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Immobilization of Heavy Metals from The Contaminated Soil Using Application of Sugarcane Organic Wastes By- Products

Osama E. Negim[1] and Atef A. A. Sweed[2*]

[1]*Soil and Water Department, Faculty of Agriculture, Sohag University, Egypt.*

[2]*Department of Soil and Natural Resources. Faculty of Agriculture, Aswan University, Egypt.*

THE AIM of this research was to evaluate the effect of three sugarcane waste by-products (pressmud PM, bagasse ash BA and molasse MO) which used as soil amendments of for immobilizing some heavy metals such as Cd, Cu, Cr, Ni and Pb in a contaminated soil near industrial factors of Qena Governorate, Egypt. The applied wastes at three levels of 0, 2.5 and 5 % (w/w) were incubated into the contaminated soil for a period of 120 days with wetting and drying cycles. The soil pH, organic matter content and DTPA extractable metals were determined after 30, 60 and 120 days of the incubation periods. The results indicated that BA addition to the contaminated soil have a high effective on reduce the soil content from DTPA-extractable of Cu, Ni and Pb compared with other treatments. Molasse (MO) application caused the best reduction in the mobility of Cd and Cr than BA and PM, while the application of pressmud (PM) had a moderate effect in reducing Ni and Pb of the soil.

Keywords: Metals, Contaminated soil, Immobilization and Organic waste

Introduction

In recent years, heavy metals contaminated environment is a widespread global problem in the world. In general, the accumulation of toxic metals such as Ag, As, Cd, Cu, Hg, Pb, Sb and Zn in soils is mainly inherited from parent materials and anthropogenic input from fertilizers, sewage sludge and pesticides in agricultural land. It may result in soil phytotoxicity as well as toxic to plants, animals, human's health and groundwater (Alloway and Ayres, 1997; Kabata-Pendias, 2001 and He et al., 2005). Therefore, it is very important to reduce or remove these metals in or from contaminated water and soils. Few decades ago, new techniques were developed to remediate heavy metals such as phytoremediation and in situ immobilization from contaminated soil, sediments and ground waters (Al-Oud & Helal 2003 and Negim et al., 2010 & 2012).

In situ immobilization or in situ stabilization techniques aim to decrease the toxic metals from sites contaminated with As, Cr, Cu, Pb, Cd, Ni and Zn by adding organic and inorganic amendments. In situ immobilization technique is simple effective approach for remediating the contaminated soils from metals, particularly when these soils are difficult or costly to be removed or treated out situ. It is also able to reduce the soluble metals through one or several processes such as metal adsorption through increased surface charge, formation of insoluble organic and inorganic metal complexes, sorption on Fe, Mn, and Al oxides, and precipitation. It also reduce the contaminant mobility and bioavailability with restoring the physical, chemical and biological properties of the soil using chemical and mineralogical agents such as industrial by-products (Mench et al., 2000; Oste et al., 2002; Bolan and Duraisamy, 2003; Adriano et al., 2004; Pérez de Mora et al., 2005; Raicevic et al., 2005 and Kumpiene et al., 2008 & 2012).

In few years, applications of organic amendments such as farmyard manure, poultry manure, pressmud, bagasse ash, molasses, tobacco dust, mushroom compost, biochar and pig manure are important in improving the physical, chemical

*Corresponding author: atefsweed@agr.aswu.edu.eg
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and biological properties as well as immobilizing metals of the contaminated soils (Gupta et al., 2003; Karaca, 2004; Gupta et al., 2009; Khan et al., 2012band Sabir et al., 2013 & 2015). Sugarcane by products such as bagasse ash, pressmud and molasses have been used for amending acid, saline sodic calcareous soils to improve chemical, physical and mineralogical soil properties and also to reduce or remove various soluble and exchangeable metals fractions in contaminated soils by increasing metal sorption and adsorption on the surface (Rasul et al., 2006; Ewulo et al., 2008; Aziz et al 2010; Zhou et al., 2012; Akkajet et al., 2013 and Negim et al., 2016). They are of low cost and rich in nutrients and organic matter as well as metals (Jamil et al., 2004 and Khan et al., 2008).

The objective of this work is to evaluate the impact of adding pressmud (PM), bagasse ash (BA), and molasse (MO) at different levels on the immobilization of Cd, Cu, Cr, Ni and Pb in the contaminated soil.

Material And Methods

An incubation experiment and analytical methods were conducted in the Department of Soils and Water and Department of Soil Science & Natural Resources, Faculty of Agriculture, Sohag and Aswan Universities respectively to evaluate the application of pressmud (PM), bagasse ash (BA), and molasse (MO) on the immobilization of heavy metals in the contaminated soil.

Soil sampling and analysis

A surface soil sample (0-30 cm) was collected from a polluted soil located near the paper industrial factory at Qena Governorate, Egypt. These soil samples were air- dried, ground and passed through a 2-mm sieve and kept for chemical and physical analysis as well as used in the incubation experiment. Data of physical and chemical properties of this soil are present in Table 1. The particle size distribution analysis was carried out using the sieving and pipette methods (Klute, 1986). Soil organic matter (OM) content was estimated by the modified Walkely-Blake method (Jackson 1973). Calcium carbonate content was determined volumetrically using the calibrated collin's calcimeter method (Jackson, 1973). Soil pH was measured in a 1:2.5 soil: water suspension by pH meter (Orion model 410A) according to Jackson (1973).

Total soluble salts were determined using electrical conductivity (EC) as dSm^{-1} in a 1:1 soil: water extract (Jackson, 1973). The available and total soil Cd, Cu, Cr, Ni and Pb concentrations were extracted using 0.05 M diethylene triamine penta acetic acid (DTPA) at pH 7.3 and aqua regia (3:1, v/v, HCl to HNO_3) respectively according to Lindsay and Norvel (1978) and Nieuwenhuize et al. (1991) respectively. All heavy metals were determined using ICP mass spectrometer (Icap6000 Series-Thermo Fisher Scientific Company).

Organic amendments

Sugarcane organic by-products such as pressmud (PM), bagasse ash (BA), and molasse (MO), were obtained from sugarcane factory, Upper Egypt. They were oven-dried (70 °C) and ground to pass through a 2 mm sieve. Each organic by-product was analyzed for some chemical characteristics according to Page et al., (1982). The obtained data are listed in Table 2.

TABLE 1. physicochemical properties of the studied contaminated soil

Soil parameter	Unit	Value
Sand	%	44
Silt	%	22
Clay	%	34
Texture		Sandy clay loam
pH (1:2.5)	-	7.65
EC (1:1)	$dS\ m^{-1}$	1.2
$CaCO_3$	$g\ kg^{-1}$	11.4
OM	$g\ kg^{-1}$	6.2
Total metals	$mg\ kg^{-1}$	
Cd		2.67
Cu		70
Cr		87
Ni		52
Pb		43
DTPA- extractable	$mg\ kg^{-1}$	
Cd		0.52
Cu		3.16
Cr		1.88
Ni		1.79
Pb		1.82

TABLE 2. Chemical characteristics of pressmud (PM), bagasse ash (BA) and Molasse (MO)

Character	Unit	PM	BA	MO
pH (1:10) (suspension)	-	7.3	8.6	6.97
EC (1:10) extract	dS m^{-1}	4.14	2.38	4.12
Total N	%	1.1	0.4	0.44
Total P	%	0.8	0.2	0.03
Total K	%	0.3	0.5	0.66
Ca	%	1.7	0.4	0.9
Mg	%	0.6	0.5	0.2
OM	%	12.5	18.2	6.8
Fe	mg kg^{-1}	568	442	847
Zn	mg kg^{-1}	107	35	21
Cu	mg kg^{-1}	55	66	12
Mn	mg kg^{-1}	236	78	24

Experimental design and remediation technique

A plastic pots with 20 cm height and 10 cm inter diameter were used in this study. Each pot was filled with 1 kg of prepared soil sample. These pots were divided into three mains groups (27 pot/main group) represented the organic wastes (pressmud (PM), bagasse ash (BA), and molasse (MO). The pots of main group were divided into three subgroups (9 pots / subgroup) representing the application rates of the used organic wastes (0. 2.5 and 5 %). The studied treatments were arranged among the experimental units in randomized completely block design in nine replicates. The pots were incubation at room temperature (25 ± 2) for 120 days. All pots were moisture by deionized water at 70% of water holding capacity of each treatment and repeated every three days. After 30, 60 and 120 days soil of three replicates of each treatment were taken and analyzed for its pH and the content of organic matter (OM) as well as the content of available heavy metals (Cd, Cu, Cr, Ni and Pb) as mentioned previously.

Statistical analysis

The results were statistically analyzed and was based on one-way analysis of variance (ANOVA) for the comparison of statistical significances among different treatments using SAS version 9.1, (SAS Institute, 2003). The least significant differences (LSD) at the 5% level of probability were computed to detect the difference among the treatments and their interactions.

Results and Discussion

Soil pH

Organic wastes applications resulted in an increase of soil pH at the three incubation time i.e. 30, 60, and 120 days (Fig. 1). In general, the soil pH significantly increased with increasing the application rate of the added organic wastes except that of MO as compared to the control treatment. After 30 days of incubation the highest soil pH (7.92) was found the treatment with BA2 followed by that found with the treatment of BA1 (7.8) and PM2 (7.72), while the lowest soil pH value (7.18) was recorded with the treatment of MO2. After 60 days of incubation organic waste application was caused slightly increase in soil pH with the treatments of PM and BA while soil pH decrease with the treatment of MO. The lowest value (7.11) in soil pH recorded with the treatment of MO2. But the highest value (7.98) was recorded for BA2.

On the other hand, after 120 days of incubation the application of MO have a low the soil pH, whilst both PM and BA treatments have a value of soil pH the wind range in the found a maximum soil pH was found with BA2 treatment (8.03) while the minimum value (7.06) was recorded for the treatment of MO2. These decreases of soil pH with increase molasses applications (MO1 and MO2) and increase incubation periods may be due to that polysaccharides in molasses rabidly decomposition to organic acids cause in decrease soil pH. The increases of soil pH at the all incubation time could be due to the high pH of both BA and PM compared to MO amendment. Several studies reported that the application of organic manure, poultry manure, pressmud, bagasse ash, biochar and activated carbon to contaminated soils caused an increase in the soil pH (Walker et al., 2003; Sabir et al., 2008; Kamari et al., 2015; Shaheen and Rinklebe, 2015 and Bashir et al., 2017).

Soil organic matter (SOM)

Additions of organic wastes have a greater effect on the soil organic matter content (%) at all incubation time (Fig. 2). The soil organic matter content was significantly increased with increasing rate of the added organic wastes compared to control. After 30 days of incubation the maximum SOM content was recorded with the treatment of BA2 (2.71 %) followed that with BA1 (2.47 %) and PM2 (2.23 %) treatments. After 60 days of incubation, the increase in SOM content was greater with using BA compared to other applications and control. The soil organic matter content after 120 days was lower than that found after 30 and 60 days of incubation. The decrease in soil organic matter content with the increase of incubation time could be due to their decomposition of the organic amendments and presence of soluble organic compounds that have varying stability of SOM content at the time incubation (Walker et al., 2003; Karaca, 2004; Clemente & and Bernal 2006 and Sabir et al., 2008).

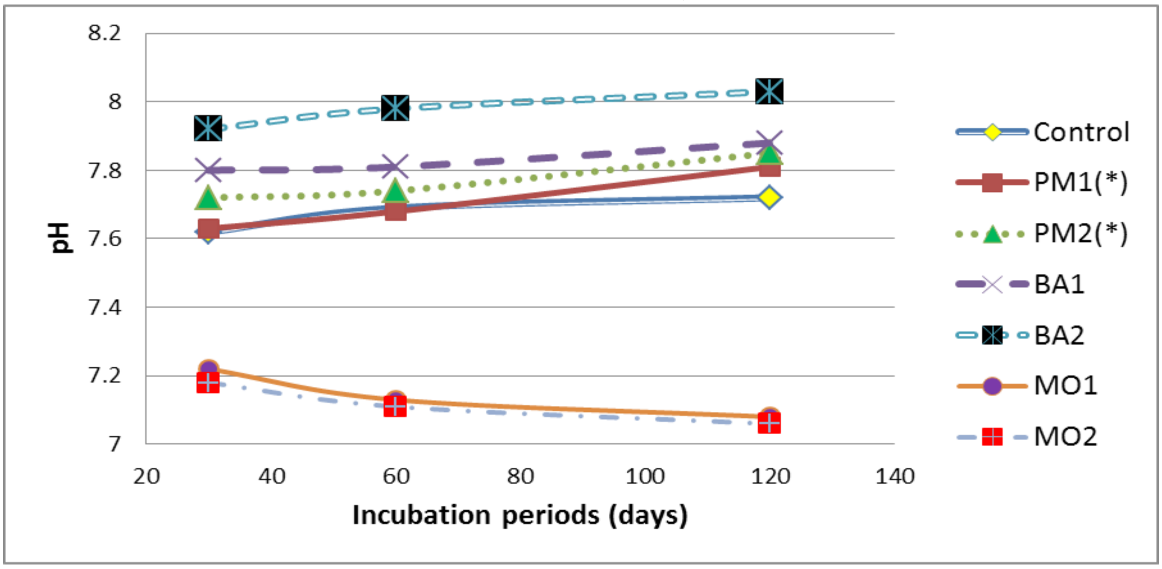

Fig. 1. Effect of pressmud (PM), bagasse ash (BA) and molasse (MO) on soil pH after 30, 60 and 120 days incubation times, LSD of treatments at 5% = 0.068

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

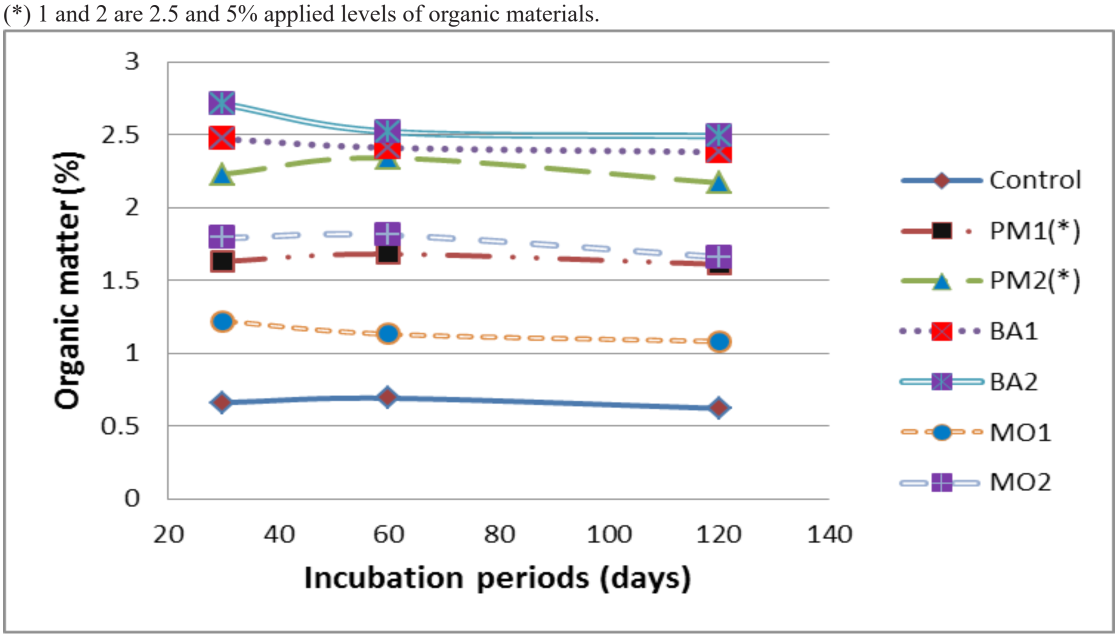

Fig. 2. Effect of pressmud (PM), bagasse ash (BA) and molasse (MO) on the soil organic matter (SOM) content (%) after 30, 60 and 120 days incubation times, LSD of treatments at 5% = 0.152

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

DTPA – extractable heavy metal contents of the soil

The impact of the applied organic wastes on the immobilization of the extractable soil Cd, Cu, Cr, Ni and Pb was presented in Figures 3, 4, 5, 6 and 7. These applications have a decrease or negative effect was on the soil content of DTPA-extractable heavy metals compared to the untreated soil (control). In addition, treating the contaminated soil with BA have a high decrease effect in the content of DTPA-extractable Cu, Ni and Pb compared to the other treatments. These finding mean that sugarcane bagasse and activated carbon application appeared high immobilization of soil heavy metals, where these applications reduced their availability and mobility in the contaminated soils (Sabir et al., 2008; Sabir et al., 2013 and Kamari et al., 2015). On the other hand, the applications of MO have a high decrease of the mobility of both Cd and Cr than the treatment of BA and PM. Moreover, the treatment of PM have a moderate effective in reducing Ni and Pb contents in the soil. In general, the reduction in the availability of heavy metals in the contaminated soil may be due to the found increases in soil pH as a result of applied organic wastes (Sabir et al., 2013).

Cadmium (Cd)

The additions of all organic wastes reduced the soil content of DTPA extractable Cd compared to the control at all incubation times (Fig. 3). More reductions the extractable Cd were found with increasing added levels of MO and BA compared to the treatment of PM for all incubation periods. Appling sugarcane bagasse-derived biochar was reported to reduce the Cd availability in the contaminated soils (Bashir et al., 2017). After 30-days of incubation, the highest decrease in the DTPA-Cd of the soil was recorded with the treatment of MO at 5.0% applied level which reached up 64.7% compared to control. Moreover, after 60-days of incubation, a maximum reduction in Cd was recorded with the addition of MO2 followed by the application of MO1 and BA2 reduced this content by 77.5, 73.4 and 55.0% respectively compared to control. The DTPA concentrations of Cd of the soil treated with all organic wastes after 120 days of incubation were lower than those after 30 and 60 days of incubation. The high reduction in the extracted DTPA- Cd was recorded for with the treatment of MO at 5.0% applied level which it was 84.6 % compared to control.

A decrease in the DTPA-extractable Cd with applying some organic wastes to a contaminated soil was also reported by Karaca, (2004) which due to formation of Cd – organic complexes. In a study of the effect of four amendments on the immobilization of some heavy metals such as Cr, Cd, Ni and Pb from sewage sludge by Gomah, (2016). It was found that, applying activated charcoal resulted in increased immobilising of Cd, Cr and Ni by Pb 52, 52, 51 and 49% for Cd, Cr, Ni and Pb, respectively.

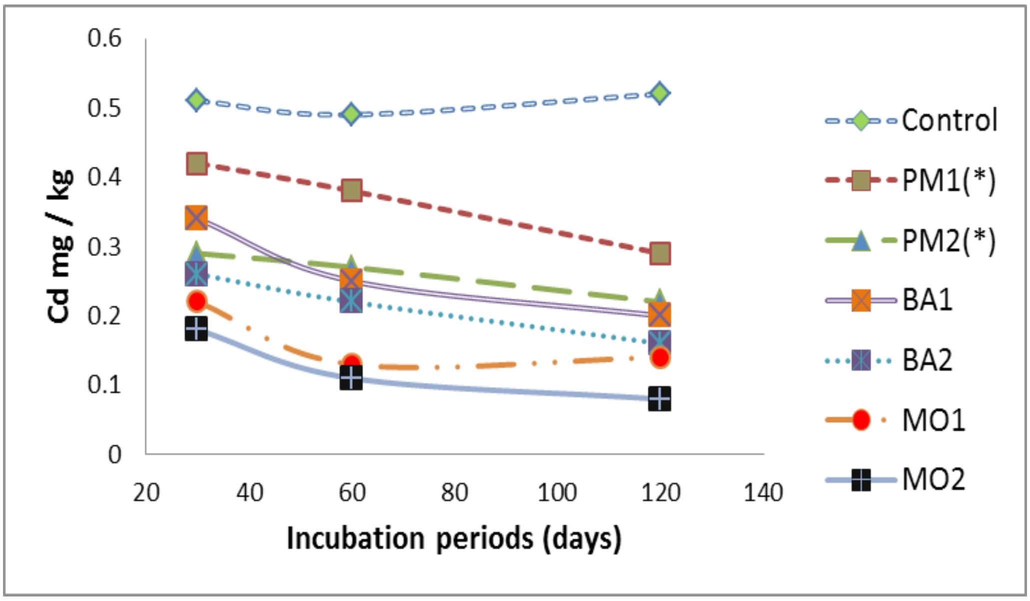

Fig. 3. DTPA extractable Cd of the soil treated with pressmud (PM), bagasse ash (BA) and molasse (MO) after 30, 60 and 120 days incubation times, LSD of treatments at 5% = 0.056

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

Copper (Cu)

Under the studied condition all applications of organic wastes reduced the DTPA-Cu concentration compared with control (Fig. 4). After 30-days of incubation, the DTPA-extractable Cu was decreased with increasing the applied level of BA and PM compared to MO applications. At that time, the BA1 and BA2 application reduced the DTPA-Cu by 83% and 89.5 %, respectively compared to control. However, PM1and PM2 additions decreased this content by 37.4 and 44 %, respectively relative to control. Similarly, after 60-days incubation, a highest reduction (91.3 %) of Cu was found with BA2 application followed by the application of BA1 (84.6 %) and PM2 (41.6 %) compared to control. On the other hand, after 120-days of incubation, BA2 treatment reduced the extracted DTPA-Cu by 92.7 % followed by these found with the treatment of BA1, PM2 and MO2 which were 87%, 52.8% and 50.4 %, respectively compared to control. The increase of soil pH and its content of organic matter have a great effect on the reduction the content of available Cu. These findings could be due to the effectives of OM and its stable form that could bind and complexes the soil Cu (Sabir et al., 2013).

Chromium (Cr)

A decrease effect of added organic wastes at different application rates as well as in the three incubation periods and the soil content of the DTPA-extractable Cr was found (Fig. 5). The DTPA-extractable Cr decreased with increasing the level of all three organic wastes added to the soil. At 30 days of incubation, the DTPA extractable soil Cr content where according to these decreases the used organic wastes may be arranged in the order of: MO > BA > PM > control.

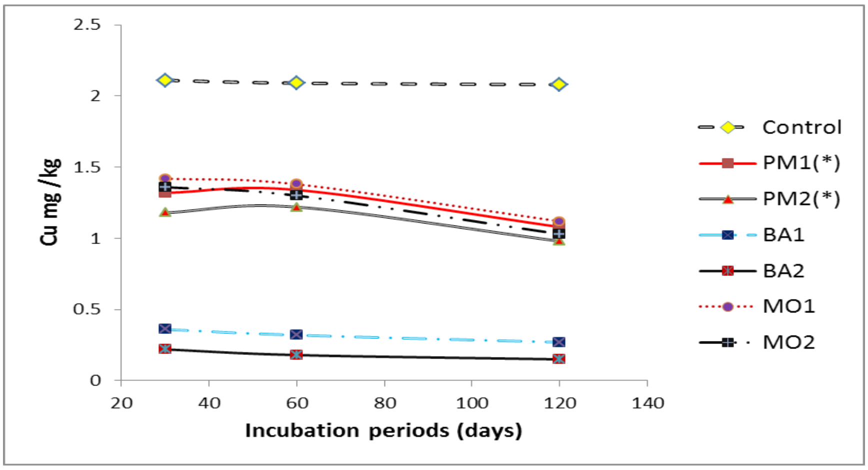

Fig. 4. DTPA extractable Cu of the soil treated with pressmud (PM), bagasse ash (BA) and molasse (MO) after 30, 60 and 120 days incubation times, LSD of treatments at 5% = 0.103

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

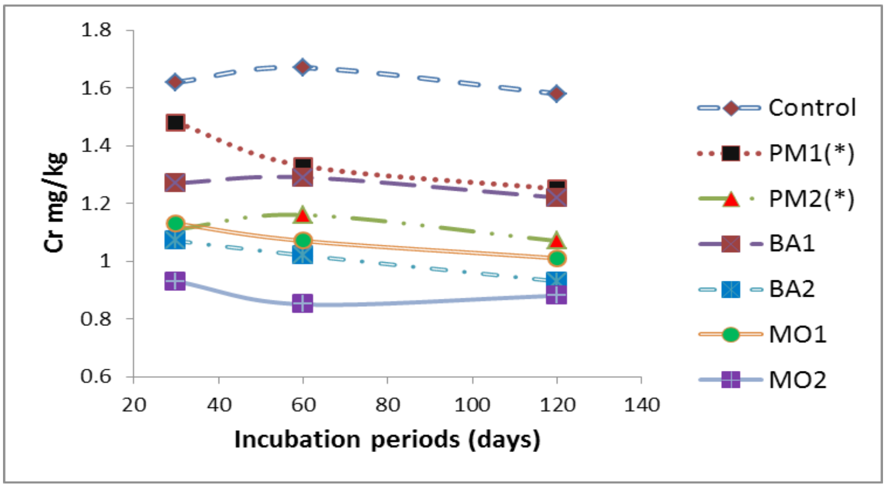

Fig. 5. DTPA extractable Cr of the soil treated with pressmud (PM), bagasse ash (BA) and molasse (MO) after 30, 60 and 120 days incubation times, LSD of treatments at 5% = 0.072

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

The highest reduction of Cr was found in the soil was observed with adding MO2 and BA2 which it was reduced by 42.5% and 33.9 %, respectively, compared to control. After 60 days of incubation, the PM1 application soil showed the highest content of the DTPA extractable Cr (79.6 %), while the application of MO2 resulted in a low decrease of extracted Cr (50.89 %) compared to the control. On the other hand, the DTPA extractable Cr after 120 days of incubation was decreased with adding all organic wastes compared to control. At that time, the DTPA extractable Cr was reduced by 44.3%, 41.3%, 36% and 32.2 % with using MO2, BA2, MO1 and PM2, respectively compared to control. Previous studies also showed that, the application of sugarcane wastes resulted in a reduce Cr availability of contaminated soils (Michailides et al., 2014; Bashir et al., 2017 and Legrouri et al., 2017).

Nickel (Ni)

Organic treatments decreased the soil content of extracted of DTPA Ni at different incubation periods compared to the control (Fig. 6). Control at 30-days of incubation, higher reductions of Ni were recorded with the high application rate of BA and PM compared to that of MO. The application of BA2 decreased the DTPA-extractable Ni by 33.5 % followed by BA1 (28.2 %) and PM2 (26.3 %) compared to control.

The bioavailability of Ni in the contaminated soil continued to decrease at 60 days incubation period with adding organic wastes especially at high application rates. The DTPA extractable Ni content was reduced in the order of: BA > PM > MO > control. After 120 days of incubation, BA2 had high reduction of extracted DTPA Ni (44 %) compared to control. The reduction in the DTPA-extractable Ni by the addition of organic wastes may be due to the increase in the pH of the BA and PM wastes compared to MO. Sabir et al., (2008) reported that, the addition of activated carbon to contaminated soils reduced Ni availability. In addition, It was seemed that, a relation between soil organic matter content, soil pH and the bioavailability of Ni in the soil (Karaca, 2004; Sabir et al., 2013). In a study of the effect of two amendments (filter cake and vinasse) on the immobilization and adsorption of Ni and Cu on loamy sand soil by Sweed (2019). It was found that, the filter cake was more ability on adsorption of both Cu and Ni compared with vinasse with increased shaking intervals and the highest adsorption amount was 4.72 mg g-1 for nickel with filter cake +soil treatment. Farid et al. (2013) who found that humic acid was more able to absorb Ni in aqueous solutions at pH 8.

Lead (Pb)

The used organic wastes had a varied influence on the extractability of Pb in the soil; however both the applications of BA and PM wastes showed higher reductions of Pb in the soil with the incubation time compared to MO and the control (Fig. 7). After 30 days of incubation, the treatment of MO2 exhibited the highest extractable Pb (2.28 mg kg^{-1}) but the treatment of BA2 displayed the lowest extractable Pb (0.64 mg kg^{-1}) followed by the treatment of BA1 (0.98 mg kg-1) and PM2 (1.05 mg kg^{-1}) relative to the control which was (1.62 mg kg^{-1}). There for, the treatment of BA1 and BA2 reduced the soil extractable Pb by 39.5% and 60.4 %, respectively, compared to the control.

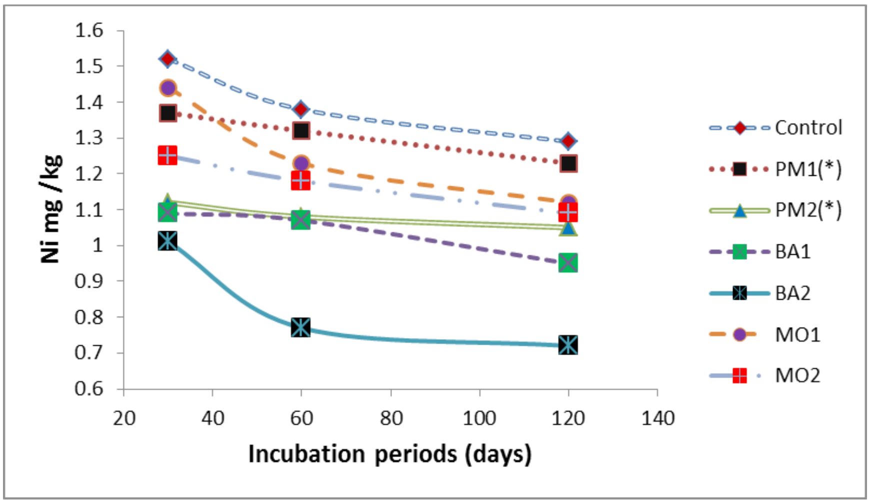

Fig. 6. DTPA extractable Ni of the soil treated with pressmud (PM), bagasse ash (BA) and molasse (MO) after 30, 60 and 120 days incubation times, LSD of treatments at 5% = 0.075

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

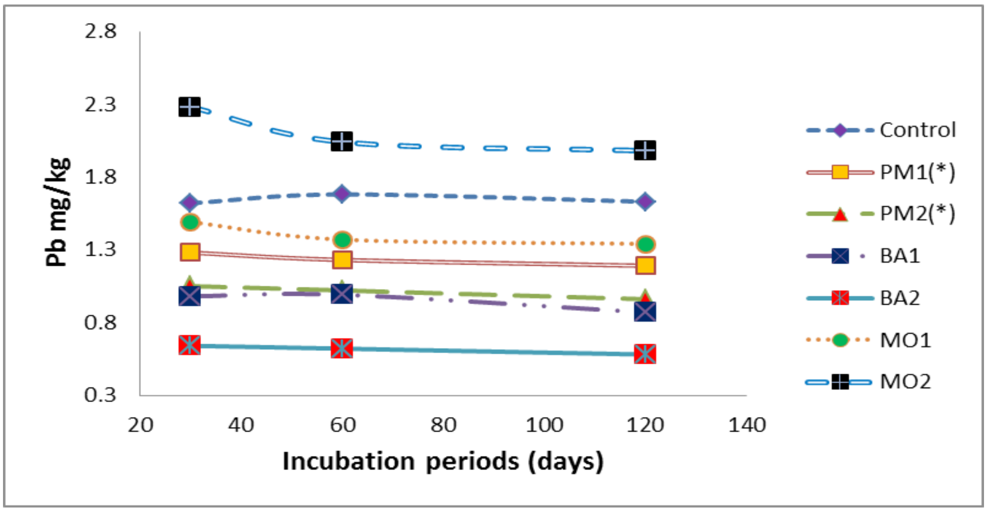

Fig. 7. DTPA extractable Pb of the soil treated with pressmud (PM), bagasse ash (BA) and molasse (MO) after 30, 60 and 120 day incubation times, LSD of treatments at 5% = 0.070

(*) 1 and 2 are 2.5 and 5% applied levels of organic materials.

Similar results were recorded after 60-days of incubation, the treatment of BA2 resulted in a high reduce in the extracted the DTPA-Pb that, these resulted from the followed by the treatments of BA1 and PM2 compared to control. However, after 120-days of incubation, highest reduction of the extracted DTPA-Pb were obtained applications of by BA1 and BA2 recorded reduction percent of 46.6% and 64.4 %, respectively, compared to control. In general, the immobilization of heavy metals in soils is greatly influenced by the soil pH (Zhou et al., 2014).

Some studies displayed that sugarcane bagasse and activated carbon were the most effective ones in immobilizing the heavy metals and reducing the bioavailability and mobility of metals in contaminated soils (Sabir et al., 2013 and Kamari et al., 2015).

Conclusion

This research was conducted to evaluate the addition effects of pressmud PM, bagasse ash BA and molasses MO on the reduction and stabilization of heavy metals (Cd, Cu, Cr, Ni and Pb) in a contaminated soil near an industrial site in Qena Governorate. Under the study conditions, the soil amended with those organic wastes at levels of 0.0, 2.5 and 5 % (w/w) was incubated for 30, 60 and 120 days with cycles of wetting and drying under laboratory the DTPA extractable Cd, Cu, Cr, Ni and Pb were determined. The results indicated that these organic wastes reduced the soil available Cd, Cu, Cr, Ni and Pb extracted by DTPA, where these decreases were increased in both added rates of organic wastes and incubation periods. Bagasse addition to this contaminated soil was more effective in reducing the DTPA-extractable Cu, Ni and Pb compared to the other wastes. However, molasses was the best one in reducing the bioavailability and mobility of Cd and Cr compared to BA and PM. Pressmud showed a moderate effect on reducing Ni and Pb in the soil. Thus, the addition of sugarcane wastes as a caused to protect the groundwater and plants from contamination.

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تقييد المعادن الثقيلة من التربة الملوثة باستخدام منتجات مخلفات قصب السكر العضوية

أسامة ابراهيم النجم* وعاطف عبد العزيز سويد **
*قسم الاراضي والمياه ، كلية الزراعة ، جامعة سوهاج ، مصر
** قسم الاراضي والموارد الطبيعية. كلية الزراعة ، جامعة أسوان ، مصر

يهدف هذا البحث لتقييم تأثير ثلاثة من نواتج مخلفات قصب السكر هم عجينة المرشحات (PM) ، رماد الباجاس (BA) والمولاس (MO) والتي تستخدم كمحسنات للتربة لتقييد بعض العناصر الثقيلة مثل Cd و Cu وCr و Ni و Pb بتربة ملوثة بالقرب من المنطقة الصناعية بمحافظة قنا ، مصر. تم تحضن المخلفات السابقة على ثلاثة مستويات هي ۰ و ۲٫٥ و ٥٪ (وزن / وزن) بالتربة الملوثة لمدة ۱۲۰ يومًا. تم تقدير pH التربة ومحتوى المادة العضوية (SOM) والمعادن المستخلصة بمادة DTPA بعد ۳۰ و ٦۰ و ۱۲۰ يومًا من فترات الحضانة. أشارت النتائج إلى أن اضافة الباجاس (BA) إلى التربة الملوثة له فعالية كبيرة في تقليل محتوى التربة من مستخلص DTPA من العناصر Cu و Ni و Pb مقارنة مع بالمحسنات الأخرى. وتسبب اضافة المولاس (MO)في أفضل اختزال في حركة كلا من Cd و Cr بالمقارنة بإضافة كلا من BA و PM ، في حين أن اضافة PM كان له تأثير متوسط .