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Measurement of Radon Exhalation Rates from Different Rock Types and Construction Materials (Gaza Strip, Palestine)

Farklı Kayaç Tipleri ve Yapı Malzemelerinde Radon Gazı Salınımı Ölçümleri (Gazze Şeriti, Filistin)

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ABSTRACT
Indoor radon increases the health hazard due to long-term exposure. Most building materials of natural origin contain small amount of naturally occurring radioactive materials. The building materials of natural origin reflect the geology of their site origin. This study was carried out to assess the radon activity concentration in rock and building materials used in construction purposes in the Gaza Strip, southwestern of Palestine. Fourteen different construction materials of imported (international) and local origin were tested, using solid state nuclear track detectors (CR-39). After 55 days of exposure, CR-39 detectors were etched chemically and then counted under an optical microscope. The radon concentration level of studied samples ranges from 94.4 to 642.5 Bq/m3. The sands (from north of Gaza Strip), black cement, gray granite and the marble show relatively highest levels with values about 642.5, 285.0, 283.6, and 257.2 Bq/m3, respectively. These values are above the international standard limits, and they are not safe for use in construction purposes. According to Ubeid and Ramadan (2017), the highest value in sands are referred to black sands, agricultural run-off and urban areas, discharges from mining activities, factories and municipal sewer systems, leaching from dumps and former industrial sites. While, the high value in gray granite is related to high percentage of silica and potassium contents, the high value of radon concentration in the marble is interpreted to high contents of organic matter in the original limestone before the metamorphism. On the other hand, values on radon concentration in the waste-dust of marble and granite from industrial quarry were 399.7 and 257.2 Bq/m3, respectively. They were above the international standard limit, and generally the ambient is not safe for workers.

Keywords: *Radon exhalation, building materials, rock fragments, Gaza Strip.*

ÖZ
Uzun süre maruz kalındığında evlerdeki radon gazı sağlık sorunlarının artışına neden olmaktadır. Yapı malzemelerinin çoğunda doğal olarak az miktarlarda radyoaktif maddeler bulunmaktadır. Doğal yapı malzemeleri bulundukları yerin jeolojisini yansıtmaktadır. Bu çalışma, güneybatı Filistin'de, Gazze Şeritinde kullanılmakta olan kayaç ve yapı malzemelerinin bu anlamda değerlendirilmesi ile ilgilidir. Uluslararası ve yerel kökenli yapı malzemelerinden alınan farklı on dört malzeme üzerinde nükleer iz detektörleri (CR-39) ile testler yürütülmüş ve optik mikroskop altında ölçmeler yapılmıştır. Ölçülen örneklerde radon konsantrasyon seviyeleri 94.4 ila 642.5 Bq/m3 arasındadır. Kumlar (Gazze Şeridinin kuzeyinden), siyah çimento, gri granit ve mermer görece yüksek, sırasıyla 642.5, 285.0, 283.6, ve 257.2 Bq/m3 değerler vermektedir. Bu değerler uluslararası standart sınırların üzerindedir ve yapı işlerinde kullanılmaları sağlıklı değildir. Ubeid ve Ramadan (2017)'a göre, kumlardaki en yüksek değer siyah kumlarda, tarımsal ve kentsel alanlarda, madencilik faaliyetlerindeki atıklardan, fabrika ve şehir kanalizasyonlarından ve eski çöplük ve sanayi alanlarından sızmalarla olmaktadır. Gri granitteki yüksek değerler yüksek silika ve potasyum içeriklerine bağlı iken mermerdeki yüksek radon konsantrasyonu metamorfizma öncesi ilksel kireçtaşındaki organik maddelerin yoğunluğuna bağlı olduğu düşünülmektedir. Öte yandan maden ocaklarındaki mermer ve granit toz atıklarında sırası ile 399.7 ve 257.2 Bq/m3 radon gazı konsantrasyonu ölçülmüştür. Bunlar uluslararası standart sınırların üzerindedir ve çalışma ortamı çalışanlar için sağlıklı değildir.

Anahtar Kelimeler: *Radon salınımı, inşaat malzemeleri, kayaç parçaları, Gazze Şeriti.*

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INTRODUCTION

Man is generally exposed to ionization from naturally occurring radioactive materials (NORM). The source of these materials is the earth`s crust, and they find their way into building materials, and the human body itself (ATSDR, 1990; Axelson, 2004). In many parts of the world, building materials containing radioactive material have been used for generations. As individuals spend more than 80% of their time indoors, the internal and external radiation exposure from building materials creates prolonged exposure situations (ICRP, 1999; UNSCEAR, 2000).

Most building materials of terrestrial origin contain small amounts of NORM, mainly radionuclides from the ^{238}U and ^{232}Th decay chains and the radioactive isotope of potassium, ^{40}K. The external radiation exposure is caused by the gamma emitting radionuclides, which in the uranium series mainly belong to the decay chain segment starting with ^{226}Ra. The internal (inhalation) radiation exposure is due to ^{222}Rn, and marginally to ^{220}Rn, and their short lived decay products, exhaled from building materials into the room air.

Generally, natural building materials reflect the geology of their site of origin. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the Earth's crust are 35, 30 and 400 Bq/kg respectively (UNSCEAR, 2000). However, elevated levels of natural radionuclides causing annual doses of several mSv were identified in some regions around the world, e.g. in Brazil, France, India, Nigeria, and Iran.

To investigate radon exhalation from different rock materials, soils, and construction materials, many authors used the sealed cup technique (e.g. Khan et al., 1992; Singh et al., 1999; Khayrat et al., 2001; Abo-Elmaged and Daif, 2010; Maged and Ashraf, 2005; Rafique and Rathore, 2013; Najam et al., 2013; Chauhan et al., 2008; 2014; Sharma et al., 2016). This method is based on using sealed cup equipped with solid-state nuclear track detector (SSNTD) such as CR-39. The track density due to alpha particles from radon that entered the air space in the hollow holder were registered in the CR-39 plastic detector, used to calculate the exhalation rates and effective radium content (Somogyi, 1990).

The aim of this study is to assess the radioactivity concentration of radon in different rock-type fragments and building materials used in construction purposes in the Gaza Strip, Palestine.

Geographical setting

In this study, all the samples for measuring the radon exhalation rate in rock fragments and materials used in constructions were collected from a local market in the Gaza Strip. The Gaza Strip is a narrow coastal region that is located at the southwestern part of Palestine, and in the south eastern coastal plain of the Mediterranean Sea (Fig. 1). Gaza Strip is bordered by Egypt from the south, Negev desert from the east and the green line from the north (GEP, 1994; Change to (Ubeid and Albatta, 2014). The width of the strip ranges between 6 km at the middle to 8 km in the north and 12 km in the south. Its length is about 40 km along the coastline and its area is about 378 km^2 (UNDP, 2009).

The population of the Gaza Strip continues to grow rapidly. With a Palestinian population growth rate of around 3.5 percent per annum that would result in a doubling of the population in 15 years. It ranks as the third most densely populated polity in the world, with around 2 million Palestinians on the Strip.

The climate of Gaza Strip is characterized by mild winters, and dry, warm to hot summers. The average mean daily temperature ranges from 26 °C in summer to 12 °C in winter. The average annual rainfall is 335 mm per year, and the average annual evaporation amounts to 1300 mm.

Buildings in the Gaza Strip are mainly constructed from bricks, cement, gypsum, lime, sand, pebbles, granite, and marble (Fig. 2 and 3). Most of these materials are imported from outside of the Palestinian Authority, e.g. from Israel, Turkey and Egypt; except the sands, which are excavated from sand dunes at the coast of the Gaza Strip.

Methodology

Fourteen samples of rock fragments and building material were collected from local markets and an industrial quarry in the Gaza Strip. Four of samples were rock fragments used in construction purposes such as granite and marble. They were crushed to fine-grained. Five of total samples were building materials such as cement and lime. Two samples of waste-dust were collected from an industrial quarry. The sources of exported countries for both gray granite and schist were unlimited while marbles are the products of the West Bank, Palestine. The rest

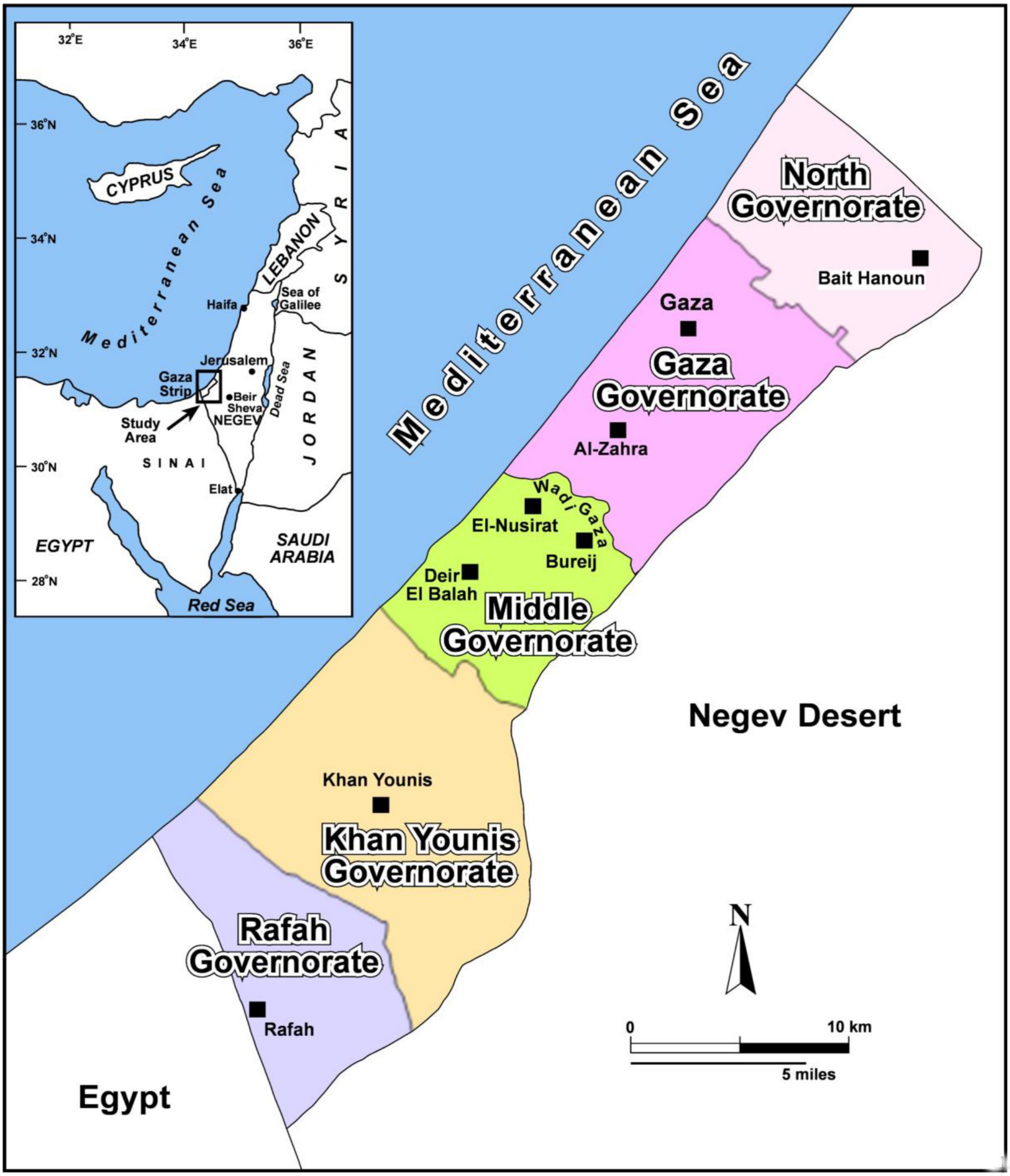

Figure 1. Location map of Gaza Strip.
Şekil 1. Gazze Şeridinin bulduru haritası.

of samples represent the local sands used in constructions from different places of Gaza Strip. All the samples were dried at 110 °C for 24 hours.

CR-39 sheets as radon detector (sealed cup technique) were used in this study. The detectors are cut in small pieces (usually 1cm x2cm) and sticked at the top of the plastic container (3liters, diameter 14 cm) (Fig. 4). Around 300 mL of the collected samples were packed and sealed at the bottom of container; the detector is exposed to radon for a time period of 55 days. The CR-39 films were chemically etched using a 6 M solution of NaOH, at a temperature of 70

Figure 2. Uses of rock types in the buildings of the Gaza Strip. A, B, C, E and F: uses of metamorphic rocks. Note in E and F, the fossils and bioclasts (circled parts) in Al-Khalili marbles, metamorphosed from bioclastic limestones. D: use of igneous rocks.

Şekil 2. Gazze Şeridinde binalarda kulanılan kayaç tipleri. A, B, C, E ve F: metamorfik kayaç kullanımı; E ve F'teki, biyoklastik kireçtaşlarından başkalaşıma uğramış Al-Halili mermerlerindeki fosillere ve biyoklastlara (halkalı kısımlar) dikkat ediniz. D: magmatik kayaç kullanımı.

^{o}C, for about 6 hours. The detectors were washed with distilled water and left to dry. Each detector was counted visually using an optical microscope through the area within 3 mm^2 in 4 distinct regions. The average number of tracks/mm^2 was determined, from the measured average track densities on the CR-39 with detector sensitivity 12.3 Bq/m^3 per tracks/cm^2/day. Radon calculations in this study are carried out using the following equation:

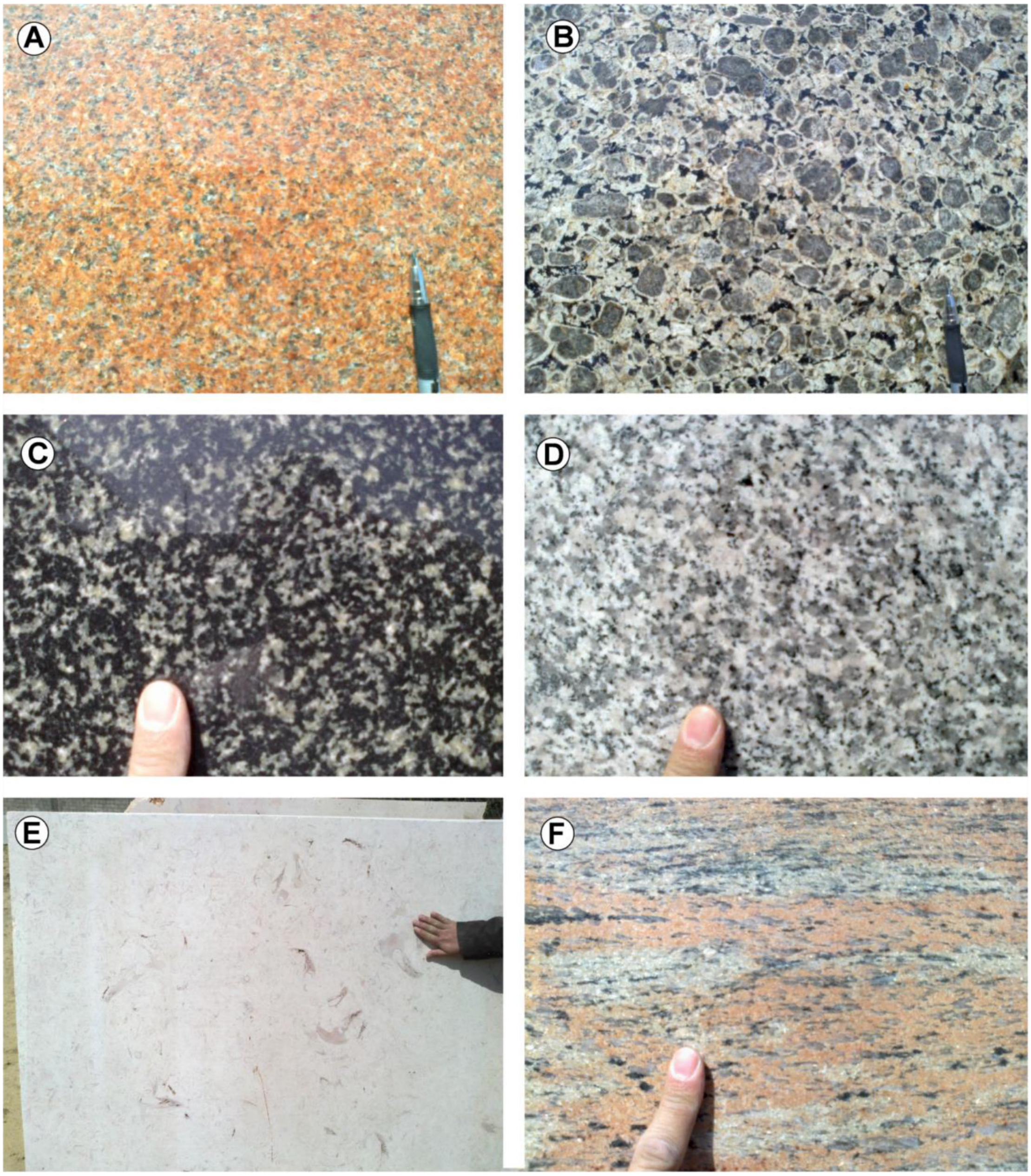

Figure 3. Some samples of rocks used in construction purposes in Gaza Strip. A, B, C and D represent plutonic igneous rocks. E and F represent metamorphic rocks.

Şekil 3. Gazze Şeridinde inşaat amaçlı kulanılan kayaçlardan bazı örnekler. A, B, C ve D magmatik, Eve F ise metamorfik kayaçları göstermektedir.

$$E = \frac{\rho}{\eta A}\left[\frac{\lambda V}{T_{eff}}\right], \qquad C_{Rn} = \frac{\rho}{\eta T_{eff}},$$

$$C_{Ra} = \frac{\rho V}{\eta M T_{eff}}, \qquad AED = 0.0252\, C_{Rn}$$

Where, E: is the radon exhalation rate (Bq/m^2/h), C_{Rn}: is the radon concentration (Bq/m^3), C_{Ra}: is the effective radium content (Bq/kg), ρ: is the track density (tracks / cm^2), η : is the detector sensitivity (tracks/cm^2/h/ Bq/m^3), λ : is the decay constant (λ= 7.56 x 10^{-3} h^{-1}), V: is the effective volume of the container (cm^3), A: is the area of the sample (cm^2), and M: is the

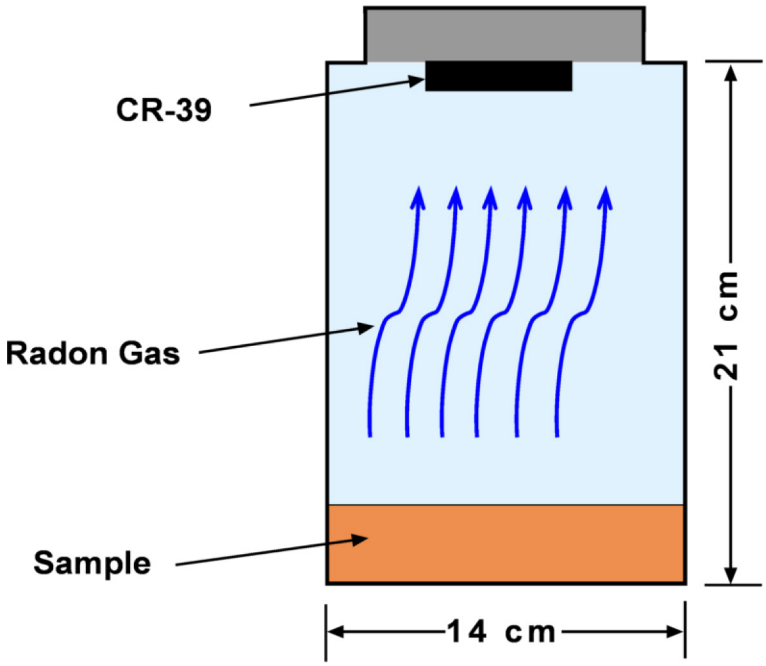

Figure 4. Experimental setup for the measurement of radon exhalation rate.
Şekil 4. Radon salınımını ölçmek için hazırlanmış deney düzeneği.

mass of the sample (kg) (Baykara and Dogru, 2006; Baykara, et al., 2005; Sroor, et al., 2001). The annual effective dose (AED) equivalent was calculated from radon concentrations in sense Guo and Cheng (2005).

The annual effective dose equivalent was calculated from radon concentrations. According to UNSCEAR (2000), the difference in radon doses and recommended a radon effective dose conversion factor of 9 nSv per (Bq h m^{-3}). Assuming 7000 hours per year indoor (an indoor occupancy factor of 80%) and an equilibrium factor of 0.4 (Chen, 2005), using the mentioned recommendation (UNSCEAR, 2000), the effective dose for one year radon exposure is calculated using the relation (Guo and Cheng, 2005),

$$AED = \varepsilon f_{Rn} T C_{Rn}$$

Where, f_{Rn}: is the conversion factor = 9 nSv / (Bq h m^{-3}), T: is the time spent indoors per year = 7000 hours, ε: is the equilibrium factor (= 0.4), C_{Rn}: is the radon concentration.

Results and Discussions

From analyzed samples for radon exhalation, the data is listed in Table 1. It shows that the plutonic acidic igneous rocks samples (granitic rocks) had radon concentration values range from 148.2 to 283.6 Bq/m^3. The highest value (283.6 Bq/m^3) was detected in gray granite where as the lowest value (148.2 Bq/m^3) was depicted in greenish granite. The sample of granite waste-dust which collected from industrial quarry shows high concentration level of radiation with value about 257.2 Bq/m^3.

The concentration of radiation in granite possible referred to constituents of uranium and thorium in acidic igneous rocks such as granite rocks (Evans and Goodman, 1941; Senetle and Keevil, 1947; Jevereys, 1952). Where, the granitic rocks characterized by their high constituent of the silica (more than 63%), which directly proportional to radioactivity. The process of crystallization form silicate minerals in such as quartz, plagioclase, and perthite from the acidic magma at high temperature. Then, uranium and thorium will tend to be concentrated in the residual liquid, eventually being incorporated in minerals like the complex rare earth silicates, uraninite, monazite and uranothorite, or perhaps remaining as films among the previously solidified crystals (Evans and Goodman, 1941).

Additionally, the potassium is a relatively abundant in most granitic rocks, which reaches up to 2.9% (Daly, 1933), especially these rocks tend to gray color. This

Table 1. Radon exhalation rate, radon concentration, effective radium content and the annual effective dose of studied samples.
Çizelge 1. Radon salınım hızı, radon konsantrasyonu, efektif radon içeriği ve ölçülen örneklerde yıllık efektif dozlar.

Sample No.	Type	Source of Sample	E ($mBq/m^2/h$)	C_{Rn} (Bq/m^3)	C_{Ra} (Bq/kg)	Dose (mSv/year)
G-1	Granite waste-dust (Industrial quarry (Gaza Strip))		341.17	257.2	1.56	6.5
G-2	Gray granite	?	321.65	283.6	2.24	7.1
G-3	Greenish granite		168.03	148.2	0.50	3.7
M-1	Granite schist	?	107.03	94.4	0.43	2.4
M-2	Marble waste-dust (Industrial quarry (Gaza Strip))	El Khalil (West Bank)	530.21	399.7	2.88	10.1
M-3	Marble	El Khalil (West Bank)	272.84	240.6	1.14	6.1
BM-1	Gypsum	Egypt	135.91	102.4	1.32	2.6
BM-2	Lime ($Ca(OH)_2$)	Egypt	140.94	106.2	2.11	2.7
BM-3	Black cement	Turkey	373.61	281.6	2.66	7.1
BM-4	White cement	Turkey	308.17	232.3	2.21	5.9
BM-5	Black cement	Israel	378.08	285.0	2.95	7.2
BM-6		South Gaza Strip	173.3	171.8	0.97	4.3
BM-7	Sand	Middle Gaza Strip	133.6	132.4	0.73	3.3
BM-8		North Gaza Strip	857.1	642.5	4.79	21.4

suggests the high concentration level of radiation in the gray granite.

On the other hand, the high level of radiation in granite waste-dust from industrial quarry could be referred to accumulation of dust with time due to working in the place, and it possibly other materials than the granite dust.

It is remarkable that the annual effective dose (AED) of the plutonic igneous rock samples range from 3.7 to 7.1 mSv/year. The highest values come from granite waste-dust from industrial quarry, and gray granite sample (6.5 and 7.1 mSv/year respectively). They were above the standard limit of the National Council on Radiation Protection and Measurements (1-5 mSv/year) (NCRP, 1987). Whereas, the AED of the rest sample were below the standard limit.

In case of metamorphic samples (marble and granite schist), it was observed that the radon concentration varied from 94.4 to 399.7 Bq/m^3. The high level of the radiation is referred to chemical composition of the marble, metamorphosed from limestone and dolomite rocks, which consist of calcium and magnesium carbonates. The Palestinian marbles, especially Al-Khalili marbles that are excavated from Al-Khalil mountains in the West Bank were metamorphosed from bioclastic limestones. That means, it was made of bioclasts and fossils that had built there skeletals from carbonates (Fig. 2E and F). This suggests that these organic matters adsorbed the radon during their sedimentation before the metamorphism (Gurari et al., 1984; Kochenov and Baturin, 2002).

The AED of the metamorphic rock samples (marble and granite schist) varied between 2.4 and 10.1 mSv/year. This indicates that the marble was above the standard limits (1-5 mSv/year) (NCRP, 1987).

The granite schist shows the lowest concentration value (94.4 Bq/m^3) comparing with studied rock fragment samples (Fig. 5), and AED values up to

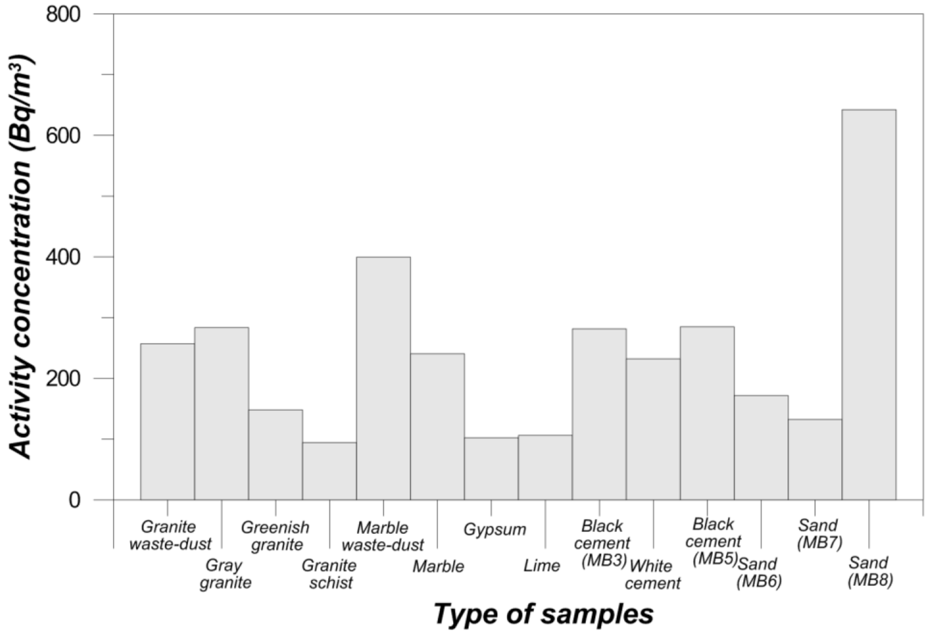

Figure 5. The radon concentration in the studied samples (MB5=sample number).
Şekil 5. Ölçülen örneklerdeki radon konsantrasyonları (MB5=örnek numarası).

2.4 mSv/year. It is below the standard limit. The low value in this type of rock possibly is referred to the effect to the heat and pressure generated within the earth depths (Jacobs, 1956). The levels of concentration of radioactivity in meteorites are low (Evans and Goodman, 1941; Arrol et al., 1942).

For construction material samples, the concentration of radon values ranges from 102.4 to 285.0 Bq/m^3 (Table 1), where higher values were observed in the Israeli and Turkish materials. The white and black cement samples from Turkey had values 232.3 to 281.6 Bq/m^3 respectively, and the black cement sample from Israel recorded the highest concentration values with up to 285.0 Bq/m^3. Whereas, the gypsum and lime from Egypt had lower concentration values up to 102.4 and 106.2 Bq/m^3 respectively.

The AED in the construction material samples (black and white cement) from Israel and Turkey range from 5.9 to 7.2 mSv/year, which were above the standard limits (1-5 mSv/year) (NCRP, 1987). Whereas, the AED in the gypsum and lime samples from Egypt were below the standard limits.

Overall high concentration values were observed in plutonic acidic igneous rock samples (granitic rocks) especially those with gray to black colors, and metamorphic rock samples (marble rock). This could indicate high contents of radium in these rocks (Durani et al., 1997; Morawsk, 1989). Therefore, the granite and marble rocks can be a significant source of radon in buildings. Thus, these types of rocks are not safe to use in constructions, where they are usually used in tiling especially in kitchens and stairs in Palestinian homes (Fig. 2). Also, the industrial quarries are not safe, where the waste-dust samples from materials that used in constructions had high radon concentration values above the standard limits.

The sands that are used in Gaza Strip as building mater were excavated from sand dunes of beach and coastal areas. Ubeid and Ramadan (2017) had observed that the sands in the northern parts of Gaza Strip had high concentration values of radon exhalation. The corresponding radon concentration value was up to 4.79 Bq/kg, and value of AED was 21.4 mSv/year. These sands had the highest radioactivity concentration between the studied samples (Fig. 5). It was above the standard limits (1-5 mSv/year) (NCRP, 1987), and they concluded these sands are not safe to use as building materials (Table 1). The authors suggested that the sources of the pollution are black sands, agricultural run-off and urban areas, discharges from mining, factories and municipal sewer systems, leaching from dumps and former industrial sites. Whereas the radon concentration levels in the middle and southern parts of Gaza Strip were 0.7 and 4.79 Bq/kg respectively, the corresponding values of AED were 3.3 and 4.3 mSv/year. They were below the standard limits, and are safe to use in constructions.

This study shows that the sands in northern part of Gaza Strip, the marble and the gray granite show highest activity concentration values, while the granite schist and the sands in middle part of Gaza Strip show the lowest activity concentration levels (Fig. 5).

Also, good correlation are found between both the radon concentration (C_{Rn}) and the annual effective dose (AED) with the effective radium content (C_{Ra}) as depicted in Figure 6A and B.

Conclusions

The study was carried out to assess the radon activity concentration in rock fragments and construction materials used in the Gaza Strip, southwestern

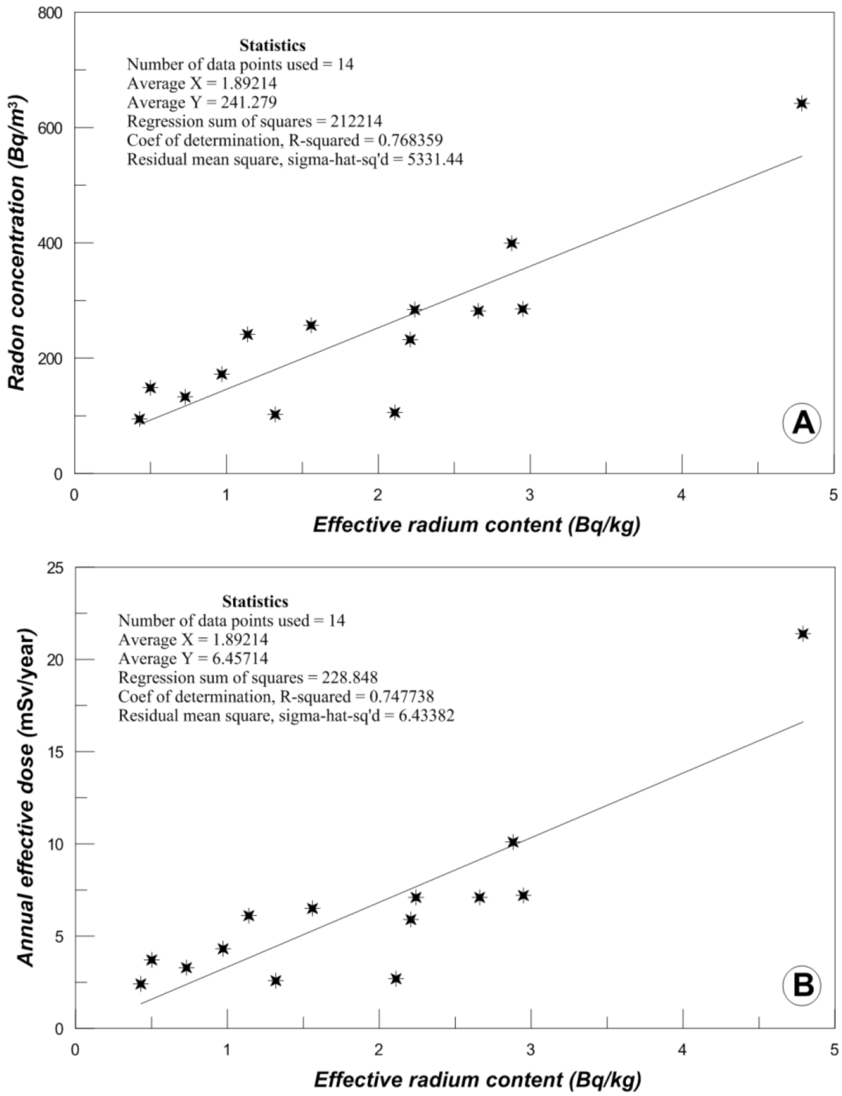

Figure 6. Correlation of the radon radioactivity results. (A) The correlation between the effective radium content (Bq/kg) and the radon concentration (Bq/m^3) of the studied samples. (B) The correlation between the effective radium content (Bq/kg) and the annual effective dose (mSv/year) of the studied samples.

Şekil 6. Radon radyoaktivitesi sonuçlarının korelasyonu. A) Örneklerdeki efektif radon içeriği (Bq/kg) ile radon konsantrasyonu (Bq/m3) arasındaki korelasyon. B) Örneklerdeki efektif radon içeriği (Bq/kg) ile yıllık efektif doz (mSv/yıl) arasındaki korelasyon.

of Palestine. Fourteen samples are collected and tested by using the closed can technique and the solid state nuclear track detectors (CR-39) to determine radon exhalation rate. The values of radon activity were ranged from 94.4 to 642.5 Bq/m^3. The highest values were observed in sands (from north of Gaza Strip), black cement, gray granite, and the marble show relatively highest levels with values about 642.5, 285.0, 283.6, and 257.2 Bq/m^3 respectively. They are above the standard international limits, and they are not safe to use as construction materials. In the sense of Ubeid and Ramadan (2017), the highest value in sands are referred to agricultural run-off and urban areas, discharges from mining, factories and municipal sewer systems, leaching from dumps and former industrial sites. While, the high value in the gray granite is related to high percentage of silica and potassium contents, the high value of radon concentration in the marble is interpret to high contents of organic matter in the original limestone before the metamorphism. In waste-dust samples from industrial quarry the radon concentration level is above the international standard limit. The ambient was polluted and is not safe for workers.

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