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Natural radioactivity in rock samples of Aravali hills in India

R. Kumari[1*], K. Kant[1], M. Garg[2]

[1]*Department of Physics, Aggarwal College Ballabgarh, Haryana, India*
[2]*Faculty of Humanities and Applied Sciences, YMCAUST, Faridabad, Haryana, India*

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Corresponding authors:
Dr. Raj Kumari,
Fax: *+ 91 129 2300769*
E-mail:
riyasisodiya007@gmail.com

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ABSTRACT

Background: Natural radionuclides in rock samples, primarily ^{238}U (uranium), ^{232}Th (thorium) and ^{40}K (potassium), are the main source of radiation exposure to workers and general public and their measurement is of paramount importance from radiation protection point of view. ***Materials and Methods***: Gamma spectrometry using HPGe detector was used to measure the radioactivity and assess the radiological hazards and annual effective dose. The data is statistically significant. ***Results:*** In the studied rock samples, the mean activity of ^{238}U, ^{232}Th, ^{40}K is 12.15 ± 1.68 Bq/kg, 45.17 ± 6.92 Bq/kg and 639.24 ± 115.86 Bq/kg, respectively. The mean values of radium equivalent activity, radiological hazards external hazard index (Hex), Internal hazard index (Hin), Gamma index (Iγ) and Alpha index (Iα) are 125.96 ± 14.94 Bq/kg, 0.34 ± 0.04, 0.37 ± 0.04, 0.48 ± 0.06 and 0.06 ± 0.00 respectively. The mean values of absorbed dose rate, indoor and outdoor annual effective dose of gamma radiation are 59.55 ± 7.16 nGy/h, 0.29 ± 0.04 mSv and 0.07 ± 0.00 respectively. A strong positive correlation was observed between the radium equivalent activity and the absorbed dose (correlation coefficient R2 = 0.99). ***Conclusion:*** The present investigations showed that the inborn radioactivity is within the world and Indian average value. And the findings indicate that the studied samples do not pose any significant radiation hazard to the public in large and the workers in particular and when used as construction material also.

Keywords: *Radiological hazards, dose, rock, radium equivalent activity, gamma spectrometry*

INTRODUCTION

Human exposure from radiation is from womb to tomb due to both terrestrial and extra-terrestrial sources and the deleterious health effects and the symptoms induced include direct chromosomal transformation, indirect free-radical formation, cancer induction, bone necrosis, etc [1]. Terrestrial radiation is due to the radionuclides present in different amounts in rocks, soils, building materials Natural radioactivity is wide spread in the earth's environment and there are several radioactive elements in this category, such as ^{238}U (uranium), ^{232}Th (thorium) and ^{40}K (potassium), also termed as primordial radioisotopes, for they were present when the earth was formed and it exists in various geological formations in soils, rocks, plants, water and air [2-4]. Natural radioactivity in rocks and soils comes from ^{238}U and ^{232}Th series and natural ^{40}K. Uranium occurs in minerals such as pitchblende, uraninite etc. It is also found in phosphate rocks, lignite and monazite sands, whereas ^{40}K is present in mineral waters and brines, and in various minerals such as carnallite, feldspar, saltpeter, greens and sylvite [5-7]. Study of radionuclide distribution and natural radioactivity levels is usually done in order to gain information about the present levels of radioactive pollutants

discharged to the environment itself or in the living creatures and is important for assessing the effects of radiation exposure to human beings [8]. Thus measurement of radioactivity in rock samples is very important from radiation protection point of view, and there has been extensive surveys in many countries for estim ating natural radiation exposure since it provides useful estimation of possible radiological hazards to human health [9-12]. Also, this type of work provides baseline data and helps in establishing baseline values for comparison with future measurements. In the light of above mentioned facts, natural radioactivity in rock samples of Aravali hills in India has been carried out in the present investigation.

MATERIALS AND METHODS

Study Area

The Geographical location of the study area is shown in the figure 1. Aravali Range (literally meaning 'line of peaks'), 28.44° N and 77.28° E is the highest peak in the is the range of oldest plateau mountains in western India running approximately 692 km in a northeastern direction across Indian states of Gujarat, Rajasthan, Haryana and Delhi and has a popular tourist attraction in India at various places. It dates back to a pre-Indian sub continental collision with the mainland Eurasian Plate. The highest peak is Guru Shikhar in Mount Abu in Rajasthan. Rising to 5650 feet (1722 meters), it lies near the south-western extremity of the range, close to the border with Gujarat state. Old Fold Mountains are characterized by having stopped growing higher due to the cessation of upward thrust caused by the stopping of movement of the tectonic plates in the Earth's crust below them. In ancient times they were extremely high but since have worn down almost completely by millions of years of weathering. Mining of copper and other metals in the Aravali range dates back to at least 5th century BC, based on carbon dating.

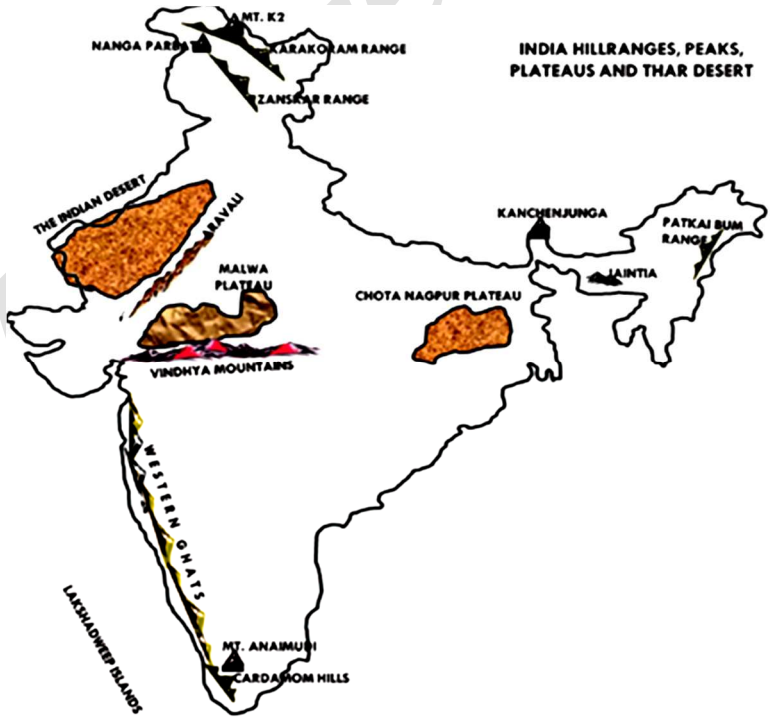

Figure 1. Geographical location of the study area (13).

Sample Collection and Preparation

In the present investigation, a total of 20 rock samples were collected from different places and altitudes of Aravali Range in Delhi, Haryana, Rajasthan and Gujarat were analyzed for the activity concentration of the natural radionuclides namely ^{238}U, ^{232}Th and ^{40}K using gamma spectrometry. All the samples were processed following the standard procedures as per International Atomic Energy Agency (IAEA) guidelines. After collection, samples were crushed into fine powder by using mortar and pestle. Fine quality of the sample was obtained using scientific sieve of 250 micron-mesh size and particles of size ≤ 250 microns were obtained. Before measurement, the samples are dried in an oven at about 1100C for 24 h. Each sample is packed and sealed in an airtight PVC container and kept for about 4-week period to allow radioactive equilibrium among the radon (^{222}Rn), thoron (^{220}Rn) and their short-lived decay products [14]. On an average 300–350 g of sample in powder form was taken for each material.

Measurement technique

The activity of samples was counted Using HPGe detector of high-resolution gamma spectrometry system. The detector is a co-axial n -type high purity germanium detector (Make EG&G, ORTEC, Oak Ridge, USA). The detector has a resolution of 2.0 keV at 1332 keV and relative efficiency of 20%. The output of the detector is analyzed using a 4K ADC system connected to PC, the spectrum is analyzed using the locally developed software "CANDLE (Collection and Analysis of Nuclear Data using Linux nEtwork)". The detector is shielded using 4'' lead on all sides to reduce the background level of the system [15]. For calibration of the low background counting system, a secondary standard was obtained, calibrated with the primary standard obtained from the International Atomic Energy Agency. Efficiency values are plotted against energy for particular geometry and are fitted by least squares method to an empirical relation that takes care of the nature of efficiency curve for the HPGe detector. The samples were counted for a period of 72000 seconds and the spectra were analyzed of the photo peak of uranium, thorium daughter products and ^{40}K. The net count rate under the most prominent photo peaks of radium and thorium daughter peaks are calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclides is calculated from the background subtracted area prominent gamma ray energies. The concentration of ^{238}U was determined using a photon peak of 609 keV (46.1%) from ^{214}Bi. The 186 keV photon peak of ^{238}Ra was not used because of interfering peak of ^{235}U, with energy of 185.7 keV. ^{232}Th concentration was determined using the gamma transitions of 583 keV (86%) from ^{208}Tl [16]. ^{40}K concentration was determined using the gamma transition of 1461 keV (10.7%).

RESULTS AND DISCUSSION

Measuring Activity Concentration of Uranium, Thorium and Potassium

The concentrations of Uranium, Thorium and Potassium were calculated using the equation (1):

$$\text{Activity (Bq)} = (\text{CPS} \times 100 \times 100/ \text{B.I.} \times E_{ff}) \pm (\text{CPS}_{error} \times 100 \times 100/ \text{B.I.} \times E_{ff}) \quad (1)$$

Where, CPS - Net count rate per second

B.I. - Branching Intensity, and Eff - Efficiency of the detector.

The concentration of the radionuclides, ^{238}U, ^{232}Th and ^{40}K in the rock samples studied in the present investigation are given in table 1. From table 1, it is clear that the activity concentration for ^{238}U, ^{232}Th and ^{40}K varies from 3.45 Bq/kg to 32.18 Bq/kg with a mean value of 12.15 ± 1.68 Bq/kg, 10.17 Bq/kg to 132.45 Bq/kg with a mean value of 45.17 ± 6.92 Bq/kg and 58.01 Bq/kg to 1822.78 Bq/kg with a mean value of 639.24 ± 115.86 Bq/kg respectively in various rock samples studied in the present work. The mean values of concentration of the radionuclides ^{238}U, ^{232}Th and ^{40}K are within the internationally recommended limit (world average is of 33, 45 and 420 Bq/kg, respectively)

and (Indian average is of 28.67, 63.83 and 400 Bq/kg respectively) except ^{40}K for which it is slightly higher [8]. The activity concentration of ^{40}K was higher in the samples than that of radium and thorium. This was probably due to the higher concentrations of ^{40}K, a micronutrient in the soil whose mobilization and subsequent migration might be the reason for these values [17-18].

Radium Equivalent Activity

The widely used radiation hazard index Ra_{eq} is called the radium equivalent activity. The radium equivalent activity is a weight sum of activities of the three natural radionuclides ^{238}U, ^{232}Th and ^{40}K based on the estimation that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the same gamma ray dose rate [19].

The radium equivalent activity was calculated using the equation (2):

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_k \quad (2)$$

Where, C_u, C_{Th} and C_k are the specific activities of ^{238}U, ^{232}Th and ^{40}K in Bq/ kg respectively.

Figure 1. Geographical location of the study area (13).

Sample code	Activity concentration (Bq/kg)			Radium equivalent activity (Ra_{eq}) (Bq/ kg)
	^{238}U	^{232}Th	^{40}K	
ARS-1	3.45	17.29	58.01	32.64
ARS -2	5.78	45.32	123.33	80.08
ARS -3	7.58	67.56	89.87	111.11
ARS -4	4.24	34.41	243.47	72.19
ARS -5	6.89	56.86	555.08	130.94
ARS -6	5.56	43.23	857.58	133.41
ARS -7	16.12	12.34	933.20	105.62
ARS -8	13.78	18.18	1547.49	158.93
ARS -9	19.21	56.25	1822.78	240.00
ARS -10	22.56	72.89	687.56	179.73
ARS -11	11.76	32.56	876.32	125.80
ARS -12	8.24	28.32	432.43	82.03
ARS -13	9.67	10.17	257.78	44.06
ARS -14	6.53	16.12	98.17	37.14
ARS -15	7.29	24.27	376.09	70.96
ARS -16	23.31	53.47	521.23	139.91
ARS -17	32.18	42.21	289.18	114.81
ARS -18	17.33	37.34	1088.28	154.52
ARS -19	11.59	102.18	1470.56	270.94
ARS -20	9.83	132.45	456.37	234.37
Range	3.45-32.18	10.17-132.45	58.01-1822.78	32.64-270.94
Mean ± SE	12.15±1.68	45.17±6.92	639.24±115.86	125.96±14.94

Where, SE is the Standard Error which is $\sigma/\sqrt{N}$, where σ is the standard deviation and N is the total no. of observations.

MADL (Minimum Activity Detection Limit) = 2Bq/kg, 2Bq/kg and 4Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K respectively.

The radium equivalent activity varied from 32.64 Bq/kg to 270.94 Bq/kg with a mean value of 125.96 ± 14.94 Bq/kg in the rock samples used in the present investigation as given in table 1. The values obtained are less than the recommended maximum value of 370 Bq/kg [8, 20-21].

From the results obtained is clear that

the concentration of Radium, Thorium and Potassium and the radium equivalent activity varied considerably in different samples.

Assessment of radiological hazards

Radiological hazards in terms of external hazard index (Hex), Internal hazard index (Hin), Gamma index (Iγ) and Alpha index (Iα) have been calculated using equations 3, 4, 5 and 6 respectively [19, 22-25].

$$Hex = ARa / 370 + ATh / 259 + AK / 4810 \quad (3)$$

$$Hin = ARa / 185 + ATh / 259 + AK / 4810 \quad (4)$$

$$I\gamma = ARa / 300 + ATh / 200 + AK / 3000 \quad (5)$$

$$I\alpha = ARa / 300 \quad (6)$$

To limit the external gamma radiation dose from materials below 1.5 mGy/y, the external hazard index, Hex should obey the following relation Hex≤1 [26-27].

The radiological hazards in terms of external hazard index (Hex), internal hazard index (Hin), Gamma index (Iγ) and Alpha index (Iα) for the samples calculated using equations 3, 4, 5 and 6 are given in table 2. Our findings are in good agreement with the findings of other researchers reported in the literature [28-33]. The Hex and Hin for the studied samples are less than unity and therefore these samples are safe from health point of view. Value of Iγ≤2 corresponds to a dose rate criterion of 0.3 mSv/y, whereas 2≤ Iγ≤6 corresponds to a criterion of 1 mSv/y [23, 34]. All the current 'Iγ' values of the studied samples follow the criterion (Iγ≤2) therefore it may be concluded that the samples are safe from health and hygiene point of view and don't pose any significant health hazards to the indigenous human population. The recommended limit for concentration of ^{226}Ra is 200 Bq/kg, for which Iα = 1. The observed values are less than unity except one sample showing that the most of the samples are safe from the health and hygiene point of view and don't pose any environmental radiation hazards.

Figure 1. Geographical location of the study area [13].

Sample code	H_{ex}	H_{in}	I_γ	I_α
ARS-1	0.09	0.10	0.12	0.02
ARS -2	0.22	0.23	0.29	0.03
ARS -3	0.30	0.32	0.39	0.04
ARS -4	0.19	0.21	0.27	0.02
ARS -5	0.35	0.37	0.49	0.03
ARS -6	0.36	0.38	0.52	0.03
ARS -7	0.29	0.33	0.43	0.08
ARS -8	0.43	0.47	0.65	0.07
ARS -9	0.65	0.70	0.95	0.10
ARS -10	0.49	0.55	0.67	0.11
ARS -11	0.34	0.37	0.49	0.06
ARS -12	0.22	0.24	0.31	0.04
ARS -13	0.12	0.15	0.17	0.05
ARS -14	0.10	0.12	0.14	0.03
ARS -15	0.19	0.21	0.27	0.04
ARS -16	0.38	0.44	0.52	0.12
ARS -17	0.31	0.40	0.41	0.16
ARS -18	0.42	0.46	0.61	0.09
ARS -19	0.73	0.76	1.04	0.06
ARS -20	0.63	0.66	0.85	0.05
Range	0.09-0.73	0.10-0.76	0.12-1.04	0.02-0.16
Mean ± SE	0.34±0.04	0.37±00.4	0.48±0.06	0.06±0.00

Where, SE is the Standard Error which is σ/√N, where σ is the standard deviation and N is the total no. of observations.

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Estimation of absorbed and annual effective dose

The measured activity concentrations of 238U, 232Th and 40K were converted into doses (nGyh-1 per Bqkg-1) and the total absorbed gamma dose rate in air at one meter above the ground level was calculated using the equation 7, [8].

D(nGyh-1)=(0.462CU+0.604CTh+0.0417CK) (7)

Where, CU, CTh and CK are the activity concentrations (Bq/kg) of uranium, thorium and potassium; 0.462, 0.604 and 0.0417 are the conversion factors for uranium, thorium and potassium, respectively in the samples. To estimate annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor.

Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv/Gy, which is used to convert the absorbed dose rate to annual effective dose with an outdoor occupancy of 20% and 80% for indoors [8,33].

The indoor and outdoor annual effective doses are determined using the equations 8 and 9 respectively.

Indoor (mSv) = (Absorbed Dose) nGyh-1 × 8760h × 0.8 × 0.7 SvGy-1 × 10-6 (8)

Outdoor (mSv) = (Absorbed Dose) nGyh-1 × 8760h × 0.2 × 0.7 SvGy-1 × 10-6 (9)

The radiation absorbed dose from rock samples calculated using equations 7, 8 and 9 are given in table 3.

Figure 2. Correlation between radium equivalent activity and absorbed dose.

Sample code	Absorbed dose rate ($nGyh^{-1}$)	Annual effective dose (mSv)	
		Indoor	Outdoor
ARS-1	14.46	0.07	0.02
ARS -2	35.19	0.17	0.04
ARS -3	48.06	0.24	0.06
ARS -4	32.90	0.16	0.04
ARS -5	60.67	0.30	0.07
ARS -6	64.44	0.32	0.08
ARS -7	53.82	0.26	0.07
ARS -8	81.88	0.40	0.10
ARS -9	118.86	0.58	0.15
ARS -10	83.12	0.41	0.10
ARS -11	61.64	0.30	0.08
ARS -12	38.94	0.19	0.05
ARS -13	21.36	0.10	0.03
ARS -14	16.85	0.08	0.02
ARS -15	33.71	0.17	0.04
ARS -16	64.80	0.32	0.08
ARS -17	52.42	0.26	0.06
ARS -18	75.94	0.37	0.09
ARS -19	128.39	0.63	0.16
ARS -20	103.57	0.51	0.13
Range	14.46-128.39	0.07-0.63	0.02-0.16
Mean ± SE	59.55±7.16	0.04±0.29	0.07±0.00

Where: SE is the Standard Error which is σ/√N, where σ is the standard deviation and N is the total no. of observations

From table 3, it is clear that the absorbed dose rate varies from 14.46 nGyh-1 to 128.39 nGyh-1 with a mean value of 59.55 ± 7.16 nGyh-1 which is very close to the World (57 nGy h–1) and Indian (56 nGy h–1) average values [8]. The annual effective dose calculated using equations 8 and 9 varies from 0.07 mSv to 0.63 mSv with a mean value of 0.29 ± 0.04 mSv and 0.02 mSv to 0.16 mSv with a mean value of 0.07 ± 0.00 mSv respectively in various rock samples studied in the present work. In all the samples, the indoor annual effective dose was less than the recommended safety limit of 1 mSy/y as recommended for general public [35]. A positive correlation with R2 = 0.99 was observed between the radium equivalent activity and the absorbed dose as shown in figure 2.

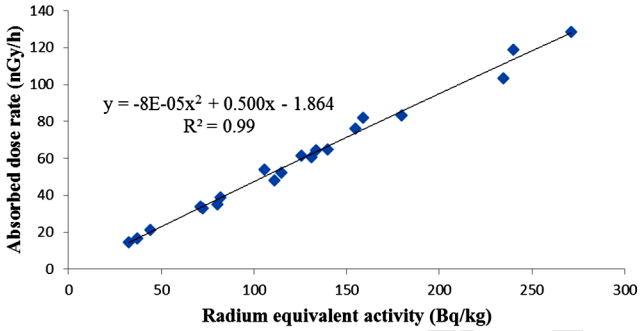

Figure 2. Correlation between radium equivalent activity and absorbed dose.

CONCLUSION

The present investigations showed that the inborn radioactivity measured using gamma-ray spectrometry system in rock samples collected from the Aravali hills are within the world and Indian average value. However, the increment of radionuclide concentrations varied amongst samples collected from various sites. These materials satisfy the universal standards limiting the radioactivity within the safe limits of 1000, 1000 and 4000 Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively. The mean value of all the calculated radiological effects (Radiological Hazards) is within the recommended safety limit [8]. The annual effective dose found in all samples also lies within the safe limit of 1 mSv/y [35]. This indicates that the rock samples of Aravali hills do not pose any significant radiation hazard to the public in large and the workers in particular and when used as construction material also.

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Conflicts of interest*: *Declared none.

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