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RESEARCH ARTICLE

Determination of radon exhalation rates in soil samples using sealed can technique and CR-39 detectors

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Abstract

Background In this study, the so-called sealed can technique dosimeters have been used to determine the radon exhalation rates in soil samples collected from different sites in Bethlehem region- Palestine.

Methods For the measurement of radon concentration emanated from these samples, alpha-sensitive, Solid State Nuclear Track Detectors (SSNTD's) have been used. A total of 82 soil samples were collected simultaneously.

Results It was found that the radon concentrations in these soil samples varied from 19.1 Bqm^{-3} to 572.9 Bqm^{-3} with an average value of 145.0 Bqm^{-3}. The radon exhalation rate in these collected samples also varied from 6.9 $mBqm^{-2}\ h^{-1}$ (0.26 $mBqkg^{-1}\ h^{-1}$) to 207.2 $mBqm^{-2}\ h^{-1}$ (7.84 $mBqkg^{-1}\ h^{-1}$) with an a total average value of 52.2 $mBqm^{-2}\ h^{-1}$ (1.97 $mBqkg^{-1}\ h^{-1}$).

Conclusions All the values of radium content in all samples under test were found to be quite lower than the corresponding the global value 30 $Bqkg^{-1}$. The present results show that the radon concentration and the resulting doses in all soil samples are below the allowed limit from ICRP. The radiological health implication to the population that may result from these doses is found to be low. The measurements have been taken as representing a baseline database of values of these radionuclides in the soils in the area. The results were compared with national and worldwide results.

Keywords Radon exhalation rate · Can technique · Radium content · CR-39 detectors

Background

About 55% of the natural radiation dose which people accepted is contributed from radon and its short-life progenies. More attentions are paid to the issue of the health risks from them [1]; therefore, the exposure of population to high concentrations of radon and its daughters for a long period leads to pathological effects like the respiratory functional changes and the occurrence of lung cancer [2].

Radon is one of the naturally occurring radioactive elements in the environment produced from the radioactive decay of radium isotopes, which are the decay products of ^{238}U, ^{232}Th and ^{235}U. Hence the concentrations of uranium and thorium in the soil types determine the amount of radon produced in the soil [3]. The infiltration of radon gas (^{222}Rn) from the soil has been identified as one of the main mechanisms influencing indoor radon levels in many buildings. It was reported that a worldwide average of 60% of indoor radon comes from the ground and the surrounding soil of buildings [4]. Therefore; it is of significance for radon control to delineate the potential high radon zone combining the local geological background. Especially in Palestine, the environmental radon survey started relatively late [5–7]. A large-scale indoor radon measurement has not been conducted yet. However, the database based on regional geological survey has been built in many localities. A great number of radiological data, such as the uranium, thorium contents and the distribution of soil texture are available [8–12]. The processes effective in transporting ^{222}Rn from the soil to the surface are related directly to the size and configuration of the space occupied by the soil gas. Radon concentrations in soil pores at depth are dependent directly upon the radium content of the soil, emanating power for radium and soil moisture content.

Exhalation designates the escape of radon from a material to the atmosphere. In the soil, radon molecules can escape from grains of soil by diffusion or recoil into the soil pores, this process is called emanation [13]. The number of radon atoms released per unit surface area per unit time from a

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material is termed as exhalation rate [11]. Normally the dominant contributor to indoor radon is the emanation from soil and fractured bedrock close to the surface. If uranium rich material lies close to the surface of the earth, there can be high radon emanation rate, resulting in high radon exposure hazards [12].

On the basis of epidemiological studies, it has been established that the enhanced levels of indoor radon in dwellings can cause health hazards and may cause serious deceases like lung cancer in human beings [13]. The epidemiological researches show the relative risk of lung cancer by 1.33 above 200 Bqm^{-3} and the relative risk increases by 0.15–0.2 per 100 Bqm^{-3} [14]. Estimating the radon risk, it is necessary to check the correlation between radium concentration and radon emanation potential in soil samples. In Palestine, a number of studies of radon concentrations and exhalation rates of radon from the soil and building materials are available [8, 15–18].

The present study aimed to measure the radon concentrations in some surface soil samples collected from many sites of Bethlehem Governorate, Palestine and to calculate the radon exhalation rate, radium contents and the effective dose equivalent, in order to detect any harmful radiation effects on the human and establish a data base for surface soil samples which, is used in agriculture and a local market.

Experimental work stages

Study area

Bethlehem is a Palestinian city located in the south central West bank, approximately 10 km south of Jerusalem with a coordinates: 31° 42′ 11″ N, 35° 11′ 44″ E (Fig. 1) (Bethlehem governorate map.com). It is the capital of the Bethlehem Governorate of Palestinian Authority, with an area of 608 Km^2. The economy is primarily a tourist-driven. The total population of Bethlehem Governorate is estimated at 180,000 individuals. Out of the total Palestinian population, 44.8% live in the rural areas and 39% in urban communities, 7.5% in refugee camps. Bethlehem city having 60,000 inhabitants and it stands at an elevation of about 770 m above sea level, 30 m higher than nearby Jerusalem [19]. The region has a Mediterranean climate, with hot and dry summers and cold winters. It receives an average of 700 mm of rainfall annually, 70% between November and January. The average annual temperature is 16.5 °C, and the average annual humidity is about 60.4% [20]. Bethlehem has a Muslim majority, but is also home to one of the largest Palestinian Christian communities. Bethlehem's chief economic is tourism, which peaks during the Christmas season when Christian pilgrims throng to the Church of the Nativity (http://www.bethlehem-city.org).

Samples collection and preparation

Eighty two surface samples of dry soil were collected from different locations in the Bethlehem region in the southern part of the West bank, Palestine as shown in Fig. 1. A 30 cm by 30 cm area was marked at four to six points in each sampling site by grading, depending on the size of the sites. The top layers of the soil which contained wastes that are yet to decompose were removed. Soil samples were collected to a depth of 5 cm using a coring tool that was thoroughly cleaned and dried before each sample was collected. Ultimate care was taken in the extraction of soil sections to avoid mixing or cross contamination of soil samples. About 1 kg of each sample was collected in a plastic bag at the sampling points and the sampling continued for about one week. Soil samples were well mixed after removing exotic materials such as pieces of stones and gravel [12]. The samples first sieved in a mesh sieve, and then dried in a hot air oven at temperature of 110 °C for 12 h to evaporate all moisture content and their bulk densities were determined. The respective net weights of the samples ready for measurement were recorded.

Dosimeters preparation

The can technique was employed for the measurement radon concentration and radon exhalation rates in soil samples from the area under investigations. Radon and its daughters reach an equilibrium concentration after a week or more and thus the equilibrium activity of emergent radon could be obtained from the time of exposure and the geometry of the container. This step was necessary to ensure that the radon gas and its daughters are confined within the sample. The samples were carefully sealed for 60 days in cylindrical containers made from a good kind of plastic with dimensions of 12.5 cm in diameter and 24 cm in depth. In each container, one CR-39 plastic detector was attached below the cork head at a certain distance (about 22.5 cm) from the surface of the material (soil sample). The sensitive part of the detector was faced to the emanating radon of the soil sample so that it could record the alpha particles resulting from the decay of radon in the whole volume of the can. Each sample container was capped tightly to an inverted cylindrical plastic cover as shown in (Fig. 2) [18]. The soil samples were put at the bottom of these containers and size of each sample were about 185 cm^3. The ratio of volumes of the containers and samples was more than 10, which reduces the probability of back diffusion [21].

During the exposure time of α-particles from the decay of radon and their daughters bombard the CR-39 detector in the air volume of the cylindrical containers. After a fixed time, the detectors were taken out, etched (6.25 M NaOH at 70 °C for 4 h) and the detector was washed in distilled water and allowed to air dry. The tracks were counted manually for ten randomly chosen fields of view, using an optical microscope

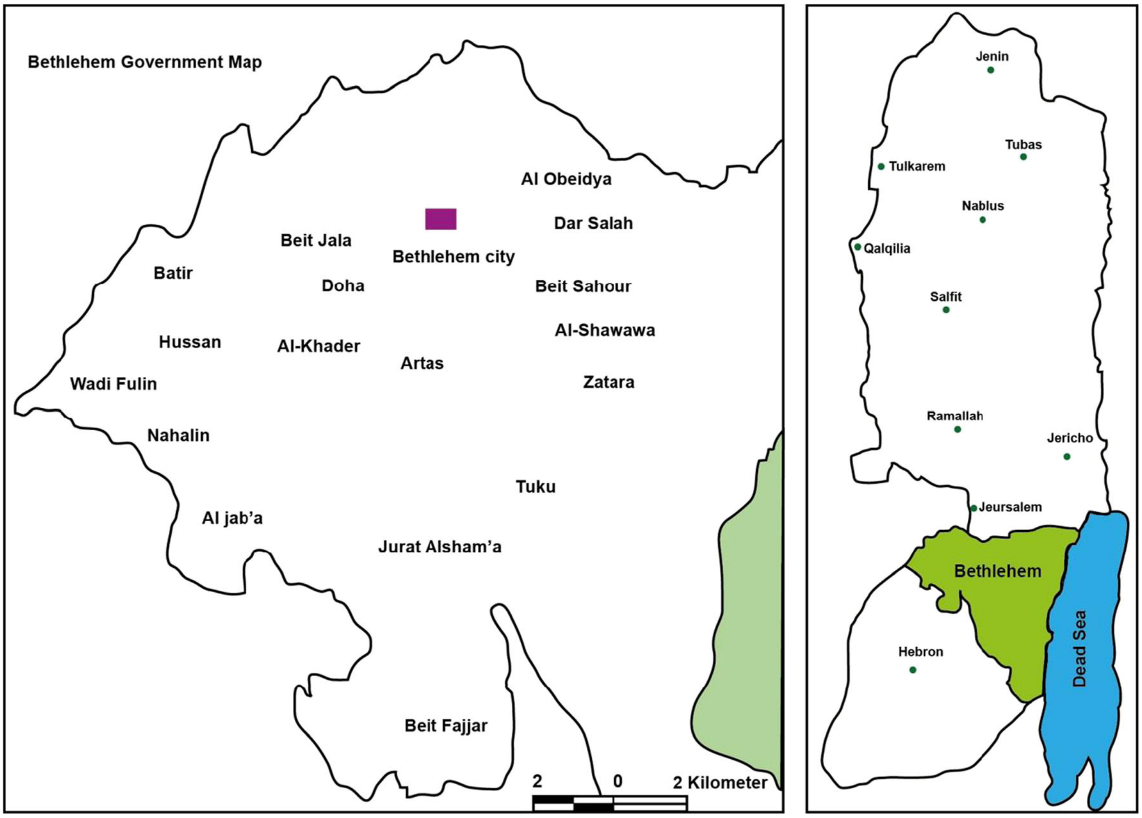

Fig. 1 The map showing the study area (Bethlehem governorate map.com)

with a magnification of 160, to obtain an average and representative value of track density for each dosimeter [8]. The area of the field of view was calculated by the digital microscope and found to be equal about (5.3×10^{-7} cm^2); the average number of tracks per field of view was used to count the track density per m^2.

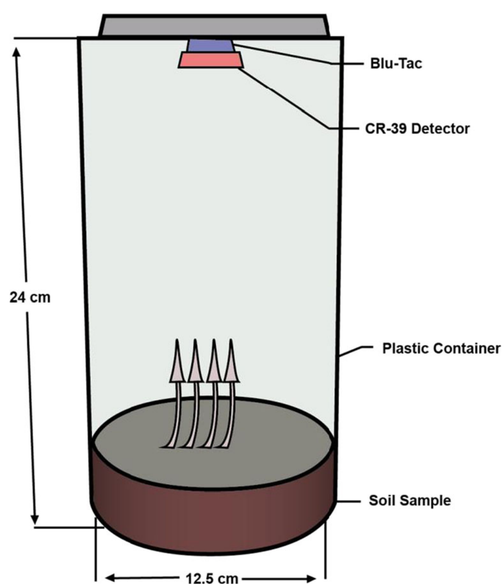

Fig. 2 Experimental set up for the measurement of radon concentration and radon exhalation rate

Results

The radon concentrations

The calculated track density was converted into radon concentrations in Bqm^{-3} using the calibration factor (k) obtained from the manufacturer, where every track per cm^2 per hour on the CR-39 detectors corresponds to an exposure of 12.3 Bqm^{-3} for the activity of radon gas and its daughters. From the measured average track densities (after background subtraction), the radon concentrations were calculated using the measured average track densities according to the following relation [22, 23]:

$$C_{Rn}\left(Bqm^{-3}\right) = K\frac{\rho}{T_{eff}}$$

And

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

Where; C_{Rn}: is the radon concentration (Bqm^{-3}); k: is the calibration factor (Bqm^{-3}/ tracks m^{-2} h^{-1}; ρ: is the track density (tracks / m^2); T_{eff}: is the effective exposure time in hour, τ is the mean life of radon (5.5 days = 132 h), t is the total exposure time and λ is ^{222}Rn decay constant (= 7.56×10^{-3} h^{-1}). This type of correction is needed only for a closed system.

Table 1 Radon concentrations in different soil samples collected from Bethlehem region- Palestine

Zone	No. of samples	C_{Rn} (Bqm^{-3})			Zone	No. of samples	C_{Rn} (Bqm^{-3})		
		Min.	Max.	Av.			Min.	Max.	Av.
Bethlehem City	6	19.1	302.1	126.7	Dar Salah	4	107.7	190.9	149.2
Beit Jala	4	74.2	144.6	109.5	Al Obeidiya	6	40.1	91.4	63.3
Beit Sahour	6	24.1	224.1	106.0	Jurat Alsham'a	4	167.5	243.2	205.2
Doha	4	159.9	478.9	319.2	Beit Fajjar	6	35.4	67.0	47.8
Al- khader	4	522.7	538.4	530.0	Al Jab'a	4	96.1	322.4	209.1
Artas	4	21.3	39.8	30.6	Nahalin	4	37.3	223.2	130.2
Tuku'	4	35.1	45.1	40.3	Wadi Fukin	4	61.0	68.2	64.5
Za'tara	4	61.0	71.4	66.0	Hussan	4	42.6	99.5	71.1
Al- Shawawra	6	56.7	572.9	231.1	Battir	4	52.9	166.8	109.6
Total Samples						82			

Min value: 19.1 Bqm^{-3} , Max value: 572.9 Bqm^{-3} ,Total average value: 145.0 Bqm^{-3}

The calculated track density was converted into radon concentrations in Bqm^{-3} using the calibration factor (k) obtained by the manufacturer, where every track per cm^2 per day on the CR-39 detectors corresponds to an exposure of 12.3 Bqm^{-3} for the activity of radon gas and its daughters [18].

Table 1, shows the values of radon concentration in soil samples collected from different sites in Bethlehem region-Palestine. It is seen that the values of radon concentration in the collected samples vary from 19.1 Bqm^{-3} to 572.9 $Bqm^{-3}$with an average of 145.0 Bqm^{-3}. It is noteworthy from Table 1 that the radon concentration of soil samples is the least in Bethlehem City (191.Bqm^{-3}) but the highest in Al- Shawawra site (572.9 Bqm^{-3}). The soil in this region is late rite that is a fine-grained soil created from weathering of rocks. Such soils contain high concentrations of iron oxides, iron hydroxides and high uranium content [24]. Its high uranium content is reflected by the high radium content; high radon concentration and the soil porosity play an important role in radon exhalation.

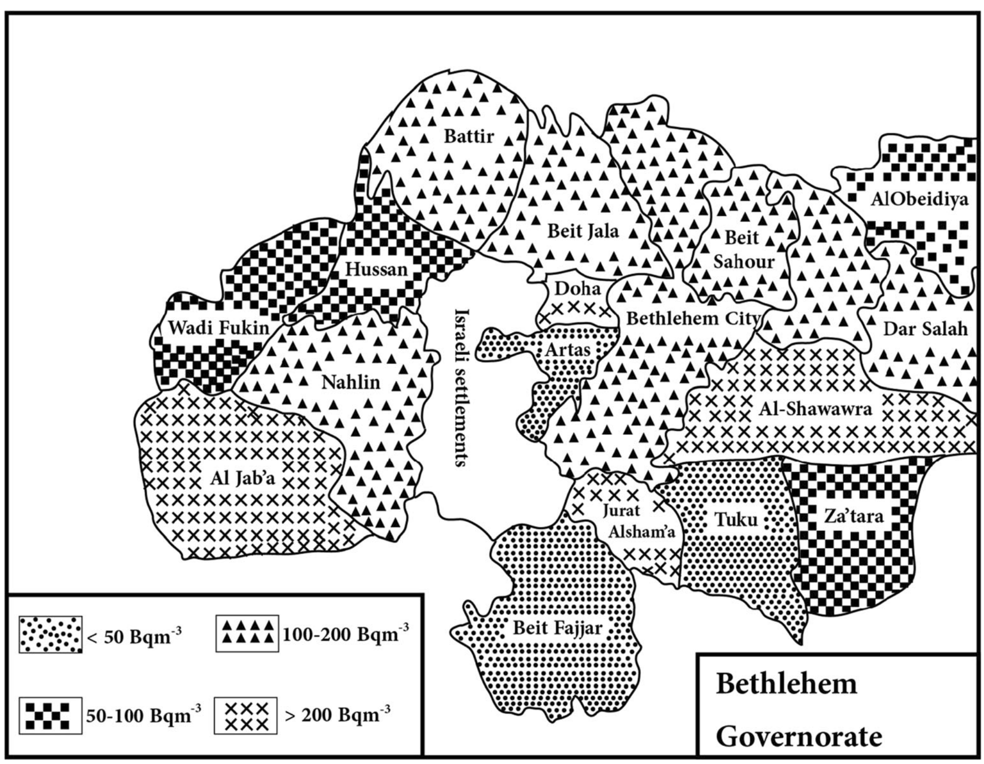

Fig. 3 The geographical distribution of ^{222}Rn concentrations in soil for Bethlehem governorate

The results show that it is possible to map the radon concentrations from soil in Bethlehem region based on the present databases. The distribution based on averaged ^{222}Rn concentrations of each site is plotted in Fig. 3.

The radon exhalation rate

The radon exhalation study is important for understanding the relative contribution of the material to the total radon concentration found in the dwellings. The equation used for surface exhalation rate is written as [25, 26]:

$$E_A\left(Bqm^{-2}h^{-1}\right) = \frac{CV\lambda}{AT_{eff}}$$

And for mass exhalation rate is written as:

$$E_M\left(Bqk^{-1}h^{-1}\right) = \frac{CV\lambda}{MT_{eff}}$$

Where; E_A: is the surface radon exhalation rate, E_M: is the mass radon exhalation rate, C: is the integrated radon exposure in (Bqm^{-3}h), V: is the void volume of the container (m^3), A: is the area of the sample (m^2), M: is the mass of the sample (kg).

Table 2, shows the values of the surface and mass exhalation rates of radon for soil samples collected from different sites in Bethlehem region. The surface exhalation rate in these collected samples varies from 6.9 mBqm^{-2} h^{-1} (Bethlehem city) to 207. 2 mBqm^{-2} h^{-1} (Al- Shawawra site) with a total average value of 52.2 mBqm^{-2} h^{-1}. The mass exhalation rate has been found to vary from 0.26 mBqkg^{-1} h^{-1} to 7.84 mBqkg^{-1} h^{-1} with an average value of 1.97 mBqkg^{-1} h^{-1} .

The effective dose equivalent

The annual exposure to potential alpha energy (effective dose equivalent, E_{eff}) is then related to the average radon concentration by following expression [27, 28]:

$$E_{eff}\left[WLMy^{-1}\right] = \frac{8760 \times F \times C_{Rn}}{170 \times 3700}$$

Where, C_{Rn} is in Bqm^{-3}; n is the fraction of time spent indoors ($n = 0.8$); 8760, the number of hours per year; 170, the number of hours per working month and F is the equilibrium factor for radon and is taken as 0.4 as suggested by UNSCEAR (2000) [29].

Radon progeny equilibrium is most important quantity, where dose calculations are to be made on the basis of the measurement of radon concentration, it may have value $0 < F < 1$. From radon exposure, the effective dose equivalent was estimated by using a conversion factor of 6.3 mSv (WLM)$^{-1}$ by ICRP [30].

Table 3, shows the values of the annual effective dose, E_{eff} of soil samples collected from different sites in the area under investigations. The effective dose equivalent, E_{eff} in these soil samples vary from 0.09 to 2.55 mSvy^{-1} with an average value of 0.65 mSvy^{-1}.

The effective radium content

The effective radium content, C_{Ra} was found from the following relation [18, 23]:

$$C_{Ra}\left(Bqkg^{-1}\right) = \frac{ChA}{MT_{eff}}$$

Where: C_{Ra} is the effective radium content of soil sample (Bqkg^{-1}) and h is the distance between the detector and the top of the soil sample.

The effective radium content, C_{Ra}, of soil samples collected from different sites in Bethlehem region are listed in Table 3. It is clear from the table that the values of effective radium content vary from 1.46 to 44.1 Bqkg^{-1} with an average value of 11.1 Bqkg^{-1}. The average effective radium content in soil

Table 2 Surface exhalation rate, E_A and mass exhalation rate, E_M in soil samples collected from Bethlehem region- Palestine

Zone	E_A (mBqm^{-2} h^{-1})			E_M (mBqkg^{-1} h^{-1})		
	Min.	Max.	Av.	Min.	Max.	Av.
Bethlehem City	6.9	108.7	45.6	0.26	4.11	1.72
Beit Jala	26.7	52.0	39.2	1.01	1.97	1.50
Beit Sahour	8.7	80.6	38.1	0.33	3.05	1.44
Doha	57.5	172.3	115.0	2.18	6.52	4.33
Al-khader	188.0	193.7	190.5	7.11	7.32	7.20
Artas	7.7	14.3	11.0	0.29	0.54	0.42
Tuku′	12.6	16.2	14.4	0.48	0.61	0.55
Za′tara	22.0	25.7	23.9	0.83	0.97	0.90
Al- Shawawra	20.4	207.2	83.4	0.77	7.84	3.16
Dar Salah	38.7	68.7	53.7	1.46	2.60	2.05
Al Obeidiya	14.4	32.9	22.8	0.55	1.24	0.86
Jurat Alsham′a	60.2	87.5	73.9	2.28	3.31	3.00
Beit Fajjar	12.7	24.1	17.2	0.48	0.91	0.65
Al Jab′a	34.6	116.0	75.3	1.31	4.39	2.85
Nahalin	13.4	80.3	46.9	0.51	3.04	1.78
Wadi Fukin	22.0	24.6	23.3	0.83	0.93	0.89
Hussan	15.3	35.8	25.6	0.58	1.35	0.97
Battir	19.0	60.0	39.5	0.72	2.27	1.50
Total	Min. value: 6.9			Min. value: 0.26		
	Max. value: 207.2			Max. value: 7.84		
	Av. value: 52.2			Av. value: 1.97		

Table 3 The effective dose equivalent, E_{eff} and soil ^{226}Racontents,C_{Ra}in some zones of Bethlehem region - Palestine

Zone	E_{eff} (mSvy^{-1})			C_{Ra} (Bqkg^{-1})		
	Min.	Max.	Av.	Min.	Max.	Av.
Bethlehem City	0.09	1.34	0.56	1.5	23.1	9.7
Beit Jala	0.33	0.64	0.49	5.7	11.0	8.4
Beit Sahour	0.11	1.00	0. 47	1.8	17.2	8.1
Doha	0.71	2.13	1.42	12.2	36.7	24.5
Al-khader	2.33	2.40	2.36	40.0	41.2	40.6
Artas	0.10	0.18	0.14	1.6	3.0	2.4
Tuku'	0.16	0.20	0.18	2.7	3.5	3.1
Za'tara	0.27	0.32	0.29	4.7	5.5	5.1
Al- Shawawra	0.25	2.55	1.03	4.3	44.1	17.8
Dar Salah	0.48	0.85	0.66	8.2	14.6	11.4
Al Obeidiya	0.18	0.41	0. 28	3.1	7.0	4.9
Jurat Alsham'a	0.75	1.08	0.91	12.8	18.6	15.7
Beit Fajjar	0.16	0.30	0.21	2.7	5.1	3.7
Al Jab'a	0.43	1.43	0.93	7.4	24. 7	16.0
Nahalin	0.17	0.99	0.58	2.9	17.1	10.0
Wadi Fukin	0.27	0.30	0.29	4. 7	5.2	4.9
Hussan	0.19	0.44	0.32	3.3	7.6	5.5
Battir	0.24	0.74	0.49	4.1	12.8	8.4
Total	Min. value: 0.09			Min. value: 1.5		
	Max. value: 2.55			Max. value: 44.1		
	Total Av. value: 0.65			Total Av. value: 11.1		

samples in the present study is found to be less than the global value 30 Bqkg^{-1} [27].

Figures 3 and 4 shows the correlation between radium concentration with both surface and mass exhalation rates for Bethlehem governorate, there is a linear correlation appeared throughout these figures, this findings is similar to that found in other studies [27, 28, 31] (Fig. 5).

A lot of data have been published regarding radon exhalation rates in open literature. The results of the current study have been compared with the already published data (Table 4). As can be seen from this table, the surface and mass exhalation rates in the study area are lower than those reported in different studies around the world, thus seem to be safe from the health aspects.

Discussion

The measurements indicate higher levels of radon concentration emanated from most soil samples collected from Bethlehem region. The levels are higher in samples collected from the west and south sites compared with other samples collected from east sites. This concentration may be due to higher radium and uranium contents in these samples or

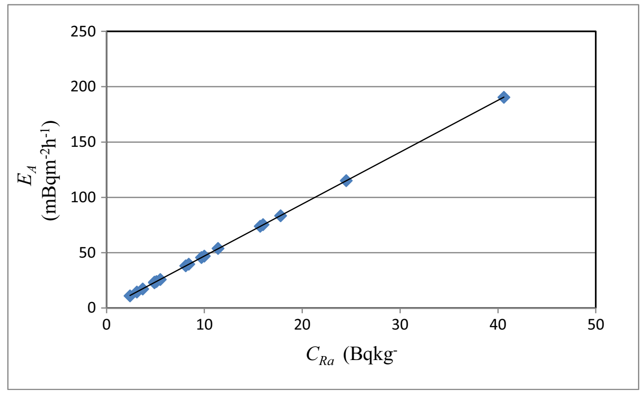

Fig. 4 The correlation between radium concentrations (C_{Ra}) with surface exhalation rate (E_A) for Bethlehem governorate

comes from-NORM which is the main sources of radiation in soils and rocks [32]. Other similar measurements performed by various researchers showed that the soil gas radon concentration may vary over a wide range depending on weather conditions, climate factors and soil type. The obtained results show that the values of radon concentrations of the most samples are below the allowed limit from ICRP. The International Commission on Radiological Protection recommended that a radon concentration from 200 to 600 Bqm^{-3} for dwelling [33]. Again, the radon levels presented above are more than the new reference level (100 Bqm^{-3}) set by WHO [34]. Hence, the result shows that this area is safe as for as the health hazards of radon are concerned.

It is assessed that of the most important parameters, which determine the radon exhalation rate from the soil, is the quantity of radium and the porosity of the soil. It is observed that the radon exhalation rate from soil is bigger than that from building materials [11]. This difference is determined by a bigger porosity of the soil.

In all the locations surveyed, the annual effective dose was less than even the lower limit of the recommended action level (3–10 mSvy^{-1}) reported by the ICRP [33] for the same period of occupancy.

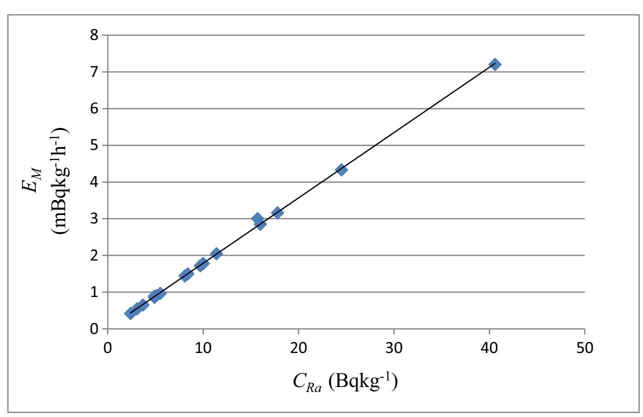

Fig. 5 The correlation between radium concentrations (C_{Ra}) with mass exhalation rate (E_M) for Bethlehem governorate

Table 4 Comparison of present data of radon exhalation rate with the values reported for different countries of the world

Country	E_A (mBqm^{-2} h^{-1})			E_M (mBqkg^{-1} h^{-1})			Reference
	Min.	Max.	Av.	Min.	Max.	Av.	
Egypt (West delta)	290	950	–	–	–	–	[25]
Sudan (El-Hosh)	–	–	4860	–	–	100	[28]
Iraq (Baghdad)	440	990	610	90	210	120	[35]
India (Rushikulya)	140	359	197	5.0	14	8.0	[36]
Pakistan (Dera Ismail Khan)	2210	3570	–	–	–	–	[37]
Saudi Arabia	4580	8400	–	135	251	–	[38]
Palestine (Hebron)	58	153	106	2.2	5.9	4.1	[39]
Palestine (Bethlehem)	6.9	207.2	52.2	0.26	7.84	1.97	Present Study

The variation of the effective radium content in the present study may be attributed to the variation of uranium concentrations according to the region of the soil and the high values in some samples belonging to soil samples nearby radioactive-rich granite, phosphate, sandstone and quartzite. Thus, our results reveal that the area is safe as far as the hazardous health effects of radium are concerned. The soil of this area is advisable for use in brick manufacturing for building construction and in agricultural matters.

Conclusion

Radon concentration levels of the soil samples collected from different sites in Bethlehem region – Palestine, were measured using the sealed can technique. The radon exhalation rates (both the surface and mass exhalation rates); the effective radium content and effective dose equivalent, in these samples were determined to assess the radiological hazards from the Palestinian different materials. The radon concentration levels and the annual effective dose are, on average, within/below the action level recommended by the ICRP (1993) and UNSCEAR (2000).

The radon exhalation study is important for understanding the relative contribution of the material to the total radon concentration found in the dwellings. The results of this study were compared with national and worldwide results.

Except some sites, the present work shows that soil materials do not pose a significant radiation hazard, and thus the use of these materials in the agriculture and construction of these are considered to be safe for the population. Hence it can be concluded that the study area is safe from the health hazard of radon and radium points of view. From this study, there is a strong positive correlation between the indoor radon concentration and the soil radium content. There is also a strong positive correlation between the radium content in soils and the area or mass exhalation rate of radon from soil.

The obtained results can be used as reference information to assess any changes in the radioactive background level due to geological processes in the investigated area.

This study can be used as a baseline for future investigations and the data obtained in this study may be useful for natural radioactivity mapping. It seems necessary to determine the radon concentrations in the soil of other parts of Palestine. The results may also be used as a reference data for monitoring possible radioactivity pollutions in future.

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Availability of data and material Data sharing not applicable to this article as no datasets were generated or analyzed during the current study.

Author contribution KM Thabayneh distributed, collected and read the dosimeters, analyzed, wrote the text, reviewed and approved the final manuscript.

Compliance with ethical standards

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The author declares that they have no competing interests.

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