METAL POLLUTION AND HEALTH RISK ASSESSMENT OF SOIL WITHIN AN URBAN INDUSTRIAL ESTATE, SOUTHWEST NIGERIA.

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ABSTRACT

This paper assessed metal contamination associated with industrial activities around Agbara Industrial Estate, Southwest, Nigeria. Thirty soil samples collected within the estate were analysed using Inductively Coupled Plasma Atomic Emission Spectrophotometry ICP-AES. The results showed that the concentrations of Mo, Ag, Cr, Ni, Cu, Pb, Zn, As, Sb, Bi, Cr, Tl, Se, Hg and Cd were higher than their background values as contained in Average Crustal Values (ACV). An evaluation of the contamination factor showed that soil samples had suffered low to moderate contamination with respect to Cu, Mn, Mo, Ag, Cd, Cr and Bi, low to considerable contamination with respect to Pb, and Th and low to very high contamination with respect to Zn, Sc, Se and Sb. The degree of contamination ranged from 2.18 to 234.28 which indicated low to high degree of contamination. The toxic elements posed between low to high risks on the environment based on the potential ecological Risk Index (RI). The soil samples with the high risks were those closed to the industrial wastewater dumpsite. The total chronic hazard quotient index (THI) of oral exposure to soil contamination in the study area ranged between 2.81 and 134.89 for children, and 1.73 and 14.06 for adults. All the samples gave THI values above 1 which depicted great potential hazard for both young and old. The critical factor for THI in both adults and children for the study area were due to exposure to Pb, Mn, Zn and Cr.

Keywords: Soil, Risk, Metals, Environment, Assessment, Industrial Estate

INTRODUCTION

Contamination of soil is dominantly related to human activities such as industrial, agricultural, mining and metallurgical processes as well as waste disposal. Pollutants, such as, toxic elements can be retained by soils or moved to soil solution by biological and chemical mechanisms with potential impact on human health (Mulligan *et al.,* 2001, Giuliano *et al.*, 2007).

Human exposure to toxic metals has increased dramatically in the last 50 years as a result of an exponential increase in the use of heavy metals in industrial processes and products (Sekabira *et al.,* 2010). Solid and liquid wastes from these industries eventually get discharged into dumps or rivers from where they enter the food chain and consequently constitute unwanted danger to health and the environment. This is a cause for major concern which necessitates continuous monitoring of the environmental pollution potential of industrial waste dumps (Odukoya and Akande, 2015).

The present study involved an assessment of the elemental concentration of soils within Agbara Industrial Estate in Lagos. This was with a view to evaluating the possible environmental and health implications of the elements on the public.

The study area, Agbara Industrial Estate, is situated within longitudes 6.508°E to 6.517° E, and latitudes 3.067°N to 3.0762°N in Ogun, Southwestern Nigeria (Fig 1). The Industrial Estate hosts lots of manufacturing companies such as pharmaceuticals, chemicals, beverages, food and drinks, electric wire and cables, among others. The climate in the area is tropical with alternating wet and dry season. The mean annual rainfall in the region ranges from 50 to 2000 mm. during the wet season. The study area has two main types of vegetation, namely, tropical rain forest and guinea savanna vegetation which is made up of trees, grasses and occasionally shrubs (Iloeje, 1987).

The geomorphic feature of the study area bears a relationship with the geology. The topographical higher areas are underlain by more resistant sandstone formation while the lowland areas are largely underlain by low resistant silt/mud stones. The undulating topography trends NW-SE, probably resulted from differential weathering and erosion of the various rock types. The highest

and lowest points in the study area are 65.5 and 15 m above sea level respectively.

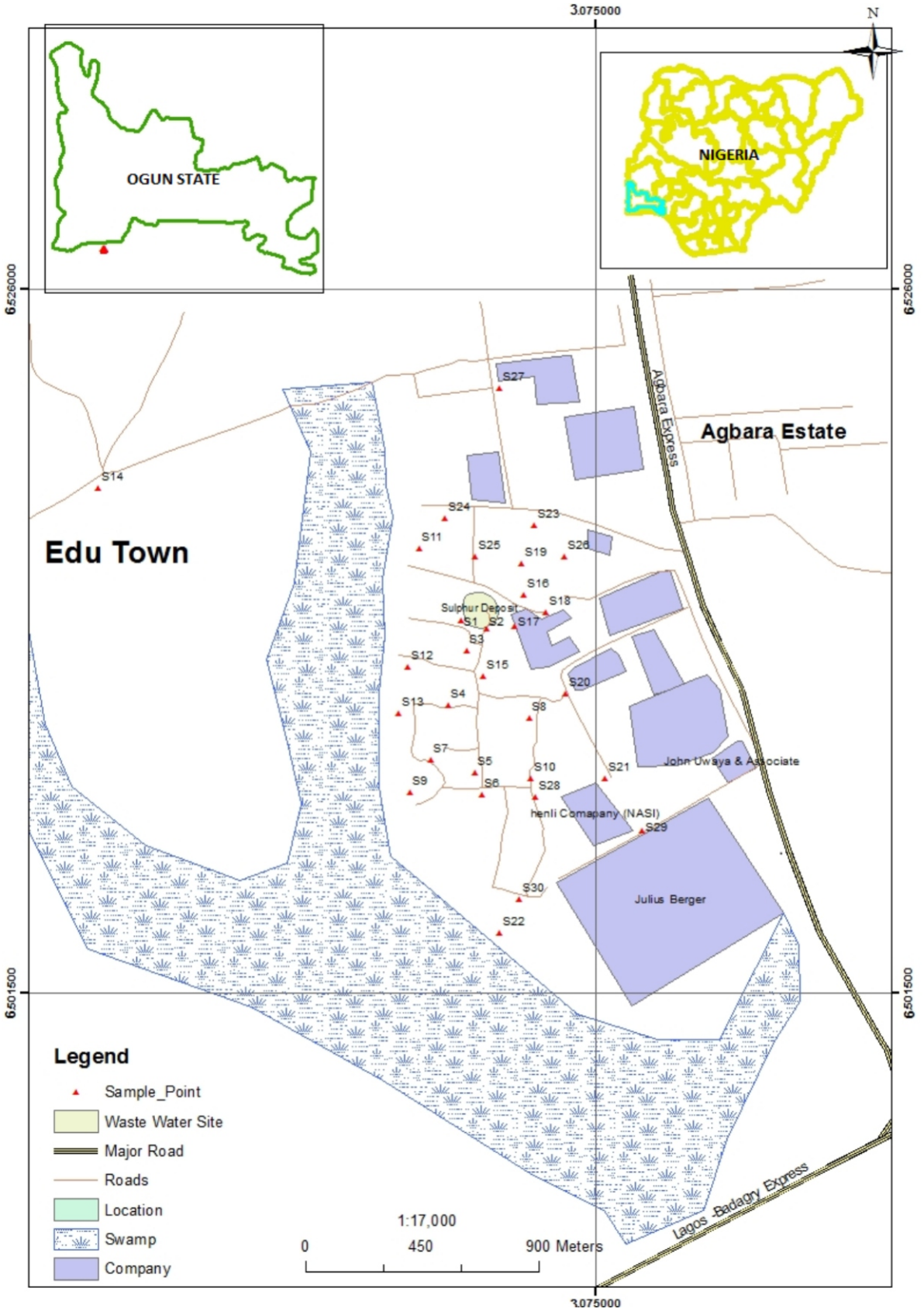

Fig.1. Map of the Study Area Showing the Sample Locations

MATERIALS AND METHODS

Thirty soil samples were collected around Agbara Industrial Estates. Composite samples (0-10 cm) were collected from the study area using an auger and stored in polyethylene bags. The soil samples were air-dried at room temperature (21-27°C) for seven days to obtain a constant weight. This was followed by mechanical grounding and sieving to obtain < 2 mm fraction. A fraction of the soil was drawn from the bulk soil (< 2 mm fraction) and reground to obtain < 200 μm fraction using a mortar and pestle. Chemical analysis was carried out at ACME Laboratories, East Vancouver, Canada. Analyses of metals were carried out using Inductively Coupled Plasma Atomic Emission Spectrophotometry (ICP-AES), after triacid total digestion. Accuracy of the analytical methods was monitored by repeated analysis of standard reference materials GSD-9 and NIM-G together with batches of soil samples.

Evaluation of Data

Some quantitative indices were used to assess the heavy metal contamination and also to allow for easy comparison among the determined metals. These indices included Contamination Factor (CF) and Contamination Degree (CD), Ecological Risk Assessment ($E^i r$), Potential Ecological Risk factor known as Risk Index (RI) and Potential Health Risk Assessment (Sekabira *et al.*, 2003, Gong *et al.*, 2008, Rapant *et al.*, 2008, Zoynab *et al.*, 2008, Ogunkunle and Fatoba 2013, Lu *et al.*, 2014, Saha and Hossain, 2015). The trace elements that were below Average Crustal Values (ACV) (Taylor, 1964) were ignored and thus presumed to pose no environmental risk.

Contamination Factor and Contamination Degree

The contamination factor (CF) and the degree of contamination (CD) were used to determine the contamination status of soil in the study area. The formula for contamination factor and Degree of contamination are stated below;

$$CF = \frac{Concentration\ of\ Heavy\ metal\ in\ sediments}{Concentration\ of\ background)} \quad [1]$$

CD = Summation of all the contamination factor (CF) [2]

The terminology for describing the contamination factor (CF) and contamination degree (CD) (Håkanson, 1980) are shown in Table 1.

Table 1. Classification of Contamination Indices

Contamination Factor		Contamination Degree	
Value	Interpretation	Value	Interpretation
Cf < 1	Low contamination factor	Cd<7	low degree of contamination
1 ≤ Cf **< 3**	Moderate contamination factor	7 ≤ Cd<14	moderate degree of contamination
3 ≤ Cf < 6	Considerable contamination factor	14 ≤ Cd<21	high degree of contamination
Cf ≥ 6	Very high contamination factor	Cd ≥ 21	very high degree of contamination

Ecological Risk Assessment ($E^i r$) and Potential Ecological Risk Factor (Risk Index (RI))

The purpose of ecological risk assessment is to assess ecological effects of human activities through scientifically credible evaluation (chemical assessment and individual bioassay) to protect and manage the environment.

The assessment of ecological risks of heavy metals in the soil samples was done using the Ecological Risk Assessment ($E^i r$) and Risk Index (RI) proposed by Hakanson (1980) and reported in Huang *et al.,* (1999) (Table 2) while grades of the

Environment by Potential Ecological Risk Index are presented in Table 3.

An ecological risk factor (E^ir) quantitatively expressed the potential ecological risk of a given contaminant (Hakanson, 1980) and the formula is

$$E^ir = Tr * Cf \quad [3]$$

where Tr is the toxic-response factor for a given substance (Table 3.) and Cf is the contamination factor.

Table 2. Grades of the Environment by Potential Ecological Risk Index (Ren *et al.,* 2007)

Grade	E^i_r value	Grade of ecological risk of single metal	RI value	Grade of potential ecological risk of the environment
A	$E^i_r < 5$	Low Risk (LR)	RI < 30	Low Risk (LR)
B	$5 \leq E^i_r < 10$	Moderate Risk (MR)	$30 \leq RI \leq 60$	Moderate Risk (MR)
C	$10 \leq E^i_r < 20$	Considerable Risk (CR)	$60 \leq RI \leq 120$	Considerable Risk (CR)
D	$20 \leq E^i_r < 40$	High Risk (HR)	$RI \geq 120$	Very High Risk (VHR)
E	$E^i_r \geq 40$	Very High Risk (VHR)		

Table 3. Pre-industrial Reference Level and Toxic- response Factor by Håkanson ((1980)

Elements	Cd	As	Ni	Cu	Pb	Cr	Zn
Pre-industrial reference level (kg/g)	1	15		50	7	90	175
Toxic-response factor	30	10	5	5	5	2	1

Assessment according to Environmental Risk Index

The calculation of the environmental risk index value (I_{ER}) for soil samples followed two steps. The first step was the calculation of the environmental risk quotient (Q_{ERI}) for each analysed chemical element or compound that exceeded the limit risk values. In the second step, the environmental risk index (I_{ER}) was determined from the summation of Q_{ERI} as expressed in the equations below.

$$Q_{ERi} = {}^{ACi}/_{RCi} - 1 \quad [4]$$

$$I_{ER} = \sum_{i=1}^{n} Q_{ERi} \quad [5]$$

where Q_{ERi} is environmental risk quotient of element i, which exceeds the concentration limit for risk values. ACi is analytical concentration of element i, RCi is the concentration limit of element i which is average crustal values (ACV) for this research, and I_{ER} is environmental risk index of the sample tested.

The Risk Index (RI) was originally introduced by Hakanson (1980) to assess the degree of heavy metal pollution in soils according to the toxicity of metals and the response of the environment. The Risk Index (RI) could evaluate ecological risk caused by toxic metals comprehensively. It is represented by the expression

RI = Summation of Q_{ER} [6]

Potential Health Risk Assessment

The risk assessment due to exposure to polluted metal in the soil was carried out to estimate the non-cancer toxicity (chronic) risk of people living and working in the study area.

Estimation of risk was calculated using USEPA exposure factors as determined by USEPA (1989) and HESP model (Veerkamp and Ten Berge, 1999). Daily Intake (DI) of soil ingested was determined using the equation below:

$$DI = \frac{C * IngR * EF * ED}{BW * AT} \quad [7]$$

where C = Mean heavy metal concentration, IngR = Soil ingestion rate, EF = exposure frequency (day/year), ED = exposure period (year), AT = average time for non-carcinogens and BW = body weight (kg) (USEPA, 1989, Grzetic and Ghariani, 2008).

Values of Daily Intake (DI) obtained are listed in Tables 4 and 5. Potential Health Risk Assessment gives the total dose entering the human body through oral ingestion of contaminated soil.

The systemic toxicity or non-carcinogenic hazard for a single element is expressed as the hazard quotient:

$$HQ = \frac{\mathbf{DI}}{\mathbf{Rfd}} \quad [8]$$

HQ = Non-cancer Hazard Quotient
where Rfd is chronic reference dose for the element (Table 4).
Total Chronic Hazard Index (THI) which is the summation of all the individual hazard quotients is represented by the equation

$$THI = \sum_{i=1}^{n} HQ \quad [9]$$

The accepted standard is 1.0 at which there will be no significant health hazard (Grzetic and Ghariani, 2008; Lai *et al.*, 2010). The probability of experiencing long-term health hazard increases with the increasing THI value (Wang *et al.*, 2012). According to Lemly (1996), THI = 1.1-10 indicate low to moderate hazard exposure while THI >10 implies high hazard exposure

Table 4. Some Toxicological Characteristics of Heavy Metals

Characteristics	Pb	Cu	Cr	Cd	Zn	Co	Mn	Ni	As
Oral minimal risk level MRL (Mg/Kg/day	-	0.01	-	0.002	-				
Oral chronic ref. dose. Rfd (Mg/Kg/day)	0.0035	0.04	0.003	0.001	0.3	0.02	0.046	0.02	0.003

Minimal Risk Level (MRL): an estimate of the daily human exposure to a hazardous substance that is likely to be without an appreciable risk of adverse non-cancer health effects over a specified route and duration of exposure.

Table 5. Risk Assessment Parameters and Values Used.

Parameters	Values used
Ingestion rate (IR) (mg/day)	0.0002 kg/day- child [ab]
	0.0001 kg/day -adult [ab]
Exposure frequency (day/yr)	350 days/year [c]
Exposure duration (yr)	6years - child [b]
	30 years - adult [b]
Average time for non-carcinogens (day/yr)	365 days/year [d]
Body weight (kg)	15 kg - child [ad]
	70 kg –adult [d]

[a]USEPA (1989); [b]Grzetic and Ghariani (2008); [c]Wang *et al.* (2012);[d] USEPA (2000)

RESULTS AND DISCUSSION

Soil Geochemistry

The statistical summary of analysed soil samples was presented in Table 6 and compared with the Average Crustal Values (ACV) (Taylor, 1964).

The results of the geochemical analysis of trace elements showed that the concentrations of nickel, cobalt, strontium, vanadium, lawrencium, barium, wallastonite, mercury, and tellurium were generally below their respective average crustal values (ACV).

On the other hand, levels (in ppm) of the

following elements had the ranges Mo (0.6-4), Ag (0.1-0.2), Mn (10-1850), As (0.5-4.6), Th (1.9-30.9), Sb (0.1-2.0), Pb (8.8 – 42.9), Zn (13 – 562), Cr (19 – 126), Cd (0.1 – 0.4), Se (0.5 – 1.5) and Sc (0.6-11.7). These elements occurred respectively at levels above Average Crustal Values (ACV) in 16.3%, 100%, 43.3%, 66.7%, 16.7%, 3.3%, 83.3%, 16.7%, 3.3%, 3.3%, 100% and 100% of the soil samples investigated. Their relative abundance as shown in Table 6 was in the order Mn > Zn > Ba > Ce > Sr > V > Cu > Pb > Co > Ni > Th > As > Ag.

Table 6. Statistical Summary of Analysed Trace Elements Compared with Crustal Average

Trace Elements	Min	Max	Mean	Std. Dev	Detection limit	ACV
Mo	0.6	4	1.5	0.6	0.1	1.5
Cu	7.5	59.1	23.4	11.6	o.1	5
Pb	8.8	42.9	18.4	7.3	0.1	12.5
Zn	13	562	65.8	109.7	1	70
Ag	0.1	0.2	0.2	0.03	0.1	0.07
Ni	0.7	11.5	7.4	2.8	0.1	75
Co	0.1	10	4.9	3	0.1	25
Mn	10	1850	770	574	1	950
As	0.5	4.6	2.3	0.9	0.5	1.8
Th	1.9	30.9	9.1	5.1	0.1	9.6
Sr	3	95	16.3	16.7	1	375
Cd	0.1	0.4	0.1	0.06	0.1	0.2
Sb	0.1	2.0	0.3	0.4	0.1	0.2
Bi	0.1	0.5	0.2	0.1	0.1	0.17
V	38	128	74	17.6	02	135
La	1	18	11	4.6	1	30
Cr	19	126	43.8	19.9	1	100
Ba	4	61	24.5	14.7	1	425
Hg	0.06	0.82	0.2	0.2	0.01	22
Sc	0.6	11.7	7.5	2.7	0.1	0.45
Tl	0.1	0.2	0.1	0.04	0.1	15
Se	0.5	1.5	0.63	0.21	0.5	0.05

Contamination Factor and Contamination Degree

Table 7 showed that the soil samples in the study area fell within low to moderate contamination factor with respect to Cu, Mn, Cr, Mo, Ag, As, Cd and Bi, low to considerable contamination factor with respect to Pb and Th and low to very high contamination factor with respect to Zn, Sc, Se and Sb.

Contamination degree (CD) ranged between 2.18 and 243.28 with an average of 111.9 and were within low to very high degree of contamination (Table 7).

Table 7. Results of Trace Elements Contamination Assessment and Ecological Risk in Soil

Indices	Major Elements		Trace Elements													
	Fe	S	Mo	Cu	Pb	Zn	Ag	Mn	As	Th	Cd	Bi	Cr	Sc	Se	Sb
EF	<1-1.30	22.56-144.56	<1-2.49	<1-1.0	<1-3.20	<1-7.50	<1-2.67	<1-1.82	<1-2.39	<1-3.01	<1-1.87	<1-2.75	<1-1.18	<1-24.27	<1-28.01	<1-1.34
Igeo	<1	0.36-6.68	<1	<1	<1-1.19	<1-2.42	<1	<1	<1	<1-1.1	<1	<1-1.18	<1	<1	<1-6.67	<1-2.74
CF	<1-1.07	1.92-154.23	0.4-2.67	0.14-1.07	0.7-3.43	0.19-8.03	1.43-2.86	0.01-1.95	0.28-2.56	0.2-3.22	0.5-2	0.59-2.94	0.19-1.26	<1-26	<1-30	0.5-10
Eir	-	-	-	0.7-5.35	3.5-17.5	0.19-8.03	-	-	2.8-25.6	-	15-60	-	0.38-2.52	-	-	-
CD	2.18-243-28															
RI	22.62-119.4															
I_{ER}	1.574-237.23															

Higher level of contamination was observed at locations very close to the site of waste water and this confirmed that soil acts as a sink for trace metals.

The sources of contamination that existed within Agbara Industrial Estate were from untreated waste water and effluents discharged from different manufacturing companies located within the estate. It is known that untreated or un-filtered emissions from most types of combustion and incineration will also carry different trace metals, such as Cr, Ni, Cu, Zn and Pb (Sanka *et al.*, 1995, Govil *et al.*, 2008, Dasaram *et al.*, 2010).

Ecological Risk Assessment (E^i_r) and Risk Index (RI)

The risk indices (E^i_r) of heavy metals for soil samples indicated that Cr fell within low risk (0.38-2.52) category for all the samples; Cu (0.7-5.35) and Zn (0.19-8.03) were within low to moderate risk, while Pb (3.5-17.5) fell within low to considerable risk category. As (2.8-25.6) and Cd (15-60) fell within low to high risk and considerable to very high risk categories respectively (Table 7). Potential ecological risk factor known as Risk Index (RI) ranged between 22.62 and 119.4 thus falling within the class of low to very high risk (Table 7).

Potential Health Risk Assessment

All the values of Daily Intake (DI) for both adult and children in this study area were greater than the recommended oral reference dose (RfDs) stipulated for the metals considered (USEPA, 2010) (Tables 4 and 8). Generally, the DI of the investigated heavy metals for children and adults daily ingestion show the following trends Pb > Mn > Zn > Cr > Cu > Ni > Co > As > Cd (Table 8).

DI for all metals in children were higher than that of the adults, meaning that children could have higher doses of these heavy metals than adults if the soil is orally ingested.

As shown in Table 8, due to their high concentrations in soils or low RfD values, Pb, Cr, Zn and Mn posed relatively higher non-carcinogenic risks to people than the other four heavy metals. Characterization of the risk of individual heavy metals (HQ) in children and adults showed that values obtained for Cu, Cd, Co, As and Ni were less than 1. Hence, no risk was posed to the health of both children and adults by these metals but HQ in children and adults showed that Pb, Cr, Zn and Mn had the following respective values (2.71-124.7 and 0.97-44.54), (1.49-9.91 and 0.27-1.77), (0.15-6.63 and 0.027-1.18) and (0.05-9.49 and 0.009-1.7) for children and adults respectively thus portending greater toxic hazards of oral exposure (Table 8).

Considering the total chronic hazard quotient index (THI) of oral exposure to soil contamination in the study area by the populace, the THI ranged between 2.81 and 134.89 for children and 1.73 and 14.06 for adults. The total hazard quotients of Pb, Mn and Cr for children accounted for 77.2%, 11.59% and 10.3% of the Total Hazard Index (THI) value respectively (Fig 2). By contrast, the total percentage of the other heavy metals for THI value was only 2%.

Table 8. Non-carcinogenic Risks for each Metal and Exposure Pathway

Metals	Summary	DI		HQ	
		Child	Adult	Child	Adult
Pb	Min	0.01	0.003	2.71	0.97
	Max	0.44	0.16	124.7	44.54
	Mean	0.09	0.03	26.31	9.4
Cu	Min	0.002	0.0003	0.044	0.008
	Max	0.014	0.0025	0.35	0.06
	Mean	0.006	0.001	0.14	0.025
Cr	Min	0.004	0.0008	1.49	0.27
	Max	0.029	0.005	9.91	1.77
	Mean	0.01	0.002	3.45	0.62
Cd	Min	0.00002	0.000004	0.024	0.004
	Max	0.00009	0.00002	0.094	0.017
	Mean	0.00002	0.000004	0.023	0.004
Zn	Min	0.003	0.0005	0.15	0.027
	Max	0.133	0.024	6.63	1.18
	Mean	0.016	0.003	0.77	0.14
Co	Min	0.00002	0.000002	0.001	0.0002
	Max	0.0024	0.0004	0.12	0.02
	Mean	0.001	0.0002	0.058	0.01
Mn	Min	0.0024	0.0004	0.05	0.0092
	Max	0.44	0.078	9.49	1.7
	Mean	0.182	0.032	3.95	0.71
Ni	Min	0.0002	0.00003	0.001	0.0002
	Max	0.003	0.0005	0.02	0.003
	Mean	0.002	0.0003	0.012	0.002
As	Min	0.0001	0.00002	0.039	0.007
	Max	0.0011	0.0002	0.361	0.065
	Mean	0.0005	0.00005	0.18	0.03
THI (Child)	2.81-134.89				
THI (Adult)	1.73-14.06				

In adult, total hazard quotients of Pb, Mn and Zn accounted for 90.9%, 6.87% and 1.35% of the total hazard quotient respectively (Fig 3).

The average hazard quotients of heavy metals for the child and adult decreased in the order of Pb > Mn > Cr > As > Cu > Cd > Ni > Zn > Co and Pb > Mn > Zn > As > Cu > Cr > Co > Cd > Ni respectively.

According to USEPA (2010), if THI <1, the exposed individual is unlikely to experience obvious adverse health effect. On the contrary, if THI > 1, there is a chance that non-carcinogenic effect may occur with a probability which tends to increase as THI increases (Man *et al.*, 2010).

It is also clear from the results that exposure to Pb, Mn, Zn and Cr in the contaminated soil constituted a critical factor for the high values of the THI in both adults and children (Table 8 and Figs 2 and 3). Therefore, it can be concluded that the heavy metals in the soils of the study area potentially have high non carcinogenic risks to the public.

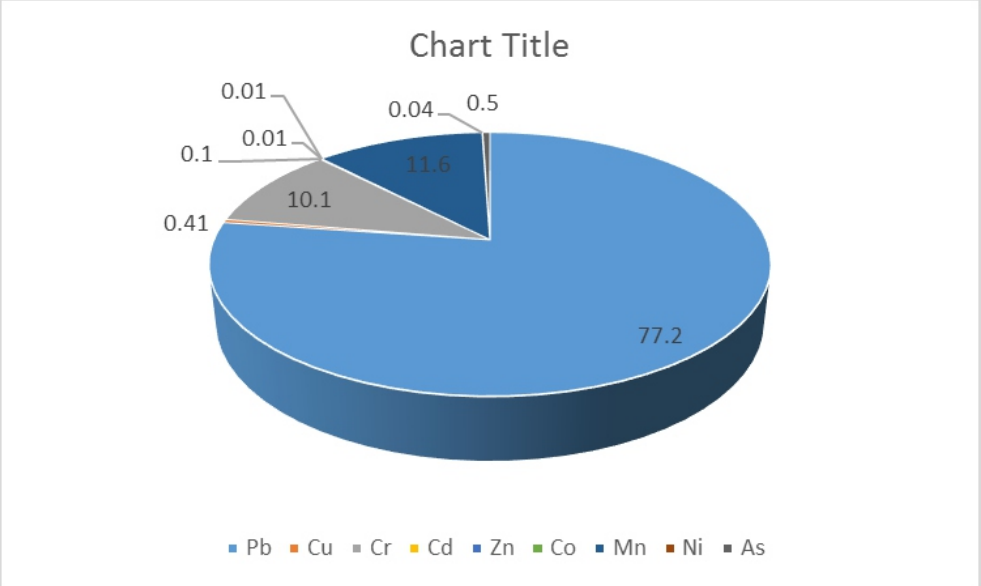

Fig 2. Average Hazard Quotient for Children

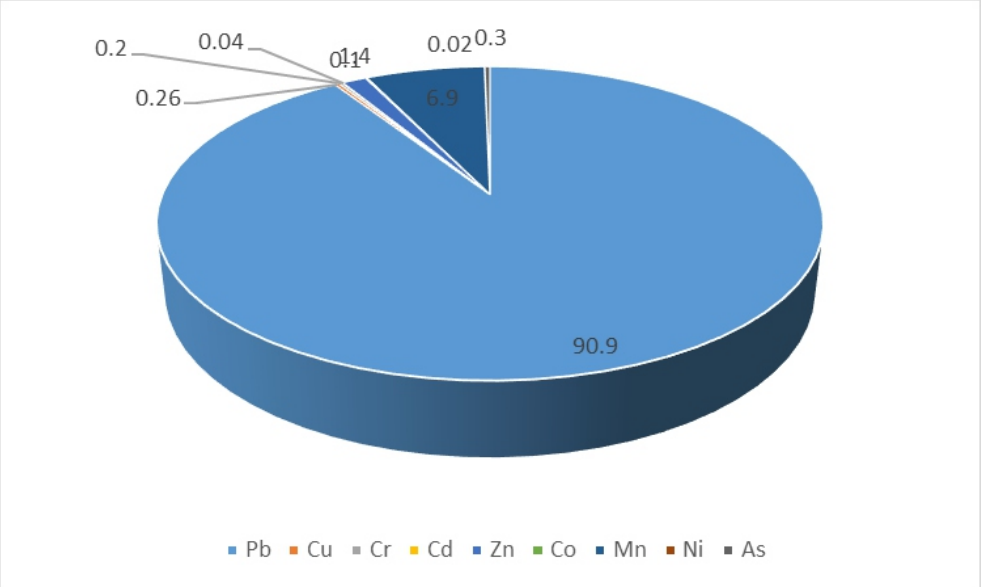

Fig 3. Average Hazard Quotient for Adults

CONCLUSION

Rapid development of industry during the past two decades is responsible for the significant increase in the amount of metal products consumed in the study area, and in turn, has led to serious environmental pollution. Through a systematic evaluation using several pollution indices and health risk assessment models, this study analysed the data set of pollution assessment in the study area and gave a comprehensive description of the overall pollution status of heavy metals in Agbara Industrial Estate soils and their exposure risks posed to the public health. According to pollution assessment, it was apparent that the soils of the study area were polluted by major and heavy metals in varying degrees with Fe, S, Cu, Mn, Cd, Cr, Pb, Ag, Mo, As, Bi, Zn, Sc, Se and Sb to varying

degrees posing low and high risk to the environment. The non-carcinogenic risk levels fell within very high risk to the public. The study area should be considered a priority control area for soil heavy metal pollution. In order to protect soil environment and human health, special attention should be paid to Pb, Mn, Zn, Cr, As, and S which have been identified in the study as the priority control components.

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