JOURNAL GEOLOGICAL SOCIETY OF INDIA
Vol.91, June 2018, pp.743-748

Assessment of Potentially Toxic Elements Pollution and Human Health Risk in Soil of Ilesha Gold Mining Site, Southwest Nigeria

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ABSTRACT

The paper presents results of research to determine the health risk of potentially toxic elements (PTEs) in soils of gold mining area in Ilesha, Southwestern Nigeria. Eighteen top soil samples were collected and analysed for PTEs using ICPMS. The pollution level in soils were assessed using pollution load index (PLI) and potential ecological risk index (PERI). The human health risk was assessed using hazard index (HI), non-carcinogenic risk index and carcinogenic risk index. All the PTEs were higher than crustal average values except Fe and follow the order Mn > Ba > Cr > Zn > Cu > Ni > Pb > Co >Fe > As > Cd. The soil samples can be classified as unpolluted to moderately polluted (0.18-1.23) and low risk (15.28-94.24) based on PLI and PERI respectively. The values of hazard Index (HI) calculated for child and adult population for all the pathways were <1 and pose no obvious non carcinogenic risk to the population except for ingestion pathway where 33% of the samples showed value >1 in child population. As, Co, Cd and Pb constituted high cancer risk in the study area with child at higher risk than adult. The Total Cancer Risk (TCR) in the study area based on all the pathways ranges between 0.19 -3.86 and 0.18-3.58 for child and adult respectively and the ingestion route seems to be the major contributor to excess lifetime cancer risk followed by the dermal pathway.

INTRODUCTION

Gold mining operations started in Ilesha area, southwest, Nigeria in early 1950s and since mid 1990s there has been continued illegal mining by artisanal miners in Itagunmodi, Ibodi, Igun, Araromi, Osu and other areas. 80% to 90% of small-scale mining is commonly illegal and artisanal in nature involving about 10 to 20 million people that has produced about 12% of the world's gold (330 tonnes) annually (UNEP, 2008). Poor performance of large scale mining sector is responsible for the ASGM (Ikenna et al., 2015).

The illegal ASGM has caused avoidable environmental pollution as a result of crude or rudimentary methods employed in extraction and processing of minerals. Mining activity has been linked to contamination of soil, water, sediments and plants which later on pose health hazards to man and livestock (Khan et al., 2008; Moreno-Jimenez et al., 2009; Li et al., 2014, Obiora et al., 2016).

Mining wastes from extraction of minerals and tailings are the main sources of toxic metals (Ji et al., 2013). The Agency for Toxic Substances and Diseases Registry (ATSDR) has categorised as potentially toxic elements (PTEs), Co, Cr, Zn, Pb, Ni, Ba, S, As, Pb and Cu due to their high toxicity and persistence in the environment and therefore are expected to be monitored worldwide (Zhao et al., 2014 and Edna et al., 2017).

Humans are exposed to these toxic metals through different pathways including ingestion of soils, plants and dusts directly, dust adhering and inhalation (Carla et al., 2014).

The present study attempts to evaluate the concentration of these toxic metals in the soil of Ilesha gold mining region and the human health risks arising from exposure to toxic metals.

METHODOLOGY

Study Area

Ilesha is the most important settlement in Ijesha region of Southwestern Nigeria. The study area lies within latitude 7º 30' and 7º 35'N and longitude 4 º 30´ and 4 º 34'E **(**Fig. 1**)**. The study area covers an area of 73.6 square kilometers. The population of Ilesha was 72,029 in 1952 when the first national census was carried out in Nigeria outside of Lagos. This increased to 165,822 in 1963 but decreased to 138,953 in 1991. At present the population of Ilesha is estimated at 210,141 (NPC, 2006). The climate in Ilesha area is characterised by high annual rainfall with the peak usually around July after which a break of 2-3 weeks of rainfall commonly referred to as August break is experienced. The wet season starts from April and early November and the dry season from November to March. The temperature ranges between 26°C to 33°C and the area represents a tropical rain forest. (Kayode 2009). The schist belts in the study area form a part of the basement complex of Nigeria and comprises biotite schist, gneiss and pegmatite. Biotite schist occur mainly in the northwestern part of the area and around Osu. They are closely associated with muscovite/sericite schists and amphibolite. Gneisses are classified into granite-gneiss, hornblende gneiss, amphibolite-muscovite gneiss and epidote gneiss (Elueze, 1977). Biotite-hornblende granite gneiss occurs sandwiched between two N-S trending quartzite ridges and quartz schists bands east of Ifewara (Ajayi and Adegoke, 1988).

Sampling and Analysis

Soil samples were collected systematically within all the communities where mining is practiced such as at Ibodi (IB), Itagunmodi (IT), Isua (IS), Okutu omo (OKU), Araromi Itagunmodi (AR), Ijana (IJ), Igbadae (IGB), Okepa (OK), Igun, Inkwe (IN) and Osu (Fig.1). 18 surface soil samples were taken at a depth between 0-15 cm after removal of superficial debris and vegetation. The samples were placed in polyethylene bags for transport to the laboratories. They were later dried in an oven at 40°C, disaggregated in a porcelain mortar, sieved (<2 mm), homogenized and pulverized to <170 µm in a pre-cleaned mechanical agate mill for chemical analysis. The geochemical analyses were carried out by the Acme Analytical Laboratories (Vancouver) Ltd., Canada, using Inductively Coupled Plasma, Mass Spectrometer (ICPMS). 0.5 g of soil samples from each site was leached in hot (95 °C) aqua regia ($HCl-HNO_3-H_2O$), and concentrations of 36 elements were determined by ICP-MS. Precision and accuracy were determined using analytical results of certified reference materials (standards C3 and G-2) and on duplicate samples in each analytical batch. The results were within the 95% confidence limits of the recommended values given for the certified

0016-7622/2018-91-6-743/$ 1.00 © GEOL. SOC. INDIA | DOI: 10.1007/s12594-018-0933-7

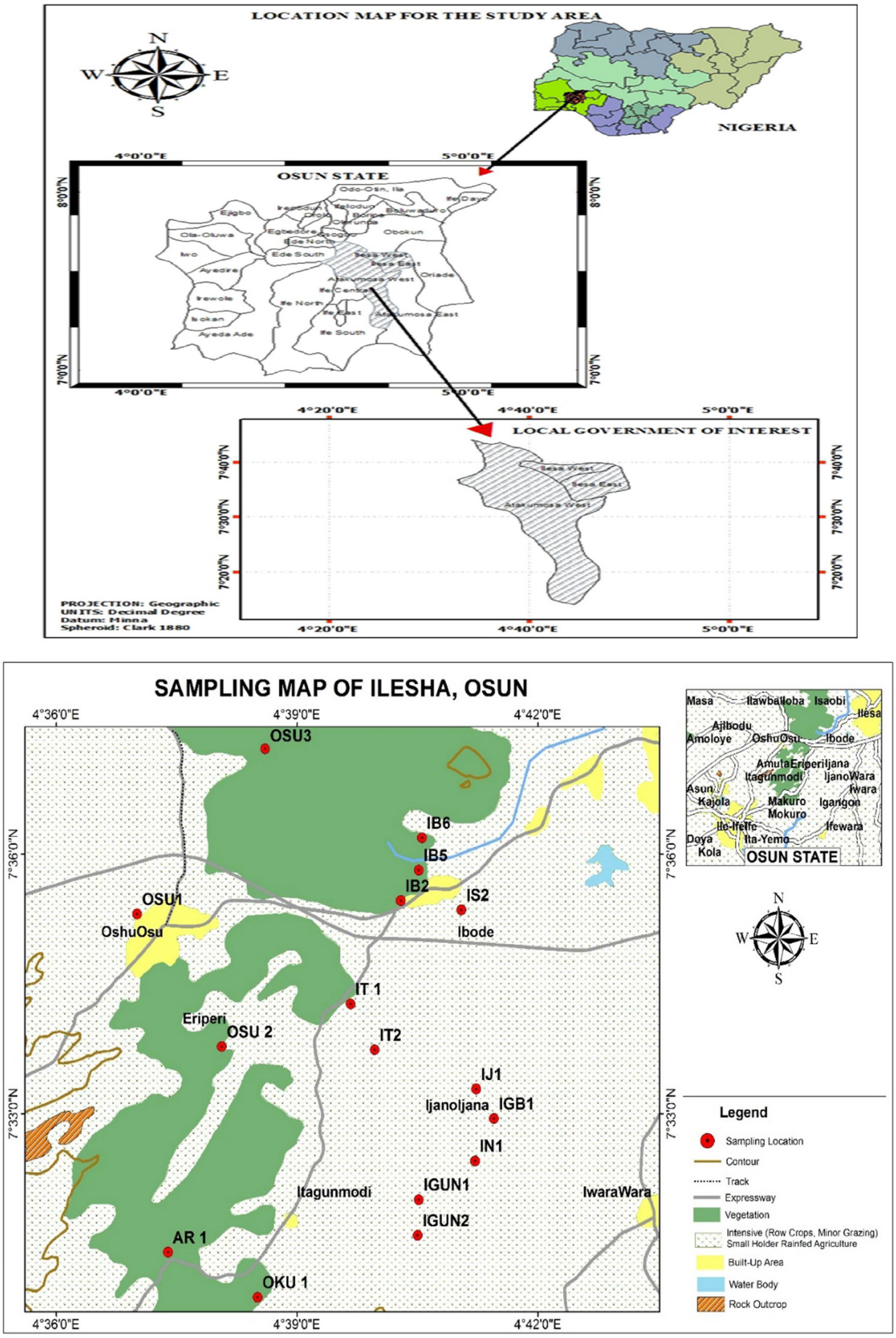

Fig.1. Map of the Study Area

materials. The Relative Standard Deviation (RSD) was between 5% and 10%.

Data Evaluation and Contamination Assessments

Statistical analysis to determine range, mean and standard deviation was done by using the Excel software (Microsoft 2013). Multivariate statistical analysis such as inter-elemental correlations and factor analysis were conducted using the SPSS software (version 16).

The level of contamination of soil samples based on some potentially toxic elements was determined using the pollution index (PI) and potential ecological risk (PER) paramenters (Wu et al., 2015, Li et al., 2014 and Xiao et al., 2015). The non-carcinogenic and carcinogenic risk of these toxic elements were also calculated using hazard quotient and hazard index parameters.

Pollution Index (PI)

Pollution Index is the ratio of element in the soil to the background concentration which is the concentration of the same element in the earth's crust.

$$PI = \frac{Cn}{Bn} \tag{1}$$

Classification of PI is presented in Table 1 (Wu et al., 2016).

Table 1. Classification of Pollution Index

$PI \leq 1$	Low Contamination
$1 < PI \leq 3$	Middle Contamination
$PI > 3$	High Contamination

In addition, to give an assessment of the overall pollution status for a sample, the integrated pollution load index (PLI) or the Nemerow integrated pollution index (NIPI) (Nemerow, 1985) can be employed (Tomlinson et al., 1980; Luo et al., 2012; Lu et al., 2014; Chen et al., 2015).

The PLI and NIPI can be calculated using

$$\text{NIPI} = (PI_1 \times PI_2 \times PI_3 \times \ldots \times PI_n)^{1/n} \quad (2)$$

According to (Zhang et al., 2011), the classification based on PLI and Nemerow integrated pollution index (NIPI) is given in Table 2.

Table 2. Classification Based on Pollution Index (PI) and Pollution Load Index (PLI) (Zhang et al., 2011)

PLI = 0	Background Concentration	NIPL = 0.7	Safe
0 < PLI = 1	Unpolluted	0.7 < NIPL = 1	Precaution
1 < PLI = 2	unpolluted to moderately polluted	1< NIPL = 2	Slight Pollution
2 < PLI = 3	Moderately polluted	2 < NIPL = 3	Moderate Pollution
3 < PLI = 4	Moderately to highly polluted	NIPL > 3	Heavy Pollution
4 < PLI = 5	Highly polluted	NIPL = 0.7	Safe
PLI > 5	Very highly polluted		

Ecological Risk Factor

An ecological risk factor ($E^i r$) quantitatively expresses the potential ecological risk of a given contaminant as suggested by Håkanson, (1980) is expressed as

$$E^i r = Tr * Cf \quad (3)$$

where Tr is the toxic-response factor for a given substance and Cf is the contamination factor. The Tr values of heavy metals given by Håkanson (1980) is given in Table 3. The terminologies used to describe the risk factor are listed in Table 4.

Human Health Risks

Carcinogenic and non-carcinogenic risks to human health can be evaluated through three exposure pathways including oral ingestion, inhalation and dermal exposure according to USEPA (1989) and (2011).

Hazard quotient (HQ) was used to assess non-carcinogenic hazards of all potentially toxic elements and exposure routes for children and adults (Li et al., 2014). When HQ <1, it means that there would not be any health hazard effects as a result of exposure; HQ > 1 indicates probability of health hazard effects due to exposure.

The average daily doses (ADDs) (mg/kg day) of potentially Toxic Elements (PTEs) through ingestion (ADDing), dermal absorption (ADDderm) and inhalation (ADDinh) for both children and adults were calculated using the equations 4-10. The values used for the calculation are presented in Table 5. ADDing, ADDderm and ADDinh are the chronic daily amount of exposure to PTEs (mg/kgday) through ingestion, dermal absorption and inhalation respectively.

Table 3. Pre-industrial reference level (kg/g) and toxic- response factor by Håkanson (1980)

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

Table 4. The terminologies used to describe the risk factor

ER	Ecological Potential Risk for Single Element	PERI	Ecological Risk
$E^i r$<40	Low potential ecological risk;	PERI < 150	Low
40≤$E^i r$<80	moderate potential ecological risk;	150 ≤ PERI <300	Moderate
80≤$E^i r$<160	considerable potential ecological risk;	300 ≤ PERI <600	Considerable
160≤$E^i r$ <320	high potential ecological risk; and	600 ≤ PERI	Very High
$E^i r$≤320	very high ecological risk		

Table 5. Parameters used for the health risk assessment through different exposure pathways for soil

Parameter	Unit	Child	Adult	References
Body weight (BW)	kg	15	70	Dept. of Environmental Affairs, 2010
Exposure frequency (EF)	days/ year	350	350	
Exposure duration (ED)	years	6	30	
Ingestion rate (IR)	mg/day	200	100	
Inhalation rate (IRair)	m^3/day	10	20	
Skin surface area (SA)	Cm^2	2100	5800	
Soil adherence factor (AF)	mg/cm^2	0.2	0.07	
Dermal Absorption factor (ABS)	none	0.1	0.1	
Dermal exposure ratio (FE)	none	0.61	0.61	
Particulate emission factor (PEF)	none	1.3*109	1.3*109	
Average time (AT)				
For carcinogens	Days	365 * 70	365 * 70	
For non-carcinogen		365 * ED	365 * ED	
Conversion factor (CF)	m^3/kg	10^6	10^6	USEPA, 2004

The carcinogenic and non-carcinogenic side risk for each PTE were computed using equations 4-10. (USEPA, 2015). Hazard Quotient (HQ) was used to calculate non-cancer risk (equation 7) for each PTE and pathway. HQ > 1 indicates possibility of non-cancer risk. Hazard Index (HI) is the total sum of all the HQ for each pathway (equation 8) (USEPA, 1989). HI < 1 indicates no significant risk of non-carcinogenic hazard while HI > 1 implies possibility of non-carcinogenic hazard which are enhanced with increasing HI values [USEPA, 2001, Carla et al., 2014]. The reference dose (RfD) for each PTE and pathway were taken from The Risk Assessment Information System (RAIS) (Mudgal et al., 2010, South Africa Department of Environmental affairs, 2010, USEPA, 2015).

Carcinogenic risk which is the probability of an individual developing any type of cancer over a lifetime, as a result of exposure to the carcinogenic hazards, was calculated for each pathway using Equations 9 and 10 (Hu et al., 2006, Bosso and Enzweiler, 2008).

A cancer risk below 1×10^{-6} is considered safe. The result of 1×10^{-6} is classified as the carcinogenic target risk. If the cancer risk is above 1×10^{-4} it is then not safe and unacceptable (USEPA, 1989, Carla et al., 2014).

$$\text{ADDing} = \text{Csoil} * \frac{IngR*EF*ED}{BW*AT} * 10^6 \quad (4)$$

$$\text{ADDderm} = \text{Csoil} * \frac{SA*AF*ABS*EF*ED}{BW*AT} * 10^6 \quad (5)$$

$$\text{ADDinh} = \text{Csoil} * \frac{Inh\ R*ER*ED}{PEF*BW*AT} * 10^6 \quad (6)$$

$$\text{HQ} = \frac{CDI\ pathway}{Rf\ D} \quad (7)$$

$$\text{HI} = \Sigma\text{HQ} = \text{HQing} + \text{HQderm} + \text{HQ inh} \quad (8)$$

$$\text{RISKpathway} = \text{CDIpathway} * \text{CSFpathway} \quad (9)$$

$$\text{RISK} = \Sigma\text{RISKpathway} = \text{RISKing} + \text{RISKderm} + \text{RISKihn} \quad (10)$$

RESULTS AND DISCUSSION

Concentrations of Toxic Elements in Soil from the Study Area

The summary of geochemical analysis results of toxic elements in ppm from the soil within and around Ilesha gold mining site is presented in Table 6. The mean values of the toxic elements in soil from the study area decreased in the order Mn > Ba > Cr > Zn > Cu > Ni > Pb > Co >Fe > As > Cd. The ranges were as follows: Mn (129–2494 ppm); Ba (30-517 ppm); Cr (29-212 ppm); Zn (17.6-204.1ppm); Cu (7.59-87.19 ppm); Ni (4.1-73.8ppm); Pb (6.06-28.61ppm); Co (35-32.8ppm); Fe (1.2-4.5 ppm); As (0.2-4.5 ppm) and Cd (0.02–0.20ppm).

All the toxic elements were higher than crustal average values used as background values except Fe in some samples (Table 6). Ni, As and Cr were higher than FAO/WHO guidelines whereas only Cr was higher than EU standard in some soil samples (Table 6).

Table 6. Summary of Toxic Elements in Soil within and around Ilesha gold mining sites

Toxic Metals (ppm).	Min	Max	Mean	Std. Dev.	FAO/ WHO Std).	EU Std.	South Africa	Crustal Avg.
Cu	7.59	87.19	36.96	20.82	100	140	16	25
Pb	6.06	28.61	14.61	6.40	100	300	20	17
Zn	17.60	204.10	72.42	42.74	300	300	240	71
Ni	4.10	73.80	28.29	19.19	50	75	91	44
Co	3.50	32.80	14.41	7.06	50	Na	300	17
Fe	1.20	16.88	5.10	3.67	na	Na	Na	35
As	0.20	4.50	1.27	1.13	1.5	Na	5.8	1.5
Cd	0.02	0.20	0.06	0.04	3	3	7.5	0.1
Mn	129.00	2494.00	983.22	549.14	na	Na	Na	600
Ba	30.00	517.00	232.22	141.14	Na	Na	na	500
Cr	29.00	212.00	89.78	53.29	100	100	6.5	83

FAO/WHO Guidelines- Chiroma et al., 2014: EU Guidelines – European Commission on environment, 2002: South Africa Guidelines -Dept. of envtal. Affairs, 2010.

CONTAMINATION ASSESSMENT OF TOXIC ELEMENTS

Pollution Index (PI), Pollution Load Index (PLI) and Potential Ecological Risk Index were used to assess the degree of contamination in the study area.

Pollution Index (PI) and Pollution Load Index (PLI)

The PIs of the toxic elements are presented in Table 7 and showed the descending order of Mn>Cu>Cr>Zn>Pb>Co>As>Ni>Cd>Ba>Fe. The PIs of Mn, Cu and As were within low to high contamination, Pb, Zn, Ni, Co, Cd and Cr were within low to middle contamination while Fe and Ba can be categorized as low contamination. Pollution Load Index showed that the soil in the study area can be classified as unpolluted to moderately polluted and ranged from 0.18 to 1.23 (Table 7).

Ecological Risk Index ($E^i r$) and Potential Ecological Risk Index (PERI)

The Ecological Risk Index ($E^i r$) calculated for six toxic elements showed that the order of pollution was Cd>As>Cu>Pb>Ni>Zn. The toxic elements showed $E^i r$ of <40 and indicated low ecological potential risk index except Cd that showed low to moderate ecological potential risk (6-60). Cd contributed highest to the Ecological Risk (PERI) which ranged between 15.28 and 94.24 and indicated low risk (Table 8).

Table 7. Results of Pollution Load Index in Ilesha Soil

Toxic Elements	Pollution Index (Range)	Pollution Index (Mean)	Interpretation
Cu	0.3-3.5	1.47	Low to High Contamination
Pb	0.4-1.7	0.85	Low to Middle Contamination
Zn	0.2-2.9	1.02	Low to Middle Contamination
Ni	0.09-1.7	0.64	Low to Middle Contamination
Co	0.2-1.9	0.84	Low to Middle Contamination
Fe	0.03-0.5	0.14	Low Contamination
As	0.13-3	0.84	Low to High Contamination
Cd	0.2-2	0.61	Low to Middle Contamination
Mn	0.2-4.2	1.64	Low to High Contamination
Ba	0.05-0.94	0.42	Low Contamination
Cr	0.35-2.6	1.08	Low to Middle Contamination
PLI	0.18-1.23	0.79	Unpolluted to moderately polluted

Table 8. Result of Potential Ecological Risk Index

Toxic Metals	$E^i r$ (Min)	$E^i r$ (Max)	$E^i r$ (Mean)	Interpretation
Cu	1.52	17.44	7.39	Low Potential Ecological risk
Pb	1.78	8.41	4.29	Low Potential Ecological risk
Zn	0.25	2.87	1.02	Low Potential Ecological risk
Ni	0.56	10.06	3.86	Low Potential Ecological risk
As	1.33	30	8.44	Low Potential Ecological risk
Cd	6	60	18.16	Low to Moderate Potential Ecological risk
PERI	15.28-94.24	43.18		Low risk

HEALTH RISK OF THE POTENTIALLY TOXIC ELEMENTS IN THE STUDY AREA

Non-Carcinogenic Risk of Toxic Elements in the study area

The Non carcinogenic risk for adults and children in Ilesha gold mining area were calculated for the ingestion, inhalation and dermal pathways and presented in Table 9 based on the toxicity threshold value known as the chronic reference dose (RfD) values and Average Daily Doses (ADD) values also presented in Table 9. The ADD for the three pathways were lower than the recommended RfDs for all the toxic elements (Table 9).

The values of hazard Index (HI) calculated for child population showed values less than 1 for dermal and inhalation pathways. The HI for ingestion pathway ranged between 0.199-1.65 with mean of 0.79 (Table 9). The value of HI for adult population was less than 1 for all the pathways. The total HI for the three pathways ranged between 0.22 -1.87 and 0.035 -0.37 for child and adult respectively (Table 9, Figs 2-3). HI <1 pose no obvious risk to the population, but if the value exceeds one, there may be concern for potential non-carcinogenic

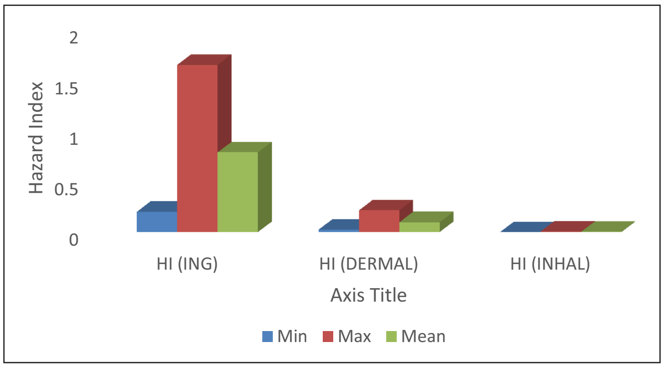

Fig.2. Hazard index for child

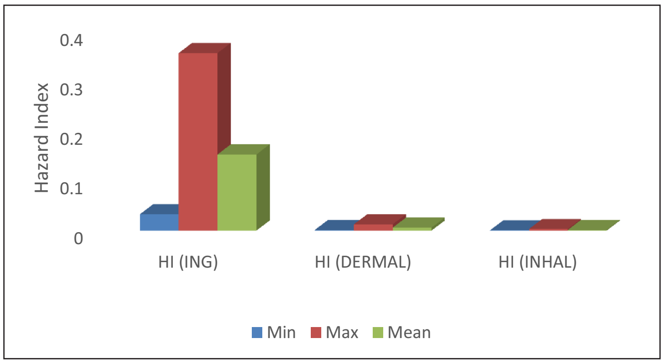

Fig.3. Hazard index for adult

effects [USEPA, 2004]. The results also showed that, in both adults and children, the ingestion pathway contributed the greatest to non-carcinogenic risk followed by the dermal pathway. Inhalation contributed the least to the risk. 33% of the samples showed HI values above 1 for child population (Fig. 4).

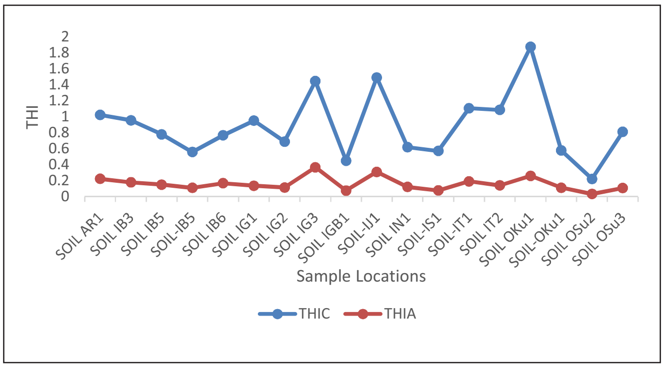

Fig.4. THI against sample locations.

Carcinogenic Risk of Toxic Elements in the study area

The Carcinogenic Risk Assessment of toxic elements calculated for child and adult in the study area showed that As contributed the highest risk to the study area with values ranging between 0.23 to 2.45. This was followed by Co (0.021-0.13), Cd (0.003- 0.3) and Pb which posed the least risk (0.0003-0.001). All the values were higher than USEPA and South Africa standards of $1x10^{-6}$ to $1x10^{-4}$ and $5x10^{-6}$ respectively (USEPA, 2004 and Govt. of South Africa, 2006). As, Co, Cd and Pb constituted high cancer risk in the study area with child at higher risk than adult. The Total Cancer Risk (TCR) in the study area based on all the pathways ranged between 0.19 -3.86 and 0.18-3.58 for child and adult respectively and pose risk to the entire population in the study area (Table 9, Fig. 5).The ingestion route seems to be the major contributor to excess lifetime cancer risk followed by the dermal pathway.

SUMMARY AND CONCLUSION

The results of geochemical analysis showed that the average

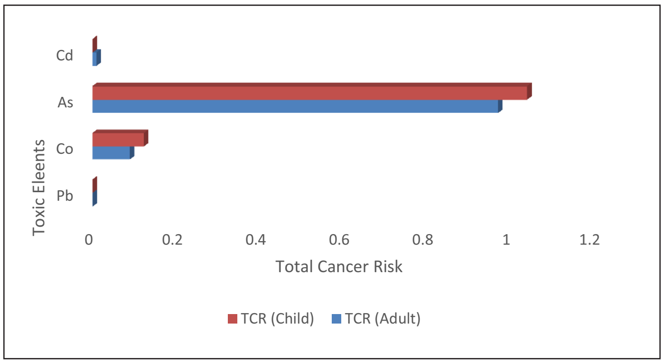

Fig.5. Total cancer risk in the study area.

concentrations of the potential toxic elements (PTEs) in soil from the Ilesha gold mining soil varied significantly and decreased in the order of Mn > Ba > Cr > Zn > Cu > Ni > Pb > Co >Fe > As > Cd. All the toxic elements were higher than crustal average values used as background excepting Fe for some samples. Ni, As and Cr were higher than FAO/WHO guidelines whereas only Cr was higher than EU standard in some soil samples. The results of Pollution Load Index and Potential Ecological Risk Index (PERI) showed that the soil in the study area can be classified as unpolluted to moderately polluted and low risk respectively. The results of health risk also showed that, in both adults and children, the ingestion pathway contributed the greatest to non-carcinogenic risk followed by the dermal pathway. Inhalation contributed the least to the risk. 33% of the samples showed HI values above 1 for child. As, Co, Cd and Pb constitute high cancer risk in the study area with child at higher risk than adult. It can be concluded that soils surrounding the gold mining area are moderately polluted by toxic elements especially from As, Co, Cd and Pb. There is therefore a critical need to put in place mining regulations to protect residents, especially children from pollution of these toxic elements in the environment.

Acknowledgments: The authors would like to thank University of Lagos Central Research Committee for the grant to undertake this research.

Table 9 Average daily intake (ADI) values in mg/kg/day for adults and children in soil from the mining area for non-carcinogenic risk calculations

	ADD	Cu	Pb	Zn	Co	As	Cd	Mn	Cr	Total
Child	Ingestion	0.000472	0.000187	0.000926	0.000184	1.62E-05	7.74E-07	0.012571	0.001148	0.015505
	Inhalation	4.34E-07	1.96E-06	1.13E-07	0.001188	1.98E-06	4.99E-07	Not Determined	0.001407	0.015505
	Dermal	1.32E-06	5.23E-07	2.59E-06	5.16E-07	1.62E-11	2.17E-08	3.52E-05	3.21E-06	4.34E-05
	Total	0.000474	0.000189	0.000929	0.001372	1.82E-05	1.3E-06	0.012606	0.002558	0.031053
	HI (Ingest.)	0.199-1.65 (0.79)								
	HI(Dermal)	0.023- 0.214 (0.097)								
	HI (Inh.)	0.0007 – 0.0058 (0.0026)								
	Total HI	0.22-1.87 (0.89)								
	TCR	0.19-3.86 (1.167)								
Adult	Ingestion	5.06E-05	2.00E-05	9.92E-05	0.000252	1.74E-05	8.30E-08	0.001347	0.000105	0.001892
	Inhalation	2.17E-07	9.81E-07	5.67E-08	0.000594	9.92E-07	2.5E-07	Not Determined	0.000703	0.001892
	Dermal	5.06E-11	2.00E-11	3.96E-07	2.82E-09	1.74E-11	3.31E-10	5.37E-06	4.21E-07	6.19E-06
	Total	5.08E-05	2.10E-05	9.97E-05	8.46E-04	1.84E-05	3.33E-07	1.35E-03	8.09E-04	3.79E-03
	RFDing	4.00E-02	3.50E-03	3.00E-01	2.00E-02	3.00E-04	1.00E-03	0.046	3.00E-03	
	RFDinh	4.00E-02	3.52E-03	3.00E-01	5.70E-06	3.00E-04	1.00E-03		2.86E-05	
	RFDderm	1.20E-02	5.25E-04	6.00E-02	5.70E-06	3.00E-04	1.00E-05	0.0035	6.00E-05	
	HI (Ing.)	0.033-0.357 (0.153)								
	HI(Dermal)	0.001 – 0.012 (0.005)								
	HI (Inh.)	0.0004 -0.003 (0.0013)								
	Total HI	0.035 – 0,37 (0.16)								
	TCR	0.18 -3.58 (1.073)								

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(Received: 2 January 2017; Revised form accepted: 17 October 2017)