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Health Risk Assessment of Some Selected Heavy Metals in Agricultural Soils from Katsina State, North-Western Nigeria

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Authors' contributions

This work was carried out in collaboration among all authors. Author AIY designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors LS, AJA, JIB, AU and AN performed the statistical analysis and manage the analysis of the study. Authors AIY and IAY managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

This work contributes to the monitoring of heavy metal pollution of Agricultural soils, Katsina State Nigeria, using Atomic Absorption Spectrophotometry. The heavy metal contamination of the soil samples were analyzed based on the Geoaccumulation index (Igeo), enrichment factor (EF), contamination factor (CF), pollution load index (PLI) and potential ecological risk index (PERI). The health risks of the evaluated heavy metals were estimated using the Hazard Quotient (HQ) and Hazard Index (HI)) to evaluate the possible non-carcinogenic effect and the Incremental Lifetime

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Cancer Risk (ILCR) for the cancer risk to the population. The results of the study had revealed that in the soil samples all the evaluated heavy metals lie within acceptable limits as set by the regulatory agencies. The evaluated soil samples pollution indices had revealed that the I-geo values for the soil samples were within the range of unpolluted to moderate pollution. Also, the heavy metal enrichment factor (EF) value for the soil samples have indicated that only the heavy metal Fe showed significant enrichment, with the soil samples being moderately contaminated with Fe. The pollution load index (PLI) also indicated unpolluted to moderate pollution. With the potential ecological risk index (PERI) values presenting low ecological risks. The calculated non-cancer risk indices in both the children and adults population for all the heavy metals were less than 1. With the exception of the ILCR for the heavy metal Pb in children from Daura zone that was in limits that environmental and regulatory agencies considered as unacceptable risk, risk values for all the heavy metals falls within the range of the threshold of the safe limit and limits regarded as safe by the regulatory agencies (10⁻⁷ to 10⁻⁴). The results of pollution indices have indicated that the Agricultural soil samples have low contamination and low health risks by the heavy metals evaluated.

Keywords: Africa; pollution; environment; heavy metals; Katsina; Nigeria; cancer.

1. INTRODUCTION

"The soil is not only a substrate for plant growth or a reservoir to dispose off of unwanted materials, but also a transmitter of many pollutants to surface water, groundwater, atmosphere and food. It is a key part of the Earth system as it controls the hydrological, erosional, biological, and geochemical cycles" in soils [6-8].

"Literature indicates that studies have been conducted on pollution by heavy metals of some areas in Nigeria [9-12], but there is paucity of data on the heavy metal levels emanating from Agricultural soils in Katsina State Northwestern Nigeria and their possible effects on the quality of soil and human health. Therefore, it is important to investigate the level of heavy metals in Katsina agricultural soil to ascertain their heavy metal pollution levels".

2. MATERIALS AND METHODS

2.1 Study Area

"The study was carried out in Katsina State Nigeria. The area is located between latitude 12^{0} 15'N and longitude of 7^{0} 30'E in the North West Zone of Nigeria, with an area of $24,192$ km² (9,341 sq. meters)" [13]. "The rainy season begins in April and ends in October, while the dry season starts in November and ends in March. The average annual rainfall, temperature, and relative humidity of Katsina State are 1,312 mm, 27.3ºC and 50.2%, respectively. Like most alluvial soils, the soil in Katsina State is the flood plain type and is characterized by considerable variations. The soils are of two main types, which

are soils with little hazards and soils with good water holding capacity" [13]. "Katsina has been divided into three agro-ecological zones (Guinea Savannah; Sudan Savannah; Sub-Sahel Savannah), with farmers in the state engaged in the production of horticultural crops, such as Maize, Sorghum, Millet, Rice, Beans, Soybeans, Cotton, Cassava, Groundnut, Sweet Potatoes, vegetables and fodder crops" [13].

2.2 Soil Sampling

The study was conducted within catchment areas that were chosen based on volume of agricultural activity [13], located within the 3 senatorial zones (Katsina, Daura and Funtua) that constitute the state. Fifty-five soil samples (Katsina zone: 15 soil samples; Daura zone: 15 soil samples; Funtua zone: 25 soil samples) were collected from 0-20 cm depths (plough layer) of cultivated farmland with a hand auger from the designated sampling areas. Five samples were collected randomly from each location. The distance from one sampling point to another was approximately 50 m at each location. The collected five samples from each location were mixed and about 250- 300 g of the soil was sampled and put into a plastic container in accordance with the method of Syed et al. [14]. The samples were properly labeled and were taken to the laboratory for analysis.

2.3 Chemical Analysis of Soil Samples

After drying, sieving and digestion of the soil samples using mixed acid $(HCI-HNO₃)$, the concentrations of the heavy metals were measured using an atomic absorption spectrometer (AA210RAP BUCK Atomic Absorption Spectrometer flame emission spectrometer filter GLA-4B Graphite furnace, East Norwalk USA) according to standard methods [15] and the results were given in part per million (ppm).

2.4 Assessment of Soil Samples Contamination Status

The heavy metal contamination of the soil samples were analyzed based on the Geoaccumulation index (Igeo), enrichment factor (EF), contamination factor (CF), pollution load index (PLI) and potential ecological risk index $(PERI)$. The geo accumulation index, $Iqeo$, is used to indicate the enrichment of metals above the baseline concentrations, where

$$
I-geo = \log_2 / (Cn / 1.5Bn)
$$
 (1)

The soil sample is classified as unpolluted if Igeo value < 0. With progressive contamination with increase in Igeo value ($0 < I$ geo < 1), ($1 < I$ geo < 2), $(2 < J$ geo < 3), and $(3 < J$ geo < 4) pointing to the soil sample being unpolluted, moderately polluted, and heavily polluted, respectively [16]. The enrichment factor (EF) give an estimate of the abundance of the heavy metals in the soil samples by comparism of their concentration with that of a reference metal [17] and is defined as follows:

$$
EF = (M/Fe)_{sample} / (M/Fe)_{Background}
$$
 (2)

Where EF is the enrichment factor, (M/Fe) sample is the ratio of metal and Fe concentration of the sample and (M/Fe) background is the ratio of metals and Fe concentration of a background. Zhang and Liu [18] proposed that EF values between 0.5 and 1.5 indicate the metal is entirely from crustal or natural processes, whereas values greater than 1.5 suggest the possible anthropogenic impact in soils. Five contamination categories are reported on the basis of the enrichment factor [19]. EF <2 deficiency to minimal enrichment, EF = 2-5 moderate enrichment, $EF = 5-20$ significant enrichment, $EF = 5-20$ = 20-40 very high enrichment, EF>40 extremely high enrichment. The contamination factor (CF) is the concentration of a given element, Csample, against the average metal in the world surface rock, Cbackground. When the CF value is < 1 it indicates low level of contamination.

The pollution load index (PLI) is calculated as

$$
= (CF1 × CF2 × CF3 × ··· CFn) 1/ (3)
$$

Where, n is the number of metals. The PLI value of >1 suggests that the soil is polluted whilst PLI < 0 indicates unpolluted soil. To calculate the PERI for individual metals, the following Equation was used;

$$
Eri = Tri \times Cfi \tag{4}
$$

Where, Tri is the toxicity coefficient of each metal whose standard values are $Cd = 30$. Ni = 5, Pb = 5, $Cr = 2$, and $Zn = 1$, $Mn = 1$ [20,21] and Cfi is the contamination factor. To describe the ecological risk index the following order: Er < 40, low; $40 \leq$ Er < 80, moderate; $80 \leq$ Er < 160, considerable; $160 \leq E r < 320$, high; and $E r \geq 320$, very high.

2.5 Health Risk Assessment (HRA)

The USEPA [22] models for risk assessment (Hazard Quotient (HQ) and Hazard Index (HI)) were employed to evaluate the possible noncarcinogenic health risk in the children and adults population from exposure to the study heavy metals. In the current study two exposure routes were used (Dermal and inhalation).

2.5.1 Dermal route (Der)

The Hazard Quotient (HQ) from exposure to the heavy metals in the study through dermal contact were evaluated using the below equation

$$
\frac{CM \times SA \times SAF \times DAF \times EF \times ED}{BW \times AT \times RFD}
$$
 (5)

The Hazard Index (HI) was evaluated using the below equation

$$
HI = \sum HQ \tag{6}
$$

2.5.2 Inhalation route (Inh)

The Hazard Quotient (HQ) from exposure to the heavy metals in the study through inhalation were evaluated using the below equation.

$$
\frac{CM \times InhR \times EF \times ED}{BW \times AT \times RFD \times PET}
$$
 (7)

The Hazard Index (HI) was evaluated using the below equation

$$
HI = \sum HQ \tag{8}
$$

HQ is the hazard quotient due to heavy metals in soil; CM is the concentration of heavy metal; EF is the exposure frequency; ED is the exposure duration; BW is the body weight; AT is the average time; RFD is the reference dose of heavy metals; SA is the skin surface area; SAF is the soil adherence factor; DAF is the dermal absorption factor; InhR is the inhalation rate and PEF is the particulate emission factor and HQ is the hazard quotient due to heavy metal. $H1 > 1$ indicates a potential for adverse effect.

2.6 Cancer Risks from Exposure to the Soil Samples

The possibility of cancer risks in the studied samples through exposure to carcinogenic heavy metals were estimated using the Incremental Lifetime Cancer Risk (ILCR) [24].

$$
ILCR = CDI \times CSF
$$
 (9)

Where, CDI is chronic daily intake of chemical carcinogen, mg/kg BW/day which represents the lifetime average daily dose of exposure to the chemical carcinogen.

The US EPA ILCR was evaluated by the use of the cancer slope factor (CSF), which is the representative risk incurred through a lifelong average dose of 1 mg/kg BW/day and is heavy metal specific [25]. The following cancer slope factor for specific heavy metals were used; $Pb =$ 0.0085 mg/kg/day [26], Cd = 0.38 mg/kg/day [27], $Cr = 0.5$ mg/kg/day [28]. The ILCR valuation result in the sample give a picture of the possibility of an individual's lifetime cancer risks from exposure to the carcinogenic heavy metals' in the sample under study [29]. The standard of acceptability for cancer risk (ILCR) as set by the regulatory bodies was considered within the range of 10^{-6} to 10^{-4} [30].

The cumulative cancer risks in the samples as a result of multiple exposures to the metallic carcinogens in the soil samples was taken to be the sum of the individual heavy metal increment risks and calculated by the following equation [24].

$$
\Sigma 1n=ILCR_1+ILCR_2+\cdots+ILCR_n \tag{10}
$$

Where, $n = 1, 2, ..., n$ are the individual carcinogenic heavy metal.

Chart 1. Parameters used to estimate health risk in the study area [23]

Chart 2. Reference doses (RFD) (mg/kg/day) of heavy metals via dermal and inhalation exposure routes used for the non-carcinogenic health risk assessment

3. RESULTS AND DISCUSSION

3.1 Heavy Metal Concentrations in Samples

"Soil samples from the 3 senatorial zones of Katsina State, were analyzed in this study. As shown in Fig. 1, among the heavy metals evaluated, the highest concentration was observed for Fe (range: 22.3363-31.2606 ppm), with Cd having the lowest concentration (0.0138- 0.0258 ppm), while the heavy metal Ni was BDL (Below detection level) in all the soil samples. The concentration range of all the evaluated heavy metals in the agricultural soil samples in the study falls within the acceptable permissible limit of heavy metals in soils as set by the regulatory agencies" [31]. The reason may likely be attributed to the low background values of these metals and lower anthropogenic influences as reported in a study conducted on heavy metals in soils from Katsina State [32].

The lower mean values of heavy metals recorded in the present study as compared to the higher mean values reported for agricultural soils along some highways in Hamedan, west of Iran [7], roadside agricultural soils from India [6] and in soils reported in studies conducted in Tarnaveni city of Romania, Birjand city of Iran, Western Rajasthan, Faisalabad, Suxian county south China, and Thrace region of Turkey, may likely be that some of the compared sites are heavy industrial areas and some are exposed to mining activities [33-38]. Various studies have shown that industrial activities contribute to pollution burden [39-41].

Higher values than the representative values of the present studies have been reported for heavy metals in sediments samples from Katsina State Nigeria [42,43]. The lower value for heavy metal concentration in the study soil samples compared to values seen in sediment samples from Katsina State may not be unconnected to location and extent of agricultural activities, as the dams used for sediment sampling are located along the busiest highways in the State, and vehicular exhaust may contribute to heavy metal pollution burden [44], and the sites are used for rain fed and irrigation farming, compared to only rain fed farming in the soil sampling areas. With poultry manure application being implicated in contributing heavy metals to the pollution load [45-47], this may explain for the difference observed.

Fig. 1. Heavy metals concentration in agricultural soils from the zones

3.2 Correlation Analysis of Heavy Metals in Soil Samples

Correlation analysis results for the agricultural soil samples are shown in Table 1. As shown on the table, the correlations between the seven heavy metals are complex with relatively high correlations between Cd and Pb, with a correlation of 0.6602 and at a P<0.01 level and exhibiting a highly significant correlation. A positive correlation also existed between Mn and Cd with a correlation of 0.5509 with the heavy metals being significantly correlated at P<0.05. Also, a weak positive correlation existed between Mn and Zn, and Mn and Fe whose values were 0.3802, and 0.3647, respectively, with the level being not significant. The observed correlations between Mn and Pb, Mn and Cr, Zn and Pb, Zn and Cr, Zn and Cd, Fe and Cr, and Fe and Cr were all negative.

3.3 Geo-accumulation Index (Igeo) for Soil Samples

"Calculations of Geo-accumulation indices from all the soil samples from the 3 senatorial zones resulted in Igeo values of the evaluated heavy metals indicating unpolluted (class 0) by the respective heavy metals. Based on the mean values of the pollution levels of the heavy metals, the order was Fe>Cd>Pb>Zn>Cr>Mn for all the senatorial zones respectively (Table 2). The Igeo values in this study were similar to the Igeo values for heavy metals in soil samples reported previously from Katsina State Nigeria" [32]. But the results are lower when compared the results

reported by Sayadi et al. [34] for soil samples from Amir Abad of Birjand city Iran, for soil samples from Korle Lagoon area in Accra, Ghana [48] and the values reported by Mihaileanu et al. [33] in a study conducted in Romania. Higher values seen in some of the compared literature may not be unconnected to the anthropogenic activities that may contribute to metal build up in those areas.

3.4 Enrichment Factor (EF) for Soil Samples

The EF of all the evaluated heavy metals were all lower than 1 with the exception of the EF values of the heavy metal Fe, the results suggested a relatively low pollution level (Table 3). With the exception of the heavy metal Fe that shows significant enrichment throughout the sampling sites, all the other heavy metals showed deficiency to minimal enrichment. This was in contrast to the report of Sayadi et al. [34] and Mihaileanu et al. [33] of significant enrichment by all the heavy metals in the soil samples they evaluated. Most of the sampling sites used in the present study were located in sub urban and rural areas as compared to the urban and industrial settings of the sampling sites of the comparative studies, and it has been reported that Municipal areas receive load of noxious waste that may contribute to pollution burden than the sub-urban or rural areas [49,50,51].

The EF values in soil samples were also indicative of a likely low exposure to heavy metals in the population from the samples.

Table 1. Correlation matrix for heavy metals in agricultural soils

*Key: * Significantly correlated at P<0.05, **significantly correlated at P<0.01*

Table 2. Heavy metals geo-accumulation values for soil samples

l-geo										
Zone	Mn	Ζn	Pb	Cd	Fe	Cr				
Katsina	-3.1118	-2.4612	-1.8293	-0.9586	-0.0680	-2.4949				
Daura	-3.2219	-2.3645	-1.5645	-0.8447	0.1065	-2.7764				
Funtua	-3.0044	-2.2280	-1.7800	-0.8683	0.0906	-2.7903				

			Enrichment	Factor (EF)		
Zone	Mn	Zn	Pb	Cd	Fe	
Katsina	0.1988	0.3707	0.2199	0.0174	14.7316	0.2210
Daura	0.1191	0.3317	0.2351	0.2160	14.1051	0.0958
Funtua	0.2348	0.5526	0.1973	0.0126	14.8850	0.0935

Table 3. Enrichment factor values for soil samples

3.5 Contamination Factor (CF) for Soil Samples

"The computed contamination factors for the evaluated heavy metals in the agricultural soil samples are shown in Table 4. From the table, the relative distributions of the contamination factor among the soil samples were: Fe > Cd > $Pb > Zn > Cr > Mn$. For all the soil samples, the heavy metal Fe has a CF values range of 1.3537-1.7264, which indicated that the Agricultural soil samples were moderately contaminated with the heavy metal Fe. In contrast, the rest of the heavy metals exhibited low contamination. The soil samples CF values (Table 4) in this study were lower than the reported CF values for agricultural soil samples from Tarutia area of Tangail District, Bangladesh" [52]. The contamination factor for the soil samples exhibited a safe use (agricultural or otherwise) of the sampling sites by the population as far as the evaluated heavy metals were concerned.

3.6 Degree of Cotamination and Pollution Load INDEX (PLI) for Soil Samples

The value of PLI as shown in Table 5, indicated unpolluted to moderate pollution. However, Funtua senatorial zone displayed the highest PLI value (0.3517) while Daura senatorial zone

(0.2636) had the lowest PLI (Table 5). The degree of contamination for all the sites sampled had the soil sample from Daura zone (1.5817) exhibiting the lowest degree of contamination and the soil sample from Funtua zone (2.1099) the highest. The PLI values recorded in the present study were lower than the values reported for Tigris river and river Turaq [53,54],and in soil samples of river Niger flood plain at Jebba, Central Nigeria [55]. Soils that are influenced by high anthrogenic factors exhibit high PLI, hence the reason for the comparative PLI disparity.

3.7 Potential Ecological Risk Index for Soil Samples

The results suggested that the potential ecological risk of the tested heavy metals in the soil samples were likely caused by the heavy metal Cd (Table 6). Based on these calculations, the order of the single ratio of the tested heavy metals for the total potential ecological hazard were; Cd>Pb>Cr>Zn>Mn for all the 3 senatorial zones.

This analysis also showed that in the agricultural soil samples, the Eri of all the heavy metals were all below 40, which placed these metals at low ecological risk level (Table 6).

Table 4. Contamination factor for soil samples

PLI value > 1 is polluted, while PLI value < 1 indicates no pollution

Table 6. Evaluations Results of Bio-available Heavy Metal, Ecological Risk Index

Table 7. Heavy metal pollution risk in children from exposure through dermal contact to the agricultural soil samples

Table 8. Heavy metal pollution risk in adults from exposure through dermal contact to the agricultural soil samples

Table 9. Heavy metal pollution risk in children from exposure through inhalation to the agricultural soil samples

Table 10. Heavy metal pollution risk in adults from exposure through inhalation to the agricultural soil samples

Table 11. Heavy metal target incremental and cumulative incremental life time cancer risk in children from exposure to the agricultural soil samples

3.8 Health Risk Index from Exposure to the Soil Samples

"For the non-cancer effects for children and adults, dermal exposure to Cd and Cr are the major exposure routes, with the distribution pattern as: Cd > Cr > Fe > Mn > Pb> Zn, with the exposure being greater for children than for adults. Children are more susceptible to a given dose of toxin and are likely to inadvertently ingest significant quantities of metals because of their hand-to-mouth behavior, which has been widely regarded as a key metal exposure pathway for children" [56]. All the same, the calculated noncancer risk indices in both the children and adults population for all the heavy metals were less than 1, a pointer to the low risk for the population.

3.9 Cancer Risks from Exposure to the Soil Samples

The carcinogenic heavy metals, Pb, Cd and Cr were analyzed, and their corresponding carcinogenic risk was assessed from calculated daily dose (CDI) multiplied by the corresponding SLF. The risk of cancer for Children from exposure ranged from 4.9300E-07 to 1.0234E-03 for the metals evaluated, while the calculated exposure risk for cancer in Adult ranged from 4.2422E-08 to8.7550E-04, respectively. With the exception of the ILCR for the heavy metal Pb in children from Daura zone that was in limits that environmental and regulatory agencies considered as unacceptable risk. The risk values for all the heavy metals falls within the range of the threshold of the safe limit and limits regarded as safe by the regulatory agencies (10⁻⁷ to 10⁻⁴). However in the correlation analysis between the heavy metals in the soil samples an observation of a significant high correlation between Cd and Pb (all being metal carcinogens) was made, which may possibly increase the enormity of the cancer risk for the children population from Daura senatorial zone.

4. CONCLUSION

The results of the study had revealed that in the soil samples all the evaluated heavy metals lie within acceptable limits as set by the regulatory agencies. The evaluated soil samples pollution indices had revealed that the I-geo values for the samples were within the range of unpolluted (class 0) to moderate pollution (class 1). Also the heavy metal enrichment factor (EF) value have indicated that only Fe showed significant enrichment, with the soil samples being moderately contaminated with Fe. The samples pollution load index (PLI) also indicated unpolluted to moderate pollution. With the potential ecological risk index (PERI) values presenting low ecological risks. With the exception of the ILCR for the heavy metal Pb in children from Daura zone that was within a level of concern, low health risks by the heavy metals evaluated were recorded in the present study.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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