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Evaluation of the Activity Concentration of ^{40}K, ^{226}Ra and ^{232}Th in Soil and Associated Radiological Parameters of Shanono and Bagwai Artisanal Gold Mining Areas, Kano State, Nigeria

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ABSTRACT: Mining in the studied area is carried out without any precautionary measure. The radiological implication of this activity was reported in this work. Soil samples were collected from the gold mines and analyzed for activity concentration of naturally occurring radioactive materials (NORMs) using NaI (Tl). The average specific activity of ^{238}U, ^{232}Th and ^{40}K were 62.73, 90.66 and 411.27 Bq/kg respectively. The radiological parameters D (nGy/h), AED (mSv/y) and Ra_{eq} had average values of 100.89, 0.13 and 224.04 respectively. Furthermore, the radiological hazard indices H_{ex}, H_{in}, I_γ and I_α had average values of 0.61, 0.78, 0.86 and 0.31 respectively, which were lower than the United Nations Scientific Committee on effects of atomic radiation (UNSCEAR) recommended limit of unity. Finally, the total cancer risk due to NORMs in soil was 7.35×10^{-06} for the whole populations which was within the USEPA acceptable range of 1.00×10^{-06} to 1.00×10^{-04}. Therefore, the gold mining activity poses no significant radiological hazard to the members of the public.

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Naturally occurring radioactive materials (NORMs) refers to all the radionuclides that exist in the environment naturally, they are the primordial radionuclides ^{40}K, ^{232}Th and ^{238}U, and their decay products which are present in varying amounts within the earth crust (UNSCEAR,1993). In most cases, the concentration of NORMs in any substance in the environment is negligible, but disposal of large quantities of certain minerals (mining) containing ^{40}K and other radionuclides in the decay series of ^{232}Th and ^{238}U from the earth crust results in the radionuclides concentrations becoming elevated (Shorabi,1998). In the studied area, miners and the members of the public may be exposed to radiation during extraction, transportation and processing of the mineral ores, they may also experience internal exposures from radon, and its short-lived decay products that are airborne or ingestible dust from their surroundings (UNSCEAR,2010). Reconnaissance survey indicated that there was high incidence of small-scale artisanal gold mining activities in some villages under Shanono and Bagwai local governments of Kano State. The mine operates within the Alajawa, Kundila and Dutsen –Bakoshi villages of Shanono Local government and Ginzo village of Bagwai Local Government, Kano State. The communities surrounding the mines depend on surface water, wells and boreholes as their sources of water in addition to farming being their major occupation. Mining in the studied area is carried out by illiterate artisans across all the age range. The crude methods of obtaining the minerals may expose the miners, immediate and adjourning environments to high levels of radionuclides. The miners brought soil and stones rich in gold mineral to the surface for processing, the tailings generated are exposed to wind and the prevailing weather conditions resulting in transportation of the particles containing radionuclides to once uncontaminated areas. The populace relies on untreated ground water (shallow wells and boreholes) for drinking and other household activities which may contain high radon concentrations (UNSCEAR, 2008). This work may call the attention of the stakeholders to give more attention to the environmental impacts of NORMs and motivate further research into controlling NORMs radiation levels in the country. Therefore, this study will also allow us to analyze how much health hazards the natural radionuclides the gold mining poses to the public and the environment. The results of this study will benefit academicians, the local community, regulatory authorities, health sector, water treatment agencies, mining industry, environmental protection agency and geological survey sectors. The objective of the present paper is to report the evaluation of the activity concentration of ^{40}K, ^{226}Ra and ^{232}Th in Soil and associated radiological parameters of Shanono and Bagwai artisanal gold mining areas, Kano State, Nigeria.

MATERIALS AND METHODS

Sample collection: Reconnaissance survey indicated that the artisanal gold mining activities within Shanono and Bagwai local Governments of Kano state were carried out in four villages namely; Alajawa, Kundila, Dutsen Bakoshi (under Shanono) and Ginzo village (under Bagwai). The selection of the sampling locations was based on the accessibility to the public and proximity to the mine. The sampling strategy that was adopted for the samples collection was random (ASTM, 1983, 1986; IAEA, 2004 and USEPA, 1989). Forty (40) soil samples were collected from within the mines area, farm lands and residential villages. Each sample was sealed in a polyethylene bag, firmly tied and labelled to avoid cross contamination. The samples were transferred to the Radiation Biophysics laboratory at Centre for energy research and training, Ahmadu Bello University, Zaria.

Sample preparation: The samples were spread on cardboard sheets and all foreign materials were removed, they were then oven dried at a temperature of about 110˚C for 12-18hours. The samples were then grinded into a fine powder and sieved using 2mm sieve. The homogenized samples were filled into 25g plastic containers (7.2cm diameter by 6cm height) which were hermetically sealed with the aid of PVC tape to prevent the escape of airborne ^{222}Rn and ^{220}Rn from the samples. The dimensions of the plastic containers were chosen in such a way that it suited the optimal soil mass of 350g for analysis of bulk samples. The samples were sealed and stored for over 24 days to allow secular equilibrium to be reached between radon and its daughters. The IAEA reference materials for gamma spectrometry (RGK-1, IAEA-448 and RGTh-1) were prepared in the same manner as the samples.

Sample analysis: The samples were analysed using NaI(Tl) detector situated at low background laboratory of center for energy research and training, Ahmadu Bello university, Zaria. The detector has a 6cm thick lead shield, cadmium lined assembly with copper sheets for the detection of background radiation. The detector has pulse resolving time of about 0.25s, an incorporated preamplifier and a 1kV external source which permits its use for high counting rates. The detector was coupled to a computer based multichannel analyser Maestro program from ORTEC for the acquisition and analysis of the gamma spectra. The detector was calibrated with the prepared IAEA reference materials RGK-1, IAEA-448 and RGTh-1 for the quantitative determination of ^{40}K, ^{238}U and ^{232}Th respectively in the soil samples. Each of the prepared samples was counted for 30,000seconds in the outlined detector geometry in order to mitigate the influence of background radiation from radioactive contaminants within the shielding materials of the detector assembly. The obtained data in counts per second were converted to conventional units of Bq/kg using calibration factors to determine the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K.

Assessment of annual effective doses: Absorbed dose rate in the air (D) in nGyh^{-1} at 1 meter above the ground surface was calculated from the mean radioactivity concentrations of the ^{226}Ra (^{238}U), ^{232}Th and ^{40K} (Bqkg^{-1}) in the soil samples using equation 1 (UNSCEAR, 2000). The annual effective dose due to external gamma radiation *(AEDγ)* from ^{226}Ra (^{238}U), ^{232}Th and ^{40}K was computed using equation 2 by employing dose conversion coefficients (0.7 Sv. Gy^{-1}) and outdoor occupancy factor (0.2) (UNSCEAR, 2000). The annual effective dose (AED_{ing}) from ingestion of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K through soil, was estimated from the mean activity concentrations of each individual radionuclides using equation 3 (UNSCEAR, 2000). The total effective dose to individual members of the public was calculated using ICRP dose formula (ICRP, 2012).

$$D\left({}^{nGy}/_{h}\right) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{k}. \quad 1$$

$$\text{AED}\gamma\left(\mu\frac{\text{Sv}}{\text{y}}\right) = \text{D}\left(\frac{\text{nGy}}{\text{h}}\right) \text{x}\, 8760\text{h x}0.2 * \frac{0.7\text{Sv}}{\text{Gy}} \text{x}\, 10^{-3} \quad 2$$

$$AED_{ing}\left(\frac{mSv}{y}\right) = A_R IR_{ing} \sum_{j=1}^{3} DCF_{ing} \quad 3$$

Where, A_{Ra} , A_{Th} and A_k are the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K, respectively. The dose coefficients in units of nGyh^{-1}per Bq.kg^{-1} were taken from the (UNSCEAR, 2000). A_R= the mean activity concentration of radionuclides in a sample (Bq/kg); IR_{ing}= the soil, consumption rate per year which had a value of 100kg/year (DEA, 2010). DCF_{ing}= the effective dose coefficient in Sv.Bq^{-1} for the ingestion of natural radionuclides ^{226}Ra (^{238}U), ^{232}Th and ^{40}K with values of $4.50 \times 10^{-08}, 2.30 \times 10^{-07}$ *and* 6.20×10^{-09}respectively (ICRP, 2012).

Assessment of radiation hazard indices: Radium equivalent activity (Ra_{eq}) was used as a common radiological index to provide the actual activity level of NORMs in the sample due to their non-uniform distribution. It is a widely used index to assess the radiation hazards which was estimated on the fact that 370 $Bq.kg^{-1}$ of ^{226}Ra, 259 $Bq.kg^{-1}$ of ^{232}Th and 4810 $Bq.kg^{-1}$ of ^{40}K produce the same gamma-ray dose rate. Ra_{eq} was computed using equation 4, it has a permissible maximum value of 370 $Bq.kg^{-1}$ which corresponds to an effective dose of 1 mSv for the general public (UNSCEAR, 2000). External and internal hazard indices (H_{ex} and H_{in}) were used to estimate the external and internal hazards that could arise from the use of the soils as building material, these indices were computed using equation 5 and 6 respectively as proposed by UNSCEAR, 2000. Furthermore, Gamma and alpha indices (I_γ *and* I_α) were used to estimate the excess γ and α radiation due to NORMs and radon inhalation originating from the soil samples that could be used as building materials. They were estimated using equation 7 and 8 respectively (Asaduzzaman *et al*., 2016; Xinwei *et al*., 2006). For radiation protection purposes, each value of external, internal, gamma and alpha hazard indices must not exceed the limit of unity. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity 370 $Bq.kg^{-1}$.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k \quad 4$$

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad 5$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad 6$$

$$I_{\gamma r} = {A_{Ra}}/{150} + {A_{Th}}/{100} + {A_K}/{1500} \quad 7$$

$$I_\alpha = \frac{A_{Ra}}{200\,(\frac{Bq}{kg})} \quad 8$$

Where, A_{Ra} , A_{Th} and A_k are the activity concentration of ^{226}Ra, ^{232}Th and ^{40K} in $Bq.kg^{-1}$

Assessment of cancer and hereditary risks: The cancer and hereditary risks due to low doses were estimated using equation 9 and 10 respectively based on ICRP, 2007 cancer risk assessment methodology. The lifetime risks (70 years) of fatal cancer were based on the hypothesis of linearity of dose and effect without any threshold. The nominal risk coefficients for low doses and low dose rates were adopted from ICRP, 2007.

$$\text{FCR} = \text{total AED (Sv)} * \text{CNR factor} \ldots .9$$

$$\text{HR} = \text{total AED (Sv)} * \text{HNR factor} \ldots 10$$

Where: FCR = fatality cancer risk; CNR: Cancer nominal risk; HR = hereditary risk, HNR = hereditary nominal risk

RESULTS AND DISCUSSION

Radiological implications: Table 3 presented the summary statistics of the activity concentration (Bq/kg) of the naturally occurring radioactive materials (^{238}U/^{226}Ra, ^{232}Th and ^{40}K) in soil from the goldmines and the associated radiological parameters. The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the goldmine soils were 62.73, 90.66 and 411.27 Bq/kg respectively; this values were higher than the control values of 26.97, 33.12, 130.41 Bq/kg and also higher than the worldwide average activity concentrations values of 35, 30 and 400 Bq/kg respectively (UNSCEAR, 2000). The absorbed dose rate in air had an average value of 100.89 nGy/hr which was higher than the recommended maximum value of 59nGy/hr (UNSCEAR, 2000). Despite that, the soils were radiologically safe due to the fact that the absorbed dose rate is inconsiderate of the occupancy factor and hence could not be a direct measure of risk. The average total AED and radium equivalent activity were 0.13 mSv/year and 224.04 Bq/kg respectively; this values were lower than the UNSCEAR recommended limits of 1mSv/year and 370 Bq/kg for members of the public (UNSCEAR, 2000). The hazard indices Hex, Hin, I_γ and Iα had their average values lower than the threshold of unity, which demonstrated the validity of using the soil from the goldmines in construction and other activities. The total fatality and hereditary cancer risk was 7.35×10^{-06} for whole population ages and 5.42×10^{-06} for adults (i.e. 7 out of 1,000,000 populations and 5 out of 1,000,000 adults may be affected by fatality or hereditary cancer); values were within the US EPA acceptable cancer risk of 1.00×10^{-06} to 1.00×10^{-04}(US EPA, 1989). The present study results were in agreement with a related study carried out at selected former uranium mining and processing areas of Kazakhstan, Kyrgyzstan, Uzbekistan and Tajikistan from Central Asian countries which reported that doses of ionizing radiation did not present any serious hazard to the health of the resident public (Stegnar *et al*., 2013). Similarly, Investigations on the concentrations and distribution of natural radionuclides in soils and water with the aim of evaluating their radiological health hazards were also carried out from Sakwa Wagusu Area in Kenya, the values of the external and internal indices were found to be less than unity signifying safe levels. The calculated outdoor mean effective dose rate was 0.17 $mSv.y^{-1}$, a value less than 1 $mSvy^{-1}$(Aguko *et al*., 2013). On the contrary, the average external hazard index and internal hazard index were 2.4 and 4.5, which were much higher than the recommended maximum permissible limit of unity for the gold mine tailings in the Province of Gauteng, South Africa (Kamunda, 2017). In addition, Mangset and Sheyin, 2009 reported a very high concentration of ^{226}Ra in mine tailings from mining and milling site in the Plateau State of Nigeria.

Table 1: Summary of radiological parameters for soil.

Parameter		mean	minimum	maximum	SD
NORMs activity concentration (Bq/kg)	^{226}Ra	62.73	27.01	89.90	23.14
	^{232}Th	90.66	63.03	109.84	16.06
	^{40}K	411.27	122.66	775.21	247.07
Annual effective dose (mSv/y)	D (nGy/h)	100.89	73.67	123.21	18.97
	AEDγ	0.12	0.09	0.15	0.02
	AED_{ing}	0.01	0.01	0.01	0.00
	Total AED	0.13	0.10	0.16	0.02
Hazard indices	Ra_{eq}	224.04	166.92	272.69	40.03
	H_{ex}	0.61	0.45	0.74	0.11
	H_{in}	0.78	0.54	0.97	0.15
	Iγ	0.86	0.58	1.08	0.18
	Iα	0.31	0.14	0.45	0.12
Fatality Cancer risk ($\times 10^{-6}$)	whole	7.10	5.26	8.60	1.28
	adult	5.29	3.92	6.41	0.95
Hereditary cancer risk ($\times 10^{-6}$)	whole	0.26	0.19	0.31	0.05
	adult	0.13	0.10	0.16	0.02
Total cancer risk ($\times 10^{-6}$)	whole	7.35	5.45	8.92	1.33
	adult	5.42	4.02	6.57	0.98

Conclusion: The activity concentration of ^{238}U, ^{232}Th and ^{40}K was higher than the world average values of 35, 30 and 400 Bq/kg respectively. The average value of AED and that of Ra_{eq} were lower than the UNSCEAR recommended limits of 1mSv/year and 370 Bq/kg respectively for members of the public. The hazard indices Hex, Hin, I_{γ} and Iα had their average values lower than the threshold of unity and the cancer risk was within the USEPA acceptable range of 1.00×10^{-06} to 1.00×10^{-04}. Hence, the mining activity poses no significant radiological hazard.

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