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Radiological Risk Measurements Due to Natural Radioactivity of Building Stones Used in Jordanian Houses

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Abstract: The radiological risk from building stone interfaces in Jordanian houses was determined depending on gamma ray spectrometric techniques. Building stone samples collected from seven types mostly used in Jordanian houses have been analyzed for the naturally occurring radioactive radionuclides. The mean specific activities of ²³⁸U, ²³²Th and ⁴⁰K were lower than 7.63±0.08, 2.77±0.12, 32.7±2.96 Bq/kg, respectively. The estimated radium equivalent activity (Ra_{eq}) in the stones was lower than 24.84±0.470 Bq/kg and the external and indoor hazard indices were also lower than unity. Moreover, different radiological hazardous parameters (the absorbed dose, the annual effective dose equivalent, the annual gonadal dose equivalent (AGDE), Excess Lifetime Cancer Risk ELCR and activity utilization index (AUI)) were calculated. The results were lower than those of published world average values. Also, the obtained values were comparable with the reported data of other building materials used in Jordan.

Keywords: Natural radioactivity, Gamma-ray spectrometry, Building stones, Hazard indices, Activity utilization index, Radiological risk.

Introduction

The effects of radiation sources on the world's population can be divided into natural and man-made effects, with dominant natural contribution from terrestrial and cosmic origins [1]. Natural radioactivity in building materials can be a main source of indoor radiation, either external or internal [2]. The former is caused by direct exposure to gamma radiation, while the latter is caused by inhalation of radon present in building materials. According to the World Health Organization, WHO, there is an association between indoor radon exposure and lung cancer, even at the relatively low concentration levels found in residential buildings [3]. For this reason, there is a growing need of controlling the use of materials derived from soils, such as phosphogypsum, cement, ceramic, granite or stones, in dwelling decoration, which cause an additional source of radiation exposure to people.

Many researchers found that natural radioactive nuclides, such as uranium (238 U), thorium (232 Th) and the radioactive isotope of potassium (40 K) in building materials originating from rocks and soils, have low-concentration amounts. A huge number of studies interested in the natural radioactivity of construction materials and those produced from industrial waste in

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different countries around the world are presented in literature, such as [4, 5, 6, 7, 8, 9], but none of them was concerned with building stones.

In Jordan, as in all other countries, there is a great interest in using various materials of rock origin as building materials. Most dwellings are being decorated from outside and inside with building materials. Recently, most buildings in all Jordanian governorates use building stones due to their rigidity and proper appearance despite of their type and source of origin in different locations in Jordan.

In view of this, there is a great need to measure the radionuclide concentrations in building materials to limit the health hazards and protect humans from environmental pollution.

Several studies have been made on the measurement of radon and natural radioactivity of the most common materials used in building construction in Jordan. Sharaf and Hamideen (2013) [10] measured the specific radioactivity of different materials used in Jordanian buildings. Also, Matiullah and Hussein (1998) [11] were engaged in the measurement of natural radioactivity of a large number of building materials of natural source and that of industrial sources used in the populated sites of Jordan. Meanwhile, Al-Jundi et al. (2009) [12] presented the indoor dose rates for a typical Jordanian concrete room, using Monte Carlo method. But, there is still a lack of regulations on the radioactivity of building materials for their safe usage.

This study was carried out to measure the ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K specific activities in seven main stones used in Jordanian houses, to compare them to the world average values for soil and to check that using these stones with other building materials does not surpass the limits of the allowed population exposure to radiation. For this purpose, collected samples from different types have been analyzed for natural radionuclides using gamma spectrometry. Also, the average radium equivalent activity, the external and indoor hazard indices, the total absorbed dose rate, the annual dose equivalent, the annual gonadal dose equivalent, Excess Lifetime Cancer Risk ELCR and activity utilization index (AUI) have been calculated in order to provide background database on the natural radioactivity levels and environmental pollution.

Materials and Methods

The stone samples were collected from seven types; namely, Ajlun, Ma'an, Basalt, Desert, Travenia, Hayan and Samic stones that are mostly used in Jordanian house interfaces. Ten samples from each type were collected, crushed, dried up in an oven at 130°C for 10 hours to avoid any moisture, pulverized to a fine powder and mixed to prepare homogenized representative samples for measurement. The powder samples were compressed in plastic taps 1.6 cm high and 7.8 cm in diameter, sealed to avoid any radon volatilization and weighed, then put away to reach radioactive equilibrium. After that, gamma analysis was performed. Sample preparation and radioactivity measurements were made in the Jordanian Atomic Energy Commission (JAEC) laboratories.

Gamma Spectrometry measurements to obtain the ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K specific activities were made using a high purity germanium (HPGe) detector (co-axial type) that was described in detail in a previous study [13]. The energy calibration and relative efficiency determination were carried out using a mixed source. The counting time for each sample and background was 60,000 s.

Activity Concentration Calculations

The natural radioactivity of the samples was determined using the count rate of each photopeak of the radionuclides ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K in the spectra detected by gamma spectroscopy. ²²⁶Ra specific activity was calculated in the samples using the peak of energy 186.3keV, ²³⁸U series by 351.9 keV peak of ²¹⁴Pb and 609.3 keV peak of ²¹⁴Bi, ²³²Th series by 911.0 keVpeak of ²²⁸Ac and 583.3 keV peak of ²⁰⁸Tl and the 1460 keV gamma-ray transition was used to determine the concentration of ⁴⁰K.

An accurate specific activity A_{Ei} (in Bq/kg), of a nuclide *i* for a peak at energy *E*, is given by the relation [14]:

$$A_{Ei} = \frac{N_{Ei}}{\varepsilon_E \times t \times f \times m_s}$$
(1)

where N_{Ei} is the net peak count inside a peak at energy E, ε_E is the detection efficiency at energy E, t is the counting live-time, f is the gamma ray yield per disintegration of the specific nuclide for a transition at energy E and m_s is the mass in kg of the measured sample. The peak activity Radiological Risk Measurements Due to Natural Radioactivity of Building Stones Used in Jordanian Houses

was averaged if there were more than one peak in the energy range of analysis and the result was the weighted average nuclide activity.

In order to prevent unnecessary exposure, radium equivalent activity (Ra_{eq}) and external hazard index (H_{ex}) were used to estimate radiation hazards. Radium equivalent activity (Ra_{eq}) was calculated using relation (2) assuming that 370 Bq/kg of ²²⁶Ra, 259 Bq/kg of ²³²Th and 4810 Bq/kg of ⁴⁰K produce the same gamma ray dose rate [1].

$$\left. \begin{array}{c} Ra_{eq} \left(Bq \, / \, kg \right) = \\ A_{226_{Ra}} + 1.43A_{232_{Th}} + 0.077A_{40_{K}} \end{array} \right\}$$
(2)

 $A_{226}{}_{Ra}$, $A_{232}{}_{Th}$, $A_{40}{}_{K}$ are the specific activities of 226 Ra, 232 Th and 40 K, respectively.

Based on the above assumption of activity world average values, the accepted radium equivalent activity (Ra_{eq}) world average value is better to be lower than 370 Bq/kg [15].

Radiological Risk Estimations

Krieger (1981) proposed a model for calculating the external hazard index, H_{ex} , assuming thick wall without any windows or doors, the external hazard index is given by the following relation [1]:

$$H_{ex} = \left(A_{226_{Ra}} / 370\right) + \left(A_{232_{Th}} / 259\right) + \left(A_{40_{K}} / 4810\right)\right\}$$
(3)

where $A_{226_{Ra}}$, $A_{232_{Th}}$, A_{40_K} are the specific activities of 226 Ra, 232 Th and 40 K, respectively.

The risk from ²²²Ra and its decay daughters to the internal respiratory systems is described by indoor hazard index, H_{in} , which must be less than 1.0 for safety requirements. In literature, a number of indices were suggested by researchers for indoor exposures as given in relations (4-7) [15, 16]:

$$H_{1} = \left(A_{226_{Ra}} / 185\right) + \left(A_{232_{Th}} / 259\right) + \left(A_{40_{K}} / 4810\right) \right\}$$
(4)

$$H_{2} = \left(A_{226_{Ra}} / 150\right) + \left(A_{232_{Th}} / 259\right) + \left(A_{40_{K}} / 4810\right)\right\}$$
(5)

$$H_{3} = \left(A_{226_{Ra}} / 1000\right) + \left(A_{232_{Th}} / 700\right) + \left(A_{40_{K}} / 10000\right)\right)$$
(6)

$$H_{4} = \left(A_{226_{Ra}} / 300\right) + \left(A_{232_{Th}} / 200\right) + \left(A_{40_{K}} / 3000\right)\right)$$
(7)

Also, $A_{^{226}Ra}$, $A_{^{232}Th}$, $A_{^{40}K}$ are the specific activities of $^{^{226}}$ Ra, $^{^{232}}$ Th and 40 K, respectively.

In order to calculate the radiological effects of any building material containing radionuclides, the absorbed dose rate D(nGy/h) in outdoor air at 1m above the ground surface in terms of ²³⁸U, ²³²Th and ⁴⁰K specific activities was calculated using the relation [17]:

$$D(nGy / h) = 0.427A_{238_U} + 0.662A_{232_{Th}} + 0.0432A_{40_K}$$
(8)

 $A_{^{238}U}$, $A_{^{232}Th}$, $A_{^{40}K}$ are the specific activities of $^{^{238}}$ U, $^{^{232}}$ Th and $^{^{40}}$ K, respectively.

The annual absorbed dose equivalent received by people can be estimated using the value of 0.7Sv/Gy as a conversion factor and 0.2 of the day for the occupancy factor [1]. The annual effective dose was calculated using the relation:

$$E\left(\mu Sv / y\right) = D\left(nGy / h\right) \times 24h \times$$

$$365.25d \times 0.2 \times 0.7Sv / Gy \times 10^{-3}$$
(9)

UNSCEAR (1988) considered the active bone marrow, the bone surface cells and the gonads, in addition to lung, breast and thyroid, as the organs of interest when humans work with radiation. One of the dosimetry models found in literature to evaluate the annual gonadal dose equivalent (AGDE) for a residential house considered as a cavity with infinitely thick walls built with a material of specific activities of radium, thorium and potassium is described by the relation [18,19]:

$$A GDE (\mu Sv / y) = 3.09 A_{226_{Ra}} + 4.18 A_{232_{Th}} + 0.314 A_{40_{K}}$$
 (10)

where $A_{226_{Ra}}$, $A_{232_{Th}}$, A_{40_K} are the specific activities of 226 Ra, 232 Th and 40 K, respectively. For a house containing the world average activity values of 226 Ra, 232 Th and 40 K in soil (35,

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30 and 400 *Bq/kg*, respectively), it will produce an AGDE of 359.15 μ *Sv/y*.

The excess lifetime cancer risk (ELCR) is another important radiological factor that is estimated as a consequence upon the evaluation of AEDE using the equation [20, 21]:

$$ELCR = E \times DL \times RF \tag{11}$$

where *E*, *DL* and *RF* are the annual effective dose equivalent, duration of life (70 years) and risk factor (0.05 Sv^{-1}), respectively. ICRP (2007) [20] defined the risk factor as fatal cancer risk per Sievert, which is assigned to a value of 0.05 for the public for stochastic effects [21].

The dose rates in indoor air according to the concentrations of different combinations of the three primordial radionuclides in soil samples used in construction materials are expressed by the activity utilization index (AUI). AUI is calculated from the following equation [22]:

$$A UI = \begin{bmatrix} (A_{226_{Ra}} / 50) f_u + (A_{232_{Th}} / 50) f_{Th} \\ + (A_{40_K} / 500) f_K \end{bmatrix} W_m$$
(12)

where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively and $f_{\rm K}$ (0.041), $f_{\rm Th}$ (0.604) and $f_{\rm U}$ (0.462)are the respective fractional contributions from the actual activities of these radionuclides to the total gamma radiation dose rate in air. w_m is a weighted factor for mass proportion of the building materials in the dwelling with the characteristic activity. Applying the appropriate conversion factor $(w_m=I)$ along with the activity concentrations of the respective radionuclides implies that all building materials used in a house are composed of this specific material. Typical activities per unit mass of 40 K, 232 Th and 226 Ra in soils A_K, A_{Th}

and A_{Ra} are reported to be 500, 50 and 50 Bq/kg, respectively [23], which gives a full utilization (AUI) of 0.5634.

Results and Discussion

The specific activities of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K in the collected stone samples are listed in Table.1. The results show very low values and most of them were under the minimum detection limits of the detector. Ajlun stones were highest characterized by the activity concentrations for all measured ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides. ²²⁶Ra specific activities were under 17.5 Bq/kg in almost all of the sample types, except in Ajlun stones, where the specific activity was 23.5 ± 2.9 Bq/kg. ²³⁸U specific activity ranged from 7.0 Bq/kg in five types to 10.6 + 1.9 Bq/kg in Ma'an stones. Meanwhile, the specific activity of ²³²Th ranged from 1.4 Bq/kg in four types to 6.3 + 0.4 Bq/kg in Basalt stones and ⁴⁰K specific activity ranged from 12.5 Bq/kg in five types to 147.5+6.2 Bq/kg in Ajlun stones. The geological locations and the geochemical characteristics of the parent rocks from which the stones were collected explain the small variation in these natural radioactivity levels [24].

The average activities of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K were found to be lower than 18.36, 7.63, 2.77 and 32.7Bq/kg, respectively. And the world average specific activities of the radionuclides ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K in all soil samples have averages of 35, 35, 30 and 400 Bq/kg, respectively [1]. Fig.1 shows the variation of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K specific activities in different stone types. The radionuclides ²²⁶Ra, ²³⁸U and ²³²Th show a uniform distribution that is lower than the accepted world average values. Meanwhile, ⁴⁰K shows an abnormal distribution in Ajlun stones depending on the geological structure of the original rocks.

TABLE 1. Natural radioactivity in Ajlun, Ma'an, Basalt, Desert, Travenia, Hayan and Samic stone types (in Bq/kg) using gamma spectrometry.

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	Stone Type	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K
	Ajlun	23.5 <u>+</u> 2.9	7.8 <u>+</u> 2.4	5.0 <u>+</u> 0.4	147.5 <u>+</u> 6.2
	Ma'an	< 17.5	10.6 <u>+</u> 1.9	< 1.4	< 12.5
	Basalt	< 17.5	< 7.0	6.3 <u>+</u> 0.4	18.9 <u>+</u> 6.8
	Desert	< 17.5	< 7.0	2.5 ± 0.5	< 12.5
	Travenia	< 17.5	< 7.0	< 1.4	< 12.5
	Hayan	< 17.5	< 7.0	< 1.4	< 12.5
	Samic	< 17.5	< 7.0	< 1.4	< 12.5
	Mean	<18.36±0.13	$< 7.63 \pm 0.08$	< 2.77±0.12	< 32.7±2.96



FIG. 1. Variation of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K specific activities in Ajlun, Ma'an, Basalt, Desert, Travenia, Hayan and Samic stone types.

All calculated values of radium equivalent Ra_{eq} (Bq/kg), external hazard index (H_{ex}) and indoor hazard indices (H₁, H₂, H₃, H₄) of Ajlun, Ma'an, Basalt, Desert, Travenia, Hayan and Samic stone types were tabulated in Table.2. The calculated Raeg results ranged from 20.46 Bq/kg in three types to 42.01 Bq/kg in Ajlun stones and the average value was less than (24.84 ± 0.470) Bq/kg which is lower than the recommended maximum value of 370 Bq/kg [15]. The estimated external hazard indices in the samples were 0.06 in four of the seven types and the maximum value was 0.11 with an average value for all types of 0.07 ± 0.001 , which is much lower than the safety recommended limit of unity. Also, the internal hazard indices show the same behavior, since their entire calculated values were very low relative to unity [1].

Table 3 presents all calculated values of absorbed dose rate D in air, annual effective dose equivalent E, annual gonadal dose equivalent AGDE, Excess Lifetime Cancer Risk ELCR and activity utilization index (AUI) in Ajlun, Ma'an, Basalt, Desert, Travenia, Hayan and Samic stone

types. The mean values of the calculated absorbed dose rate D in air, annual effective dose equivalent E and annual gonadal dose equivalent AGDE were (6.50±0.18) nGy/h, (7.98±0.22) μ Sv/y and (78.57 \pm 1.65) μ Sv/y, respectively. A general overview of the previous results indicates that all of the calculated parameters have average values below the safety limits recommended of 57 nGy/h, 70 μ Sv/y and 359.15 μ Sv/y for the previous parameters, respectively [1]. Also, the estimated value of Excess Lifetime Cancer Risk ELCR ranged from 19.14×10^{-6} to 55.89×10^{-6} with a mean value of 27.94×10^{-6} , which is below the world average value of 290×10^{-6} for soils [1]. This means that the excess risk of cancer among the population living inside these decorated houses due to using the building stones is insignificant. Finally, the activity utilization index (AUI) values ranged from 0.16 to 0.24 with a mean value of 0.18. This means that even for full utilization of the stones in building; i.e., $w_m = I$, the associated AUI was much lower that the recommended limit of 0.5634.

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ΤÆ	ABLE 2.	. Radium	equival	ent Ra _{eq} ,	external	hazard	index	(H_{ex})	and	indoor	hazard	indices	$(H_1,$	H ₂ ,	H3,
	H_4) in A	Ajlun, Ma	a'an, Bas	salt, Dese	ert, Travo	enia, Ha	ayan an	id Sai	mic s	stone typ	pes.				

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Stone Type	$Ra_{eq}(Bq/kg)$	H_{ex}	H_{l}	H_2	H_3	H_4		
Ajlun	42.01	0.11	0.177	0.207	< 0.045	< 0.153		
Ma'an	< 20.46	< 0.06	< 0.103	< 0.125	< 0.021	< 0.069		
Basalt	< 27.96	< 0.08	< 0.123	< 0.145	< 0.028	< 0.096		
Desert	< 22.04	< 0.06	< 0.107	< 0.129	< 0.022	< 0.075		
Travenia	< 20.46	< 0.06	< 0.103	< 0.125	< 0.021	< 0.069		
Hayan	< 20.46	< 0.06	< 0.103	< 0.125	< 0.021	< 0.069		
Samic	< 20.46	< 0.06	< 0.103	< 0.125	< 0.021	< 0.069		
Mean	<24.84±0.470	<0.07±0.001	<0.12±0.002	<0.14±0.002	< 0.026 ± 0.001	<0.086±0.001		

TABLE 3. Absorbed dose rate D in air, annual effective dose equivalent E, annual gonadal dose equivalent AGDE, Excess Lifetime Cancer Risk ELCR and activity utilization index (AUI) in Ajlun, Ma'an, Basalt, Desert, Travenia, Hayan and Samic stone types.

Stone Type	D(nGy/h)	$E(\mu Sv/y)$	AGDE(µSv/y)	ELCR ($\times 10^{-6}$)	AUI
Ajlun	13.01	15.97	139.83	55.89	0.24
Ma'an	< 5.99	< 7.35	< 63.85	< 25.74	< 0.16
Basalt	< 7.98	< 9.79	< 86.34	< 34.26	< 0.17
Desert	< 5.18	< 6.36	< 68.45	< 22.27	< 0.17
Travenia	< 4.46	< 5.47	< 63.85	< 19.14	< 0.16
Hayan	< 4.46	< 5.47	< 63.85	< 19.14	< 0.16
Samic	< 4.46	< 5.47	< 63.85	< 19.14	< 0.16
Mean	$< 6.50 \pm 0.18$	< 7.98±0.22	< 78.57±1.65	< 27.94±5.09	$< 0.18 \pm 0.01$

In literature, Matiullah et al. [11] found that the radium equivalent activity of Ajloun stones was 109.4 Bq/kg and of Ma'an stones was 88.1Bq/kg. Meanwhile, Al-Jundi et al. [25] estimated the radium equivalent activity of Desert stone to be 70.7Bq/kg, Halabat stones 53.2 Bq/kg and Ajloun stones 64.7 Bq/kg. A comparison between the calculated radium equivalents with other building constructions

used in Jordanian buildings is given in Fig.2. It is shown that Ra_{eq} due to the building stones was lower than the measured values of 123.20, 117.90, 86.22 and 54.96 Bq/kg for fine aggregates, coarse aggregates, sand and marble, respectively [25]. Also, it is lower than the average value of 79.9 Bq/kg measured for the general Jordanian building materials [11].



FIG. 2. Comparison of radium equivalent Ra_{eq} in the stone types with other materials used in Jordanian buildings.

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Conclusion

Seven kinds of building stones used in Jordanian building constructions, considered as the most popular ones used, were measured for their natural radioactivity in order to determine their radiological impact when they are used as building materials. The specific activities of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K activity levels in the stone samples were lower than the world average values and the corresponding values in other construction materials used in Jordanian buildings. The corresponding external hazard index, indoor hazard indices, absorbed dose rate in air, annual effective dose equivalent, gonadal dose equivalent, Excess Lifetime Cancer Risk ELCR and activity utilization index (AUI) don't

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exceed significantly the average dose limit for all stone types. The results obtained indicate no significant radiological hazards arising from using such stones in decoration of building interfaces, since the health effects due to natural radiation are very low.

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