Assessment of potential radiological risks due to natural gamma radiations in some selected rock samples using γ-ray spectrometry

Abdu Hamoud Al-khwalny$^{1,2}$, A R Khan$^{3}$, J M Pathan$^{4}$ and Ifra Fatema$^{4}$

$^1$Department of Physics, Dr. Babasaheb Ambedkar University, Aurangabad, India-431001

$^2$Department of Physics, Faculty of Education & Languages, Amran University, Amran, Yemen

$^3$Department of Computer Science, Maulana Azad College, Dr. Babasaheb Ambedkar University, Aurangabad, Maharashtra, India-431001

$^4$Department of Physics, Maulana Azad College, Dr.Babasaheb Ambedkar University, Aurangabad, Maharashtra, India-431001.

Corresponding author’s e-mail: abdualkhwalny@gmail.com

Abstract. One of the sources of radiation risk in dwellings made of rocks is primordial radionuclides. It is imperative to assess the levels of radioactivity due to gamma rays from these materials and consequently to determine the dose rate from these materials. Fourteen samples of rock were collected from various sites of Aurangabad-India and the level of radioactivity and elemental compositions of samples have been measured utilizing γ-ray spectroscopy and EDXRF technique. The findings showed that the mean activity concentrations of the 226Ra, 232Th, and 40K were 6.883, 10.841 and 128.616 Bq/kg, lower than the global average values of 35, 30, and 400 Bq/kg. The obtained results were compared with the worldwide average as well as with similar studies. The ratios among the detected radioisotopes in rocks were computed for the spatial distribution of natural radionuclides in the studied area. The radiological risk parameters were estimated and compared with internationally recommended values. The Pearson correlation was used to determine the relationship between the radionuclide concentration and radioactive variables, as well as with the elemental compositions. EDXRF results indicate that SiO$_2$, Al$_2$O$_3$, K$_2$O, CaO, TiO$_2$, Na$_2$O, MgO, P$_2$O$_5$, MnO, and Fe$_2$O$_3$ are the major oxides present in the samples. Moreover, heavy metals such as V, Cr, Co, Ni, Cu, Zn, As, Ba, Cd, Hg, and Pb with varying concentrations were detected. The radiological assessment indicated that the rocks of the study area are radiologically safe and may be used for construction without causing any threat to human health.
1. Introduction

Human beings are exposed to ionizing radiation both inside and outside their dwellings every day from natural radionuclides in the ground, building materials, air, food, the universe and even elements in their own bodies [1, 2]. Natural radioactivity in the environment is mainly caused by primordial radionuclides such as $^{40}\text{K}$, and the radionuclides from the $^{232}\text{Th}$ and $^{238}\text{U}$ series and their decay products, which exist at trace levels in all ground formations. Primordial radionuclides are formed in stars by the cycle of nucleosynthesis and are distinguished by half-lives comparable to the age of the earth. The most commonly encountered radionuclides are $^{238}\text{U}$ and $^{232}\text{Th}$, their decay products, and $^{40}\text{K}$. Most of the countries within the world carry out extensive surveys of natural radiation and environmental radioactivity in order to collect data on potential radiological risks and take appropriate protection [3, 4]. Knowledge of the distribution of natural radioactivity and levels of radiation in the environment is important for assessing the effects of radiation exposure from both terrestrial and extraterrestrial sources. The earth's crust contains natural radionuclides of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$. When these radionuclides and their daughters undergo decays, $\gamma$-rays, $\beta$ and $\alpha$-radiation are released into the environment [5, 6]. Human beings have always been exposed to natural radiation arising from the earth as well as from outside the earth [7, 8]. Radioactivity levels and associated radiological hazards depend mainly on the geological and geographical conditions of the environment and occur at various levels in the soils of each region of the world [9]. Most of the soil, rock, and building material contain different quantities of natural radionuclides [10]. Exposure to radiation from construction materials can be classified into external and internal exposure [11]. The external exposure to the public is caused by direct $\gamma$-radiation, while internal exposure is caused by inhalation of $^{222}\text{Rn}$, $^{220}\text{Rn}$, and their short-lived decay products [12]. Radon is a noble gas, which can be easily transported by porous media such as building materials, while only a fraction of that contained in the material reaches the surface and enters the indoor air [13].

Studying the effect of radiation on the environment is significant in both physics as well as health care. This study is relevant not only for a scientific reason but also for a practical reason in terms of hazards of radiation to the living being, particularly human life. Detailed information on nuclear radiation exposure and its permissible limits will help to avoid the nuclear hazard to humans. Adequate information on natural radioactivity and dose rates in our environment is very important for risk awareness and can be used as a basis for planning management strategy to achieve better environmental quality and provide information to the general public. Many studies were being carried out to map natural radioactivity levels in rocks and gamma dose rate all over the world. Currently, there was no available data from this study area on the natural radioactivity levels in rocks and radiological risks evaluation. This study has been conducted and reported for the first time on natural radioactivity measurements in rocks of Aurangabad district. The major purpose of this study is to assess radiological hazards to humans from the radioactivity in the rock samples from the outskirts of Aurangabad, India. In this study, the collected rock samples were analyzed using $\gamma$-ray spectroscopy (HPGe) detector to determine activity concentrations of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in the samples. As a result, radiological risk parameters were computed and compared to acceptable safety limits and worldwide average values. Moreover, the elemental compositions of samples have been determined using the EDXRF technique.

2. Materials and Method

2.1. Sample collection and preparation

A total of fourteen of rock samples were collected from various sites of Aurangabad, Maharashtra-India (Figure 1) as this is a historic city with recorded history and having monuments of world heritage for tens
of centuries. Area of study located between 19.926436°–20.009619°N latitudes and 75.329620°–75.261971°E longitudes. Rock sampling was carried out during the July-September 2017 period which corresponds to the raining season in this area. Rock samples were crushed into small pieces using a crushing machine and ground to suitable powder, then samples were sifted through 200 meshes with a grinding machine which is the optimum size enriched with heavy minerals [14]. After sieving and prepared optimum powder of rock samples, the samples were dried in a muffle furnace at 120°C for each sample for 24 hours to remove moisture. The samples were packed in plastic containers dimensions of 60 mm in diameter and 100 mm in height. The samples were weighed and left for at least one month at room temperature before analysis by γ-ray spectrometry in order to ensure a secular equilibrium between 226Ra, 232Th, and their respective progeny products [15].

Figure1. Location map of the study area (Aurangabad-India)

2.2. Radioactivity determination

The activity concentrations of 226Ra, 232Th, and 40K in rock samples were determined by utilizing HPGe γ-ray spectrometry (p-type, coaxial type, model BE3825, Canberra, USA) with energy resolution (FWHM) at 1330 keV 60Co is 2.2 keV and relative efficiency is 50%. The detector was coupled with a multi-channel analyzer (MCA) for data acquisition via electronic modules. The detector was shielded from the surrounding environment in a tightly cylindrical lead shield, to avoid background radiation. Experiments were carried out at the Centre for Advanced Research in Environmental Radioactivity Measurements Laboratory (CARER), India. Energy calibration is carried out to ensure a linear relationship between energy and the number of channels corresponding to that energy, and to determine the energy of each channel in a spectrum. The calibration of the HPGe detector (energy, efficiency) was performed using reference materials from the IAEA quality assurance system: RGU-238, RGTh-232, RGK-1, and SOIL-6. These standard reference materials were taken in plastic containers of 300 ml (similar to the containers used to fill the samples for the γ-spectrometric analysis). The same materials and samples were taken in
the same size and form of containers so that the geometry remained the same. For a long time, the source of the calibration was counted to obtain clear defined photo peaks. These three standards gamma-ray sources of the same volume were used to generate efficiency versus the energy curve. Figure 2 demonstrates the variation of the efficiency of the HPGe detector versus the energy curve. The background radiation due to the naturally occurring radionuclides in the environment around the detector was measured immediately before the sample counting using an empty plastic container and counted for the same counting time used for the samples (30000 sec) to obtain the net count rate. The obtained spectrum was analyzed using a 16 K multichannel analyzer connected to the computer using GENIE-2000 software. The activity concentration of $^{226}$Ra was determined using $\gamma$-ray emission lines of $^{214}$Pb (295.22, 351.93 keV), $^{214}$Bi (609.31, 1120 keV) and $^{226}$Ra (186.1 keV). Similarly, $^{232}$Th activity concentration was measured using $\gamma$-ray emission lines of $^{228}$Ac (911.2, 209.25, 338.32 keV) and $^{208}$Tl (2614, 583.19, 860.56 keV). The $^{40}$K activity was directly determined using the following equation [16]:

$$A \text{ (Bq/kg)} = \frac{N}{T_c \times I_{\gamma}(E_\gamma) \times \varepsilon(E_\gamma) \times M}$$

(1)

Where, $N$ is the number of counts in a given peak area corrected for background peaks at energy $E_\gamma$ ($N = N_p - N_b$), $T_c$ is the sample counting time in seconds, $I_{\gamma}(E_\gamma)$ is the $\gamma$-ray emission probability (gamma yield), $\varepsilon(E_\gamma)$ is the detection efficiency at energy $E_\gamma$, and $M$ is the mass of measured sample (kg).

![Efficiency Calibration Curves](image)

Figure 2. Efficiency curve of HPGe detector generated using GENIE-2000 software

### 2.3. EDXRF technique

EDXRF is one of the most effective nuclear techniques used to analyze major and trace elements and commonly used in the applications of science and industry. Using the Energy Dispersive X-ray Fluorescence (EDXRF) spectrometric analysis, the concentrations of major elements and heavy metals present in the studied rock samples were determined. The principal work of the X-ray fluorescence technique is based on releasing the X-ray photons with characteristic energy or wavelength when the individual atoms are excited by an external energy source. Therefore, the elements present in the sample
can be determined and quantified by counting the number of photons of each energy emitted from a sample. In the present study, five grams of the fine powder of rock sample was mixed with one gram of boric acid. The mixture was carefully pressed into circular pellets of 30 mm diameter using a 20-ton hydraulic press. The prepared pellets were then loaded into the sample chamber then, the chemical contents of the rock samples were analyzed using an X-ray fluorescence spectrometer (XRF, SPECTRO XEPOS, AMETEK). The instrument (SPECTRO XEPOS) was controlled by a software computer based on the X-LAB pro menu. This instrument characteristic included: 50 watts end-window X-ray tube, up to eight polarization, and secondary targets, automatic sample changer, SSD detection system. Standards used in XRF peak calibration are as follows: FLX-SP1 glass tablets, 38.0 voltage/kV, 15.0 current/μA for MCA peak calibration, and FLX-SP2 along with the global calibration. The typical form of the fluorescent spectral lines obtained for rock samples (typical sample-R4) is shown in Figure 3.

Figure 3. A typical EDXRF spectrum for the rock sample-R4

3. Estimation of radiological health risk parameters

3.1. Radium equivalent activity (Raeq)
The radium equivalent activity is a significant index representing the specific activities of natural radionuclides $^{226}$Ra, $^{232}$Th, and $^{40}$K by a single quantity which takes into account the radiation hazards associated with them. This index was calculated using the following formula [17]:

$$Ra_{eq} \text{ (Bq/kg)} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_{K}$$

(2)

Where $A_{Ra}$, $A_{Th}$, and $A_{K}$ are the activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K, respectively.

3.2. Absorbed dose rate in air ($D$)
The measured activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K can be converted it into doses (nGy/h per Bq/kg) by adding radium, thorium and potassium 0.462, 0.604 and 0.042 factors respectively [3]. Using
the following equation, these factors determine the total absorbed gamma dose rate in air at 1 meter above ground level [18].

\[ D_{ab} = 0.462 A_{Ra} + 0.604 A_{Th} + 0.0417 A_{K} \]  \hspace{1cm} (3)

3.3. Annual effective dose equivalent (AEDE)

The conversion coefficient from the absorbed dose in air to the effective dose must be made for the indoor and outdoor occupancy factors respectively, for measurement the annual effective doses (indoors & outdoors). United nations scientific committee on the effects of atomic radiation (UNSCEAR) recommended the conversion coefficient, 0.7 Sv Gy\(^{-1}\). Adults spend about 80% of their time indoors, while the remaining 20% of their time outdoors is spent. So the factors of indoor and outdoor occupancy were provided by UNSCEAR as 0.8 and 0.2, respectively [3]. The annual effective doses (indoor & outdoor) in mSv were therefore calculated by the following equations:

\[ AEDE_{\text{(indoor)}} = D_{ab} \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ Sv Gy}\(^{-1}\) \times 10^{-6} \]  \hspace{1cm} (4)

\[ AEDE_{\text{(outdoor)}} = D_{ab} \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv Gy}\(^{-1}\) \times 10^{-6} \]  \hspace{1cm} (5)

3.4. Annual gonadal dose equivalent (AGDE)

This index is a measure of the genetic significance of the annual dose equivalent obtained by the reproductive organs of the population since the gonads are known as the organs of interest, along with the active bone marrow and bone surface cells. AGDE (mSv y\(^{-1}\)) due to the specific activities of \(^{226}\)Ra, \(^{232}\)Th, and \(^{40}\)K was determined using the following equation [19]:

\[ AGDE \text{ (mSv y}\(^{-1}\)) = 3.09 A_{Ra} + 4.18 A_{Th} + 0.31 A_{K} \]  \hspace{1cm} (6)

3.5. External and internal hazard index (H\(_{ex}\) & H\(_{in}\))

The external hazard index is given as follows for the measurement of the gamma radiation dose expected to be delivered externally from building materials [20].

\[ H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \]  \hspace{1cm} (7)

Radiation risk is negligible if the external hazard index maximum value is less than unity (H\(_{ex}\) \(\leq 1\)), which is equivalent to a maximum value of the Ra\(_{eq}\) activity < 370 Bq kg\(^{-1}\). The inhalation exposure for radon (\(^{222}\)Rn) gas and its progeny products or other radionuclide ingestion is called internal exposure. Radon is present in all building materials and is a carcinogenic material. The internal hazard index for the measurement of radon exposure is therefore given as follows [20]:

\[ H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \]  \hspace{1cm} (8)

3.6. Gamma index (I\(_{\gamma}\))

This index called the representative level index and commonly used to estimate \(\gamma\)-radiation arising from the natural radionuclide in the environmental samples. The European Commission’s proposed gamma
index \( I_y \) has been calculated from the activity concentrations of \( ^{226}\text{Ra}, \ ^{232}\text{Th}, \) and \( ^{40}\text{K} \) in the samples as indicated in the following equation [10]:

\[
I_y = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500}
\]

### 3.7. Alpha index \( I_\alpha \)

The radioactive alpha particles are emitted due to radon progeny disintegration and attach to aerosols, dust, and other particles in the air. As we inhale, the progeny of radon is deposited on the airways lining cells where the alpha particles can damage DNA and potentially cause lung cancer. The excess alpha radiation due to inhalation of radon from building materials is estimated using the following formula [21]:

\[
I_\alpha = \left( \frac{A_{Ra}}{200} \right)
\]

### 3.8. Excess lifetime cancer risk (ELCR)

Long-term radiation exposure is expected to present some cancer-causing risks. That means that people are at risk of having cancer. ELCR is another risk that somebody might have of getting cancer if that person is exposed to cancer-causing materials for a longer period of time. Therefore, the following relation was calculated on the basis of estimated values of the annual effective dose rate for excess lifetime cancer risk.

\[
ELCR \ (mSv/yr) = AEDE \times DL \times RF
\]

Where AEDE is the annual effective dose equivalent, DL duration life or average human age (approximately 70 years) and RF risk factor \((Sv^{-1})\) is a fatal cancer risk per sievert. For stochastic effects, ICRP-60 uses values of 0.05 for the public [22].

### 3.9. Activity utilization index (AUI)

The main factors affecting the dose absorbed indoors are the activity concentrations of natural radionuclides in the building materials. Radiation emitted from outdoor sources is absorbed well enough by the walls. The dose rate in indoor air will increase depending on the radionuclide activity concentration present in construction materials [23]. The concentrations of radionuclides in building materials are the main factors that affect the indoor absorbed dose. Radiation emitted by sources outdoors is well enough absorbed by the walls. The dose rate in air indoors will be increased according to the activity concentration of radionuclides present in construction materials [23]. To promote the determination of dose rates in air from various combinations of the three radionuclides in rock samples and by applying the correct conversion factors, the following formula can be used to calculate an activity utilization index (AUI).

\[
AUI = \left( \frac{A_{Ra}}{50 \ Bq/kg} \right)f_{Ra} + \left( \frac{A_{Th}}{50 \ Bq/kg} \right)f_{Th} + \left( \frac{A_{K}}{500 \ Bq/kg} \right)f_{K}
\]

Where \( A_{Ra}, A_{Th}, \) and \( A_{K} \) are the activities and where \( f_{Ra}, f_{Th} \) and \( f_{K} \) are the fractional contributions due to these radionuclides to the total gamma dose rate in air \((f_{Ra}= 8.09\%, \ f_{Th}= 47.98\%, \ f_{K} = 43.98\%)\) [23].

### 3.10. Criteria formula (CF)
Kreiger [21] investigated a value of 1.5 mGy on the basis of models proposed by Krisiuk [24] when estimating the annual external dose of radiation in dwellings constructed from construction materials with a Ra\text{eq} value of 370 Bq kg\(^{-1}\). Such authors later updated their equations to compensate for the existence of windows and doors, taking into account a wall of finite thickness and adding a weighting factor of 0.7 [25]. Their results, based on the following formula, can be used as a standard for limiting the annual radiation dose from construction materials.

\[
CF = \left(\frac{A_{Ra}}{740 \text{ Bq/kg}}\right) + \left(\frac{A_{Th}}{520 \text{ Bq/kg}}\right) + \left(\frac{A_{K}}{9620 \text{ Bq/kg}}\right)
\]  

4. Results and Discussion

4.1. Activity concentration

The specific activity concentrations of \(^{226}\text{Ra}\), \(^{232}\text{Th}\), and \(^{40}\text{K}\) were measured in rock samples and the results obtained were listed in Table 1. The concentration of activity of \(^{226}\text{Ra}\) ranges from 3.603 to 12.191 Bq kg\(^{-1}\) with an average value of 6.883 Bq kg\(^{-1}\). On the other hand, \(^{232}\text{Th}\) activity concentration ranges from 7.217 to 13.984 Bq kg\(^{-1}\) with an average value of 10.841 Bq kg\(^{-1}\). Similarly, \(^{40}\text{K}\) activity ranged between 67.904 to 190.362 Bq kg\(^{-1}\) with an average value of 128.616 Bq kg\(^{-1}\). It has been observed that the \(^{232}\text{Th}\) activity present in all the samples is higher than the \(^{226}\text{Ra}\) activity. The variation in the concentration of radionuclides depends on the type of rocks, the location where the rocks were collected and the geological formation of these rocks. However, the mean activity concentrations obtained for the samples of \(^{226}\text{Ra}\), \(^{232}\text{Th}\), and \(^{40}\text{K}\) are 6.883, 10.841, and 128.616 Bq kg\(^{-1}\), respectively, which are also lower than the global average values of 35, 30, and 400 Bq kg\(^{-1}\), respectively, as recommended by the United Nations Scientific Committee on Atomic Radiation Effects (UNSCEAR) [3]. The activity concentration of \(^{40}\text{K}\) is found higher than the \(^{226}\text{Ra}\) and \(^{232}\text{Th}\) activities. The findings showed that the average value of \(^{40}\text{K} >^{232}\text{Th} >^{226}\text{Ra}\). It is clear that potassium contributes to the area's most important activity as compared to radium and thorium. This may be due to the high content of potash feldspars such as microline and orthoclase, or micas such as muscovite and biotite since rocks rich in these minerals have high potassium content.

| Sample ID | Sample Location | Latitude | Longitude | \(^{226}\text{Ra}\) | \(^{232}\text{Th}\) | \(^{40}\text{K}\) | \(^{226}\text{Ra} /^{232}\text{Th}\) | \(^{232}\text{Th} /^{226}\text{Ra}\) | \(^{232}\text{Th} /^{40}\text{K}\) |
|------------|-----------------|----------|-----------|----------------|----------------|----------------|----------------|----------------|----------------|
| R1         | Harsul          | 19.926436| 75.329620 | 3.603          | 7.985          | 190.362        | 0.451          | 2.216          | 0.042          |
| R2         | Ohar            | 19.946103| 75.313918 | 5.393          | 7.975          | 156.755        | 0.676          | 1.479          | 0.051          |
| R3         | Jatwada         | 19.956592| 75.285846 | 6.827          | 13.205         | 93.674         | 0.517          | 1.934          | 0.141          |
| R4         | Turn            | 19.966837| 75.273615 | 5.555          | 10.430         | 99.780         | 0.533          | 1.878          | 0.105          |
| R5         | Sarai           | 20.019780| 75.223154 | 7.640          | 13.075         | 128.029        | 0.584          | 1.711          | 0.102          |
| R6         | Maratha Dhaba   | 20.023803| 75.186384 | 10.830         | 11.003         | 67.904         | 0.984          | 1.016          | 0.162          |
| R7         | Takli Phata lamangaon | 20.048840| 75.196791 | 6.661          | 12.120         | 115.787        | 0.550          | 1.820          | 0.105          |
| R8         | Mhaismal Begin  | 20.052340| 75.188994 | 7.051          | 13.703         | 140.736        | 0.515          | 1.943          | 0.097          |
4.2. Activity ratios

The activity ratios were determined from the measured activity of radionuclides in the samples. It can be seen from Table 1 that the $^{226}\text{Ra}/^{232}\text{Th}$, $^{232}\text{Th}/^{226}\text{Ra}$, $^{232}\text{Th}/^{40}\text{K}$, and $^{226}\text{Ra}/^{40}\text{K}$ ratios vary in the ranges $0.451$–$0.984$, $1.016$–$2.216$, $0.042$–$0.162$, and $0.019$–$0.159$ with corresponding mean values of $0.636$, $1.646$, $0.091$, and $0.060$, respectively. These ratios can be used as an indicator of the relative occurrence of these radionuclides. Figure 4 illustrates the spatial distribution of radionuclides.

|   |   |   |   |   |   |
|---|---|---|---|---|---|
| R9 | Mhaismal TV Tower | 20.091809 | 75.180060 | 6.224 | 8.367 | 101.841 | 0.744 | 1.344 | 0.082 | 0.061 |
| R10 | Khuldabad Guest | 20.016350 | 75.182750 | 4.641 | 7.217 | 80.146 | 0.643 | 1.555 | 0.090 | 0.058 |
| R11 | Kagazipura | 19.974661 | 75.211669 | 6.044 | 11.999 | 144.610 | 0.504 | 1.985 | 0.083 | 0.042 |
| R12 | Daultabad | 19.947353 | 75.220381 | 12.191 | 13.984 | 154.884 | 0.872 | 1.147 | 0.090 | 0.079 |
| R13 | Mhaismal Lake | 20.076728 | 75.186162 | 7.104 | 10.706 | 169.058 | 0.664 | 1.507 | 0.063 | 0.042 |
| R14 | Ghodegaon | 20.009619 | 75.261971 | 6.603 | 10.005 | 157.057 | 0.660 | 1.515 | 0.064 | 0.042 |
| Minimum | – | – | 3.603 | 7.217 | 67.904 | 0.451 | 1.016 | 0.042 | 0.019 |
| Maximum | – | – | 12.191 | 13.984 | 190.362 | 0.984 | 2.216 | 0.162 | 0.159 |
| Average | – | – | 6.883 | 10.841 | 128.616 | 0.636 | 1.646 | 0.091 | 0.060 |
| SD | – | – | 2.244 | 2.290 | 36.280 | 0.150 | 0.341 | 0.033 | 0.032 |
| Recommended value | – | – | 35 | 30 | 400 | – | – | – | – |

Figure 4. Spatial distribution of $^{226}\text{Ra}$ and $^{232}\text{Th}$ in rock samples
The ratio $^{232}\text{Th}/^{226}\text{Ra}$ was calculated to define the oxidizing or reducing state of radium and thorium. This ratio is between 1.016 and 2.216. Uranium (radium) is generated by weathering and/or leaching, due to surface runoff. Rock is consists of heavy and light minerals. An activity ratio ($^{232}\text{Th}/^{40}\text{K}$) was computed to identify the presence of heavy or light minerals in the rock samples. Calculated $^{232}\text{Th}/^{40}\text{K}$ values range from 0.042 to 0.162. These values are low in all sampling locations indicating the presence of light minerals in the study area. However, there are no high values (> 0.5) of $^{232}\text{Th}/^{40}\text{K}$ ratio which indicates the absence of heavy minerals. Hence, it can be concluded that light minerals such as quartz, mineral feldspar group, and kaolinite are abundant in the study region.

4.3. Radioelements concentration

The elemental concentrations of radium (ppm), thorium (ppm), and potassium (%) were calculated from measured activity concentrations of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in Bq/kg using conversion factors recommended by the IAEA [26] [1 ppm Ra = 11.1 Bq/kg of $^{226}\text{Ra}$, 1 ppm Th = 4.06 Bq/kg of $^{232}\text{Th}$, 1% K = 313 Bq/kg of $^{40}\text{K}$]. The distribution of elemental concentrations is shown in Table 2 with concentration ranging from 0.325 to 1.098 ppm, and between 1.778 to 3.444 ppm, respectively, for $^{226}\text{Ra}$ and $^{232}\text{Th}$, and between 0.217 to 0.608% for $^{40}\text{K}$. The radium to thorium ratio was calculated and listed in Table 2 to assess the elemental abundances of these natural radionuclides within the study area. The mean value of the ratio $^{226}\text{Ra}/^{232}\text{Th}$ was 0.232. The radium-thorium relationship can be considered in terms of ratio Th/Ra. The elemental concentration ratio (Th/Ra) was computed. This ratio is between 2.777 and 6.052, with an average value of 4.501. This result indicates that the ratio is higher than the average of 3.82 for the continental crust [27]. The theoretical Th/Ra ratio for a typical continental crust is approximately 3.0 and in this study, the ratio values obtained for all samples were greater than 3.0. Elemental ratio analysis of Ra/Th and Th/Ra can also be useful in studying the enrichment/depletion process due to complex metamorphic history, alteration, and weathering processes that affect the rocks.

| Sample ID | $^{226}\text{Ra}$ (ppm) | $^{232}\text{Th}$ (ppm) | $^{40}\text{K}$ (%) | $^{226}\text{Ra}/^{232}\text{Th}$ | $^{232}\text{Th}/^{226}\text{Ra}$ |
|-----------|-------------------------|-------------------------|---------------------|-----------------------------|-----------------------------|
| R1        | 0.325                   | 1.967                   | 0.608               | 0.165                       | 6.052                       |
| R2        | 0.486                   | 1.964                   | 0.501               | 0.247                       | 4.041                       |
| R3        | 0.615                   | 3.252                   | 0.299               | 0.189                       | 5.288                       |
| R4        | 0.500                   | 2.569                   | 0.319               | 0.195                       | 5.138                       |
| R5        | 0.688                   | 3.220                   | 0.409               | 0.214                       | 4.680                       |
| R6        | 0.976                   | 2.710                   | 0.217               | 0.360                       | 2.777                       |
| R7        | 0.600                   | 2.985                   | 0.370               | 0.201                       | 4.975                       |
| R8        | 0.635                   | 3.375                   | 0.450               | 0.188                       | 5.315                       |
| R9        | 0.561                   | 2.061                   | 0.325               | 0.272                       | 3.674                       |
| R10       | 0.418                   | 1.778                   | 0.256               | 0.235                       | 4.254                       |
| R11       | 0.545                   | 2.955                   | 0.462               | 0.184                       | 5.422                       |
| R12       | 1.098                   | 3.444                   | 0.495               | 0.319                       | 3.137                       |
| R13       | 0.640                   | 2.637                   | 0.540               | 0.243                       | 4.120                       |
| R14       | 0.595                   | 2.464                   | 0.502               | 0.241                       | 4.141                       |
Results of the activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in the present study were compared with the global average concentration as well as with similar investigations in different places of India and summary findings were listed in Table 3. It is found that the concentrations of the radionuclides in the present study are lower than the worldwide average and lower than the reported values of different regions of India. As shown in Table 3, the radioactivity concentration in rock samples varied from one area to another depends on the locality geological conditions.

Table 3. Comparison of natural radioactivity concentration (Bq/kg) in the present rock samples with the reported values from different areas of India.

| Location                      | Average activity concentration (Bq/kg) | References |
|-------------------------------|---------------------------------------|------------|
|                               | $^{226}$Ra | $^{232}$Th | $^{40}$K |
| World average                 | 50        | 50        | 500     | [3]       |
| Indian average                | 32        | 64        | 400     | [3]       |
| Hassan district, Karnataka    | 50.9      | 79.6      | 609.2   | [28]      |
| Coorg District, Karnataka     | 41.08     | 86.26     | 869.29  | [29]      |
| Ramanagara and Tumkur         | 41.08     | 86.26     | 869.29  | [30]      |
| Dhanbad city of Jharkhand     | 3.01      | 18.45     | 311.74  | [31]      |
| Thiruvannamalai, Tamilnadu    | 25.88     | 42.82     | 560.6   | [32]      |
| Mandya district               | 30.5      | 34.4      | 700.2   | [33]      |
| North Karnataka               | 52.76     | 71.51     | 1035    | [34]      |
| Bangalore rural district      | 93.2      | 306.2     | 1074.4  | [35]      |
| Kaiga, India                  | 14.2      | 11.5      | 866.2   | [36]      |
| Mysore city                   | 52.9      | 73.8      | 750.1   | [37]      |
| Aurangabad city               | 6.883     | 10.841    | 128.616 | Present study |

4.4. Radiological health risk assessment

The rocks of the study area are used for various purposes, mainly as building materials; hence the need to assess the radiological hazards related with the radionuclides by calculating the radiological risk parameters such as radium equivalent activity, absorbed dose rate, annual effective doses, annual gonadal dose equivalent, etc., which given in Tables 4 & 5. Table 4 indicates that the ($Ra_{eq}$) value ranges from 21.1326 to 44.1142 Bq kg$^{-1}$ with a mean value of 32.2851 Bq kg$^{-1}$ and a standard deviation of 5.5261. The mean radium equivalent activity value was below the 370 Bq kg$^{-1}$ safe limits set by the organization for
economic cooperation and development (OECD) [38]. The absorbed dose rate ranged between 9.8453 and 20.5372 nGy h⁻¹ with a mean value of 15.0893 nGy h⁻¹. This mean value was lower than the world average value of 58 nGyh⁻¹ provided by UNSCEAR [3]. The annual effective dose values indoors ranged from 0.0483 to 0.1007 mSv yr⁻¹ with a mean value 0.0740 mSv y⁻¹ while outdoors ranged from 0.0121 to 0.0252 mSv y⁻¹ with a mean value 0.0185 mSv y⁻¹. The annual effective dose is noted below the international recommended value 1 mSv y⁻¹ for the general public [3]. The annual effective dose (indoor & outdoor) values ranged from 0.0483 to 0.1007 mSv y⁻¹ with a mean value 0.0740 mSv y⁻¹. The AGDE values range from 0.0694 to 0.1441 mSv/y with an average of 0.1065 mSv y⁻¹. The average AGDE values of rock samples do not exceed the acceptable limits of 0.36 mSv y⁻¹, suggesting that the harmful effects of these radiances are insignificant. Figure 5 shows a comparison of the effective annual dose (indoor & outdoor) and annual gonadal dose equivalent of the rock sample. It can be noted that the average annual gonadal dose has the highest values in comparison with the effective annual dose (indoor & outdoor). The external hazard index ranges from 0.0571 to 0.1191 with a mean value of 0.0872, while the internal hazard index values in rock samples vary from 0.0696 to 0.1203 with an average value of 0.1058. These values are less than unity, indicating that the rock samples are safe and can be used as building material without significant harmful effects to the general public. Figure 6 provides a comparison of calculated values of internal and external radiation hazard indices at each sampling site which indicates that the internal radiation hazard index is higher than the external radiation hazard index.

Table 4. Radium equivalent activity, absorbed dose, annual effective dose (indoor & outdoor), annual gonadal dose equivalent and hazard indices for the investigated rock samples

| Sample ID | Raeq (Bq kg⁻¹) | D (nGy h⁻¹) | AEDE (indoor) (mSv y⁻¹) | AEDE (outdoor) (mSv y⁻¹) | AGDE (mSv/y) | H_down | H_int |
|-----------|----------------|-------------|--------------------------|---------------------------|--------------|--------|-------|
| R1        | 29.6645        | 14.4186     | 0.0707                   | 0.0177                    | 0.1035       | 0.0801 | 0.0898|
| R2        | 28.8530        | 13.8385     | 0.0679                   | 0.0169                    | 0.0986       | 0.0779 | 0.0925|
| R3        | 32.9231        | 15.0361     | 0.0738                   | 0.0184                    | 0.1053       | 0.0889 | 0.1074|
| R4        | 28.1530        | 13.0270     | 0.0639                   | 0.0160                    | 0.0917       | 0.0760 | 0.0910|
| R5        | 36.1955        | 16.7658     | 0.0823                   | 0.0206                    | 0.1180       | 0.0978 | 0.1184|
| R6        | 31.7929        | 14.4809     | 0.0710                   | 0.0178                    | 0.1005       | 0.0859 | 0.1151|
| R7        | 32.9082        | 15.2262     | 0.0747                   | 0.0187                    | 0.1071       | 0.0889 | 0.1069|
| R8        | 37.4830        | 17.4029     | 0.0854                   | 0.0213                    | 0.1227       | 0.1012 | 0.1203|
| R9        | 26.0306        | 12.1759     | 0.0597                   | 0.0149                    | 0.0858       | 0.0703 | 0.0871|
| R10       | 21.1326        | 9.8453      | 0.0483                   | 0.0121                    | 0.0694       | 0.0571 | 0.0696|
| R11       | 34.3375        | 16.0699     | 0.0788                   | 0.0197                    | 0.1137       | 0.0927 | 0.1091|
| R12       | 44.1142        | 20.5372     | 0.1007                   | 0.0252                    | 0.1441       | 0.1191 | 0.1521|
| R13       | 35.4140        | 16.7903     | 0.0824                   | 0.0206                    | 0.1191       | 0.0956 | 0.1148|
| R14       | 32.9890        | 15.6361     | 0.0767                   | 0.0192                    | 0.1109       | 0.0891 | 0.1069|
| Minimum   | 21.1326        | 9.8453      | 0.0483                   | 0.0121                    | 0.0694       | 0.0571 | 0.0696|
|         | Maximum | Mean     | SD      | Permissible limits |
|---------|---------|----------|---------|-------------------|
| 2020    | 44.1142 | 32.2851  | 5.5261  | 370               |
| 2020    | 20.5372 | 15.0893  | 2.5517  | 57                |
| 2020    | 0.1007  | 0.0740   | 0.0125  | 0.46              |
| 2020    | 0.0252  | 0.0185   | 0.0031  | 0.07              |
| 2020    | 0.1441  | 0.1065   | 0.0179  | 0.36              |
| 2020    | 0.1191  | 0.0872   | 0.0149  | < 1               |
| 2020    | 0.1203  | 0.1058   | 0.0196  | < 1               |

**Figure 5.** A comparison of annual effective dose (indoor & outdoor) and annual gonadal dose.

**Figure 6.** Comparison of external (H\text{ex}) and internal (H\text{in}) radiation hazard indices.
The gamma index ($I_\gamma$) was calculated for the rock samples and given in Table 5. The values ranged from 0.1565 to 0.3244 with the mean value of 0.2400 which is less than unity and thus the rock samples are safe for building houses. However, $\alpha$-index was found to range from 0.0180 to 0.0609 with an average value of 0.0344 and a standard deviation of 0.0112. All values of $I_\alpha$ are noted to be below the maximum permissible value of $I_\alpha = 1$. The calculated values of excess lifetime cancer risk ranged from 0.0424 to 0.0882 mSv/y with an average of 0.0648 mSv/y. This average value of ELCR is lower than the worldwide recommended value of 0.290 mSv/y [3]. The AUI values were between 0.1473 to 0.2902 with an average of 0.2283. These values matched AUI < 2, which corresponds to an annual effective dose of $\leq$ 0.3 mSv y$^{-1}$ [39]. It means that these rocks can be used safely for the construction of buildings. The calculated criteria values from the sum of the three quotients for the annual radiation dose associated with the studied rock samples are given in Table 5. The values for criterion range from 0.0285 to 0.0595 with an average of 0.0435. The mean value of the analyzed samples is well below the recommended maximum value < 1. This indicates that $\gamma$-activity does not exceed the proposed criterion limit in the rock samples, and these rocks can be used safely in the construction of dwellings. Figure 7 shows the variation of excess lifetime cancer risk, activity utilization index, and criteria formula in the investigated rock samples.

### Table 5. Radiological hazard parameters: Gamma index ($I_\gamma$), Alpha index ($I_\alpha$), Excess lifetime cancer risk (ELCR), Activity utilization index (AUI) and Criteria formula (CF) for the samples

| Sample ID | $I_\gamma$ | $I_\alpha$ | ELCR (mSv/y) | AUI  | CF  |
|-----------|------------|------------|--------------|------|-----|
| R1        | 0.2307     | 0.0180     | 0.0620       | 0.2499 | 0.0400 |
| R2        | 0.2201     | 0.0269     | 0.0592       | 0.2231 | 0.0389 |
| R3        | 0.2400     | 0.0341     | 0.0644       | 0.2202 | 0.0444 |
| R4        | 0.2079     | 0.0278     | 0.0560       | 0.1968 | 0.0379 |
| R5        | 0.2670     | 0.0382     | 0.0721       | 0.2504 | 0.0488 |
| R6        | 0.2275     | 0.0542     | 0.0623       | 0.1828 | 0.0429 |
| R7        | 0.2428     | 0.0333     | 0.0655       | 0.2289 | 0.0443 |
| R8        | 0.2779     | 0.0353     | 0.0746       | 0.2667 | 0.0505 |
| R9        | 0.1931     | 0.0311     | 0.0522       | 0.1799 | 0.0351 |
| R10       | 0.1565     | 0.0232     | 0.0424       | 0.1473 | 0.0285 |
| R11       | 0.2567     | 0.0302     | 0.0690       | 0.2521 | 0.0463 |
| R12       | 0.3244     | 0.0609     | 0.0882       | 0.2902 | 0.0595 |
| R13       | 0.2670     | 0.0355     | 0.0721       | 0.2629 | 0.0478 |
| R14       | 0.2487     | 0.0330     | 0.0672       | 0.2448 | 0.0445 |
| Minimum   | 0.1565     | 0.0180     | 0.0424       | 0.1473 | 0.0285 |
| Maximum   | 0.3244     | 0.0609     | 0.0882       | 0.2902 | 0.0595 |
| Mean      | 0.2400     | 0.0344     | 0.0648       | 0.2283 | 0.0435 |
| SD        | 0.0404     | 0.0112     | 0.0109       | 0.0396 | 0.0075 |
| Permissible limits | $\leq 1$ | $< 1$ | 0.29 | 2 | $< 1$ |
4.5. Chemical Analysis by EDXRF

The elemental analysis by X-ray fluorescence (XRF) has been performed to determine the major and trace element concentrations and their oxides present in the rock samples. Table 6 shows the percentage of concentrations of the major oxide elements. The obtained analysis data indicated that the major oxides present in samples are SiO$_2$, Al$_2$O$_3$, K$_2$O, CaO, TiO$_2$, Na$_2$O, MgO, P$_2$O$_5$, MnO, and Fe$_2$O$_3$. The SiO$_2$ concentration was found to be the greater dominant element than other main elements in the rock samples. Its concentration ranged from 36.07 to 61.28% with an average percentage value of 53.40%. This element has a much higher value relative to the crustal average. This may be due to the rock structure and type of rocks. Iron oxide was the second dominant element in the samples and their concentration ranged from 1.248 to 16.97% with an average of 13.176% which is higher than the crustal average value of 4.72% suggesting that a large amount of the element may have resulted from the weathering of silicate-bearing rocks rich in feldspar and mica. The measured concentrations of major elements oxides were compared with the crustal average values which found to be higher except K$_2$O, which reflects the low activity of $^{40}$K in these rocks. Moreover, we noted that rocks from the study area are rich with the oxides of the elements indicating that the presence of natural radionuclides with different concentrations in these rocks can be attributed by elements oxides. The concentrations of SiO$_2$, Fe$_2$O$_3$, Al$_2$O$_3$, and CaO account for more than 80% of major oxides present in the rocks, while MgO, Na$_2$O, TiO$_2$, K$_2$O, P$_2$O$_5$, and MnO account for less than 15% of the oxide concentration in samples. Generally, the average concentration of major oxides in the samples was in this formed order: SiO$_2$$>$Fe$_2$O$_3$$>$Al$_2$O$_3$$>$CaO$>$MgO$>$Na$_2$O$>$TiO$_2$$>$K$_2$O$>$P$_2$O$_5$$>$MnO. The variability in the major element concentrations may be due to the difference in their mineral content.

**Figure 7.** Bar chart representing value of ELCR, AUI and CF in rock samples
The heavy metal content in the rock samples was measured in the ppm unit using EDXRF and findings summarized in Table 7. It involves V, Cr, Co, Ni, Cu, Zn, As, Ba, Cd, Hg, and Pb. The elemental concentrations measured were compared with the crustal average. The mean concentration of V, Co, Cu, Zn, Cd, and Hg was found to be above average shale concentration value, while the other elemental concentration was below the average shale concentration. The most important heavy metal detected in the samples was barium. The Ba concentration ranged from 160 to 565 ppm with an average of 294.93 ppm that is lower than the average crustal value of 580 ppm. It had the highest value of concentration in the samples. The variation in Ba concentration may be due to the formation of a considerable amount of barites (BaSO₄) in the carbonates [41]. Vanadium (V) was the second most abundant heavy metal detected in the samples and their concentration ranged from 21.2/430 ppm with an average value of 269.01 ppm above the average crustal value of 130 ppm. The variation of the elemental concentrations in this study may be due to the nature of weathering processes as well as the velocity of transporting media. The total concentrations of trace elements in rocks depend not only on the input of the trace element but also on the mineral content of the rocks, which may vary from region to region. This is the reason refer to the variation of elemental concentration in rock samples. Generally, the relative abundance of the heavy metals in the samples was as this formed order: Ba>V>Cu>Zn>Cr>Ni>Cd>Pb>As>Hg. The variation of the heavy metals concentrations in the rock samples was presented in Figure 8.
Table 7. Heavy metals concentration of the investigated rock samples (ppm)

| Sample No | V  | Cr  | Co  | Ni  | Cu  | Zn  | As  | Ba  | Cd  | Hg  | Pb  |
|-----------|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| R1        | 93 | 98.2| 36.7| 50.2| 186.6| 74.9| 0.4 | 268 | 2.70| 0.96| 2.70|
| R2        | 324| 95.6| 46.8| 52.6| 196.8| 97.2| 1.1 | 324 | 1.90| 0.81| 4.70|
| R3        | 174| 68.3| 63.9| 76.1| 289.8|109.6| 0.80| 242 | 1.86| 0.58| 4.00|
| R4        | 290| 44.7| 43.5| 66.1| 249.2|123 | 0.50| 543 | 4.40| 0.79| 5.50|
| R5        | 430| 106 | 66.80| 58.30| 241.2|129.4| 0.40| 322 | 1.54| 0.81| 5.10|
| R6        | 417| 64.5| 57  | 50.1| 101.5|108.2| 1.5 | 211 | 1.88| 0.77| 1.50|
| R7        | 351| 98.1| 66.4| 92.2| 270.70|100.10| 1.3 | 229 | 1.93| 0.37| 4.50|
| R8        | 343| 71.10| 60.5| 77.5| 245.9|178.7| 2.80| 288 | 1.75| 0.61| 11.60|
| R9        | 417| 132.7| 65.4| 94.3| 303.8|133.4| 0.34| 160 | 1.90| 0.48| 6.40|
| R10       | 21.2| 11.9| 11| 9.20| 7.6 |165.3| 0.65| 565 | 1.84| 0.32| 15 |
| R11       | 207| 59.99| 42.05| 34.75| 219 |143 | 1.12| 260 | 2.05| 0.92| 12.74|
| R12       | 153| 76.02| 61.43| 27.84| 86.31|101 | 0.32| 306 | 1.65| 1.09| 17 |
| R13       | 240| 82.08| 54.91| 18.53| 117 |86.97| 0.60| 232 | 2.42| 0.16| 8.05|
| R14       | 306| 90.13| 44.85| 71.43| 228 |106.64| 1.04| 179 | 3.05| 0.99| 10.45|
| Min       | 21.2| 11.9| 11| 9.20| 7.6 |74.9 | 0.32| 160 | 1.54| 0.16| 1.50|
| Max       | 430| 132.7| 66.8| 94.3| 303.8|178.7| 2.8 | 565 | 4.4 | 1.09| 17 |
| Mean      | 269.01| 78.58| 51.52| 55.65| 195.96|118.39| 0.92| 294.93| 2.21| 0.69| 7.80|
| Crustal Average[40] | 130| 90| 19| 68| 45| 95| 13| 580| 0.3| 0.4| 20 |

Figure 8. Variation of heavy metals concentration in the investigated rock samples
4.6. Correlation Analysis

Correlation studies were performed to reveal the possible relationship between radionuclide concentrations and with radioactive variables. The Pearson correlation matrixes between radionuclide concentrations with itself as well as with radiological hazard parameters were calculated and presented in Table 8. The correlation terms were given as weak, moderate, and strong for coefficients of 0.20–0.40, 0.40–0.60, and r > 0.60, respectively [42]. There is a strong positive correlation between $^{226}$Ra and $^{232}$Th in the samples. The $^{232}$Th concentration was correlated with $^{40}$K negative. There is a negative correlation between $^{226}$Ra and $^{40}$K. The correlation value between $^{226}$Ra and $^{232}$Th was significantly higher as compared to that between $^{226}$Ra and $^{40}$K. $^{40}$K and $^{232}$Th, this significant positive correlation indicates that their rock content is mostly influenced and controlled by the similar source origin [42]. A high good positive correlation observed between $^{226}$Ra and $^{232}$Th because radium and thorium decay series occur combined together in nature. But a weak negative correlation between $^{40}$K and $^{226}$Ra, $^{232}$Th was observed, since the origins of $^{40}$K are in different decay series. There exists a strong positive correlation between $^{232}$Th and absorbed dose with the correlation coefficient ($r = 0.78$) (Figure 9). There is also a strong positive correlation between $^{226}$Ra and the absorbed dose. It is observed, a moderate positive correlation exists between $^{40}$K and the absorbed dose. There exists a very strong positive correlation with the correlation coefficient ($r = 0.83$) between $R_{eq}$ and $^{232}$Th. To determine ELCR dependence on the activity concentration and ELCR dependence on other radiological hazard indices, correlation analysis was performed between the $R_{eq}$ and ELCR, absorbed dose rate and ELCR. There was a very strong correlation between $R_{eq}$ and ELCR ($r = 1.0$), the absorbed dose rate and ELCR ($r = 1.0$) (Figure 10). There is no safe limit and all doses of radiation carry some form of risk. Hence, radiation-induced cancer and lifetime cancer cannot be prevented, only can be reduced by minimizing the radiation dose.

**Table 8.** Pearson correlation matrix of radionuclides concentration and radioactive variables

| Variables | $^{226}$Ra | $^{232}$Th | $^{40}$K | $R_{eq}$ | D | AEDE | AGE D | H$_{ex}$ | H$_{in}$ | I$_{a}$ | I$_{q}$ | ELCR |
|-----------|------------|------------|----------|---------|---|------|-------|---------|---------|--------|-------|------|
| $^{226}$Ra | 1.00       | -          | -        | -       | - | -    | -     | -       | -       | -      | -     | -    |
| $^{232}$Th | 0.59       | 1.00       | -        | -       | - | -    | -     | -       | -       | -      | -     | -    |
| $^{40}$K   | -0.22      | -0.29      | 1.00     | -       | - | -    | -     | -       | -       | -      | -     | -    |
| $R_{eq}$   | 0.66       | 0.83       | 0.34     | 1.00    | - | -    | -     | -       | -       | -      | -     | -    |
| D          | 0.61       | 0.78       | 0.45     | 0.99    | 1.00 | -    | -     | -       | -       | -      | -     | -    |
| AEDE       | 0.61       | 0.78       | 0.45     | 0.99    | 0.99 | 1.00 | -     | -       | -       | -      | -     | -    |
| AGED       | 0.57       | 0.75       | 0.50     | 0.99    | 0.99 | 0.99 | 1.00  | -       | -       | -      | -     | -    |
| H$_{ex}$   | 0.66       | 0.83       | 0.34     | 1.00    | 0.99 | 0.99 | 0.98  | 1.00    | -       | -      | -     | -    |
| H$_{in}$   | 0.82       | 0.83       | -0.06    | 0.97    | 0.95 | 0.95 | 0.97  | 0.97    | 1.00    | -      | -     | -    |
| I$_{a}$    | 0.58       | 0.78       | 0.47     | 0.99    | 0.99 | 0.99 | 0.99  | 0.99    | 0.94    | 1.00   | -     | -    |
| I$_{q}$    | 0.99       | 0.59       | -0.22    | 0.66    | 0.61 | 0.61 | 0.57  | 0.66    | 0.82    | 0.58   | 1.00  | -    |
| ELCR       | 0.61       | 0.78       | 0.45     | 0.99    | 0.99 | 0.99 | 0.99  | 0.99    | 0.95    | 0.99   | 0.61  | 1.00 |
Figure 9. Correlation between absorbed dose in air and activity of $^{232}$Th

Figure 10. Correlation between excess lifetime cancer risk and absorbed dose rate
The correlations between the radionuclides and elemental compositions of the rock samples were computed and the results were presented in Tables 9 & 10. $^{226}$Ra activity concentration showed weak correlation with all the major elements except Na$_2$O and MgO which had a moderate positive correlation with this radioisotope. $^{232}$Th activity concentration had a strong positive correlation with MgO ($r = 0.74$) and a moderate positive correlation with TiO$_2$, P$_2$O$_5$, and Fe$_2$O$_3$ however weak correlation with other major elements was noted in the samples. On the other hand, $^{40}$K showed a moderate negative correlation with Na$_2$O, and CaO and a weak correlation with other major elements. An interesting relation was observed between Al$_2$O$_3$ and Fe$_2$O$_3$ in the samples. Al had a strong positive correlation with Fe ($r = 0.84$). It is well known that an increase in some elements in the rock leads to a decrease in others. MnO is found to have a strong positive correlation with P$_2$O$_5$ and a moderate linear positive correlation with Na$_2$O. While MgO had a moderate positive correlation with Al$_2$O$_3$ and TiO$_2$ in rock samples. A significant correlation between Fe$_2$O$_3$ with Al$_2$O$_3$, K$_2$O, TiO$_2$, and MgO was observed.

**Table 9.** The Pearson correlation coefficients between radionuclides and major elements in rocks

| Variable | $^{226}$Ra | $^{232}$Th | $^{40}$K | SiO$_2$ | Al$_2$O$_3$ | K$_2$O | CaO | TiO$_2$ | Na$_2$O | MgO | P$_2$O$_5$ | MnO | Fe$_2$O$_3$ |
|----------|------------|------------|----------|--------|------------|-------|-----|--------|--------|-----|----------|------|-----------|
| $^{226}$Ra | 1.00       | -          | -        | -      | -          | -     | -   | -      | -      | -   | -        | -    | -         |
| $^{232}$Th | 0.59       | 1.00       | -        | -      | -          | -     | -   | -      | -      | -   | -        | -    | -         |
| $^{40}$K  | -0.22      | -0.29      | 1.00     | -      | -          | -     | -   | -      | -      | -   | -        | -    | -         |
| SiO$_2$   | 0.12       | 0.07       | -0.02    | 1.00   | -          | -     | -   | -      | -      | -   | -        | -    | -         |
| Al$_2$O$_3$ | 0.32      | 0.25       | -0.16    | -0.18  | 1.00       | -     | -   | -      | -      | -   | -        | -    | -         |
| K$_2$O   | -0.24      | -0.09      | -0.15    | 0.08   | -0.60      | 1.00  | -   | -      | -      | -   | -        | -    | -         |
| CaO      | 0.13       | 0.02       | -0.44    | -0.29  | 0.12       | 0.26  | 1.00| -      | -      | -   | -        | -    | -         |
| TiO$_2$  | 0.38       | 0.46       | -0.02    | 0.12   | 0.38       | -0.68 | -0.50| 1.00   | -      | -   | -        | -    | -         |
| Na$_2$O  | 0.42       | 0.19       | -0.51    | 0.38   | 0.57       | -0.17 | 0.29 | 0.27   | 1.00   | -   | -        | -    | -         |
| MgO      | 0.42       | 0.74       | -0.04    | -0.22  | 0.54       | -0.32 | -0.13| 0.58   | 0.31   | 1.00| -        | -    | -         |
| P$_2$O$_5$ | 0.27      | 0.40       | 0.27     | 0.06   | 0.02       | 0.04  | -0.05| 0.37   | 0.27   | 0.26| 1.00     | -    | -         |
| MnO      | 0.11       | 0.38       | 0.05     | 0.25   | 0.33       | 0.00  | -0.03| 0.32   | 0.44   | 0.24| 0.75     | 1.00 | -         |
| Fe$_2$O$_3$ | 0.21      | 0.42       | 0.05     | -0.12  | 0.84       | -0.67 | -0.31| 0.68   | 0.35   | 0.75| 0.13     | 0.38 | 1.00      |

The Pearson correlation between the natural radionuclides and the heavy metal contents in the samples were calculated and presented in Table 10. Radium-226 has a weakly negative correlation with Ni, Cu, Zn, Ba, and Cd. A moderate positive relation was noted between $^{226}$Ra and Co. There are no significant correlations between $^{226}$Ra and heavy metals. $^{232}$Th shows a significant correlation with Co ($r = 0.67$) and a negative correlation with Ba and Cd but has an insignificant positive relationship with Cr, Ni, Cu, Zn, As, V, Hg and Pb. However, $^{40}$K has no significant correlation with any of the heavy metals except Zn. Furthermore; it was found that there is no correlation between the $^{40}$K radionuclide and Co. The absence of correlations could be interpreted by the common independence or different behavior of the elements. From the result, it can be said that heavy metals may not be responsible for emitting radiation because heavy metals (Cr, Ni, Cu, Zn, As, Ba, V, Cd, Hg, and Pb) show weakly correlation with $^{226}$Ra, $^{232}$Th, and $^{40}$K in most of the cases. The heavy metals had strong positive correlations: Co with Cr, Cu and Ni, and V with Co and Ni and Cu with Ni. These strong correlations between heavy metals showing that the presence of one parameter may increase the concentration of the other and suggest their common origin. Similarly, Ba has strong or moderate negative correlations with Cr, Co, Ni, V, and Cu and for Pb with Cr,
Ni, Cu, and V. These indicate that if one of the elements in a given pair is elevated, the amount of other decreases.

Table 10. The Pearson correlation matrix for natural radionuclides and heavy metals

| Variable | $^{226}$Ra | $^{232}$Th | $^{40}$K | Cr | Co | Ni | Cu | Zn | As | Ba | V | Cd | Hg | Pb |
|----------|-----------|-----------|---------|----|----|----|----|----|----|----|----|----|----|----|----|
| $^{226}$Ra | 1.00 | - | - | - | - | - | - | - | - | - | - | - | - | - |
| $^{232}$Th | 0.59 | 1.00 | - | - | - | - | - | - | - | - | - | - | - | - |
| $^{40}$K | -0.22 | -0.29 | 1.00 | - | - | - | - | - | - | - | - | - | - | - |
| Cr | 0.03 | 0.02 | 0.39 | 1.00 | - | - | - | - | - | - | - | - | - | - |
| Co | 0.54 | 0.67 | 0.00 | 0.66 | 1.00 | - | - | - | - | - | - | - | - | - |
| Ni | -0.11 | 0.17 | -0.18 | 0.57 | 0.59 | 1.00 | - | - | - | - | - | - | - | - |
| Cu | -0.26 | 0.24 | 0.04 | 0.58 | 0.57 | 0.88 | 1.00 | - | - | - | - | - | - | - |
| Zn | -0.10 | 0.13 | -0.43 | -0.41 | -0.20 | 0.03 | 0.01 | 1.00 | - | - | - | - | - | - |
| As | 0.09 | 0.32 | -0.07 | -0.14 | 0.15 | 0.28 | 0.14 | 0.46 | 1.00 | - | - | - | - | - |
| Ba | -0.28 | -0.23 | -0.28 | -0.73 | -0.63 | -0.42 | -0.40 | 0.37 | -0.18 | 1.00 | - | - | - | - |
| V | 0.29 | 0.23 | -0.20 | 0.57 | 0.69 | 0.62 | 0.52 | 0.05 | 0.30 | -0.44 | 1.00 | - | - | - |
| Cd | -0.36 | -0.27 | 0.09 | -0.21 | -0.30 | 0.09 | 0.16 | -0.21 | -0.21 | 0.34 | -0.07 | 1.00 | - | - |
| Hg | 0.24 | 0.17 | 0.34 | 0.11 | 0.00 | 0.01 | 0.07 | -0.19 | -0.08 | -0.07 | -0.01 | 0.17 | 1.00 | - |
| Pb | 0.20 | 0.17 | 0.16 | -0.41 | -0.33 | -0.48 | -0.44 | 0.48 | 0.04 | 0.28 | -0.46 | -0.17 | 0.09 | 1.0 |

5. Conclusion

The concentrations of primordial radionuclides and chemical compositions of the selected rock samples from the study area have been measured using γ-ray spectrometry and the EDXRF technique. The average concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K were well below the permissible limits of 35, 30, and 400 Bq kg$^{-1}$, respectively reported by UNSCEAR-2000. The present study showed a non-uniform distribution in the study region of the natural radionuclides which depends on the geological formation and geochemical composition of the area in which the rock is formed. The rock samples in the study area are classified as basaltic rocks, and low-level radioactivity was detected from every site, as basaltic rocks have low background radioactivity. The variation found in the activities from $^{226}$Ra, $^{232}$Th and $^{40}$K in the samples, possibly due to rock properties such as porosity, density, geological formation, humidity, and chemical composition of samples. The activity ratios of the radionuclides around unity for most samples, indicating that radioactive secular equilibrium has been attained among their respective daughter nuclides. It was found that the total absorbed dose rate is lower than the world average of 60 nGr/h. The effective dose rates indoor and outdoor are below the recommended safety level of 1.0 mSv y$^{-1}$ proposed by UNSCEAR-2000. The annual gonadal dose equivalent is less than the recommended average world level of 0.36 mSv y$^{-1}$. The external and internal hazard indices are less than the recommended safe level unity and thus do not pose any radiation hazards to the inhabitants of the study area. The ELCR estimated was below the world average of 0.29. The calculated results obtained from AUI and Cf are less than the world average value (AUI < 2 & Cf < 1) and thus the samples are safe according to the report on radiation safety. From EDXRF analysis, indicated that the major elements present in samples are SiO$_2$, Al$_2$O$_3$, K$_2$O, CaO, TiO$_2$, Na$_2$O, MgO, P$_2$O$_5$, MnO, and Fe$_2$O$_3$. 


Furthermore, heavy metals were detected in the rock samples such as V, Cr, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb. Analysis of rocks by EDXRF help geochemists research the processes involved in forming various rocks by studying the major and trace elements and other rare earth elements. The data obtained here are reference values to be used as a database for drawing a radiological map of the study area and could be used as a database for future investigation. Overall, it can be concluded that there is no serious radiological threat to the public from the rocks in the study area, and it is safe to use for construction purposes.

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