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Assessment of radiological hazards of travertine rocks as building materials

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ABSTRACT

Introduction: Although travertine is now widely used as a building material, the effects of its radiological hazards to public health should be a matter of concern. Therefore, this study aimed to determine its radiological parameters with regard to human health.
Material and Methods: In this research 10 travertine samples were collected from exploitation zone in Haji Abad Mine in Mahallat, Iran. Specific radionuclide activities were determined by gamma spectrometry using a high purity germanium detector. In addition, radiological parameters, such as radium equivalent, air doses, internal and external risk factors (H_{in}, H_{ex}), yearly gonadal dose equivalent (AGDE), and additional cancer risk (ELCR) throughout life were calculated.
Results: The specific activities of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides ranged within 3.08-9.16, 1.22-6.45, and 20.15-91.04 Bq/kg, respectively. The obtained means of internal and external annual effective dose of samples were 0.03 and 0.007 mSv/y. External and internal hazard indices for samples were within the range of 0.003-0.01 and 0.01-0.03, respectively.
Conclusion: The results of this study show that the amount of radionuclides in travertine rocks is very small compared to its global average in soil and rock. Therefore, it is suggested to use travertine as a building material, which is not a threat to public health.

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Introduction

There are many active mines of travertine stones in Mahallat, Iran. Hajiabad Mine is one of the largest and most qualified travertine quarries in West Asia. Travertine is a kind of limestone that has a chemical origin and is formed by the deposition of calcium carbonate near springs, caves, and marshy ponds. In other words, its formation is mainly due to the activity of hot springs. Travertine is layer-shaped with porous texture [1]. Travertine stones are widely used in buildings as the interior or exterior cladding and modern architecture. Therefore, it is of great importance to measure the radiation dose exposure to the inhabitants of these buildings. Radioactive nuclei exist naturally and artificially in our surrounding environment, and all people are exposed to nuclear radiation. Different types of materials, such as soil, stones, water, and plants contain a small amount of radionuclides [2]. Therefore, it is important to determine their amount and effects on living creatures.

Specific activities of radionuclides in the Earth's crust reported to range within 16-110 Bq/kg (mean= 35 Bq/kg) for ^{238}U, 17-60 Bq/kg (mean= 35 Bq/kg) for ^{226}Ra, 11-64 Bq/kg (mean= 30 Bq/kg) for ^{232}Th, and 140-850 Bq/kg (mean: 400 Bq/kg) for ^{40}K [2, 3]. It is, therefore, important to analyze gamma radiation spectrometry and determine the number of radioactive nuclei in rocks and soil both from the perspectives of geologists and radiologists, in order to form the basis for estimating the dose absorbed by inhabitants [4, 5]. Accordingly, this study aimed to determine specific activities of radionuclides ^{226}Ra, ^{232}Th, and ^{40}K, radiological parameters, including radium equivalent (Ra_{eq}), absorbed dose rate in air (D) at 1 m above the ground, indicators of internal and external hazards (H_{in}, H_{ex}), annual effective dose equivalent indoor and outdoor (AEDE), annual gonadal dose equivalent (AGDE), and the excess lifetime cancer risk (ELCR) in the samples.

Materials and Methods

Sampling and Sample Preparation

In this research, ten active walls under exploitation in 100 hectares' area of Hajiabad Mine in western Iran, where Mahallat district in Markazi Province is located, were selected for collecting the data samples. Ten samples were collected from these sites. Sampling method in this research was performed randomly and experimentally, through which the researchers took up

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to 2 kg of each sample. The geographical locality of sampling sites is shown in Figure1.

Firstly, travertine stone samples were powdered through the use of a jaw crusher to remove possible moisture. Secondly, each sample was dried by an oven for 6 hours at 120 ° C to maintain constant weight during the measurement. Thirdly, the samples were sieved through a 50-mesh screen until a fine powder was achieved [6]. Finally, the powdered samples were packed and sealed in the standard Marinelli Baker's containers with net mass of 950 grams due to the fact that radon gas always tends to escape from the surface of the samples. In order to attain equilibrium between ^{226}Ra and its daughter ^{222}Rn, the samples were kept for at least 50 days at the laboratory [6]. Samples were given T1-T10 codes.

Gamma-ray spectrometry was performed using high purity germanium detector (GCD30195BSI model, manufactured by Baltic Scientific Instruments Ltd [Riga, Lv-1005, Latvia] with a relative efficiency of 30% and spectra were registered by Lsrmbsi software (005- Latvia). The energy resolution of this detector was 1.95 keV for the gamma line at 1332.520 keV ^{60}Co and its operating voltage was 3000 V. Each spectrum was registered for one day (86400 seconds). Energy calibration and system efficiency were estimated by using standard source containing ^{152}Eu, ^{137}Cs, and ^{241}Am radionuclides for each exact activities. The analysis of the spectra was performed by using the Maestro II Gamma Vision 32 software (manufactured by EG&G Ortec Company, Tennessee 37831, USA). In order to reduce the effects of background radiations, the detector was shielded by a 10-cm thick layer of lead and two inner layers of copper and cadmium with the thickness of 2 and 1 mm, respectively. This shield reduced the cosmic soft beams containing low-energy photons and electrons to a very low level. The copper layer effectively absorbed X-rays produced in lead shield with the energy of 73.9 keV and prevented them from entering the detector [7]. Thermal neutrons were effectively absorbed by the cadmium layer and did not reach the detector. Background radiation correction was measured by the subtraction of the spectrum recorded for the empty container from each spectrum under the same conditions.

To determine the gamma-ray efficiency of the detector, *Equation 1* was used [8].

$$\varepsilon(\%)=\frac{N_i}{Act\times P_n(E_i)\times t}.100 \qquad (1)$$

Where, the N_i is net counts under the full-energy peak corresponding to the E_i energy, *Act* refers to the activity of radionuclide in the standard container in Bq, $Pn\ (E_i)$ denotes the probability of the gamma photon emission with E_i, per decay in terms of percentage and t is the spectral time in second. The energy efficiency curve of the detector versus that of gamma-rays was plotted using mathematical Table Curve 2D software [9].

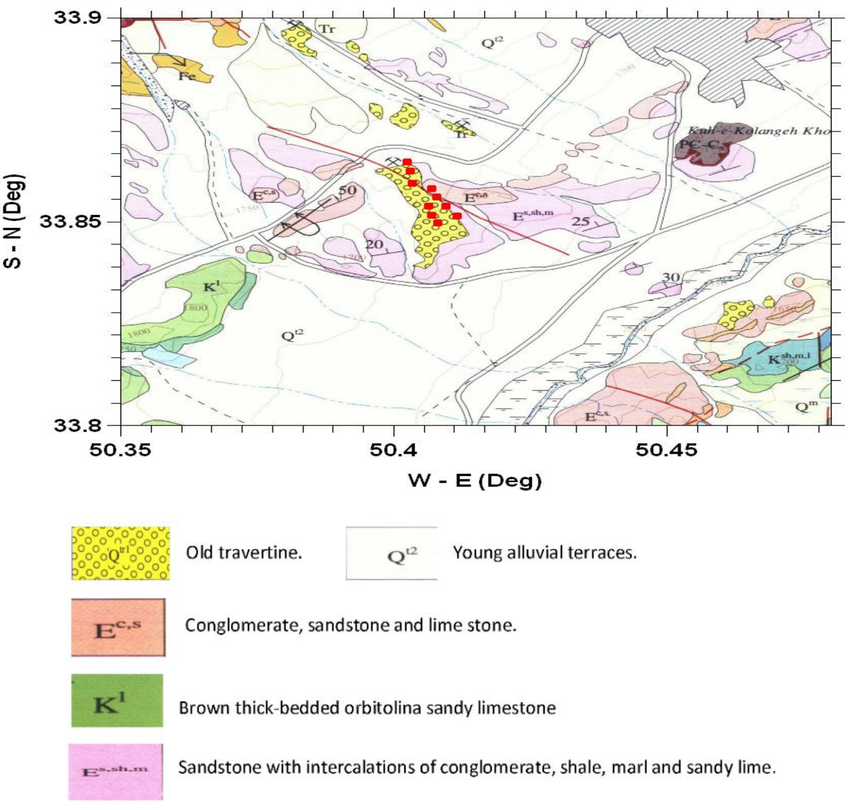

Figure 1. Sampling location map of Hajiabad mine marked with red color point

Specific activity measurements of radionuclides

Equation (2) was used to calculate the specific activity of radionuclide [10]:

$$\mathrm{Act} = \frac{\mathrm{Ni}}{\varepsilon\,.\times \boldsymbol{P_n(E_i)} \times \mathrm{t} \times \mathrm{m}} . 100 \qquad (2)$$

Where, the *Act* is the specific activity of radionuclide, *Ni is the net area under the peak corresponding to the specified energy, and P_n refers to the probability of gamma radiation emission in E_i energy in percent*. ε signifies the efficiency of the energy detector. Moreover, *t* is the spectral time of the sample in seconds, and *m* is the sample mass in kilograms [10]. In order to determine the specific activity of ^{226}Ra in samples, we used gamma-ray lines with 361.95 keV and 609.31 keV for ^{214}Pb and ^{214}Bi, respectively. The gamma-ray lines 911.21 and 968.78 keV (^{228}Ac) with emission probability of 100 per decay (26.6% and 17.4%, respectively) were used to determine the specific activity of ^{232}Th. The specific activity of ^{40}K was determined from its gamma line with 1460.70 keV energy. Moreover, the gamma line of 661.66 keV was used to determine the specific activity of ^{137}Cs in samples [11].

Radiological hazard parameters

Radium equivalent

The distribution of natural radioactive nuclei is not uniform in the environment, so an indicator for determining and comparing their total level of radioactivity is defined, which is named radium equivalent. With regard to gamma radiation, 98.5% of the radiological hazards of the uranium series are due to the ^{226}Ra and its progeny daughters. Therefore, the total radiation of the sample is assessed in terms of the activity of the radium by using equation 3. This indicator was determined based on the fact that 10 Bq/kg ^{226}Ra, 7 Bq/kg ^{232}Th, and 130 Bq/kg ^{40}K were given the equal dose rate [12].

$$Ra(eq) = A_{Ra} + 1.43\, A_{Th} + 0.077 A_K \qquad (3)$$

Figure 2 shows the radiological map of the radium equivalent using the SURFER15 software [13], where its amount is given for equivalent lines in Bq/kg.

Absorbed gamma dose rate

The amount of D in each zone depends on the contamination of ^{226}Ra, ^{232}Th, and ^{40}K in soil and rocks. The D at 1 m above the ground originates from natural radioactivity due to gamma radiation, which was calculated by using *Equation 4* [14, 15]:

$$D(nGy/h)=0.462A_{Ra}+0.604A_{Th}+0.0417A_K \qquad (4)$$

Where, 0.462, 0.604, and 0.0417 are the conversion factors in nGy/h per 1 Bq/kg of specific activities of the radionuclides, respectively [12].

Internal Hazard Index

As a result of the breakdown of a series of uranium and thorium, a certain amount of radon gas is formed, some of which come into the air and can enter the human body through breathing. In order to estimate the amount of exposure from radon gas and its products, H_{in} was employed based on *Equation 5* [12]:

$$H_{in}= A_{Ra}/ 185+A_{Th}/ 259+A_K/ 4810 < 1 \qquad (5)$$

External Radiation Hazard (Hex)

External threat (hex) means exposure to the external radiation associated with gamma-rays as a result of the breakdown of ^{226}Ra, ^{232}Th series, and ^{40}K. In order to ensure that the environment is safe and does not require radiation protection intervention, this coefficient should not exceed one. This amount is obtained on the basis of the permissible equivalent radium limit for building materials (370 Bq / kg) [16]. The external risk indicator is presented in *Equation 6* [8].

$$H_{ex}= A_{Ra}/ 370+A_{Th}/ 259+A_K/ 4810 < 1 \qquad (6)$$

Annual Gonadal Dose Equivalent

The AGDE is an annual equivalent dose absorbed by reproductive organs, as well as sensitive parts of the body, such as the breast, bone marrow, and surface bone cells in the population that United Nations Scientific Committee on the Effects of Atomic Radiation is considering [1, 2]. The amount of AGDE resulting from the decay of ^{226}Ra, ^{232}Th series, and ^{40}K was calculated using Equation 7 [17].

$$AGDE\ (mSv/y)=(3.09A_{Ra}+4.18A_{Th}+0.314A_K)\times10^{-3} \qquad (7)$$

Where, A_{Ra}, A_{Th}, and A_K in equations 3 to 7 are specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively.

Annual effective dose equivalent outdoor and indoor

The annual effective dose equivalent rate for outdoor and indoor occupancy can be calculated by considering the values of the absorbed dose rate in ambient air 1 m above ground using, as presented in Equations *8* [1]:

$$AEDE\ (mSv/y)=D\times F\times 8766\times 0.7\times 10^{-3} \qquad (8)$$

Where, *D* is dose rate in nGy/h, and 8766(h/y) is the annual time. Moreover, *F* defined occupancy factor as 0.2 and 0.8 for outdoor and indoor annual effective dose equivalent, respectively. Additionally, 0.7×10^{-3} refers to the conversion coefficient in mSv/Gy [18]. According to the European Convention on radiation protection, building material can be used when the annual effective dose absorption by residents is less than one mSv/y. Therefore, the risks of nuclear radiation from the natural radioactivity can be neglected. On the report of the European Union, if the annual dose absorption exceeds 1 mSv/y, it should be taken into account from the radiation protection point of view.

Excess lifetime cancer risk

Equation 9 was used to calculate the developing of lifetime cancer risk due to gamma-ray radiation exposure to the resident population by radionuclides content of the soil.

$$ELCR=AEDE\times DL\ (75.8y)\times RF\ (0.05Sv^{-1}) \qquad (9)$$

In this regard, the AEDE, the annual effective dose equivalent, DL, the median lifespan of 75.7 years and RF were the risk factors for 1 Sv absorbed dose [19-20].

Results

Table 1 shows results of recorded gamma-ray spectra, the specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in all the samples. The results of the radiological parameters calculations for samples, including Ra_{eq}, H_{in}, H_{ex}, D, AEDE indoor, AEDE outdoor, and ELCR are presented in Table 2. Figure 3 shows the radiological map of dose rate distribution derived by SURFER15 software, which is similar to radium equivalent map. On the curves, the amount of dose rate in air was given in nGy/h.

Table 1. Geographical position of sampling points and specific activities of radionuclides in the samples in Bq/kg

Sample code	Geographical coordinate		Specific activities (Bq/kg)			
	Longitude E	Latitude N	^{226}Ra	^{232}Th	^{232}Th	^{137}Cs
T1	44.5241	37.45656	4.08±0.22	2.07±0.29	23.95±0.80	ND*
T2	44.5576	37.45830	6.89±0.50	6.45±0.60	84.19±1.80	0.23±0.09
T3	44.5378	37.46105	3.21±0.22	1.22±0.21	17.44±0.90	ND
T4	44.5143	37.45861	5.94±0.42	3.98±0.46	55.93±1.40	ND
T5	44.5099	37.46100	9.16±0.58	6.83±0.67	91.04±2.00	ND
T6	44.5211	37.46369	3.82±0.37	1.38±0.42	26.27±1.30	ND
T7	44.4794	37.47067	3.08±0.31	2.65±0.49	32.60±1.10	ND
T8	44.4801	37.46735	4.04±0.31	2.18±0.36	31.99±1.10	ND
T9	44.5133	37.46735	5.18±0.30	1.42±0.25	20.15±0.60	ND
T10	44.4731	37.47333	4.39 ±0.25	2.26±0.29	31.78±0.80	ND
Mean	-----	-----	4.98±0.35	3.07±0.47	41.53±1.20	-------

* ND: not detected

Table 2. Radiological hazards of the soil samples: Ra_{eq} (Bq/kg), D (nGy/h), Hin, H_{ex}, outdoor and indoor AEDE (mSv/y), AGDE (mSv/y), and ELCR

Sample code	Ra_{eq} (Bq/kg)	D (nGy/h)	H_{in}	H_{ex}	AEDE outdoor (mSv/y)	AEDE indoor (mSv/y)	AGDE (mSv/y),	ELCR ($\times10^{-3}$)
T1	8.88	4.15	0.004	0.02	0.005	0.02	0.003	0.02
T2	22.59	10.59	0.005	0.05	0.013	0.05	0.004	0.04
T3	6.29	2.95	0.003	0.01	0.003	0.01	0.002	0.01
T4	15.75	7.48	0.006	0.03	0.009	0.04	0.005	0.03
T5	20.06	12.26	0.01	0.05	0.015	0.06	0.009	0.05
T6	7.81	3.70	0.003	0.02	0.004	0.02	0.002	0.02
T7	9.37	4.41	0.004	0.02	0.005	0.02	0.003	0.02
T8	9.61	4.54	0.003	0.02	0.005	0.02	0.002	0.02
T9	7.37	4.11	0.004	0.01	0.005	0.02	0.003	0.02
T10	10.54	4.65	0.004	0.02	0.006	0.02	0.004	0.02
Mean	11.83	5.61	0.005	0.02	0.007	0.03	0.004	0.02

Discussion

In this study, the specific activities of radionuclides ^{226} Ra, ^{232}Th, and ^{40}K in travertine samples varied within the range of 3.08 ± 0.31 to 6.28±0.89, 1.22±0.22 to 6.83±0.86, and 17.44±0.90 to 91.04±2.00 in Bq/kg, respectively. These ranges were significantly lower than the global average for these nuclei (35, 40 and 412, respectively) [2]. Accordingly, travertine rocks can be used safely without being a threat to public health. Travertine rocks are formed in the bed of hot and cold springs, through which the salt from radium, thorium, and potassium is dissolved in water and transferred to other areas. Therefore, the low level of radioactivity of these types of rock confirms the theory of their formation [1].

The ^{137}Cs as an artificial radionuclide was observed only in the T2 sample (0.92±0.23 Bq/kg). This seems reasonable due to the depth of the soil samples and low annual raining in this region, while the soil of Markazi Province is polluted to this radionuclide [21]. This particular nucleus has been transmitted by atmospheric flows from abroad, and it has been found mostly in the soil and the surface of rocks, which is due to the low rainfall and a lower depth of penetration into rocks. The obtained results for the radium equivalent activity values ranged within 6.29-22.59 with the mean of 11.83 Bq/kg, which was lower than the world average (131.69 Bq/kg) and the maximum permissible value of building materials (370 Bq/kg) [16]. Therefore, this type of rocks can be used favorably as building materials along with other rocks. The mean of H_{in}, H_{ex} for travertine samples were 0.005 and 0.02, respectively, which is due to their safety and does not pose any risk to inhabitants. The mean of absorbed dose rate in air at 1 m height above ground, the mean of AEDE indoor, AEDE outdoor for travertine samples were 5.61 nGy/h, 0.03 and 0.007 in mSv/y, respectively, which was lower than the global average (55 nGy/h and 1 mSv/y) [1]. Accordingly, its risk for the population is negligible. The amounts of worldwide average of AGDE and ELCR is 460 µSv/y and 0.29×10^{-3} respectively, the results obtained in this study were very lower than the global average [1]. In Table 3, the results of this study were compared with the reported results of some other studies in different countries. As you can see, the radioactivity of travertine rocks in the current study is consistent with the results of other countries, and its level is much lower than in the case of granite [21-25]. Moreover, the reported ratio of radium equivalent (R) for different types of stones in comparison with the results of the current study shows in most cases R> 1, which refers to the better quality for building materials.

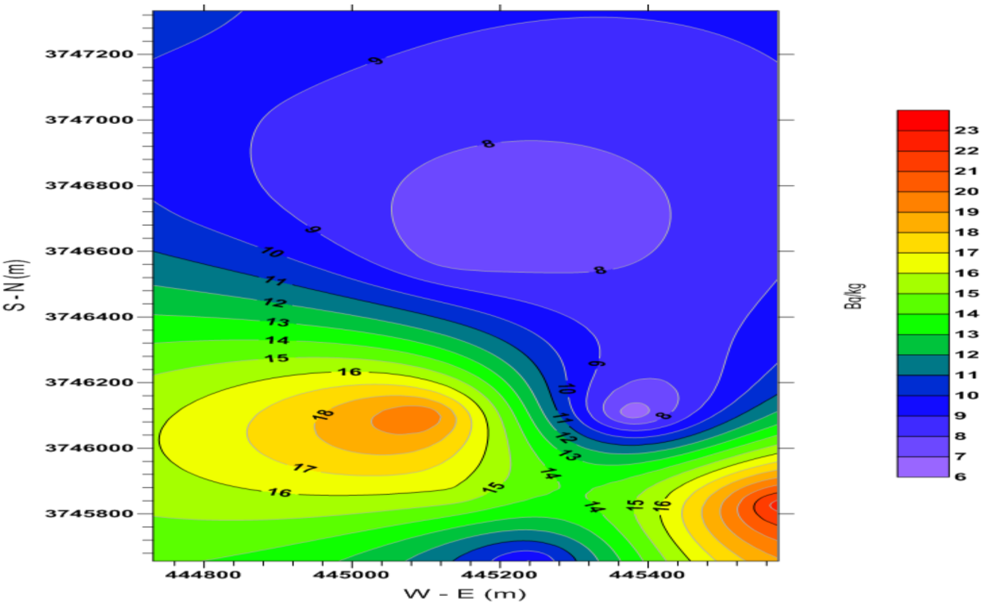

Figure 2. Radioactivity map of Hajiabad travertine min basis on radium equivalent in Bq/kg

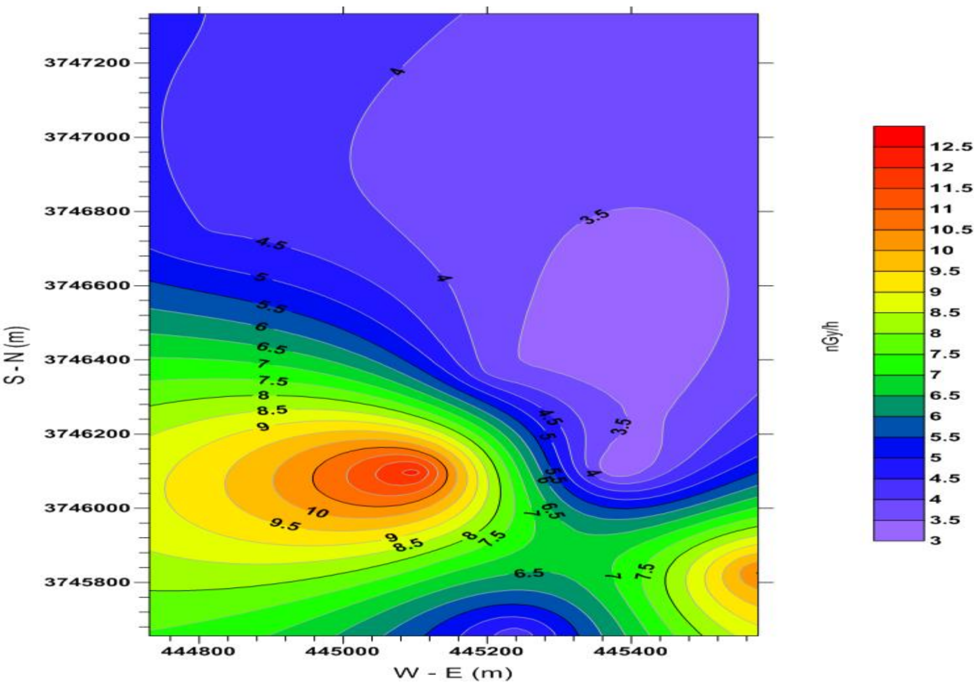

Figure 3. Absorbed dose rate distribution of Hajiabad travertine mine in nGy/h

Table 3. Comparison of the mean specific activities of radionuclide ^{226}Ra, ^{232}Th, ^{40}K, and radium equivalent with different types of rocks used as building materials

Kind of rock	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K(Bq/kg)	Ra_{eq} (Bq/kg)	$R=\frac{Ra_{eq}\,(other\ work)}{Ra_{eq}\,(this\ work)}$	reference
Travertine	4.98	3.1	41.5	11.88	1	This work
Travertine (Turkey)	0.8	0.9	4.1	2.16	0.18	22
Travertine (Germany)	4.0	19.0	20.0	32.63	2.75	23
Granit	134.0	108.0	1210	758.59	63.85	24
Gabbrodiorite Iran	11.1	13.2	257.2	49.78	4.19	24
Quartz diorite Iran	30.1	44.0	709.9	96.41	8.12	24
Diorite Iran	37.1	10.0	79.1	52.22	4.37	24
Monzodiorite Iran	37.2	26.1	235.4	76.49	6.44	24
Granodiorite Iran	41.6	85.0	668.7	169.68	14.28	24
Granodiorite, (Poland)	45.0	----	----	45	3.79	25
Granite, (Germany)	76.1	70.0	1465.4	181.61	15.29	26

Conclusion

The mean of specific activities of ^{226}Ra, ^{232}Th and ^{40}K were 4.98, 3.07, and 41.53 Bq/kg, respectively, which are about ten times less than the world average value (35, 30 and 400 Bq/kg). Moreover, the calculated radiological parameters were lower in comparison with the maximum permissible value. Therefore, using this kind of stone as a building material is better than igneous rocks. Moreover, its radiation is no hazard to the inhabitants of the building.

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