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Assessment of health risk due to the exposure of heavy metals in soil around mega coal-fired cement factory in Nigeria

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ABSTRACT

Mobilization and dispersion of potentially toxic elements into the atmosphere and human environment due to industrial and anthropogenic activities have been associated with significant human health challenges. In this investigation, 20 surface soil samples collected around a coal-fired cement factory in northeast Nigeria were analysed for their heavy metal (Cr, Pb, Ni, Cu, Zn and Mn) concentrations using inductively coupled plasma mass spectrometry. The results showed that mean concentrations of heavy metals, except for Cr were lower than their normal backgrounds ($Cr = 76.44 > 64\,mg\,kg^{-1}$, $Pb = 19.32 < 70\,mg\,kg^{-1}$, $Ni = 29.09 < 50\,mg\,kg^{-1}$, $Cu = 5.03 < 63\,mg\,kg^{-1}$, $Zn = 10.15 < 200\,mg\,kg^{-1}$) provided in the Canadian soil quality guidelines. Potential health risk assessment for adults and children for lifetime exposure through ingestion, inhalation and dermal contact were estimated. Statistical analysis identified anthropogenic activities as the principal source of metal contamination in the studied soils. Risk assessments indicated that ingestion pathway is the primary exposure route for both adults and children. Children were found to be prone to higher health risk possibly due to their hand-to-mouth dietary habits. Carcinogenic and non-carcinogenic health risk values were within safety limits for all the metals, though Cr showed a high potential for occurrence of non-carcinogenic health effects in the subpopulations.

Introduction

Mobilization of heavy metals and potentially toxic pollutants in human environment by anthropogenic activities has attracted much attention due to their indestructible and non-degradable nature, couple with their toxicity and impacts on human health [1–4]. Coal combustion and cement production have been identified as two principal sources of heavy metals mobilization in the atmosphere [5–9]. They have become the primary anthropogenic routes by which human population is exposed to higher level of metal loads above normal background [10,11]. Coal combustion has long been associated with soil contamination and environmental pollution problems, ranging from air and water contamination to human health hazards [8,12]. During combustion process, potentially toxic metals bound in coal are mobilized and released into the surrounding environment through atmospheric emissions from the stack and from leaching of combustion products [12–17]. Similarly, cement production has a long history of environmental unfriendliness due to emissions of cement dust laden with toxic air pollutants [18,19]. These metals when released, are spread over large areas by the wind and finally deposited in soils. Human population are exposed to soil pollutants through three principal pathways: direct ingestion of contaminated soil; inhalation and dermal contact through exposed skin [20–23]. Heavy metals when finally localized in human tissues and circulatory system induces respiratory and cardiovascular diseases, including asthma and lung cancer [24–26]. Some are associated with organ failures and nervous/endocrine breakdown [23,27]. Okedeyi et al. [19], reported that impaired reproduction and retardation in children development could be linked to heavy metal contamination while Freedman et al. [28] associated heavy metal pollution with brain damage and nervous

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breakdown. Heavy metal risks generally are more prevalent in children possibly due to their hand-to-mouth dietary habits [22].

Ashaka Cement factory Plc (AshakaCem) is one of the leading cement production industries in North-eastern Nigeria that depends solely on coal for power generation. Dust emissions from cement production couple with emissions of organic and inorganic pollutants from coal combustion makes AshakaCem an industry of great environmental concern. Although emission control systems have been installed in the factory, the collection efficiency of the dust filters cannot be sufficient enough to prevent the escape of gaseous pollutants into the soil. Emissions from vehicles and trucks engaged in transportation activities around AshakaCem also contributed significantly to the total soil contamination which the factory workers and the public are exposed to continually. The health risk associated with the long term exposure can be mutagenic, teratogenic or carcinogenic which can lead to high mortality rate [23,29]. It has therefore become necessary to investigate the concentration of these heavy metals in soil around AshakaCem and to assess their human health risk. Although many studies all over the world have focused on concentration and health risk assessment of heavy metals in the environment, little is known about the level of metal pollution and the associated human health hazards from Ashaka cement production factory. This is therefore a pilot study with the objective of assessing the severity of soil pollution due to emissions from the coal-fired AshakaCem and to ascertain the extent of exposure through the three exposure pathways for both children and adults. The results of this investigation will help in developing a quantitative estimate of the probability that any of the risks associated with metal toxins will be realised in different subpopulations. The findings will assist the factory workers in adopting effective and safe protective measures against industrial pollution. It will also help the relevant regulatory authorities in their policy formulation and planning of control mechanisms to achieve a better environmental quality.

Materials and method

Sampling and sample preparation

Ashaka cement factory Plc (AshakaCem) is located at $10^0 55'49''$N and $11^0 28'34''$E in Gombe, northeast Nigeria. AshakaCem which assumed full continuous operation since 1979 is currently the largest cement producing factory in Northern Nigeria. About 90% of the energy requirements of the factory is realised from coal combustion. The company installed dust bag filters as pollution control systems to reduce the hazardous emissions from coal combustion and cement production processes in the factory.

Twenty soil samples (0–15 cm depth) were randomly collected around AshakaCem factory in Gombe state, northeast Nigeria. The coordinates of each point were recorded using the global positioning system (GPS). The samples were carefully packed in neatly labelled polyethylene bags to avoid sample mixture and transported to the laboratory for analysis.

The soil samples were air-dried at room temperature for 72 h in the laboratory after which they were sieved through < 2 mm mesh, pulverized and thoroughly homogenized before digestion. Each sample was digested on hot plate following a 3-step acid digestion procedure. 10 ml of 1:1 HNO_3 were added to 1.00 ± 0.01 g of soil sample in 250 ml volumetric flask. The slurry was covered with a watch glass and refluxed for 15 min without boiling at 95 ± 5 °C. The sample was refluxed again after cooling with addition of 5 ml concentrated HNO_3 (70%) repeatedly until no brown fumes were observed and was allowed to cool after which 2 ml of deionized water and 3 ml of 30% H_2O_2 in steps of 0.5 ml were added carefully without any loss (to a maximum of 10 ml). The entire mixture was heated on hot plate until effervescence subsided. The sample was allowed to cool and was filtered through Whatman No. 41 filter. The filtered digestate was transferred into 100 ml volumetric flask and filled up to mark with deionized water. The final solutions were stored at 4 °C for analysis.

Instrumentation

The elemental analysis was carried out at the ICP-MS laboratory, Chemistry department, University of Malaya, Malaysia using 7500 series inductively coupled plasma mass spectrometry (ICP-MS) supplied by Agilent Technologies, USA. All equipments used for sample preparation were thoroughly washed with 15% HNO_3 (v/v) and distilled water. Blanks and standards were prepared in the same way as the samples with accurate dilution. Multi-element standard solutions ($1\,g\,l^{-1}$) for ICP-MS (Agilent Technologies, USA, part No. 8500-6940) were diluted appropriately to prepare calibration stock solution of $10\,mg\,l^{-1}$ for each of the analysed elements. This was used for instrument calibration prior to analysis. Minimum elemental detection limit (MEDL) were computed as concentrations analogous to three times the standard deviation of the blank signals for each analyte. The MEDL ($\mu g\,g^{-1}$) of 0.03, 0.01, 0.03, 0.02 and 0.02 were obtained for Cr, Pb, Ni, Cu and Mn respectively. Each sample (prepared in triplicate) were analyzed for Cr, Pb, Ni, Cu, Zn and Mn. The sample injection system for the ICP-MS consists of a nebulizer and spray chamber (temperature controlled) which are coupled to an auto-sampler. The responsiveness of the ICP-MS was preserved by keeping the operation conditions constant throughout the period of measurement. For purpose of reproducibility, all samples were prepared and analysed in triplicate and results expressed as the mean with 95% level of confidence. The obtained results were compared with SLR-4 reference material (National Research Council, Canada) of certified values for numerous trace metals and certified reference material for ICP (Merek). The results showed good agreement with certified values with range of recoveries varying between 83 and 103%.

Potential health risk assessment model

The probability that any of the hazards associated with heavy metal pollution could be realized in any exposed population is the basis for health risk assessments [30]. Health risk assessment model developed by the United States Environmental Protection Agency, USEPA for soil screening guidance was adopted in this study to estimate the degree of metal exposure for children and adults from soil samples around AshakaCem. The risk assessment parameters adopted in this research are presented in Table 1, while reference doses (*RfDs*) and cancer potency factors (CPFs) are shown in Table 2. Human exposure to metals in soils occur via three principal pathways: direct ingestion (DI_{ing}), inhalation (DI_{inh}) and dermal absorption through exposed skin (DI_{derm}). Health risks suffered from metal contamination are dependent on the toxicity of the pollutants and the extent of exposure [34]. Human exposure is estimated by the daily intake of each potential toxic metal separately, through individual exposure pathway.

The daily intake (DI) for each pathway, expressed in $mg\,kg^{-1}\,day^{-1}$ is calculated from the equation [31]:

$$DI_{ing} = C \times \frac{R_{ing} \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{1}$$

$$DI_{inh} = C \times \frac{R_{inh} \times EF \times ED}{BW \times PEF \times AT} \tag{2}$$

$$DI_{derm} = C \times \frac{ABF \times SA \times SAF \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{3}$$

where R_{ing} and R_{inh} are the ingestion and inhalation rates respectively and other parameters are as defined in Table 1.

The lifetime average daily dose (LADD) in $mg\,kg^{-1}\,day^{-1}$, used for the assessment of carcinogenic risks for Cr, Pb and Ni inhalation pathway for both children and adults is calculated from the equations [26]

Table 1
Risk assessment parameters of heavy metals adopted in this study.

ID	Parameter	Value used			References
		Common	Child	Adult	
AT	Average time for non-carcinogens (days/year)	365	–	–	USEPA [31]
EF	Exposure frequency (days/year)	350	–	–	USEPA [31]
DF	Dilution factor indoor	0.4	–	–	Grzetic and Ghariani [32]
PEF	Particulate emission factor (m^3/kg)	$1.36x10^9$	–	–	USEPA [31]
ABF	Dermal absorption factor for all metals	0.001	–	–	De Miguel et al [33]
IR	Ingestion rate (mg/day)	–	200	100	USEPA [38]; Grzetic and Ghariani [33 31,32]
ED	Exposure duration (years)	–	6	30	Grzetic and Ghariani [32]
IN	Inhalation rate (m^3/day)	–	7.6	20	Zheng et al [22]
SA	Exposed skin surface area (cm^2)	–	2800	5700	USEPA [31]
SAF	Skin adherence factor for soil ($mg/cm^2/h$)	–	0.2	0.07	USEPA [31]
BW	Body weight (kg)	–	15	70	USEPA [31]

Table 2
*RfDs (*reference dose) and CPFs (cancer potency factor) of heavy metals ($mg\,kg^{-1}\,day^{-1}$) adopted in this study, values adopted from De Miguel et al. [33]; Xu et al. [26].

Metals	Cr	Pb	Ni	Cu	Zn	Mn
RfD_{ing}	3.00×10^{-3}	3.50×10^{-3}	2.00×10^{-2}	4.00×10^{-2}	3.00×10^{-1}	4.60×10^{-2}
RfD_{inh}	2.86×10^{-5}	3.52×10^{-3}	2.06×10^{-2}	4.02×10^{-2}	3.00×10^{-1}	1.43×10^{-5}
RfD_{dermal}	6.00×10^{-5}	5.25×10^{-4}	5.40×10^{-3}	1.20×10^{-2}	6.00×10^{-2}	1.84×10^{-3}
CPFs	$4.20 \times 10^{+1}$	4.20×10^{-1}	8.40×10^{-1}			

$$LADD_{child} = C \times \frac{EF \times R_{inh-child} \times ED_{child}}{BW_{child} \times PEF \times AT} \tag{4a}$$

$$LADD_{adult} = C \times \frac{EF \times R_{inh-adult} \times ED_{adult}}{BW_{adult} \times PEF \times AT} \tag{4b}$$

where C ($mg\,kg^{-1}$) is the metal concentration in the soil sample. Other parameters are as defined in Table 1.

Risk characterization

The non-carcinogenic health risk to any exposed human being which expresses the systematic toxicity for a single metal in soils is a dimensionless quantity called the hazard quotient (HQ). Toxicological risk of exposure to metals is assessed by comparing the estimated daily intake of each metal to its corresponding reference dose (*RfD*) in a given exposure pathway. It is expressed as:

$$HQ = \frac{DI}{RfD} \tag{5}$$

Human health risk due to carcinogenic elements is calculated as "the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen" [32]. It is a dimensionless level of probability expressed as:

$$Risk_{cancer} = DI \times CPF \tag{6}$$

where DI ($mg\,kg^{-1}\,day^{-1}$) is the daily intake calculated for each metal for a particular pathway using Eqs. (1)–(3), *RfD* and CPF ($mg\,kg^{-1}\,day^{-1}$) are the reference dose and cancer potency factor respectively defined for each metal (Table 2). *RfD* is the chronic reference dose for a given toxic metal above which any daily human (including sensitive subpopulations) exposure through any of the exposure pathways could result in deleterious risk during a lifetime [32,35]. Thus, if a DI value for a given pollutant through any exposure pathway is higher than its corresponding *RfD* (i.e. HQ > 1), there is a likelihood of adverse non-carcinogenic health effect through that exposure route [36,37]. Cancer potency factor (CPF) on the other hand relate exposure to the probability of incurring any carcinogenic effect [34,38]. All risks whether non-carcinogenic or carcinogenic, are additive. The sum of HQ values for different pollutants and/or multiple exposure routes is called the chronic non-carcinogenic hazard index (HI) and is expressed as [22,26,39]:

$$HI = \sum_{i=1}^{n} HQ_i = \sum_{i=1}^{n} \frac{DI_i}{RfD_i} \tag{7}$$

where HQ_i, DI_i and RfD_i are the HQ value, DI value and RfD respectively for the i^{th} metal. If HI < 1, the risk of non-carcinogenic health effects is negligible. HI > 1 indicates the likelihood of occurrence of non-carcinogenic effects, the probability which increases with increasing HI [31].

Similarly, the cumulative carcinogenic risk which describes the probability of an individual developing cancer from simultaneous exposure to multiple carcinogenic pollutants is expressed as [32]:

$$Total\ \ cancer\ \ risk = \sum_{i=1}^{n} DI_i \times CPF_i \tag{8}$$

where DI_i and CPF_i are the daily intake and cancer potency factor respectively for the i^{th} pollutant. The precautionary range for carcinogenic risk provided for regulatory purposes is $1E-06 – 1E-04$ [40,41]. Hazard index and cancer risk methods were used in this study to assess cumulative human health exposure risks from heavy metals via the three exposure pathways in soil samples around AshakaCem. This study assume all site-base parameters for all metals (Table 1) to be constant for all calculations made. The risks were also estimated based on total lifetime population exposure approximation of 70 years. Although there are uncertainties with risk assessment models, they have become indispensable tools in establishing the relationship between human health and metal toxicity by which we can quantify both carcinogenic and non-carcinogenic health effects through any exposure framework [23].

Results and discussion

Metal concentrations

Concentrations of six heavy metals contained in the soil samples around AshakaCem, northeast Nigeria are presented in Table 3. Descriptive summary including the minimum, the maximum, the mean and the standard deviation along with their respective kurtosis and

Table 3
Descriptive statistics of heavy metal concentrations (mg kg^{-1}) in soil samples around AshakaCem.

	Co-ordinate			Metal concentration (mg/kg)					
Sample ID	Long	Lat	pH	Cr	Pb	Ni	Cu	Zn	Mn
ASS1	10° 56′ 06″	11° 28′ 33″	7.4	101	15.6	36.5	5.8	6.3	814.5
ASS2	10° 55′ 46″	11° 28′ 21″	7.5	104	26.6	44.7	9.6	11.7	570.7
ASS3	10° 55′ 38″	11° 28′ 44″	7.8	97.2	19.7	34.1	15.1	23.5	995.6
ASS4	10° 55′ 35″	11° 28′ 21″	7.7	95.8	29.2	35.6	4.01	20.3	361.6
ASS5	10° 55′ 37″	11° 28′ 59″	7.2	92.7	37.2	32.6	6.5	9.0	322.2
ASS6	10° 55′ 35″	11° 29′ 01″	7.7	85.9	23.9	30.7	4.5	3.2	141.1
ASS7	10° 55′ 28″	11° 28′ 33″	7.0	90.4	39.7	37.7	6.9	23.1	1080
ASS8	10° 55′ 16″	11° 28′ 37″	7.7	82.7	10.5	31.3	2.3	4.1	338.5
ASS9	10° 55′ 34″	11° 28′ 17″	7.2	81.7	15.3	31.8	3.5	4.0	240.0
ASS10	10° 55′ 33″	11° 28′ 15″	8.0	74.5	15.0	28.1	2.8	3.7	284.1
ASS11	10° 55′ 38″	11° 28′ 14″	7.8	80.7	14.4	31.2	4.9	3.4	373.0
ASS12	10° 55′ 40″	11° 28′ 17″	7.6	61.6	20.3	24.1	7. 8	11.6	298.1
ASS13	10° 55′ 42″	11° 28′ 51″	7.5	63.3	24.6	22.4	4.8	35.7	326.9
ASS14	10° 55′ 45″	11° 28′ 48″	7.6	67.4	13.9	22.5	2.9	4.6	266.6
ASS15	10° 56′ 00″	11° 28′ 53″	7.1	55.0	6.1	21.0	3.4	0.4	336.7
ASS16	10° 56′ 03″	11° 28′ 54″	7.3	62.6	20.1	30.1	6.8	15.7	822.0
ASS17	10° 56′ 12″	11° 28′ 47″	8.0	57.7	12.3	22.7	1.5	4.8	744.6
ASS18	10° 56′ 26″	11° 28′ 43″	7.7	60.1	19.1	23.3	2.8	7.8	363.1
ASS19	10° 53′ 39″	11° 28′ 08″	7.1	56.3	10.8	20.2	3.1	8.0	325.6
ASS20	10° 53′ 20″	11° 28′ 11″	7.1	58.3	12.3	21.3	1.6	1.8	304.9
Mean ± SD				76.4 ± 16.6	19.3 ± 8.8	29.1 ± 6.8	5.03 ± 3.2	10.1 ± 9.2	466 ± 271
Skewness				0.20	0.94	0.47	1.78	1.46	1.19
Kurtosis				−1.51	0.56	−0.34	4.17	1.83	0.16

skewness are also presented. The pH values obtained for the studied soils were relatively similar, varying between 7.0 and 8.0 (Table 3), which give an indication of neutral to sub-alkaline soil conditions around AshakaCem.

The most abundant metal in the studied soils is Mn followed by Cr and Ni. Pb, Zn and Cu recorded average concentrations below 20 mg kg^{-1}.The mean concentration values showed a trend of low metal loads in ascending order of Cu < Zn < Pb < Ni < Cr < Mn and were all below the baseline (B-value) values (Pb = 19.32 < 70 mg kg^{-1}, Ni = 29.09 < 50 mg kg^{-1}, Cu = 5.03 < 63 mg kg^{-1}, Zn = 10.15 < 200 mg kg^{-1}) provided in the Canadian soil quality guidelines for agricultural soils (CCME, 2007), except for Cr (Cr = 76.44 > 64 mg kg^{-1}). These values also falls within the range reported for similar studies by other authors as seen in Table 4. Furthermore, all the studied heavy metals except Cr and Ni recorded kurtosis values above zero suggesting a steeper than normal distribution of the metals in the soil samples. Also, the skewness values recorded for Cu, Zn and Mn were above 1, indicating that they are positively skewed towards lower concentrations (Table 3).

Concentration of Cr varies from 55.03 to 103.90 mg kg^{-1} with an average value of 76.44 mg kg^{-1}. Concentration of Cr in the studied soil samples can be attributed to its release by friction, wear and tear from the linings of rotaries used in cement industries [42]. According to Gržetić and Ghariani [32], Cr (VI) will always dominate in a system with redox potential of 400 mV for a pH of 7.0–8.0. Aerated soils exhibits redox potential of up to 400 mV. Therefore, since pH plays a vital role in the distribution and mobility of heavy metals in soils [6,34], it could be assumed that Cr (VI) is the dominant chromium specie in the studied soil samples.

The concentrations of Pb were found to be in the range of 6.06–39.67 mg kg^{-1} with a mean value of 19.32 mg kg^{-1}. Pb in the studied soils is traceable to its release from coal during combustion for energy generation. Mining and processing of lime stone within the vicinity of the cement plant could have contributed in no small measure to the presence of Pb in the studied soil samples. Odoh et al. [9] reported that leaded gasoline still remains the major energy source for most vehicles and trucks in Nigeria, which accounted for about 2800 metric tons of Pb deposited annually on urban soils from vehicular emissions. Pb accumulation in the studied soils may therefore have resulted from emissions from exhausts of heavy trucks and vehicles that transports raw materials into, and finished products out of the cement factory [42,43].

Cu concentration varied between 1.48 and 15.11 mg kg^{-1}, with an average value of 5.08 mg kg^{-1} (Table 3). The level of Cu in the studied soil samples might have been influenced by emissions from the cement plant and from exhausts of trucks and vehicles involved in different operations around the cement factory [9]. Corrosion of metallic wastes from the cement factory that are dumped all around the factory and mechanical aberrations of the vehicles are additional potential sources of Cu in soils [44].

Average concentration of Zn in all the studied soil samples was 10.14 mg kg^{-1}. This value was found to be lower than those reported in similar investigations (Table 4). Carreras and Pignata [43], Al-Khashman [45] and Ellis and Revitt [46], associated Zn in soils with vehicular emissions, mechanical aberrations and wear and tear of vulcanized vehicle tyres. Corrosion of galvanized automobile parts,

Table 4
Comparison of heavy metal concentrations (mg kg^{-1}) of the present study and other authors.

Country	Technique(s) used	Cr	Pb	Ni	Cu	Zn	Mn	References
Belgrade, Serbia	AAS	70.23	350.06	123.67	122.29	268.37	641.8	Gržetić and Ghariani [32]
Catalonia, Spain	ICP-MS, AAS	10.3	16.4	11.3	27.6	38.2	213.7	Schuhmacher et al. [34]
Riyadh, Saudi Arabia	ICP-AES	9.5	4.27	–	3.8	15.22	–	Al-Oud et al. [18]
Volta Region, Ghana	EDXRF	961	13.3	245.26	27.97	35.02	544.92	Addo et al. [3]
Sagamu (Nigeria)	AAS	156.6	666.1		613.4	188.5		Ogunkunle and Fatoba [50]
Gombe (Nigeria)	ICP-MS	76.44	19.32	29.09	5.03	10.14	465.49	Present study

lubricants and atmospheric depositions from coal burning processes are additional probable sources of accumulation of Zn and Cu in soils [47].

Ni in the studied soil samples recorded concentration values in the range of 20.19–44.72 mg kg^{-1} with mean concentration value of 29.09 mg kg^{-1}. Cement production and other anthropogenic activities around the factory could be responsible for Ni accumulation in the studied soils. Cempel and Nikel [48] reported that industrial depositions and agricultural activities accumulate Ni in surface soils. Human exposure to Ni at elevated dose in soils can results in some pathological effects including lung fibrosis, skin allergies and cancer. Ni concentration in soil samples around AshakaCem should therefore be given strict attention.

Mn was found to be the most abundant element in the studied soil samples with average concentration of 465.49 mg kg^{-1}. Mn is usually present under natural conditions in relatively high concentration in soils [49]. Hence, its presence in the studied soils may not be attributable to any anthropogenic processes neither can Mn be considered a contaminant. The spatial distribution of these metals demonstrates the influence of cement production activities, coal combustion processes and vehicular emissions around the vicinity of AshakaCem

Statistical analysis

Multivariate statistical analysis which involves Pearson's correlation matrix and Factor analysis was performed on the heavy metals composition. This is to enable us establish any inter-element relationships existing among the metals in the studied soil samples. These relationships provides very useful information about the source (s) and pathways of metals in any system.

Pearson's correlation

Pearson's correlation matrix for heavy metals in soil samples around AshakaCem are presented in Table 5. Cr showed significantly positive correlation ($r^2 = +0.930$) with Ni. This strong relationship indicated a common contamination source possibly anthropogenic and coal combustion processes for the metals. Pb exhibited relatively strong positive relationship with Ni ($r^2 = +0.571$) and Zn ($r^2 = +0.585$), attesting to their common origin from corrosion of metallic wastes, vehicular emissions and pollution due to cement production processes. Mn on the other hand showed weak correlation with all the studied metals except Cu ($r^2 = +0.563$), indicating that Mn and Cu were obtained from the local soil (natural source) [8,49], though Cu accumulation could be influenced by anthropogenic activities.

Factor analysis (FA)

For the purpose of clarity and accuracy of information about the source and distribution of heavy metals in soils, FA was performed in which the extracted correlation matrix was subjected to Varimax rotation with Khaisar normalization. The results of the factor loadings obtained for the Varimax rotation along with their respective percentage of variance and communalities are presented in Table 5. Two significant factors which explained a cumulative variance of 75.20% were extracted. Factor 1 explains about 39% of the total variance and is heavily loaded on the elements Cr and Ni. This factor identified the influence of anthropogenic activities including coal combustion and lime stone processing activities within the vicinity of the factory as the principal source of the metals. This is further corroborated by the significantly positive correlation coefficient between Cr and Ni (Table 5). Zn, Mn, Cu and Pb showed strong positive loadings in factor 2 and

Table 5
Pearson correlation matrix and Varimax rotated factor loadings of heavy metals in soil samples around AshakaCem.

Pearson correlation matrix							Factor analysis			
	Cr	Pb	Ni	Cu	Zn	Mn		Factor 1	Factor 2	Communality
Cr	1.000							**0.96**	0.17	0.95
Pb	0.545	1.000						0.50	0.57	0.58
Ni	0.930	0.571	1.000					**0.95**	0.23	0.95
Cu	0.545	0.424	0.556	1.000				0.47	**0.67**	0.67
Zn	0.204	0.585	0.222	0.515	1.000			0.00	**0.91**	0.83
Mn	0.322	0.285	0.441	0.563	0.424	1.000		0.26	**0.68**	0.53
							Variance (%)	39.28	35.92	
							Cumulative (%)	39.28	75.20	

Table 6
Daily intake of heavy metals from the studied soil samples by children and adults through the three exposure routes.

		Children					Adults				
Element		DI_{ing}	DI_{inh}	DI_{derm}	Total	LADD	DI_{ing}	DI_{inh}	DI_{derm}	Total	LADD
Cr	Min	4.24×10^{-3}	1.18×10^{-7}	1.18×10^{-5}	4.25×10^{-3}	1.18×10^{-7}	2.26×10^{-3}	3.33×10^{-7}	9.02×10^{-6}	2.27×10^{-3}	3.32×10^{-7}
	Max	8.00×10^{-3}	2.23×10^{-7}	2.23×10^{-5}	8.02×10^{-3}	2.22×10^{-7}	4.27×10^{-3}	6.28×10^{-7}	1.70×10^{-5}	4.29×10^{-3}	6.28×10^{-7}
	Mean	5.89×10^{-3}	1.64×10^{-7}	1.64×10^{-5}	5.90×10^{-3}	1.64×10^{-7}	3.14×10^{-3}	4.62×10^{-7}	1.25×10^{-5}	3.15×10^{-3}	4.62×10^{-7}
Pb	Min	4.67×10^{-4}	1.30×10^{-8}	2.25×10^{-6}	4.69×10^{-4}	1.30×10^{-8}	2.49×10^{-4}	3.66×10^{-8}	1.72×10^{-6}	2.51×10^{-4}	3.66×10^{-8}
	Max	3.04×10^{-3}	8.50×10^{-8}	8.52×10^{-6}	3.05×10^{-3}	8.49×10^{-8}	1.63×10^{-3}	2.40×10^{-7}	6.50×10^{-6}	1.64×10^{-3}	2.40×10^{-7}
	Mean	1.49×10^{-3}	4.14×10^{-8}	4.05×10^{-6}	1.49×10^{-3}	4.14×10^{-8}	7.94×10^{-4}	1.17×10^{-7}	3.09×10^{-6}	7.97×10^{-4}	1.17×10^{-7}
Ni	Min	1.55×10^{-3}	4.33×10^{-8}	4.34×10^{-6}	1.55×10^{-3}	4.32×10^{-8}	8.30×10^{-4}	1.22×10^{-7}	3.31×10^{-6}	8.33×10^{-4}	1.22×10^{-7}
	Max	3.44×10^{-3}	9.58×10^{-8}	9.61×10^{-6}	3.45×10^{-3}	9.57×10^{-8}	1.84×10^{-3}	2.70×10^{-7}	7.33×10^{-6}	1.85×10^{-3}	2.70×10^{-7}
	Mean	2.24×10^{-3}	6.23×10^{-8}	6.25×10^{-6}	2.25×10^{-3}	6.23×10^{-8}	1.20×10^{-3}	1.76×10^{-7}	4.77×10^{-6}	1.20×10^{-3}	1.76×10^{-7}
Cu	Min	1.14×10^{-4}	3.17×10^{-9}	3.18×10^{-7}	1.14×10^{-4}		6.08×10^{-5}	8.95×10^{-9}	2.43×10^{-7}	6.11×10^{-5}	
	Max	1.16×10^{-3}	3.24×10^{-8}	3.25×10^{-6}	1.17×10^{-3}		6.21×10^{-4}	9.13×10^{-8}	2.48×10^{-6}	6.24×10^{-4}	
	Mean	3.87×10^{-4}	1.08×10^{-8}	1.08×10^{-6}	3.88×10^{-4}		2.07×10^{-4}	3.04×10^{-8}	8.24×10^{-7}	2.07×10^{-4}	
Zn	Min	2.70×10^{-5}	7.50×10^{-10}	7.52×10^{-8}	2.70×10^{-5}		1.44×10^{-5}	2.12×10^{-9}	5.74×10^{-8}	1.44×10^{-5}	
	Max	2.74×10^{-3}	7.66×10^{-8}	7.68×10^{-6}	2.75×10^{-3}		1.47×10^{-3}	2.16×10^{-7}	5.86×10^{-6}	1.48×10^{-3}	
	Mean	7.80×10^{-4}	2.17×10^{-8}	2.18×10^{-6}	7.83×10^{-4}		4.17×10^{-4}	6.13×10^{-8}	1.66×10^{-6}	4.18×10^{-4}	
Mn	Min	1.08×10^{-2}	3.02×10^{-7}	3.03×10^{-5}	1.09×10^{-2}		5.80×10^{-3}	8.53×10^{-7}	2.31×10^{-5}	5.82×10^{-3}	
	Max	8.28×10^{-2}	2.31×10^{-6}	2.32×10^{-4}	8.31×10^{-2}		4.44×10^{-2}	6.53×10^{-6}	1.77×10^{-4}	4.46×10^{-2}	
	Mean	3.57×10^{-2}	9.98×10^{-7}	1.00×10^{-4}	3.58×10^{-2}		1.91×10^{-2}	2.81×10^{-6}	7.63×10^{-5}	1.92×10^{-2}	

accounted for 36% of the total variance. This factor suggested that both natural (local soil) and anthropogenic inputs are the principal sources of accumulation of these metals in the studied soil samples.

Health risk assessment

The daily intake (DI) and the LADD of heavy metals from soil samples around AshakaCem for both children and adults via the three exposure pathways were calculated using Eqs. (1)–(4b) and the results presented in Table 6. The total maximum DI of Cr and Pb for the children are 8.02×10^{-3} and 3.05×10^{-3} mg kg^{-1} day^{-1} respectively while for the adults are 6.28×10^{-3} and 2.40×10^{-7} mg kg^{-1} day^{-1} respectively. The cumulative maximum exposure doses from Ni and Cu are 3.45×10^{-3} and 1.17×10^{-3} mg kg^{-1} day^{-1} respectively for children and 1.85×10^{-3} and 6.24×10^{-4} mg kg^{-1} day^{-1} respectively for adults. The overall maximum DI for Zn are 2.75×10^{-3} and 1.48×10^{-3} mg kg^{-1} day^{-1} respectively for children and adults.

From the children perspective, the DI of all the studied metals via ingestion route are 2–4 order of magnitude higher than the inhalation and dermal routes. Also, average values of the total daily intake of Cr, Pb and Ni are one order of magnitude higher than for Cu and Zn. Similarly, the daily intake of the metals are 2–4 order of magnitude higher for ingestion pathway compared to inhalation and dermal contacts in adults. However, the mean values of total exposure dose for Cr and Ni are in the same order of magnitude (10^{-3} mg kg^{-1} day^{-1}) which is higher than that of Pb, Zn and Cu (10^{-4} mg kg^{-1} day^{-1}) for adults.

Mn on the other hand recorded the highest mean daily intake of 3.58×10^{-2} and 1.92×10^{-2} respectively for children and adults. Comparison of the DIs with their respective *RfDs* for the three exposure routes showed that DI values for both subpopulations were lower than the respective *RfD* values for all the studied metals except for Cr. This showed that both children and adults were not under any threat of potential non-carcinogenic risk from contamination by these metals. The trend is however different for Cr. The DI_{ing} values for Cr for both children and adults are 1.96 and 1.05 mg kg^{-1} day^{-1} respectively. These values were above the *RfD* of 3.00×10^{-3} mg kg^{-1} day^{-1} for Cr provided by USEPA [40]. Serious attention should therefore be given to the ingestion of Cr which is present in its carcinogenic form (Cr^{+6}) in the studied soil samples. General observation of the results (Table 6) showed that ingestion route contributed about 99% of the total daily intake of metals for both children and adults. Soil ingestion is therefore the dominant metals exposure pathway for both subpopulations. A comparison of the exposure rate between the two subpopulations showed that children are more exposed to metal contamination in the studied soil samples than the adults. This may be due to their daily hand-to-mouth dietary habits.

For carcinogenic metals (Table 6), the average value for daily intake (LADD) of 1.64×10^{-7} mg kg^{-1} day^{-1} recorded for Cr was found to be higher than for Pb (4.18×10^{-8} mg kg^{-1} day^{-1}) and Ni (6.23×10^{-8} mg kg^{-1} day^{-1}). The LADD for Cr, Pb and Ni were however in the same order of magnitude (10^{-7} mg kg^{-1} day^{-1}) for adults.

Health risk levels

Hazard quotients (HQ) for the three exposure routes together with the non-carcinogenic and carcinogenic risk (HI) estimates for both children and adults were calculated using Eqs. (5)–(8). The results obtained are presented in Table 7. The HQ and HI for non-carcinogenic risk for both children and adults followed the same decreasing trend as Zn < Cu < Ni < Pb < Mn < Cr.

Additionally, the HQ values for the three exposure pathways portrayed an increasing order of ingestion > dermal contact > inhalation, except for Mn whose increasing order was ingestion > inhalation > dermal contact for both adults and children.

Table 7
Non-carcinogenic (three exposure routes) and carcinogenic (inhalation) risk for children and adults.

	Children					Adults				
Elements	HQ_{ing}	HQ_{inh}	HQ_{derm}	$HI = \sum HQ_i$(non⁻carcinigenic)	Carcinogenic risk	HQ_{ing}	HQ_{inh}	HQ_{derm}	$HI = \sum HQ_i$(non– carcinigenic)	Carcinogenic risk
Cr	$1.96 \times 10^{+00}$	5.73×10^{-3}	2.74×10^{-1}	$2.24 \times 10^{+00}$		$1.05 \times 10^{+00}$	1.62×10^{-2}	2.09×10^{-1}	$1.27 \times 10^{+00}$	
Cr_{cancer}					6.87×10^{-6}					1.94×10^{-5}
Pb	4.25×10^{-1}	1.18×10^{-5}	7.71×10^{-3}	4.33×10^{-1}		2.27×10^{-1}	3.32×10^{-5}	5.89×10^{-3}	2.33×10^{-1}	
Pb_{cancer}					1.74×10^{-9}					4.90×10^{-9}
Ni	1.12×10^{-1}	3.03×10^{-6}	1.16×10^{-3}	1.13×10^{-1}		5.98×10^{-2}	8.54×10^{-6}	8.83×10^{-4}	6.07×10^{-2}	
Ni_{cancer}					5.23×10^{-8}					1.48×10^{-7}
Cu	9.68×10^{-3}	2.68×10^{-7}	9.00×10^{-5}	9.77×10^{-3}		5.16×10^{-3}	7.56×10^{-7}	6.87×10^{-5}	5.23×10^{-3}	
Zn	2.60×10^{-3}	7.24×10^{-8}	3.63×10^{-5}	2.64×10^{-3}		1.39×10^{-3}	2.04×10^{-7}	2.77×10^{-5}	1.42×10^{-3}	
Mn	7.76×10^{-1}	6.98×10^{-2}	5.43×10^{-2}	9.00×10^{-1}		4.16×10^{-1}	1.97×10^{-1}	4.15×10^{-2}	6.54×10^{-1}	
Total				$3.70 \times 10^{+00}$					$2.23 \times 10^{+00}$	
Mean				6.17×10^{-1}	2.31×10^{-6}				3.71×10^{-1}	6.51×10^{-6}

Contribution to the total risk from soil ingestion pathway (QI_{ing}) is about 89% for children and 79% for the adults. This further confirmed that ingestion of metals from the studied soil samples is the main exposure route responsible for non-carcinogenic risk in both subpopulations. The results agree with those from similar studies [22,26,35]. Furthermore, the HI values for Pb, Ni, Cu, Zn and Mn for both children and adults were found to be lower than unity (safety limit), indicating that the non-carcinogenic risks to adults and children due to these metals in soil samples around AshakaCem are insignificant. Cr however recorded higher toxicological risk of exposure for both children and adults in this study. The cumulative hazard indexes due to Cr intake via the three exposure routes was 2.24 for children and 1.27 for adults (Table 7). These values exceed the safety limit of unity which therefore make Cr an element of great concern. This concern becomes more severe as Cr is present in studied soils in its highly toxic Cr (VI) specie which has greater absorption rate of 2–8% when ingested [50].

Cumulative hazard index due to metal exposure via the three exposure pathways studied were 3.70 for children and 2.23 for adults (Table 7), of which ingestion of Cr contributed about 99%. This results showed that the potential non-carcinogenic risk due to heavy metal exposure is higher for the children than for the adults. This could probably be due to their hand-to-mouth dietary habits [22], whereby contaminated soil can readily be ingested [51].

The results of carcinogenic risks resulting from exposure to Cr, Pb and Ni in the studied soil samples presented in Table 6, showed that cancer risk levels for Cr, Pb and Ni were 6.87×10^{-6}, 1.74×10^{-9} and 5.23×10^{-8} respectively for children, and 1.94×10^{-5}, 4.9×10^{-9} and 1.48×10^{-7} respectively for adults. The average cumulative cancer risks from exposure to Cr, Pb and Ni in the studied soil samples were 2.31×10^{-6} and 6.51×10^{-6} respectively for children and adults. This showed that the probability of children getting cancer from exposure to these three carcinogenic metals is about 0.4 times higher than for adults. The cancer risk levels of Cr for children (6.87×10^{-6}) and adults (1.94×10^{-5}) were found to exceed the lowest precautionary limit of 1×10^{-6} set by USEPA [40]. Ingestion of Cr constituted about 99% of the total cancer risk. This make Cr an element of concern in the studied soils. Exposure for very long period may accumulate Cr in human tissues resulting in gastrointestinal effects. Any excessive build-up of Cr in human bodies can trigger lung and stomach cancer [23].

Generally however, the carcinogenic risk levels for both children and adults falls within the acceptable safety range of 1×10^{-6} to 1×10^{-4} set by USEPA [40,41]. Hence these metals cannot possibly induce any carcinogenesis in both children and adults living around AshakaCem.

Conclusions

Twenty soil samples collected at random around a coal-fired cement factory (AshakaCem) in north-eastern Nigeria were examined for their heavy metal concentrations. The results showed a decreasing order in metal concentrations as Mn > Cr > Ni > Pb > Zn > Cu, though their mean concentration values are within the range reported in literature for similar studies. Statistical analysis identified coal combustion, cement production processes and vehicular emissions as main sources of these metals in the studied soils. Human health risk analysis identified soil ingestion route as the exposure pathway with the highest risk level for both children and adults. Estimated non-carcinogenic risk for both children and adults via the three exposure routes were within acceptable safety levels except for Cr whose HI is higher than 1. This suggested that Cr should be given attention and deep concern. The overall cumulative HI value was found to be higher for children, indicating that children are more at risk of metal contamination than the adults. The cancer risk levels for the carcinogenic metals was not outside the safety range provided by USEPA, showing that the probability of carcinogenesis as a result of metals in the studied soils is negligible. Although there are uncertainties associated with risk assessment models, they have proved to be useful tools in identifying potent exposure pathway(s) that may be of serious concern in evaluating the overall potential health impacts to any population from metal contamination. The overall results of this research will assist government in their policy formulation and in adopting a risk-oriented approach for the protection of man and his environment from the effects of heavy metal pollution.

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Conflicts of interest

No conflicts of interest to be declared.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rinp.2018.10.003.

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