EVALUATION OF RADIOACTIVITY AND HEAVY METAL CONCENTRATIONS IN SOIL AROUND A COAL-FIRED CEMENT FACTORY OF NORTHEAST NIGERIA: IMPLICATIONS ON HUMAN HEALTH AND ENVIRONMENT

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ABSTRACT

Anthropogenic activity has become a major source of radiological contamination and heavy metal pollution in the environment. Coal from Maiganga coal mine Gombe is currently the main source of energy for AshakaCem, a mega cement factory in northeast Nigeria. Periodic monitoring of radioactive and heavy metal concentrations in soil around AshakaCem is thus of utmost importance from human health and environmental protection perspective. In this study, natural radioactivity and heavy metal contents in soil around AshakaCem were assessed using gamma-ray spectrometry and inductively coupled plasma mass spectrometry (ICP-MS). Mean specific activities of ^{226}Ra, ^{232}Th and ^{40}K were 6.41, 16.63 and 167.31 Bq kg^{-1} respectively. Mean values of the evaluated hazard parameters were within safety limits provided for human and environmental protection. Average concentrations of Cr, Pb, Ni, Cu, Zn and V were below their respective background level provided by the Canadian soil quality guidelines for agricultural soils, except for Cr. Average pollution load index for all the studied heavy metals showed lower values than unity. Therefore, activities of the coal-fired cement factory does not make any significant change to the radioactivity and heavy metal contents in the surrounding environment. Results of this study could serve as an important baseline for subsequent radiological studies and environmental monitoring activities in future.

Keywords: Natural radioactivity, heavy metals, gamma absorbed dose, pollution load index, AshakaCem, northeast Nigeria.

Introduction

Increasing demand for energy to satisfy the growing challenge of industrialization and urbanization has led to rapid consumption of fossil and nuclear fuels. As a results, pollutants which are mostly heavy metals and radioactive nuclides are continuously released into the environment sometimes at concentrations that become dangerous to human health and environmental quality. Soil has become the primary sink for these pollutants in terrestrial environment (Çayır *et al.*, 2012). When present in agricultural soils at concentrations above the normal threshold, they impede natural

biological phenomena and enhance soil toxicity (Ogunkunle and Fatoba, 2013). Direct ingestion of crops grown on polluted soils thus become potential sources of human exposure to radioactive nuclides and heavy metal contamination. Continuous monitoring of agricultural soils with regards to natural radioactivity and heavy metal contents is therefore necessary for protection of man and the environment.

Anthropogenic activities have been identified as major sources of radionuclide and heavy metal mobilization in the atmosphere (Odoh, 2014; Yi *et al.*, 2008). Coal combustion and cement production in particular are critical anthropogenic pathways for population exposure to heavy metals and radioactivity above normal background (Gbadebo and Bankole, 2007). Environmental unfriendliness of coal combustion and cement production has been due to emission and release of coal and cement dust loaded with toxic heavy metals and radioactive nuclides into the atmosphere. Once released, they are dispersed over large areas by atmospheric processes and subsequently deposited in the surrounding soils (Agrawal *et al.*, 2010; Al-Khashman and Shawabkeh, 2006; Al-Oud *et al.*, 2011), thereby enhancing their concentrations in surface soils. It is therefore paramount to investigate the radionuclide and heavy metal contents of soil samples around a coal powered cement production industry and to evaluate their contribution to soil pollution and possible risk to human health. While this has become a major focus in many countries, very little is known about the environmental impact of Ashaka cement factory in north-eastern Nigeria. The primary objective of this study therefore is the assessment of natural radioactivity and heavy metal concentrations in soil samples around Ashaka cement factory, northeast Nigeria. Radiation hazard parameters and pollution load index will be evaluated to determine the level of soil pollution and human exposure. Data from this study will form part of baseline guide for further studies in assessing the environmental impacts of cement production in Nigeria.

Materials and Methods

Sample site

Ashaka cement factory Plc (AshakaCem) is located at $10^0 55' 49'' N$ and $11^0 28' 34'' E$ in Bajoga, Funakaye Local Government Area of Gombe State, 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 especially in firing the cement kiln is realised from coal combustion. Cement production and coal combustion processes releases huge quantities of potentially hazardous air pollutants into the surrounding environment thereby endangering the health of factory workers and the surrounding communities from continuous constant exposure. The company has however installed dust bags filters as pollution control systems to reduce the hazardous emissions from the factory.

Sample collection, preparation and analysis

10 surface soil samples (0-15 cm) were collected at random around AshakaCem in Gombe, northeast Nigeria. Each sample, about 1.0 kg in weight was thoroughly screened of all foreign matters and carefully packed in well labelled polyethylene bags and transported to the laboratory for analysis. The soil samples were air-dried at room temperature for 72 hours in the laboratory to attain constant weight, after which they were sieved, pulverized and thoroughly homogenized.

Homogenized samples were carefully prepared for radiometric analysis using HPGe gamma spectrometer at the radiation laboratory, department of Physics, University of Malaya, Malaysia. 370±1.0g of the homogenized samples were neatly packaged into well-labelled Marinelli beakers at the laboratory and tightly sealed to prevent escape of radon.

Sealed samples were kept for about thirty seven days to attain radiological equilibrium between daughter nuclides and their respective long-lived parents (Amin *et al.*, 2013; Khandaker *et al.*, 2012).

Radiometric analysis of the samples was carried out using a P-type Coaxial ORTEC, GEM-25 HPGe gamma ray detector with 57.5 mm crystal diameter and 51.5 mm thickness. The detector was enveloped in thick cylindrical lead shield to reduce the interference of background radiation from terrestrial and extra-terrestrial sources with the measured spectrum (Amin *et al.*, 2013; Kolo *et al.*, 2015). The detector which has relative efficiency of 28.2% and 1.67 keV FWHM energy resolution at 1.33 MeV peak of ^{60}Co, was coupled to ADCM data acquisition system with PCAII multi-channel analyser and set at operating voltage of +2800 V. Energy and efficiency calibrations were done on the detector prior to analysis using a cylindrical multi-nuclide gamma ray source with homogenously distributed activity in the same container geometry as the samples. The calibration source which has initial activity of 5.109 μCi contain the following nuclides: ^{241}Am (59.541 keV), ^{109}Cd (88.040 keV), ^{57}Co (122.061 keV, 136.474 keV), ^{203}Hg (279.195 keV), ^{113}Sn (391.698 kεV), ^{85}Sr (514.007 keV), ^{137}Cs (661.657 keV), ^{88}Y (898.042 keV, 1836.063 keV), and ^{60}Co (1173.22 keV, 1332.492 keV).The source was supplied by Isotopes Products Laboratories, (Valencia, CA 91355). Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the analysed soil samples were computed using the equation (Cevik *et al.*, 2010; Khandaker *et al.*, 2012; Kolo *et al.*, 2015):

$$A(Bq\ kg^{-1}) = \frac{CPS \times 1000}{\varepsilon_\gamma \times I_\gamma \times W} \qquad (1)$$

where A (Bq kg^{-1}) is the specific activity, CPS is the net counts per second for each sample investigated, ε_γ (E) is the detector photo-peak efficiency at respective gamma-ray peak, I_γ is the corresponding gamma ray intensity and W is the sample mass in g.

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. 1.0±0.01 g of each soil sample was digested for heavy metal determination in 250 ml flask on hotplate following a 3-step acid digestion procedure. 1.00±0.01 g of soil sample was mixed with 10 ml of 1:1 HNO_3 in 250 ml volumetric flask. The mixture was covered with a watch glass and refluxed for 15 minutes at 95^{0}C±5^{0}C without boiling. The sample was refluxed again after cooling with addition of 5 ml concentrated HNO_3 (70 %) repeatedly until no brown fumes were observed. It was then allowed to cool after which 2 ml of deionized water and 3 ml of 30 % H_2O_2 were added carefully in steps of 0.5 ml 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^{0}C for analysis. The instrument was calibrated prior to analysis with multi element calibration standard 2A solution with analyte concentration of 10 mg kg^{-1} (Agilent Technologies, USA, part No. 8500-6940). The elemental detection limit was 0.0001 mg kg^{-1}.

Radiation hazard indices

Radium equivalent activity (Ra_{eq}) was defined to account cumulatively for the hazards associated with ^{226}Ra, ^{232}Th and ^{40}K in soil samples (Beretka and Mathew, 1985; UNSCEAR, 2000). Ra_{eq} was estimated by the equation (Ravisankar *et al.*, 2014; UNSCEAR, 2000):

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq kg^{-1}.

Absorbed dose rate in air (D_R) at 1 m above ground level due to gamma ray emissions from ^{226}Ra, ^{232}Th and ^{40}K in soils was calculated from the equation (UNSCEAR, 2008):

$$D_R(nGy\ h^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq kg^{-1}.

Exposure risk to any individual due to absorbed dose rate is estimated in terms of the annual effective dose equivalent (AEDE). AEDE was calculated by applying the conversion factors of 0.70 Sv Gy^{-1} which converts absorbed dose rate in air to effective dose and the outdoor occupancy factor of 0.2, assuming that an individual spends average of 20 % of his time outdoors (UNSCEAR, 2000). AEDE in outdoor air, measured in mSv y^{-1} was evaluated using the equation:

$$AEDE\ (mSv\ y^{-1}) = D_R \times 1.21 \times 10^{-3} \quad (4)$$

Excess lifetime cancer risk is evaluated to determine the probability of risk of cancer to any population from exposure to radiation. It was calculated based on the estimated AEDE using the equation (Ravisankar *et al.*, 2014; Taskin *et al.*, 2009):

$$ELCR = AEDE \times DL \times RF \quad (5)$$

where DL is the life duration of 70 years and RF is risk factor given to be 0.05Sv^{-1} for stochastic effects in any given population (ICRP-60, 1990; Taskin *et al.*, 2009).

Results and Discussion

Natural radioactivity and radiation hazard parameters

Descriptive statistical results (minimum, maximum, mean, standard deviation, skewness and kurtosis) and the coefficients of variance of natural radionuclides in soil samples around AshakaCem are presented in Table 1. Also presented in Table 1 are the radium equivalent activity (Ra_{eq}), gamma absorbed dose rate (D_R), annual effective dose equivalent (AEDE) and excess lifetime cancer risk (ELCR) incurred by the population from exposure to natural radionuclides in soil samples. Mean specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples were 6.41, 13.63 and 167.31 Bq kg^{-1} respectively. These values are lower than their respective world average values of 35, 30 and 400 Bq kg^{-1} for normal soils set by UNSCEAR (2000). Distribution of radionuclides in the studied soil samples appears to be steeper than normal as demonstrated by the skewness and the kurtosis values. Ra_{eq} values ranged between 22.52 and 47.83 Bq kg^{-1} with an average of 38.78 Bq kg^{-1} which is below the recommended precautionary limit of 370 Bq kg^{-1} (UNSCEAR, 2000). Furthermore, calculated D_R has a mean value of 18.15 nGy h^{-1}, with a corresponding mean AEDE value of 0.02 mSv y^{-1}. These values are lower than their respective mean global safety limits of 59 nGy h^{-1} and 0.07 mSv y^{-1} (UNSCEAR, 2000). Similarly, average of 0.8 x 10^{-4} calculated for ELCR is below world average value of 0.29 x 10^{-3} set by UNSCEAR (UNSCEAR, 2000). The overall results of radioactivity contents of soil samples around AshakaCem are generally low, thus reflecting the normal radiation background trend.

Table 1. Descriptive statistics of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, Ra_{eq}, D_R, AEDE and ELCR of soil samples around AshakaCem

	Activity (Bq kg^{-1})				Dose		
	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	D_R (nGy h^{-1})	AEDE (mSv y^{-1})	ELCR (10^{-3})
Min	3.68	9.15	74.77	22.52	10.30	0.01	0.04
Max	8.83	15.66	222.87	47.83	22.50	0.03	0.10
Mean	6.41	13.63	167.31	38.78	18.15	0.02	0.08
SD	1.39	2.27	47.92	7.72	3.72	0.01	0.02
Skew	-0.34	-0.89	-1.07	-1.20	-1.22	-0.13	-0.94
Kurt	1.11	-0.25	0.47	1.01	1.06	0.18	0.87
CV (%)	21.68	16.65	28.64	19.90	20.49	50.00	25.00

SD = standard deviation
CV = coefficient of variance

Heavy metal concentration and pollution level assessment

Descriptive statistics of heavy metal concentrations and pollution load index of heavy metals in soil samples around AshakaCem are presented in Table 2. Also presented in Table 2 are the baseline values (B-value) for heavy metals. There is yet no established screening levels for heavy metals in Nigerian soils, thus the B-values used in this study were adopted from the Canadian soil quality guidelines for the protection of environmental and human health (CCME, 2007). Concentrations of heavy metals in the studied soil samples followed and increasing order of Cu<Zn<Pb<V<Ni<Cr with mean concertation values of 6.09, 10.88, 23.26, 24.88, 34.32 and 90.56 mg kg^{-1} respectively. These values are lower than their corresponding B-values in Canadian agricultural soils except for Cr whose mean concentration is about 1.2 times its B-value (Table 2).

Table 2. Descriptive statistics of heavy metal concentrations (mg kg^{-1}) of soil samples around AshakaCem

	Cr	**Pb**	**Ni**	**Cu**	**Zn**	**V**	**PLI**
Min	74.50	10.48	28.09	2.27	3.15	16.39	0.15
Max	103.90	39.67	44.72	15.11	23.47	39.80	0.34
Mean	90.56	23.26	34.32	6.09	10.88	24.88	0.25
SD	9.29	9.87	4.67	3.85	8.33	7.72	0.08
Skew	-0.27	0.53	1.14	1.59	0.73	0.64	0.00
Kurt	-0.77	-0.88	1.91	2.76	-1.36	-0.26	-1.86
CV (%)	10.26	42.43	13.61	63.21	76.56	31.03	32.00
B-Value	64.00	70.00	50.00	63.00	200.00	130.00	

SD = standard deviation
CV = coefficient of variance
B-Value = Baseline values for heavy metals in agricultural soil (CCCME, 20017

High concentration of Cr may be due to its release from rotary linings by friction, wear and tear during cement production processes (Al-Khashman and Shawabkeh, 2006). The influence of anthropogenic activities on heavy metal contents and their degree of dispersion in soils are demonstrated by their respective standard deviations (SD) and coefficients of variation (CV) (Lu *et al*., 2013; Suresh *et al*., 2011). Zn, Cu and Pb recorded higher CV values of 76.6, 63.1 and 42.4% respectively. This is attributable to the influence of coal combustion and cement production activities in AshakaCem. Lu *et al* (2013), and Al-Oud *et al* (2011) also attributed enhanced concentrations of Pb, Cu and Zn in soils to anthropogenic activities such as cement production and coal combustion.

Pollution load index (PLI) was evaluated to assess the environmental quality and total degree of contamination of soil samples around AshakaCem by heavy metals. PLI is a comparative index which defines the pollution status and toxicity of any given environmental medium (Suresh *et al*., 2011). PLI was evaluated following the equation (Lu *et al*., 2013):

$$PLI = \left[\prod_{j=1}^{n} \frac{C_j}{BV_j}\right]^{\frac{1}{n}} \qquad (6)$$

where C_j is concentration of heavy metal j in the studied soil and BV_j is the B-value for heavy metal j. PLI <1 indicates insignificant soil pollution while PLI >1 show that the soil is polluted. Calculated PLI values of heavy metals in soil samples around AshakaCem are generally low, ranging between 0.15 and 0.34 with an average value of 0.25 (Table 2). This low PLI values show that heavy metal contamination of soil around AshakaCem is relatively insignificant.

Multivariate Statistical Analysis

Radiological variables, heavy metal concentration and PLI for soil samples around AshakaCem were subjected to statistical analysis using a statistical software package, SPSS 22.0. This was necessary to establish any interdependency and mutual relationships that may exist among all the variables.

Pearson correlation analysis

Pearson correlation coefficients for the analysed variables are presented in Table 3. Very strong positive relationship is observed between Cr and Ni (r^2 = 0.83), Cr and V (r^2 = 0.70) and Ni and V (r^2 = 0.76). This highly positive relationship presupposes that Cr, Ni and V in the investigated soil samples have a common origin and exhibits identical behaviours during their transport through soil medium (Suresh *et al*., 2011). All the studied heavy metals correlates positively with PLI ($0.66 \leq r^2 \leq 0.83$), indicating that Cr, Pb, Ni, Cu, Zn and V contributes mutually to metal pollution in soil samples around AshakaCem.

Similarly, significantly positive correlation exist between the primordial radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the soil samples and the radiation hazard parameters ($0.68 \leq r^2 \leq 0.98$). This shows that gamma dose incurred by the population in the vicinity of the AshakaCem is principally due to exposure to ^{226}Ra, ^{232}Th and ^{40}K present in the surrounding soil.

Principal component analysis (PCA)

The data set obtained in this study was subjected to principal component analysis to identify any possible underlying factor responsible for the relationship that may exist. The PCA was performed using varimax rotation method with Kaiser Normalization. The obtained factor loadings with their respective percentage of variance and communalities are shown in Table 4. Three principal components were delineated with eigenvalues greater than 1 which accounts for 87.71% of the total variance. Component 1 accounts for 46.02% of the total variance and is heavily loaded positively on activity

concentrations of primordial radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in association with radiological variables. This indicate that the radioactivity level in the surrounding soil of AshakaCem is determined by the concentrations of ^{226}Ra, ^{232}Th and ^{40}K. Component 2 which explains 21.25% of the total variance is very strongly correlated with Pb, Zn and PLI, with positive loading values of 0.72, 0.96 and 0.81 respectively (Table 4). Component 3 is characterized primarily by high positive loading of Cr, Ni and V and accounts for 20.4% of the total variance. This component show a common contamination source possibly anthropogenic for these metals. This result is further corroborated by significantly positive relationship between Cr, Ni and V (Table 3). Figure 1 gives a three-dimensional pictorial representation of the three loading components. The results of the PCA are well corroborated with Pearson correlation analysis.

Table 3. Pearson correlations matrix of measured variables in soil samples around AshakaCem

	Cr	Pb	Ni	Cu	Zn	V	PLI	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	D_R	AEDE	ELCR
Cr	1.00													
Pb	0.37	1.00												
Ni	**0.83**	0.40	1.00											
Cu	**0.64**	0.24	0.48	1.00										
Zn	0.51	0.57	0.47	0.65	1.00									
V	**0.70**	0.33	**0.76**	0.53	0.17	1.00								
PLI	0.78	0.69	0.75	0.80	0.83	0.66	1.00							
^{226}Ra	0.53	-0.10	0.29	0.40	0.04	0.37	0.24	1.00						
^{232}Th	0.44	-0.11	0.31	0.44	0.08	0.29	0.24	0.92	1.00					
^{40}K	0.46	0.06	0.11	0.26	0.08	0.20	0.20	0.80	0.70	1.00				
Ra_{eq}	0.50	-0.03	0.24	0.38	0.08	0.28	0.24	0.95	0.92	0.92	1.00			
D_R	0.51	-0.02	0.23	0.37	0.09	0.28	0.24	0.94	0.91	0.93	1.00	1.00		
AEDE	0.57	0.13	0.24	0.48	0.51	0.16	0.46	0.79	0.68	0.81	0.82	0.83	1.00	
ELCR	0.50	0.00	0.21	0.35	0.13	0.20	0.23	0.95	0.91	0.90	0.98	0.98	0.86	1.00

The overall results together with statistical analysis show that coal combustion and cement production activities does not have overbearing influence on the natural radionuclide and heavy metal concentrations in soil samples around AshakaCem. Coefficients of variation of ^{226}Ra, ^{232}Th and ^{40}K are generally low indicating their natural abundance in soil. Positive relationship between Pb and Zn (Tables 3 and 4) suggest their common origin. Activity concentrations of primordial radionuclides in the soil samples are lower than their mean values for normal soils documented by UNSCEAR (2000).

Table 4. Verimax rotated factor loadings of measured variables in soil samples around AshakaCem

	Component			
Variables	1	2	3	Communalities
Cr	0.40	0.44	**0.70**	0.85
Pb	-0.13	**0.72**	0.21	0.58
Ni	0.09	0.35	**0.86**	0.86
Cu	0.31	0.58	0.45	0.64
Zn	0.06	**0.96**	0.09	0.93
V	0.13	0.12	**0.93**	0.89
PLI	0.13	**0.81**	0.57	0.99
226Ra	**0.94**	-0.05	0.27	0.95
232Th	**0.89**	-0.04	0.25	0.86
40K	**0.91**	0.09	0.02	0.84
Raeq	**0.98**	0.02	0.16	0.99
DR	**0.98**	0.03	0.15	0.99
AEDE	**0.85**	0.45	-0.01	0.92
ELCR	**0.98**	0.07	0.09	0.98
Eigenvalues	7.55	3.52	1.21	
% of variance	46.02	21.25	20.44	
Cummulative %	46.02	67.27	87.71	

Extraction method: Principal Component Analysis

Rotation method: Varimax with Kaiser Normalization

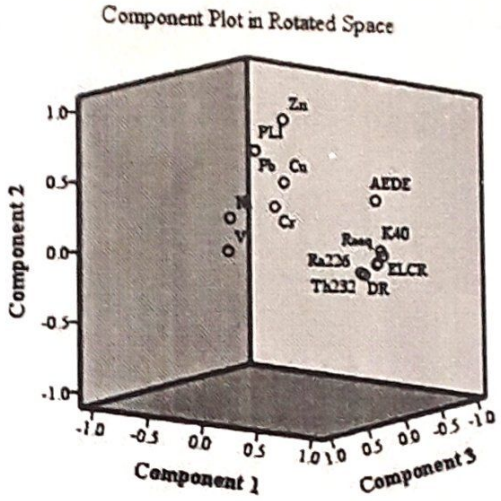

Figure 1. Rotated factor loadings of component-1 (46.02%), component-2 (21.25%) and component-3 (20.44%).

Furthermore, concentrations of heavy metals in the studied soil samples are lower than their respective B-values for agricultural soils given by CCME (2007). Higher concentration of Cr in the soil samples may be due to coal combustion and cement production activities.

Conclusion

Mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples were below the world average values for normal soils. Estimated radiation hazard parameters were lower than the precautionary limits for radiation protection. Furthermore, concentration of heavy metals in soil samples around AshakaCem were below the background values for Canadian agricultural soils except for Cr. The corresponding PLI values for heavy metals were below unity. Contamination of soil samples around AshakaCem from natural radionuclides and heavy metals is therefore insignificant. The soil is thus fit for agricultural activities and other domestic or industrial use.

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