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Radiometric and radon exhalation rate analysis of Gahirat marble, Chitral Khyber Pakhtunkhwa, Pakistan

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ABSTRACT

Background: Geological materials usually contain trace amounts of radioactive materials and may serve as a natural source of background radiation exposure to the general public. This study presents results of radiometric and radon exhalation rate (RER) analysis of 28, export quality marble samples taken from various quarries of Gahirat Chitral area. ***Materials and Methods:*** The marble specimens were investigated using gamma spectroscopy by HPGe detector. Samples were also analyzed for radon exhalation rate using closed CAN technique. ***Results and Discussion:*** The mean values of ^{226}Ra, ^{232}Th and ^{40}K were found as 31.598± 0.989, 1.529± 0.308 and 5.273± 1.593Bqkg^{-1} respectively. Average value of Ra_{eq} was estimated as 34.19±1.55 Bqkg^{-1}. Radiation risk parameters viz. internal (H_{in}), external (H_{ex}), alpha (I_α) and gamma (I_γ) hazard indices were estimated and found less than unity value. The values for effective indoor ($\dot{D}_{in}$) and outdoor gamma dose rates ($\dot{D}_{out}$) due to the contents of primordial radionuclides were also estimated. The contribution of radon towards radiation exposure was assessed by estimating RER, which was found in the range (1.01±0.07 to 9.67±0.27) ×10^{-2} Bqm^{-2} h^{-1} with mean value of (5.84±0.002) ×10^{-2} Bqm^{-2} h^{-1}. ***Conclusion:*** The surface radon exhalation rate values estimated in the current study were found smaller than as reported for many other countries. The results obtained for gamma emitting radionuclides have been compared with the data available in the literature. Measurements shows that marble samples investigated have low concentrations of radionuclides and uses of marbles in dwellings do not pose significant threat to the inhabitants.

INTRODUCTION

Geological stones contain trace amounts of radionuclide's that may pose potential health threat to human beings in case of sustained exposure. Natural rock materials quarried for the purpose of obtaining blocks, tiles or slabs and their use for interior, exterior decoration and construction of buildings may serve as a source for radiation exposure [1-3]. Naturally occurring radionuclides viz. ^{238}U, ^{234}Th and ^{40}K are present in various rock formations, alluvium, vegetation cover, rivers and marine water [4]. Beside presence of naturally occurring radionuclides, anthropogenic radionuclides viz. ^{137}Cs etc. are also found in the environment. Existence of anthropogenic radionuclides in the environment is subject to either nuclear reactor accidents or atomic bomb testing.

The occurrence of the radioactive isotopes in stones can affect directly to the society living in the closed buildings environment. The existence of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in the stones are continuous sources of radiation including radon gas (^{222}Rn) and its decaying products. The building stones with higher assemblages of radionuclide concentrations may raise the levels of radiations within the indoor and outdoor environments and thus making the environment vulnerable for the inhabitants [5-7]. In Earth's crust, the standard global concentration levels of ^{232}Th, ^{226}Ra and ^{40}K are about 50, 50 and 500 Bqkg^{-1}, respectively [8-9]. Construction materials with higher levels of ^{232}Th, ^{226}Ra and ^{40}K are not only sources of external gamma ray radiations but are also the cause of internal radon and its decaying products exposure to the public [10].

The ^{222}Rn gas within indoor environments can be inhaled by inhabitants followed by the emission of alpha particles and decay products that may deposit their energy to the tissues and ultimately leading to the lung cancer [11-12].

Keeping in view the importance of the subject, many researchers across the globe have conducted radiometric and radon measurement surveys to get an estimate of natural radionuclides and radon exhalation rate in rocks, building materials, water and environmental samples [13-18]. Researchers have investigated environmental samples for primordial and anthropogenic radionuclides. They have also investigated the impact of seasonal variations, building age and age dependent risk factors associated with the sustained exposure to radioactivity arising from radionuclides [19-20].

Awareness about the source of radioactivity in dimension stones is important for the general public. All dimension stones, consisting of marbles, have variety of radionuclides as their constituent's elements, and the concentration of these natural radionuclides is high in these samples when compared to the rocks of mantle and Earth's crust [21]. In Pakistan, marble is used in majority of the houses as decorative stones. And keeping in view the quality of locally produced marbles it is also exported to other countries and is a source of revenue generation. Marble resources of Pakistan are mostly distributed over three provinces, viz. Khyber Pukhtunkhawa (KP), Balochistan and Punjab. Along with Gadanai, Mohmand Agency, Risalpur, Loralai, Chitral have been declared as marble cities. Marbles produced from these reserves are not only used within the country, as decorative stones, but also exported to other countries.

The primary purpose of the current study is to get an assessment for the contents of primordial radionuclides viz. ^{232}Th, ^{226}Ra, ^{40}K and estimation of radon exhalation rate in the Gahirat marble specimens. Health hazards associated with the presence of radionuclide in marble samples have also been calculated and assessed for the level of health threat to the inhabitants.

MATERIALS AND METHODS

Geology of the Area

The study area lies in district Chitral, Northern Pakistan. Geologically, the Chitral area is characterized through the occurrence of thick sedimentary and metamorphic succession comprising carbonate to arenaceous rocks of Paleozoic and Mesozoic Eras. The stratigraphy of the area represents the sediments from continental shelf to flysh basin of Neo-Tethys Ocean. The flysh sediments in north of Chitral constitute the Karakoram and Pamir Block and deposits of Kohistan Magmatic arc in the south. The geological map (see figure 1) shows the rock unit and sample location of the Chitral area. The rock units exposed in the area are ranging from Devonian to Cretaceous age. These rocks consist of low to medium grade metamorphic rocks along with the intrusion of granitic rocks. The marble is interbedded with calcareous mica schist and contains about 10 ft thick quartz vein [22-25].The estimated reserves of marble in the KP province is approximately 3.0 billion tonnes. About more than 1000 million tonnes of marble deposits occurred in Chitral [26]. The locality of Gahirat Marble is 3.2 km east of Gahirat village exposed along the bank of Chitral River.

Sample Collection and Treatment

The marble is a metamorphic rock, and extensively used as a building and decorative stones. Twenty Eight marble samples were collected from various quarries of Gahirat near Chitral Valley for radiometric investigation and radiological hazard assessment. Pretreatment of the rock specimens was carried out before their spectroscopic characterization. For the purpose of particle size characterization (PSC) a 40 –mesh sieve was used to mesh the samples and converted into the powdered form.

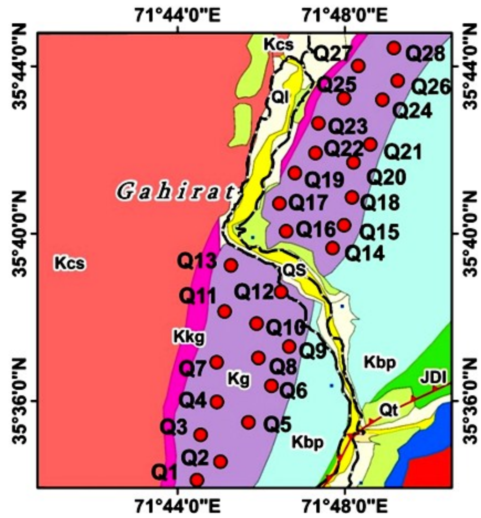

Figure 1. Geology and location map of the area; Queries Location: Q1-Q3 (Chinar), Q4-Q6 (Goja Lasht), Q7-Q9 (Khairabad), Q10-Q12 (Kesu), Q13-Q14 (Gang), Q15-Q20 (Gahirat), Q21-Q23 (Gumbaz), Q24-Q26 (Ayun), Q27-Q28 (Chitral). **Source Line:** Abbreviation:Q= Querry: Kg= Gahirat Marble: Kkg = Koghaz Foramtion: Kcs= Chitral Slates: Ql=Alluvial Deposits, JDl= Lawi Formation: Qt= Terrace Deposits: Qs= Stream channel deposits.

All marble samples were heated in an oven, while keeping its temperature at 110 ^{0}C, for the time period of four hrs in order to eliminate the content of moisture, if present any. These rock samples, each having a mass of 200g, were then placed into plastic Merinelli beakers [27]. The Merinelli beakers were perfectly sealed to retain the radon gas originating from the powdered samples enclosed in the beaker.

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The tightly sealed beakers were left for 28 days to allow the daughter nuclide of ^{238}U and ^{232}Th decay series to achieve secular radioactive equilibrium. Using gamma ray spectroscopy, the concentrations for primordial radionuclides were calculated for all the samples [28].

Statistical analysis

Data analysis, for the results of all samples under investigation, was carried out using Minitab® software, product version was Minitab® 20.4 and application run requirement was 64 bit machines (Minitab Inc. USA). For two set of data viz. ^{226}Ra and ^{222}Rn, we have used 2 sample t-tests for statistical analysis and for the purpose of obtaining p-value. Details are mentioned in discussion section.

Gamma spectrometric analysis

The samples of Gahirat marble were analyzed by gamma spectrometric methods [29]. High Purity Germanium (HPGe) detector with P-type closed-end coaxial geometry was used as a measuring system. The HPGe detector has relative efficiency of 30% as compared with thallium-activated sodium iodide detector (NaI(Tl) detector). The energy resolution of the detector was 2.0 keV (FWHM), for 'γ-ray' photon of energy 1.332 MeV, originating from a radioactive source of ^{60}Co. The effects of background radiations were minimized by placing the detector within 15 cm dense lead shield closed environment containing with the internal coating of 3 mm copper plate and 4 mm thick tin coatings. For the purpose of calibration of the γ- ray spectrometer, IAEA soil-326 was used and in order to confirm the reliability of counting efficiency, IAEA soil-375 was used as reference material. Each sample was counted for 6500 s and γ-spectrum obtained from multichannel analyzer (MCA) was analyzed through Genie 2000 version 2.1 (Canberra, USA). Gamma lines with energies 351.99, 911.07, 1460.75 and 661.62 keV, were respectively used to find activity contents of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs.

Empty Marinelli beakers were used for the determination of background contributions at the same pattern as the procedure was adopted for the other investigated samples. The activity concentrations were determined by the measurement of the background. Each sample was crushed into the powder form while keeping the size of particles less than 1 mm. 200 gm of each sample were placed into standard Marinelli beaker and the radioactive contents of ^{226}Ra, ^{232}Th and ^{40}K in the marble specimen were calculated using equation (1) [30].

$$A = \frac{(CS)Net}{\gamma I \times Ef\gamma \times M\,(kg)} \qquad (1)$$

Where, 'A' stands for activity contents, measured in the unit of $Bqkg^{-1}$, *'(CS)Net'* are net counts per second which is equivalent to {(cps) sample - (cps) background}, γI is the absolute intensity of the γ-ray, 'Efγ' is the detector efficiency and M(kg) is sample mass in kilograms.

The lower limit of detection (LLD) was estimated, for all radionuclides under investigation using the equation (2) [31],

$$LLD = \frac{4.66\,(Continum\ Counts + Background\ Peak\ Counts)^{1/2}}{Sample\ Mass\,(kg) \times Efficiency \times Live\ time\ (s) \times Yield} \qquad (2)$$

Where, 'LLD' is measured in Bq kg^{-1} and the number 4.66 appear as statistical coverage factor (SCF). LLD for the cesium, thorium, radium, and potassium radionuclides were estimated as 1.35, 2.25, 3.60, and 6.70 Bq Kg^{-1} respectively.

Radiological Hazards Assessment

Measurement of Radium Equivalent Activity (Ra_{eq})

To evaluate the hazards related with the radiation originating from the decorative stones, a parameter called radium equivalent activity (Ra_{eq}) has been calculated. Calculations were based upon the assumption that progenies of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their originators. The estimation of Ra_{eq} was carried out by the Equation (3) [32].

$$Ra_{eq} = \left(A_{Ra} + \frac{370}{259}A_{Th} + \frac{370}{4810}A_k\right) \qquad (3)$$

It is assumed that the compliance of the criterion $Ra_{eq} \leq 370\frac{Bq}{Kg}$ must be achieved to control the external dose D≤1.5mG/y [32].

The radiation hazard indices, external (H_{ex}) and internal (H_{in}), have been evaluated by the Equations 4 and 5 respectively [33].

$$H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \qquad (4)$$

While following criterion should be met i.e., H_{ex} £ 1, and Ra_{eq} £ 370 Bq kg^{-1}, for maintaining dose D £ 1.5 mGy y^{-1}.

$$H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \qquad (5)$$

For keeping D £ 1.5 mGy y^{-1}, H_{in} must be less than unity and Ra_{eq} £ 370 Bq kg^{-1}.

Estimation of gamma dose rate ($\dot{D}$)

For indoor air, the absorbed gamma dose rate, $\dot{D}_{in}$ (nGy h^{-1}), arising from ^{226}Ra, ^{232}Th and ^{40}K radionuclide's exposures was estimated using equation (6) [34].

$$\dot{D}_{in} = (0.462 \times A_{Ra}) + (0.604 \times A_{Th}) + (0.0417 \times A_K) \qquad (6)$$

$\dot{D}_{in}$ was calculated with the assumption that all the progenies of radium and thorium radionuclide's are in radioactive equilibrium with their precursors.

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For the outdoor environment, the external absorbed dose rate ($\dot{D}_{out}$), coming from the natural occurrence of radionuclides in the samples, was estimated by the equation (7) [34].

$$\dot{D}_{out}(\text{nGy h}^{-1})=0.427A_{Ra}+0.662A_{Th}+0.0432A_K(\text{nGy h}^{-1}) \quad (7)$$

UNSCEAR 2000 reports that $\dot{D}_{in}$ is greater than the $\dot{D}_{out}$ by the factor 1.4. Equation (8) has been used for the estimation of the indoor absorbed dose rate ($\dot{D}_{in}$).

$$\dot{D}_{in} = 1.4\ \dot{D}_{out} \quad (8)$$

Determination of Annual Effective Dose Equivalent (E, mSv y-1)

Annual Effective Dose Equivalent, *E (mSv y⁻¹)* received by the public due to exposure of radiations coming from the Gahirat Marble sample, was estimated using equation (9) [35].

$$E(\text{mSv y}^{-1})=\dot{D}(\text{nGy h}^{-1})\times 8766\ \text{hrs}\times 80\%\times 0.7\ \text{SvGy/y (Conversion factor)}\times 10^{-6} \quad (9)$$

Gamma Index (I_γ)

Mathematical expression mentioned in the Equation (10) was used for the estimation of gamma activity index [34].

$$I_{(\gamma)} = \left(\frac{A_{Ra}}{150}+\frac{A_{Th}}{100}+\frac{A_K}{1500}\right) \quad (10)$$

The 'I_γ' is associated with the cause of excess external radiation triggered by superficial material and the value of annual dose rate. Gamma index values i.e., $I\gamma \leq 2$, is equivalent to a dose rate criterion of 0.30, and for gamma index value in the range of $2 < I\gamma \leq 6$ is equivalent to dose rate criterion of 1 and similarly for $I\gamma \leq 0.5$ the equivalent dose rate criterion is 0.3 mSv y^{-1} [36]. The suitability or selection of building materials can be made based upon the gamma dose criterion value. In order to avoid exposure from higher values of dose rates, higher than the recommended value of 1 mSv y^{-1}, only those building materials should be used with I_γ values less than 6 [37].

Estimation for Alpha index (I_α)

The 'I_α' was calculated by equation (11) [38]. 'I_α' accounts for the excess radiation exposure, due to alpha emitters present in building stones resulting from inhalation.

Where A_{Ra}(Bqkg^{-1}) is the activity produced by ^{226}Ra.

$$I_\alpha = \frac{A_{Ra}}{200}(\text{Bqkg}^{-1}) \quad (11)$$

Radon activity concentration and radon exhalation rate

'CAN' technique [33] was used to get an estimate for radon exhalation rate from twenty eight marble samples (See figure 2). The samples were crushed and dried, to remove moisture, while placed in the oven for four hours at 110°C. Then samples, each weighing 200g, were put in plastic CANS having volume 8.55×10^3 cm^3. Polyallyldiglycol carbonate (CR-39) polymer plastic sheets, with thickness of 1 mm and 1×1 cm^2 area, were attached at the upper part of National Radiological Protection Board (NRPB) dosimeters. CANs were made completely airtight and detectors were permitted to get exposed with the radon coming from samples for 28 days. Four weeks' time and geometry of CAN make ^{222}Rn and its progenies to reach equilibrium with ^{226}Ra.

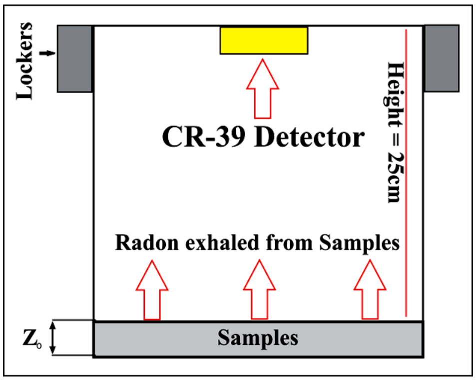

Figure 2. Experimental set up for RER measurement.

After the completion of exposure period, detectors were retrieved and etched in 6M NaOH solution at the temperature of 70 °C for 6 hours. Thereafter, CR-39 detectors were cleaned with the distilled water. Optical microscope was used for counting alpha tracks. Thereafter, track densities were measured using equation (12).

$$\text{Track Density } (\rho) = \frac{\text{Total Number of Tracks}}{\text{Area of the Field of View}} \quad (12)$$

Track densities, after background correction, were used to get the radon concentrations with the help of equation (13) and calibration factor (K) of 2.7 tracks-cm^{-2} .h^{-1}. kBq^{-1} .m^{-3} [39-40].

Radon exhalation rate

Before estimating radon exhalation rate, radon gas concentration was measured. $C_{222_{Rn}}$ (in Bq m^{-3}) (in air) was related with the track density 'ρ (in tracks cm^{-2})' and exposure time 'T (in hours)' using the Equation (13);

$$C_{222_{Rn}}(\text{in Bq m}^{-3}) = \left(\frac{\text{Track Densities (in tracks cm}^{-2})}{\text{Caliberation Factor} \times \text{Exposure Time (in hours)}}\right) = \frac{\rho\ (\text{in tracks cm}^{-2})}{K \times T\ (\text{in hours})} \quad (13)$$

After estimating radon concentration, radon exhalation rate was calculated using the equation (14) [33]

$$E = \frac{C_{222_{Rn}}[\omega A+\lambda V]}{A\left[1-\left(e^{-\left(\frac{\omega A}{V}+\lambda\right)T}\right)\right]} \quad (14)$$

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Where the symbols 'λ' stands for decay constant measured in h^{-1}, 'T' is for ^{222}Rn exposure time (in hours), 'V' is volume of CAN in m^3, 'A' is surface area of the sample in m^2. We have also corrected radon exhalation rate for back diffusion parameters. Corrected values of radon exhalation rate were measured using equation (15).

$$E_{corrected} = E - \omega C_{222_{Rn}} \quad (15)$$

Where, $\omega = \varepsilon\lambda Z_0$, is back diffusion constant for any particular material, Z_0 is the depth of sample within CAN, C_{222Rn} is the activity concentration of ^{222}Rn just over the surface of sample.

RESULTS

Results obtained from the measurements carried out for the detection of radionuclide's viz., ^{226}Ra, ^{232}Th and ^{40}K in twenty eight marble samples are displayed in table 1. The concentration of ^{226}Ra in the Gahirat Marble varied from 5.57±0.39 to 51.98±1.47 $Bqkg^{-1}$ with the mean value of 31.60±0.99 $Bqkg^{-1}$. The concentration of ^{232}Th ranged from below lower limit of detection to 12.41±2.67 $Bqkg^{-1}$ with the mean value of 1.53±0.31 $Bqkg^{-1}$. The concentration of ^{40}K ranged from below the lower limit of detection to 5.27±1.59 $Bqkg^{-1}$, with the mean value of 33.68±8.09 $Bqkg^{-1}$.

Table 1. The activity contents of naturally occurring radionuclides in the Gahirat Marble, Queries Location: Q1-Q3 (Chinar), Q4-Q6 (Goja Lasht), Q7-Q9 (Khairabad), Q10-Q12 (Kesu), Q13-Q14 (Gang), Q15-Q20 (Gahirat), Q21-Q23 (Gumbaz), Q24-Q26 (Ayun), Q27-Q28 (Chitral).

Sample ID	Activity of ^{226}Ra ($BqKg^{-1}$)	Activity of ^{232}Th ($BqKg^{-1}$)	Activity of ^{40}K ($BqKg^{-1}$)
Q1	12.37±0.47	Below LLD	30.83±7.77
Q2	21.66±1.31	Below LLD	Below LLD
Q3	13.28±0.47	Below LLD	Below LLD
Q4	12.5±0.46	Below LLD	26.77±7.42
Q5	16±1.27	Below LLD	Below LLD
Q6	10.97±1.06	Below LLD	Below LLD
Q7	5.57±0.39	Below LLD	33.68±8.09
Q8	10.87±0.45	Below LLD	Below LLD
Q9	17.33±1.29	Below LLD	Below LLD
Q10	22.52±1.34	Below LLD	Below LLD
Q 11	36±0.68	Below LLD	25.66±7.41
Q12	51.98±1.47	9.98±2.67	Below LLD
Q13	36.54±0.68	Below LLD	Below LLD
Q14	35.81±0.66	Below LLD	Below LLD
Q15	44.49±1.30	9.19±2.37	Below LLD
Q16	48.28±1.43	11.24±2.65	Below LLD
Q17	41.72±1.36	Below LLD	Below LLD
Q18	39.75±1.35	Below LLD	Below LLD
Q19	42.95±1.38	Below LLD	Below LLD
Q20	43.95±1.36	Below LLD	Below LLD
Q21	38.42±0.69	12.41±0.93	16.16±7.11
Q22	41.41±0.72	Below LLD	Below LLD
Q23	35.01±0.65	Below LLD	Below LLD
Q24	38.52±0.67	Below LLD	14.55±6.79
Q25	44.22±1.35	Below LLD	Below LLD
Q26	40.77±0.68	Below LLD	Below LLD
Q27	43.29±1.41	Below LLD	Below LLD
Q28	38.56±1.34	Below LLD	Below LLD
Mean	**31.60±0.99**	**1.53±0.31**	**5.27±1.59**
Max value	**51.98±1.47**	**12.41±2.67**	**33.68±8.09**
Min value	**5.57±0.39**	**Below LLD**	**Below LLD**

Radium equivalent activity (Ra_{eq}) have been estimated to assess radiation hazards associated with the use of Gahirat marble as decorative building stones. Table 2 shows that the value of Ra_{eq} activity, in samples of Gahirat Marble, ranging from 8.163±10.45 to 66.25±5.29 $Bqkg^{-1}$ with the mean value of 34.19±1.55$Bqkg^{-1}$. It is observed that values of Ra_{eq} are smaller as compared to the standard value, for the harmless use of building materials, which is 370 $Bqkg^{-1}$ [32].

The suitability of stones, in terms of possible radiological effects, for their use as building materials can be further envisaged from the estimated values of H_{ex}. The radiation hazard indices, external and internal hazard indices, were calculated and found with very low values. H_{ex} for current marble samples varied from 0.022±0.0027 to 0.179±0.014 with mean value of 0.092±0.004. Values of H_{ex}, for all marble samples, were found lower than unity (see table 2). The values of H_{in} in marble samples varied from 0.037±0.0037 while the mean value was found as 0.178±0.0034 (table 2). These values are less than unity, so Gahirat Marbles may be considered safe for possible public exposure and can be used as a safe building stone [14]. Results for the Ra_{eq}, H_{ex} and H_{in} are displayed in table 2.

In order to further investigate the radiological hazards associated with the use of Gahirat marbles, gamma dose rate $(\dot{D})$ have been evaluated. The absorbed dose rate, for indoor air, $\dot{D}_{in}$ (nGy h^{-1}) arising from radium, thorium and potassium radionuclide's exposures was estimated using equations (6) and (7) and results are displayed in table 3. It can be seen that the values of indoor dose rates ranges from 3.98±0.52 to 30.04±2.29 nGy h^{-1} and with mean value of 15.78±0.30 nGy h^{-1}. The range of values obtained for gamma dose rate, in current study, was found to be less than the world range from 10 to 200 nGy h^{-1} [14, 41].

The numeric values of outdoor external absorbed dose rate ($\dot{D}_{out}$) calculated due to the occurrence of ^{226}Ra, ^{232}Th and ^{40}K are displayed in table 3. The values of $\dot{D}_{out}$ (see table 3) in marble samples varied from 2.84±0.37 to 21.46±1.64 nGy h^{-1} with the mean value of 11.27±0.22 nGy h^{-1}. The values of total dose rate ($\dot{D}$) are also displayed in table 3. The values of $\dot{D}$ shown in table 3, ranged from 6.82±0.89 to 51.5±3.93 nGy h^{-1} with the mean value of 27.05±0.51 nGy h^{-1}.

The annual indoor effective dose equivalent $(E,\ mSv\ y^{-1})$ received by the population, due to exposure of radiation, from the Gahirat Marble sample, was estimated and results are displayed in table 4. Measured values of $E\ (mSv\ y^{-1})$ ranged from 0.02±0.003 to 0.18±0.014 mSv y^{-1} and with average value of 0.1±0.002 mSv y^{-1}. We have used an indoor occupancy factor of 8760 hrs (80%) for a complete year and a dose conversion factor of 0.7 SvGy y^{-1} in calculations. The gamma activity index (I_γ) was calculated and results of 'I_γ' for the marble samples

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are displayed in table 4. The I_γ is associated with the cause of excess external radiation triggered by superficial material and the values of annual dose rates. Results for (I_γ) are displayed in table 4. The values of gamma index (I_γ) in the marble samples ranged from 0.06±0.004 to 0.446±0.018 with the mean value of 0.229±0.002.

The Alpha index (I_α) was calculated which accounts for the excess alpha radiation exposure, originated from building stones, resulting from inhalation and are displayed in table 4. For the current study, the estimated I_α values in the marble varied from 0.028±0.002 to 0.26±0.007 with the average value of 0.158±0.003.

Radon exhalation rate (RER)

Table 5 shows activity concentration of radon gas and surface radon exhalation rates. Radon concentration was found in the range 1.6±0.11 to 17.11±0.48 Bq m^{-3} with mean value 10.43±0.33 Bq m^{-3} (see figure 3 a & b). The values of radon exhalation rates were found in the range (1.01±0.07 to 9.67±0.27) $\times 10^{-2}$ Bq m^{-2} h^{-1} with mean value of (5.84±0.002)$\times 10^{-2}$ (Bq m^{-2} h^{-1}). Range of radon, radium and relationship between radon and radium and radium and radon exhalation rate are shown in figure 3(a,b,c,d). Estimated values of radon and RER for marble samples are given in table 5.

Table 2. Radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal hazard (H_{in}) indices.

Sample ID	Radium Equivalent Activity (Ra_{eq}) (Bq kg^{-1})	External hazard (H_{ex})	Internal hazard (H_{in})
Q1	14.74	0.039842	0.073274
Q2	21.66	0.058541	0.117081
Q3	13.28	0.035892	0.071784
Q4	14.56129	0.039349	0.073133
Q5	16	0.043243	0.086486
Q6	10.97	0.029649	0.059297
Q7	8.16336	0.022056	0.03711
Q8	10.87	0.029378	0.058757
Q9	17.33	0.046838	0.093676
Q10	22.52	0.060865	0.12173
Q 11	37.97582	0.102632	0.199929
Q12	66.2514	0.179019	0.319506
Q13	36.54	0.098757	0.197514
Q14	35.81	0.096784	0.193568
Q15	57.6317	0.155726	0.275969
Q16	64.3532	0.173884	0.304371
Q17	41.72	0.112757	0.225514
Q18	39.75	0.107432	0.214865
Q19	42.95	0.116081	0.232162
Q20	43.95	0.118784	0.237568
Q21	57.41062	0.155113	0.25895
Q22	41.41	0.111919	0.223838
Q23	35.01	0.094622	0.189243
Q24	39.64035	0.107133	0.211241
Q25	44.22	0.119514	0.239027
Q26	40.77	0.110189	0.220378
Q27	43.29	0.117	0.234
Q28	38.56	0.104216	0.208432
Mean value	34.19±1.55	0.092±0.004	0.178±0.0034
Max. value	66.25±5.29	0.179±0.014	0.32±0.0180
Min. Value	8.163±10.45	0.022±0.0027	0.037±0.0037

Table 3. Absorbed dose rate, external and internal dose rate (nGy h-1).

Sample code	Indoor Dose Rate $\dot{D}_{in}$ (nGyh^{-1})	Outdoor Dose Rate $\dot{D}_{out}$ (nGyh^{-1})	Total Dose Rate $\dot{D}$ (nGyh^{-1})
Q1	8.09±0.54	5±0.39	13±0.93
Q2	10.01±0.61	7.15±0.43	17.16±1.04
Q3	6.14±0.22	4.38±0.16	10.52±0.37
Q4	6.89±0.52	4.92±0.37	11.81±0.89
Q5	7.39±0.59	5.28±0.42	12.67±1.01
Q6	5.07±0.49	3.62±0.35	8.69±0.84
Q7	3.98±0.52	2.84±0.37	6.82±0.89
Q8	5.02±0.21	3.59±0.15	8.61±0.36
Q9	8.01±0.6	5.72±0.43	13.73±1.02
Q10	10.4±0.62	7.43±0.44	17.84±1.06
Q 11	17.7±0.62	12.64±0.45	30.35±1.07
Q12	30.04±2.29	21.46±1.64	51.5±3.93
Q13	16.88±0.31	12.06±0.22	28.94±0.54
Q14	16.54±0.3	11.82±0.22	28.36±0.52
Q15	26.11±2.03	18.65±1.45	44.75±3.48
Q16	29.09±2.26	20.78±1.62	49.88±3.88
Q17	19.27±0.63	13.77±0.45	33.04±1.08
Q18	18.36±0.62	13.12±0.45	31.48±1.07
Q19	19.84±0.64	14.17±0.46	34.02±1.09
Q20	20.3±0.63	14.5±0.45	34.81±1.08
Q21	25.92±1.18	18.51±0.84	44.43±2.02
Q22	19.13±0.33	13.67±0.24	32.8±0.57
Q23	16.17±0.3	11.55±0.21	27.73±0.51
Q24	18.4±0.59	13.14±0.42	31.55±1.02
Q25	20.43±0.62	14.59±0.45	35.02±1.07
Q26	18.84±0.31	13.45±0.22	32.29±0.54
Q27	20±0.65	14.29±0.47	34.29±1.12
Q28	17.81±0.62	12.72±0.44	30.54±1.06
Mean	15.78±0.30	11.27±0.22	27.05±0.51
Max value	30.04±2.29	21.46±1.64	51.5±3.93
Min value	3.98±0.52	2.84±0.37	6.82±0.89

Table 4. Values of annual effective dose (E), gamma activity index (I_γ) and alpha index (I_α) for marble samples.

Sample ID	Annual Effective Dose Eq. E (mSvy^{-1})	GammaHazard index (I_γ)	Alpha Hazard index (I_α)
Q1	0.04±0.003	0.103±0.004	0.062±0.002
Q2	0.06±0.004	0.144±0.004	0.108±0.007
Q3	0.04±0.001	0.089±0.002	0.066±0.002
Q4	0.04±0.003	0.101±0.004	0.063±0.002
Q5	0.05±0.004	0.107±0.004	0.08±0.006
Q6	0.03±0.003	0.073±0.004	0.055±0.005
Q7	0.02±0.003	0.06±0.004	0.028±0.002
Q8	0.03±0.001	0.072±0.002	0.054±0.002
Q9	0.05±0.004	0.116±0.004	0.087±0.006
Q10	0.06±0.004	0.15±0.004	0.113±0.007
Q 11	0.11±0.004	0.257±0.005	0.18±0.003
Q12	0.18±0.014	0.446±0.018	0.26±0.007
Q13	0.1±0.002	0.244±0.002	0.183±0.003
Q14	0.1±0.002	0.239±0.002	0.179±0.003
Q15	0.16±0.012	0.389±0.016	0.222±0.007
Q16	0.18±0.014	0.434±0.018	0.241±0.007
Q17	0.12±0.004	0.278±0.005	0.209±0.007
Q18	0.11±0.004	0.265±0.005	0.199±0.007
Q19	0.12±0.004	0.286±0.005	0.215±0.007
Q20	0.12±0.004	0.293±0.005	0.22±0.007
Q21	0.16±0.007	0.391±0.009	0.192±0.003
Q22	0.12±0.002	0.276±0.002	0.207±0.004
Q23	0.1±0.002	0.233±0.002	0.175±0.003
Q24	0.11±0.004	0.267±0.004	0.193±0.003
Q25	0.13±0.004	0.295±0.005	0.221±0.007
Q26	0.12±0.002	0.272±0.002	0.204±0.003
Q27	0.12±0.004	0.289±0.005	0.216±0.007
Q28	0.11±0.004	0.257±0.004	0.193±0.007
Mean value	0.1±0.002	0.229±0.002	0.158±0.003
Max. value	0.18±0.014	0.446±0.018	0.26±0.007
Min. Value	0.02±0.003	0.06±0.004	0.028±0.002

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Table 5. Estimated values of radon and RER for Gahirat marble samples.

Sample ID	Radon Concentration ($Bq\ m^{-3}$)	Radon Exhalation Rate ($Bq\ m^{-2}\ h^{-1}$)×10^{-2}
Q1 Q2 Q3 Q4 Q5 Q6 Q7 Q8 Q9 Q10 Q 11 Q12 Q13 Q14 Q15 Q16 Q17 Q18 Q19 Q20 Q21 Q22 Q23 Q24 Q25 Q26 Q27 Q28	3.97±0.15	2.26±0.09
	6.56±0.4	3.88±0.24
	4.66±0.16	2.43±0.09
	4.45±0.16	2.3±0.08
	5.01±0.4	2.84±0.23
	3.63±0.35	2±0.19
	1.6±0.11	1.01±0.07
	3.45±0.14	2.04±0.08
	6.47±0.48	3.15±0.24
	9.53±0.57	4.08±0.24
	13.12±0.25	6.54±0.12
	17.11±0.48	9.67±0.27
	11.34±0.21	6.82±0.13
	10.21±0.19	6.52±0.12
	12.55±0.37	8.24±0.24
	15.64±0.46	9.14±0.27
	12.84±0.42	8.38±0.27
	13.19±0.45	7.15±0.24
	16.77±0.54	7.95±0.26
	15.16±0.47	7.97±0.25
	11.71±0.21	7.09±0.13
	11.63±0.2	7.63±0.13
	12.35±0.23	6.51±0.12
	12.96±0.23	7.08±0.12
	15.51±0.47	8.11±0.25
	13.16±0.22	7.54±0.13
	14.41±0.47	7.94±0.26
	12.99±0.45	7.19±0.27
Mean value	**10.43±0.33**	**5.84±0.002**
Max. value	**17.11±0.48**	**9.67±0.27**
Min. Value	**1.6±0.11**	**1.01±0.07**

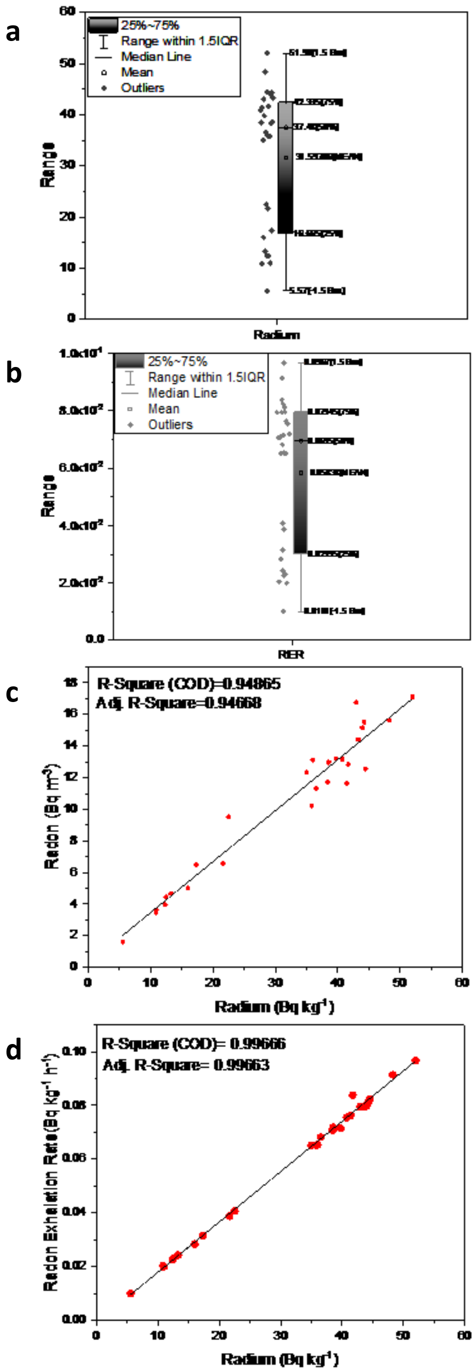

Figure 3. a. Range of ^{226}Ra activities, **b.** Range of ^{222}Rn activities, **c.** ^{226}Ra versus ^{222}Rn, **d.** 226Ra versus radon exhalation rate.

The surface RER values reported in current study, for export quality marble samples ranged from (1.01±0.07) ×10^{-2} to (9.67±0.27)×10^{-2} $Bqm^{-2}\ h^{-1}$ with mean value of (5.84±0.002)×10^{-2} $Bqm^{-2}\ h^{-1}$. Two sample t-tests for the mean of ^{226}Ra and ^{222}Rn were performed with Minitab®. The p-value obtained in this case was found less than 0.001 (i.e., $p<0.001$). As p-value in current case is less than 0.05 so it can be concluded that mean value of ^{226}Ra differs ^{222}Rn at the 0.05 level of confidence. Ninety five percent (95%) confidence interval (CI) have been estimated for the difference. CI quantifies the uncertainty associated with estimating the difference in means from the sample data. From the current study we are 95% confident that the true difference is between 15.525 and 26.815. No outliers were detected in both sample data for ^{226}Ra and ^{222}Rn.

DISCUSSION

Figure 3c shows that the relationship between radon and radium. A linear relationship, with coefficient of determination (CoD) value 0.94 exists

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between radon and radium. Likewise, the relationship between RER and radium is also found as linear with CoD value of 0.99 (figure 3d). Both CoD values obtained from radon and radium and then radon exhalation rate and radium relations are justified due to the reason that ^{222}Rn is an immediate decay product of ^{226}Ra. Radon and radium are part of ^{238}U radioactive series and radon is obtained whenever radium decays with the emission of alpha particles. The ^{222}Rn dependence on ^{232}Th has not been investigated by virtue of the fact that ^{222}Rn does not fall in the decay chain of ^{232}Th radioactive series.

Occurrence of radionuclides in marble samples is due to the fact that uranium is present to some extent in all types of rocks. In most rocks uranium minerals, viz. coffinite, uraninite, carnotite, tyuyamunite, autunite, brannerite and uranophane along with heavy minerals viz. titanite, allanite, zircon and monazite are found in predictable abundances. Usually, those rocks having uranium concentration greater than 5 parts per million are considered to pose a threat of high concentrations of indoor radon exposure. These rocks may include carbonaceous black shales, metamorphic rocks with granitic composition, uranium-bearing granites, glauconite-bearing sandstones, pegmatites, pyroclastic volcanic rocks, felsic and alkalic volcanoclastic and many other sheared or faulted rocks. On the other hand rock types having the composition of marine quartz sands, metamorphic and igneous rocks of mafic composition, non-carbonaceous shales and siltstones, and mafic volcanic rocks are considered to pose less threat of radon exposure. Average values of uranium concentrations in metamorphic rocks are usually 2 ppm [(42)]. For the current study, lower values of radionuclide concentration are reported which is due to the reason that natural origin of Gahirat marble samples belongs to metamorphic rock type.

The surface radon exhalation rate values obtained in the current study were found considerably lower than that are reported for white marbles of Egypt (range 0.03 ±0.01 Bqm^{-2} h^{-1}), Iraq (mean value 1.21 Bqm^{-2} h^{-1}) and Nigeria (range 0.72 to 1.71 with mean value 1.06± 0.56 Bqm^{-2} h^{-1}) [(43-48)].

In table 6, for the current study, the concentration of ^{226}Ra in Gahirat Marble was found higher than that reported for countries viz. Algeria [(43)], Kuwait [(44)], Cameroonian [(45)], Jordan [(46)], Saudi Arabia [(47)] and less than as compared to the values reported for Egypt [(48)]. The mean activity concentration of ^{232}Th and ^{40}K were found marginally higher than that reported for the marble samples of Kuwait and Cameroonian, while lower than the values reported for the countries like Algeria, Egypt, Saudi Arabia and Jordan.

Table 6. Comparison of current study results with other studies conducted in different countries.

Country	^{226}Ra ($Bqkg^{-1}$)	^{232}Th ($Bqkg^{-1}$)	^{40}K ($Bqkg^{-1}$)	Reference
Pakistan	31.60±0.99	1.53±0.31	5.27±1.59	Present study
Algeria	23±2	18±2	310±2	(55)(43)
Kuwait	3.9±0.5	0.22±0.08	19±2	(56)(44)
Cameroonian	8±2	0.35±0.02	19±2	(57)(45)
Jordan	20.1	11.4	85	(58)(46)
Saudi Arabia	12.7±3.4	13.2±1.4	64±3.6	(59)(47)
Egypt	205±83	115±60	865±3.92	(60)(48)

CONCLUSION

Radiological hazards due to exposure of radiations originating from natural radionuclides present in marble samples have been assessed. Radon exhalation rate was also estimated using the CAN passive detection method in order to find contribution of radon to the exposure. The levels of radionuclides viz. ^{232}Th, ^{226}Ra and ^{40}K, were found as 31.598± 0.989, 1.529± 0.308 and 5.273± 1.593 $Bqkg^{-1}$ respectively, which were observed lower than the standard values of 50, 50, and 500 Bq kg^{-1} respectively. The mean value of radon exhalation rate was found as (5.84±0.002) $\times 10^{-2}$ Bqm^{-2} h^{-1}. Radon exhalation rate was found reasonably smaller as compared to data available for most of the countries. The Ra_{eq} was found lower than the acceptable limits for safety. It is concluded from the study that Gahirat marble samples are safe for the use as decorative stones.

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