and chicken bone biochars effectively decreased lead solubility in soil. These substances were also considered as the most effective immobilizing amendments. Baby orange juice and commercial citric acid were the best mobilizing amendments which significantly enhanced soluble Pb. Amendment of chicken manure and chicken bone biochars lowered, yet but citric acid increased the environmental risk to the level of high-risk status. Important properties of amendment in influencing soil-Pb mobility were pH, P, and Ca content. Amendment of commercial citric acid in lead-contaminated soil should be followed by the cultivation of plants to prevent risk of contamination.

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