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Measurement of Radon Exhalation Rate from Building Materials

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Abstract

Indoor radon has been recognized as one of the health hazards for mankind because long-term exposure to radon increases the risk of developing lung cancer. This study aims at assessing the contribution of building materials towards the total indoor radon exposure to the inhabitants of Nablus district, Palestine. The radon exhalation rate has been carried out for different building materials of international origin used in construction in Nablus district. The "closed-can technique" has been employed in this study using solid state nuclear track detectors (CR-39). After 100 days of exposure, CR-39 detectors were etched chemically and then counted under an optical microscope. Results show that Radon exhalation rates from granite and marble have relatively high values as compared to other building materials followed- in order- by cement, ceramic, concrete, building stones, and porcelain, while gypsum, sand, gravel and bricks contribute less to radon exhalation rate which was found to range from (55.37 ± 15.01) mBq/m^{2}h for gypsum samples to (589.54 ± 73.24) mBq/m^{2}h for granite samples, with a total average value of (268.56 ± 166.21) mBq/m^{2}h. The corresponding radon concentration, effective radium content, and annual effective dose average values were (148.49 ± 91.13) Bq/m^3, (1.93 ± 1.20) Bq/Kg and (3.74 ± 2.30) mSv/y, respectively. In general, the radon exhalation rate from the investigated building materials is low and under the global value except for granite, marble and some cement samples and thus except for the excluded, the studied materials are safe as construction materials.

Keywords: *closed-can technique, radon exhalation rate, CR-39, building materials*

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INTRODUCTION

During the last few decades researchers were concerned with the effect of natural radioactivity on human health. Radon gas is by far the most important source of ionizing radiation among those that are of natural origin [1].

The most important isotope of radon, in terms of environmental effects is (^{222}Rn) which is formed from the α-decay of radium (^{226}Ra), which is a decay product of Uranium (^{238}U). Uranium has been around since the earth was formed and it's most common isotope ^{238}U (Natural abundance = 99.284%) has a very long half-life (4.5 billion years). Uranium, radium, and thus radon, will continue to occur for millions of years at about the same concentrations as they do now [2].

The damage of Radon lies in that it decays quickly, giving off tiny radioactive high energy alpha- particles which cause local ionization damaging the tissue with a subsequent risk for cancer development. β- and γ- radiation are also present from some of the decay products but this much lower energy content compared to α- radiation makes the effect relatively marginal [3]. Those radioactive particles when inhaled, can damage the cells that line the lung. And so, long-term exposure to radon can lead to lung cancer [4].

Radon exhalation measurement using the "Closed-can Technique" have been carried out by many researchers worldwide as well as at national level and extensive data are available in literature [5–13].

The purpose of this study is to measure the Radon exhalation rates from building materials used in Nablus district. Our study will include samples of marble, granite, ceramic, porcelain, concrete, cement, sand, gravel, bricks, gypsum, and building stones from different international origins used in the mentioned area of study in order to get some insight on its impact on the health of the Palestinian people residing in Nablus area.

AREA OF STUDY

In this study, we present our data concerning measurement of the radon exhalation rate from building material samples collected from Nablus district in the West Bank in Palestine. The location of this district is shown in Figure 1. Houses in this district are mainly constructed from soil, bricks, cement, sand, granite and marble. Nablus is located in the northern part of the West Bank of Palestine in a strategic position at a junction in the shade of two mountains: Ebal (940 m height) to the North and Gerzim (881 m height) to the South. The city stands at an elevation of around 550 m above sea level, in a narrow valley between the two mentioned mountains. Nablus is located 110 km west of Amman, capital of Jordan, 63 Km north of Jerusalem, the capital of Palestine, and 42 Km east of the Mediterranean. It lies on the latitude 14°–32° to the north of the Equator, and the longitude 15°–35° to the east of Greenwich.

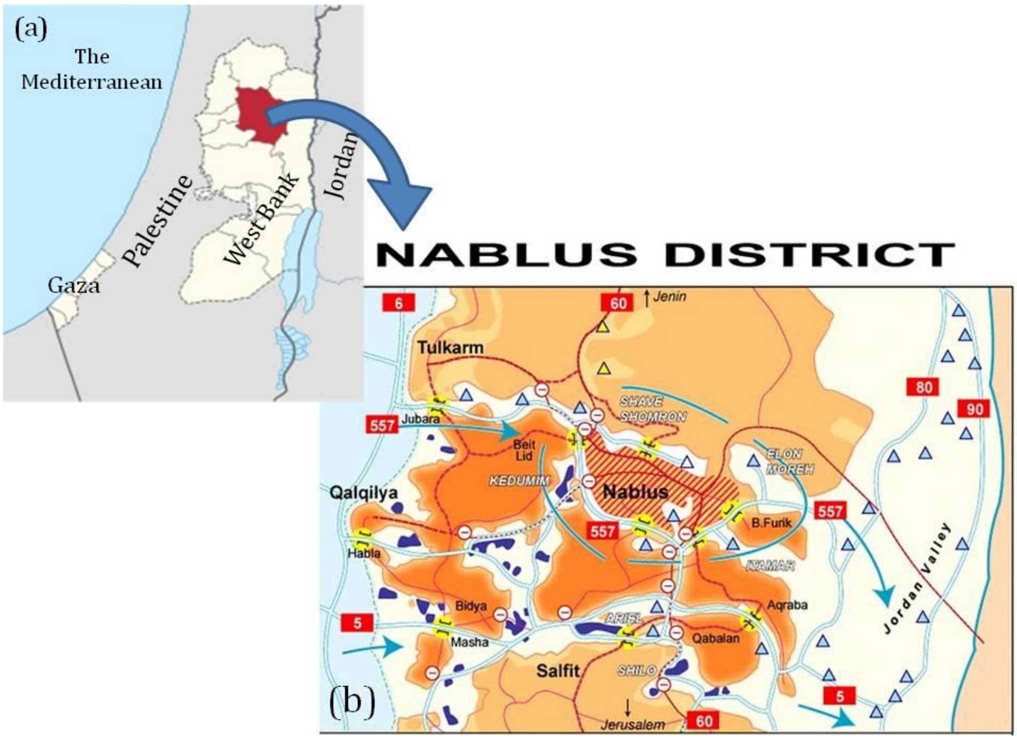

Fig. 1: *(a) Palestine Map, (b) The Study Region.*

The relatively temperate Mediterranean climate brings hot, dry summers and cool, rainy winters to Nablus. Spring arrives around March–April and the hottest months in Nablus are July and August with the average high being 29.4°C. The coldest month is January with temperatures usually at 7.2°C. Rain generally falls between October and March. The period during which this study was performed was in the cold months including December, January and February.

MATERIALS AND METHODS

Forty seven samples of building materials from different international origins used in buildings in Nablus District were collected randomly from different organizations, quarries and commercial companies all around the area of study during October and November.

Samples included five types of granite and marble each, seven of ceramic, four from each of porcelain, sand, bricks, gravel, cement, and building stones, three from concrete and gypsum each. These materials were from American, Chinese, Italian, French, Spanish, Turkish, Arabian, and Palestinian's origins. Bulk samples were crushed and milled to a fine powder with a uniform particle size, while the powder samples were used in their natural form. The respective net weights of the samples ready for measurement were recorded. Samples were then identified and given a number and an identifying code, and then dried in an oven at about 100°C for two hours to evaporate all moisture content. Building material samples were put at the bottom of plastic cylindrical vessels of volume (2.945 X 10^{-3}) m^3 with cross sectional area of (0.01227 m^2) as shown in Figure 2. Square pieces (1 cm x 1 cm) of solid-state nuclear track detectors (SSNTDs) CR-39 (Intercast Europe S.p.A, Italy), were then fixed on the inside cover of the vessels using blue-tac paste at a distance of about (22.5 cm) from the surface area of the samples so as to count only the contribution of ^{222}Rn and to evade the role of thoron from the surface of sample [14, 15]. Cans were hermitically sealed and stored in the months from November 2011 through February 2012 for about 100 days.

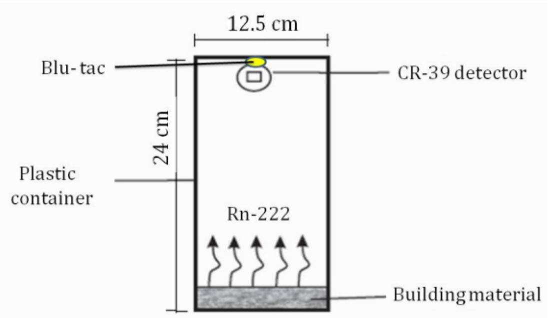

Fig. 2: Experimental Setup for the Measurement of Radon Exhalation Rate.

The detectors received variable level of radon exposure. Consequently, effective exposure time needs to be determined. The effective exposure time was calculated using the following relation [16]:

$$T_{eff} = t + \tau(e^{-\lambda t} - 1) \qquad (1)$$

where, τ is the mean life time of radon (5.5 days), t is the total exposure time and λ is ^{222}Rn decay constant. This type of correction is needed only for a closed system [14]. After the mentioned period, detectors were taken out of the dosimeters and exposed to chemical etching in (6.25 N) solution of Sodium Hydroxide (NaOH) at a temperature of 75°C for 5 h. Detectors were then washed by running and distilled water and dried to remove any remaining amount of the etchant from the surface of the detectors. A digital optical microscope with 400 times magnification was used to count the number of tracks per field of view. About thirteen fields of view were scanned randomly for each detector, then the average number of tracks per field of view, corrected to the background, was used to count the track density per cm^2.

The calculated track density was converted into radon concentrations in Bq/m^3 using the calibration factor obtained by the manufacturer, where every track per cm^2 per day on the CR-39 detectors corresponds to an exposure of 12.3 Bq/m^3 for the activity of radon gas and its daughters. From the measured average track densities, the radon exhalation rate was calculated via the relation [17]:

$$E = \frac{\rho}{\eta A}\left[\frac{\lambda V}{Teff}\right] \qquad (2)$$

where, E is radon exhalation rate (Bq m^{-2} h^{-1}), ρ: is the track density (tracks/ cm^2), η: is the detector sensitivity (tracks cm^{-2} h^{-1}/Bq m^{-3}), λ: is ^{222}Rn decay constant (h^{-1}), V: is the void volume of the container, A: is the area of the sample, and T_{eff}: is the effective exposure time (h) as defined by Eq. (1).

Radon concentration was calculated using the average track densities according to the relation [6]:

$$C_{Rn} = k\frac{\rho}{Teff} \qquad (3)$$

where, C_{Rn} is radon concentration (Bq/m^3), k: is the calibration factor (Bq m^{-3}/ tracks cm^{-2} h^{-1}), ρ: is the track density (tracks/ cm^2).

And the following relation was used to compute the effective radium content [18]:

$$C_{Ra} = \frac{\rho V}{\eta M Teff} \qquad (4)$$

where, C_{Ra}: is the effective radium content (Bq/Kg), M: is the mass of the sample (Kg), and the other symbols carry the same meaning as in Eq. (2)

The annual effective dose equivalent was calculated from radon concentrations using the UNSCEAR recommended conversion factor of 9 nSv per (Bq h m^{-3}) [19]. Assuming 7000 h per year indoor (an indoor occupancy factor of 80%) and an equilibrium factor of 0.4 [20], the effective dose for one year radon exposure is calculated using the relation [21]:

$$Dose = \epsilon\, f_{Rn}\, T\, C_{Rn} \quad (5)$$

where, f_{Rn}: is the conversion factor, T: is the time spent indoors per year (7000 h), ϵ: is the equilibrium factor, and C_{Rn}: is the radon concentration.

RESULTS AND DISCUSSION

As can be noted from the data listed in Table 1, radon exhalation rate from granite samples ranged from 138.92 mBq/m^2 h for G4 sample which is from Saudi Arabian origin, to 949.46 mBq/m^2 h for the American granite sample (G1), while Radon exhalation rate from marble samples ranged from a minimum value of 277.82 mBq/m^2 h for a local origin sample (M2) manufactured at Hebron city, to 646.35 mBq/m^2 h for the Spanish sample (M5). Taking the results for each type of the studied samples by group (Table 2), the average values for radon exhalation rate, radon concentration, effective radium content, and the effective dose were calculated.

Table 1: *Radon Concentration, Exhalation Rates, Effective Radium Content and the Annual Effective Dose from Studied Samples.*

Sam No.	Sam. Code	Country of Origin	E (mBq/m^2 h)	C_{Rn} (Bq/m^3)	C_{Ra} (Bq/Kg)	Dose (mSv/y)	Sam. No.	Sam. Code	Country of Origin	E (mBq/m^2 h)	C_{Rn} (Bq/m^3)	C_{Ra} (Bq/Kg)	Dose (mSv/y)
1.	G1	America	949.46	516.73	7.43	13.02	12.	C2	China	330.54	183.53	2.40	4.62
2.	G2	China	429.41	235.20	3.12	5.93	13.	C3	Spain	707.25	395.57	5.44	9.97
3.	G3	Italy	703.97	383.09	4.89	9.65	14.	C4	Italy	345.75	190.66	2.65	4.80
4.	G4	Saudi Arabia	138.92	76.62	0.84	1.93	15.	C5	China	304.18	171.06	2.21	4.31
5.	G5	Spain	725.97	399.13	4.85	10.06	16.	C6	Spain	220.57	121.16	1.71	3.05
6.	M1	Turkey	395.28	215.60	2.78	5.43	17.	C7	Japan	160.26	89.09	1.17	2.25
7.	M2	Palestine/ Hebron	277.82	151.46	2.13	3.82	18.	P1	China	187.31	103.35	1.36	2.60
8.	M3	Turkey	378.40	206.69	2.63	5.21	19.	P2	Spain	206.33	114.04	1.49	2.87
9.	M4	China	496.09	272.62	3.60	6.87	20.	P3	China	277.63	153.24	2.03	3.86
10.	M5	Spain	646.35	356.37	3.90	8.98	21.	P4	Italy	313.30	172.84	2.28	4.36
11.	C1	Turkey	363.42	204.91	2.56	5.16							

Sam.: sample, E: exhalation rate, C_{Rn}: radon concentration, C_{Ra}: radium content, Dose: annual effective dose.
Types of samples: G: granite, M: marble, C: ceramic, P: porcelain.

Comparing these results, one can see from Table 2 that granite have on the average- the highest radon concentration of all other samples followed by marble. The annual effective doses (see Table 1) from granite and marble samples (except for G4 and M2 samples) are higher than the National Council on Radiation Protection and Measurements (NCRP) maximum value of 5 mSv/y [22].

Thus, it can be concluded that most of granite and marble samples are not safe to be used as building materials which are usually used in tiling the kitchens of Palestinian homes. This may be due to the high radium content in granite and marble samples and the low level in other samples [23, 14]. Therefore, granite and marble can be a significant source of radon in houses, when used in tiling large enclosed areas.

Radon exhalation rate from ceramic and porcelain samples ranged from a minimum value of 160.26 mBq/m^2 h for the Japanese ceramic sample (C7), and 187.31 mBq/m^2 h for Chinese porcelain (P1), to a maximum value of 707.25 mBq/m^2 h for the Spanish ceramic (C3), and 313.3 mBq/m^2 h for Italian porcelain (P4), with a mean value of (347.42 ± 79.95) mBq/m^2 h and (246.14 ± 59.29) mBq/m^2 h for ceramic and porcelain respectively (Table 2). The corresponding radon concentration, effective radium content and the annual effective dose average values for ceramic and porcelain samples, respectively, were (193.71 ± 45.10) and (135.86 ± 32.67) Bq/m^3, (2.59 ± 0.57) and (1.79 ± 0.44) Bq/Kg, and (4.88 ± 1.14) and (3.42 ± 0.82) mSv/y. In comparison it can be seen that ceramic samples have a slightly higher radon concentrations than porcelain and lower than both of granite and marble. Also the corresponding average annual effective doses from both of ceramic and porcelain are lower than the NCRP maximum value except for the Spanish ceramic sample. In general it can be concluded that both of ceramic and porcelain are safe to be used as building materials.

Table 2: Radon Concentration, Exhalation Rate, Effective Radium Content and Annual Effective Dose Average Values from Studied Samples Used in Nablus District.

Sample Type	$E_{ave.}$±SD (mBq/m^2 h)	$C_{Rn\ Ave.}$± SD (Bq/m^3)	$C_{Ra\ Ave,}$ ± SD (Bq/Kg)	$Dose_{Ave.}$± SD (mSv/y)
Granite	589.54 ± 73.24	322.16 ± 40.61	4.23 ± 0.37	8.12 ± 1.02
Marble	438.79 ± 89.38	240.55 ± 49.60	3.01 ± 0.61	6.06 ± 1.25
Ceramic	347.42 ± 79.95	193.71 ± 45.10	2.59 ± 0.57	4.88 ± 1.14
Porcelain	246.14 ± 59.29	135.86 ± 32.67	1.79 ± 0.44	3.42 ± 0.82

Cement as a building material is a fine-grained compound that turns into a solid when mixed with water. Cement is used to bind mixtures of materials into a composite solid. From Table 3 one can see that radon exhalation rate from cement samples ranged from 250.89 mBq/m^{2}h for the Jordanian cement (Ce2), to 497.30 mBq/m^{2}h for the Israeli cement (Ce3) with a mean value of (363.38 ± 58.77) mBq/m^2 h. The corresponding radon concentration, effective radium content and the annual effective dose average values for cement samples, respectively, were (204.91 ± 33.45) Bq/m^3, (2.61 ± 0.49) Bq/Kg, and (5.16 ± 0.84) mSv/y. It is obvious that cement samples have medium radon concentration between marble and ceramic. The corresponding annual effective doses from cement are lower or at the boundary of the NCRP maximum value except for the Israeli cement with a high annual effective dose value of 7.1 mSv/y, so more attention is required in choosing the type of cement that must be used in building in Nablus district.

Radon exhalation rate from sand, brick, and gravel samples (Table 4) have average values of (80.62 ± 21.8), (112.92 ± 26.58) and (126.05 ± 31.53) mBq/m^2 h, respectively. The corresponding radon concentration average values for them, respectively, are (44.99 ± 12.15), (62.36 ± 14.77), and (69.49 ± 17.00) Bq/m^3, the annual effective dose average values are (1.13 ± 0.31), (1.57 ± 0.37), and (1.75 ± 0.43) mSv/y, while the effective radium content average values are (0.48 ± 0.13), (0.86 ± 0.22), and (0.91 ± 0.23) Bq/Kg. From the measured values it is clear that radon concentrations and exhalation rates from brick, sand and gravel samples are very low, also the corresponding average annual effective doses from all of them are much lower than the NCRP maximum value and at the boundary of the recommended minimum value (of 1 mSv/y), so it can be concluded that each of brick, sand and gravel used in Nablus district from all origins do not pose a radiation danger when used as building materials.

Table 3: *Radon Concentration, Exhalation Rates, Effective Radium Content and the Annual Effective Dose from Cement Samples.*

Sample code	**Country of origin**	**E (mBq/m^2 h)**	**C_{Rn} (Bq/m^3)**	**C_{Ra} (Bq/Kg)**	**Dose (mSv/y)**
Ce1	Turkey	353.28	199.57	2.56	5.03
Ce2	Jordan	250.89	140.76	1.81	3.55
Ce3	Israel	497.30	281.53	3.35	7.09
Ce4	France	352.06	197.78	2.72	4.98
	Ave.	363.38	204.91	2.61	5.16
	SD	58.77	33.45	0.49	0.84

Results obtained for radon concentration and exhalation rates for gypsum (which is a soft powder used as a decorative material in Nablus district) (Table 4) have the lowest values compared with all other building materials with radon exhalation rate average value of (55.37 ± 15.01) mBq/m^2 h, radon concentration of (31.48 ± 8.59) Bq/m^3 and a corresponding effective radium content and annual effective dose average values around (0.37 ± 0.09) Bq/Kg and (0.79 ± 0.22) mSv/y, respectively.

Radon exhalation rate average value for concrete samples (Table 4) was (325.38 ± 32.43) mBq/m^2 h, while the corresponding radon concentration, effective radium content and the annual effective dose average values, respectively, were (179.37 ± 16.94) Bq/m^3, (2.46 ± 0.26) Bq/Kg, and (4.52 ± 0.43) mSv/y. In comparison it can be seen that concrete samples have radon concentration value close to cement samples. This is to be expected as the main ingredient of concrete is cement. However, the annual effective dose from concrete is lower than the NCRP maximum value, so concrete samples are safe as a building material. Building stones are stones that are used in Nablus district to cover the buildings from outside after finishing the construction which increases their durability. Building stone samples recorded an acceptable radon exhalation rate with an average value of (268.59 ± 54.27) mBq/m^2h, radon concentration, effective radium content and annual effective dose average values of (147.00 ± 29.90) Bq/m^3, (1.95 ± 0.37) Bq/Kg and (3.70 ± 0.75) mSv/y, respectively.

The average value of radon exhalation rate from all building materials was (268.56 ± 166.21) mBq/m^2h, the corresponding radon concentration, effective radium content, and annual effective dose average values were (148.49 ± 91.13) Bq/m^3, (1.93 ± 1.20) Bq/Kg, and (3.74 ± 2.30) mSv/y, respectively.

Table 4: *Radon Concentration, Exhalation Rates, Effective Radium Content and the Annual Effective Dose from Building Material Samples Used Fin Nablus District.*

Sample Type	**$E_{ave.}$±SD (mBq/m^2 h)**	**$C_{Rn\ Ave.}$± SD (Bq/m^3)**	**$C_{Ra\ Ave.}$ ± SD (Bq/Kg)**	**$Dose_{Ave.}$± SD (mSv/y)**
Sand	80.62 ± 21.80	44.99 ± 12.15	0.48 ± 0.13	1.13 ± 0.31
Brick	112.92 ± 26.58	62.36 ± 14.77	0.86 ± 0.22	1.57 ± 0.37
Gravel	126.05 ± 31.53	69.49 ± 17.00	0.91 ± 0.23	1.75 ± 0.43
Gypsum	55.37 ± 15.01	31.48 ± 8.59	0.37 ± 0.09	0.79 ± 0.22
Concrete	325.38 ± 32.43	179.37 ± 16.94	2.46 ± 0.26	4.52 ± 0.43
Building stone	268.59 ± 54.27	147.00 ± 29.90	1.95 ± 0.37	3.70 ± 0.75

The histogram in Figure 3 clearly shows that granite and marble are the most radon radiant followed by cement, ceramic and concrete while gypsum, sand, gravel and bricks have low radon and radium contents. Also, a good correlation is found between both of radon exhalation rate and radon concentration with the effective radium content in building material samples as depicted in Figures 4 and 5, respectively, with a correlation coefficient $R^2 = 0.996$.

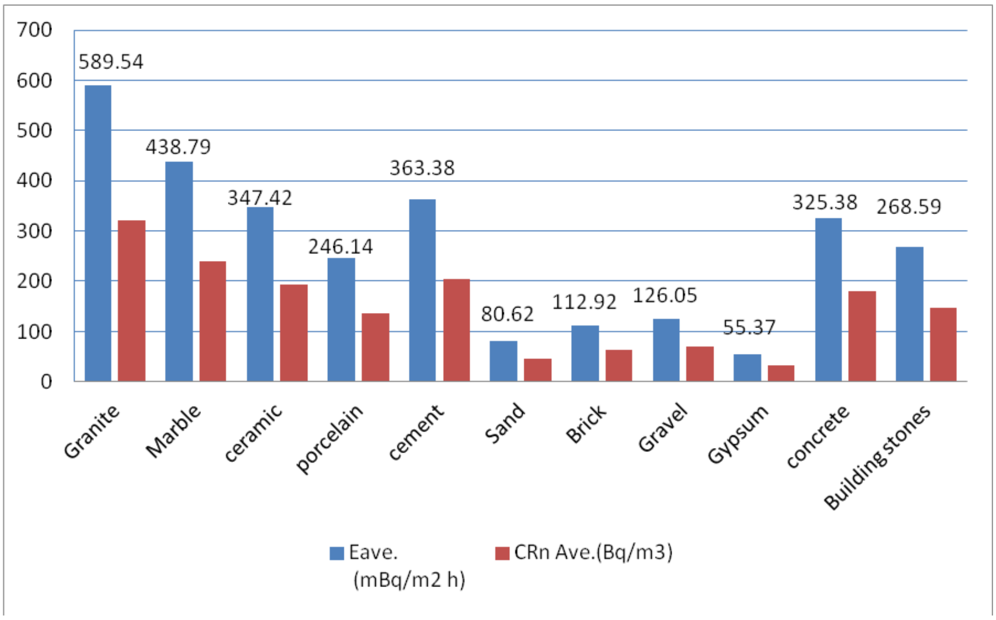

Fig. 3: *Comparing Histogram for the Average Radon Concentrations ($C_{rn\ Ave.}$) and Exhalation Rates ($E_{Ave.}$) from Building Materials.*

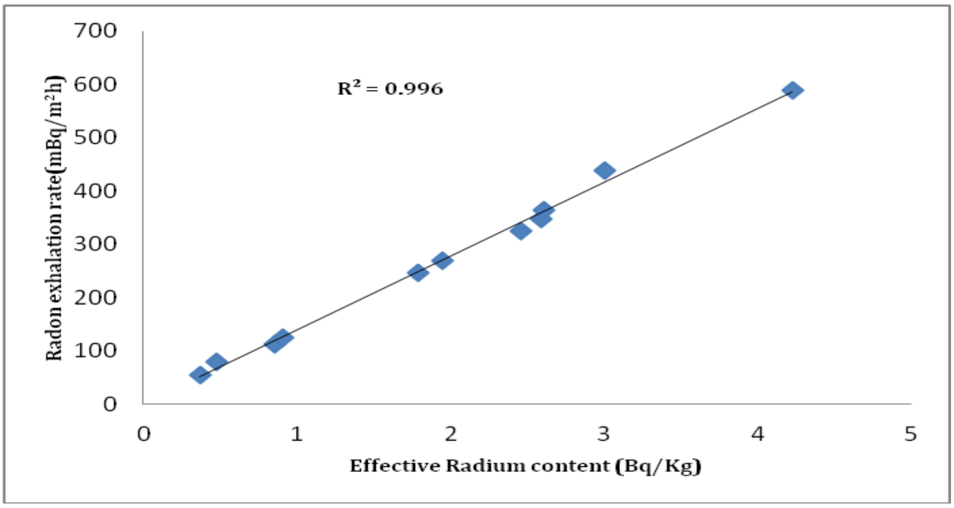

Fig. 4: *Correlation Between Radon Exhalation Rate from Building Material Samples and the Effective Radium Content.*

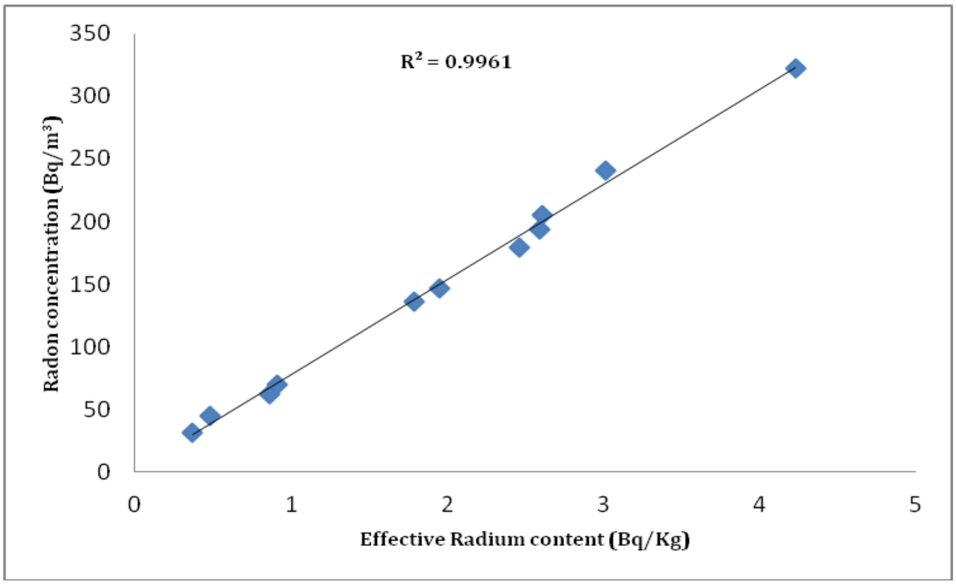

Fig. 5: *Correlation Between Radon Concentration of Building Material Samples and the Effective Radium Content.*

Finally, the average values of the exhalation rate results, obtained in the current study, show reasonable agreement when compared with those reported in the literature as given in Table 5.

Table 5: *Comparison of Present Results of Building Materials' Radon Exhalation Rate with the Values Reported for Different Countries.*

Building material	Country	E_{Rn} (mBq/m^{2}h)	Reference
Granite	Saudi Arabia Canada Greece Palestine/Hebron Palestine/Nablus	700 1750 1240 ± 119 146 589.54 ± 73.24	[24] [9] [25] [12] Current study
Marble	Egypt Palestine/ Hebron Palestine/Nablus	189 333 to 1250 127 438.79 ± 89.38	[26] [16] [12] Current study
Ceramic	Egypt Palestine Hebron Palestine/Nablus	224 75 347.42	[26] [12] Current study
Sand	Pakistan Palestine/ Hebron Palestine/Nablus	366 ± 8 to 649 ± 8 261 48 80.62 ± 21.80	[27] [28] [12] Current study
Gravel	Pakistan Palestine/ Hebron Palestine/Nablus	168 ± 17 to 322 ± 11 143 126.05 ± 31.53	[29] [12] Current study
Bricks	Pakistan India Greece Palestine/ Hebron Palestine/Nablus	184 ± 14 to 231 ± 14 292 112.4 210 ± 18 90 112.92 ± 26.58	[29] [28] [30] [25] [12] Current study

CONCLUSION

Using the closed can technique and the solid state nuclear track detectors (CR-39), radon exhalation rate from building material samples used in Nablus district was determined with the aim to assess the contribution of individual material (e.g., granite, marble, ceramic, cement, concrete, sand, gravel, and bricks) to the total indoor radon exposure of the inhabitants of Nablus district as well as international peoples since the construction materials are of international origin.

The values of radon concentration, effective radium content and the annual effective dose were also determined. The annual effective dose values were compared with the effective dose limit values recommended by the National Council on Radiation Protection and Measurements (from 1 to 5 mSv/y). Results obtained from the current study show that the radon exhalation rates from granite and marble have relatively high values as compared to other building material samples followed by cement, ceramic and concrete while gypsum, sand, gravel and bricks contribute less to the indoor radon. The radon exhalation rate in the studied samples ranged from a maximum value of (589.54 ± 73.24) mBq/m^2h for granite samples to a minimum value of (55.37 ± 15.01) for gypsum samples with an average value (268.56 ± 166.21) mBq/m^2h.

In general, the radon exhalation rate from the investigated building materials is low and under the global value except for granite, marble and some cement samples and thus except for the excluded, the studied materials are safe as construction materials.

Future work is planned to establish a database for all building materials available in the Palestinian market with the aim of increasing the awareness of the citizens on what kinds of building materials are most safely to be used and those that must be restricted according to this study and the forthcoming studies. However, restricting the use of certain building materials (e.g. building stones) might have economical consequences at the national level and should be assessed when establishing binding regulations.

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