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Effects of Sludge on Some Soil Properties and Heavy Metals Uptake by some Vegetables Grown on Tannery Sludge Amended Soils in Kano Metropolis, Nigeria

[1]Mahadi Garba Doka*, [2]Mohammed M. I.
[1]Department of Chemistry,
Federal College of Education (Tech.),
Bichi. Kano Nigeria

[2]Department of Pure and industrial Chemistry,
Bayero University,
Kano Nigeria
Email: gmahadidoka@gmail.com

Abstract
Tannery sludge waste is currently being used by many farmers for the cultivation of vegetables on large scale for human consumption. The study investigated the effect of tannery sludge on some soil properties and levels of heavy metals in Amaranthus hybridus and Corchorus olitorius grown on the tannery amended soil. Pot experiment was conducted and the metals (Ni, Pb, Cd and Cr) were analyzed using Atomic Absorption Spectrophotometer. The results showed the soil pH, organic matter content and conductivity of the amended soils increased as compared to the controls. The metal concentrations in the amended soil were higher than 12.90 ± 0.07mg/kg, 44.80 ± 0.05mg/kg, 1.90 ± 0.32mg/kg and 11.40 ± 0.55mg/kg obtained in the unamended soil for nickel, lead, cadmium and chromium respectively. Also higher concentration of nickel (15.73 ± 1.17mg/kg) in the leave, lead (60.41 ± 9.97mg/kg) and cadmium (1.99 ± 0.85mg/kg) in the root of Corchorus olitorius were observed. While higher concentration of chromium (348.99 ± 50.31mg/kg) was found in the root of Amaranthus hybridus. The concentrations, except of nickel in Amaranthus hybridus tissues were above the World Health Organization recommended safe limits. But, the transfer factors of all the tested metals in the vegetables tissues ranged from 0.01 – 0.81, showing none of the vegetables is hyperaccumulator. Still the concentrations are not safe for dietary consumption. Hence, stringent guidelines set for tannery sludge applications on agricultural land should be totally enforced.

Keywords: Amended soil, Heavy metals, Sludge, Tannery, Vegetables.

INTRODUCTION
Over the years, an increase in population, as well as human activities, generates wastes, which alters the natural characteristics of the environment. Waste is defined as some materials discarded after use or as moveable materials that are perceived to be of no use (Njoku, & Agwu, 2017). Which releases a high amount of heavy metals into surface and groundwater, soil and ultimately in the biosphere. Thus, the metals in plants or animals can be dangerous at high concentrations due to their chronic toxicity and carcinogenetic effects as well as fatality (Reeves & Baker, 2000).

*Author for Correspondence

Land contamination is common in urban and peri-urban areas as a result of industrial and municipal wastes released. Industrial wastes of major pollutants are generated from tannery industries (Ezike *et al*., 2012; Ademir *et al*., 2013; Ana *et al*., 2014; Luna *et al*., 2014), where hides and skin are converted into leather. The operations generate a large amount of organic and inorganic wastes known as sludge. Sludge continually increase because of the high demand in leather materials and its status in the Nigerian economic development (FME, 2012). Thus, indiscriminate disposal of tannery sludge increases and due to these facts, the safe disposal of waste is becoming one of the main environmental problems. Probably due to the associated costs and lack of appropriate disposal regulations. Hence, this gives an opportunity to most farmers sourcing for cheap fertilizers. Thus the application of tannery sludge wastes as soil amendments to agricultural land has being a common practice over the past several years. Since this scenario is a characteristic of most farmers closed to wastes disposal sites in northern Nigeria, there is the need to characterize waste quality especially tannery sludge waste.

Studies have revealed that waste (Sludge) can transfer the significant amount to toxic metals such as lead, chromium, cadmium, nickel, iron, mercury, and others into the soil ((Taek-keun, *et al*., 2010; Njoku & Agwu, 2017), where plants absorb their minerals including the heavy metals. Eventually, these metals are taken up by human beings and animals through the food chain.

Vegetables are the fresh and edible portions of herbaceous plants, which are eaten raw or cooked. They contained valuable food ingredients such as carotene, ascorbic acid, riboflavin folic acids, minerals, and dietary fibres, which have marked health effects (Funke, 2011). It's also a source of antioxidants, vitamins and other phytochemicals with antioxidant characteristics that give protections against many diseases (Fasola & Ogunsola, 2014). Indeed, vegetables constitute an important part of the human diet since they contain carbohydrates, proteins, as well as vitamins, minerals and trace elements (Uwah *et al*., 2012).

In this study, the effects of using tannery sludge on the irrigation of Amaranths (*Amaranthus hybridus*) and Jew mallow (*Corchorus olitorius*) were investigated for heavy metals (Cd, Pb, Cr, and Ni) accumulation. This becomes necessary considering the importance of the vegetables to health and the fact that many people have now resulted in eating vegetables for their healthy living. The aim of the study is to evaluate the effect of tannery sludge amendment on heavy metals availability and physiological responses of the above vegetable species.

The study Area

The study area is the Tannery sludge disposal pits located behind Kano Coca – Cola industry, Challawa industrial estate in Kumbotso local Government area. Kumbotso falls within the Northern part of Kano, Nigeria. It is bordered in the West by Madobi local government, in the Southwest by Rimin Gado, in the South by Dala and Gwale, in the East by Kano municipal and Dawakin kudu local government areas. Tanneries are the dominant industries in the area.

The types of waste found in the area were mainly tannery sludge wastes and some leather remains. Poor wastes management services and lack of law enforcement to control disposal are the main factors that accounted for the continuous indiscriminate dumping in and around the pits environs. All the industries located in the area dump their sludge in the pits. This practice has been going on for a longer period without any caution from the authorities to stop it. Hence, the sludge accumulated over the years and being a major source of organic fertilizer for the inhabitants.

MATERIALS AND METHODS

Experimental design

The experiment was conducted with analytical grade reagents and deionized water. Heavy metal concentrations were determined with Variant Spectra Atomic Absorption Spectrophotometer (Agilent Technology 240 model) equipped with a digital readout system. All measurements were carried out in triplicate.
Seeds of Amaranths (*Amaranthus hybridus*) and Jute or Jews mallow (*Corchorus olitorius)* were purchased from Sharada Market in Kano metropolis. These were screened, labeled and stored in the laboratory for planting.
Sandy loam soil was obtained (0 – 20cm) from an abandoned farm at the new site of Bayero University, Kano Nigeria. The sample was air dried and screened for pebbles, stones and leaves and then thoroughly mixed to establish homogeneity (Olayinka *et al*., 2011). The sample was labeled in plastic container for laboratory analysis.

The tannery sludge was obtained from ten different locations in the tannery disposal sites. Each sample was separately air-dried, gently crushed with mortar and pestle and sieved using 2mm plastic and stored in a labeled container for planting and subsequent laboratory analysis.

Tannery sludge amended soil samples were formed by mixing sludge and soil samples in a 1:10 ratio. The homogenous mixture was transferred into ten plastic Pots measuring 16.5cm × 12cm × 14.5cm. The control pot contained only the soil (uncontaminated).

Planting was conducted in the Botanical garden of Bayero University, Kano. Seeds of the vegetables were sown in each pot separately, depending on the vegetable species. The pots were arranged in a complete block design, in eleven columns and two rows. Each row contained a particular vegetable species. The eleventh column served as the control.

Two weeks after planting, vegetables were thinned and left to grow under natural conditions with watering at regular intervals. After eight weeks, the plants were harvested by uprooting (when the soil was moist) using plastic hand trowel and gently removed (Ogunkunle *et al*., 2013). The vegetable samples were then wrapped, labeled and transported to the laboratory.

Sludge and Soil physiocochemical analysis

The pH and Electrical Conductivity (EC) were separately determined by dissolving 10g of the amended soil in 50cm^3 deionized water (1:5w/v) and shaken until homogeneity was reached. A glass electrode pH meter (Jenwsay 3510 model) and conductivity meter (Jenway 4010 model) were used to measure pH and conductivity respectively (Rayment & Higginson, 1992).
Organic Matter (OM) content was determined using the modified Walkley – Black method as described by Bruce and Schulte (2009) using 10cm^3 of 0.167M potassium dichromate ($K_2Cr_2O_7$), 10cm^3 of concentrated H_2SO_4, 10cm3 of orthophosphoric acid (30% H_3PO_4) and titrated with 0.5M Ferrous ammonium sulphate; ($Fe(NH_4)_2(SO_4).6H_2O$) using Ferroin indicator. The same was repeated to the control samples.

Table 1: Mean physico-chemical properties of the tannery sludge

Parameters	Tannery sludge	Safe limits
pH	8.19 ± 0.18	-
Electrical conductivity (mS/cm)	13.46 ± 8.81	-
Organic carbon (%)	12.67 ± 2.89	-
Organic matter (%)	21.91 ± 4.99	-
Nickel (mg/kg)	28.10 ± 4.09	420
Lead (mg/kg)	265.27 ± 27.40	300
Cadmium (mg/kg)	5.48 ± 0.48	39
Chromium (mg/kg)	16032.30±121.87	1200

Limits source: USEPA (1994)

Heavy metals analysis

Soil and sludge samples (1g) were digested with a mixture of HCl and HNO_3 in a 3:1 ratio and the total concentration of heavy metals in soil and were determined using Atomic Absorption Spectrophotometer (Association of Analytical Chemist, 1995).

The harvested vegetable tissues were measured one gram separately into porcelain crucibles. The crucibles were placed in a muffle furnace and heated to 500^0C for 8 hours. The completely ashed (clean white ash) sample was allowed to cool and then removed from the furnace. The ash was dissolved with 5cm^3 of 6M hydrochloric acid, warmed and then filtered through number 42 Whatman filter paper into 100cm^3 volumetric flask. The crucible was washed as well as the filter paper several times and the solution was then made up to the mark. The procedure was repeated to all the samples including the control sample. The average values of all the properties analyzed were summarized in Tables.

RESULTS AND DISCUSSION

The selected physicochemical properties of the soil used in the study are shown in Table 1. The soil pH was slightly acidic (6.67 ±0.08), Electrical conductivity (0.44 ± 0.66mS/cm) and Organic matter (0.90 ± 0.23%) were low indicating the soil is sterile to agricultural activities. While the tannery sludge was alkaline in nature (8.19 ± 0.18), Electrical conductivity (13.46 ± 8.81mS/cm), organic matter (21.91 ± 4.99%) and heavy metals were observed high in the tannery sludge. The high EC and OM indicated that the sludge have enough nutrients to support plants (George & John, 2013). Organic matter contributes to the structural stability of the soil and resistance to erosion (Ortiz &Alcanic, 2006) as well as improve soil aeration, water holding capacity and aggregate stability of soil (Gupta *et al*., 2010). This makes the sludge preferable to synthetic fertilizer and also revealed its potential in supporting agricultural practices.

The results of heavy metals were in the order Cr >Pb> Ni > Cd in the tannery sludge (Table 1). The concentrations showed only chromium (16032.30 ±121.87mg/kg) was above the allowable limit recommended for land application as specified by USEPA (1994). Thus indicates the tannery sludge was highly polluted with chromium. The relative higher concentration may be attributed to the fact that tannery industries use chromium salts as the major tanning agents. This confirmed the report of Akan *et al*. (2009) which stated the poor conditions of the tanneries' treatment plants, and the report of Garba & Mohammed, (2017) that tannery sludge as the source of chromium contamination of agricultural farms soil of Challawa and its environs.

Table 2: Mean physico-chemical properties of unamended and amended tannery sludge soil

Parameters	Unamended soil	Amended sludge	Safe limits*
pH	6.67 ± 0,08	7.40 ± 0.20	-
Electrical conductivity (mS/cm)	0.44 ± 0.66	1.12 ± 0.27	-
Organic carbon (%)	0.57 ± 0.04	1.06 ± .28	-
Organic matter (%)	0.90 ± 0.23	1.83 ± 0.47	-
Nickel (mg/kg)	12.90 ± 0.07	19.40 ± 1.52	100
Lead (mg/kg)	44.80 ± 0.05	90.64 ± 66.81	1000
Cadmium (mg/kg)	1.90 ± 0.32	2.79 ± 1.61	3.0 - 6.0ˣ
Chromiu (mg/kg)	11.40 ± 0.55	1438.64 ± 37.45	200

Limit values source: FAO/WHOˣ; Habib*et al.,* (2016)*

Effects of Tannery sludge on soil properties and heavy metal concentrations

The result of the analysis revealed that the tannery sludge amendment increased the soil properties. (Table 2). The increase in the pH may be attributed to mineralization of carbon and the subsequent production of OH^- ions by ligand exchange as well as the introduction of basic cations, such as K^+, Ca^{2+}, and Mg^{2+} (Gupta *et al.,* 2010). Ogunkunle *et al.,* (2013) reported that, use of sludge for land application increased the soil pH besides increased uptake of plant nutrients.

The sludge neutralized the slightly acidic nature of the soil, whereas the organic matter and electrical conductivity (EC) both increased compared to the unamended soil (control). The trend may be related to the organic acids produced in the mineralization process (Habib *et al*., 2016). Also, the expressive increase in the conductivity may be due to the high salts content of tannery sludge, and the higher the EC the higher the salinity hazards. This indicates the treatment had affected the soil's electrical conductivity.

Also, significant increase in the concentration of heavy metals (Ni, Pb, Cr, and Cd) were observed in the amended soil compared to the control (unamended). It was found (as recorded in Table 2) that chromium was the most abundant metal in the amended soil (1438.64 ± 37.45mg/kg) followed by lead (90.64 ± 66.81mg/kg), then nickel (19.40 ± 1.52mg/kg) and cadmium (2.79 ± 1.61mg/kg) was the least. Thus, chromium with the highest concentration was the only metal above the recommended limit. Its higher concentration may be attributed to its concentration in the sludge (Garba & Mohammed, 2017). Higher concentration of heavy metals in soil is unsafe and may pose health risks associated with irregular heartbeat, stomach upset, kidney and liver damage (Paul *et al.*, 2012). Thus increase of heavy metals in the soil as a result of amendment is the major setback to the use of tannery sludge in agricultural land.

Effects of tannery sludge on heavy metal concentration in the vegetables

The concentration of heavy metals in *Amaranthus hybridus* and *Corchorus olitorius* increase (Table 3) compared to the control. The control recorded the lowest uptake of all the metals. This indicated that sludge enhanced heavy metal uptake by the vegetables.

Table 3: Mean heavy metal concentrations in vegetable tissues(mg/kg)

Metal	Sample	Amaranthushybridus			Corchorusolitorius			safe
		Root	Stem	Leaf	Root	Stem	Leaf	limits
Ni	Control	2.6 ± 0.02	1.6 ± 0.04	1.25 ± 0.08	13.00 ± 0.08	11.60 ± 0.01	12.73 ± 0.11	
	Amendment	6.15 ± 1.91	4.38 ± 1.36	8.46 ± 1.18	15.41 ± 1.47	14.10 ± 1.55	15.73 ± 1.17	10.00
Pb	Control	15.90 ± 0.25	27.10 ± 0.01	28.70 ± 0.98	43.20 ±0.23	32.00 ± 1.87	38.10 ± 0.02	
	Amendment	42.83 ± 9.63	50.36±20.83	51.91±15.97	60.41 ±9.97	60.37 ± 4.61	58.25 ± 5.56	2.00
Cd	Control	1.10 ± 0.42	1.00 ± 0.58	0.71 ± 0.13	1.30 ±0.85	0.80 ± 0.07	0.80 ± 0.21	
	Amendment	2.05 ± 0.56	1.73 ± 0.23	1.62 ± 0.46	1.99 ±0.85	0.99 ± 0.44	1.58 ± 0.41	0.02
Cr	Control	ND	ND	ND	14.22 ±0.21	3.51 ± 0.02	4.19 ± 0.07	
	Amendment	348.99±50.31	14.70±10.84	30.17±20.08	260.20±21.17	33.24±22.97	31.45±12.94	1.30

Safe limits source: Ruqia *et al.*, (2015) ND: not detected

The results revealed that the root of both vegetables contained higher concentrations of chromium, cadmium, and lead than the stem and leaf, except the concentration of nickel which showed higher values in the leaves of the vegetables. Many factors such as nature and degradability of organic matter, type of soil and pH may be attributed to the trends. Among the factors, pH is the most important characteristic for determining the availability of heavy metals (Naidu *et al*., 2003; Habib *et al*., 2016). It plays a significant role in the solubility of metals in soil solution (Erikson, 1989). Once the availability of the metal in the soil is low the uptake by the plant will below as well. This probably accounted for the low level observed in the control soil. However, Kabata – pendias and Pendias (1992) have reported that an increase in soil pH in some cases does not lead to a decrease in heavy metal availability.

Ekiyoyo & Akinola, (2006) reported this same trend of roots having a higher concentration of metals than shoot, while in some plant species there were higher concentrations in the shoots than the roots as observed in the case of nickel in this study.

The roots are indeed the primary sites for the absorption of water and minerals including heavy metals (Ekiyoyo & Akinola, 2006). The roots will thus contain more heavy metals load than the leaves and acts as a storage organ in this case. Chromium showed high concentration (348.99±50.31mg/kg) as compared to nickel, cadmium, and lead in the roots of the two vegetables with highest observed in *Amaranthus hybridus*, which may be attributed to factors such as organic chelator, presence of other metals and salinity. The large difference showed that the metal had a restriction of movement from the roots to other tissues, probably due to complexation of metals with sulfhydryl groups resulting into less translocation of the metals to the upper part of the vegetable which varies from one metal to another (Gupta *et al*., 2010).

In this study, chromium in the amended soil was probably in a trivalent form (Cr^{3+}), which is more stable and has low solubility and mobility (Ademir *et al.*, 2013; Garba & Mohammed, 2017), this makes it generally hardly available to vegetable shoots (stem and leaves). The mechanism of this process is probably due to the trivalent chromium affinity to form complexes and chelates with cell wall components (Luna *et al.,* 2014). The process limits Cr penetration into cells and its translocation to the shoots, these explain the low concentrations of the metal observed in the vegetable shoots.

Also, the result showed a greater proportion of nickel (15.73 ± 1.17mg/kg) and lead (51.91±15.97mg/kg) in the *Corchorus olitorius* and *Amaranthus hybridus* shoots respectively. The result indicated that *Corchorus olitorius* had a greater ability to transfer nickel and *Amaranthus hybridus* to transfer lead to the aerial parts. The study confirmed the report which stated that stem of leafy vegetables serves not only the function of metal transfer from roots to the leaf through its conducting tissues but also stores some quantities of the heavy metals (Rangnekar *et al.*, 2013). The concentrations, except for Nickel in *Amaranthus hybridus* tissues were above the World Health Organization recommended safe limits.

Table 4: Transfer factor of vegetable tissues for heavy metals.

Metal	*A. hybdridus*			*C. olitorius*		
	Root	Stem	Leaf	Root	Stem	Leaf
Nickel	0.32	0.23	0.44	0.79	0.73	0.81
Lead	0.47	0.56	0.57	0.67	0.67	0.64
Cadmium	0.73	0.62	0.58	0.71	0.35	0.57
Chromium	0.24	0.01	0.02	0.18	0.02	0.02

However, the transfer factors which are an index demonstrating the potential of the whole plant or its tissues to accumulate metal from the amended soil provide supporting values (Table 4). The values ranged from 0.01 – 0.81 in the vegetable tissues, which indicatedthatall the values were less than 1.00. The sequence accumulation in *Amaranthus hybridus* tissues is Cd >Pb> Ni > Cr, while the order in *Corchorus olitorius* is Ni > Cd >Pb> Cr. Thus indicates Cd and Ni are the most bioavailable in the tissues. Hence, since the values were less than 1.00, the vegetables are not hyper accumulator species. However, the danger in this observed characteristic isthat, these vegetables are readily consumed by human beings. Though human does not normally eat roots where the higher concentrations were observed, but are eaten by other animals which in turn are being fed by humans and invariably heavy metals contained in the roots and shoots can get into the food chain. Therefore, continuous consumption might be a great threat to human health.

CONCLUSION

The study clearly showed that the tannery sludge amendment affects soil properties such as pH, organic matter, electrical conductivity, and heavy metals concentration. It also revealed a higher concentration of chromium above the recommended limits in the amendment. However, low transfer factors of the metals were observed in the vegetable tissues. Hence, due to the dietary importance of the vegetables to humans, the inflow of sludge into agricultural lands need to be avoided. The study recommends the Government to enforce measures on the disposal of tannery sludge and its use for agricultural purposes. Further research should also be carried out to study the effects of heavy metals on the inhabitants whose agricultural land is being cultivated with tannery sludge.

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