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A Comparison Study of Radon exhalation Rate from Fresh and Destroyed Building Materials in Jabalia-Gaza, Palestine

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This study aims at assessing the contribution of fresh and destroyed building materials in war 2014 towards the total indoor radon exposure to the inhabitants of Jabalia district in Gaza. 40 Samples have been collected from common destroyed building materials in 2014 war in Jabalia district, and another similar 40 samples from fresh building material from Gaza. The closed-can technique has been employed in this study using solid state nuclear track detectors (CR-39). After 124 days of exposure to radon, CR-39 detectors were etched chemically by (6N) NaOH solution at 75° C for 4.20 hours and then counted under an optical microscope. Results show the average radon exhalation rate in term of area from the fresh building materials in the studied samples ranged from (27.27) mBq.m-2.h-1 for glass samples to (107.48) mBq.m-2.h-1 for Asbestos samples, while the destroyed materials ranged from (86.51) mBq.m-2.h-1 for glass samples to (463.90) mBq.m-2.h-1 for Asbestos samples. On the basis of these values the annual effective dose for each sample was also determined and compared with the effective dose limit values recommended by the National Council on Radiation Protection and Measurements (from 1 to 5 mSv/y). In general, the annual effective doses from the investigated destroyed building materials are low and under the global value except for Concrete and asbestos of destroyed samples with average values(9.464) and (9.3528) mSv/y, respectively, and from samples of fresh building materials the effective dose are (2.25) for Concrete and (2.71) for Asbestos. There are big differences between results from fresh materials and destroyed materials, which may influenced by war pollution.

Keywords:
CR-39, Radon Concentration, Radon Exhalation, destroyed, Fresh, Building materials.

1. Introduction:

Most building materials of natural origin contain small amount of Naturally Occurring Radioactive Materials (NORM), mainly radionuclides from the ^{238}U and ^{232}Th decay chains. 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. People are continuously exposed to ionizing radiation from NORM. The origin of these materials is the Earth's crust, but they find their way into building materials. Radiation exposure due to building materials may be classified into external and internal exposures. It is well known that radiation exposure due to building materials in building is caused mostly by the external γ-rays and α-particles emitted from radio nuclides of the uranium (^{238}U) and thorium (^{232}Th) decay series as well as from the potassium radionuclide (^{40}K). The contribution of building materials to indoor radon and thoron concentration are usually low and can be measured by passive and active methods [1.2.3.4.5]

All building materials contain various amounts of main natural radionuclides of the (^{238}U) and (^{232}Th) series, and since those radionuclides are sources of Radon gas then the knowledge of the natural radioactivity of building materials is important for the determination of population exposure to radiations. For the aforementioned reasons we intend to study the concentration of Radon and the exhalation rate from destroyed building. It will then be compared to results obtained with the results of previous studies.

In this study, we present our data concerning measurement of the radon exhalation rate from destroyed and fresh building material samples collected from Jabalia district in the Gaza strip in Palestine using close vessel technique. The purpose of this study is to measure and compare the Radon

exhalation rates from destroyed building materials during 2014 war against Gaza, Palestine and from fresh building materials. Our study will include samples of a red brick, marble, ceramic, concrete, tiles, and asbestos, glass and building stones from different origins used in the mentioned area of study.

The location of this district is in the northern part of the Gaza strip of Palestine. Houses in this district are mainly constructed from soil, bricks, cement, sand, granite and marble..

In a neighboring country, Egypt, a study on ^{222}Rn exhalation rate from Egyptian building materials was performed in 2009 and found that the radon exhalation rate in the studied samples ranged from (2.2 x 104 ± 7.2 x102) μBq m^{-2} s^{-1} , for granite sample, to (3.4x101 ± 9.0x100) μBqm^{-2} s^{-1}, for portland cement with an average value (1.8x103 ± 6.5x101) μBq m^{-2} s^{-1} [3]. El-Ghossain et. al.[1] , the activity of alpha, beta and gamma radiation in tap water in the north-east of Gaza (Al-Naser area) were measured. For this purpose we used a solid state nuclear track detectors (CR-39) and some other detectors (Geiger counter, NaI detector). The average gross alpha concentration from C4-39 is 35.50 Bq/m3 (0.95 pci/L), the maximum concentration is 64.67 Bq/m3, and minimum concentration is 24.20 Bq/m3 [4]. The radon concentration in Air at middle of Gaza Strip was measured, the average radon concentration 37.83 Bq/m^3 [4]. In Gaza, Palestine, the radon concentration in soil in at north of Gaza Strip, was measured by N. M. Hammed (2005) [5]. The results of the average radon concentration was 207.24 Bq/m^3. M. Rasas (2003) [4] measured the radon concentration in Air at middle of Gaza Strip, the average radon concentration in Air was 37.83 Bq/m^3 [6]. Then radon concentration values have been measured using passive integrated solid-state nuclear track devices. The overall average radon concentration for all water samples is found to be (14.24 ±3.62) Bq/L [7]. In Nablus district, Palestine , The measured 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^2h for gypsum samples to (589.54 ± 73.24) mBq/m^2h for granite samples, with a total average value of (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 [8].

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2. Materials and Experimental Methods:

Different samples of destroyed building materials after the 2014 war against Gaza were collected randomly, where 40 different destroyed building materials, and 40 samples of fresh buildings materials, like, houses, commercial companies, and factories, all around the area of study during the month of March to July. Samples were a red brick (F), marble (D), ceramic (G), concrete (B), tiles (E), asbestos (H), glass (C) and building stones (A), samples were from different origins, used in construction of building in Jabalia district, Gaza Strip, Palestine. Samples were then identified and given a number and an identifying symbol which identify the location of the samples, as in table 1. Then 5 kg from each sample were collected and dried in a temperature controlled furnace (oven) at a temperature100°C for two hours to ensure that moisture was completely removed. And then the samples were crushed to a fine powder and sieved through a small mesh size to remove the larger grains size and render them more homogenous. The respective net weights of the samples ready for measurement were recorded.

The close vessel technique was used in this study "cans technique" or we call them "Dosimeters". Dosimeters are plastic cylindrical vessels of volume (7.93×10^{-4}) m^3 with cross sectional area of (5.02×10^{-3} m^2) as shown in figure 1. The destroyed building material samples were put at the bottom of these vessels. About 200 g of each sample was placed in a plastic can of dimensions15.8 cm in height and 8 cm in diameter.

The use of plastic solid-state nuclear track detectors, SSNTDs of type CR-39, which were cut into small pieces, 2 cm × 2 cm and fixed on the top of inner surface of the can, in such a way that its sensitive surface always facing the sample. The can was sealed air tight with adhesive tape and kept for assessment of radon exhalation for exposure evaluation over four months. During the exposure period (one hundred and twenty four days), the detector was exposed freely to the emergent radon from the sample in the can so that it could record alpha particles resulting from the decay of radon in the remaining volume of the can [3, 4, 5, 6, 7, 8]

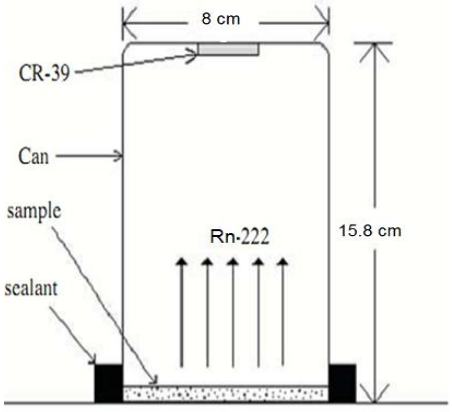

Figure 1 CR-39 Set up for Radon Detection

After the mentioned period, forty detectors were taken out of the dosimeters. The detectors were then chemically etched in 6 N-solution of Sodium Hydroxide (Na OH) at a temperature of 75 C for four hours and one third of an hour. The etching process was performed at chemistry Laboratories at Islamic University of Gaza using the setup. In addition, the function of the condenser is to keep the concentration of the NaOH solution constant, and the function of the thermometer is to make sure that the temperature is constant during the whole period of the etching process. After four hours and one third of an hour

detectors were washed by running and distilled water and then dried to remove any remaining amount of the etchant from the surface of the detectors. By now alpha tracks formed on the detectors were ready for scanning and counting

A digital optical microscope with 400 times magnification was used to count the number of tracks per field of view; about ten fields of view were scanned randomly for each detector. Tracks of alpha particles emitted by radon in a CR-39 detector were scanned by the microscope as shown in Figure 2. The area of the field of view was calculated by the digital microscope and found to be equal about 5.3×10^{-3} cm^2 ; the average number of tracks per field of view was used to calculate the track density. The calculated track density was converted into radon concentrations in Bq/m^3 using the calibration factor (k) obtained by the standard manufacturer, where every track per cm^2 per day on the CR-39 detectors corresponds to an exposure of 12.5 Bq/m^3 for the activity of radon gas and its daughters and we use previous calibrations [6, 7 ,8]

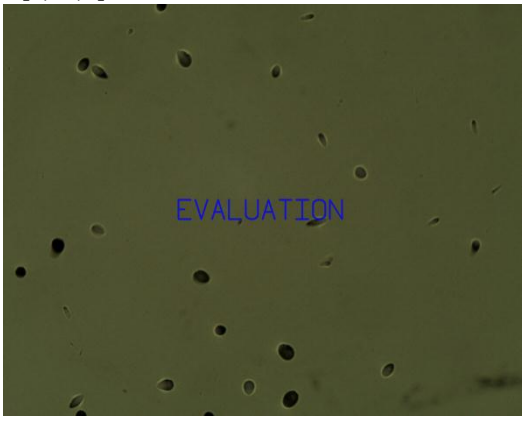

Figure 2:Tracks of alpha particles emitted by radon in a CR-39 detector.

3. Calculations:

The radon concentrations, radon exhalation rate were calculated using the experimental measured average track densities according to the following relations from previous studies [7, 8, 9, 10].

3.1 Determination Radon Concentration:

$$C_{Rn} = K\frac{\rho}{T_{eff}} \qquad 1$$

C_{Rn}: is the radon concentration (Bq/m^3)
K: is the calibration factor = 12.5 Bqm^{-3}/tracks $cm^{-2}d^{-1}$.

ρ : Is the track density (tracks/ cm^2)
T_{eff} : effective time = $[t + (e^{-\lambda t} - 1)/\lambda]$
t : exposure time

3.2 Determination radon exhalation rate per area:

The radon exhalation rate (Ex) of any sample is defined as the flux of radon released from the surface of material. The surface exhalation rate in the building material samples was calculated using equation (2), the radon exhalation rate per area (surface exhalation rate) in units of $Bq\cdot m^{-2}\cdot h^{-1}$ can be obtained by as [8, 9, 10,11, 12].

$$E_x = \frac{CV\lambda}{A[t + (e^{-\lambda t} - 1)/\lambda]} \qquad 2$$

Where:
C: is the integrated radon exposure ($Bq\cdot m^{-3}\cdot h$);
V: is the volume of air in the cup (m^3) = $7.942\times10^{-4}m^3$
λ: is the decay constant for Rn^{222} (h^{-1}) = $7.56\times10^{-3}h^{-1}$
A: is the surface area of the sample (m^2) = $5.0265\times10^{-3}m^2$
t: is the exposure time (h) =124 days =2976 hours

3.3 Determination Radon Exhalation Rate per Mass:

The mass exhalation rate ($Bqkg^{-1}\cdot h^{-1}$) in the building material samples is calculated using the following formula 3:

$$E_M = \frac{CV\lambda}{M[t + (e^{-\lambda t} - 1)/\lambda]} \qquad 3$$

Where E_M is the mass exhalation rate in ($Bqkg^{-1}\cdot h^{-1}$) and M is the mass of sample (kg) **[**8, 9, 10, 11, 12, **13**, **14**, **15].**

3.4 Determination the Annual Effective Dose

The following equation was used to calculate the annual effective dose as giving in equation 4, which used to calculate the dose accumulated in one year of exposure to radon gas as follow:

$$Dose = \epsilon f_{Rn} T_y C_{Rn} \qquad 4$$

Where:
f_{Rn}: is the conversion factor = 9 nSv / (Bq h m-3).
T_y: is the time spent indoors per year = 7000 hours
ϵ: is the equilibrium factor (= 0.4)
C_{Rn} : is the radon concentration.
Substituting the previous parameters in equation (4) we can evaluate the annual effective dose simply according to the following relation 5 [16,17,18].

$$Dose\ (mSv/y) = 0.0252 \times C_{Rn} \qquad 5$$

4. Results and Discussion:

Results and discussion for radon concentrations, radon exhalation rate E_x, and radon exhalation rate E_m for destroyed and fresh building material samples used are given in this section. Equations 1, 2, 3 and 5 respectively were used for calculating radon concentrations, radon exhalation rate in term of area , E_x, radon exhalation rate in terms of mass, E_m, and Annual Dose for destroyed building material samples used in this study which include a red brick, marble, ceramic, concrete, tiles, asbestos, glass, and building stones. The results of Radon concentration only is shown in table 2 for fresh building materials.

a) Results from Fresh building Materials

Table 1: Summary of results of the average radon exhalation rate in terms of area Ex, radon concentration, radon exhalation rate in terms of mass Em and the annual effective dose from all fresh building materials used in [6, 7, 8 ,9, 10].

Dose ($mSv.y^{-1}$)	E_m ($mBq.Kg^{-1}.h^{-1}$)	E_x ($mBq.m^{-2}.h^{-1}$)	C_{Rn} (Bq/m^3)	Sample Type
1.42942464	1.790	71.130	56.7232	Stone
0.83759256	1.040	41.750	33.2378	Marble
2.7083826	3.390	135.020	107.4755	asbestos
2.25698508	2.830	112.520	89.5629	concrete
1.1284938	1.410	56.260	44.7815	tile
0.68712588	0.860	34.260	27.2669	glass
0.92787156	1.160	46.260	36.8203	ceramic
0.9178344	1.150	45.760	36.422	Red Brick
1.36	12.59	67.87	54.04	Aver.

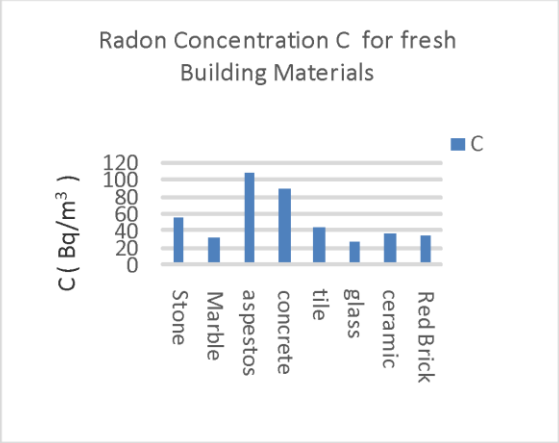

Figure 3 shows the Radon concentration for fresh building materials

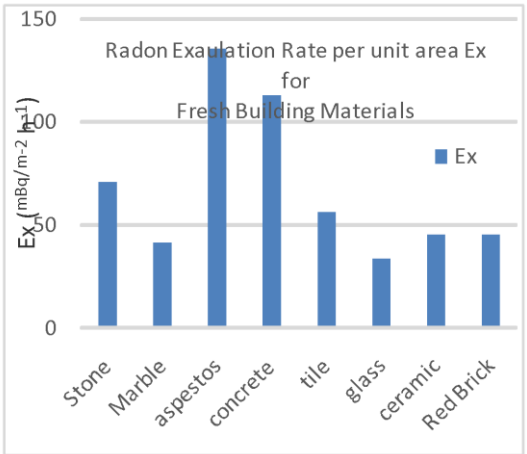

Figure 4 shows the Radon Exaltation rate per unit area for different fresh materials

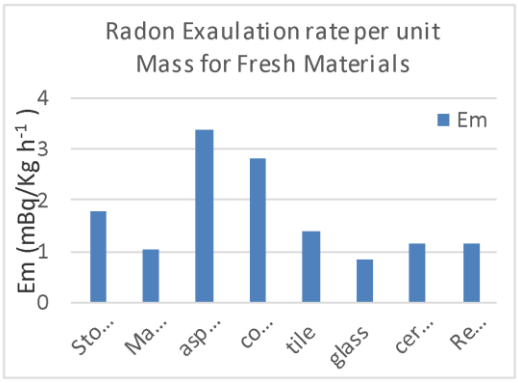

Figure 5 shows the radon exaltation rate per unit mass for fresh materials

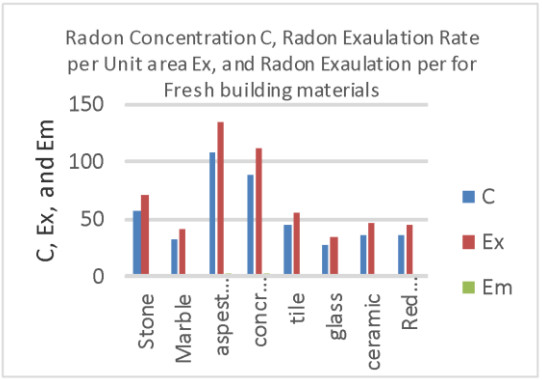

Figure 6 shows a comparison between radon concentration C, Radon exaltation per unit are Ex, and Radon exultation per unit mass Em, for fresh building materials

b) The Results from destroyed building Materials

The radon exhalation rate E_x , and radon exhalation rate E_m , and annual effective dose for each destroyed sample are summarized in Table 2.

Table 2: Summary of results of the average radon exhalation rate in terms of area Ex, radon concentration, radon exhalation rate in terms of mass Em and the annual effective dose from all destroyed building materials used in [6, 7, 8 ,9, 10].

Dose ($mSv.y^{-1}$)	E_m ($mBq.Kg^{-1}.h^{-1}$)	E_x ($mBq.m^{-2}.h^{-1}$)	C_{Rn} (Bq/m^3)	Sample Type
9.464	11.799	469.017	375.580	Concrete
9.3528	11.659	463.895	371.14	Asbestos
4.1144	5.129	204.087	163.28	Tiles
3.831	4.776	190.025	152.03	building stones
2.7126	3.570	133.92	107.65	Ceramic

2.1324	2.658	105.945	84.630	red brick
2.1194	2.642	105.130	84.110	Marble
1.744	2.174	86.506	69.210	glass
4.433	5.550	219.815	175.95	Aver.

The data listed in Table 2 clearly show that concrete, tiles, building stones and asbestos are have high radon exhalation rate in terms of area E_x, radon concentration, radon exhalation rate in terms of mass E_m and the annual effective dose. But the glass have low radon exhalation rate in terms of area E_x, radon concentration, radon exhalation rate in terms of mass E_m and the annual effective dose.

The Figure 7 shows the comparison between destroyed building materials in terms of the average radon exhalation rates in term of area where the concrete have the highest value.

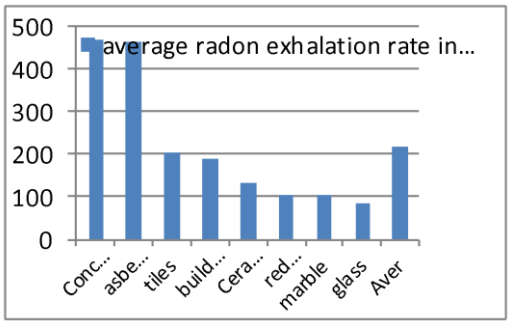

Figure 7: This figure shows the Comparing histogram for the average radon exhalation rates in term of area.

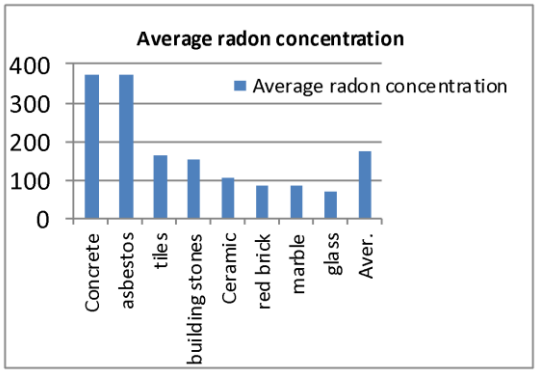

Figure 8: This figure shows the Comparing histogram for the average radon concentration rates

The Figure 8 shows the comparison between destroyed building materials in terms of the average radon concentration rates where the concrete have the highest value. The Figure 9 shows the comparison between destroyed building materials in terms of the average radon exhalation rates in term of mass where the concrete have the highest value with 11.79 $mBq.kg^{-1}.h^{-1}$ then asbestos with 11.65 $mBq.kg^{-1}.h^{-1}$ then (tiles, building stones, ceramic, a red brick, marble and glass) with(5.12, 4.77, 3.57, 2.65, 2.64 and 2.17) $mBq.kg^{-1}.h^{-1}$respectively. Note that the glass has the lowest value of the materials studied.

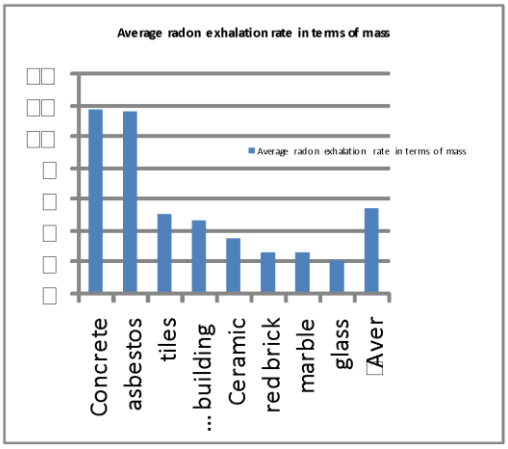

Figure 9: This figure shows the Comparing histogram for the average radon exhalation rates in term of mass

The Figure 10 shows the comparison between destroyed building materials in terms of the average annual effective dose for radon gas where the concrete have the highest value with 9.46 $msv.y^{-1}$ then asbestos with 9.352 $msv.y^{-1}$ then (tiles, building stones, ceramic, a red brick, marble and

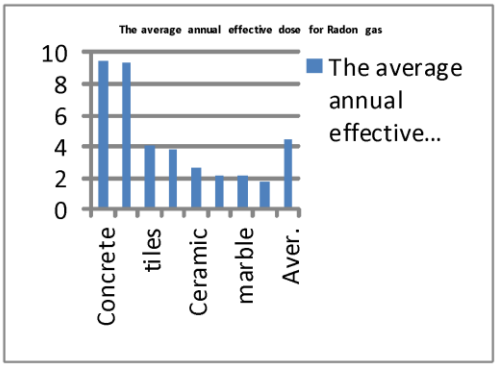

Figure 10: This figure shows the Comparing histogram for the average annual effective dose for radon gas.

Glass) with (4.11, 3.83, 2.71, 2.13, 2.11 and 1.74) $msv.y^{-1}$ respectively. Note that the glass has the lowest value of the materials studied.

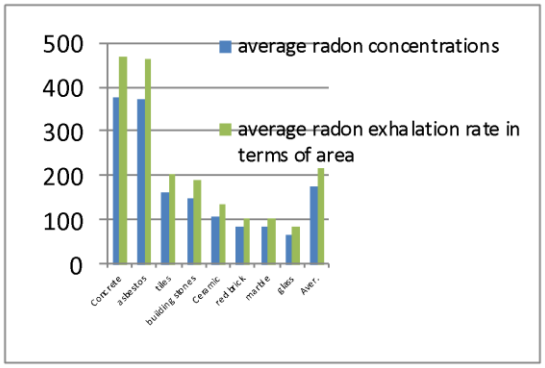

Figure 11: This figure shows the Comparing histogram for the average radon concentrations (C_{Rn} Ave.) and exhalation rates (E_x Ave.) from destroyed building materials

In Figure 11 We notice that, the concrete have the highest value of the average radon concentration and the average radon exhalation rate in term of area E_x, then (asbestos, tiles , building stones , ceramic, a red brick , marble and glass) respectively.

c) **Comparison Between fresh and destroyed building materials:**

Figure 12 show comparison between fresh and destroyed building materials for the Radon concentration C

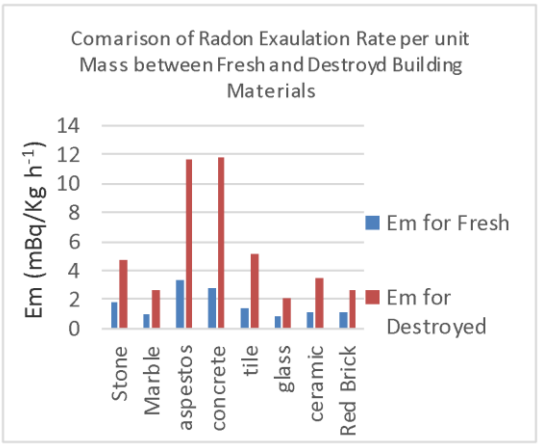

Figure 12 show a comparison between the fresh and destroyed building materials for the Radon Exultation rate per unit area

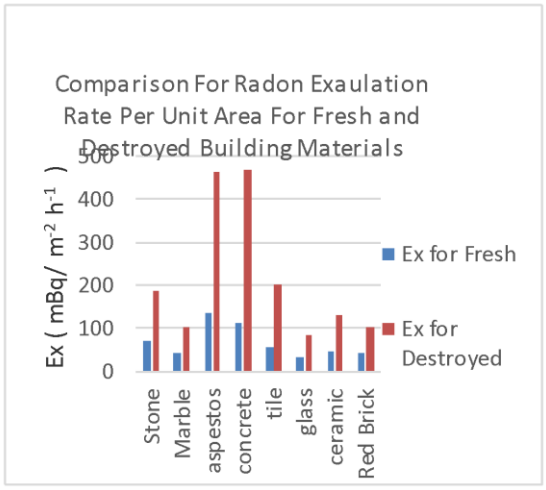

Figure 13 show the comparison of radon Exultation rate per unit mass for fresh and destroyed building materials

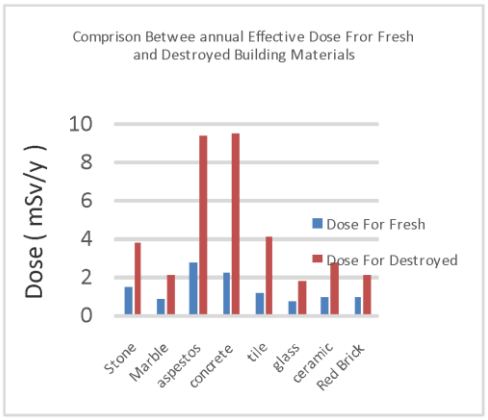

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Figure 14 shows the comparison of the annual effective dose between fresh and destroyed building materials

4. Conclusion

Using the closed can technique and the solid state nuclear track detectors (CR-39), we measured the radon exhalation rate from building material samples used in Jabalia in order to assess the contribution of individual material (e.g. red brick , marble, ceramic, concrete, tiles, asbestos, glass, and building stones) to the total indoor radon exposure of the inhabitants of Jabalia district. The corresponding radon concentration, and the annual effective dose were determined and compared with the effective dose limit values recommended by the National Council on Radiation Protection which (from 1 to 5 mSv/y). Results obtained from the current study show that the radon exhalation rates from asbestos and concrete have relatively high values as compared to other building material samples followed red brick, marble, ceramic, tiles, glass, and building stones contribute less to the indoor radon. From the results of our study we can conclude that the Concrete have the maximum values of radon concentrations 375.58Bq/m^3, radon exhalation rate in term of area469.017mBq.m^{-2}.h^{-1}, radon exhalation in term of mass 11.799 mBq.Kg^{-1}.h^{-1} and the annual effective dose 9.464 mSv.y^{-1},also asbestos have maximum values of radon concentrations 371.14 Bq/m^3, radon exhalation rate in term of area 463.895 mBq.m^{-2}.h^{-1}, radon exhalation rate in term of mass 11.659 mBq.Kg^{-1}.h^{-1} and the annual effective dose 9.3528 mSv.y^{-1}. But the glass have the minimum values radon concentrations 69.21 Bq/m^3, radon exhalation rate in term of area 86.506 mBq.m^{-2}.h^{-1}, radon exhalation rate in term of mass 2.174 mBq.Kg^{-1}.$h^{-1}$and the annual effective dose 1.744 mSv.y^{-1}. In comparison with the annual effective dose of Radon by NCRP, we found that concrete and asbestos are 9.46 and 9.35 mSv/y, are much higher than the proposed limit which is 1 to 5 mSv/y, and all other material are below the limit. There are many researchers studied radon gas for building materials, comparison with previous studies will be shown in following tables, the results obtained in Sudan are in table 4[19]:

Table 3: Results from Sudan (Elzain) [19]:

Dose ($mSv.y^{-1}$)	Em ($mBq.Kg^{-1}.h^{-1}$)	Ex ($mBq.m^{-2}.h^{-1}$)	CRn (Bq/m^3)	Sample Type
3.59	2.84	240	128	Ceramics
5.32	4.21	355	190	Red brick
5.52	4.37	369	197	Block
6.01	4.76	402	214	Ispistos

The results obtained in Palestine are in table 4 [7]:

Table 4: Results from Palestine, Nabulus (Shoqwara) [7]:

Dose ($mSv.y^{-1}$)	Em ($mBq.Kg^{-1}.h^{-1}$)	Ex ($mBq.m^{-2}.h^{-1}$)	CRn (Bq/m^3)	Sample Type
6.06	3.01	438.79	240.55	marble
4.88	2.59	347.42	193.71	ceramic
4.52	2.46	325.38	179.37	concrete
3.70	1.95	268.59	147.00	building stones

In comparison with values we measured with other people values, we see that we are very close in numbers, the differences are due to the different in origin of building materials, and the different in calibration numbers from place to other place. Also we compare the destroyed building materials with the fresh one, we see a big difference, which mean there is be might a big pollution from the war and we may need more accurate devices to detect radio nuclei like Uranium.

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دراسة مقارنة عن معدل انبعاث غاز الرادون من مواد البناء الجديدة والمدمرة في جباليا-غزة، فلسطين

وتهدف هذه الدراسة إلى تقييم مساهمة مواد البناء الجديدة والمدمرة في الحرب عام 2014 نحو إجمالي التعرض للرادون في الأماكن المغلقة لسكان منطقة جباليا في غزة. تم جمع 40 عينه من مواد البناء المدمرة في عام 2014 في منطقة جباليا، و 40 عينة أخرى مماثلة من مواد البناء الجديدة من غزة .وقد استخدمت نظام العلب المغلقة في هذه الدراسة باستخدام كاشفات المسار النووي للحالة الصلبة (CR-39) . بعد 124 يوما من التعرض للرادون، تم معالجة الكواشف كيميائيا من قبل (6N) هيدروكسيد الصوديوم في 75 درجة مئوية لمدة 4.20 ساعة، ثم تم عد المسارات باستخدام المجهر الضوئي .أظهرت النتائج أن متوسط معدل إنبعاث الرادون في المساحة من مواد البناء الجديدة في العينات المدروسة تراوحت بين ($mBq.m^{-2}.h^{-1}$) 27.27[1]) للعينات الزجاجية إلى ($107.48\ mBq.m^{-2}.h^{-1}$) لعينات الأسبستوس ، في حين تراوحت المواد المدمرة من ($86.51 mBq.m^{-2}.h^{-1}$) للعينات الزجاجية إلى ($mBq.m^{-2}.h^{-1}$ 463.90) لعينات الأسبستوس .واستنادا إلى هذه القيم، تم أيضا تحديد الجرعة الفعالة السنوية لكل عينة ومقارنتها بقيم الجرعة الفعالة التي أوصى بها المجلس الوطني للحماية من الإشعاع والقياسات (من 1 إلى 5 ملي سيفرت / السنة) . وبصفة عامة، فإن الجرعات الفعالة السنوية من مواد البناء المدمرة التي تم التحقيق فيها منخفضة وتحت القيمة العالمية باستثناء الخرسانة والأسبستوس من العينات المدمرة بمتوسط قيم (9.464) و (9.3528) ملي سيفرت / سنة على التوالي، ومن عينات مواد البناء الجديدة الجرعة الفعالة هي (2.25) للخرسانة و (2.71) للأسبستوس .هناك اختلافات كبيرة بين النتائج من المواد الجديدة والمواد المدمرة، والتي قد تكون قد تأثرت بتلوث الحرب.

كلمات مفتاحية:

CR-39- قياس تركيز غاز الرادون – معدل إنبعاث غاز الرادون – مواد البناء المدمرة والجديدة