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IMMOBILIZATION AND REDUCTION OF LEAD (II) CONCENTRATION IN HIGHLY CONTAMINATED SOIL APPLIED WITH PEANUT HULL BIOCHAR

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Abstract

Biochar produced from waste biomass is increasingly being recognized as a green, cost-effective amendment for environmental remediation. The study was conducted to determine the effectiveness of biochar in immobilizing and reducing concentration of heavy metal particularly Lead (Pb) in contaminated soil. Biochar prepared from peanut hull was incubated with contaminated soils at rates of 0, 2.5, 5.0 and 10g kg^{-1} soil by weight for 55 days. Sub-sampling also was done at 25, 40, 50 and 55 days after incubation to determine the Pb reduction effect of biochar with time. Lead concentration was thoroughly quantified by following the aqua-regia leaching method at Soil Pedological Laboratory, Department of Agronomy and Soil Science, Visayas State University, Visca, Baybay City, Leyte. Results revealed that biochar was significantly increases the phosphorus content of the soil which enhances its effectiveness in immobilizing and reducing Pb concentration. Moreover, the effectiveness was enhanced with increasing incubation time from day 0 to day 55 and biochar rates (0 g to 10 g kg^{-1} soil). However, no significant differences in the pH and organic matter content of the soil were observed after addition of peanut hull biochar. After 55 days of incubation, soils treated with the highest rate of 10g kg^{-1} soil biochar showed the highest reduction in Pb concentrations in aqua - regia leaching method. This is due to the high phosphorus originally contained in biochar reacted with soil Pb to form insoluble hydroxypyromorphite $Pb_5(PO_4)_3(OH)$ which was presumably responsible for soil Pb immobilization. The results highlighted the potential of peanut hull biochar as one of the unique amendment for immobilization of heavy metal particularly lead in contaminated soil.

Key words: *Peanut hull biochar, lead immobilization, contaminated soil*

INTRODUCTION

Industrialization and technical advances have led to an increase of the discharge of environmental contaminants from industrial, residential, and commercial sources which degrades the surrounding ecosystems. Soil and water media in an ecosystem are frequently subject to contamination by organic and inorganic contaminants mainly due to anthropogenic activities. Accordingly, inorganic contaminants, particularly metals in the environment, originate mostly from a range of anthropogenic sources, such as mining, smelting, metal finishing, fertilizers, animal manure, pesticides, leaded gasoline, battery manufacture, power plants, waste water, and sewage sludge (Okatan et al., 2015; Usman et al., 2012; Lim et al., 2013; Okatan, 2018) while some soils can have a high background level of heavy metals due to volcanic activity or weathering of parent materials. In contrary to organic substances, heavy metals are non-degradable and hence accumulate in the environment having the potential to contaminate the food chain and threatens soil quality, plant and human survival.

Resulting for soil heavy metal pollution that poses a risk to the environment and to human health (Roy and McDonald, 2013) due to biomagnification (increases in metal concentration as the element passes from lower to higher trophic levels). Additionally, it has a pernicious effect on soil microbial properties (Yang et al., 2012) and on the taxonomic and functional diversity of soils (Vacca et al., 2012).

To reduce the risk related with the presence of high concentration of heavy metals in soils, actions are undertaken with the aims of immobilization and reduction of the toxicity of contaminants occurring in the environment. In view of the expense related with the measures commonly applied (e.g. dredging), the aim of which is the removal of contaminants; there appears the need for a search for more cost-effective solutions (Ghosh et al., 2011). The application of treatments aimed at reducing the bioavailability of contaminants can be considered as one of such solutions (Rakowska et al., 2012). Hence, technologies are advancing to remediate contaminated soil and water in which one of the most important technologies is to reduce the bioavailability of contaminants, and consequently decrease their accumulation and toxicity in plant and animals.

Recently, environmental remediation has been recognized as a promising area where biochar can be successfully applied (Ahmad et al., 2014). This is a carbonaceous material which has been deliberately used for in situ remediation of metal contaminated soil and water (Park et al., 2011a). Biochar is also recognized as a very significant tool of environmental management (Lehmann and Joseph, 2009). Therefore, numbers of studies have demonstrated that biochar has a high capacity to absorb pollutants in soils and sediments, and is known as a ‘supersorbent’ (Bornemann et al., 2007). Moreover, it has been used for a long time as sorbents for organic and inorganic contaminants in soil and water (Ahmad et al., 2012c). It is emerging also as an ameliorant to reduce the bioavailability of contaminants in the environment with additional benefits of soil fertilization and mitigation of climate change (Sohi, 2012).

However, contradicting explanations on the mobility of metals within biochar have been reported (Beesley et al., 2010) and it is not clear whether the use of biochar derived from agricultural crop wastes would provide qualitative or quantitative differences in efficacy through contrasting surface area and sorptive capacity (Zanzi et al., 2002).That is why it is indeed to have specific investigations into the mechanisms related to metal binding, transformation, and release. On the other hand, this study on heavy metal pollution is focused on Pb (II) as it is non-essential heavy metal which can cause health problems in humans or can result in phytotoxicity at high concentrations. With an increasing amount of literature on heavy metal remediation, the study aimed to determine the effectiveness of peanut hull derived biochar as a sorbent in immobilizing and reducing Pb (II) concentration in highly contaminated soil. Therefore it is hypothesized that application of peanut hull derived biochar could effectively immobilized and reduced the Pb (II) concentration in contaminated soil.

METHODOLOGY

Soil Collection and Preparation

Highly contaminated soil samples were collected at 0 - 20 cm soil depth at the dump site/garbage site near the copra drier of National Coconut Research Center (NCRC), Visayas State University, Visca, Baybay City, Leyte. Immediately after collection, the soils were air-dried and liberated from stones, litters, roots, and plastics. After two to three days of air-drying, sieving was done using a 2-mm wire mesh to ensure uniformity of the soil in terms of aggregate size and weight. Thereafter, the soil samples were set aside for initial analysis of pH (water), % OC, extractable P and Pb (mg kg^{-1}) concentration. Soil pH was analyzed potentiometrically using 1:2.5 soil-water ratio (PCARR, 1980); Organic Carbon (OC) using the modified Walkley–Black method (Nelson and Sommers, 1982), and extractable Pusing Bray P method and Pb (mg kg^{-1}) concentration using aqua-regia leaching method (Research Center for Toxic Compounds).

Peanut Hull Biochar Processing

Charring was done using a modified Top Lit Updraft Double Barrel processing (Quayle, 2010). Samples of air-dried peanut hull were placed into a tin can measuring 35 cm in height x 20 cm diameter (the carbonization chamber) at about ¾ height of the can. The carbonization chamber was tightly covered and placed in a drum measuring 80 cm in height x 40 cm diameter (the combustion chamber) with three holes (measuring 5 cm diameter) at its lower side. Three small cans measuring 5.8 cm in height x 5.2 cm diameter was placed under the carbonization chamber to facilitate airflow. After the set-up of the peanut hull and combustion chamber, the combustion chamber was filled with rice hull and wood chips at 3:1 ratio which acted as the combustion fuel to achieve an even burn. After a few minutes, a tin lid with a chimney (measuring 62 cm height x 8 cm diameter) was placed on top center of the drum to achieve a sufficient draft for a clean burn. During the 4 hours period of charring, temperature of about (500 ^{0}C and above) was monitored every 10 minutes using a thermometer attached to the external wall of the combustion chamber. After the peanut hull converted into char, the combustion and carbonization chambers were allowed to cool overnight at room temperature (28 ^{0}C) and removed from the carbonization chamber; weighed and placed in a sealed polyethylene bags ready for chemical analysis and incubation set-up. After charring, peanut hull biochar (PHB) was ground and passed through to a 2 mm sieve. The sieved PHB was also analyzed for pH, % OC, extractable P following the standard method of analysis above and Pb (mg kg^{-1}) using aqua-regia leaching method.

Set-up and Design

The study was carried out at the Soil Pedological Laboratory of the Department of Agronomy and Soil Science, Visayas State University from June to November, 2015. There were four treatments with three replications and laid out in a Randomized Complete Block Design. The treatments used were T_1 = Control (0 g Peanut hull biochar kg^{-1} air-dried soil), T_2 = 2.5 g Peanut hull biochar kg^{-1} air-dried soil, T_3 = 5.0 g Peanut hull biochar kg^{-1} air-dried soil, T_4 = 10.0g Peanut hull biochar kg^{-1} air-dried soil.

Contaminated soil incubation

Before amendment, 1 kg air-dried contaminated soil (unamended) was placed in a pot measuring 15 cm diameter and 25 cm height and allowed to equilibrate at room temperature for 24 hr. After which, the specified rates of peanut hull biochar were mixed well with pre-equilibrated soil. After addition, the soil moisture was adjusted to 75% of its field capacity. Thereafter, all pots were inserted with saucer at the bottom and covered also with a saucer respectively to control the moisture content of the soil during incubation. During the entire incubation period, sub-sampling of soil samples were done at 25th, 40th, 50th and 55th day of incubation and the soil samples were prepared, air-dried and set aside for Pb concentration determination (aqua-regia leaching method).

Data Analysis

Statistical analysis was done using the Sirichai Statistic Version no. 6. Data collected in pH, % OC, extractable P and Pb were subjected to analysis of variance (ANOVA) at 5% significance level to determine whether there were differences on their concentration in the contaminated soil as affected by different rates of peanut hull biochar. Duncan's Multiple Range Test (DMRT) was used to determine which of the treatment means were significantly different from the other.

RESULTS AND DISCUSSION

Properties of the soil and biochar

The properties of the peanut hull biochar and soil studied are presented in Table 1. The carbon content in soil and biochar were observed in higher percentage of 22.36% and 51.34% respectively. The higher organic carbon content of the studied soil could be due to regular burning of garbages in the dumpsite of Visayas State University. This is also related to the highest input of black carbon into the soil from different materials such as plastics, clothes, batteries, timbers and other organic materials and other wastes as well. Hence, higher organic carbon in the soil can be observed. Similarly, higher pH values on both biochar and soil in Table 1 were also observed. The higher pH value on biochar is attributed from pyrolysis process of peanut hull which is naturally high in organic carbon thereby increasing the pH of the material. On the other hand, biochar had also higher phosphorus value because aside from nitrogen fixation, peanuts have the inherent capacity to absorb more phosphorus nutrients from the soil.

Moreover, the presence of heavy metal (Pb) concentration in the studied soil showing a concentration of 1.38 mg kg^{-1}soil (Table 1). The results indicate that the soil is contaminated with heavy metal specifically Pb due to its accumulation from wastes such as plastics, cans and other sources in the dumpsite. Peanut hull biochar also contain very low concentration of Pb (0.06 mg kg^{-1} soil) because unlike other plant species which is considered as hyperaccumulator plants which have the mechanism in tolerating and higher absorption capacity of heavy metals.

Effect of biochar on soil pH, organic carbon and extractable phosphorus

The pH value of the soil remained unchanged during 55 d incubation (Figure 1). Although there were no significant differences ($p<0.05$) observed between the rates of biochar addition in changing the pH of the soil. However, the soil pH gradually increased afterward ranging from 7.30 -7.34 after addition of peanut hull biochar at a rate of 5.0g to 10g kg^{-1} soil. The increase in pH until 55 days of incubation was most likely due to dissolution of calcite ($CaCO_3$) present in biochar (Cao et al., 2011). In addition, the initial analysis also of peanut hull biochar was found to have alkaline pH value of 10.52 in Table 1 rendering its liming effect into the soil.

As a soil amendment, the peanut hull biochar should be stable for long term effectiveness. It was found out in the study that addition of peanut hull char biochar at 2.5 to 10g peanut hull biochar positively increased soil OC contents in contaminated soils (Figure 2). However, there were no significant differences ($p<0.5$) observed between the treatments, indicating no significant loss of biochar C and thereby showing its stability during 55 days of incubation. This observation was expected since the presence of Pb probably reduced microbial activity and slowed OC degradation. Similarly, a study of Cao et al. (2011) found to have no significant loss of organic carbon content in both soils due to less microbial activity and slow degradation of organic carbon due to the presence of Lead and atrazine in the soil. Furthermore, soil extractable P was significantly increase with the addition of increasing levels of peanut hull biochar (Figure 3). The higher increase of phosphorus in soil was mainly due to the inherent phosphorus content of the biochar applied.

Table 1. Chemical properties of contaminated soil and biochar before incubation

Chemical properties	Contaminated soil	Peanut hull char
pH_{water} (1: 2.5)	7.24	10.52
Organic C (%)	22.36	51.34
Extractable P (mg kg^{-1})	0.08	763.78
Pb (mg kg^{-1})	1.38	0.06

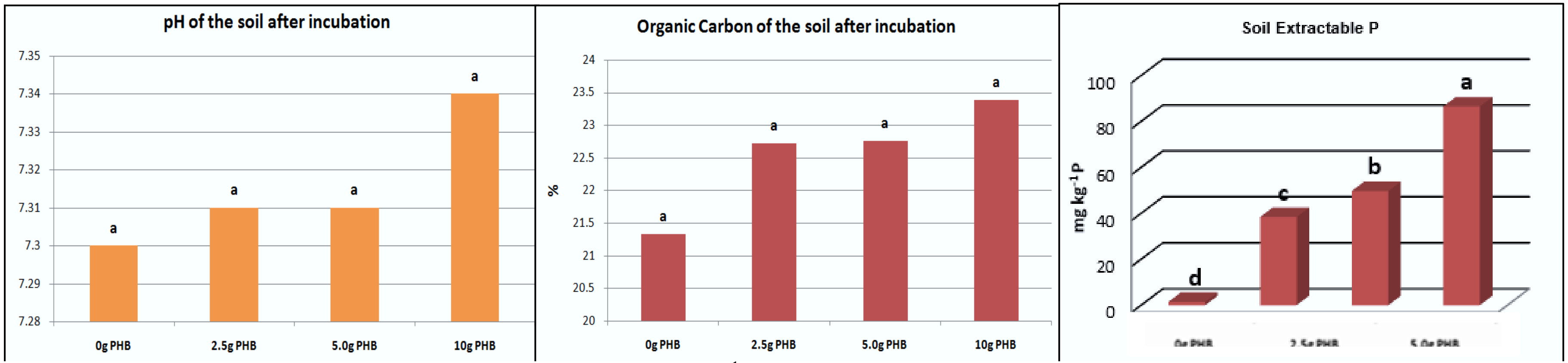

Figure 1,2 and 3. Soil pH, % OC and extractable P (mg kg^{-1}) after 55 days of incubation as affected with levels of peanut hull biochar

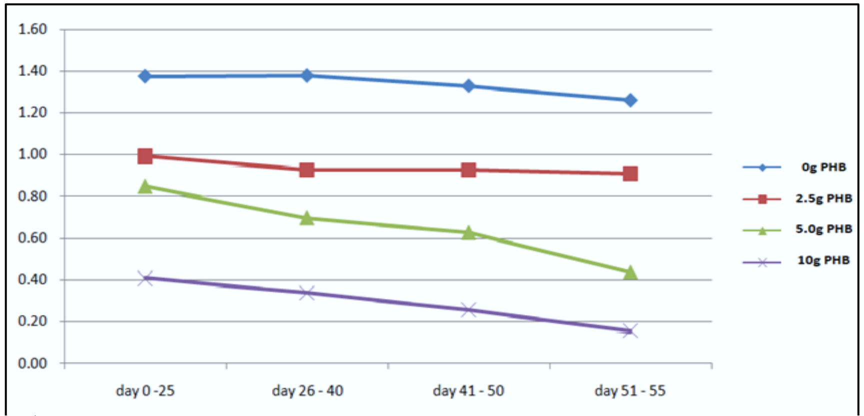

Figure 4. Soil Lead (mg kg^{-1} soil) concentration from day 0 to 55 days of incubation as affected with levels of peanut hull biochar

Effect of biochar on Pb concentration from day 0 to day 55 of incubation

The figure showed the Pb concentrations in the soil after 55 days of incubation (Figure 4). Results revealed that lead concentration was significantly ($p<0.05$) reduced in the biochar-treated soils, and the reduction increased with increasing biochar rates (2.5 – 10g PHB kg^{-1} soil) and incubation time 0 – 55 days (Figure 3). The results indicated that biochar was effective in immobilizing soil Pb. The results suggest that the ash component of the biochar may be primarily responsible for Pb immobilization and the most probable reason for this is that peanut hull biochar contains a much higher extractable P concentration causing more $Pb_5(PO4)_3OH$ resulting to the formation of hydroxypyromorphite and $Pb_5(PO4)_3Cl$ precipitates (Zheng et al., 2013). This was thought to be the main pathway by which peanut hull biochar immobilized Pb in contaminated soil.

Moreover, Ahmad et al. (2013) reported that in soil amended with biochar, rise in soil pH favored the sorption of Pb onto kaolinite making charge on kaolinite more negative. Since the pH of the soil is at pH > 5, Pb forms strong inner sphere bidentate surface complexes with kaolinite (Grafe et al., 2007). In the study also of Cao et al. (2011) reported the simultaneous immobilization of Pb and atrazine by dairy-manure biochar in soil. Those authors demonstrated that Pb was immobilized as a result of precipitation to insoluble hydroxypyromorphite due to the P content in the biochar, whereas atrazine was adsorbed onto biochar surfaces.

CONCLUSION

Therefore, application of peanut hull biochar is positively immobilize and reduce the concentration of Pb (II) and increases the phosphorus content of contaminated soil. However its application did not significantly increases the pH and organic carbon content of the soil.

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