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Effect of Poultry Manure Amendment on the Distribution and Mobility of Heavy Metals in Naturally Contaminated Dump Soil

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***Abstract*— *In this study, the effect of poultry manure amendment on the availability of some heavy metals, (Cu, Cr, Mn and Zn) was evaluated. The uptake of the metals by Ricinus communis (castor oil) with and without amendment was conducted in a green house. Soil sample was treated with 5%, 10% and 20% of poultry manure in a pot experiment. There was an increase in physicochemical properties of the soil such as pH, organic matter content and ECEC on treatment. Chemical speciation of the parent soil indicated that there was appreciable concentration of the metals in the extractable fraction. After three months of planting, the results showed that the extractability of the metals decreased significantly mostly with increase in percentage amendment. Residual fractionsgave the highest concentration of the metals and extractable having the least. 20% amendment has the best immobilization potential for Cu (7.07%), Cr (9.68%) and Mn (15.17%). The results also showed that amendment decreased plant metal uptake, generally decreasing as percentage amendment increased. These findings will be useful in the assessment and remediation of heavy metal-contaminated soils.*

***Keywords*— poultry manure, soil amendment, heavy metal.**

I. INTRODUCTION

Soil contamination by heavy metals is a major problem that has attracted much research interest the world over. In 2005 for instance, metals were the main contaminants reported to affect over 6000 sites studied in Denmark (Jensen *et al.*, 2009) just as about 20000 sites were reported in England and wales (Environmental-Agency, 2004). Similarly, over 100000, 80000 and 50000 pollution sites were reported in USA, European Union and Australia respectively (He *et al.* 2015). China may be the worst hit with about 14000000 ha sites reportedly contaminated by heavy metals (Sun *et al*. 2009). Not only do heavy metals occur naturally in parent rocks, but they emanate from other specific sourcesand anthropogenic activities such as mining, smelting, use of pesticides, fertilizers, sludge and emission from industries (Sawidis*et al.*, 2014).

Following lessons from historic catastrophic heavy metal contamination, public awareness has grown with focus on the implications of contaminated soils environment on human and animal health (Mulligan *et al*., 2001; Bolan *et al*., 2003). Soil remediation has become expedient and has been so promoted. The remediation of metal contaminated sites is howevercost intensive and employs environmentally invasive practices. Different remediation strategies have been employed for metal contaminated soils, includingphysical and chemical treatments such as acid leaching and electro-reclamation, excavation and landfilling and thermal treatments(EPA 2006). These are however only effective enough to lower the risk (Jiang *et al*., 2009) and would be more ideal for relatively small contaminated sites (Basta and McGowen, 2004; Debela *et al*., 2012) since these techniques are expensive.

Soil remediation using low-cost organic materials and wastes has been considered as a promising solution. Numerous amendments have been proposed and tested for soil stabilization, including agricultural and industrial by-products. Lee *et al. (*2013) studied immobilization in a contaminated rice paddy soil using egg shell waste and reported that the toxicity of Pb and Cd was reduced by 93.2% and 67.9% respectively. Walkers *et al. (*2004) reported that amending a metal contaminated soil with cow manure increased the growth of *Chenopodiumalbum* and reduced shoot concentration of Cu, Mn and Zn. Sato *et al. (*2010) evaluated Cd phytoavailibilty in soils as effected by application of chemical fertilizer and three types of animal waste compost derived from cattle, swine and poultry wastes and reported that Cd concentration in spinach grown on soil amended with animal waste compost was 38% lower than in plants cultivated on chemical fertilizer-treated soil. The effectiveness of stabilization strategy depends on the nature of the contaminants, physical and chemical characteristics of the amendment, and type of the soil.

Organic soil amendment harnesses the*in situ* stabilization of soils whereby an amendment is incorporated into a contaminated soil in order to immobilize heavy metals and reduce the uptake by plants without any side effect(Hartley and Lepp, 2008; Hosseini *et al*., 2013;). This method decreases the hazard potential of the contaminants into their least soluble, mobile or toxic form (Karbassin*et al*., 2014). Amendments may bind, absorb or co-precipitate metal contaminants, reducing metal mobility and availability. Organic amendments have been reported to decrease heavy metals bioavailability, shifting them from plant available forms that is extractable with water or neutral salts such as calcium chloride, to fractions associated with organic matter, carbonates or metal oxides (Walker *et al*., 2004). Consequently, functional groups present on the surface of organic amendments would provide binding sites for heavy metals.

II. METHODOLOGY

Sample collection

Soil samples were collected into plastic bags from several dump sites at Nnewi, Nigeria at 0-15cm depth using a spade. The samples were air-dried, sieved with 2mm sieve and then thoroughly mixed together before storing for further treatment. Poultry droppings were collected from a local farm in the study area,dried and ground for further assessment.

Pot Experiment

Pots with diameter of 14.0 cm and a height of 12.0 cm were filled with 2.0kg of soil in a greenhouse. The poultry droppings were added to the soil at 5%, 10% and 20% treatment in replicates. The treated soil samples were then thoroughly mixed, watered with deionized water and kept for two days in order to get equilibration of heavy metals between the amendments and the soils. Seeds of castor oil (*Ricinus communis*) were sown in each of the amended soil with thinning carried out on germination. The plants were uprooted after three months of germination, washed thoroughly with running water and deionized water respectively.

Plant Analysis

The harvested plants were dried for 72 hours at 60^{o}C and later ground. The plant materials were digested with concentrated nitric acid (Lina *et al*., 2009) and heavy metals (Pb, Cr, Mn, and Cu) were determined using AAS (model-PG 990).

Soil Analysis

The physico-chemical properties (pH, organic matter, exchangeable cation exchange capacity, particle size) of the parent soil samples and amended soil samples were determined by standard methods. Soil samples collected from each pot after plant harvesting and sequential extraction done for the soils. The various extracts wereanalysed for heavy metals using AAS (model-PG 990).

Sequential extraction

Sequential extraction was doneas described by Tessier *et al. (*1979) with modification as described by Sebasthiar*et al.*, (2005), replacing percloric acid with aqua regia. All extracts were analyzed using AAS (model: PG-990).

Exchangeable

To 1g of amended soil sample, 8 mL of 1M $MgCl_2$ was added with pH adjusted to 7.0 with agitation for 1hr before centrifuging for 15 mins. The supernatant was filtered into a polypropylene bottle for AAS analysis, while the residue was used for further extraction.

Carbonate bound

1M NaOAc (8mL) was added to the residue obtained from the exchangeable fraction above and then adjusted to pH 5.0 with concentrated acetic acid and agitated for 5 hrs. The mixture was then centrifuged at 15 rpm for 15 mins. The supernatant was filtered into a polypropylene bottle for AAS analysis.

Fe-Mn Oxides

20 mL of 0.04M $NH_2OH.HCl$in 25% HOAc was added to the residue obtained from the carbonate bound fraction and placed in water bath for 6 hours at 96±3ºC. The mixture was then centrifuged at1500 rpm for 15 mins beforethe supernatant was filtered into a polypropylene bottle for metal analysis.

Bound to organic

3 mL of 0.02M HNO_3 and 5mL of 30%H_2O_2 adjusted to pH 2.0 was added to the residue obtained from the step above and mixture was heated to 85±2ºC for 2 hours.3mL of 30% H_2O_2 was later added and mixture heated to 85±2ºC for 3hrs before centrifuging at 1500rpm for 15 mins. The supernatant was filtered into a polypropylene bottle for metal analysis.

Residual fraction

Residue from the organic bound extraction was digested with 8mL of aqua regia for 2 hrsbefore collecting for analysis.

Determination of mobility factor

The mobility factor which is the percentage fraction of heavy metals that are mobile or available for plant absorption was calculated thus:

$$MF = (F1 + F2)/(F1+F2+F3+F4+F5) \times 100$$

Where F1 = exchangeable fraction, F2 = bound to carbonate, F3 = bound to Fe-Mn Oxide, F4 = bound to organic, F5 = residual.

DATA ANALYSIS

Results are presented as mean value ± standard deviation and analyzed by analysis of variance (ANOVA) using SPSS software package. Multivariate analysis was also used for the comparative immobilization effect of different manure amendments and forms of heavy metals in the dump soil. Statistically significant differences between means were determined by Least Significant Difference (LSD) at 95% confidence limit.

III. RESULTS AND DISCUSSION

Physicochemical properties

Table 1 shows the physicochemical properties of the amended and unamended soil samples. The pH, ECEC and organic matter of the dump soil increased with increase in manure amendment in the order, 20%>10%> 5%resulting in change inconcentrations. The variation may be due to the different concentrations of manure especially since the change in values were statistically significant ($p < 0.05$). Increased application of poultry manure resulted to the reduction in the mobility factors of the metals. The increasein percentage organic matter detectedwhich is statistically significant ($p < 0.05$) may be due to varying concentrations of poultry amendment. Mohammadi *et al.*,(2011) andHou *et al.*, (2012) suggested that manure is an organic source of nutrients which increases the soil organic matter and enhances soil quality.

	Manure Amendment			
	Control	**5%**	**10%**	**20%**
PH	6.72±0.02	6.73±0.01	6.95±0.04	7.10±0.05
%Organic matter	3.77±0.03	4.21±0.00	3.88±0.12	4.15±0.01
ECEC (cmol/kg)	14.58±0.07	15.41±0.49	19.11±2.50	25.82±0.31
% Sand	93.8±0.00	93.80±0.00	93.80±0.00	93.80±0.00
% Silt	2.8±0.00	3.40±0.00	3.40±0.00	3.40±0.00
% Clay	3.4±0.07	2.80±0.00	2.80±0.00	2.80±0.00

Metal Distribution in Amended soils

Tables 2 to 5 gives the distribution of various metals in the amended and unamended soils.

Chromium

The residual fraction of the extract contained the highest Cr for all amendments giving54.00%, 56.40 %, and 61.70 % for 5%, 10% and 20% amendment respectively. The concentrations of Cr in the 5% sequential fraction followed the order; residual > bound to Fe-Mn oxide> bound to organic > exchangeable > bound to carbonate; 10% wasresidual > bound to organic >bound to Fe-Mn oxide> exchangeable > bound to carbonate and residual > bound to organic >bound to Fe-Mn oxide> bound to carbonate> exchangeable. For 20% amendment. The mobile fractions of Cr available for plant absorption in dump soil amended with 5%, 10% and 20% of Poultry manure ranges from 9.68% - 13.02% with the 20% amendment of poultry manure having the smallest amount.

Copper

The highest fraction obtained for Cu in dump soil amended with 5%, 10% and 20% of Poultry manure was obtained in the residual fraction while the lowest fraction was obtained in exchangeable fraction. For all the percentage amendments, the concentration of sequential fractions of Cu (mg/kg) followed the order; residual > bound to organic > bound to Fe-Mn oxide > bound to carbonate > exchangeable. Residual fraction of Cu obtained in soil amended with 5% of Poultry manure is 56.50 %, lower than residual fraction obtained in the soil amended with 10% and 20 % of Poultry manure that recorded 53.70% and 53.70% respectively. The exchangeable fraction of Cu obtained in the dump soil amended with 5% of Poultry manure is 3.70%, higher than the exchangeable fraction obtained in the soil amended with 10 % and 20% of Poultry manure that recorded 1.60% and 0.90 % respectively.The mobile fractions available for plant absorption were 10%, 7.29 %, and 7.07 % for soil amended with 5%, 10% and 20% of Poultry manure respectively. It was observed that increase in % amendment resulted to decrease in concentration of mobile fractions of Cu in dump soil.

Manganese

The residual fractions contained the highest fraction of Mn in all percentage amendments at 37.0% and 32.7% for 10% and 20% amendment respectively. Based on the Mn concentration (mg/kg), the sequential fraction as observed followed the order; residual > bound to Fe-Mn oxide > bound to organic > exchangeable > bound to carbonate fraction for 5% amendment, residual > bound to Fe-Mn oxide > bound to organic > exchangeable > bound to carbonate fraction for 10% amendment and residual > bound to organic > bound to Fe-Mn oxide > exchangeable > bound to carbonate fraction for 20% amendment. The mobile fractions of Mn available for plant absorption in dump soil in order of decreasing fraction is 17.09 % > 15.70 % > 15.17 % for soil amended with 10%, 5% and 20% of poultry manure respectively.The mobile fractions of Mn was highest in the soil amended with 10% of Poultry manure but lowest in the soil amended with 20 % of poultry manure.

Zinc

The fraction with highest concentration of Zn in dump soil amended with 5%, 10% and 20% manure was obtained in the residual fraction while the lowest fractions were obtained in the exchangeable fractions. For all the percentage amendment, the concentration of sequential fractions of Zn (mg/kg) followed the order; residual > bound to organic > bound to Fe-Mn oxide > bound to carbonate > exchangeable. Residual fraction of Zn obtained in soil amended with 5% of Poultry manure is 52.80 %, higher than residual fraction obtained in the soil amended with 10% and 20 % of Poultry manure that recorded 50.20 % and 49.20 % respectively. The mobile fractions of Zn available for plant absorption are 14.42%, 15.43 %, and 15.04 % for soil amended with 5%, 10% and 20% of poultry manure respectively. The order of mobile fractions of Zn available for plant absorption with respect to amendment with poultry manure is, 10% > 20% > 5 % poultry manure amendment.

Table 2: Distribution of Cr in soil samples amended with 5%, 10% and 20% poultry manure

	Control		5%		10%		20%	
	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction
F1	5.25±1.05	11.41	2.58±0.36	6.50	2.62±0.37	6.60	1.79±0.29	4.50
F2	9.63±1.29	20.93	2.2±0.78	5.50	2.55±0.17	6.40	2.01±0.46	5.10
F3	6.24±0.90	13.56	7.49±2.46	18.80	5.26±1.15	13.20	3.66±0.86	9.30
F4	11.67±2.07	25.35	6.1±1.24	15.30	6.87±1.27	17.30	7.6±2.21	19.40
F5	13.23±2.59	28.75	21.58±3.6	54.00	22.41±5.05	56.40	24.21±4.71	61.70
Sum	46.03		39.95		39.71		39.27	
Mf%	32.34		11.96		13.02		9.68	

Key: F1=Exchangeable, F2= Bound to Carbonate, F3= Bound to Fe-Mn oxide, F4= Bound to Organic, F5= Residual, Mf= Mobility factor

Table 3: Distribution of Cu in soil samples amended with 5%, 10% and 20% poultry manure

	Control		**5%**		**10%**		**20%**	
	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction
F1	8.28±1.07	15.88	1.41±0.25	3.70	0.66±0.06	1.60	0.46±0.08	0.90
F2	11.28±2.60	21.64	2.4±0.41	6.30	2.36±0.54	5.70	2.96±0.39	6.10
F3	7.30±0.34	13.99	4.76±0.72	12.50	7.16±1.34	17.30	8.43±1.02	17.40
F4	2.81±0.60	5.40	7.99±2.91	21.00	9.02±1.79	21.80	10.55±2.72	21.80
F5	22.47±3.41	43.10	21.55±3.93	56.50	22.25±7.67	53.70	26±3.97	53.70
Sum	52.15		38.11		41.45		48.40	
Mf%	37.52		10.00		7.29		7.07	

Key: F1=Exchangeable, F2= Bound to Carbonate, F3= Bound to Fe-Mn oxide, F4= Bound to Organic, F5= Residual, Mf= Mobility factor

Table 4: Distribution of Mn in soil samples amended with 5%, 10% and 20% poultry manure

	Control		**5%**		**10%**		**20%**	
	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction
F1	21.64±3.54	13.37	14.26±3.87	10.40	14.8±3.21	10.70	15.34±2.37	8.6
F2	17.38±1.73	10.74	7.33±2.69	5.30	8.83±0.52	6.40	11.66±1.11	6.5
F3	54.27±6.31	33.52	36.16±11.87	26.30	36.33±6.02	26.30	43.46±4.08	24.4
F4	37.90±4.41	23.41	30.46±11.93	22.10	27.15±3.25	19.60	49.38±5.46	27.7
F5	30.70±4.19	18.96	49.31±14.71	35.90	51.15±11.66	37.00	58.17±18.34	32.7
Sum	161.89		137.52		138.26		178.01	
Mf%	24.10		15.70		17.09		15.17	

Key: F1=Exchangeable, F2= Bound to Carbonate, F3= Bound to Fe-Mn oxide, F4= Bound to Organic, F5= Residual, Mf= Mobility factor.

Table 5: Distribution of Zn in soil samples amended with 5%, 10% and 20% poultry manure

	Control		**5%**		**10%**		**20%**	
	Mean (mg/kg)	% fraction	Mean (mg/kg)	% fraction	Mean (mg/g)	% fraction	Mean (mg/kg)	% fraction
F1	36.03±4.13	19.04	4.44±1.11	4.40	4.38±1.13	3.90	3.21±0.35	2.40
F2	42.81±5.19	22.62	9.94±2.95	10.00	13.15±3.25	11.60	16.56±2.31	12.60
F3	26.36±4.47	13.93	15.76±3.75	15.80	17.24±4.68	15.20	23.02±31.91	17.50
F4	18.48±3.46	9.77	16.95±3.95	17.00	21.81±6.91	19.20	23.97±1	18.20
F5	65.56±7.56	34.64	52.62±11.15	52.80	57.06±17.62	50.20	64.7±21.91	49.20
Sum	189.24		99.71		113.64		131.46	
Mf%	41.66		14.42		15.43		15.04	

Key: F1=Exchangeable, F2= Bound to Carbonate, F3= Bound to Fe-Mn oxide, F4= Bound to Organic, F5= Residual, Mf= Mobility factor

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Mobility Factor and Plant Metal Uptake

The effect of poultry manure amendment on mobility factor and plant metal uptake is shown in Figures 1 and 2.The total concentrations of the heavy metals available for absorption by *Ricinus communis* after amendment with different percentages of the manures used are calculated and presented as mobility factor. An increase inpoultry manure amendment decreased the mobility factor just as metal uptake by plant decreased. Plant metal uptake was decreased in the poultry manure amended soil compared with plant metal uptake in the unamended soil. Lowest mobility was recorded in the plant with lowest plant metal uptake. Alloway and Jackson (1991) corroborated this results in a similar study where it was reported that the metal uptake from soil to plant was slow because organic matter introduces new binding sites to the soil and therefore present fewer risk for plants, as compared to unamended soils. Similar effects for biochars and biosolids of poultry manure was studied by Uchimiya*et al*. (2012) and Wuana*et al*. (2012).

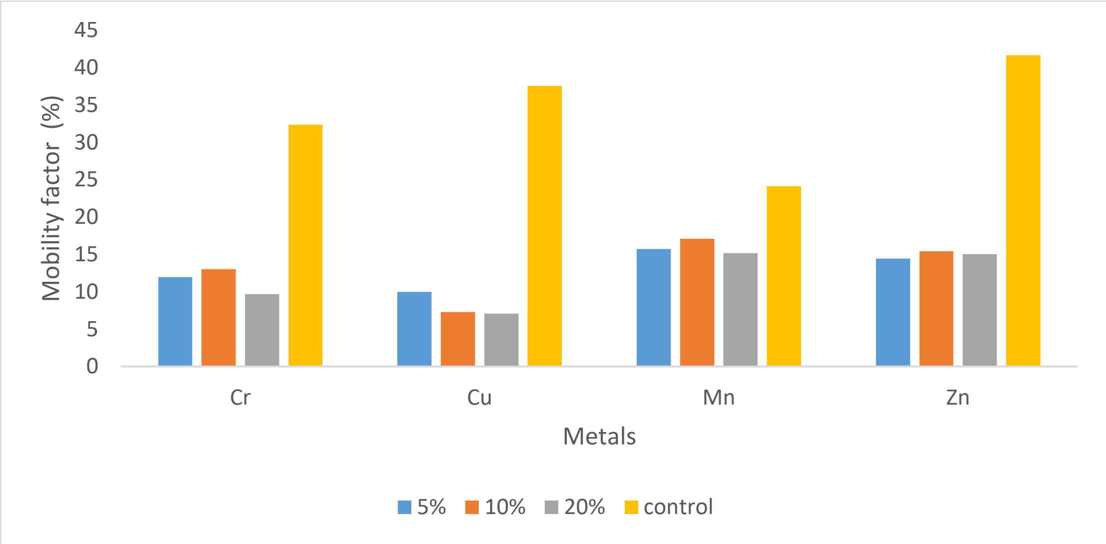

Fig.1: Effect of manure amendment on mobility factor (Mf)

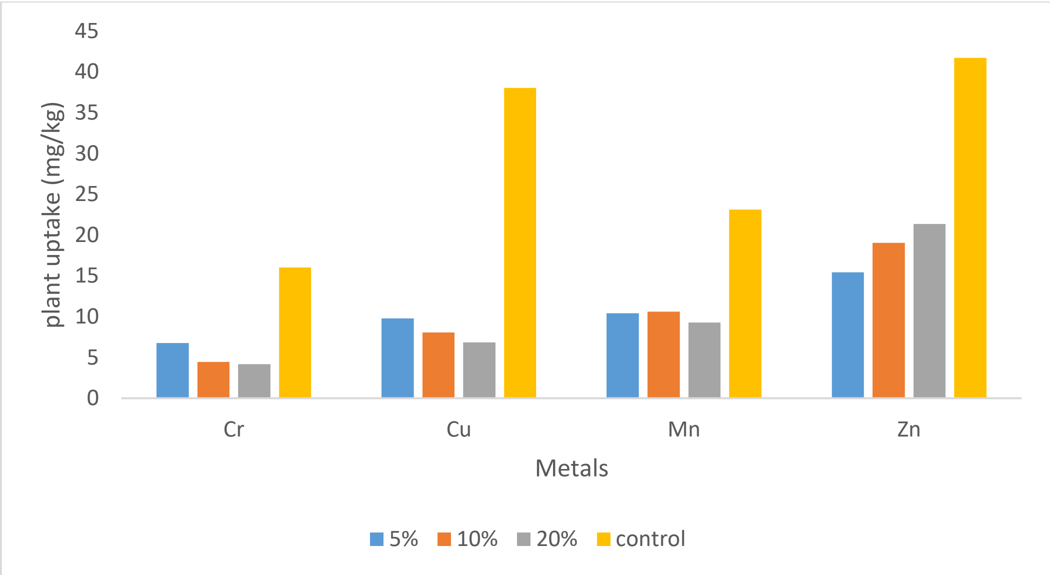

Fig.2: Effect of manure amendment on plant metal uptake

IV. DISCUSSION

Application of poultry manure increased pH, ECEC and organic matter resulting in change inconcentrations as well as reduction in the mobility factors of the metals. Generally, the concentrations of the heavy metals present in the dump soil changed in increasing order of residual >

bound organic > bound to Fe-Mn oxide >bound to carbonate > exchangeable. Results from Azeez *et al.* (2019) however disagrees with the current findings reporting anorder of abundance of the fractions in dump soil amended with poultry manure was carbonate bound >Fe-Mn oxides bound > exchangeable. Nonetheless, they reported that the application of poultry manure resulted in reduction in the available Zn, exchangeable Zn and oxide-bound Zn, Pb and Cd in line with the finding of this research. According to Azeez and his colleagues, the reason for the high carbonate-bound heavy metals in the dumpsites reported in their work could be attributed to the fact that most of the heavy metals were in association with carbonate salts. Poultry manure amendments has been reported for Cu optimization in the contaminated soil (Thomas and Dauda, 2015);but according to Walker *et al*. (2003) the release of phosphate, carbonates and other salts after the application of composted poultry manure may transform into metal insoluble compounds and decreased metal solubility.

The immobilization effect of poultry manure on various heavy metals as seen in this research is in agreement with the reports of Irshad *et al*. (2014) and Wuana*et al.* (2012). This may be due to the presence of trace metal (TM) sorbents capable of reducing TM solubility and enhancing immobility in the soil as reported by Haroon *et al.* (2019). Hanc *et al*. (2008) reported that poultry manure and compost decreased the available Cd and Cu content of contaminated soil. Lina, *et al*. (2009) also reported a decrease of soluble/exchangeable fraction of Cd and increase in organic-bound fraction when compared with the control. This also agreed with the current findings. Okieimen *et al*. (2011) while reporting stated that total uptake of Cr and Cu by maize plant decreased with increased loads of poultry amendment to the contaminated soil. These reductions were as associated with the capacity of the amendment to immobilize metals in soils. They stated that the organic matter and phosphorus content of organic amendment could account for the immobilization of metals.

Plant metal uptake shows that increase in percentage manure amendment decreased the mobility factor which also resulted in decrease in plant metal uptake this in agreement to the report of Angelove*et al.* (2010) who reported that organic amendment reduced the concentration of Pb, Zn, Cu and Cd in potato tubers . Italso showed that the immobilization potential of the manure increased as their concentration in the soil was increased implying that poultry manure is a good immobilizing agent for remediation of heavy metal contaminated soil. The manure amendment significantly ($p < 0.05$) decreased the metalconcentrations in the extractable fraction (exchangeable and bound to carbonate) when compared with un-amended soil.

V. CONCLUSION

The concentrations of all the metals in each of the soil fractions significantly ($p<0.05$) varied for amendment with poultry manure. It was observed that % amendment with poultry manure significantly affected the forms of Cd, Cu, Mn, and Zn ($P<0.05$). Multiple comparison between 5%, 10%, and 20% of poultry manure revealed that 20% of poultry manure has more effect on the forms of Mn, and Cd while 10% of poultry manure has more effect on geochemical forms of Zn and Cu.The finding also shows that higher percentage amendment immobilizes metals more than lower percentage amendment except for Cd where immobilization effect decreases with percentage amendment.

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