

Radiological Impact Assessment of Soil and Rock Samples in Underground Goldmine

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ABSTRACT

Soil and rock samples were collected monthly over a period of one year in 10 different levels in an underground goldmine to determine the radiological impact on workers. The samples were analysed using a high pure germanium detector. The average permissible value for the radionuclides were below the limits apart from Th-232. The lowest concentration of the natural radionuclides in soil and rock samples was observed in level 100YOD and the highest concentration was observed in level 880. The hazard indices, exposure rate, excess lung cancer risk and annual effective dose from the rock and soil samples possess no radiological hazard if used for construction of buildings but the radium equivalent activity for level 880 were above the permissible limit and the mean excess life cancer risk (ELCR) for the naturally occurring radioactive materials in the soil and rock samples were above the average world value.

Keywords: Radon Concentration, NORM, Underground goldmine, High Pure Germanium Detector.

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Introduction

The environment around us always contains small amounts of Natural Occurring Radioactive Materials (NORMs), which have existed since the formation of the earth. Their availability in the environment is generally at levels that are not potentially harmful to human health. A major concern comes when the levels are elevated as a result of human practices like mining or natural hazards like earth quakes (Ahmed & El-Arabi, 2005).

Accumulations of materials containing radionuclides of the uranium and thorium decay chain can build up in underground and surface operations. The radionuclides of these decay chains include a wide variety of gamma emitters and emission energies. Strong gamma emitters include ²²⁶Ra and its short-lived decay products (uranium decay chain) and ²⁰⁸Tl (thorium decay chain).

Gamma exposures are of particular importance where ores are mined or where there is a significant enhancement of radionuclide activity in the surface plants (e.g. scales, sediments and wastes). For unprocessed material, U and Th decay chains can be assumed to be in secular equilibrium. In these cases, only the parent radionuclides (ie; U and Th) require to be analyzed, but for processed materials there is a need to understand what radionuclides are present and their concentrations. The most important NORMs in radiation protection are radionuclides from the Uranium-238 (²³⁸U) and Thorium-232 (²³²Th) decay series. Potassium-40 (⁴⁰K), a non-series radionuclide, also contributes significantly to human exposure in the environment (Ziajahromi, Khanizadeh, & Nejadkoorki, 2015). Therefore, the knowledge of ²³⁸U, ²³²Th and ⁴⁰K activity concentrations is important in the evaluation of absorbed doses that can lead to the estimation of their radiological hazard to the population.

Materials and Method

Soil and Rock Samples

Forty (40) Sampling points within the 10 levels were selected using the non-probability quota sampling technique soil and rocks samples. The choice of sampling was considered since the underground mine is sub divided into sections and sampling points were chosen based on areas that human activities take place most within the sections.

The soil and rock samples were taken quarterly from the sampling points making a total of 40 soil and rock samples in a quarter and 160 soil and rock samples for the year monitored. A total number of Forty (40) composite soil samples were made from the 160 samples collected from the 10 levels in a year for analysis

Sample Preparation

The samples collected were used as representative samples of the soil and rock at the 10 levels of the underground mine and were prepared and processed in the following steps for the reproducible results.

Soil and Rock

Drying

Soil and rock Samples were spread on clean stainless-steel trays and air dried for 24hrs. Initial sieving of soil was made and the pebbles, grasses and any residual roots, leaves and branches of plants removed. After the initial sieving the samples were dried in an electric oven at a temperature of 110°C for 10 hours until the moisture of the soil could not further be removed.

Grinding

If the chemical and radiometric analysis is to be performed, it is essential that all surfaces coming into contact with the soil be stainless steel, plastic or wooden, preferably of a material, which cannot contaminate the sample. The dried samples were further crushed into fine powder at the Food and Environmental Laboratory of the Radiation Protection Institute, Ghana Atomic Energy Commission, until they passed through a sieve of mesh size 20µm so that clay and material particles may be homogenized into fine powder size of reference material (Ahmed, 2005; Tufail, Ahmad, Almakky, Zafar, & Khan, 1992). (The IAEA reference materials IAEA-RGU-1(U-ore), IAEA-RGTh-1 (Th-ore) and IAEA-RGK-1 (K-ore) with densities (1.33 ± 0.03) similar to the soil samples to be measured were prepared into same cylindrical containers) to be used for Quality control and validation of analytical technique. The proper homogenization of the soil sample is essential for gamma spectroscopy system. The homogenised samples are transferred to Marinelli Beakers, which were completely sealed and stored for 1 month to allow the short-lived daughters of ^{238}U and ^{232}Th decay series to attain equilibrium with their long-lived parent radionuclides (ASTM, 1983; 1986). The sealed samples were weighed and each counted using a high purity germanium detector.

Direct gamma spectrometry analysis without pre-treatment (non-destructive) was used for the measurement of gamma rays for the soil samples using a High Purity Germanium detector (HPGE). The gamma spectrometry system consists of HPGE detector coupled to a computer based multi-channel analyser (MCA). The relative efficiency of the detector is 40 % with energy resolution of 2.0 keV at gamma ray energy of 1332 keV of ^{60}Co . The identification of individual radionuclides was performed using their characteristic gamma-ray energies and the quantitative analysis of radionuclides was performed using the Genie 2000 gamma acquisition and analysis software. The detector is housed in a 100 mm passive shielding of lead lined with copper, cadmium and plexiglass (3mm each) to reduce the background radiation. The detector is cooled in liquid nitrogen at a temperature of $-196\text{ }^{\circ}\text{C}$ (77 k). In order to determine the background distribution in the environment around the detector (quality control), ten (10) empty Marinelli beakers were thoroughly cleaned using nitric acid and counted for 36000 s in the same geometry as the samples. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. The background spectra were also used to determine the minimum detectable activities of radionuclides at each photo peak for the detector.

The activity concentrations of ^{226}Ra were determined using the γ -ray emissions and the respective γ -yield of ^{214}Pb at 351.9 keV (35.8%) and ^{214}Bi at 609.3 keV (44.8%). The ^{228}Ra activity concentrations were determined through the gamma emissions of ^{228}Ac at 911 keV (26.6%), and the ^{228}Th activity concentrations were determined through the gamma emissions of ^{212}Pb at 238.6 keV (43.3%) and ^{208}Tl at 583 keV (30.1%) and 2614.7 keV (35.3%) taking into consideration a branching ratio of 33.7% from ^{212}Bi towards ^{208}Tl . The ^{40}K activity concentration was determined directly from its emission line at 1460.8 keV (10.7%) while the ^{137}Cs and ^{210}Pb activity concentrations were determined directly from the gamma emission lines at 661.67 keV (85.1%) and 46.5 keV (4.3%) respectively. Finally, the ^{238}U activity concentrations were determined through the gamma-ray emission of its daughter ^{234}Th (4.8%). All the energies and intensities of the different radiations mentioned were taken from a well-known library.

Calibration of gamma spectrometry system

Prior to the measurements, the detector and measuring assembly were calibrated for energy and efficiency to enable both qualitative and quantitative analysis of the samples to be performed. A multi – element standard (supplied by the Eckert and Ziegler-calibration source number AJ-9177) containing radionuclides with known energies (^{241}Am (59.54 keV), ^{109}Cd (88.03 keV), ^{57}Co (122.06 keV), ^{139}Ce (165.86 keV), ^{203}Hg (279.20 keV), ^{113}Sn (391.69 keV), ^{85}Sr (514.01 keV), ^{137}Cs (661.66 keV), ^{60}Co (1173.2 keV and 1332.5 keV) and ^{88}Y (898.04 keV and 1836.1 keV) and activities in a 1L Marinelli beaker was used.

Energy Calibration

The energy calibration was performed by matching the principal gamma ray peaks observed in the spectrum of the standard to the channel numbers. The formulae relating the energy and the channel number was expressed as equation 1

$$E = A_0 + A_1 \cdot CN \quad (1)$$

Where E is the energy, CN is the channel number for a given radionuclide, and A₀ and A₁ are calibration constants for a given geometry.

Efficiency Calibration

The efficiency calibration was performed by acquiring a spectrum of the standard until the count rate of total absorption could be calculated with a statistical uncertainty of <1% at a confidence level of 95%.

The net count rate was determined at the photo peaks for all the energies to be used for the calculation of the efficiency. The efficiency was then related to the count rate and the activity of the standard by

$$\eta = \frac{N_T - N_B}{P_E \cdot A_{STD} \cdot T_{STD}} \quad (2)$$

Where P_E is the gamma ray emission probability for the energy E, η(E) is the efficiency of the detector, N_T is the total count under a photopeak in a peak range, N_B is the background counts, A_{STD} is the activity of the calibration standard for a given radionuclide in Bq at the time of measurement and T_{STD} is the counting time (Gazineu & Hazin, 2008).

The efficiency is related to the energy by the expression (Adjirackor et al., 2014)

$$\ln \eta(E) = B_0 + B_1 \ln E + B_2 (\ln E)^2 \quad (3)$$

Where, B₀, B₁, B₂ are calibration constants for a given geometry and the other symbols have the usual meaning given earlier in the passage.

From the efficiency calibration curve the following expression was obtained:

$$y = -0.0808x^4 + 2.0208x^3 - 18.856x^2 + 77.082x - 121.08 \quad \text{for } E_\gamma > 100 \text{ keV} \quad (4)$$

Where y is lnη(E) and x is lnE_γ

Calculation of activity concentration

The specific activities of samples were calculated from the expression

$$A_{SP} = \frac{N_{sam} \cdot e^{(\lambda T_d)}}{P_E \cdot \eta(E) \cdot T_c \cdot M_{sam}} \quad (5)$$

Where M_{sam} is the mass of sample (Kg), N_{sam} is the net counts for the sample in the peak range, P_E is the gamma emission probability, T_c is the counting time, η(E) is the photopeak efficiency and T_d is the delay time between sampling and counting. The factor exp(λT_d) is the correction for decay between sampling and counting. The world accepted criteria of for ²²⁶Ra, ²³²Th and ⁴⁰K are 35, 35 and 370 Bq kg⁻¹ respectively.

Minimum Detectable Activity

The minimum detectable activity (MDA) of the γ-ray measurements were calculated according to the formula:

$$MDA = \frac{\sigma \sqrt{B}}{\epsilon PTW} \quad (\text{Bqkg}^{-1}) \quad (6)$$

Where σ is the statistical coverage factor equal to 1.645 confidence level 95%, B is the background counts for the region of interest of a certain radionuclide, T is the counting time in seconds, P is the gamma yield for any element, W is the weight of the empty Marinelli beaker and ε is the efficiency of the detector. The minimum detectable activity (MDA) derived from background measurements was approximately 0.12 Bq kg⁻¹ for ²²⁶Ra, 0.11 Bq kg⁻¹ for ²³²Th and 0.15 Bq kg⁻¹ for ⁴⁰K. Concentration values below these detection limits have been taken in this work to be below the minimum detection limit (MDL).

Radiological Impact assessment of Soil and Rock Samples

Estimation of hazard indices

Radium equivalent activity (Raeq)

The exposure due to γ-radiation is usually defined in terms of radium equivalent activity Raeq is given by Eq. (7) (Murugesan, Mullainathan, Ramasamy, & Meenakshisundaram, 2011)

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (7)$$

The above equation is based on the assumption that 370 Bq kg⁻¹ of ²²⁶Ra, 259 Bq kg⁻¹ of ²³²Th, and 4810 Bq kg⁻¹ of ⁴⁰K produce the same gamma-ray dose rate. The radium equivalent is related to both the external γ-dose and the internal γ-dose from radon and its progeny. The permissible maximum value of the radium equivalent activity is 370 Bq kg⁻¹, which corresponds to an effective dose of 1mSv yr⁻¹ for to the inhabitants of dwellings (Esirole, Ibeanu, Garba, & Onoja, 2019).

External hazard index (H_{ex})

Some authors proposed a model for a room in the house where the inhabitants live with infinitely thick walls without windows and doors and calculated Hex using the following relation (Model I) (Esiolo et al., 2019):

$$H_{EX} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (8)$$

Other authors modified such model to a room with windows and doors and calculated Hex using the following equation (Model II) (Baykara, Karatepe, & Dođru, 2011):

$$H_{EX} = \frac{A_{Ra}}{740} + \frac{A_{Th}}{518} + \frac{A_K}{9620} \leq 1 \quad (9)$$

where the three factors of Eq. (9) are decreased to half their values in Eq. (8). In model II the presence of doors and windows will cause some kind of ventilation in the room which will decrease the exposure of inhabitants to radiation and decrease all kinds of doses (Mattsson, Johansson, & Liniecki, 2008). The value of this index must be less than unity in order to keep the radiation hazard insignificant. The prime objective of this index is to limit the radiation dose to the accepted dose limit of 1 mSv yr⁻¹ (Hewamanna, Sumithrarachchi, Mahawatte, Nanayakkara, & Ratnayake, 2001).

Internal hazard index (H_{in})

Inhalation of alpha particles emitted from the short-lived radionuclides (radon 222Rn, the daughter product of 226Ra) and thoron (220Rn, the daughter product of 224Ra) is also hazardous to the respiratory organs. This hazard can be controlled by the internal hazard index (H_{in}), (Righi & Bruzzi, 2006) which is given by the following Eq:

$$H_{IN} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (10)$$

For the safe use of a certain building material in the construction of dwellings, the index (H_{in}) should be less than unity.

External (γ -radioactivity) level index I_γ

This index is also known as the representative level index and was calculated from the following relation (El-Gamal, Nasr, & El-Taher, 2007; NEA-OECD, 1979):

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \leq 1 \quad (11)$$

The National Environment Agency (NEA) and the Organisation for Economic Co-operation and Development (OECD) group of experts suggested some criteria for a definition of different levels to be (representative, first enhanced, second enhanced) (NEA-OECD, 1979).

$I_\gamma = 1$ as an upper limit

$I_\gamma \leq 1$ corresponds to 0.3mSvy⁻¹

$I_\gamma \leq 3$ corresponds to 1mSvy⁻¹

Concerning different building materials, the ranges I_γ are

- ❖ Materials used in bulk amounts like bricks $I_\gamma \leq 0.5$ to $I_\gamma \leq 1$
- ❖ Superficial and other materials $I_\gamma \leq 0.2$ to $I_\gamma \leq 6$

Internal (α -radioactivity) level index I_α

The excess alpha radiation due to radon inhalation originating from building materials is estimated using the relation below (El Galy, El Mezayn, Said, El Mowafy, & Mohamed, 2008):

$$I_\alpha = \frac{A_{Ra}}{200} \leq 1 \quad (12)$$

I_α should be lower than the maximum permissible value of $I_\alpha = 1$ which corresponds to 200 Bq kg⁻¹. For alpha radiation and taking into consideration that a building material with Ra concentration lower than 200 Bq kg⁻¹ could not cause indoor radon concentration higher than 200 Bq m⁻³.

Exposure rate and annual effective dose equivalent at 1m from a radioactive source

Exposure rate (ER)

The exposure rate was calculated using the following relation (Akhtar, Tufail, Ashraf, & Iqbal, 2005):

$$ER(\mu R h^{-1}) = 1.90A_{Ra} + 2.82A_{Th} + 0.0179A_K \quad (13)$$

Dose rate (DR) and its relation with (ER)

The dose rate was calculated using the following two relations (O'Brien & Sanna, 1976)

$$DR \text{ (mSv yr}^{-1}\text{)} = 0.0833ER \text{ (}\mu\text{Svh}^{-1}\text{)} \quad (14)$$

Estimation of Dose Parameters

Estimation of Annual Dose Rate

Air absorbed gamma dose rate (D_{air})

The absorbed gamma dose rate in air 1 m above the ground surface for the uniform distribution of radionuclides (226Ra, 232Th, and 40K) was computed on the basis of provided guidelines (Baykara et al., 2011; Hewamanna et al., 2001). The conversion factors used to compute absorbed gamma dose rate in air (D_{air}) per unit activity concentration in (1 Bq kg⁻¹) are 0.462 for 226Ra, 0.621 for 232Th, and 0.0417 for 40K. Such dose parameter was calculated applying the following relation (Kurnaz et al., 2007):

$$D_{air} \text{ (nGyh}^{-1}\text{)} = 0.462A_{Ra} + 0.621A_{Th} + 0.0417K \quad (15)$$

The population-weighted values give an absorbed dose rate in air outdoor from terrestrial gamma radiation a value of 57 nGy h⁻¹.

Annual effective dose equivalent (AEDE)

To estimate the AEDE the conversion factor (0.7 Sv Gy⁻¹) from absorbed dose rate in air in nGy h⁻¹ to effective dose rate in mSv yr⁻¹ is used with outdoor occupancy factor of 0.2 and indoor occupancy factor of 0.8. The AEDE was calculated using the following formulae (Esirole et al., 2019):

$$AEDE \text{ (Indoor)} \text{ (mSvy}^{-1}\text{)} = D_{air} \text{ (nGyh}^{-1}\text{)} \times 8766h \times 0.8 \times 0.7SvGy^{-1} \times 10^{-6} \quad (16)$$

$$AEDE \text{ (outdoor)} \text{ (mSvy}^{-1}\text{)} = D_{air} \text{ (nGyh}^{-1}\text{)} \times 8766h \times 0.2 \times 0.7SvGy^{-1} \times 10^{-6} \quad (17)$$

These indices measure the risk of stochastic and deterministic effects in the irradiated individuals. The recommended value of the annual effective dose equivalent is 0.48 mSv yr⁻¹ and the criterion of the total annual effective dose equivalent (indoors + outdoors) should be less than 1 mSv yr⁻¹ (Esirole et al., 2019).

The annual gonadal dose equivalent (AGDE)

The gonads, the active bone marrow and the bone surface cells are considered to be the organs of importance. An increase in AGED has been known to affect the bone marrow, causing destruction of the red blood cells that are then replaced by white blood cells. The annual gonadal dose equivalent (AGDE) due to the specific activities of 226Ra, 232Th and 40K was calculated using the following relation (Kurnaz et al., 2007).

$$AGDE \text{ (}\mu\text{Svhy}^{-1}\text{)} = 3.09A_{Ra} + 4.18A_{Th} + 0.0314K \quad (18)$$

The world averages of AGDE of a house containing activity concentrations of 226Ra, 232Th and 40K are 35, 35 and 370 mSv yr⁻¹, respectively. The standard UNSCEAR value for AGED is 300 mSv yr⁻¹.

Excess lifetime cancer risk (ELCR)

This gives the probability of developing cancer over a lifetime at a given exposure level. The ELCR has been calculated using the following equation (Kurnaz et al., 2007):

$$ELCR \text{ (mSvy}^{-1}\text{)} = AEDE \times DL \times RF \quad (19)$$

where DL is the duration of life (70 years average) and RF is the risk factor (Sv) i.e. fatal cancer risk per Sievert. For stochastic effects, the ICRP 106 used a value of RF = 0.05 for the public.

Results and Discussion

Activity Concentration of Radionuclides in Rock and Soil Samples

The distribution of activity concentration of radionuclides in all the 10 levels in the underground goldmine are shown in table 1. Activities concentrations ranges from a minimum of 18.93±1.68 Bqm⁻³ to a maximum of 45.13±3.91 Bqm⁻³ for Ra-226 with a mean concentration of 32.19±6.98 Bqm⁻³. The lowest activity concentration was found at level 1000 YOD and the highest activity concentration was found at level 880. The mean activity concentration of Ra-226 is above the world average value of 35 Bq kg⁻¹ from the United Nations Scientific Committee on Atomic radiation as well as other measurements recorded in some mining companies in Ghana as shown in table 2.

Th-232 recorded a minimum of 17.6±2.32 Bq/kg to a maximum of 51.9±3.85 Bq/kg with a mean concentration of 39.08±10.66 Bq/kg. The lowest concentrations were found at level 1000YOD and the highest levels were found at level 880. The mean activity concentration of Th-232 is below the world average value of 40 Bq/kg from the United Nations Scientific Committee of Atomic radiation as well as other measurement recorded in some mining companies in Ghana apart from measurements recorded in Adamus mine as shown in table 2.

Table 1: Activity Concentration of Radionuclides at Various Levels

| Levels | Ra-266 (Bq/kg) | Th-232 (Bq/kg) | K-40 (Bq/kg) |
|-------------|-------------------|-------------------|-----------------|
| 800 | 34.13±1.58 | 43.83±2.27 | 267.43±10.28 |
| 810 | 30.5±1.48 | 44.2±2.47 | 233.40±18.97 |
| 840 | 33.55±1.83 | 46.35±0.91 | 256.43±16.18 |
| 880 | 45.13±3.91 | 51.9±3.85 | 364.00±14.41 |
| 790 | 32.55±1.42 | 42.78±1.32 | 252.93±10.50 |
| 820 | 37.03±1.37 | 43.5±4.61 | 310.55±7.70 |
| 960 | 25.3±1.00 | 23.15±2.78 | 159.95±20.10 |
| 1000YOD | 18.93±1.68 | 17.6±2.32 | 156.35±36.81 |
| 920 | 27.98±1.13 | 34.33±1.26 | 221.35±9.34 |
| 1000SKY DEC | 35.4±1.99 | 44.15±2.92 | 239.20±8.47 |
| Average | 32.19±6.98 | 39.08±10.66 | 247.18±61.65 |

K-40 ranges from a minimum of 156.35±36.81 Bq/kg to a maximum of 364.00±14.41 Bq/kg with a mean concentration of 247.18±61.65 Bq/kg. The lowest concentrations were found at level 1000 YOD and the highest levels were found at level 880. The mean activity concentration of K-40 is below the world average value of 400 Bq /kg from the United Nations Scientific Committee of Atomic radiation and measurements recoded in Obuasi and Adamus mine as shown in table 1. Rock and soil samples from level 880 must undergo further radiological test for it to be considered as a NORM free material due to the significance increase in activity concentration of radionuclides. However, the higher activity concentrations of Ra-266, Th-232 and K-40 in level 880 as compared to the other levels could be due to the presence of a rich gold ore since rich ores have high uranium content.

Table 2: Comparison of Activity Concentration Ra-226, Th-232 and K-40 in Soil and Rock Samples with Worldwide Average and other Published Work

| Agency/ Country | Location | Ra-226 | Th-232 | K-40 | Reference |
|---|-------------------------|--------|--------|--------|----------------------|
| United Nations Scientific Committee on Atomic Radiation (UNSCEAR) | Worldwide average | 35 | 40 | 400 | UNSCEAR, 2000 |
| Ghana | Obuasi Underground mine | 29.3 | 26.8 | 770.0 | Darko et al., 2005 |
| Ghana | Newmont Akyem | 11.28 | 12.23 | 113.78 | Wiseman et al., 2000 |
| Ghana | Tarkwa Mine | 13.61 | 24.22 | 162.18 | Faanu et al., 2008 |
| Ghana | Chirano Mines | 9.79 | 9.18 | 237.40 | Faanu et al., 2008 |
| Ghana | Bibiani Adamus Mine | 8.03 | 43.98 | 395.05 | Awudu et al., 2016 |
| This Study | Nzema SUG Mine | 32.19 | 39.08 | 247.18 | PhD Thesis |
| | Newmont Ahafo | | | | |

Table 2 shows the descriptive statistics of activity concentration of Ra-226, Th-232 and K-40 distributions in the soil and rock samples.

A positively skewed data distribution of Ra-226 and K-40 indicates that low values of data for the radionuclides were recorded as compared to high values and a negatively skewed data distribution of Th-232 indicates that high values of data for the radionuclide were recorded as compared to low values. In addition, a platykurtic data distribution of Th-232 indicates that different Th-232 concentrations were recorded, or the concentrations are not comparably the same and a leptokurtic data distribution of Ra-266 and K-40 indicates that the concentrations of Ra-226 recorded in the underground mine are similar and that of the K-40 were also similar.

However, the data for all the radionuclides is normally distributed since the p-value in the Kolmogorov-Smirnov test as shown in table 2 is greater than the 5% level of significance ($P > 0.05$) thus; the representative values give a true reflection of the population.

Table 2: Descriptive Statistics of Activity Concentration Distribution in Soil and Rock Samples

| Statistics | Ra-226 | Th-232 | K-40 |
|------------------------------|--------|--------|-------|
| Skewness | 0.001 | -0.959 | 0.054 |
| Kurtosis | 0.661 | -0.055 | 0.242 |
| Shapiro Wilk Test (P values) | 0.405 | 0.485 | 0.461 |

Table 3 shows the external hazard indices for all the levels in the underground mine. The external hazard indices for both rooms in the house where the inhabitants live with infinitely thick walls without windows and doors (H_{EX1}) and a room with windows and doors (H_{EX2}) shows an insignificant radiological effect to inhabitants who may use the soil and rock from the underground mine.

Table 3: External Hazard Indices of Rock and Soil Samples (H_{EX})

| Levels | H_{EX1} | H_{EX2} |
|--------------|-----------|-----------|
| 800 | 0.32 | 0.16 |
| 810 | 0.30 | 0.15 |
| 840 | 0.32 | 0.16 |
| 880 | 0.40 | 0.20 |
| 790 | 0.31 | 0.15 |
| 820 | 0.33 | 0.17 |
| 960 | 0.19 | 0.10 |
| 1000 YOD | 0.15 | 0.08 |
| 920 | 0.25 | 0.13 |
| 1000 SKY DEC | 0.32 | 0.16 |
| Average | 0.29 | 0.14 |

Table 3 shows the internal hazard indices for all the 10 levels in the underground mine. The internal hazard indices (H_{IN}) are the cause of harmful effects to the lungs due to the internal contact of α -particles of a higher ionization power to the sensitive tissues of the lungs and other parts of the respiratory system. Values of internal hazard indices are less than unity, rock and soil samples from the underground mine that may be used for construction of dwellings may not pose any radiological hazard.

Table 4: Internal Hazard Indices of Rock and Soil Samples (H_{IN})

| Levels | H_{IN} | $I\alpha$ |
|--------------|----------|-----------|
| 800 | 0.41 | 0.17 |
| 810 | 0.38 | 0.15 |
| 840 | 0.41 | 0.17 |
| 880 | 0.52 | 0.23 |
| 790 | 0.39 | 0.16 |
| 820 | 0.43 | 0.19 |
| 960 | 0.26 | 0.13 |
| 1000 YOD | 0.20 | 0.09 |
| 920 | 0.33 | 0.14 |
| 1000 SKY DEC | 0.41 | 0.18 |
| Average | 0.38 | 0.16 |

The Internal level index ($I\alpha$) is used to access the excess alpha radiation due to radon inhalation originating from building materials (El Galy et al., 2008): Since all the values found at the 10 different levels in the underground goldmine are lower than the maximum permissible value ($I\alpha < 1$) as shown in table 4. The soil and rock samples which may be used for building material will have a radon concentration lower than 200 Bq kg^{-1} and when used for construction of buildings may not pose any radiological health to inhabitants. The internal and external hazard indices are set to limit the radiation dose to the acceptable dose limit to 1 mSv yr^{-1} for the members of the public (Hewamanna, Sumitharachchi, Mahawatte, Nanayakkara, & Ratnayake, 2001).

The radium equivalent activity (R_{aeq}) is related to exposure of both the external γ -dose and the internal γ -dose from radon and its progeny. The obtained results in table 5 shows that the radium equivalent activity (R_{aeq}) of all soil and rock samples from the underground goldmine have values less than the permissible maximum value of 370 Bq/kg (UNSCEAR, 2000) apart from level 880 (399.62 Bq/kg) which was slightly higher. The permissible maximum value of the radium equivalent activity corresponds to an effective dose of 1 mSv yr^{-1} for to the inhabitants of dwellings (Esiolo et al., 2019). Hence soil and rock samples from the underground mine are radiologically safe for construction of buildings.

Table 5: Radium Equivalent Activity and External Level in Rock and Soil Samples

| Levels | Raeq (Bq/kg) | $I\gamma$ |
|--------------|-----------------|-----------|
| 800 | 302.71 | 0.42 |
| 810 | 273.42 | 0.40 |
| 840 | 297.28 | 0.43 |
| 880 | 399.622 | 0.53 |
| 790 | 288.4705 | 0.41 |
| 820 | 338.35 | 0.44 |
| 960 | 181.57 | 0.25 |
| 1000 YOD | 164.48 | 0.20 |
| 920 | 247.50 | 0.34 |
| 1000 SKY DEC | 282.72 | 0.42 |
| Average | 277.61 | 0.38 |

The Nuclear Energy Agency (NEA) and Organisation for Economic Co-operation and Development (OECD) group of experts suggested some limits for external level index ($I\gamma$) for different building materials for the members of the public to be radiologically safe. The limit for materials used in bulk amounts like bricks should be less than 1 ($I\gamma < 1$) and that of superficial and other materials like tiles should be less than 6 ($I\gamma < 6$). The values $I\gamma$ as shown in table 5 shows that soil and rock samples from the underground mine are radiologically safe and may be used for bricks and superficial materials such tiles for construction of buildings.

Table 6: Annual Dose Parameters

| Levels | ER mR h ⁻¹ | DR mSv yr ⁻¹ | Dair nGy h ⁻¹ | AEDEind mSv yr ⁻¹ | AEDEout mSv yr ⁻¹ | AGDE μSv yr ⁻¹ | ELCR % |
|--------------|--------------------------|----------------------------|-----------------------------|---------------------------------|---------------------------------|------------------------------|-----------|
| 800 | 63.54 | 5.29 | 54.13 | 0.27 | 0.07 | 297.03 | 0.93 |
| 810 | 70.87 | 5.90 | 51.27 | 0.25 | 0.06 | 286.33 | 0.88 |
| 840 | 71.55 | 5.96 | 54.98 | 0.27 | 0.07 | 305.46 | 0.94 |
| 880 | 67.14 | 5.59 | 68.26 | 0.34 | 0.08 | 367.81 | 1.17 |
| 790 | 63.31 | 5.27 | 52.14 | 0.26 | 0.06 | 287.32 | 0.90 |
| 820 | 57.88 | 4.82 | 57.07 | 0.28 | 0.07 | 305.99 | 0.98 |
| 960 | 20.08 | 1.67 | 32.73 | 0.16 | 0.04 | 179.97 | 0.56 |
| 1000 YOD | 16.47 | 1.37 | 26.19 | 0.13 | 0.03 | 136.96 | 0.45 |
| 920 | 47.61 | 3.97 | 43.47 | 0.21 | 0.05 | 236.87 | 0.75 |
| 1000 SKY DEC | 61.52 | 5.13 | 53.75 | 0.26 | 0.07 | 301.44 | 0.92 |
| Average | 53.99 | 4.50 | 49.40 | 0.24 | 0.06 | 270.52 | 0.85 |

Concerning exposure rate (ER), in table 6, it was observed that all the soil and rock samples have values less than the criterion of 600 mR h⁻¹ of exposure rate to naturally occurring radioactive materials. The dose rate (DR) due to naturally occurring radioactive materials from the soil and rock samples are lower than the maximum permissible limits (50 mSv yr⁻¹) (Darwish, Abul-Nasr, & El-Khayatt, 2015).

The absorbed gamma dose rate in air 1m above the ground surface (Dair), in soil and rock samples are lower than the permissible maximum limit apart from level 880 (68.26 nGy h⁻¹) and level 820 (57.09 nGy h⁻¹), which recorded a value higher than the population-weighted value of 57 nGy h⁻¹ (Baykara et al., 2011; Hewamanna et al., 2001). However, it may not be advisable to use soil and rock samples from these levels for construction of buildings.

The annual effective dose equivalent (AEDE) measures the risk of stochastic and deterministic effects in the irradiated individuals. The recommended value of the annual effective dose equivalent is 0.48 mSv yr⁻¹ and the criterion of the total annual effective dose equivalent (indoors + outdoors) should be less than 1 mSv yr⁻¹ (Esirole et al., 2019). The values for both indoor and outdoor annual effective dose equivalent from soil and rock samples were less than the recommended values and the sum of the indoor and outdoor annual effective dose equivalent is less than 1 mSv yr⁻¹. Thus, there may not be any significant radiation effect on individuals who will be exposed to soil and rock samples from the underground mine.

The gonads, the active bone marrow and the bone surface cells are considered to be the organs of importance. An increase in AGDE has been known to affect the bone marrow, causing destruction of the red blood cells that are then replaced by white blood cells. The annual gonadal dose equivalent (AGDE) from soil and rock samples ranged from a minimum of 179.97 μSv yr⁻¹ in level 960 to a maximum of 367.81 μSv yr⁻¹ in level 880 with a mean of 270.52 μSv yr⁻¹ which was less than the standard UNSCEAR value for AGED of 300 mSv yr⁻¹. This shows a low probability of radiation effect on the bone marrow of individuals exposed to the rock and soil samples from the underground mine apart from rocks from level 880.

The excess lifetime cancer risk (ELCR) gives the probability of developing cancer over a lifetime at a given exposure level. The ELCR ranges from a minimum of 0.45 at level 1000YOD to a maximum of 1.17 at level 880

with a mean of 0.85 are higher than the world's average value of 0.29×10^{-3} (Uosif et al., 2014). Thus, the chance of developing using soil and rock samples from the underground mine is higher as compared to the world average.

Conclusion

Industries like mining and oil & gas companies do not allow their waste to be used commercially due to chemicals, radiological or other types of contamination but some of the rock sample that are segregated from the rich ore are used internally. Most of the mining sectors are mostly found in remote areas and when these mines are abandoned residents from nearby villages could use some of these soil and rock samples for buildings, road construction etc. The analysis of these soil and rock samples from the underground goldmine is to assess their radiological impact in case they are being used for construction of buildings. The primary aim of radiation protection is to protect humans and the environment from the harmful effect of ionizing radiation.

The mean activity concentration of Ra-226 and K-40 were below the permissible limit of 35 Bq kg⁻¹ and 370 Bq kg⁻¹ respectively but Th-232 was above the limit of 35 Bq kg⁻¹. The dose rate due to NORMS in rock and soil samples, absorbed dose rate, annual effective dose, annual gonadal dose equivalent from the soil and rock samples from the underground goldmine possess no radiological hazard and the materials may be used for building materials. The radium equivalent dose and excess life cancer risk from soil and rock samples in level 880 was above the permissible limit and this may be attributed to a rich gold ore at that level since rich ores have higher uranium content. The values of the external level index ($I\gamma$) is also known as the representative level index from the rock and soil samples suggest that they can be used for bulk amounts like bricks since the values obtained were between $I\gamma \leq 0.5$ to $I\gamma \leq 1$.

The values of the internal (α -radioactivity) level index $I\alpha$ also suggest that when the soil and rock samples are used for construction of buildings there will be no risk of excess alpha radiation due to radon inhalation originating from building materials. The exposure rate, dose rate, dose rate in air and the indoor and outdoor annual effective dose were all below the permissible limit apart from the excess lifetime cancer risk (ELCR) which is higher than the world's average value of 0.29×10^{-3} (0.00029).

Recommendation

The levels of Radon concentration in the soil and rock samples appears to be low but may present a potential long-term health risk to occupationally exposed workers due stochastic effect of radiation. Nonetheless, as a new underground goldmine continues monitoring of soil and rock samples for deeper levels is recommended. This will ensure that the occupationally exposed workers at the underground goldmine work with no radiological health risk.

References

- Adjirackor, T., Darko, E., Emi-Reynolds, G., Kpeglo, D., Awudu, R., & Owusu, J. (2014). Radiological study of soil, fertilizer and foodstuffs in some selected farming communities in the greater Accra region, Ghana. *Elixir Nuclear and Radiation Physics*, 77, 29112-29118.
- Ahmed, N. K. (2005). Measurement of natural radioactivity in building materials in Qena city, Upper Egypt. *Journal of environmental radioactivity*, 83(1), 91-99.
- Ahmed, N. K., & El-Arabi, A. G. M. (2005). Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, Upper Egypt. *Journal of environmental radioactivity*, 84(1), 51-64.
- Akhtar, N., Tufail, M., Ashraf, M., & Iqbal, M. M. (2005). Measurement of environmental radioactivity for estimation of radiation exposure from saline soil of Lahore, Pakistan. *Radiation Measurements*, 39(1), 11-14.
- Baykara, O., Karatepe, Ş., & Dođru, M. (2011). Assessments of natural radioactivity and radiological hazards in construction materials used in Elazig, Turkey. *Radiation Measurements*, 46(1), 153-158.
- Darwish, D., Abul-Nasr, K., & El-Khayatt, A. (2015). The assessment of natural radioactivity and its associated radiological hazards and dose parameters in granite samples from South Sinai, Egypt. *Journal of Radiation Research and Applied Sciences*, 8(1), 17-25.
- El-Gamal, A., Nasr, S., & El-Taher, A. (2007). Study of the spatial distribution of natural radioactivity in the upper Egypt Nile River sediments. *Radiation Measurements*, 42(3), 457-465.
- El Galy, M., El Mezayn, A., Said, A., El Mowafy, A., & Mohamed, M. (2008). Distribution and environmental impacts of some radionuclides in sedimentary rocks at Wadi Naseib area, southwest Sinai, Egypt. *Journal of environmental radioactivity*, 99(7), 1075-1082.
- Esiolo, S., Ibeanu, I., Garba, N., & Onoja, M. (2019). Determination of radiological hazard indices from surface soil to individuals in Angwan Kawo Gold Mining Sites, Niger state, Nigeria. *Journal of Applied Sciences*

- and Environmental Management*, 23(8), 1541-1547.
- Gazineu, M., & Hazin, C. (2008). Radium and potassium-40 in solid wastes from the oil industry. *Applied radiation and isotopes*, 66(1), 90-94.
- Hewamanna, R., Sumithrarachchi, C., Mahawatte, P., Nanayakkara, H., & Ratnayake, H. (2001). Natural radioactivity and gamma dose from Sri Lankan clay bricks used in building construction. *Applied radiation and isotopes*, 54(2), 365-369.
- Kurnaz, A., Küçükömeroğlu, B., Keser, R., Okumusoglu, N., Korkmaz, F., Karahan, G., & Çevik, U. (2007). Determination of radioactivity levels and hazards of soil and sediment samples in Firtina Valley (Rize, Turkey). *Applied radiation and isotopes*, 65(11), 1281-1289.
- Mattsson, S., Johansson, L., & Liniecki, J. (2008). Radiation dose to patients from radiopharmaceuticals. Addendum 3 to ICRP Publication 53. ICRP Publication 106. Approved by the Commission in October 2007. *Annals of the ICRP*, 38(1-2), 1-197.
- Murugesan, S., Mullainathan, S., Ramasamy, V., & Meenakshisundaram, V. (2011). Radioactivity and radiation hazard assessment of Cauvery River, Tamilnadu, India. *Iran J Radiat Res*, 8(4), 211-222.
- NEA-OECD, O. (1979). for EC and D. Ex posure to Ra di ation from the Nat u ral Ra dio ac tiv ity in Build ing Ma teri als, Re port by a Group of Ex perts of the OECD Nuclear En ergy Agency: OECD, Paris.
- O'Brien, K., & Sanna, R. (1976). The distribution of absorbed dose-rates in humans from exposure to environmental gamma rays. *Health physics*, 30(1), 71-78.
- Righi, S., & Bruzzi, L. (2006). Natural radioactivity and radon exhalation in building materials used in Italian dwellings. *Journal of environmental radioactivity*, 88(2), 158-170.
- Tufail, M., Ahmad, N., Almakky, S., Zafar, M., & Khan, H. A. (1992). Natural radioactivity in the ceramics used in dwellings as construction material. *Science of the total environment*, 127(3), 243-253.
- Uosif, M., Issa, S. A., Ebrahim, A., Zahran, E. M., & Moussa, M. (2014). Determination of natural radioactivity in building raw materials from the quarries of Assiut cement company, Assiut, Egypt. *Int. J. New. Hor. Phys*, 1(1), 25-32.
- Ziajahromi, S., Khanizadeh, M., & Nejadkoorki, F. (2015). Using the RESRAD code to assess human exposure risk to 226Ra, 232Th, and 40K in soil. *Human and Ecological Risk Assessment: An International Journal*, 21(1), 250-264.