



SPECIATION, MOBILITY AND POTENTIAL TOXICITY OF METALS (CR, CO, CU AND MN) IN SOIL SAMPLES FROM DUMPSITES IN KANO METROPOLIS

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ABSTRACT

Soil pollution by heavy metals is a global environmental problem. The need to keep the environment clean is increasingly becoming a global concern. Heavy metals are one group of pollutants that are widespread in our environment. They tend to persist indefinitely once they enter the soil. Their mobility and toxicity depend on their forms and oxidation states in the soil. This research presents the speciation, mobility and potential toxicity of Cr, Co, Cu and Mn in soils from some dumpsites in Kano, using a six-step sequential extraction method and AAS analysis. The results revealed the pH of the soils to range from 6.20-7.80. Percentage composition of clay and organic matter ranged from 6.00-16.58 % and 0.48- 4.30 % respectively. The results showed that the metals are bioavailable at different forms evident from their distribution pattern in the different fractions and therefore the soils should not be used for cultivation to avoid the metals getting into the food chain. The mobility factor values ranging between 14.38 % and 61.48 % for dry season; 15.63 % and 74.74 % for rainy season showed the metals to be very mobile especially Co, Cu, and Mn. This could lead to potential pollution of the agricultural produce and ground water in this area and hence have toxic effects on human lives. This result can serve as a guide to researchers on the appropriate method to reclaim the soils in this area based on the pattern of distribution of the metals in different geochemical phases.

Keywords: Heavy metals, speciation, mobility, mobility factor, Jakara River dumpsites

INTRODUCTION

Environmental pollution is a global nightmare, due to its hydra headed nature as its devastating effects are felt in every corner of the world. Environmental pollution came about as a result of industrial revolution, urbanization, advances in science and technology. Pollution can be seen as the introduction of various pollutants into the environment which cause discomfort to human beings and other living organisms, or that causes environmental destruction, and may come in different forms such as chemical substances. The ultimate fate of some of these pollutants, such as chromium, cobalt, lead, nickel, copper, manganese, mercury, cadmium; metalloids like arsenic, selenium and antimony, in the environment is a food for thought for everyone. Recently the World Health Organization, (WHO) attributed most of the global health challenges being observed today to long term exposure to environmental pollution (Kimani, 2011). This pollution leads to environmental imbalance which also results in other environmental problems including soil erosion which leads to flooding.

Pollution of soil, water channels and air in cities became severe as a result of industries, human and vehicular activities. The rise in agricultural wastes production, growth in the processing methods in the industries plus the types of compounds utilized, either natural or synthetic, have compounded environmental problems (ICEPR, 2012). Substantial amounts of heavy metals get into the environment through either pedogenic or anthropogenic processes. However, the heavy metals that are parts of the parent soil materials are not bioavailable and therefore have less effect on soil organisms (Adriano, 2001; Kunhikrishnan, 2011).

The toxic effects of a few trace inorganics have been demonstrated throughout history. Some effects of heavy metal toxicity are confusion, muscle pain and headaches. Prolong exposure to these metals is linked with abnormal development, different types of cancers, damage to the kidney and sometimes death (Adriano, 2001). The toxicity of heavy metals in soils depends on a number of factors like overall

concentration, chemical form, their binding states, their properties, environmental factors, as well as soil properties such as pH, and organic matter content (Lu *et al.*, 2003). The presence of heavy metals and metalloids in soil is transitory because soil is just one part of the natural pathways for movement of elements in the ecosystem. The oxidation states of these metals determine the type of chemical or physical reactions they undergo in the soil. This in turn determines their mobility, toxicity and transportation between the soil components (Violante *et al.*, 2010). They are sometimes retained in the soil through adsorption, precipitation or complexation reactions (KunhiKrishnan, 2011). However, the ultimate fate of these metals in the soil environment rest on both the soil characteristics and environmental factors (Kunhikrishnan, 2011). The negative effects of refuse dumpsites to the soil environment and the quality of air are well documented (Salami *et al.*, 2014; Abdourahamane *et al.*, 2015). Most of the refuse dumpsites along the Jakara River are used extensively by the locals as cheap and alternative manure for growing a number of vegetables and more without the necessary assessments. Previous studies have focused on toxicity and remediation of heavy metals in soil. This research is aimed at studying the mobility, distribution pattern and potential toxicity of some heavy metals once they enter the soil environment and assess their bioavailability so as to proffer an appropriate remediation technique for the soil reclamation.

MATERIALS AND METHODS

Description of the Study Area

Samples for the study were collected from dumpsites along Jakara river in Kano metropolis in January and June, 2021. Jakara river is located between latitude 12° 25' to 12° 40' N and longitude 8° 35' to 8° 45' E. Some of the people around this river are farmers, using the water from the river to irrigate their farmland, especially during the dry season.

Sample Collection and Preparation

All samples were collected based on established procedures (Preparation of soil sampling protocols: sampling techniques and strategies. EPA 600 R-92/128, (1992) and were stored in appropriate containers before analysis to avoid contamination. Thirteen (13) soil samples were collected at thirteen (13) different dumpsites along Jakara River, Kano together with control samples 50 m away from each dumpsite but on a higher slop. From each location, four samples were pulled in a square area of 5 m using an auger and composite samples made in the laboratory. Each of the composite soil samples including the control was divided into two portions and were each air dried and sieved to pass through a 2-mm sieve and stored in plastic bags at room temperature prior to laboratory analysis. One portion of the sieved sample was used for routine analysis (physico-chemical parameters) while the remaining portion was used for the extraction scheme.

Samples Analysis

All chemicals used were of analytical grade. Distilled / deionized water was used throughout. Cation exchange capacity (CEC) of the soil was obtained using the method described by Gillman and Sumpter, (1986). Electrical conductivity was carried out according to the method described by Kunhikrishnan, (2011). Moisture content by ISO 11465, (1993). Organic matter by Walkley- Black, (1934); particle size analysis was achieved by the hydrometer method (Day, 1965). The pH of the soil was measured using ISO 10390, (1994). The phosphate content was obtained by the method described by Udo *et al.*, (2009). Sulphate content was estimated by the method described by Pillai *et al.*, (2006). Total metal content was estimated by the method described by Nguyen *et al.*, (2010).

For the chemical speciation, a six-step sequential extraction procedure as described by Finzgar *et al.*, (2007), was adopted. The relative index of metal mobility calculated as "mobility factor" was calculated using the following equation

$$Mf = \frac{F1+F2+F3}{F1+F2+F3+F4+F5+F6} \times \frac{100}{1} \quad (1)$$

Mf = mobility factor, F1 = Soluble form, F2 = Exchangeable, F3 = Carbonate, F4 = Fe – MnO, F5 = Organic, F6 = Residual. F1, F2, F3, F4, F5 and F6 represent the percentage geochemical forms of the metals in the soils from the dumpsites.

Analysis of all the fractions was carried out using atomic absorption spectrophotometer (AAS). Buck Scientific 210 VGB.

All analyses were carried out in triplicate and the mean values reported.

RESULTS

The values of the selected soil properties varied from sample to sample. The pH values ranged from 6.2 to 7.8 which shows the soil to be slightly acidic in some cases and slightly basic in others. This values are comparable to those obtained by Obasi *et al.*, (2012) for similar study. The pH of the soil influences the mode of soil retention of metal because their adsorption is tagged to pH. Therefore, the pH values obtained could influence the bioavailability and uptake of these metals by plants and organisms in the soil. Electrical conductivity (EC) of the soils ranged between 340 and 6000 $\mu\text{S}/\text{cm}$. These high values may be the result of accumulated soluble salts in the soil. The results compare favorably with those reported in literature (Obasi *et al.*, 2012). The content of soil organic matter varied from 0.48 to 4.30 %. These values are in good agreement with those reported in literature (Finzgar *et al.*, 2007; Obasi *et al.*, 2012). Soil organic matter adsorbs metals through varied mechanisms such as ion exchange or complexation; therefore the values obtained in this study could have implications on the mobility of the metals. The PO_4^{3-} and SO_4^{2-} contents of the soils ranged from 23.0 mg/kg to 58.3 mg/kg, and 3.65 to 38.9 mg/kg respectively. Soil phosphates usually form sparingly soluble salts and may affect heavy metal bioavailability in the soil. These results also compare very well with those reported by Finzgar *et al.*, (2007).

The cation exchange capacity (CEC) of the soils range from 3.4 to 13.1 cmol/100g. CEC is responsible for the buffering capacity of the soil which could limit the leaching of nutrient cations (Yoo and James, 2002). According to Awode *et al.* (2008), the CEC of soil is related to the quality and nature of clay and organic matter contents because the former gives a permanent surface negative charge whilst the latter gives a pH dependent surface negative charge. Both the two scenario put limitation on metal mobility in soil. The mean moisture content which is related to the water retention capacity of the soil was relatively low, ranging between 0.200 to 2.881%. The low moisture contents of the samples from the study area are to be expected, due to the overall climatic predisposition of the area.

Table 1 (a): Soil properties of the samples

Parameters	Samples						
	A	B	C	D	E	F	G
pH	7.20	6.30	7.20	7.80	6.70	6.50	6.70
EC (Ms/cm)	1922	729	3511	6000	390	3869	1920
OM (%)	3.36	4.02	2.88	3.15	1.23	2.88	1.57
PO_4^{3-} (ppm)	36.6	42.1	38.5	27.9	29.2	48.4	29.1
MC (%)	0.80	0.81	1.01	0.20	0.60	0.81	1.01
Clay (%)	12.5	12.2	14.4	14.4	12.4	14.5	16.3
Silt (%)	12.6	18.6	12.7	16.6	0.70	14.7	6.97
Sand (%)	74.9	69.0	73.0	69.1	86.7	70.8	76.8
Cr (mg/kg)	144	170	190	131	184	176	150
Co (mg/kg)	62.2	49.1	49.2	50.0	49.0	50.0	52.1
Cu (mg/kg)	151	158	125	101	131	160	98.3
Mn (mg/kg)	190	133	176	284	76.3	275	113
SO_4^{2-} (ppm)	27.3	35.4	30.9	32.9	10.2	38.1	22.2
CEC (cmol/100g)	9.11	11.8	10.3	1.10	3.41	12.7	7.40

EC = Electrical conductivity; OM = Organic matter; MC = Moisture content; CEC = Cation exchange capacity

Table 1 (b): Soil properties of the samples

Parameters	Samples						
	H	I	J	K	L	M	N
pH	6.50	7.20	6.70	7.20	6.50	6.20	6.50
EC (Ms/cm)	1123	710	1050	340	1182	1169	2202
OM (%)	4.30	1.37	3.63	2.14	0.48	2.40	1.66
PO ₄ ³⁻ (ppm)	42.0	23.0	58.3	34.1	47.2	36.6	28.9
MC (%)	1.01	1.01	2.46	1.83	0.81	0.60	1.83
Clay (%)	14.5	16.5	12.9	16.6	13.0	12.5	6.00
Silt (%)	6.57	16.2	12.6	12.5	4.54	10.6	16.0
Sand (%)	73.9	66.9	74.5	70.9	82.5	77.0	78.0
Cr (mg/kg)	39.4	44.1	76.1	115	107	130	4.15
Co (mg/kg)	62.1	59.0	63.1	61.1	72.1	74.2	6.41
Cu (mg/kg)	66.2	70.1	147	120	133	141	3.98
Mn (mg/kg)	154	97.8	166	165	125	127	62.2
SO ₄ ²⁻ (ppm)	38.9	28.8	36.9	30.9	19.8	33.6	3.65
CEC (cmol/100g)	13.1	9.61	12.3	10.3	6.61	11.2	7.20

EC = Electrical conductivity; OM = Organic matter; MC = Moisture content; CEC = Cation exchange capacity

The texture of the soil samples also varied substantially from 6.00 to 16.48%. Soil composition is an important parameter in determining the level of soil contamination with heavy metals. According to literature, soil with high clay content aggregates high levels of heavy metals due to the binding capacity of the clay mineral. On the other hand, sandy soils which have little clay minerals have low sorption capacity and therefore weakly bound heavy metals, and this may lead to their contamination of ground and surface waters (Sheoran *et al.*, 2009).

Therefore, going by the clay contents of the soil samples studied, it may have a profound effect on the heavy metal distribution and mobility in the study area. The textural class of the soil samples was mainly sandy loam.

Heavy Metals in Soil Geochemical Phases

In an attempt to predict the form, mobility and bioavailability of the studied metals in the soil, a six-step sequential

extraction scheme was used. This sequential step reveals how the metals are distributed among the soil components and helps to predict the behavior of these metals in the soil (Maiz *et al.*, 2000). The heavy metals are partitioned among solid components of the soil via phases including water soluble, exchangeable, carbonate, oxides and hydroxides, and organics.

The results of the sequential extraction of Cr in the sample of waste soil and control soil for dry and rainy seasons are as presented in Figures 1 and 2 respectively. Total Cr content ranged from 4.15 – 189.90 mg/kg and 1.21 – 184.40 mg/kg for dry and rainy seasons respectively. The Cr content of most of the samples were above USEPA, (1986) limit of 100 mg/kg but below 750 mg/kg limit permissible by CCME, (1991) for domestic gardens, agricultural and residential areas.

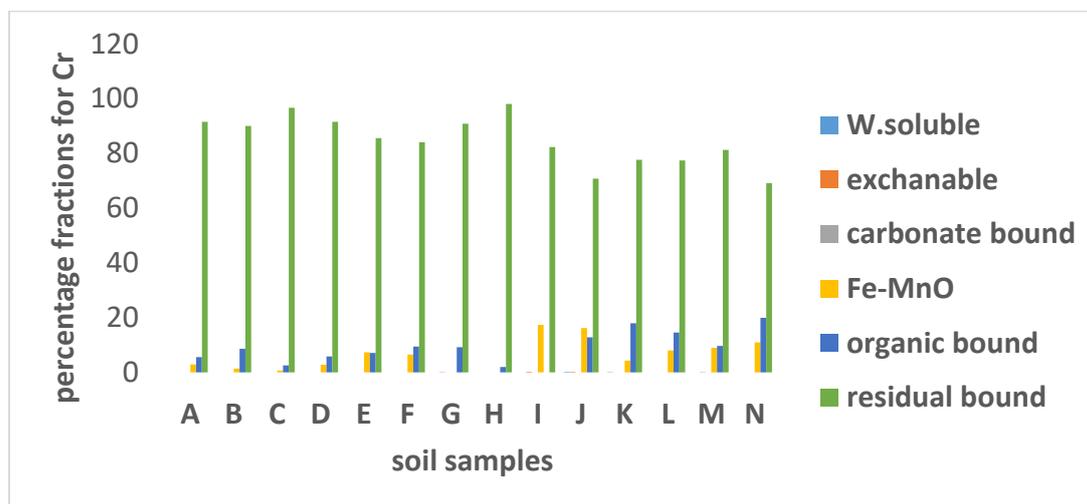


Figure1: Percentage fractions for Cr (dry season)

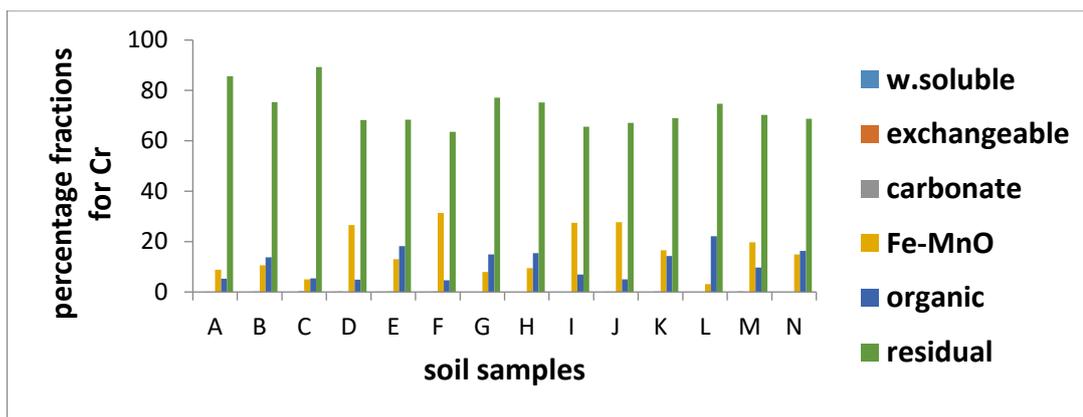


Figure 2: Percentage fractions for Cr (rainy season)
 A – abattoir, B- Akija, C- koki, D- Malafa, E- Kabo holdings 2, F- Jagwal, G- Airport read 1, H- Airport read 2, I- Gidan ruwa gama 3, J- Kaura goje, K- kabo holdings 1, L- gidan ruwa gama 2, M- gidan ruwa gama 1, N- control.

The fractionations of Co in the studied samples for dry and rainy seasons are presented in Figures 3 and 4 respectively. Total Co concentrations in the soils ranged between 6.41 to 74.20 mg/kg and 4.14 to 66.10 mg/kg for dry and rainy seasons respectively.

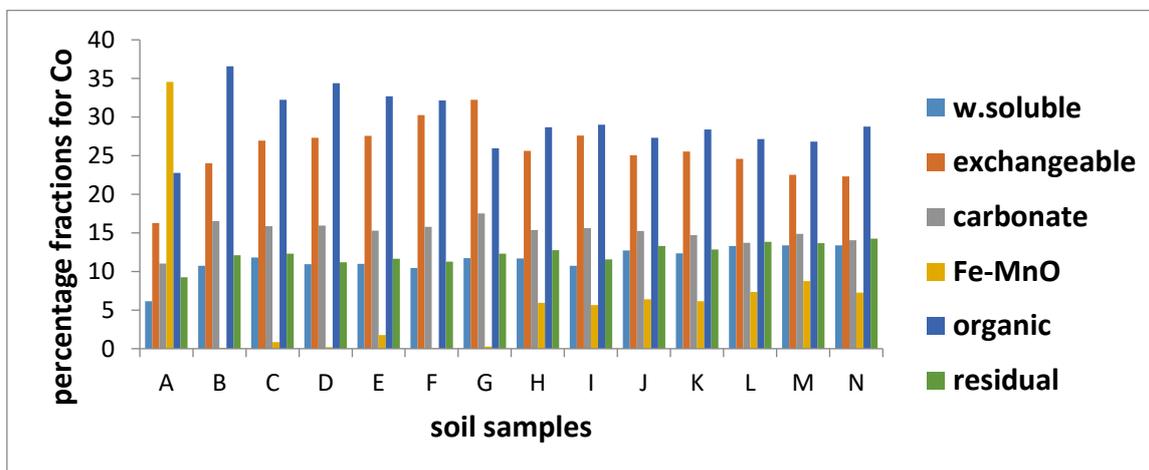


Figure 3: Percentage fractions for Co (dry season)

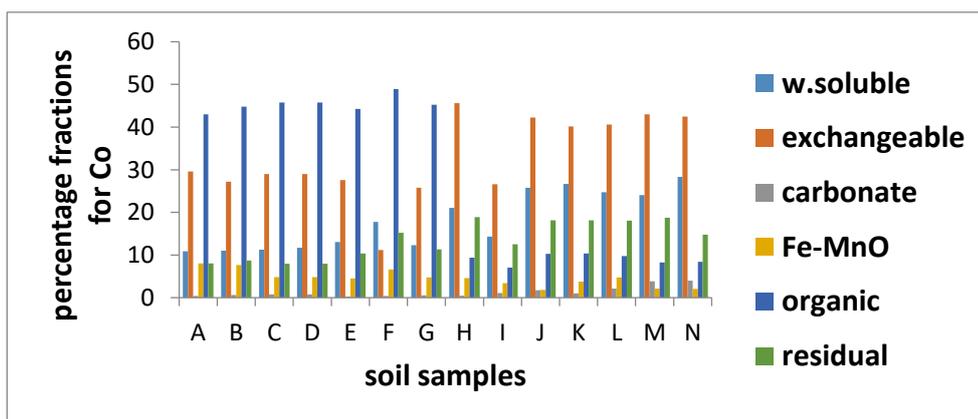


Figure 4: Percentage fractions for Co (rainy season)

The results of the sequential extraction of copper in the soil samples are shown in Figures 5 and 6 for dry and rainy seasons respectively. Total Cu concentrations in the soils ranged from 3.48 – 160.00 mg/kg for dry season and 3.71 – 154.16 mg/kg for rainy season. The concentrations of this metal in most of the studied samples were below the toxic limit of 250 mg/kg set by USEPA, (1986) for agricultural lands.

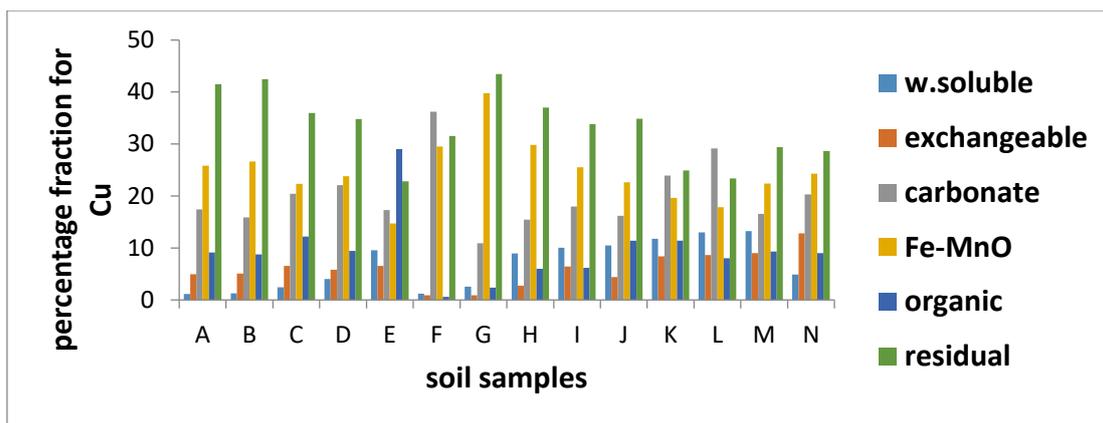


Figure 5: Percentage fractions for Cu (dry season)

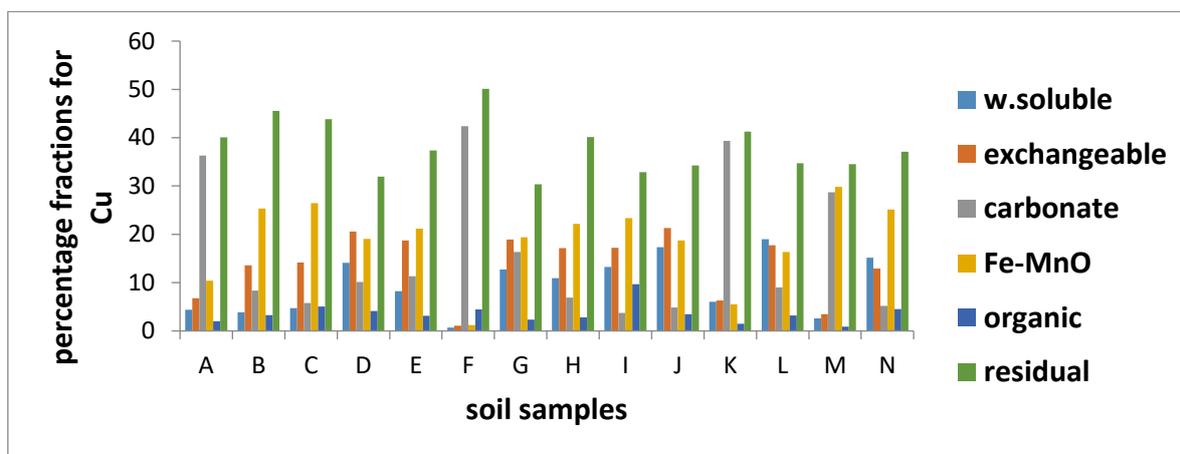


Figure 6: Percentage fractions for Cu (rainy season)

Fractionation of Mn in soil samples is shown in Figures 7 and 8 for dry and rainy seasons respectively. Total Mn concentrations in soils ranged from 62.20-284.11 mg/kg and 34.84-117.41 mg/kg for dry and rainy seasons respectively.

The concentrations of this metal in all the samples were within the tolerable limits (100 – 300 mg/kg) set by USEPA (1986) for agricultural lands, even though the dry season concentrations were almost out of the limit allowable.

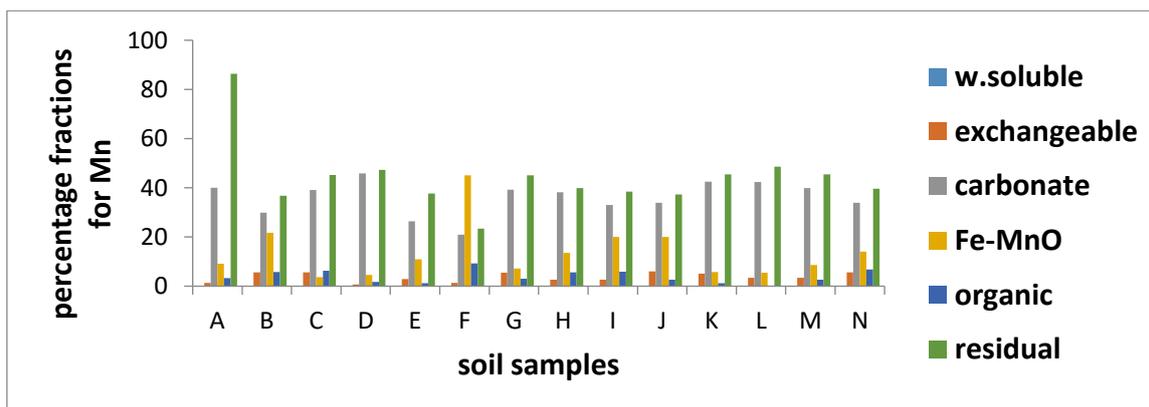


Figure 7: Percentage fractions for Mn (dry season)

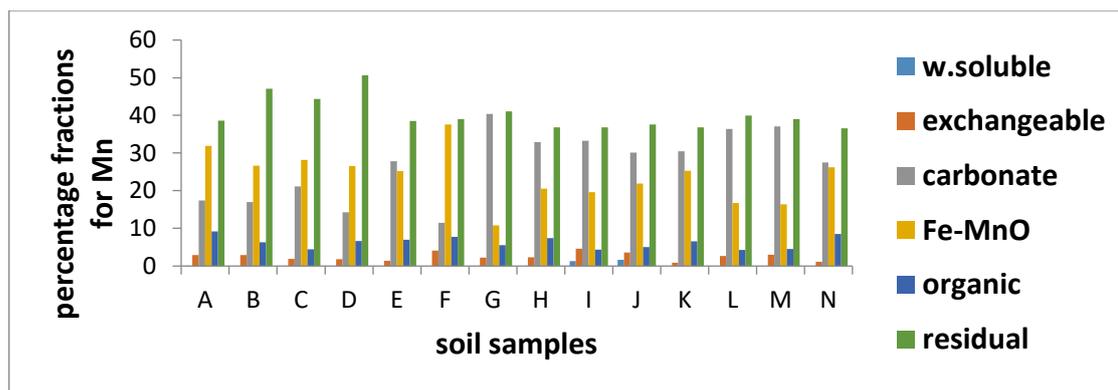


Figure 8: Percentage fraction for Mn (rainy season)

The mobility factor values assigned to the heavy metals studied are presented in Tables 2 and 3 for dry and rainy seasons respectively. It is observed that the relative mobility of the metals varied considerably.

Table 2 (a): Mobility factor (%) for dry season

	A	B	C	D	E	F	G
Cr	0.00	0.00	0.00	0.00	0.00	0.00	0.07
Co	33.44	51.25	54.61	54.21	53.85	56.51	61.46
Cu	23.50	22.18	29.48	31.91	33.41	38.36	14.38
Mn	41.61	35.71	44.87	46.43	29.23	22.28	44.64

Table 2 (b): Mobility factor (%) for dry season

	H	I	J	K	L	M	N
Cr	0.00	0.26	0.34	0.11	0.00	0.09	H
Co	52.64	53.95	53.01	52.58	51.62	50.79	47.74
Cu	27.12	34.46	31.12	44.09	50.78	38.89	37.99
Mn	40.92	35.65	39.90	47.63	43.83	43.25	39.54

Table 3 (a): Mobility (%) for raining season

	A	B	C	D	E	F	G
Cr	0.23	0.27	0.28	0.30	0.34	0.35	0.00
Co	40.91	38.85	41.26	41.49	40.84	29.25	38.67
Cu	47.49	25.83	24.64	44.89	38.29	44.17	48.00
Mn	20.33	19.96	23.06	16.08	29.25	15.63	42.61

Table 3 (b): Mobility (%) for raining season

	H	I	J	K	L	M	N
Cr	0.00	0.00	0.14	0.16	0.00	0.34	0.00
Co	67.13	41.94	69.75	67.79	67.42	70.88	74.74
Cu	34.90	34.12	43.53	51.69	45.78	34.73	33.22
Mn	35.18	39.21	35.42	31.32	39.01	40.13	28.69

DISCUSSION

The ultimate aim of extracting heavy metals with assorted reagents from soil composite is to determine the relative distribution and hence identify the form(s) that has high phytoavailability and water contamination potential. According to literature, heavy metals found in the soluble, exchangeable and organically bound fractions are highly phytoavailable (He *et al.*, 1992). The results of Cr fractionation showed strong association with the residual (69.15 – 98.04 %) and organic matter fractions which are in agreement with results reported in literature (Tokalioglu *et al.*, 2000; Alvarez *et al.*, 2002; Obasi *et al.*, 2012). Although the result was in contrast with that reported by Olubunmi and Olorunsola, (2010) who observed a strong association of Cr with the exchangeable fraction. It is evident from Figures 1 and 2 that the percentage associated with the water soluble,

exchangeable and carbonate fractions is very negligible in this study. This was also similar to those reported by Zhao *et al.*, (2006), where no Cr was detected in the first three fractions. High levels of Cr associated with the residual fraction compared to other fractions indicates lower mobility of the metal in the environment. However, its substantial association with the organic phase in this study, could make it available for plant uptake.

The low percentage of Co associated with the residual fractions (Figures 3 and 4), in this research indicates the possible availability for plant uptake. The pattern of metal distribution among fractions was similar in both dry and rainy seasons. The results for the fractionation of copper indicated that, Cu was mostly found in the residual phase. This agrees well with the results of Zhao *et al.*, (2006), but in contrast with the works reported by Nasr *et al.*, (2006) as well as Olubunmi

and Olorunsola, (2010) who reported more Cu association with the organic matter fraction. The percentage of Cu associated with residual fraction ranged between 23.35 and 43.47%, 30.33 and 50.15% for dry and rainy seasons respectively. From this result it can be deduced that the copper in this samples was less mobile due to the fact that less amounts were found in the mobile phases. Among non-residual fractions, the fraction associated with Fe – MnO occurred in the highest percentage with average values ranging between 14 – 39.75 % of the total Cu. The fractions of Cu associated with exchangeable and the water soluble could become available for plants uptake upon solubilization. Manganese was mostly associated with the residual and carbonate fractions. This agrees with the works of Ramirez *et al.*, (2005), who reported a strong association of Mn with the residual phase. However, Obasi *et al.*, (2012) reported a strong association of Mn with the Fe – MnO and organic matter fractions, due to the precipitation of amorphous hydrous oxides of manganese during aging of dumpsites. It is generally believed that metals in the residual fractions are tightly bound to silicate materials in the soil and not readily released into the environment and therefore unavailable for plant uptake. Going by the amount of Mn associated with the residual fraction in this research, it shows that the pollution level due to this metal is low. However, considering the amount of Mn found in the mobile phase (carbonate) for the samples, it could be suggested that Mn could potentially be bioavailable for plants uptake.

The magnitude of Mf values are indicators of metal lability and bioavailability in soils. From Tables 2 (a), 2 (b), 3 (a) and 3 (b) of all the metals studied, Cr had the lowest Mf values which indicates low mobility and least bioavailable metal in this study. According to Ramirez, (2005), metals originating from anthropogenic sources are usually found in the water soluble fraction. Hence it could be concluded that Cr in these dumpsites did not originate from anthropogenic sources. This implies that these dumpsite soils are not likely to be contaminated by Cr metal.

From the same Tables, the high Mf Values of Co, Cu and Mn is indicative of their relative mobility and biological availability. Going by Ulrich *et al.*, (1999), metals with Mf values above 10 % are deemed potentially available for plant uptake. The combination of low organic matter content and slightly acidic nature of the studied soils could be responsible for the bioavailability and mobility of these metals. The dumpsites are likely to be contaminated by Co, Cu and Mn.

CONCLUSION

The heavy metal pollution levels of the soils from dumpsites along river Jakara, Kano was assessed. The pH of the dumpsite soils ranged between slightly acidic and slightly basic in nature. The soils have low organic matter and clay contents as well as moderate cation exchange capacity. The metals were found to partition themselves between different fractions of the soil in different amounts. Their mobility also varied from metal to metal. The results also showed that these metals could be available for plant uptake in the soil with the exception of Cr. As a result, continued use of the dumpsites soils as amendment for growing edible vegetables and foodstuff as presently practiced pose health risk to the consumers. Due to the bioavailability of Co, Cu and Mn in the studied soils, phytoremediation technique would be an appropriate method for the soil reclamation, which boils down to the selection of the appropriate plants. But for Cr perhaps an alternative method such as chemical treatments which involve the application of agents to promote extraction of the metal may be required.

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