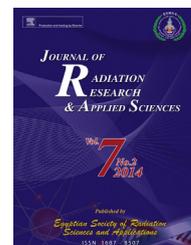


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Assessment of natural radiation exposure and radon exhalation rate in various samples of Egyptian building materials

M.Y. Shoeib^{a,*}, K.M. Thabayneh^b^aBasic Science Department, Modern Academy for Engineering and Technology in Maadi, Cairo, Egypt^bFaculty of Sciences and Technology, Hebron University, PO.Box40, Hebron, Palestine

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ABSTRACT

The aim of this investigation was to determine the amount of γ -decay of several building materials used in Egypt, in terms of Bq kg^{-1} , and to calculate the radiological effect caused by this radioactivity. Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in 30 samples of manufactured building materials were measured using gamma-spectroscopy system based on high-purity germanium detector with an efficiency of 40 %. The activity concentrations for ^{226}Ra , ^{232}Th , and ^{40}K , from the selected building materials, ranged from $(8.15 \pm 2.81$ to $288.5 \pm 17.49 \text{ Bq kg}^{-1})$, $(3.59 \pm 1.36$ to $77.77 \pm 15.61 \text{ Bq kg}^{-1})$ and $(4.09 \pm 4.72$ to $1314 \pm 15.30 \text{ Bq kg}^{-1})$, respectively. Radium equivalent activities, absorbed dose rate, Excess lifetime cancer risk and the values of hazard indexes were calculated for the measured samples to assess the radiation hazards arising from using those materials in the construction of dwellings. These results show that annual dose absorbed by inhabitants from construction materials used in Egypt (except cement bricks) are below 1.0 mSv y^{-1} . Therefore, the types used in the current study are quite safe to be used as building materials, except the cement brick, granite and ceramic samples which are critical points for safety in construction. Finally, the so-called can technique has been used to measure radium content and exhalation rates of radon in these building materials samples. Positive correlation was found between radium concentration and radon exhalation rates.

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1. Introduction

Man is continuously exposed to ionizing radiation from naturally occurring radioactive materials (NORM). Natural

radioactivity is widely spread in the earth's environment and it exists in various geological formation e.g. soils, rocks, plants, water, air and in building materials (Ramassay, Dheenathayalu, Ravishankar, & Ponnusamy, 2004; Rati et al., 2010). Measurement of activity concentrations of

* Corresponding author. Tel.: +20201270199267 (mobile).

E-mail addresses: drmarwashoeib@hotmail.com (M.Y. Shoeib), drkaleelt@yahoo.com (K.M. Thabayneh).

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radionuclides in building materials is important in the assessment of population exposures, as most individuals spend 80% of their time indoors (Merle & Enn, 2012). Naturally occurring radionuclides in building materials are source of external radiation exposure in dwellings. This radiation is caused by gamma radiation originating from the uranium and thorium series and from ^{40}K (Amrani & Tahtat, 2001).

The population-weighted average of indoor absorbed dose rate in air from terrestrial source of radioactivity is estimated to be 84 nGy h^{-1} (UNSCEAR, 2000). The worldwide average indoor effective dose due to gamma ray from building materials is estimated to be about 0.4 mSv y^{-1} (UNSCEAR, 1977, 1993). The average activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in the earth's crust are 35, 30, and 400 Bq kg^{-1} , respectively. However, elevated levels of natural radionuclides causing annual doses of several mSv have been identified in some regions around the world, e.g. in Brazil, France, India, Nigeria, Iran (UNSCEAR, 2000, 1977, 1993). This external radiation exposure, caused by gamma emitting radionuclides in building materials, can be assessed either by direct exposure measurements in the existing buildings or by radionuclide analyses of building materials with the dose rate modeling (Merle & Enn, 2012).

External gamma dose estimation due to the terrestrial sources is essential as these doses vary depending upon the concentrations of the natural radionuclides, ^{226}Ra , ^{232}Th , and their daughter products and ^{40}K , present in soils and rocks, which further depend upon the local geology of each region in the world (Rati et al., 2010). Radium is present everywhere in the earth's crust so radon is found everywhere in varying quantities. It can move freely from the place of its origin through pores in soil and cracks in walls. Radon transportation is mainly due to diffusion and forced flow (Shakir Khan, Naqvi, Azam, & Srivatsava, 2011). Radon is an alpha-emitting radioactive gas. It is a daughter product of ^{226}Ra and decay with a half-life of 3.82 days emitting alpha particles of energy 5.49 Mev. The radioactive daughter product of radon via ^{218}Po and ^{214}Po emit alpha particles. These daughter products are solid and have a tendency to attach themselves to aerosols in ambient air. When we breathe or inhale radon and its daughter product along with the normal air, most of the radon is exhaled, its daughter products get logged to the inner walls and membranes of our respiratory system and continue causing constant damage due to their alpha activity (Khan, Tariq, & Rawat, 2012).

Lung cancer, skin cancer and kidney diseases are the hazards caused by the inhalation of radon decay products. The radiological impact caused by nuclides is due to radiation exposure of the body by the gamma rays and irradiation of the lung tissues from inhalation of radon and its progeny. Therefore, keeping in view the natural risk, it is necessary to know the dose limits of public exposure (Nain, Chauhan, & Chakravarti, 2006). Indoor radon exposure has become a problem all over the world due to the fact that it accounts for approximately 60% of the total natural background radiation (UNSCEAR, 1993). Radon concentration measurements are nowadays routinely performed and different laboratories around the world have developed several types of radon radiation detectors. The suitable choice of detector depends on several factors: study purpose, sensitivity, cost, ect (Nidal, Ghassan, Mousa, & Toshiyuki, 2007).

The technique of track etch is widely applied in Europe for measuring the total indoor radon level. Kodalpha film type (LR-115) radon dosimeter is a small, black box of dimensions $4 \times 7.5 \times 0.5 \text{ cm}$. The radon-sensitive part, which is the actual dosimeter, is a small film badge which is housed on the inside section of the hinged lid of the dosimeter. These film badges are LR-115 type nuclear track films produced by KODAK and they consist of a $100 \mu\text{m}$ thick polyester substrate that is coated with a $12 \mu\text{m}$ thick layer of red colored cellulose nitrate. Kodalpha film type (LR-115) has been used for radon measurement by many laboratories throughout the world and by most important radiation safety institutes like the United States of America-Environmental Protection Agency (USA-EPA) and United Kingdom-National Radiological Protection Board (UK-NRPB). Kodalpha film type (LR-115) has several characteristics: (1) very sensitive to alpha particles only; (2) can be used for short-term measurement at minimum 10–30 days of exposure and also for long-term measurement, 3 months up to 1 year; (3) insensitive to environmental changes such as humidity, water and temperature up to $60 \text{ }^\circ\text{C}$; (4) suitable to be used for radon measurements in stagnant or flowing water and in oil (Nidal et al., 2007).

2. Materials and methods

2.1. Sample collection

For this study, thirty different samples of Egyptian building materials were collected. Mostly, the sample selection consisted of the commonly available materials, which were obtained from the building material stores. All samples were crushed into grains, dried, homogenized, and put into PVC Marenilli with capacity of hundreds cm^3 , the average samples volume around 100 cm^3 and their masses vary from 77.45 to 151.37 g. The samples were sealed for 4 weeks to reach secular equilibrium between ^{226}Ra and ^{232}Th with their decay products.

2.2. Spectroscopic analysis

The radionuclide activity concentrations in the prepared samples were measured using an n-type coaxial high-purity germanium (HPGe) detector, Canberra model in Gamma Irradiation Unit – Nuclear Research Center – Atomic Energy Authority – Egypt. The detector has an efficiency of about 40%, energy resolution of 1.9 keV, full width at the half maximum (FWHM) for the 1332.3 keV gamma line of ^{60}Co and MCA with 8000 channels. An empty Marenilli beaker was used for the same period of time to assess the background concentrations of the γ -rays. To reduce the γ -rays background from building and cosmic rays, a cylindrical lead shield of 100 mm thickness is used to shield the detector from the surroundings environment. This shield is composed of three inner concentric shells of lead, cadmium and copper.

Radon concentration and exhalation rate was measurements were done for the collected samples using the “Can technique” (Abu-Jarad, 1988). For the measurement of exhalation rate, 100 cm^3 of samples were taken. Fine quality of samples obtained by using a scientific sieve of $150 \mu\text{m}$ mesh

size and then dried in oven at 50 °C for twenty four hours so that moisture can be removed from samples then packed and sealed in impermeable air tight polyvinyl chloride (PVC) container to prevent the escape of radon gas. LR-115 type II foils were used in this study cut into 2 × 2 cm². Foils were exposed for a stipulated period of 150 days (long term exposure). Samples were retrieved and chemically etched in 2.5 N of NaOH (sodium hydroxide) solutions at 60 °C temperature for about 1.5 h for developing the tracks registered in the films. After etching, the detector was washed in distilled water, dipped for a few seconds in 3% acetic acid solution, washed again and dried in hot air. Using an optical microscope at 400× objective lens, the number of tracks in 30 fields was scanned for each detector to determine the track density per m³. The tracks were counted and the concentrations of both radon and its progeny were determined according to [Thabayneh \(2013\)](#).

3. Results and discussion

3.1. Radium, thorium and potassium in various samples

The concentrations of radium, thorium and potassium were calculated using the following equation ([Rati et al., 2010](#)):

$$\text{Activity (Bq)} = \frac{\text{CPS} \times 100 \times 100}{I \times \xi_{ff}} \pm \frac{\text{CPS}_{\text{error}} \times 100 \times 100}{I \times \xi_{ff}} \quad (1)$$

where CPS is counts per second; I is the intensity and ξ_{ff} is the efficiency of the detector.

[Table 1](#) presents the average concentration of radionuclides, ²²⁶Ra, ²³²Th and ⁴⁰K. The concentration for ²²⁶Ra, ²³²Th and ⁴⁰K varies from 8.15 ± 2.81 to 288.5 ± 17.49 Bq kg⁻¹, 3.59 ± 1.36 to 77.77 ± 15.61 Bq kg⁻¹ and 4.09 ± 4.72 to 1314 ± 15.30 Bq kg⁻¹, respectively in building material samples used in this study. The activity found for ²²⁶Ra is the lowest for gypsum and maximum for cement bricks, ²³²Th concentration found for cement plaster is the lowest and cement brick is the highest, ⁴⁰K concentration found in granite is the maximum and in white cement it is the lowest. The samples showed that the specific activity values of natural source nuclear materials, ²²⁶Ra, ²³²Th and ⁴⁰K were within the permissible limits 35, 30 and 370 Bq kg⁻¹ respectively ([UNSCEAR, 2000](#)), except samples of cement brick, cement, ceramic and cement plaster where the specific activity values of ²²⁶Ra were 288.5 ± 17.49, 44.61 ± 2.83, 51.12 ± 2.74 and 50.61 ± 2.48 Bq kg⁻¹. The specific activity values ²³²Th was higher than the permissible limits for

samples of cement brick, granite and ceramic with values of 77.77 ± 15.61, 47.76 ± 1.89 and 40.52 ± 2.5 Bq kg⁻¹ respectively. Also the specific activity values ⁴⁰K were higher than the permissible limits for samples of red brick, cement brick, granite and ceramic with values of 447.84 ± 10.16, 909.5 ± 59.73, 1314.82 ± 15.30 and 682.60 ± 10.13 Bq kg⁻¹ respectively. For the sake of comparison, the levels of the natural radionuclides in the building material samples of various other countries are exhibited in [Table 2](#). Most of the results obtained in the present study are within the international values measured in many countries, while some of the results are higher.

3.2. Calculation of radium equivalent

The radium equivalent index, Ra_{eq} is generally introduced as the weighed sum of ²²⁶Ra, ²³²Th and ⁴⁰K activities based on the assumption that 10 Bq kg⁻¹ of ²²⁶Ra, 7 Bq kg⁻¹ of ²³²Th and 130 Bq kg⁻¹ of ⁴⁰K will produce the same dose rates of gamma rays. Values of Ra_{eq} were calculated using the equation ([Bereka & Mathew, 1985](#); [Thabayneh, 2013](#)):

$$Ra_{eq} (\text{Bq kg}^{-1}) = C_{Ra} + (C_{Th} \times 1.43) + (C_K \times 0.077) \quad (2)$$

where C_{Ra} , C_{Th} and C_K are the activity concentrations (Bq kg⁻¹) of radium, thorium and potassium in the samples. [Table 3](#), represents the radium equivalent values which were found to vary from 26.04 to 469.74 Bq kg⁻¹. The lowest value was found in gypsum while the maximum value was found in cement brick. The radium equivalent values for the analyzed samples are less than the international average value 370 Bq kg⁻¹ ([UNSCEAR, 2000](#)) except the sample of cement brick, while the value is 469.74 Bq kg⁻¹.

3.3. Estimation of absorbed dose rate

A quantity, while considering the radiation risk to human and other, is absorbed dose rate. Assessment of gamma radiation hazard to human associated with the building materials can be done by calculating the different radiation hazard indices ([Rati et al., 2010](#)). [Table 3](#), presents the absorbed dose rate D (nGy h⁻¹) due to terrestrial gamma rays at one meter above the ground level can be estimated by the concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K by applying the factors 0.462, 0.604 and 0.0417 for radium, thorium and potassium, respectively ([El-Shershaby, El-Bahy, Walley El-Din, & Dabayneh, 2006](#); [Thabayneh, 2013](#)).

Table 1 – The average activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclides in some building material samples used in Egypt.

| Sample type | No. of samples | ²²⁶ Ra (Bq kg ⁻¹) | ²³² Th (Bq kg ⁻¹) | ⁴⁰ K (Bq kg ⁻¹) |
|----------------|----------------|--|--|--|
| Red-brick | 3 | 23.06 ± 2.60 | 23.11 ± 2.99 | 447.84 ± 10.16 |
| Sand | 3 | 16.63 ± 1.54 | 13.36 ± 1.58 | 118.9 ± 4.79 |
| Cement bricks | 3 | 288.5 ± 17.49 | 77.77 ± 15.61 | 909.5 ± 59.73 |
| Cement | 3 | 44.61 ± 2.83 | 10.39 ± 1.54 | 51.12 ± 5.96 |
| White cement | 3 | 17.45 ± 2.33 | 8.44 ± 1.49 | 4.09 ± 4.72 |
| Gypsum | 3 | 8.15 ± 2.81 | 7.93 ± 2.90 | 85.1 ± 10.01 |
| Granite | 3 | 32.46 ± 1.56 | 47.76 ± 1.89 | 1314.82 ± 15.30 |
| Ceramic | 3 | 51.12 ± 2.74 | 40.52 ± 2.54 | 682.6 ± 10.13 |
| Cement plaster | 3 | 50.61 ± 2.48 | 3.59 ± 1.36 | 57.57 ± 5.61 |
| Hydrated lime | 3 | 16.14 ± 1.97 | 5.33 ± 2.00 | 70.22 ± 6.00 |

Table 2 – Comparison between the activity concentrations of our building materials with that of other countries of the World.

| Sample type | Country | C _{Ra} (Bq kg ⁻¹) | C _{Th} (Bq kg ⁻¹) | C _K (Bq kg ⁻¹) | Ref. |
|--------------|---------------|--|--|---------------------------------------|---------------------------|
| Red-brick | Palestine | 32.8 | 7.2 | 1139 | [Dabayneh, 2007] |
| | Algeria | 65.0 | 51.0 | 675 | [Amrani and Tahtat, 2001] |
| | Cameroon | 49.6 | 91.0 | 172 | [Ngachin et al., 2007] |
| | Hong Kong | 143.0 | 158.0 | 850 | [Dabayneh, 2007] |
| | Albania | 33.4 | 42.0 | 644 | [Xhixha et al., 2013] |
| | Egypt | 23.06 | 23.11 | 447.84 | Present study |
| Sand | Palestine | 20.6 | 18.8 | 26.3 | [Dabayneh, 2007] |
| | Algeria | 12.0 | 7.0 | 74.0 | [Amrani and Tahtat, 2001] |
| | Saudi Arabia | 12.3 | 19.7 | 260 | [El-Taher, 2012] |
| | Brazil | 14.3 | 18.0 | 807 | [Dabayneh, 2007] |
| | Cameroon | 14.0 | 31.0 | 586 | [Ngachin et al., 2007] |
| | Hong Kong | 24.3 | 27.1 | 841 | [Dabayneh, 2007] |
| | Cuba | 16.7 | 15.6 | 188 | [Dabayneh, 2007] |
| | Egypt | 16.63 | 13.36 | 118.9 | Present study |
| | Palestine | 85.1 | 67.6 | 18.5 | [Dabayneh, 2007] |
| Cement | Algeria | 41.0 | 27.0 | 422 | [Amrani and Tahtat, 2001] |
| | Saudi Arabia | 38.4 | 45.3 | 86 | [El-Taher, 2012] |
| | Brazil | 61.7 | 58.5 | 564 | [Dabayneh, 2007] |
| | Cameroon | 27.0 | 15.0 | 277 | [Ngachin et al., 2007] |
| | Hong Kong | 19.2 | 18.9 | 127 | [Dabayneh, 2007] |
| | Cuba | 22.8 | 10.6 | 467 | [Dabayneh, 2007] |
| | Albania | 55.0 | 17.0 | 179.7 | [Xhixha et al., 2013] |
| | Egypt | 44.61 | 10.39 | 51.12 | Present study |
| | Palestine | 29.8 | 28.9 | 31.5 | [Dabayneh, 2007] |
| | Egypt(Qena) | 72.0 | 46.0 | 250 | [Dabayneh, 2007] |
| White cement | Cuba | 44.5 | 21.8 | 99.0 | [Dabayneh, 2007] |
| | Egypt | 17.45 | 8.44 | 4.09 | Present study |
| | Palestine | 78.2 | 9.8 | 2.7 | [Dabayneh, 2007] |
| | Saudi Arabia | 33.3 | 47.2 | 88 | [El-Taher, 2012] |
| | Brazil | 6.3 | – | 18.1 | [Dabayneh, 2007] |
| Gypsum | Italy | 6.0 | 2.0 | 12 | [El-Taher, 2012] |
| | Egypt | 8.15 | 7.93 | 85.1 | Present study |
| | Palestine | 35.1 | 20.5 | 639.5 | [Dabayneh, 2007] |
| | Saudi Arabia | 23.0 | 30.0 | 340 | [El-Taher, 2012] |
| | Brazil | 48.6 | 288.2 | 1335 | [Dabayneh, 2007] |
| | Hong Kong | 202.0 | 140.0 | 1030 | [Dabayneh, 2007] |
| Granite | France | 90.0 | 80.0 | 1200 | [El-Taher, 2012] |
| | Egypt | 32.46 | 47.76 | 1314.82 | Present study |
| | Palestine | 73.7 | 58.2 | 624 | [Dabayneh, 2008] |
| | Egypt (Qena) | 126.0 | 72.0 | 300 | [Dabayneh, 2008] |
| | Algeria | 55.0 | 41.0 | 410 | [Amrani and Tahtat, 2001] |
| | Egypt | 51.12 | 40.52 | 682.6 | Present study |
| | Palestine | 58.2 | 78.6 | 2.9 | [Dabayneh, 2007] |
| Ceramic | Algeria | 16.0 | 13.0 | 36 | [Amrani and Tahtat, 2001] |
| | Saudi Arabia | 28.6 | 49.2 | 66 | [El-Taher, 2012] |
| | Brazil | 24.3 | 7.0 | 205 | [Dabayneh, 2007] |
| | Egypt | 16.14 | 5.33 | 70.22 | Present study |
| | Hydrated lime | | | | |

$$D(\text{nGy h}^{-1}) = (0.462 C_{Ra} + 0.621C_{Th} + 0.0417 C_K) \tag{3}$$

As shown in Table 3, the absorbed dose rate values were found to vary from 12.24 to 219.51 nGy h⁻¹. The lowest value was found in gypsum while the maximum value was found for cement brick. The absorbed dose rate values are below the permissible level 55 nGy h⁻¹ (UNSCEAR, 2000) except the samples of cement brick, granite and ceramic, where the values are 219.51, 99.48 and 77.24 nGy h⁻¹ respectively.

3.4. External hazard index

Consideration of external radiation exposure is usually associated with gamma radiation emitted by radionuclides of

concern. The value of H_{ex} should be below one to be sure of the safe use of building materials, which corresponds to the upper limit of R_{eq} (370 Bq kg⁻¹) and to keep the radiation hazard insignificant (Rati et al., 2010):

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \tag{4}$$

As shown in Table 3, the values of external hazard index have been found to be lower for gypsum 0.07; which is under permissible limits and 1.27 for cement brick which is higher than permissible limits (UNSCEAR, 1993). The external hazard index for all samples is under the permissible limits except sample of cement brick.

Table 3 – Radium equivalent (Ra_{eq}), absorbed dose rate (D), health hazard index (H_{ex}), radioactivity level index (I_γ) and alpha level index (I_α) for different building material samples.

| Sample type | Ra_{eq} (Bq kg ⁻¹) | D (nGy h ⁻¹) | (H_{ex}) | (I_γ) | (I_α) |
|----------------|----------------------------------|----------------------------|------------|--------------|--------------|
| Red-brick | 90.59 | 43.68 | 0.24 | 0.68 | 0.12 |
| Sand | 44.89 | 20.94 | 0.12 | 0.32 | 0.08 |
| Cement bricks | 469.74 | 219.51 | 1.27 | 3.31 | 1.44 |
| Cement | 63.40 | 29.19 | 0.17 | 0.44 | 0.22 |
| White cement | 29.83 | 13.47 | 0.08 | 0.20 | 0.09 |
| Gypsum | 26.04 | 12.24 | 0.07 | 0.19 | 0.04 |
| Granite | 202.00 | 99.48 | 0.55 | 1.57 | 0.16 |
| Ceramic | 161.62 | 77.24 | 0.44 | 1.20 | 0.26 |
| Cement plaster | 60.18 | 28.01 | 0.16 | 0.41 | 0.25 |
| Hydrated lime | 29.17 | 13.69 | 0.08 | 0.21 | 0.08 |

3.5. Radioactivity level index

The gamma radiation hazardous levels associated with the natural radionuclides in the building material samples were assessed by means of radioactivity level index, I_γ . According to the European Commission guidelines, the representative level of I_γ values was estimated according to the following equation (Beretka & Mathew, 1985; Dabayneh, Mashal, & Hasan, 2008; Thabayneh, 2013):

$$I_\gamma = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \quad (5)$$

Table 3, represents the radioactivity level index, where the lowest value 0.19 was found in gypsum while the maximum value 3.31 was found in cement brick. The radioactivity level index for all studied samples is below the permissible level except samples of cement brick, granite and ceramic were higher than the permissible level (UNSCEAR, 1993). If external hazard exceeds unity, we conclude that external doses exposed individuals will exceed the acceptable levels. The radioactivity level index values for cement brick, granite and ceramic were 3.31, 1.57 and 1.20 respectively.

3.6. Alpha index

The excess alpha radiation due to the radon inhalation originating from the building materials is assessed through the alpha index (I_α) which is defined as follows (Rafique et al., 2011):

$$I_\alpha = \frac{C_{Ra}}{200} \quad (6)$$

The recommended exemption and recommended upper levels of ²²⁶Ra concentrations in building materials are 100 Bq kg⁻¹ and 200 Bq kg⁻¹. When the ²²⁶Ra activity concentration of building materials exceeds the value of 200 Bq kg⁻¹, it is possible that radon exhalation from this material may cause indoor radon concentration greater than 200 Bq m⁻³. On the other hand, if ²²⁶Ra concentration is less than 100 Bq kg⁻¹, then resulting indoor radon concentration is less than 200 Bq m⁻³. These considerations are reflected in the alpha index. The recommended limit concentration of ²²⁶Ra is 200 Bq kg⁻¹, for which $I_\alpha = 1$ (Rafique et al., 2011). Table 3, represents the alpha index I_α where the values range from 0.04 to 1.44. These observed values are less than unity showing that construction materials are safe from the point of view of environmental radiation hazards, except the sample of cement brick, where $I_\alpha = 1.44$.

3.7. Annual effective dose rates

To estimate annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose 0.7 Sv Gy⁻¹ and the indoor occupancy factor of 0.2 proposed by UNSCEAR, 2000 were calculated (UNSCEAR, 2000). The annual effective doses are determined as follows (Rati et al., 2010):

$$\text{Indoor (mSv y}^{-1}\text{)} = \text{Absorbed dose (mGy h}^{-1}\text{)} \times 24 \text{ h} \times 365.25 \text{ d} \\ \times 1.4 \times 0.8 \times 0.7 (\text{Sv Gy}^{-1}) \times 10^{-6} \quad (6)$$

and

$$\text{Outdoor (mSv y}^{-1}\text{)} = \text{Absorbed dose (mGy h}^{-1}\text{)} \times 24 \text{ h} \times 365.25 \text{ d} \\ \times 0.2 \times 0.7 (\text{Sv Gy}^{-1}) \times 10^{-6} \quad (7)$$

The results of indoor, outdoor and total annual effective dose rates for Egyptian building material samples are listed in Table 4. It can be seen that the total values for each sample are less than the corresponding worldwide value of 1 mSv y⁻¹, except the sample of cement brick, where $D_{tot} = 1.78$ mSv y⁻¹. Therefore, the building materials used in the current study are quite safe to be used as building materials.

3.8. Total gamma radiation dose rate

The total gamma radiations dose rate (D_γ) is modified to include the contributions from natural radionuclides, ¹³⁷Cs

Table 4 – Indoor (D_{in}) and outdoor (D_{ou}) annual effective dose equivalent, total annual effective dose equivalent (D_{tot}), total gamma radiation dose rate (D_γ) and the excess lifetime cancer risk (ELCR) for different building material samples.

| Sample type | D_{in} (mSv y ⁻¹) | D_{ou} (mSv y ⁻¹) | D_{tot} (mSv y ⁻¹) | D_γ (nGy h ⁻¹) | ELCR ($\times 10^{-3}$) |
|----------------|---------------------------------|---------------------------------|----------------------------------|-----------------------------------|---------------------------|
| Red-brick | 0.30 | 0.05 | 0.35 | 78.40 | 1.259 |
| Sand | 0.14 | 0.03 | 0.17 | 55.06 | 0.597 |
| Cement bricks | 1.51 | 0.27 | 1.78 | 247.78 | 6.061 |
| Cement | 0.20 | 0.04 | 0.24 | 62.12 | 0.797 |
| White cement | 0.09 | 0.02 | 0.11 | 47.21 | 0.375 |
| Gypsum | 0.08 | 0.02 | 0.10 | 46.39 | 0.351 |
| Granite | 0.68 | 0.12 | 0.8 | 136.01 | 2.892 |
| Ceramic | 0.53 | 0.09 | 0.62 | 112.00 | 2.211 |
| Cement plaster | 0.19 | 0.03 | 0.22 | 60.46 | 0.75 |
| Hydrated lime | 0.09 | 0.02 | 0.11 | 47.44 | 0.381 |

Table 5 – Radon activity (C_{Rn}), radon surface exhalation rate (E_s) and mass exhalation rate (E_m) in different building material samples.

| Sample type | Track density ($Tr\ cm^{-2}\ d^{-1}$) | C_{Rn} ($Bq\ m^{-3}$) | E_s ($mBq\ m^{-2}\ h^{-1}$) | E_m ($mBq\ kg^{-1}\ h^{-1}$) |
|----------------|---|---------------------------|---------------------------------|----------------------------------|
| Red-brick | 2.89 | 137.62 | 88.94 | 22.73 |
| Sand | 2.28 | 108.57 | 70.17 | 10.76 |
| Cement bricks | 4.56 | 217.14 | 140.34 | 29.34 |
| Cement | 3.03 | 144.29 | 93.26 | 19.50 |
| White cement | 2.86 | 136.19 | 88.02 | 18.40 |
| Gypsum | 1.50 | 71.43 | 46.17 | 13.27 |
| Granite | 2.86 | 136.19 | 88.02 | 15.57 |
| Ceramic | 4.05 | 192.86 | 124.65 | 28.67 |
| Cement plaster | 3.71 | 176.67 | 114.18 | 20.20 |
| Hydrated lime | 3.3 | 152.38 | 98.48 | 25.17 |

and cosmic radiation according to the following equation (Thabayneh & Jazzar, 2013):

$$D_\gamma\ (nGy\ h^{-1}) = 0.427 C_U + 0.662 C_{Th} + 0.043 C_K + 0.03 C_{Cs} + 34 \quad (8)$$

where C_{Cs} are the activity concentrations ($Bq\ kg^{-1}$) of cesium in the samples; in this study the samples did not contain cesium concentration ($C_{Cs} = 0\ Bq\ kg^{-1}$ for all samples). Table 4, represents the values of total gamma radiation dose rate (D_γ). The lowest value of the total gamma radiation absorbed dose was $46.39\ nGy\ h^{-1}$ found in gypsum and the maximum value was $247.78\ nGy\ h^{-1}$ found in cement brick. The annual effective dose values are below the permissible limits except the sample of cement brick.

3.9. Excess lifetime cancer risk

The excess lifetime cancer risk (ELCR) is calculated using the following equation (Thabayneh & Jazzar, 2013):

$$ELCR = AEDE \times DL \times RF \quad (9)$$

where AEDE, DL and RF are the total annual effective dose equivalent ($\mu Sv\ y^{-1}$), the duration of life (70 years) and risk factor (Sv^{-1}) (fatal cancer risk per sievert) for stochastic effects. ICRP 60 uses values of 0.05 for the public (Thabayneh & Jazzar, 2013). Table 4, shows the values of ELCR ranged from 0.381×10^{-3} to 6.061×10^{-3} ; where the samples of cement bricks, granite, ceramic and red brick show ELCR higher than the world average (0.29×10^{-3}) (Taskin et al., 2009); according to these results the cancer risk increases with increasing the exposure time to these materials.

3.10. Radon activity and radon exhalation rate

The activity of radon was obtained by track density from the etched detectors using calibration factor $0.021\ Tr\ cm^{-2}\ d^{-1} = 1\ Bq\ m^{-3}$ obtained from the experiment (Nain et al., 2006). Exhalation rate of radon is calculated by using the equations (Nain et al., 2006; Rati et al., 2010):

$$E_s\ (Bq\ m^{-2}\ h^{-1}) = \frac{CV\lambda/A}{\left[T + 1/\lambda(e^{-\lambda T} - 1)\right]} \quad (10)$$

and

$$E_m\ (Bq\ kg^{-1}\ h^{-1}) = \frac{CV\lambda/M}{\left[T + 1/\lambda(e^{-\lambda T} - 1)\right]} \quad (11)$$

where E_s is the radon surface exhalation rate; E_m is the mass exhalation rate; C the integrated radon exposure as measured by LR-115 detector ($Bq\ m^{-3}\ h$); V the effective volume of air in Can; λ the decay constant for radon (h^{-1}); T the exposure time (h); A the area covered by Can (m^2) and M is mass of sample in Can (kg).

Table 5 shows the activity of radon varies from 71.43 to $217.14\ Bq\ m^{-3}$. Using equations (10) and (11), radon surface and mass exhalation rates are found. Surface and mass exhalation rates vary from 46.17 to $140.34\ mBq\ m^{-2}\ h^{-1}$ and 13.27 to $29.24\ mBq\ kg^{-1}\ h^{-1}$, respectively. Radon activity, surface and mass exhalation rates found for Gypsum are the lowest values and for cement brick are found to be the maximum. A positive correlation was found between radium concentrations which is determined using HPGe gamma ray spectrometric system and radon concentration which is determined with the passive technique (LR-115); the correlation coefficient = 0.492. Graph for the correlation is shown in fig. 1. Table 6 compares the recorded values of the radon surface exhalation rate ($Bq\ m^{-2}\ h^{-1}$) and the mass exhalation rate ($Bq\ kg^{-1}\ h^{-1}$) with those obtained in other countries (Dabayneh, 2008; Ismail, Abumurad, & Al-Bataina, 1996; Kumar & Singh, 2004). Most of the results obtained in the present study are within the international values measured in many countries, while some of the results are higher.

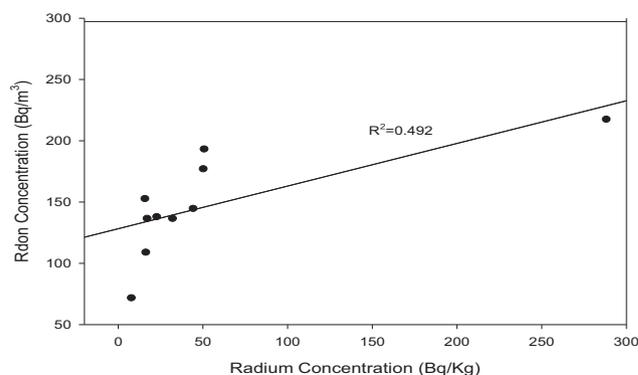


Fig. 1 – Linear regression of the concentration of radium and radon.

Table 6 – Comparison between surface exhalation rates (E_s) and mass exhalation rates (E_m) of some building materials in different countries.

| Material | Country | E_s (mBqm ⁻² h ⁻¹) | E_m (mBq kg ⁻¹ h ⁻¹) | References |
|--------------|-----------|---|---|-------------------------|
| Brick | Jordan | 90.0 | 6.0 | [Ismail et al., 1996] |
| | India | 258.0 | 119.0 | [Nain et al., 2006] |
| | Palestine | 90.0 | 4.5 | [Dabayneh, 2008] |
| | Egypt | 88.9 | 22.7 | Present study |
| Cement | Jordan | 83.0 | 5.00 | [Ismail et al., 1996] |
| | India | 288.0 | 13.3 | [Nain et al., 2006] |
| | Palestine | 99.0 | 4.9 | [Dabayneh, 2008] |
| | Iraq | 215.4 | 18.6 | [Zakariya et al., 2013] |
| | Egypt | 93.3 | 19.5 | Present study |
| White cement | India | 112.40 | 3.40 | [Kumar and Singh, 2004] |
| | Palestine | 65.0 | 3.6 | [Dabayneh, 2008] |
| | Egypt | 88.0 | 18.0 | Present study |
| Sand | Palestine | 48.0 | 2.4 | [Dabayneh, 2008] |
| | Iraq | 345.9 | 22.3 | [Zakariya et al., 2013] |
| | India | 93.4 | 2.8 | [Kumar and Singh, 2004] |
| | Egypt | 70.2 | 10.8 | Present study |
| | Palestine | 162.2 | 8,7 | [Dabayneh, 2008] |
| Ceramic | Iraq | 115.9 | 14.5 | [Zakariya et al., 2013] |
| | Egypt | 124.7 | 28.7 | Present study |
| | Palestine | 146.0 | 7.2 | [Dabayneh, 2008] |
| Granite | Egypt | 88.0 | 15.6 | Present study |
| | Palestine | 85.0 | 4.9 | [Dabayneh, 2008] |
| Gypsum | Iraq | 115.9 | 14.5 | [Zakariya et al., 2013] |
| | Egypt | 46.2 | 13.3 | Present study |

4. Conclusion

Gamma ray spectrometry has been used to determine the radioactivity concentrations ²²⁶Ra, ²³²Th and ⁴⁰K in the studied samples of various Egyptian building materials. The specific activity values of the natural source nuclear materials, ²²⁶Ra, ²³²Th and ⁴⁰K are normal and within the international levels, except samples of cement brick, cement and ceramic, were the recorded specific activity levels of which are higher than the permissible level.

The activity concentrations were obtained to estimate several radiological parameters that served to qualify and quantify the radiological hazard associated with the studied building materials. It is concluded that the radiological parameters obtained are normal and within the international levels except the samples of cement brick, granite and ceramic which showed the highest quantity, compared to other building materials analyzed in this work. Therefore, the use of these building material samples under investigation in the construction of dwellings is considered to be safe for inhabitants, except the cement brick, granite and ceramic samples which are critical points for safety in construction.

The ELCR values for cement brick, red brick, granite and ceramic are higher than the world average. The radon concentration measured by the passive technique showed a linear correlation with radium concentration measured by HPGe spectrometry. The radon concentration, alpha index, surface and mass exhalation rates for cement brick are the maximum values.

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REFERENCES

- Abu-Jarad, F. (1988). Application of nuclear track detectors for radon related measurements. *Nuclear Tracks and Radiation Measurements*, 15, 525.
- Amrani, D., & Tahtat, M. (2001). Natural radioactivity in Algerian building materials. *Applied Radiation and Isotopes*, 54, 687.
- Beretka, J., & Mathew, P. (1985). Natural radioactivity of Australian building materials, industrial wastes and by-products. *Health Physics*, 48, 87.
- Dabayneh, K. (2008a). Natural radioactivity in different commercial ceramic samples used in Palestinian buildings as construction materials. *Hebron University Research Journal A (Natural Science)*, 3(2), 49.
- Dabayneh, K. (2007). Radioactivity measurements in different types of fabricated building materials used in Palestine. *Arab Journal of Nuclear Sciences and Applications*, 40(3), 207.
- Dabayneh, K. M. (2008b). ²²²Rn concentration level measurements and exhalation rates in different types of building materials used in Palestinian buildings. *Isotopes and Radiation Research Journal*, 40(2), 277.
- Dabayneh, K. M., Mashal, L. A., & Hasan, F. I. (2008). Radioactivity concentration in soil samples in the southern part of the West Bank, Palestine. *Radiation Protection Dosimetry*, 131(2), 265.

- El-Shershaby, A., El-Bahy, S., Walley El-Din, N., & Dabayneh, K. (2006). Assessment of natural and man-made radioactivity levels of the plant leaves samples as bio-indicators of pollution in Hebron district – Palestine. *Arab Journal of Nuclear Sciences and Applications*, 39(2), 232.
- El-Taher, A. (2012). Assessment of natural radioactivity levels and radiation hazards for building materials used in Qassim area, Saudi Arabia. *Romanian Journal of Physics*, 57(3/4), 726.
- Ismail, A., Abumurad, K., Kullab, M., & Al-Bataina, B. (1996). Measurement of Rn-222 concentrations in building materials used in Jordan. *Mutah Journal for Research and Studies*, 11(3), 213.
- Khan, M., Tariq, M., & Rawat, R. (2012). Preliminary measurements of radon radiations in Bare Mode in Rampur district of Western U.P. India. *IOSR Journal of Applied Physics*, 1(4), 4.
- Kumar, A., & Singh, S. (2004). Radon exhalation studies in building materials using solid-state nuclear track detectors. *Pramana – Journal of Physics*, 62(1), 143.
- Merle, Lust, & Enn, Realo (2012). Assessment of natural radiation exposure from building materials in Estonia. *Proceedings of the Estonian Academy of Sciences*, 61(2), 107.
- Nain, M., Chauhan, R., & Chakarvarti, S. (2006). Alpha radioactivity in Indian cement samples. *Iranian Journal of Radiation Research*, 3(4), 171.
- Ngachin, M., Garavaglia, M., Giovani, C., Kwato, M., & Nourreddine, A. (2007). Assessment of natural radioactivity and associated radiation hazards in some Cameroonian building materials. *Radiation Measurements*, 42, 61.
- Nidal, Dwaikat, Ghassan, Safarini, Mousa, El-hasan, & Toshiyuki, Iida (2007). CR-39 detector compared with Kodalphafilm type (LR-115) in terms of radon concentration. *Nuclear Instruments and Methods in Physics Research Section A*, 574, 289.
- Rafique, M., Rehman, H., Matiullah, Malik, F., Rajput, M., Rahman, S., & Rathore, M. (2011). Assessment of radiological hazards due to soil and building materials used Mirpur Azad Kashmir, Pakistan. *Iranian Journal of Radiation Research*, 9(2), 77.
- Ramassay, V., Dheenathayalu, M., Ravisankar, R., & Ponnusamy, V. (2004). Natural radioactivity measurements in Beach-rock samples of South-East Coast of Tamilnadu, India. *Radiation Protection Dosimetry*, 111, 229.
- Rati, V., Mahur, A., Sonkawade, R., Suhail, M., Azam, A., & Prasad, R. (2010). Evaluation and analysis of ^{226}Ra , ^{232}Th , ^{40}K and radon exhalation rate in various grey cements. *Indian Journal of Pure and Applied Physics*, 48, 473.
- Shakir Khan, M., Naqvi, A., Azam, A., & Srivastava, D. (2011). Radium and radon exhalation studies of soil. *Iranian Journal of Radiation Research*, 8(4), 207.
- Taskin, H., Karavus, M., Ay, P., Topuzoglu, A., Hindiroglu, S., & Karahan, G. (2009). Radionuclide concentrations in soil and lifetime cancer risk due to the gamma radioactivity in Kirklareli, Turkey. *Journal of Environmental Radioactivity*, 100, 49.
- Thabayneh, K. M. (2013). Measurement of natural radioactivity and radon exhalation rate in Granite samples used in Palestinian buildings. *Arabian Journal for Science and Engineering*, 38, 201.
- Thabayneh, K. M., & Jazzar, M. M. (2013). Radioactivity levels in plant samples in Tulkarem district, Palestine and its impact on human health. *Radiation Protection Dosimetry*, 153(4), 467.
- United Nations Scientific Committee on the Effects of Atomic Radiation. (1977). *Sources, effects and risks of ionizing radiation*. Report to the general assembly, 1, Annex B.UNSCEAR.
- United Nations Scientific Committee on the Effects of Atomic Radiation. (1993). *Sources and effects of ionizing radiation*. Report to the general assembly, 1, Annex B.UNSCEAR.
- United Nations Scientific Committee on the Effects of Atomic Radiation. (2000). *Sources and effects of ionizing radiation*. Report to the general assembly, 1, Annex B.UNSCEAR.
- Xhixha, G., et al. (2013). First characterization of natural radioactivity in building materials manufactured in Albania. *Radiation Protection Dosimetry*, 153(2), 1.
- Zakariya, A., Mohamad, S., Asaad, H., & Ammar, A. (2013). Radon exhalation rate from building materials using Passive Technique Nuclear Track Detectors. *International Journal of Scientific and Engineering Research*, 4(7), 1276.