

Heavy Metals Source Apportionment and Human Health Risk Assessment of Contaminated Soils of Zamfara State, Nigeria

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Abstract. Progressive illegal artisanal mining activities threaten public health without functional law enforcement on pollution control and proper management practices. This is not an exception of Zamfara State, Nigeria, where a large portion of the populace participates in artisanal mining. The study was conducted to assess the level of health risk associated with heavy metals contaminated soils of Zamfara state, Nigeria. Soil samples were collected from five mining locations (Abare, Bagega, Daret, Sunke, and Tunga Kudaku) and Anka-town (control site) with no record of mining activities. In each place, bulked soil samples were collected from three sites (mining site, processing site, and village), and the concentration of six heavy metals (Fe, Pb, Cd, Cr, Zn, and Ni) in all the samples were analyzed. The result of the principal component analysis and correlation analysis revealed that Pb, Zn, Cr, and Ni originated from the same source, i.e., anthropogenic/mining activities. While Fe and Cd originated from the geogenic processes because of their high abundance in the soil of the study area, as Anka-town (control site) also recorded high concentrations of Fe and Cd. Health risk assessments were carried out in two groups of population (adult and children) through three exposure pathways (i.e., ingestion, dermal contact, and inhalation). The results showed that ingestion dominated dermal contact and inhalation pathways, and Fe is the riskiest metal while Cd and Ni have the lowest risk of exposure for daily intakes. The non-cancer hazard quotient (HQ) values were all recorded below 1. For the total hazard index (THI), all the adult's exposure pathways were negligible, while for children, only Bagega has ingestion of heavy metals exceeding one (1.10), indicating that non-cancer health risks for children exist. The other four mining locations, Abare, Sunke, Tunga, and Daret, have values approaching one (i.e., 0.71, 0.60, 0.50, and 0.74, respectively). While for Anka town, which is the control site, it has a value far less than one (0.16). These indicate that all the study locations have the potential for children's health risk through ingesting food produced from contaminated soils. Therefore, there is an urgent need to apply remediation measures immediately to combat complications raised due to heavy metal contaminations.

Keywords: artisanal mining; heavy metal pollution; human-risk assessment; Zamfara; Nigeria.

INTRODUCTION

Nigeria is the country heavenly bestowed with numerous natural resources. Within the country, Zamfara state has been recognized among the top artisanal mining centers, producing massive solid mineral resources for decades, despite farming being the primary source of livelihood for local people. The abundance of solid mineral resources attracted illegal mining and mineral explorations in the regions producing waste products in a large quantity, accompanied by the discharge of lethal elements to the environment (Nuhu et al., 2014), particularly heavy-metals (Abdu & Yusuf, 2013; Yahaya et al., 2021). The existence of heavy metals in the soil may not be confirmed as contaminants because of the geological

nature of parent materials from which the soil originated (Giusti, 2013). Apart from artisanal mining, many industries, such as textile, dairies, recycling facilities, fertilizer industries, tin and drug industries, and agricultural activities, also contribute to heavy metals pollution in the soil (Patil & Kaushik, 2016). Due to the persistent, non-degradable, and toxic nature in soils and water bodies; heavy metals become a subject of several studies that draw global attention (Yap & Al-Mutairi, 2022). The heavy metal contaminants are known to be a problem in developed countries due to advanced industrialization but are increasingly experienced in developing countries where illegal mining activities become rampant, because law enforcement on pollution

controls is seriously lacking in the developing countries (Addey et al., 2018).

The accumulation of heavy metals in an environment is a consequence of either natural (weathering) or anthropogenic (illegal mining) processes. While the accumulation through weathering process is rarely toxic, the anthropogenic processes, on the other hand, contaminate the soils more rapidly with one or more heavy metals above the threshold limits to cause menaces in humans, plants, animals, or the entire ecosystems (Masindi & Muedi, 2018).

Heavy metals are often transported from mining areas to neighboring environmental locations with higher risks of direct human exposure. The discharged products relatively contain high concentrations of metals compared to those in the receiving environment. Subsequently, other biological entities may render the metal more bioavailable in the receiving environmental system (D'amore et al., 2005; Mohammed & Abdu, 2014). Therefore, these enhanced the accumulation of heavy metals in both mining and their neighboring communities. Without enforcement of environmental pollution control laws and proper management practices, these may trigger the movement of heavy metals in the environment through the food chain and food web (Chang & Cockerham, 2019), causing a threat to public health and the environment.

Different kinds of literature assessed the level of contamination and distribution of heavy metals in various areas of the study locations (Akpanowo et al., 2021; Orisakwe et al., 2017; Sulaiman et al., 2019). However, studies on human health risk assessments exposed to heavy metals in contaminated areas are limited.

This study is essential in evaluating the non-cancer health risk hazard caused indirectly; by consuming contaminated products or directly by dermal contact and inhaling contaminated dust. In addition, information from this study is also vital to public health workers and scientific management of pollution control in both

mining and processing sites in developing strategies for site remediation concerning human health risks.

METHODS

Study Area

Zamfara state has been a center of artisanal mining activities for decades. Apart from mining, farming is considered the primary source of income for the inhabitants (Orisakwe et al., 2017). Five villages (Abare, Bagega, Dareta, Sunke, and Tungar Kudaku), where mining activities are taking place and Anka-town with no record of mining activities were selected for the study. All villages are under the Anka Local Government Area of Zamfara State, Nigeria (Figure 1).

Soil Sample collection, preparation, and analysis

Soil sampling was carried out in six (6) different locations. In each location, soil samples were collected from the processing site, mining site, and the villages except in Anka-town (control site), where only one sample was collected. At each sampling site, five random subsamples were collected at a 0-30 cm sampling depth and made a composite sample.

These make sixteen (16) samples, and each is replicated into three. The collected soil samples were separately placed in a polythene bag and taken to the laboratory, spread, and air-dried. Some portions of the prepared samples were used for physical and chemical analyses, while the remaining portion was used for heavy metal analysis.

Soil analysis conducted for soil pH, electric conductivity, total nitrogen, available phosphorus, organic carbon, exchangeable acidity, exchangeable cations, and cation exchange capacities, all using standard methods.

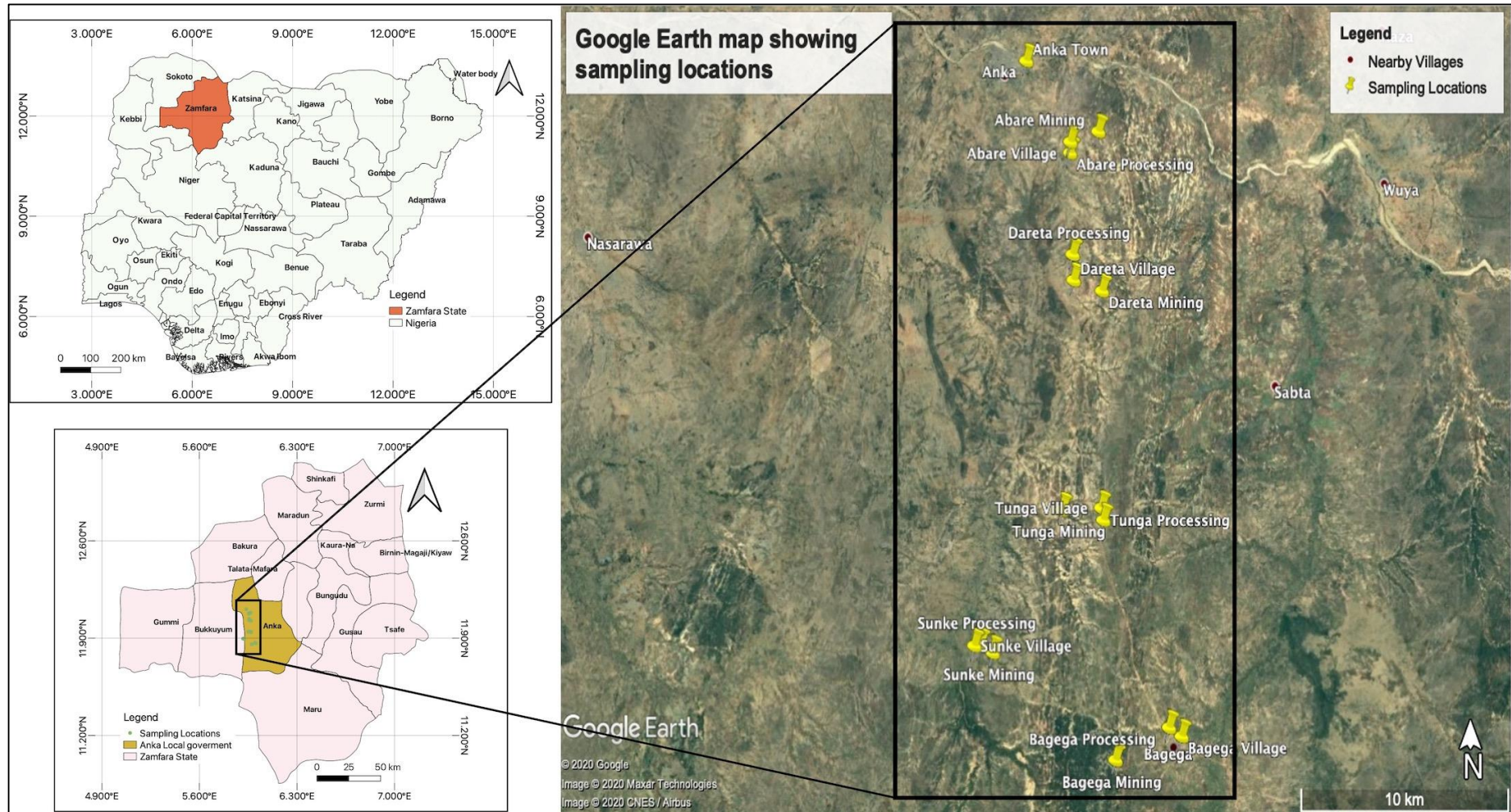


Figure 1. Map of sampling locations – Zamfara state, Nigeria

Analysis of heavy metals concentration

In analyzing heavy metals concentration, stainless steel was used to grind the air-dried soil samples. Soil sample (1 gram) was digested using a mixture of HF: HClO₄: HNO₃ acids in a 250 ml conical flask (Nwajei & Gagophien, 2000). After digestion on a hot plate for 3 hours at 80°C, the digests were filtered and made to 100 ml mark with deionized water in 100 ml standard plastic bottles.

Flame atomic absorption spectrophotometry (Varian model-AA240FS) was used to determine the concentrations of the heavy metals (iron, zinc, lead, chromium, and cadmium) in the sample solutions.

$$ADD_{inh} = \frac{C \times R_{inh} \times ED \times EF}{BW \times PEF \times AT} \quad \text{Eq. (1)}$$

$$ADD_{ing} = \frac{C \times R_{ing} \times EF \times ED \times 10^{-6}}{AT \times BW} \quad \text{Eq. (2)}$$

$$ADD_{derm} = \frac{C \times SL \times SA \times EF \times ABS \times ED \times 10^{-6}}{BW \times AT} \quad \text{Eq. (3)}$$

Where:

Table 1. Definition of equations variables

Variable	Meaning	Reference
ADD _{inh, ing, and derm}	average daily dose for inhalation, ingestion, and dermal contact, respectively	Variable
C	concentration (mg/kg) of each metal in soil	Variable
R _{ing}	ingestion rate (200 mg/day) for children and (100 mg/day) for adults	(USEPA, 2001)
R _{inh}	inhalation rate is 10m ³ /day for children and 20m ³ /day for adult	(Van den Berg, 1995)
EF	exposure frequency is 250 days/year	(Ferreira-Baptista & De Miguel, 2005)
ED	exposure duration is 6 years for children and 25 years for adult	(USEPA, 2001)
SA	exposed skin area is 2800 cm ² for children and 3300 cm ² for adult	(USEPA, 2001)
SL	skin adherence factor is 0.2mg/cm ² /h for children and adult	(USEPA, 2001)
ABS	dermal absorption factor (unitless) is 0.001	(USEPA, 1989)
PEF	particle emission factor is 1.316 x 10 ⁹ m ³ /kg	(USEPA, 2001)
BW	average body weight is 15 kg for children and 70 kg for adults	(USEPA, 1989)

Human health risk assessment

Average Daily Doses (ADDs)

The human can encounter heavy metals directly via three exposure pathways: ingestion of particles containing heavy metals; dermal absorption of trace elements in particles adsorbed to exposed skin; and inhalation of the suspended particles containing heavy metals through the nose and mouth.

The average daily dose or intake (ADD, mg/kg/day) for non-carcinogens via each of the three exposure pathways were used for the assessment and were computed using the equations proposed by USEPA (1989, 1996b).

Hazard Quotient (HQ)

Hazard quotient is an index used to estimate the potential health risk to human from soil heavy-metals. It has an assumption that there is a daily intake rate (reference dose (RfD)) of heavy metal that poses no appreciable non-cancer risk or adverse health effects even if a sensitive population is exposed to it over a 70 year lifetime (USEPA, 2005a). It is calculated using equation (4) proposed by USEPA (2001).

$$HQ = \frac{ADD}{RfD} \quad \text{Eq. (4)}$$

ADD (average daily dose) is measured in mg/kg/day for each exposure pathway; RfD (reference dose) is also measured in mg/kg/day.

RfD_{inhalation} is calculated by multiplying RfC with 20 m³ per day per 70 kg for adults (USEPA, 1994b, 2005b); and with 10 m³ per day per 15 kg for children (USEPA, 1994b, 2005b).

RfC is the reference concentration obtained from USDOE (2011).

Total Hazard Index (THI)

Based on the calculated hazard quotients, the total hazard index is determined to summate all the different heavy metals HQs; this is the combined effect of heavy metals' potential risk. It is calculated thus by using equation (5).

$$THI = HQ1 + HQ2 + \dots + HQn \quad \text{Eq. (5)}$$

If THI is < 1.0, there is no potential risk because of the heavy metals. When the THI is > 1.0, there is a potential non-cancer health effect in the study area (Ihedioha et al., 2017).

Quality assurance and Statistical analysis

For the accuracy of experimental data, all reagents used were of analytical grade. Distilled water was used throughout the experimental procedure. Each soil sample was analyzed in triplicate, and one standard sample was analyzed after every three experimental samples. Field blank and

experimental blank samples were also analyzed to ensure the accuracy of the data obtained.

Analysis of variance using completely randomized design (CRD) was used to analyze the means, while Duncan Multiple Range Technique (DMRT) tests were used to separate the means. Computation of the risk factors was done using Microsoft Excel. Principal component analysis and correlation analysis were used to establish the relationship between the heavy metals and establish the factor responsible for the metal concentration and source apportionment (Sarvade et al., 2016). The number of significant principal components was selected in the PCA based on varimax orthogonal rotation with Kaiser Normalization at Eigenvalues greater than 1. All the analyses were conducted using SAS 9.0 (SAS, 2002) and Microsoft Excel (version 2016).

RESULTS AND DISCUSSION

Physical and chemical properties of the soil

Table 2 shows results of the physical and chemical analysis of the experimental soils. Using the USDA soil classification system, the soil textural classes at the ploughing depth (0-30cm) was predominantly sandy loam, fewer falls under loam, and only one is silty loam. This proved the suitability of soils in all the regions for agricultural production (Weil and Brady, 2017), as most field crops grow better on loam textured soils. The pH (H₂O) values ranged from 6.2 – 7.7 (slightly acidic to slightly basic), which is in agreement with findings of Raji et al. (2015) in some soils of the Nigerian savanna and falls within the optimum pH range for most plants growth (Queensland, 2021). The electrical conductivity and cation exchange capacity ranged from 0.10 – 0.35 dSm⁻¹ and 3.97 – 14.89 cmol₍₊₎kg⁻¹ respectively, these indicate that the salinity and retention capacity of the soils would not significantly affect plant growth in the regions (Hazelton & Murphy,

2016; USSLS, 1954). The values of the organic carbon ranged from 0.20 – 1.22 gkg⁻¹; based on the ratings of Sigarf et al. (2003), these can be described as very low ($\leq 2\%$). This is undoubtedly due to lower organic matter contents in the soils because of lower vegetation, and these could be a sign of poor biological activities in the soils (Seid & Genanew, 2013; Sarvade et al., 2014; Chaturvedi et al., 2016; Sarvade et al., 2019). The available phosphorus and total nitrogen content range from 1.58 – 17.68 mg/kg to 0.7 – 2.8 gkg⁻¹, rated low to medium and very low to low, respectively (FMANR, 1990; Sigarf et al., 2003). The nitrogen deficiency might be attributed to lower organic matter contents in the soils (Pawar et al., 2014; Asmamaw et al., 2018), which is a feature of typical Nigerian Savanna soils.

Principal component analysis and correlation coefficients

Table 3 shows the result of correlation and principal component analysis. The correlation matrix revealed that Pb has a highly significant positive correlation with Zn, Cr, and Ni ($p < 0.001$) with a correlation coefficient of 0.757, 0.375, and 0.652. Furthermore, Cd has a positive correlation with Fe having a correlation coefficient of 0.224. Based on the result of the principal component analysis, three (3) principal

components (PCs) cumulatively account for 87 % of the total variation (

Table 3). The PC1 mainly contain Cr, Ni, Pb, and Zn with variance values of 0.807, 0.917, 0.798, and 0.850, respectively, and accounted for 58 % of the total variation, PC2 mainly contain Cd and Fe with a variance value of 0.564 and 0.669 respectively and accounted for 17 % of the total variation, and PC3 mainly have Fe with a variance value of 0.581 and accounted for 13 % of the total variation.

The results of the correlation analysis agree with that of the principal component analysis. This indicates that the significant correlation between the heavy metals suggested that they originate from a common origin (Qu et al., 2012; Rahman et al., 2018). Thus, Cr, Ni, Pb, and Zn with the same principal component and a highly significant positive correlation originated from the same source, i.e., artisanal mining, since is the common anthropogenic activity in the study area. This agreed with Yahaya et al. (2021) finding of the soil of the exact study locations. As for Cd and Fe that were also positively correlated and had the same principal component, they originated from the abundance of the heavy metals in the soils/parent material of the study area. Ankatown (control site) also recorded a significant amount of Cd. For Fe, earlier reported that it's the most abundant element in Nigerian soils (Amusan et al., 2005; Solomon et al., 2016).

Table 2. Physical and chemical properties of the experimental soils

Locations	pH		OC. (CaCl ₂)	EC. (dSm ⁻¹)	Available P (mg/kg)	Total N (gkg ⁻¹)	Ca	Mg	K	Na	EA	ECEC	Clay	Silt	Sand	Textural Classes
	(H ₂ O)	(gkg ⁻¹)														
Abare Mining	6.8	0.80	6.2	0.14	1.78	1.8	7.23	0.88	0.32	0.00	0.40	8.83	6	24	70	SL
Abare Processing	7.1	0.26	6.3	0.18	4.38	1.4	7.64	0.92	0.32	0.14	0.60	9.62	8	40	52	L
Abare Village	7.2	0.32	6.4	0.19	3.85	1.1	10.00	0.80	0.24	0.11	0.40	11.55	8	30	62	SL
Bagega Mining	7.2	0.26	6.7	0.14	3.50	2.8	7.78	0.62	0.39	0.26	0.40	9.45	14	28	58	SL
Bagega Processing	7.4	0.42	6.8	0.13	1.58	2.5	7.92	1.32	0.42	0.64	0.40	10.69	16	18	66	SL
Bagega Village	7.7	0.66	6.7	0.12	2.98	1.4	4.71	0.65	0.34	0.00	0.60	6.30	10	42	48	L
Dareta Mining	6.6	0.52	5.2	0.13	2.63	1.8	3.69	0.57	0.25	0.02	0.40	4.92	16	28	56	SL
Dareta Processing	6.2	0.08	5.5	0.30	2.28	1.4	6.01	1.12	0.19	0.19	0.40	7.90	6	40	54	SL
Dareta Village	7.1	0.62	5.8	0.10	2.63	1.4	7.51	1.07	0.24	0.30	0.60	9.71	10	40	50	L
Sunke Mining	6.9	0.80	5.8	0.12	2.10	2.1	3.20	0.15	0.20	0.02	0.40	3.97	10	42	48	L
Sunke Processing	6.6	0.20	6.1	0.35	2.10	2.1	5.09	0.37	0.77	0.51	0.40	7.13	10	22	68	SL
Sunke Village	7.2	1.22	6.5	0.17	3.15	2.1	9.63	0.28	0.37	0.00	0.40	10.68	10	52	38	SiL
Tunga Mining	6.5	0.30	6.0	0.14	2.45	1.4	11.70	1.53	0.24	0.00	0.60	14.07	16	24	60	SL
Tunga Processing	6.4	0.22	6.3	0.31	2.10	0.7	9.63	1.03	0.23	0.28	0.40	11.57	10	22	68	SL
Tunga Village	7.0	0.80	6.2	0.12	7.88	1.4	10.30	1.48	0.44	0.00	0.40	12.62	12	46	42	L
Anka town	7.2	0.32	6.6	0.18	4.20	1.4	12.00	2.00	0.29	0.00	0.60	14.89	6	22	72	SL

Textural class key: L – Loam; SiL – Silt Loam; SL – Sandy Loam.

Table 3. Principal component analysis (PCA) and correlation coefficients of heavy metals in Anka soils

Metal	Principal Component Analysis			Pearson's Correlation Coefficient							
	PC1	PC2	PC3	Metal	Cd	Cr	Fe	Ni	Pb	Zn	
Cd	-0.628	0.564	-0.371	Cd	1						
Cr	0.807	-0.191	0.249	Cr	-0.02	1					
Fe	0.453	0.669	0.581	Fe	0.224	0.345**	1				
Ni	0.917	-0.258	-0.171	Ni	-0.21	0.754***	0.156	1			
Pb	0.798	0.255	-0.294	Pb	-0.09	0.375***	0.368**	0.652***	1		
Zn	0.85	0.28	-0.36	Zn	0.031	0.582***	0.359**	0.771***	0.757***	1	
Eigenvalue	3.45	1.013	0.783								
Variability (%)	57.504	16.875	13.048								
Cumulative (%)	57.504	74.379	87.428								

** Correlation is significant at the 0.01 level; *** Correlation is significant at the 0.001 level.

Health risk assessments of heavy metals contaminated soils

Average daily doses (ADDs)

Tabel 4-9 shows the average daily dose of adult and child for the three exposure pathways (ingestion, dermal, and inhalation) across the study locations. The result revealed that the average daily doses (ADDs) of the heavy metals for children are greater than that of adults for all the study locations' exposure pathways. At Bagega (**Error! Not a valid bookmark self-reference.**) the ADD ingestion decreases in the order of Fe > Pb > Cr > Zn > Ni > Cd for both adults and children. For ADD inhalation it follows the order Cr > Ni > Fe > Pb > Zn > Cd for children, and Fe > Ni > Pb > Cr > Zn > Cd for adults. While for ADD dermal contact, it follows the order Fe > Pb > Cr > Zn > Ni > Cd for both adults and children. At Dareta and Sunke (table 5 and table 6 respectively), all the ADDs decreased in the order: Fe > Pb > Cr > Zn > Cd > Ni. There is an observed slight difference in the order of decreasing

ADDs at Abare (table 8): Fe > Pb > Zn > Cr > Cd > Ni for both children and adults. Moreover, at Anka-town (table 9), all the ADDs recorded decreased in the order: Fe > Cd > Ni > Cr > Pb > Zn. At Tunga, all the observed ADDs differed in the three exposure pathways, as shown in table 7.

Generally, it could be observed from the results that Cd risk intake is the lowest at Bagega and second-lowest after Ni at Dareta, Sunke, and Abare for both adults and children through all the exposure pathways. Although Cd mainly contributed the highest to the overall potential ecological risk as reported in the previous study of Mohammadi et al. (2020) in Neyshabur, Iran, and Yahaya et al. (2021) in the exact study locations, here Fe recorded the highest ADDs values in all the study locations, indicating that it has the highest risk of intake to both adults and children through all the three exposure pathways. This could be attributed to being the most abundant metal in Nigerian soils (Amusan et al., 2005).

Table 4. Average Daily Doses of heavy metals in Bagega-Anka Local Government, Zamfara State

Metals	Average Metal Conc. (mg/kg)	Age	Ingestion	Inhalation	Dermal
Fe	23892.73	Child	2.12E-01	8.29E-06	6.11E-04
		Adult	2.34E-02	1.77E-06	1.54E-04
Pb	336.47	Child	3.07E-03	1.17E-07	8.60E-06
		Adult	3.29E-04	2.50E-08	2.17E-06
Cd	1.34	Child	1.23E-05	4.67E-10	3.44E-08
		Adult	1.32E-06	9.10E-11	8.68E-09
Zn	25.47	Child	2.33E-04	8.84E-09	6.51E-07
		Adult	2.49E-05	1.89E-09	1.64E-07
Cr	81.1	Child	7.41E-04	2.81E-05	2.07E-06
		Adult	7.94E-05	6.03E-09	5.24E-07
Ni	3.77	Child	3.44E-05	1.31E-05	9.63E-08
		Adult	3.69E-06	2.80E-08	2.43E-08

Conc.: concentration; E-: x 10[^]

Table 5. Average Daily Doses of heavy metals in Dareta-Anka Local Government, Zamfara State

Metals	Average Metal Conc. (mg/kg)	Age	Ingestion	Inhalation	Dermal
Fe	21619.07	Child	1.97E-01	7.50E-06	5.53E-04
		Adult	2.12E-02	1.61E-06	1.40E-04
Pb	177.83	Child	1.62E-03	6.17E-08	4.55E-06
		Adult	1.74E-04	1.32E-08	1.15E-06
Cd	1.73	Child	1.58E-05	6.01E-10	4.43E-08
		Adult	1.70E-06	1.29E-10	1.12E-08
Zn	11.73	Child	1.07E-04	4.07E-09	3.00E-07
		Adult	1.15E-05	8.72E-10	7.58E-08
Cr	40.73	Child	3.72E-04	1.41E-08	1.04E-06
		Adult	3.99E-05	3.03E-09	2.63E-07
Ni	1.5	Child	1.37E-05	5.21E-10	3.84E-08
		Adult	1.47E-06	1.12E-10	9.69E-09

Conc.: concentration; E-: x 10[^]

Table 6. Average Daily Doses of heavy metals in Sunke-Anka Local Government, Zamfara State

Metals	Average Metal Conc. (mg/kg)	Age	Ingestion	Inhalation	Dermal
Fe	25636.67	Child	2.34E-01	8.90E-06	6.56E-04
		Adult	2.51E-02	1.91E-06	1.66E-04
Pb	108.5	Child	9.91E-04	3.76E-08	2.77E-06
		Adult	1.06E-04	8.07E-09	7.01E-07
Cd	2	Child	1.83E-05	6.94E-10	5.11E-08
		Adult	1.96E-06	1.49E-10	1.29E-08
Zn	16.7	Child	1.53E-04	5.79E-09	4.27E-07
		Adult	1.63E-05	1.24E-09	1.08E-07
Cr	32.97	Child	3.01E-04	1.14E-08	8.43E-07
		Adult	3.23E-05	2.45E-09	2.13E-07
Ni	0.001	Child	1.32E-08	5.01E-13	3.69E-11
		Adult	1.41E-09	1.07E-13	9.33E-12

Conc.: concentration; E-: x 10[^]

Table 7. Average Daily Doses of heavy metals in Tunga-Anka Local Government, Zamfara State

Metals	Average Metal Conc. (mg/kg)	Age	Ingestion	Inhalation	Dermal
Fe	18283.8	Child	1.67E-01	6.34E-06	4.68E-04
		Adult	1.79E-02	1.36E-06	1.18E-04
Pb	107.5	Child	9.82E-04	3.73E-08	2.75E-06
		Adult	1.05E-04	7.99E-09	6.94E-07
Cd	1.73	Child	1.58E-05	6.01E-10	4.43E-08
		Adult	1.70E-06	1.29E-10	1.12E-08
Zn	16.03	Child	1.46E-04	5.56E-09	4.10E-07
		Adult	1.57E-05	1.19E-09	1.04E-07
Cr	36.27	Child	3.31E-04	1.26E-08	9.27E-07
		Adult	3.55E-05	2.70E-09	2.34E-07
Ni	0.002	Child	1.42E-08	5.40E-13	3.98E-11
		Adult	1.52E-09	1.16E-13	1.00E-11

Conc.: concentration; E-: x 10[^]

Table 8. Average Daily Doses of heavy metals in Abare-Anka Local Government, Zamfara State

Metals	Average Metal Conc. (mg/kg)	Age	Ingestion	Inhalation	Dermal
Fe	19014.57	Child	1.74E-01	6.60E-06	4.86E-04
		Adult	1.86E-02	1.41E-06	1.23E-04
Pb	210.87	Child	1.93E-03	7.32E-08	5.39E-06
		Adult	2.06E-04	1.57E-08	1.36E-06
Cd	1.43	Child	1.31E-05	4.97E-10	3.67E-08
		Adult	1.40E-06	1.07E-10	9.26E-09
Zn	13.6	Child	1.24E-04	4.72E-09	3.48E-07
		Adult	1.33E-05	1.01E-09	8.78E-08
Cr	2.43	Child	2.22E-05	8.45E-10	6.22E-08
		Adult	2.38E-06	1.81E-10	1.57E-08
Ni	0.001	Child	5.07E-09	1.93E-13	1.42E-11
		Adult	5.44E-10	4.13E-14	3.59E-12

Conc.: concentration; E-: x 10[^]

Table 9. Average Daily Doses of heavy metals in Anka town-Anka Local Government, Zamfara State

Metals	Average Metal Conc. (mg/kg)	Age	Ingestion	Inhalation	Dermal
Fe	11576.8	Child	1.06E-01	4.02E-06	2.96E-04
		Adult	1.13E-02	8.61E-07	7.48E-05
Pb	0.0007	Child	6.09E-09	2.31E-13	1.70E-11
		Adult	6.52E-10	4.96E-14	4.31E-12
Cd	1.2	Child	1.10E-05	4.16E-10	3.07E-08
		Adult	1.17E-06	8.92E-11	7.75E-09
Zn	0.0003	Child	3.04E-09	1.16E-13	8.52E-12
		Adult	3.26E-10	2.48E-14	2.15E-12
Cr	0.0017	Child	1.52E-08	5.78E-13	4.26E-11
		Adult	1.63E-09	1.24E-13	1.08E-11
Ni	0.002	Child	1.83E-08	6.94E-13	5.11E-11
		Adult	1.96E-09	1.49E-13	1.29E-11

Conc.: concentration; E-: x 10[^]

Hazard quotient (HQ.)

Table 10 – 15. In general, the trends of the order of decreasing HQ values were Pb>Fe>Cd>Cr> Zn>Ni and observed to be regular at Bagega, Daret, Tunga Kudaku, and Abare, while in Sunke and Anka town, the HQ was in the order Fe>Pb>Cd> Cr> Zn>Ni, respectively. These shown that the contamination in Anka town and Sunke was due to the abundance of Fe in the study locations, as Fe is the most abundant element in Nigerian soils (Amusan et al., 2005). The order of the hazard quotient (non-cancer risk) was ingestion > dermal contact > inhalation and was observed to be consistent in all the study locations.

The values for soil particles' inhalation through mouth and nose is almost negligible than oral ingestion and dermal contact, and it is quite not likely to pose a substantial hazard. Also, the health risk exposure of children to the soil in the locations was consistently higher than that of adults in all three exposure pathways. This could be attributed to the high possibility of children absorbing high amounts of heavy metals during their outdoor play activities, resulting in more

Results of the magnitude of heavy metals' toxic effects (HQ) were presented in susceptibility to soil toxic metals than adults (Karim, 2019). Result is similar to the result of Ihedioha et al. (2017) in the soil of a municipal solid waste dump in Uyo, Nigeria; Wu et al. (2018) in the soil of an electronic manufacturing site in Hubei, China; and Mohammadi et al. (2020) in soils from the industrial zone in Neyshabur, Iran. Nevertheless, it is contrary to Kamunda et al. (2016) finding in Witwatersrand Gold Mining Basin, South Africa soil, where the inhalation pathway has the highest risk.

All the calculated HQ of the heavy metals was below 1, indicating that the current level of exposure is not likely to cause adverse human health effects (USEPA, 2000). Nonetheless, Pb and Fe revealed the highest values in all the study locations, earlier documented by Salisu et al. (2016) and Yahaya et al. (2021) in their investigation for the distribution and concentration of heavy metals in the study locations. Therefore, gradual increased human activities and exploitation of these heavy metals across the locations may increase the chance of ingestion (Venkateswarlu & Venkatrayulu,

2020), thereby leading to adverse health effects in the near future.

Table 10. Hazard quotients (HQ) of heavy metals in Bagega-Anka Local Government, Zamfara State

Metals	Mean metal Conc. (mg/kg)	Age Group	Ingestion	Inhalation	Dermal
			HQ	HQ	HQ
Fe	23892.73	Child	3.11E-01	1.00E-06	8.73E-04
		Adult	3.34E-02	2.15E-07	2.20E-04
Pb	336.47	Child	7.68E-01	3.34E-05	1.64E-02
		Adult	8.23E-02	7.15E-06	4.14E-03
Cd	1.34	Child	1.23E-02	8.18E-06	6.88E-05
		Adult	1.32E-03	1.75E-06	1.74E-05
Zn	25.47	Child	7.75E-04	1.36E-04	1.09E-05
		Adult	8.31E-05	2.91E-05	2.74E-06
Cr	81.1	Child	4.94E-04	9.38E-04	1.06E-04
		Adult	5.29E-05	2.01E-04	2.69E-05
Ni	3.77	Child	1.72E-03	1.45E-05	1.72E-05
		Adult	1.84E-04	3.11E-06	4.34E-06

Conc.: concentration; E-: x 10[^]

Table 11. Hazard quotients (HQ) of heavy metals in Dareta-Anka Local Government, Zamfara State

Metals	Mean metal Conc. (mg/kg)	Age group	Ingestion	Inhalation	Dermal
			HQ	HQ	HQ
Fe	21619.07	Child	2.82E-01	9.09E-07	9.09E-07
		Adult	3.02E-02	1.95E-07	1.95E-07
Pb	177.83	Child	4.06E-01	1.76E-05	1.76E-05
		Adult	4.35E-02	3.78E-06	3.78E-06
Cd	1.73	Child	1.58E-02	1.06E-05	1.06E-05
		Adult	1.70E-03	2.26E-06	2.26E-06
Zn	11.73	Child	3.57E-04	6.26E-05	6.26E-05
		Adult	3.83E-05	1.34E-05	1.34E-05
Cr	40.73	Child	2.48E-04	4.71E-04	4.71E-04
		Adult	2.66E-05	1.01E-04	1.01E-04
Ni	1.5	Child	6.85E-04	5.78E-06	5.78E-06
		Adult	7.34E-05	1.24E-06	1.24E-06

Conc.: concentration; E-: x 10[^]

Table 12. Hazard quotients (HQ) of heavy metals in Sunke-Anka Local Government, Zamfara State

Metals	Mean metal Conc. (mg/kg)	Age Group	Ingestion	Inhalation	Dermal
			HQ	HQ	HQ
Fe	25636.67	Child	3.34E-01	1.08E-06	9.36E-04
		Adult	3.58E-02	2.31E-07	2.37E-04
Pb	108.5	Child	2.48E-01	1.08E-05	5.28E-03
		Adult	2.65E-02	2.30E-06	1.33E-03
Cd	2	Child	1.83E-02	1.22E-05	1.02E-04
		Adult	1.96E-03	2.61E-06	2.58E-05
Zn	16.7	Child	5.08E-04	8.91E-05	7.12E-06
		Adult	5.45E-05	1.91E-05	1.80E-06
Cr	32.97	Child	2.01E-04	3.81E-04	4.32E-05
		Adult	2.15E-05	8.17E-05	1.09E-05
Ni	0.001	Child	6.60E-07	5.57E-09	6.60E-09
		Adult	7.07E-08	1.19E-09	1.67E-09

Conc.: concentration; E-: x 10[^]

Table 13. Hazard quotients (HQ) of heavy metals in Tunga-Anka Local Government, Zamfara State

Metals	Mean metal Conc. (mg/kg)	Age Group	Ingestion	Inhalation	Dermal
			HQ	HQ	HQ
Fe	18283.8	Child	2.39E-01	7.69E-07	6.68E-04
		Adult	2.56E-02	1.65E-07	1.69E-04
Pb	107.5	Child	2.45E-01	1.07E-05	5.24E-03
		Adult	2.63E-02	2.28E-06	1.32E-03
Cd	1.73	Child	1.58E-02	1.06E-05	8.86E-05
		Adult	1.70E-03	2.26E-06	2.24E-05
Zn	16.03	Child	4.88E-04	8.56E-05	6.83E-06
		Adult	5.23E-05	1.83E-05	1.73E-06
Cr	36.27	Child	2.21E-04	4.19E-04	4.76E-05
		Adult	2.37E-05	8.99E-05	1.20E-05
Ni	0.002	Child	7.10E-07	6.00E-09	7.10E-09
		Adult	7.61E-08	1.29E-09	1.79E-09

Conc.: concentration; E-: x 10[^]

Table 14. Hazard quotients (HQ) of heavy metals in Abare-Anka Local Government, Zamfara State

Metals	Mean metal Conc. (mg/kg)	Age group	Ingestion	Inhalation	Dermal
			HQ	HQ	HQ
Fe	19014.57	Child	2.48E-01	8.00E-07	6.95E-04
		Adult	2.66E-02	1.71E-07	1.75E-04
Pb	210.87	Child	4.81E-01	2.09E-05	1.03E-02
		Adult	5.16E-02	4.48E-06	2.59E-03
Cd	1.43	Child	1.31E-02	8.73E-06	7.33E-05
		Adult	1.40E-03	1.87E-06	1.85E-05
Zn	13.6	Child	4.14E-04	7.26E-05	5.80E-06
		Adult	4.44E-05	1.56E-05	1.46E-06
Cr	2.43	Child	1.48E-05	2.82E-05	3.19E-06
		Adult	1.59E-06	6.03E-06	8.06E-07
Ni	0.001	Child	2.54E-07	2.14E-09	2.54E-09
		Adult	2.72E-08	4.59E-10	6.41E-10

Conc.: concentration; E-: x 10[^]

Table 15. Hazard quotients (HQ) of heavy metals in Anka town-Anka Local Government, Zamfara State

Metals	Mean metal Conc. (mg/kg)	Age group	Ingestion	Inhalation	Dermal
			HQ	HQ	HQ
Fe	11576.8	Child	1.51E-01	4.87E-07	4.23E-04
		Adult	1.62E-02	1.04E-07	1.07E-04
Pb	0.0007	Child	1.52E-06	6.61E-11	3.25E-08
		Adult	1.63E-07	1.42E-11	8.20E-09
Cd	1.2	Child	1.10E-02	7.30E-06	6.14E-05
		Adult	1.17E-03	1.57E-06	1.55E-05
Zn	0.0003	Child	1.01E-08	1.78E-09	1.42E-10
		Adult	1.09E-09	3.81E-10	3.59E-11
Cr	0.0017	Child	1.01E-08	1.93E-08	2.19E-09
		Adult	1.09E-09	4.13E-09	5.52E-10
Ni	0.002	Child	9.13E-07	7.71E-09	9.13E-09
		Adult	9.78E-08	1.65E-09	2.31E-09

Conc.: concentration; E-: x 10[^]

Total hazard index (THI)

The health risk exposure (THI) obtained from the cumulative HQ (Figure 2) showed that the children are the most vulnerable category for health risk exposure to soils across all the study locations. Children's THIs were consistently higher than adults' through all the analyzed exposure (ingestion, dermal contacts, and inhalation) pathways. All the THIs observed for the studied heavy metals in adults and THI inhalation and dermal contact for children were generally less than 1.0. This implies that a little or no adverse health risk in all the study locations from heavy metals through the above pathways will be observed. However, only the THI value through ingestion for children exceeded 1.0 (1.10) at the Bagega study location, indicating an adverse non-cancer health risk for children due to ingestion of heavy metals (Tepanosyan et al., 2017).

Moreover, the THI through ingestion for children in the other artisanal mining study

locations were all approaching one (i.e., 0.74, 0.71, 0.6, and 0.5 at Abare, Dareta, Sunke, and Tunga location, respectively). This indicates that a steady increase in human activities in the abovementioned locations could pose adverse health risks in the near future. Hence, immediate remediation measures need to be taken to curb the trends. Uwah et al. (2020) reported similar findings $HI < 1$ in their study locations except for Agip, which was greater than 1. Moreover, Anka town revealed the lowest THI through ingestion for children, which is by far < 1 , showing that the higher values obtained in other locations were due to mining. The values recorded in all the locations (except Anka town) were within the values range (0.5 – 1.2) reported in disturbed agricultural soil by pipeline construction in China (Shi et al., 2014). Although, Ying et al. (2016) reported much higher values (1.16 – 1.36) in soil from a free open dumpsite. The assessment was only based on soil exposure; other pathways like food and water intakes were not included.

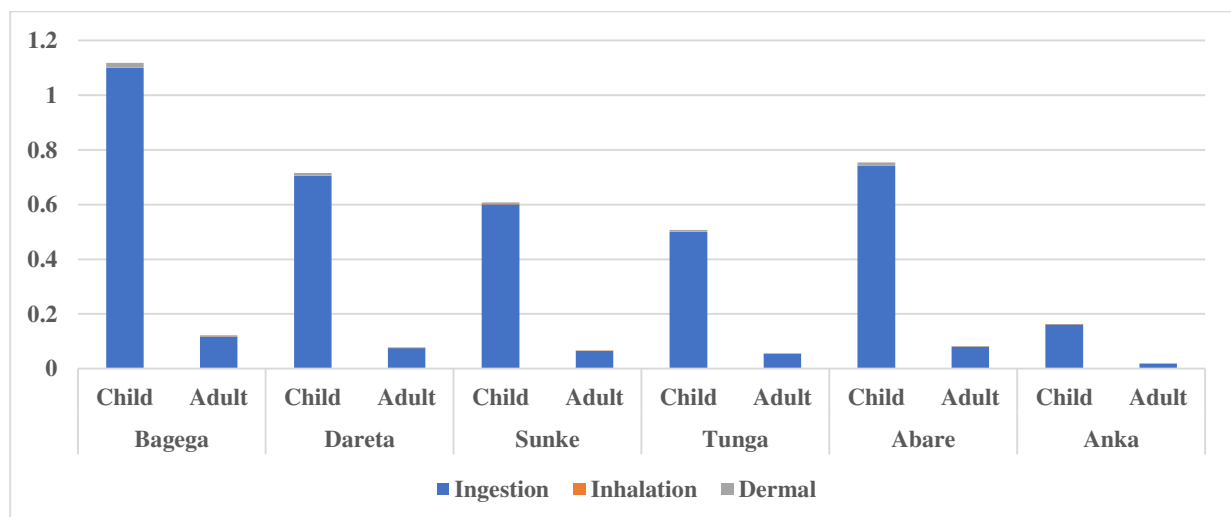


Figure 2. Total hazard index of heavy metals contamination Anka Local Government, Zamfara State.

CONCLUSION

Human health risk assessment indices (i.e., Hazard quotient and Total hazard index) were considerably below the potential to cause human risk hazards, except the THI at Bagega that has a value > 1.0 for children ingestion of heavy metals (i.e., a non-cancer

health risk for ingestion of heavy metals exists). Also, all the values recorded in the remaining study locations Dareta, Sunken, Tunga, and Abare (except for Anka Town) have a THI ingestion of children approaching one (1.0). This indicates that most human health risks will be due to ingestion of soil

particles rather than inhalation or dermal contact with the soil particles. This can be inferred that, in both the studied group of population (i.e., adults and children), the ingestion pathway was the highest contributor to the non-cancer risk followed by dermal contact then inhalation pathway. Therefore, policymakers need a systematic approach to establish and strengthen mining regulations to protect the environment and inhabitants, more importantly, children from heavy metals pollution in the area. Application of appropriate remediation measures is also recommended to restore the contaminated areas.

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