

## **EFFICIENCY OF SOME SOIL AMENDMENTS TO REMEDIATE ARTIFICIALLY POLLUTED SOIL WITH LEAD\*\*\***

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### **ABSTRACT**

A column experiment was conducted to study the mobility of lead through soil layers. Lead was applied to the soil at 3 rates (0, 200 and 400 ppm) as solution of lead acetate. The effect of the following five soil amendments: [Farmyard manure (FYM), Polymer (Super-Hydro), Gypsum (G), triplesuperphosphate (TSP) and ethylene-diamin-tetra-acetic acid (EDTA)] at rates of [( 0.9%, 0.2%, 1.92%, 0.007% and 0.19% w/w )] the mobility and concentration of lead in soil layers was studied. Three concentrations of lead were added and mixed with the five types of soil amendments at starting from the surface soil layer in columns. In the present study the total and available lead concentrations were determined in three soil layers (0-15, 15-30, 30-45 cm). The effect of studied parameters showed that the interaction between pollutant (P), soil layers (L) and amendments (A) effect on total and available lead which were highly significantly in all treatments. The results showed that in soil without lead, total and available Pb had highest values with found by using FYM (0.9% w/w) followed by gypsum (1.92% w/w) treatment while, the lowest mean values were found with EDTA (0.19% w/w). In the artificially polluted soil with 200 and 400 ppm Pb the highest mean values of total and available Pb contents were recorded with unamended soil and with FYM (0.9%w/w), it was followed by gypsum treatment (1.92% w/w) while, the lowest mean values of total and available Pb contents was recorded with EDTA (0.19% w/w). It is clear that the concentration of total and available lead increased by increasing the level of Pb applying in the experimental soil. The highest mean values of total and available Pb were accumulated were found in the surface soil layers (top soil 0-15 cm).

**\*\*\*Part of M. Sc. Thesis, Soil Dept., Fac. Of Agric., Mansoura Univ., Egypt (2008).**

### **INTRODUCTION**

The study was conducted at Mansoura, within the heart of the Nile Delta. In Egypt, most of the industrial factories began to creep into and around the agricultural land. Thus, the occurrence of soil pollution with heavy metals such as Cd, Pb and Ni is likely to expected. The behavior of these metals in some soils of Egypt must be taken in consideration. Heavy metals are natural constituents in the soil; however, during the past years since the beginning of industrialization, huge changes have been occurred in the global budget of critical chemicals at the earth's surface (Derome and Lindroos, 1998). It is well known that lead (Pb) is strongly immobilized in the soil due to adsorption and precipitation. However, the reversibility of these reactions is poorly documented. Lead has a low mobility in soil equilibrium. Calculations predict that Pb solubility in the soil can be controlled by mineral phase, such as chloropyromorphite,  $Pb_5(PO_4)_3Cl$  (Degryse *et al.*, 2007).

Lead is a high toxic element and is one of the major chemical pollutants of the environment. Thus, attention has been given recently to the potential hazard of lead from various sources of contaminates to air, soil, water and plant. Removing heavy metals from soil is an energy and time consuming process. On the other hand, reducing the mobility of heavy metal species by means of a chemical additive (Stabilization/solidification technique) is a much more, expensive solution for heavy metal contaminated soils. There is a type of remediation, namely chemical remediation. Removal of heavy metal contaminates from soils is necessary in order to reduce the risk of contaminations in order to meet related to the environmental acceptable level (Abdel-Bary, 2000). Therefore, the present work was conducted to study the mobility and concentration of lead through the soil layers and in drainage water using column experiment, and non polluted and polluted Pb treated with the five amendments were examined [Farmyard manure (FYM), Polymer (SH), Gypsum (G), Triplesuperphosphate (TSP) and ethylene-diamin-tetra-acetic acid (EDTA)].

## **MATERIALS AND METHODS**

### **Experimental layout and objectives:-**

A column experiment was conducted to study the mobility of lead through soil layers along with the effect of the five amendments (FYM, P "SH", G, TSP and EDTA) on the mobility and concentration of lead in soil layers. The amount of total and available lead contents were determined in three soil layers (0-15, 15-30, 30-45 cm). The experimental design adapted was split split plot where the main plots were the soil amendments and in sub-main plots were the concentrations of lead. The experiments was divided in to two parts:-

- I. Amendments lead interaction and the amounts of total Pb in soil.
- II. Amendments lead interaction and the amounts of available Pb in soil.

### **Soil Sampling and Analyses:-**

Part one of experimental study; soil samples were collected, air dried; crushed with a wood roller, sieved to pass through a 2-mm sieve and preserved for analyses of some physical and chemical analyses in (Table 1).

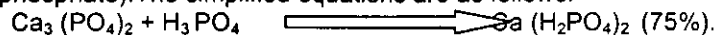
**Total lead content:** the total Pb was determined by wet digesting using mixture of sulfuric and perchloric acids according to Jackson, (1973). Lead in digested soil solution was measured by using Atomic Absorption Spectrophotometer.

**DTPA-extractable lead:** Available Pb was determined in all the soil samples by extracting with 0.005 M DTPA (diethylenetriaminepenta acetic acid) reagent using the method of Lindsay and Norvell (1978) and Pb was measured by Atomic Absorption Spectrophotometer.

### **Amendments:**

1-Ethylene-diamine-tetraaceticacid (EDTA.2H<sub>2</sub>O): EDTA powder was brought from El- Gomhoria Company. 2-Gypsum: The used gypsum was obtained from the soil improvement Soil Dept., Ministry of Agriculture and calculated as soil Gypsum Requirements (G.R). 3-Farmyard manure (FYM): Farmyard

manure (FYM) was brought from the farm of animal production, dried and wet digested (Jackson, 1973) for chemical analysis. 4- Polymer: Polymer in the form of Super-Hydro was brought from AI-DOHASA RVE CEARD GENEV. 5- Triple Super Phosphate (TSP) is formed when only phosphoric acid is formed firstly and reacts with the crude phosphate to form Ca-dihydrogenphosphate = (Triple phosphate):The simplified equations are as follows:



**Table (1): The main physical and chemical characteristics of the soil:**

Soil Properties	Mean Values
<b>Particle size distribution:</b>	
Sand %	14.60
Silt %	29.60
Clay %	55.80
Texture	Clay
Ca CO <sub>3</sub> %	1.40
Organic matter %	0.89
pH (1:2.5) soil: water suspension*	7.60
EC (1:5) **dS.m <sup>-1</sup>	0.97
S.P. %	67
<b>Soluble ions, meq/100g</b>	
Ca <sup>++</sup>	1.28
Mg <sup>++</sup>	1.40
Na <sup>+</sup>	2.10
K <sup>+</sup>	0.14
CO <sub>3</sub> <sup>--</sup>	00.0
HCO <sub>3</sub> <sup>-</sup>	1.56
Cl <sup>-</sup>	2.13
SO <sub>4</sub> <sup>--</sup>	1.23
<b>Available macro-nutrient (ppm)</b>	
N	32.00
P	3.74
K	375
Total Pb (mg/Kg soil)	42.32
Available Pb (mg/Kg soil)	31.91

\* pH was measured 1:2.5 soils: water suspension.

\*\* EC was measured in soil water extract.

\*\*\* Soluble ions (meq/100g) in 1:5 soils: water extract.

All of these amendments were added and mixed with the upper layer of each column at the rate of 0.19, 1.92, 0.9, 0.2 and 0.007 %, respectively. Some chemical and physical properties of these soil amendments were determined, and the amount of total and available lead and nickel in soil amendments, and the data are presented at Table (2):-

**Table (2): Total and available lead (mg/Kg soil) in soil amendments:**

Amendments	Lead Pb (mg/Kg soil)	
	Available	Total
FYM	1.59	1.71
P (SH)	00.0	2.27
G	0.47	1.8
T.S.P.	1.02	1.10
EDTA.2H <sub>2</sub> O	0.66	1.30

**Preparation of columns:**

Ninety cm plastic (PVC) column of 60 cm length and 11.5 cm diameter was used. The after column was layer into three categories; 0-15 cm; 15-30 cm; 30-45 cm height, leaving the uppermost 10 cm for applying irrigation water and at the bottom of column; 5 cm was filled with coarse sand as filter layer. Each column was filled with 7 kg oven-dried (at 105 °C soil) as follows: 5 kg were placed without any addition, then 2 kg soil were mixed with the soil amendments and Pb at the rates (0, 200 and 400 ppm) as solution of lead acetate of study as previously. Then, soil moisture was kept out at the rate of 60 % of water holding capacity throughout the experimental soil.

In part one of the experiment, representative soil samples were taken from each layer of the column (60 cm length) which were divided into 3 sections every one is 15 cm length then it was prepares for analysis; dried at (105 °C soil) and kept out for determining of total and available lead contents. These soil samples were brought of each layer and analysed after finishing of the experiment.

**Experimental design:**

The main plots were the applications of the different soil amendments. The sub main plots were the application to the soil surface layer concentrations of lead as soluble salts with different soil amendments as types of remedies.

Soil analyses: Soil mechanical analysis was used to determine the particle size distribution as described by Gee and Bauder, (1986). Saturation percentage of the soil was determined using the method described by Davies and Freitas, (1970). Calcium carbonate was determined using Collin's calcimeter according to Page et al., (1982). Organic matter, pH, and EC were determined as described by Jackson, (1973). Available nitrogen and available potassium were determined according to Hesse, (1971). Available phosphorus in the soil was extracted with 0.5 N Na HCO<sub>3</sub> solutions (pH 8.5) and determined using spectrophotometer after treating with ammonium molybdate and stannous chloride at a wavelength of 725 nm as described by Jackson, (1967).

**Statistical analysis:-**

The results were subjected to statistical analysis according to the complete randomized block design with two factors as described by Gomes and Gomez, (1984).

## RESULTS AND DISCUSSION

### ➤ Part I of the experiment:-

#### 1- (Lead – Amendment Interaction):-

This experiment investigate the effect of the interaction between pollutant (p), soil layers (l) and amendments (A) on total and available lead in artificially polluted soil:-

#### 1- Total lead (Pb):-

Table (3) showed that the interaction between soil pollutant (P), soil layers (L) and amendments (A) has a highly significant and positively affected the total and available Pb in polluted sections of the soil columns.

These data reveal that soils without lead and with three layer and with different soil amendments had the highest significant mean values of total Pb 56.28, 56.09 and 55.95 mg/Kg soil with FYM (0.9% w/w), followed by 56.18, 55.49 and 55.09 mg/Kg soil with gypsum (1.92% w/w) treatment in 0-15, 15-30 and 30-45 cm, respectively. On the other hand, the lowest mean values of total Pb was found in the soil without amendments and with EDTA (0.19% w/w). This decrease was found at rates of 11.71 and 8.76 % in the surface layer 0-15 cm but in 15-30 and 30-45 cm soil layer the decrease was at rates of (13.82 and 8.89 %) and (14.07 and 10.24 %), respectively. These results may be due to the fact that Pb is concentrated on soil surface and decrease downwards in the column. This is due to the low mobility of Pb in soil and the physical properties of the soil and also due to soil alkalinity. These results agree with those reported by Mills and Zwartich, (1975), and Al-Khashman and Shawabkeh, (2001). Williams *et al.*, (1984) found that Pb, Cd movement within the profile was limited to a depth of 5 cm below the zone of sludge application (0-20cm). Chlopecka (1996) indicated that the extractable concentrations of heavy metals from irrigated soils were all significantly higher than those of non-irrigated soils and that the concentrations of heavy metals found are highest in the topsoil.

Table (3) show that the highest significant mean values of total Pb in the Pb-treated soil with 200 Pb ppm were 198.16, 194.23 and 180.94 mg/Kg soil in the soil without amendments. These amounts of total Pb were 163.28, 155.52 and 147.90 mg/Kg soil with FYM (1.92% w/w) in soils collected at 0-15 cm, 15-30 cm and 30-45 cm, respectively. However, the lowest mean values in the polluted soil with 200 Pb ppm and recorded with EDTA (0.19% w/w) as relative values were 23.05, 26.81 and 28.58 % in the 0-15, 15-30 and 30-45 cm, respectively. The obtained data reveal that the pH of the soil decreased at the highest concentration of added organic matter (FYM) consequently, leading to the precipitation of lead as carbonate according by Shalaby *et al.*, (1996). These results are in parallel with those obtained by El-Sokkary, (1994) who indicated that atmospheric deposition of metals cause accumulated in the surface layer with little downward movement through the soil profile. This may be due to both high soil pH and carbonate content which lead to lower metal solubility and consequently reduce metal mobility in the soil. El-Shebiny and Khalifa (2001) found that organic matter fertilize (FYM) and Gypsum treatment which may cause accumulation of the lead in the soil

and consequently, increase its adsorption by soil particles which could be dangerous to human health.

Table (3) showed also that high significant mean values of total Pb in the artificially polluted soil with 400 Pb ppm and without amendments which were 400.27, 398.64 and 384.29 mg/Kg soil. These values were 378.41, 368.33 and 356.90 mg/Kg soil with FYM (0.9% w/w) in 0-15, 15-30 and 30-45 cm soil layers, respectively. Gypsum (1.92% w/w) treatment showed similar effect. The lowest mean values of total Pb in the artificially polluted soil with 400 Pb ppm and with EDTA (0.19% w/w) as relative decreased at rates of 7.84, 9.69 and 9.49 % in the 0-15, 15-30 and 30-45 cm. These followed with triple superphosphate (0.007% w/w), respectively. This reduction increased with the layers along with depth. The ability of synthetic chelators for enhancing the remediation of lead contaminated soil was decreased. Ethylenediaminetetraacetic acid (EDTA) was applied to the soil to elevate metal mobility. Results showed the decrease of the amount of metal accumulation. These results are in agreement with those obtained by Chen and Cutright (2001) who used gypsum in order to reduce the toxic effect of Pb especially where, pH was high consequently, the lead was accumulated. These results are in accordance with those obtained by Madhavi and Charyulu (1998) who used triple superphosphate due to the high soil pH, decreased the available Pb. This was also described by Abd El-Aziz *et al.*, (1993).

The same trends were observed for available Pb. The content of available Pb in the soil without lead with three layers and with different soil amendments was higher in the surface layer 0-15 cm than in the layer depths 15-30 and 30-45 cm.

Table (3) reveals that the soil without lead and with the three layer and with different soil amendments the interaction among it is a highly significant. In this treatment the highest significant mean values of available Pb were 46.93, 46.63 and 45.78 mg/Kg soil with FYM (0.9% w/w). These followed by 46.82, 46.55 and 45.45 mg/Kg soil with gypsum (1.92% w/w) in 0-15, 15-30 and 30-45 cm, respectively. The mean values in the soil without amendments and with EDTA (0.19% w/w) had decreased at the rate of 27.21 and 17.28 % in the surface layer 0-15 cm while in 15-30 and 30-45 cm soil layers the mean values of available Pb decreased at the rate of (27.32 and 17.56 %) and (27.28 and 18.57 %), respectively. In contrast, the high content of O.M in the soil was accompanied by the decrease in Pb content of the soil. Similar results were obtained by Abd El-Aziz *et al.*, (1993). The ability of synthetic chelators for enhancing the remediation of the contaminated soil decreased the amount of metal accumulation. These results are in agreement with those obtained by Chen and Cutright, (2001). This may be due to the fact that Pb is concentrated in the surface layer and decrease in the lower part of the soil. This is due to its mobility and the physical properties of the soil and its alkaline pH values. These results are in agreement with those reported by Mills and Zwartich, (1975) and Al-Khashman and Shawabkeh, (2001).

**Table (3): The amount of total and available Pb (mg/Kg soil) as affected by the interaction between pollutant (P), soil layers (L) and amendments (A) in soil.**

Soil layer (cm)	Amend	Total Pb			Available Pb		
		Control	200 Pb	400 Pb	Control	200 Pb	400 Pb
0-15	Control	49.69	198.16	400.27	34.16	148.25	247.41
	FYM	56.28	163.28	378.41	46.93	138.57	239.64
	Polymer	53.55	154.38	369.47	41.55	99.83	218.98
	Gypsum	56.18	162.41	372.37	46.82	125.52	230.39
	Triple P	55.85	158.65	371.80	44.78	109.43	227.58
	EDTA	51.35	152.49	368.88	38.82	90.89	197.22
15-30	Control	48.34	194.23	398.64	33.89	139.93	235.40
	FYM	56.09	155.52	368.33	46.63	129.32	231.65
	Polymer	52.22	143.35	362.25	40.38	87.17	207.59
	Gypsum	55.49	148.54	365.45	46.55	112.94	221.45
	Triple P	55.30	145.47	364.85	43.39	98.15	219.59
	EDTA	51.11	142.15	359.99	38.44	86.78	189.78
30-45	Control	48.08	180.94	384.29	33.29	129.16	223.83
	FYM	55.95	147.90	356.90	45.78	112.02	216.35
	Polymer	50.30	129.24	350.39	40.08	79.51	196.59
	Gypsum	55.09	140.57	352.41	45.45	104.19	207.87
	Triple P	55.03	133.16	350.69	43.11	89.89	206.63
	EDTA	50.22	129.22	347.81	37.28	77.72	163.13
<b>F.Test</b>		<b>**</b>			<b>**</b>		
<b>LSD</b>	<b>5 %</b>	<b>2.30</b>			<b>1.06</b>		
	<b>1 %</b>	<b>3.05</b>			<b>1.41</b>		

Control = unamended.

Control\*\* = non-polluted soil (soil without lead).

Table (3) indicated that regarding that the amount of available Pb in the artificially polluted soil with 200 and 400 Pb ppm was increased compared to soil without lead addition, the increase of Pb added into the surface layer increased the available Pb in soil. The highest significant mean values of available Pb were 148.25, 139.93 and 126.16 mg/Kg soil and 247.41, 235.40 and 223.83 mg/Kg soil in the artificially polluted soil with 200 and 400 Pb with FYM (0.9% w/w) as a type of soil amended in 0-15 cm, 15-30 and 30-45 cm soil layers, respectively. These followed by gypsum (1.92% w/w) treatment. The lowest mean values of available Pb in the artificially polluted soil were with 200 and 400 Pb ppm with EDTA (0.19% w/w). This decrease was at rates of 38.69, 37.98 and 39.83 %. The mean values of available Pb decreased at rates of 20.29, 19.38 and 27.12 % in 0-15, 15-30 and 30-45 cm soil layers, respectively.

It can be suggested that high soil pH values caused precipitate of Pb as hydroxide, phosphate or carbonate as reported as it promotes the formation of Pb-organic complex. These results are in a parallel with those by Leshber and Davis, (1985). Abd El-Aziz *et al.*, (1993) showed that used triplesuperphosphate, due high soil pH, the amount of available Pb decreased in soils.

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### كفاءة بعض محسنات التربة في معالجة تربة ملوثة صناعيا بالرصاص.

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أجريت تجربة أعمدة لدراسة حركة الرصاص خلال طبقات التربة. وقد تم إضافة الرصاص إلى التربة بثلاثة معدلات (صفر-200-400 جزء في المليون) على صورة محلول من أسيتات الرصاص. كما أضيفت محسنات التربة وهي: (السماد العضوي "FYM"، البوليمر "Super-Hydro"، الجبس "G"، تربل سوبر فوسفات "TSP"، إيثيلين داي أمين تتراسيتيك أسيد "EDTA") وذلك بالمعدلات الآتية: (0.9%، 0.2%، 1.92%، 0.007%، 0.19% و/و) على التوالي وذلك لدراسة حركة وتركيز الرصاص في طبقات التربة وهي (صفر-15، 15-30، 30-45 سم). حيث تم إضافة وخلط ثلاث تركيزات من الرصاص مع خمسة أنواع من محسنات التربة المضافة في الطبقة السطحية للتربة في عمود الدراسة. في هذه الدراسة تم تقدير الرصاص الكلي والصالح خلال طبقات التربة الثلاثة (صفر-15، 15-30، 30-45 سم). حيث أوضحت نتائج الدراسة أن تأثير التفاعل بين الملوثات وطبقات التربة والمحسنات المضافة على الرصاص كان تأثيره معنوياً في كل المعاملات حيث أوضحت النتائج أن التربة غير الملوثة بالرصاص (كنترول) كان الرصاص الكلي والصالح فيها أعلى تركيزاً مع معاملة السماد العضوي (0.9% و/و) يتبعها مع معاملة الجبس (1.92% و/و). أما في التربة الملوثة صناعياً بتركيزي (200، 400 جزء في المليون رصاص) وجد أن أعلى قيم للرصاص الكلي والصالح سجلت مع التربة غير المعاملة بأي محسن يليها مع المعاملة بالسماد العضوي (0.9% و/و) يليها مع معاملة الجبس (1.92% و/و) بينما كانت أقل القيم تركيز الرصاص الكلي والصالح مع معاملة الـ EDTA (0.19% و/و). يتضح من ذلك زيادة تركيز الرصاص الكلي والصالح مع زيادة تركيز الملوثات في التربة. كما وجد أنه يزداد تراكم الرصاص الكلي والصالح في الطبقة السطحية للتربة (صفر-15 سم) عن باقي الطبقات.

\*\*\*جزء من رسالة الماجستير بقسم الأراضي - كلية الزراعة - جامعة المنصورة.