Natural radioactivity estimation in local and imported granite used as building materials in Kanyakumari District, Tamil Nadu, India

F. S. **Karolin Mary**[1], **G. Shanthi**[2]

[1]Research Scholar (20213282132012), Department of Physics & Research Centre, Women's Christian College, Nagercoil-629001. Affiliated to Manonmaniam Sundaranar University, Abishekapatti, Tirunelveli -627012, Tamil Nadu, India.

[2] Assistant Professor, Department of Physics & Research Centre, Women's Christian College, Nagercoil-629001. Affiliated to Manonmaniam Sundaranar University, Abishekapatti, Tirunelveli -627012, Tamil Nadu, India.

(Corresponding Author: Email: karolinmary1996@gmail.com; Phone: +91-9445694211)

Abstract:

Natural radioactivity in building materials is a source of ongoing exposure for humans. The distribution of natural radionuclide γ-ray activities produced by ^{238}U, ^{232}Th and ^{40}K were analyzed using gamma ray spectrometer with NaI (Tl) detector for 10 granite samples collected from several local suppliers in Kanyakumari District. Every sample contained ^{40}K and radionuclides from the uranium and thorium series. The radionuclide activity concentrations measured were compared with the data that had been published globally. The geometric mean of ^{238}U, ^{232}Th, ^{40}K were 10.04, 101.54 and 222.30 Bq/kg respectively. Radiological hazard indices such as absorbed dose rate (D_R), radium equivalent activity (R_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), indoor and outdoor annual effective dose (mSv/y), were evaluated. The obtained results showed that the mean absorbed dose rate, radium equivalent activity, external hazard index, internal hazard index, indoor and outdoor annual effective dose were: 91.93nGy/h, 206.03 Bq/kg,0.51, 0.60, 0.14 $mSvy^{-1}$, 0.45 $mSvy^{-1}$ respectively.

Keywords

Building materials, granite, gamma ray spectrometer, activity concentration, radiological hazard parameters.

INTRODUCTION:

The world is naturally radioactive, and natural sources account for around 90% of human radiation exposure (Rani et al., 2005). The principal radioactive nuclides found in the earth's crust are uranium, thorium, and potassium. The ^{238}U and ^{232}Th series are made up of several daughter products produced by the sequential decay of parent radionuclides. These decay cascades generate radioactive daughter nuclides, which eventually culminate in the stable isotopes ^{208}Pb and ^{206}Pb. The naturally occurring uranium contains 99.2745% by weight ^{238}U, 0.7200% ^{235}U, 0.0055% ^{234}U (Harb et al.,2014). Higher-level radiation is associated with volcanic rocks such as granites. Some granite rock shale contains a high concentration of radionuclides.

Radionuclides are known kidney toxins and carcinogens in humans. Once in the system, radionuclides accumulate in many organs. Because of their extended half-lives (^{232}Th: 1.41010 years, ^{238}U: 4.47109 years, and ^{40}K: 1.28109 years) and chemical behaviour, they provide radiation dosages that cause changes in the structure of chromosomes, leading to the development of many disorders, including cancer. Even though a tiny amount of stable potassium is nutritionally vital to the human system, ^{238}U, ^{232}Th, and ^{40}K are radiotoxic.

Granite, like most natural stones, is a natural source of radiation. It contains veins of naturally occurring radioactive materials such as uranium and thorium, as well as the radioactive decay products of these elements. Because of its toughness, elegant appearance, and availability in a variety of colours, granites are utilised as building materials, decoration materials, and luxury materials. Granite comes in a variety of colours, including white and grey, as well as pink, red, green, blue, and black. The mineral makeup of the rock, the rate at which the magma cools and solidifies, and the presence of other minerals or contaminants all contribute to colour differences in granite. Granite that is mostly composed of feldspar and quartz will be white or grey in colour. The presence of trace amounts of other minerals can cause the rock to look speckled, with darker or lighter areas. Granite can be pink or red due to the presence of potassium feldspar. Depending on the percentage of potassium feldspar, the tint of pink or red can vary. Granite can be green due to the presence of minerals such as chlorite or epidote. These minerals are commonly found in granite that has been subjected to extreme heat and pressure. Granite can be blue due to the presence of minerals such as sodalite or lazurite. These minerals are commonly found in granite subjected to hydrothermal activity. Granite can be black due to the presence of minerals such as biotite or hornblende. These minerals' concentrations can vary, resulting in varying colours of black.

According to a review of the available literature, some researches have determined the levels of exposure due to natural radiation in Kanyakumari District, while others have studied the radioactivity of building materials in other Tamilnadu districts. Radioactivity levels in construction materials have been reported in several Indian states and in a number of other countries. However, no comprehensive investigation on exposure to natural radiation emitted by building materials has been conducted in Kanyakumari District. As therefore, because the people spend approximately 80% of their time indoors, it is critical to assess the concentration of radionuclides in granite before selecting material for use in the decoration of the home (Al-Zahrani 2017). The study's objectives are (i) to determine the specific radioactivity concentrations of ^{238}U, ^{232}Th, and ^{40}K in granites used as building materials in Kanyakumari District, and (ii) to assess the potential radiological risks to human health and compare the results to UNSCEAR data.

MATERIALS AND METHODS:

Background of the study area

Kanyakumari is one of Tamil Nadu's 38 districts and the southernmost district on Indian continent. When it comes to Tamil Nadu districts' population densities, it is ranked second. The Western Ghats mountains border the district of Kanniyakumari on its northern side, while the sea is present on three of its sides. The district is located between 8°03' and 8°35' north latitude and between 77°15' and 77°36' east longitude. The district shares borders with the Thiruvananthapuram District (Kerala) in the west, the Arabian Sea in the west, the Indian Ocean in the south, and the Tirunelveli district in the north and northeast. 1,870,374 people were living in the Kanniyakumari district as of the 2011 census. The district had a total of 483,539 households ("Kanyakumari." Wikipedia). A very high intrinsic anomalous radioactivity of 41.03 μSvh^{-1} was observed, in a famous tourist spot in the coastal belt of Kanyakumari District (Jeni Chandar Padua, et al., 2013)

Sample collection and preparation

Ten granite samples were acquired from several local vendors in the Kanyakumari District. The samples were cleaned and crushed into small pieces once they were collected. The crushed pieces were dried for 24 hours at 110°C before being ground into fine powder, sieved to produce roughly the same grain size (2mm), and placed in a 100 ml vial for gamma ray spectrometry. The samples were sealed for four weeks to allow the current radionuclides to reach equilibrium with their decay offspring. The jars were hermetically and externally sealed to prevent gas leakage caused by the internal overpressure caused by Ra decay. The concentration of natural radionuclides in the samples was determined using the gamma ray spectrometric technique after confirming secular equilibrium between Ra and its near daughter products.

Gamma ray spectrometry

The gamma ray spectrometer was utilised in the current study to assess the activity levels of ^{238}U, ^{232}Th, and ^{40}K in granite samples. The gamma spectrum was obtained using a 3"3" NaI (Tl) detector-based 1 k Multichannel analyzer (MCA). Using the linked laptop computer, the photomultiplier tube operating voltage (630 V) and pre amplifier gain (08) were tuned to yield roughly 3 keV/channel. To decrease the system's background, the detector was encased inside a large lead shield.

The energy and efficiency calibrations are always included in the calibration of the gamma ray detection system. The energy calibration converts channel numbers to gamma-ray energy in MeV, whereas the efficiency calibration attempts to calculate gamma ray counting efficiencies across the whole energy range of measurement. Here the system is calibrated using Co-60 and Cs-137. The polynomial curve fitting was done with observed channel number and corresponding photopeaks. IAEA reference materials in 100 ml geometry(RGU-1, RGTh-1 and RGK-1) were used for efficiency calibration.The minimum detectable activity for each radionuclide was derived from the background radiation spectrum using a counting period of 20000 seconds, which was estimated to be 5.05 $Bqkg^{-1}$ for ^{238}U, 5.04 $Bqkg^{-1}$ for ^{232}Th ,and 17.3 $Bqkg^{-1}$ for ^{40}K.The ^{232}Th concentration was measured from 208 Tl (2614 keV) concentrations in the samples, the ^{238}U content was obtained from ^{214}Bi (1764 keV) concentrations, and the ^{40}K concentration was determined directly using its 1460 keV gamma line.

Activity measurement:

Using the following equation, the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K were determined:

$$A\,(Bqkg^{-1}) = \frac{N}{\varepsilon \times \beta \times M}$$

where, N is the net gamma counting rate (counts per second), ε is the detector efficiency of the specific gamma ray, β is the absolute transition probability of Gamma-decay and M is the mass of the sample (Kg) (Al-Zahrani 2017).

Radiation hazard indices

Radium equivalent activity (Ra_{eq}):

Distribution of ^{238}U, ^{232}Th and ^{40}K in environment is not uniform, so that with respect to exposure to radiation, the radioactivity has been defined in terms of radium equivalent activity (Ra_{eq}) in $Bqkg^{-1}$ to compare the specific activity of materials containing different amounts of ^{238}U, ^{232}Th and ^{40}K (Beretka & Mathew, 1985; UNSCEAR, 2000, Al-Zahrani 2017).

$$R_{eq}\,(Bqkg^{-1}) = A_{Ra} + 1.43\,A_{Th} + 0.077A_K$$

where, A_U, A_{Th} and A_K are the specific activities of ^{238}U, ^{232}Th and ^{40}K in $Bqkg^{-1}$, respectively. This calculation is based on the assumption that 1 $Bqkg^{-1}$ of ^{238}U, 0.7 $Bqkg^{-1}$ of ^{232}Th, and 13 $Bqkg^{-1}$ of ^{40}K result in the same gamma-ray dose. The maximum value of R_{eq} in building materials must be less than 370 Bqkg-1 in order to keep the external dosage below 1.5 $mSvy^{-1}$ (UNSCEAR, 2000, Al-Zahrani 2017).

Absorbed dose rate from the granite:

UNSCEAR has given the dose conversion factors for converting the activity concentrations of ^{238}U, ^{232}Th and ^{40}K into doses ($nGyh^{-1}$ per $Bqkg^{-1}$) as 0.462, 0.604 and 0.0417, respectively (Asaduzzaman et al., 2016; UNSCEAR, 2000, Al-Zahrani 2017).

$$D\,(nGy/h) \;=\; 0.462\,A_{Ra} + 0.604\,A_{Th} + 0.0417\,A_K$$

where D is the dose rate in $nGyh^{-1}$ and A_U, A_{Th} and A_K are the concentrations of uranium, thorium and potassium, respectively.

Annual effective dose rate (D_{eff}):

To estimate the annual effective dose the conversion factor (0.7 $SvGy^{-1}$) from absorbed dose rate in air in $nGyh^{-1}$to effective dose rate in $mSvy^{-1}$ is used with outdoor occupancy factor of 0.2 and indoor occupancy factor of 0.8. The annual effective dose equivalent was calculated using the following formula (UNSCEAR, 2000):

$$D_{eff(out)}(mSv/y) \;=\; D(nGyh^{-1}) \,\times\, 8760 \times 0.2 \times 0.7 \times 10^{-6} SvGy^{-1}$$

$$D_{eff(in)}(mSv/y) \;=\; D(nGyh^{-1}) \,\times\, 8760 \times 0.8 \times 0.7 \times 10^{-6} SvGy^{-1}$$

where D is the absorbed dose rate in air, D_{eff} is the annual effective dose.

External hazard index (H_{ex}):

The ultimate use of the measured activities in building materials is to estimate the radiation dose expected to be delivered externally if a building is constructed using these materials. The external hazard index (H_{ex}) is given by the following equation (Al-Zahrani 2017):

$$H_{ex} = \left(\frac{A_{Ra}}{370\ \mathrm{Bqkg^{-1}}}\right) + \left(\frac{A_{Th}}{259\ \mathrm{Bqkg^{-1}}}\right) + \left(\frac{A_{K}}{4810\ \mathrm{Bqkg^{-1}}}\right)$$

To limit the annual external gamma-ray dose to 1.5 $\mathrm{mSvy^{-1}}$, the external hazard index (H_{ex}) should be less than unity.

Internal hazard index (H_{in}):

The internal exposure to ^{222}Rn and its radioactive progeny is assessed by the internal hazard index (H_{in}), which is given by (Al-Zahrani 2017).

$$H_{in} = \left(\frac{A_{Ra}}{185\ \mathrm{Bqkg^{-1}}}\right) + \left(\frac{A_{Th}}{259\ \mathrm{Bqkg^{-1}}}\right) + \left(\frac{A_{K}}{4810\ \mathrm{Bqkg^{-1}}}\right)$$

For the safe usage of a material in home building, the maximum value of (H_{in}) should be smaller than unity.

RESULTS AND DISCUSSIONS:

The activity concentration of granite samples was estimated (Fig 1). ^{238}U range varies from $\leq$ 5.05 (MDA) $\mathrm{Bqkg^{-1}}$ to 103.89 $\mathrm{Bqkg^{-1}}$ with geometric mean of 10.04 $\mathrm{Bqkg^{-1}}$, which is lower than the world average value of 50 $\mathrm{Bqkg^{-1}}$; ^{232}Th varies from $\leq$ 5.04 (MDA) $\mathrm{Bqkg^{-1}}$ to 634.28 $\mathrm{Bqkg^{-1}}$ with geometric mean of 101.54 $\mathrm{Bqkg^{-1}}$ which is higher than the world average value of 50 $\mathrm{Bqkg^{-1}}$, ^{40}K concentration varies from $\leq$ 17.3 (MDA) $\mathrm{Bqkg^{-1}}$to 1243.59 $\mathrm{Bqkg^{-1}}$ with geometric mean of 222.3 $\mathrm{Bqkg^{-1}}$ which is lower than the world average value of 500 $\mathrm{Bqkg^{-1}}$ (UNSCEAR, 2008).

Table 1: ^{238}U, ^{232}Th and ^{40}K activity concentrations of the granite samples used as the interior building materials in Kanyakumari District

Sample code	Sample name	Color	Activity concentrations ($\mathrm{Bqkg^{-1}}$)		
			^{238}U	^{232}Th	^{40}K
G_1	Kuppam white	White with black	5.05	50.16	144.45

			(MDL)		
G_2	Galaxy black	Black	5.05 (MDL)	5.04 (MDL)	121.73
G_3	Honey blue	Honey blue	5.05 (MDL)	154.74	657.81
G_4	Mini green	Green	103.89	179.02	17.3 (MDL)
G_5	Pista green	Green	13.12	203.49	18.60
G_6	SK blue	Blue with grey and black	5.05 (MDL)	270.32	466.00
G_7	Multi	Pink with black and white	5.05 (MDL)	634.28	1243.59
G_8	Steel grey	Dark grey with black	90.90	496.03	1116.94
G_9	G20 black	Dark black	5.05 (MDL)	12.45	122.90
G_{10}	English catseye	Grey with white	5.05 (MDL)	77.19	995.42
Geometric mean			10.04	101.54	222.30
UNSCEAR 2008			50	50	500
Minimum			5.05	5.04	17.30
Maximum			103.89	634.28	1243.59
SD			38.71	209.16	480.81
Skewness			1.79	1.20	0.57
Kurtosis			1.56	0.67	-1.51
Mean			24.33	208.27	490.47
Median			5.05	166.88	305.23

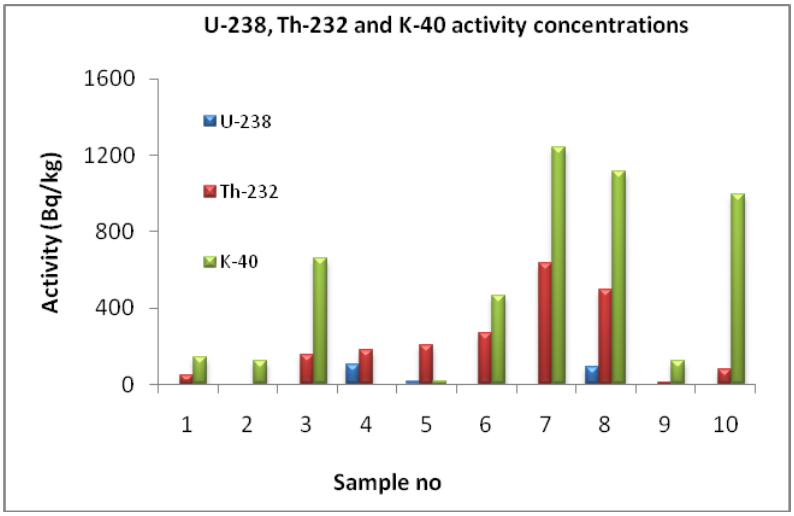
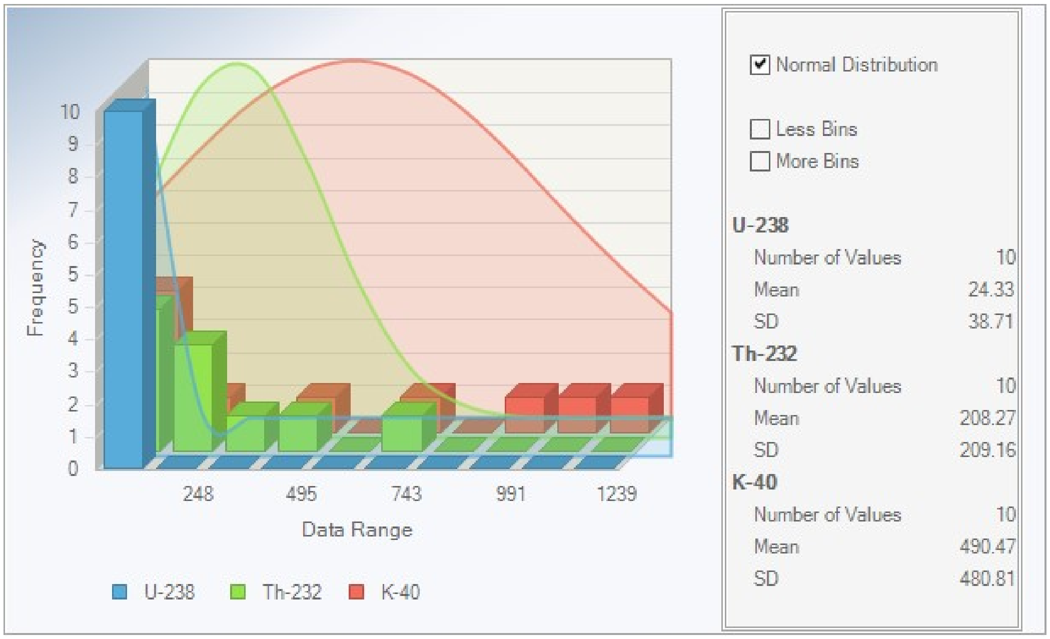


Fig 1. ^{238}U, ^{232}Th and ^{40}K Activity concentrations of granite samples.



☑ Normal Distribution

☐ Less Bins
☐ More Bins

U-238
Number of Values 10
Mean 24.33
SD 38.71
Th-232
Number of Values 10
Mean 208.27
SD 209.16
K-40
Number of Values 10
Mean 490.47
SD 480.81

Fig 2: Normal distribution of ^{238}U, ^{232}Th and ^{40}K activity (Bq/kg)

Table 2: The calculated radiological hazard parameters of the studied samples

Sample code	Radiological hazard indices					
	Radium equivalent activity Ra_{eq} ($Bqkg^{-1}$)	External Hazard Index (H_{ex})	Internal Hazard Index (H_{in})	Absorbed dose D ($nGyh^{-1}$)	Annual effective dose $D_{eff\ (0ut)}$ ($mSvy^{-1}$)	Annual effective dose $D_{eff\ (In)}$ ($mSvy^{-1}$)

G_1	87.9	0.24	0.25	38.65	0.19	0.47
G_2	21.63	0.06	0.07	10.45	0.05	0.01
G_3	276.98	0.75	0.76	123.23	0.6	0.15
G_4	361.23	0.98	1.26	156.85	0.77	0.19
G_5	305.54	0.83	0.86	129.75	0.64	0.16
G_6	427.49	1.16	1.17	185.04	0.91	0.23
G_7	1007.83	2.73	2.73	437.3	2.15	0.54
G_8	886.23	2.4	2.64	388.17	1.9	0.48
G_9	32.32	0.09	0.10	14.98	0.07	0.02
G_{10}	192.12	0.52	0.53	90.48	0.44	0.11
Geometric mean	206.03	0.56	0.60	91.93	0.45	0.14
Minimum	21.63	0.06	0.07	10.45	0.05	0.01
Maximum	1007.83	2.73	2.73	437.30	2.15	0.54
SD	339.13	0.92	0.96	147.10	0.72	0.19
Skewness	1.13	1.13	1.04	1.14	1.14	0.59
Kurtosis	0.35	0.34	0.00	0.35	0.36	-1.17
Mean	359.93	0.98	1.04	157.49	0.77	0.24
Median	291.26	0.79	0.81	126.49	0.62	0.18

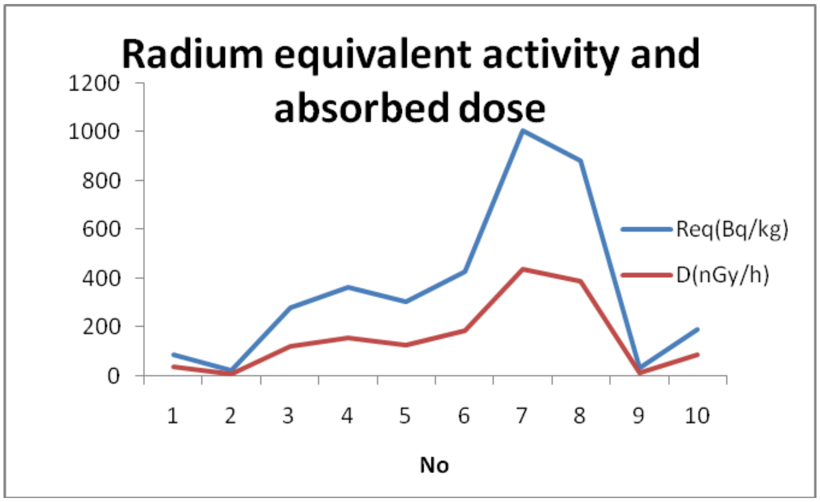


Fig 3: Radium equivalent activity and absorbed gamma dose

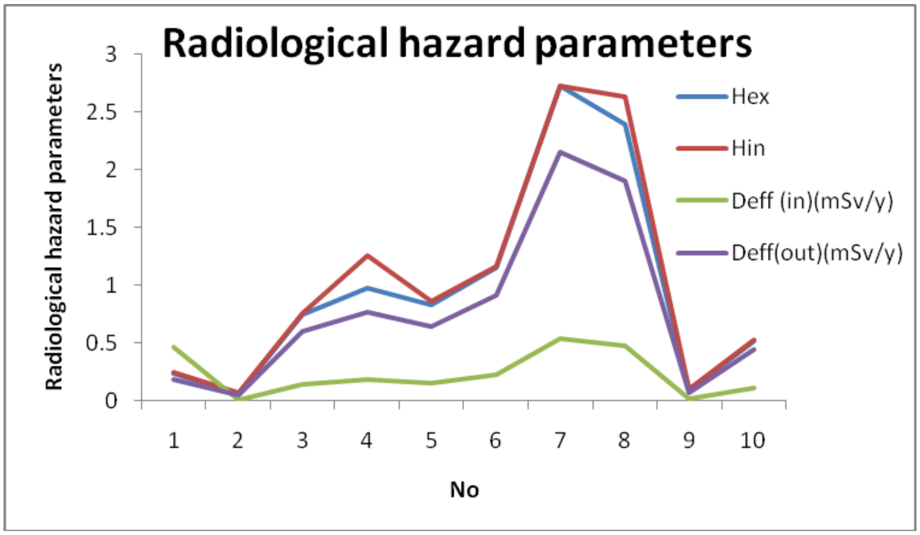


Fig 4: Radiological hazard parameters ((i) internal hazard index, (ii) external hazard index (iii) indoor annual effective dose (iv) and outdoor annual effective dose)

The radium equivalent activity (Ra_{eq}) values ranged from 21.63 to 1007.83 $Bqkg^{-1}$ with geometric mean of 206.03 $Bqkg^{-1}$. The radium equivalent activity is above the recommended limit of 370 $Bqkg^{-1}$ for three samples. The estimated absorbed dose rate ranged from 10.45 to 437.3 $nGyh^{-1}$. The calculated geometric mean of absorbed dose was 91.93 $nGyh^{-1}$ which is higher than the population-weighted value 84 $nGyh^{-1}$. The indoor annual effective dose values ranged from 0.05 to 2.15 $mSvy^{-1}$, with geometric mean of 0.45 $mSvy^{-1}$ and the outdoor annual effective dose values ranged from 0.01 to 0.54 $mSvy^{-1}$ with geometric mean of 0.14 $mSvy^{-1}$, which are within the annual effective dose equivalent limit of 1 $mSvy^{-1}$ (Cherry et. al., 2012). The geometric mean of internal and external hazard index is 0.56 and 0.60 respectively. The external hazard index values are higher than unity for three samples and the internal hazard index values are higher than unity for four samples respectively.

Conclusion:

Natural radioactive elements such as uranium, thorium, and potassium may be found in granite rock. Depending on the chemical makeup and the creation of the molten rock, some granite includes more of these than others. From the experimental work on natural radioactivity, the collected granite samples contain ^{238}U, ^{232}Th, and ^{40}K radionuclides with concentrations higher, comparable and lower than the set limits. The geometric mean of radiological hazard parameters such as internal and external hazard indices, radium equivalent activity, and indoor and outdoor annual effective dose are within the recommended safe limit. Since some samples have the radiological hazard parameters slightly higher than the recommended limit those granites can be used for exterior construction while others can be used for interior decorations in buildings. Also, these results can be used as reference values for distribution of natural radioactivity in granites while measuring the radioactive concentrations in other granite samples.

Reference:

Al-Zahrani, J. H. Estimation of natural radioactivity in local and imported polished granite used as building materials in Saudi Arabia. J. Radiat. Res. Appl. Sci. 10, 241–245 (2017) (DOI 10.1016/j.jrras.2017.05.001)

Asaduzzaman, K., Khandaker, M. U., Amin, Y. M., & Bradley, D. A. Natural Radioactivity, Radon Exhalation Rates and Indoor Radon Concentration of Some Granite Samples Used as Construction Material in Turkey. Radiation Protection Dosimetry, nct110, 2016

Beretka, J. & Matthew, P. J. Natural radioactivity of Australian building materials, industrial wastes and by-products. Health Phys. 48, 87–95 (1985) (DOI 10.1097/00004032-198501000-00007) (Pubmed:3967976)

Cherry, S. R. & Phelps, M. E. Physics in Nuclear Medicine. 4th ed, 2012

Harb, S., et al. Measurements of Naturally Occurring Radioactive Materials for some Granite Rocks samples in the Eastern Desert Egypt, IOSR. J. Appl. Phys., e-ISSN: 2278-4861 6, 40–46 (2014)

Padua, J. C. & Rose, M. R. B., Natural gamma radioactivity in the villages of Kanyakumari District, Tamil Nadu, India. Radiat. Prot. Dosimetry., 156, 42–48 (2013)

Rani, A. & Singh, S. Natural radioactivity levels in soil samples from some areas of Himachal Pradesh, India using γ-ray spectrometry. Atmos. Environ. 39, 6306–6314 (2005) (DOI 10.1016/j.atmosenv.2005.07.050)

UNSCEAR. 1 (United Nations, & Scientific Committee on the effects of Atomic Radiation (United Nations Publications, 2000). Sources and effects of ionizing radiation: sources

Kanyakumari Wikipedia (Wikimedia Foundation, June 11 2023), Available from: https://en.wikipedia.org/wiki/Kanyakumari_district. Wikipedia

UNSCEAR (United Nations, & Scientific Committee on the effects of Atomic Radiation (United Nations Publications, July 10–18 2008). Report of the United Nations Scientific Committee on the effects of atomic radiation: Fifty-sixth Session (No.46)