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Determination of Radiological Hazard Indices from Surface Soil to Individuals in Angwan Kawo Gold Mining Sites, Niger state, Nigeria

[1*]ESIOLE, SO; [2]IBEANU, IGE; [1]GARBA, NN; [1]ONOJA, MA

[1]*Physics Department, Ahmadu Bello University, Zaria, Nigeria.*
[2]*Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria*
**Corresponding Author Email: stephen.esiole102@gmail.com; Tel. +2348030850208*

ABSTRACT: The determination of radiological health hazard indices from soil samples has been carried out to assess the environmental impact assessment of gold mining activities in Angwan Kawo community in Niger state, Nigeria using NaI (TI) gamma ray spectroscopy. The radionuclide concentrations were determined using the sodium iodide [NaI (TI)] detector with a low background configuration. The results obtained show that the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K ranged from (20.43±0.04 to 86.45±3.83) (Bq/kg), (19.79±0.83 to 69.80±2.60) (Bq/kg) and (52.63±1.07 to 714.32±16.25) (Bq/kg) at the goldmine and (30.54±0.48 to 82.39±5.83) (Bq/kg), (54.63±0.83 to 114.92±2.91) (Bq/kg) and (290.74±7.08 to 600.44±16.24) (Bq/kg) at the processing zone, respectively. The results obtained were compared with the world mean values of 35, 30 and 400 Bq/kg, for ^{226}Ra, ^{232}Th and ^{40}k, respectively. To assess the radiological hazard of the gold mining site the radiological hazard indices i.e., absorbed dose (D), annual effective dose equivalent (AEDE), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}) and gamma representative index (I) were calculated and found to be below the internationally recommended values. Based on this study finding, for the purpose of protection of mine workers and the public it is highly recommended that these sites should be regularly monitored and controlled in order to avoid any radiological impact to the population.

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Background radiation in soil is a fact of life, as such varies greatly depending on the geological location man finds himself. Man has being exposed to radiation continuously right from the creation of the earth. The exposure of man to ionizing radiation from natural sources is an inevitable phenomenon of life on Earth. The natural source of ionizing radiation originates from the terrestrial and cosmic origin. The assessment of these doses from natural materials is important as external radiation exposures from natural materials significantly contribute to the average annual dose to man from all radiation sources (Taskin, *et al.,* 2009). Naturally occurring radioactive materials (NORMs) are widely spread in the environment, as it tends to exist in various geological formations. Gamma radiation emitted from naturally occurring radionuclides exist as trace levels in the earth's formations, refers to the main external source of irradiation to man. This radionuclides present in soil depends mainly on geological formation of a particular region and it significantly affect terrestrial gamma radiation levels. The development of natural resources worldwide involves the manipulation of the environment to arrive at specific objectives. Mineral resources are the most exploited natural resource and it involves extracting, grinding, ore concentration and dispersal of tailings (Ferreira, *et al* ., 2004). Generation of chemical waste as a result of mining activity occurs world-wide and may severely affect natural resources such as vegetation, streams and the ecosystem in general (OECD, 1970). Radiological hazards are one of the least known effects which can be generated by these tailings through contamination processes (Esiole, 2016). Impacts of goldmines can be environmentally detrimental and can constitute serious radiological hazards to exposed or contaminated individuals. The knowledge of radionuclide distribution levels in the environment is important in assessing the effects of radiation exposure due to natural and artificial sources. It has been established by (Taskin*, et al.,* 2009) that the natural radioactivity concentration depends mainly on geological and geographical conditions and appears at different levels in soils of different geological regions. Assessment of radionuclide in soils and rocks in many parts of the world has been on the increase in the past two decades because of their hazards on the health of the populace (Belivermis, *et al.,* 2009). The assessment of gamma

**Corresponding Author Email: stephen.esiole102@gmail.com*

radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of world population (UNSCEAR, 2000). When rocks disintegrate through natural process, radionuclides are carried to soil by rain and water flow (Rahaman, 1988). The effects of radiation on humans, could lead to lung, pancreas, hepatic, bone, skin and kidney cancers, cataracts, sterility, atrophy of the kidney and leukemia (Rahaman, 1988). Many studies have been carried out Worldwide in order to determine the risks associated with long term, short term, low level and high level natural radiation exposure. It has been reported by (Ibeanu, 1999) that mining areas are highly unsafe radiological. Following the recent illegal mining in the country, that reared its ugly head once again in rural Rafi local government area in Niger state with reports of numerous deaths of children and women from lead poisoning (Fidelis, 2015). Majority of the people working in the mines and those associated with various activities within the vicinity of the mines as well as the mill processing zones are largely unaware of the health implications they are likely to be subjected to due to radiations from the exposed rock surfaces, crushed and generally, from mill tailing deposit. Ibeanu (2003) published a paper titled "Tin Mining and Processing in Nigeria: Cause for Concern". This work though made reference to only tin mining but the issue is that all radioactive minerals share the same radiological problem and otherwise. However, the degree of ignorance exhibited by the miners in small scale mining to the possibilities of exposure to ionizing radiation shows in their attitudes in and around the mines. Mining of radioactive minerals such as gold and valuable minerals, properly planned and executed in compliance with regulatory body recommendations usually yields net benefits to the licensees, the government, the residents and the society in general (Ibeanu, 2003). The knowledge on natural radioactivity present in soil in a particular region would enable one to assess any possible radiological hazard to man and his environment, hence the present study is therefore aimed at establishing a base line data on the activity concentration of natural radionuclides and the radiological risk associated with the gold mining and ore processing activities in the area. Results of this study are important for assessment of the risk for human health, planning process and policy making in Nigeria.

MATERIALS AND METHODS

Study Area: Angwan Kawo is located in Niger state, it lies between latitude $10^0$1'29''N and $10^0$1'30''N and longitude 6^0 28' 30''E to $6^0$28'31''E of Nigeria. Two sites were selected to carry out the present study. The horizontal profile soil sampling strategy was chosen for the study, since the mine site is a virgin site, hence depth profile sampling not needed. The materials used for the field measurements were hand auger, metre rule, global positioning system (GPS), gas mask, water-tight polythene bag. Soil samples were collected from 50 outdoor locations both from the gold mine, processing zone and control sites, respectively. The gold mine is 120m^2 which was divided into 40 grid points of 20m^2, 20 grid points at each horizontal level for both right and left direction of the mining pit. Also for the processing zone, the dimension of sheds on average were about 3m by 3m hence, the mill tailing site was divided into 9 grid points of 3m^2. The field measurements were made at the artisan mines and mill tailing sites, respectively. For the purpose of adequate data interpretation the locations were marked with a portable hand held global positioning system (GPS). The mine site was partitioned into $20m \times 20m$ grid point. Twenty (20) soil samples, each were taken from both right and left direction in the mine site. Soil samples were collected from 40 outdoor locations from the gridded area. Four (4) locations were measured from the direction of gridded areas in the mining pit. In the processing zone, the dimension of the sheds on average was about 3m by 3m. Most of this space is swallowed by the milling machine and its accessories, the sacs of crushed rocks for milling as well as sacs of milled materials.

Sample collection and preparation: A total of 50 samples were collected from the mine, mill tailing and undisturbed sites. The samples were taken from the depth of 10-15 cm within marked cleared area of 80m^2 and mixed thoroughly, to obtain the representative of the soil type of the area. Samples were taken in open field (i.e. away from roads, trees and buildings) in an attempt to curtail the effects of human activity (like road construction), which has been confirmed to cause artificial variation in natural environmental radioactivity in one of the world's high background natural radiation area of brazil (Garba, *et al.,* 2016); (Aliyu and Ramli, 2015). The geographical coordinates of each sampling point was recorded with a global positioning system (GPS). The collected samples were packed in polythene bags and labeled. In the laboratory, the samples were completely left open for 24 hours to dry under ambient temperature and were pulverized into a fine powder and passed through a standard mesh for the grain size fraction < 2mm. The samples were packed to fill into 25g plastic cylindrical containers with dimensions of 7.2 cm diameter and 6 cm height, which suited the optimal soil mass of 300-350g for spectrometric analysis of bulk samples. This configuration and geometry was maintained throughout the analysis. The samples were then carefully weighed and stored for a minimum of 28 days prior to measurement in order to allow radium to reach equilibrium with its daughters.

Sample analysis: The samples analysis was carried out at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria. Sodium iodide [NaI (TI)] detector in a low background configuration was used for the measurements, counted for 29,000 seconds using a low level gamma counting spectrometer comprising a 7.6 cm × 7.6 cm NaI (TI) detector housed in a 6 cm thick lead shield, cadmium lined assembly with copper sheets for reduction of background radiation which is coupled to a computer based multi-channel analyzer (MCA) through a pre-amplifier base. Calibration was done using IAEA standard reference materials i.e., RGK-1, RGU-1 and RGTh-1 for the quantitative determination of K, U and Th of the soil samples, respectively. The activity concentrations of the natural radionuclides were calculated after analyzing the spectral lines of various emissions using MAESTRO software. The system was set at a working energy range of 0-3000 KeV and energy resolution of 7.2%

RESULTS AND DISCUSSION

*Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K***:** The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of each sample from mine site, mill tailing site and control area, were determined and presented in Table 1. It can be observed that activity concentrations of radionuclides in samples from the goldmine and processing zone are strongly enhanced with respect to the soils collected from the control area. High activity concentrations of ^{232}Th and ^{40}K were observed in all samples collected from the mill tailing sites, this may be attributed to the mined minerals in the processing zone, which are characterized by a significant activity concentrations for the ^{232}Th and ^{40}K, indicating the dimensions of radionuclide contamination of the site as a result of the mining activities in the area. One possible explanation for the high activity concentrations in the goldmine and processing zone, is that during gold mining and ore processing, the radionuclides are transferred into a mill tailing deposits; as such they can even get enriched in the settled suspension. In contrast, the activity concentrations of the soil collected from the mine site were also observed to be high. It should also be noted that, compared to the control soils, the activity concentrations were observed to be lower, this observation could be attributed either to the geological factors or as a result the impact of the mining activity in the area.

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of each sample from the goldmine and processing zone are shown in Fig. 1 and 2, respectively. The highest activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K found to be (76.67 ± 3.83), (59.80 ± 2.60) and (714.32 ± 16.25) at the goldmine and (72.39 ± 5.83), (94.92 ± 2.91) and (590.44 ± 16.24) at the processing zone, respectively. Conversely, the lowest activity concentrations (Bq/kg) were observed to be (20.43 ± 0.04), (19.79 ± 0.83) and (52.63 ± 1.07) at the goldmine and (30.54 ± 0.48), (54.63 ± 0.83) and (290.74 ± 7.08) at the processing zone, respectively. It is apparent that ^{40}K exhibited the highest activity concentrations for all measured radionuclides in all of the soil samples. It can also be observed that the activity concentrations of the processing zone were higher than that of the goldmine; this variation can be attributed to the variable dumping of mill tailing deposit.

Table 1 compares the results obtained in this work with the world mean values presented by (Taskin, *et al.,* 2009). The obtained results showed that the activity concentrations of Ra-226 for the soil samples ranged from (20.43 ± 0.04) to (86.67 ± 5.83) and (30.54 ± 0.48) to (46.70 ± 2.88), respectively, which fall within the worldwide range, although the mean concentration of ^{226}Ra at the goldmine and processing zone was found to be higher than the worldwide mean. The activity concentrations (Bq/kg) of ^{232}Th and ^{40}K ranged from (19.79 ± 0.83) to (69.80 ± 2.60) and (52.63 ± 1.07) to (724.32 ± 16.25) at the goldmine, and (54.63 ± 0.83) to (104.92 ± 2.91) and (290.74 ± 7.08) to (600.44 ± 16.24) at the processing zone, respectively. The activity concentrations of ^{232}Th and ^{40}K are above the upper range of the World mean value due to the high concentrations observed at the processing zone. The mean activity concentrations (Bq/kg) of ^{232}Th and ^{40}K are (48.61 ± 2.06) and (314.17 ± 23.03), of the soil samples, are comparable with worldwide mean value while the mean activity concentrations (Bq/kg) of ^{232}Th and ^{40}K are (82.41 ± 7.18) and (482.16 ± 35.26), at the processing zone, respectively are above the worldwide mean value.

Assessment of Radiological hazard: Absorbed Dose rate in air (D): The Absorbed Dose rate (D) due to gamma radiation in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides are calculated based on guidelines provided by (Taskin, *et al.,* 2009). A direct connection between radioactivity concentrations of natural radionuclide and their exposure is known as the absorbed dose rate in the air at 1 meter above the ground surface. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bqkg^{-1}) in the soil samples are used to calculate the absorbed dose rate given by the following formula:

$D\ (nGy/h) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_k$ (1)

Where D is the absorbed dose rate in $nGyh^{-1}$, A_{Ra},A_{Th} and A_k are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The dose coefficients in units of $nGyh^{-1}$per $Bq.kg^{-1}$ were taken from the (Taskin*, et al.,* 2009).

Table 1. Mean activity concentrations of Ra-226, Th-232 and K-40 at the goldmine, processing zone and control site with the world wide mean

	Mean Activity concentration (Bq/Kg)		
Sample location	**Ra-226**	**Th-232**	**K-40**
Goldmine	53.94±3.02	48.61±2.06	314.17±23.03
Processing zone	55.39±5.07	82.41±7.18	482.16±35.26
Control site	22.25±0.48	35.42±1.07	167.84±5.71
Worldwide Range	**17-60**	**11-64**	**140-850**
Worldwide Mean	**35**	**30**	**400**

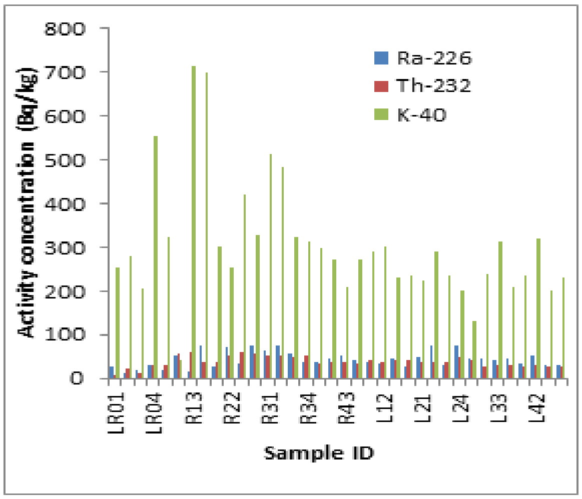

Fig. 2 Mean activity concentrations of Ra-226, Th-232 and K-40 at the goldmine

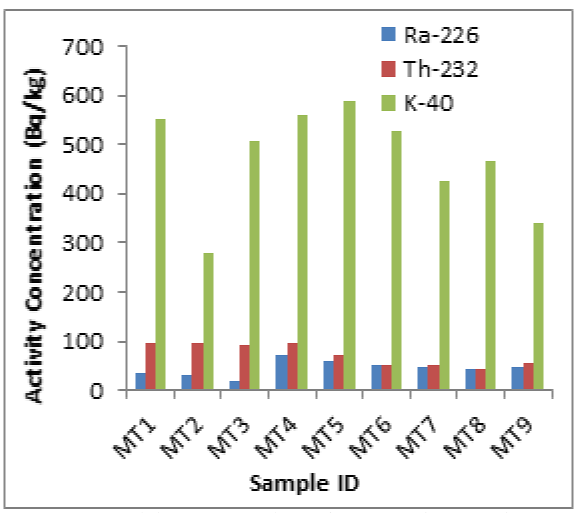

Fig. 3 Mean activity concentrations of Ra-226, Th-232 and K-40 at the processing zone

The estimated absorbed dose rates (nGy/h) of the soil samples ranged from (11.78 ± 1.55) to (86.38 ± 4.87) with a mean value of (53.94 ± 2.84) at the goldmine and ranged from (66.86 ± 1.55) to (114.18 ± 3.32) with a mean value of (84.39 ± 5.21) at the processing zone, respectively. ^{40}K is the main contributor to the absorbed dose rate in most samples in this study. Comparing the results obtained with the worldwide values, the average mean value of absorbed dose rate from all the samples in this work are higher than the worldwide mean value. Also the absorbed dose rate of the in-situ were observed to be higher than those of the computed, however this could be attributed as a result of the effect of cosmic radiation from the in-situ measurement.

Annual effective dose equivalent (AEDE): The absorbed dose rate in air at 1 meter above ground surface does not directly provide the radiological risk to which an individual is exposed (ICRP, 1990; Jibiri, 2011). The absorbed dose can be considered in terms of the annual effective dose equivalent from outdoor terrestrial gamma radiation which is converted from the absorbed dose by taking into account two factors, namely the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv/Gy) and the outdoor occupation factor (0.2) as proposed by (Taskin, *et al.,* 2009). The annual effective dose equivalent can be estimated using the following formula (Belivermis, *et al.,* 2009) , (Taskin, *et al.,* 2009):

$AEDE\ (\mu Sv/y) = D\ (nGy/h) \times 8760h \times 0.7Sv/Gy \times 0.2 \times 10^{-3}$ (2)

The values of those parameters used in the (Taskin, *et al.,* 2009) are 0.7 $Sv.Gy^{-1}$ for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.2 for the outdoor occupancy factor (Taskin, *et al.,* 2009). According to (Taskin, *et al.,* 2009), report, the dose rate in air outdoors from terrestrial gamma rays in normal circumstances is about 60nGy/h while the worldwide average effective dose is approximately 70 µSv. The annual effective dose equivalent from the outdoor terrestrial gamma radiation was estimated by taking into account the conversion coefficients from the absorbed dose in air to effective dose and outdoor occupancy factor. The effective dose (mSv/y) of the soil and mill tailing samples in the study varied from (0.01 ± 0.004) to (0.11 ± 0.005), with the mean value of (0.07 ± 0.003) at the goldmine and also ranged from (0.08 ± 0.002) to (0.14 ± 0.004), with a mean value of (0.10 ± 0.002), which is higher compared with the worldwide effective dose of 0.07 mSv/y by (Taskin, *et al.*, 2009). The acceptable annual effective dose for members of the public without constraint should be 1.0 mSv/y for safety purposes (Ibeanu, 2003). As such the radiological constraints for an adequate protection of potential users of 0.5 mSv/y, in which the results obtained from this study was compared with as recommended by (EC, 1999).

Radium Equivalent activity (Ra_{eq}): The radium equivalent activity takes into account the radiation hazards associated with ^{226}Ra, ^{232}Th and ^{40}K, which provides a useful guideline in regulating the safety standards on radiation protection for the general public. Due to a non- uniform distribution of natural radionuclide in the soil samples, the actual activity level of ^{226}Ra, ^{226}Th and ^{40}K in the samples can be evaluated by means of a common radiological index named the radium equivalent activity (Ra_{eq}) (Bertka & Matthew, 1995). It is a widely used index to assess the radiation hazards. This estimates 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. This index is mathematically defined by (Taskin, *et al.,* 2009) as:

$$Ra_{eq} = A_{Ra} + 1.43_{Th} + 0.077A_k \quad (3)$$

Where A_{Ra}, A_{Th} and A_k are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively. The permissible maximum value of the radium equivalent activity is 370 Bq.kg^{-1} (Taskin, *et al.*, 2009), which corresponds to an effective dose of 1 mSv for the general public (Ajayi, *et al.,* 2009).

The results of the radium equivalent activity (Bq/kg) of the soil and mill tailing samples ranged from (51.51 ± 3.23) to (186.35 ± 10.66), with a mean value of (122.57 ± 5.71) in the mine site and ranged from (114.01 ± 3.23) to (251.35 ± 7.36), with a mean value of (185.29 ± 11.73) in the mill tailing site, respectively. From the results obtained from this study, it can be observed that the Ra_{eq} values were lower than the accepted safety limit value of 370 Bq/kg as recommended by the Organization for Economic Cooperation and development (NBIRR, 2003), (UNSCEAR,2000), (Belivermis, *et al.,* 2009). The radium equivalent of 370 Bq/kg corresponds to the dose limit of 1mSv/y for the general population (Jibiri and Biere, 2011). Hence, the use of the soils of Angwan Kawo goldmines does not constitute a health hazard of radiation

Hazard indices (H_{ex} and H_{in}): The decay of naturally occurring radionuclides (^{226}Ra, ^{226}Th and ^{40}K) in soil produces a radiation field that transcends the soil and air interface to produce significant human exposure. To limit the radiation exposure attributable to natural radionuclide in the sample to the permissible dose equivalent limit of 1 mSv.y^{-1}. The external hazard index and internal hazard index were estimated as:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

In order to keep the radiation hazard insignificant, the value of external hazard index 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} (Belivermis, *et al.,* 2009). The external hazard index (H_{ex}) and internal hazard index (H_{in}) was evaluated and are presented in Table 2.The results of the external hazard index (H_{ex}) of the soil samples ranged from (0.14 ± 0.01) to (0.50 ± 0.02) with a mean value of (0.33 ± 0.02), at the goldmine and (0.39 ± 0.01) to (0.68 ± 0.03), with the mean value of (0.50 ± 0.03) at the processing zone, respectively. The results of the internal hazard index (H_{in}) of the soil and mill tailing samples ranged from (0.19±0.01) to (0.71±0.02) with a mean value of (0.45±0.02), at the goldmine and (0.50±0.01) to (0.87±0.04), with the mean value of (0.62±0.04) at the processing zone, respectively. The obtained results of the (H_{ex}) and (H_{in}) shows that the soil samples are below the limit of unity, as such the radiation dose is within the permissible limit of 1 mSv/y as recommended by (UNSCEAR, 2000).

Gamma Index (I): The gamma index is used to estimate the gamma radiation hazard associated with natural radionuclides in specific investigated samples. The representative level of gamma index (I) was estimated using the following equation (Taskin, *et al.,* 2009):

$$I = 0.0033A_{Ra} + 0.005A_{Th} + 0.0005A_k \quad (6)$$

According to (Taskin, *et al.,* 2009), for radiological effects to be considered negligible, the values of each of I must be less than 1.The gamma index (I) was evaluated and presented in Table 2. The gamma index (I) of the soil samples ranged from (0.19 ± 0.01) to

(0.67 ± 0.04) with mean value of (0.43 ± 0.02) at the goldmine and ranged from (0.53 ± 0.01) to (0.90 ± 0.04) with a mean value of (0.53 ± 0.02) at the processing zone, respectively.

Table 2. The mean radiation hazard parameters obtained from all the soil samples with that of control and worldwide

Sample	D(nGy/h)	AEDE(mSv/y)	Ra_{eq}(Bq/kg)	H_{ex}	H_{in}	I
Goldmine	53.94±2.84	0.07±0.003	122.57±5.71	0.33±0.02	0.45±0.02	0.43±0.02
Processing zone	84.39±5.21	0.10±0.006	185.29±11.73	0.50±0.01	0.62±0.04	0.67±0.04
Control site	27.60±1.11	0.03±0.001	60.76±2.45	0.16±0.01	0.22±0.01	0.22±0.01
Worldwide mean	**57**	**0.07**	**< 370**	**≤ 1**	**≤ 1**	**≤ 1**

Conclusion: The present study presented the results on activity concentration for ^{226}Ra, ^{232}Th and ^{40}K and the radiological hazard parameters of the soil samples collected from the study area. It can be concluded that the radium equivalent activity, external and internal hazard indices, the absorbed dose rate and annual effective dose for all the soil and mill tailing samples that have been analyzed are within the range of acceptable limits set by regulatory agencies, as such the radiological health risks to the people living in the studied areas is not significant

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