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Natural of radon gas concentrations in fertilizers and soils in Middle Governorate (Gaza Strip, Palestine)

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التركيزات الطبيعية لغاز الرادون في الأسمدة والتربة بمحافظة الوسطى (قطاع غزة- فلسطين)

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Abstract:

It has become a need and a tool used around the world to apply inorganic fertilizers to treat crop soil and improve its yield. There are varying concentrations of uranium and thorium in these fertilizers. Therefore, due to the high use of fertilizer in agriculture, lots of radionuclides of the natural radioactive series ^{238}U, ^{235}U and ^{232}Th are dispersed into the environment. In this work, the radioactivity levels were determined in five samples of three types of fertilizers (Triple superphosphate-46% (TSP), Nitrogen, Phosphorus, and Potassium (NPK), Potassium sulfate (SOP)) were imported from four countries and were available in local markets. In addition, eleven samples of soils from ten localities in Middle Governorate of Gaza Strip, Palestine were analyzed to determine the radioactivity levels using the sealed-cup technique and CR-39 detectors.

The radioactivity concentration levels in fertilizers were used in agricultural lands for cultivation had the maximum radon concentration 1099 Bq/m^3 and effective radium 8.7 Bq/kg, with exhalation rate 1108 mBq/m^{2}h from the NPK from Jordan. While the minimum radon concentration, effective radium, and exhalation rate values were found to be 304 Bq/m^3, 1.81 Bq/kg, and 308 mBq/m^{2}h respectively in SOP fertilizer made in China .

The soil in the study area were classified into two main textural classes, the sandy loam and loamy sand. In the sandy loam the radon concentration levels in the sandy loam class range from 970.10 to 2295.73 Bq/m^3 and effective radium 6.66 to 17.13 Bq/kg, with exhalation rate 0.98 to 2.315 mBq/m^{2}h, whereas in the loamy sand were 538.25 to 1823.78 Bq/m^3, 3.18 to 11.34 Bq/kg, and 0.545 to 1.84 mBq/m^{2}h respectively. The radioactivity concentration were higher in the sandy loam class due the their finer texture contents (silt and clay) comparing with loamy sand class. Additionally, this study found that the higher concentration levels referred to high contents of organic matter and water contents in the soil samples.

Keywords: radon concentration, pollution, fertilizer, soil, Gaza Strip

الملخص:

استخدام الأسمدة الغير عضوية لمعالجة التربة وتحسين جودة المحاصيل الزراعية بات وسيلة ملحة حول العالم. تختلف تركيزات عنصر اليورانيوم والثوريوم في هذه الأسمدة الزراعية ونظراً للاستخدام المفرط للأسمدة الزراعية في الزراعة ترتفع العناصر الطبيعية لسلسة اليورانيوم 238 والثوريوم 232. في هذا البحث تم تحديد مستويات الاشعاع الطبيعي في خمس عينات لثلاث أنواع من الأسمدة المستوردة (السوبر فوسفات الثلاثي46 % – نيتروجين فوسفات بوتاسيوم - كبريتيد البوتاسيوم) من اربع دول والمتاحة في السوق المحلي. بالإضافة الى تحديد المستوى الإشعاعي لأحدى عشر عينة تربة زراعية من عشر مواقع بمحافظة الوسطي بقطاع غزة فلسطين بتقنية الوعاء المغلق وكاشف. CR-39.

اعلى مستويات تركيز غاز الرادون في الأسمدة المستخدمة في الأراضي الزراعية 1099 بيكرل /م3 والراديوم الفعال 8.7 بيكرل/كيلو جرام ومعدل انطلاق لغاز الرادون 1108 ملي بيكرل/م2 ساعة لعينة نيتروجين فوسفات بوتاسيوم من الأردن في حين ان اقل مستويات تركيز لغاز الرادون والراديوم الفعال و معدل انطلاق غاز الرادون 304 بيكرل /م3 و 1.81 بيكرل/كيلو جرام و 208 ملي بيكرل/م2 ساعة بالترتيب وجدت لعينة كبريتيد البوتاسيوم من الصين.

صنفت عينات التربة الزراعية في مجموعتين رئيسيتين الرملية الطينية والطينية الرملية وفي التربة الطينية الرملية تراوح مستوى التركيز لغاز الرادون من 970.10 الى 2296.73 بيكرل /م3 والراديوم الفعال من 6.66 الى 17.13 بيكرل/كيلوجرام ومعدل انطلاق غاز الرادون من 0.98 الى 2.315 بيكرل/م2 ساعة في حين التربة الرملية الطينة تراوح مستوى لتركيز لغاز الرادون من 538.78 الى 1823.78 بيكرل /م3 والراديوم الفعال من 3.18 الى 211.34 بيكرل/كيلوجرام ومعدل انطلاق غاز الرادون من 0.545 الى 1.84 بيكرل/م2 ساعة. تركيزات الاشعاع في عينات التربية الطينية الرملية هي الاعلى بسبب المحتويات الدقيقة من الطفل والطمي بالإضافة الى ارتباط التركيزات الاشعاعية بالمحتوي العضوي للعينات والمحتوى المائي في عينات التربة.

كلمات مفتاحية: تركيز الرادون, التلوث ,الأسمدة الزراعية, التربة , قطاع غزة .

Introduction:

People are exposed to ionizing radiation from radionuclides found in various forms of natural sources (Ghosh et al., 2008), one of the most important of which is soil, which may thus be exposed to radiation either indirectly from a near source of radiation or internally from contaminated material that has reached the body. Soil not only serves as a source of continuous human exposure to radiation, but also as a means of migration to biological systems to pass radionuclides (Senthilkumar and Narayanaswamy, 2016), which can cause harmful biological effects such as cancer and DNA damage.

At present, reports of health effects related to ionizing radiation have provided considerable evidence that sickness or even death may be caused by exposure to a high dose of radiation. Despite the well-known cancer effect, researchers have long known that high-dose ionizing radiation can also induce mental retardation in children of mothers exposed to radiation during pregnancy (Schnug and Lottermoser, 2013; Rafik et al., 2014).

All food types contain a measurable amount of radioactivity which, through the ingestion pathway, successively relocates into the human body. We all recognize that food activity is strictly related to the activity of the soil where the food is produced. Natural radionuclides occurring in soil and fertilizers consist predominantly of isotopes ^{238}U, ^{232}Th, and their daughter products, as well as ^{40}K.

The absorption and distribution of radionuclides in the soil depends on a range of factors, such as the pH of the soil, the type and quantity of clay, the exchangeable content of Ca and K and organic matter, the physicochemical properties of radionuclides, the nature of crop (crop species and diversity and cultural practices), the application of fertilizers, drainage, plowing, liming and environment conditions (Asaduzzaman et al., 2015).

Relatively high levels of natural radionuclides detected in phosphate fertilizers during cultivation contaminate the atmosphere and farm property (Chauhan et al., 2013; Saueie and Mazzilli, 2006; Sabiha-Javied et al., 2010; Hassan et al., 2016). Therefore, the exposure of employees and the public to phosphate rock and fertilizer radiation is not impossible (Scholten and Timmermans, 1995) because the use of fertilizers, particularly phosphates, greatly increases the amount of radioactive activity of the cultivated soil relative to the soil of the arid soil (Ahmed and El-Arabi, 2005). The high value of radionuclide content in fertilizers, especially phosphates derived from phosphate fertilizers, has been discovered by many researchers (Chauhan et al., 2013; Khater and AL-Sewaidan, 2008; Kumar and Chauhan, 2014), uranium-rich-238 (Asaduzzaman et al., 2015; Fathivand et al., 2014). The radiological influence of the use of soil phosphate fertilizers is due to the internal irradiation of alpha particles into the lungs, the short-lived progeny of radonthoron, and the external irradiation of the body by in situ gamma rays released by radionuclides.

The purposes of this study is to measure the radon gas concentration, exhalation rates and effective radium mass from fertilizer and soil samples using the sealed-cup technique and CR-39 detectors, and to provide useful information in the monitoring of environmental contamination by natural radioactivity, and this is due to the lack of studies concerning the assessment of radium contents and radon exhalation rates. risks. Moreover, as the Gaza Strip depends on seawater desalination for drinking purposes, seawater quality monitoring is an essential routine in seawater desalination procedures. Furthermore, by completing the final stages of the Central Desalination Plant, open intake for feeding the plant will be the only source for desalination. Therefore, regular monitoring of seawater quality will help rapid response in emergence cases and lock down the plant to avoid exposing the desalination systems to polluted seawater and ensure optimal performance.

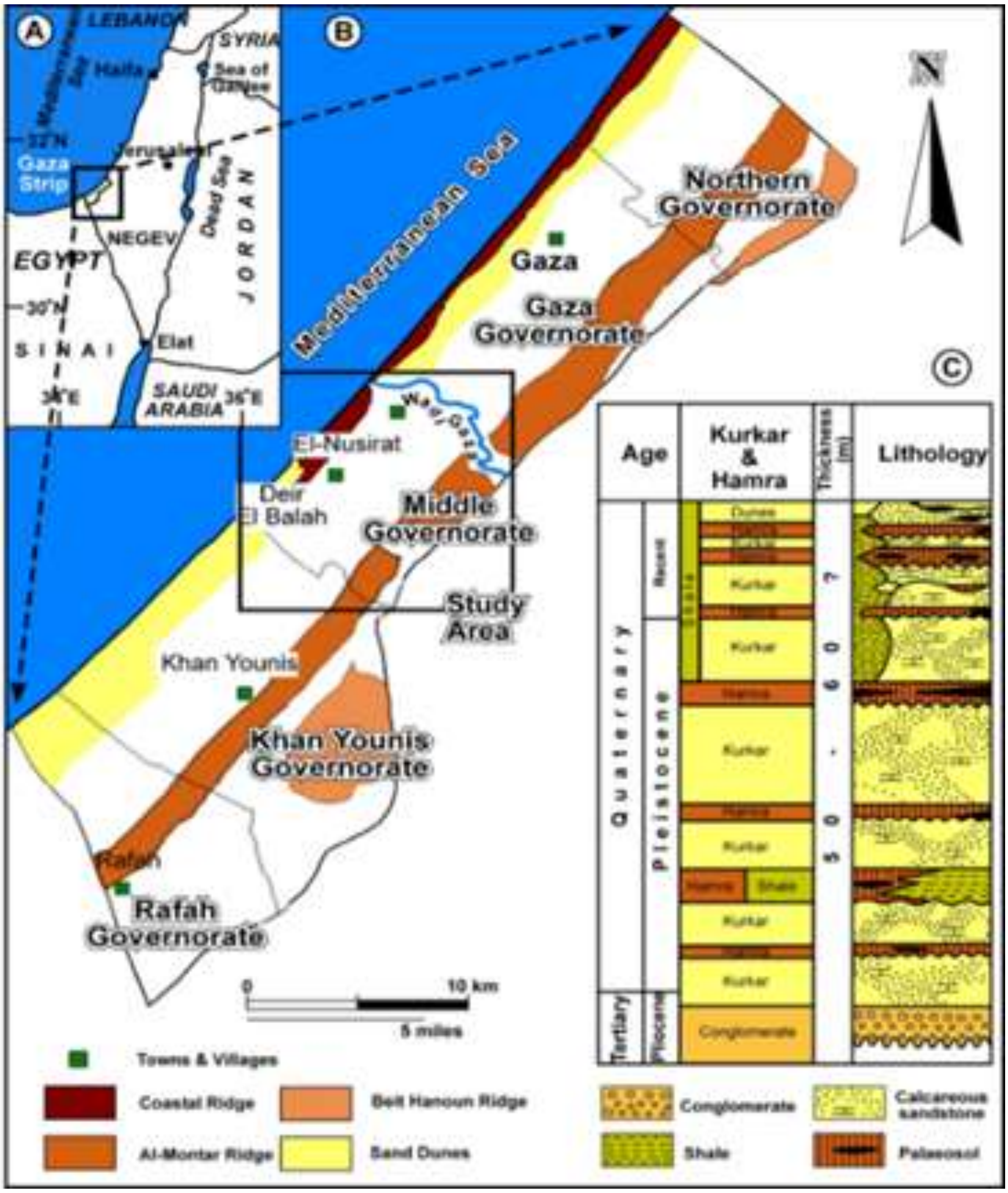
Study area

The Gaza Strip is situated on the southeast coast of the Mediterranean Sea in southwestern Palestine, between 34o 2' East and 31o 45' North (Fig. 1A). It occupies an area of roughly 365 km2, is 45 km long in the coastal plain, and ranges from 5 to 8 km wide in the central and northern regions to 12 km wide in the south. It lies in the transition region between the temperate Mediterranean climate to the west and north, and the arid climate to the east and south of the Sinai Peninsula. Throughout the year, the temperature steadily changed, reaching its highest during the summer in August, and its lowest during the Winter in January. The maximum monthly average temperature ranges from 17.6 Co for January to 29.4 Co for August. The average monthly minimum temperature in January is roughly 9.6 C^{o} and 22.7 C^{o} in August, respectively. The mean annual precipitation is 335 mm per year and the mean annual evaporation is 1300 mm per year. The topography of Gaza is characterized by three ridges (locally called kurkar ridges) and depressions (Fig. 1B). In the NE-SW direction, parallel to the coastline, the ridges and depressions usually extend. The elevation of the surface varies from the mean sea level to about 110 m above the mean sea level. The depressions containing alluvial deposits are about 20-40 m above the mean sea level.

The Gaza Strip is split into five provinces: the Governorate of the North, the Governorate of Gaza, the Governorate of the Middle East, the Governorate of Khan Younis, and the Governorate of Rafah. In the Middle Governorate, the research region is (Fig. 1B). The kurkar ridges belonging to the Gaza Fm are stratigraphically composed of calcareous sandstones (locally called kurkar) alternating with brown reddish fine-grained soils (locally called hamra)

(Fig. 1C) (Abed and Al Weshahy 1999, Ubeid 2010, 2011). Sand dunes appear to mask these elements in the southern part of the Gaza Strip. The Gaza Strip's soil consists primarily of three types: sandy soil, clay soil, and loose soil (Ubeid 2011, 2013). Sandy land, in the shape of sand dunes, was discovered along the coast and in the middle of the Gaza Strip. The artificial soil is located along the northeast side of the Gaza Strip. Around the Wadis, loess soil is found.

Figure 1, Location map of study area.



Methodology

Outdoor sampling

Five soil fertilizer samples were collected from local markets of the Gaza Strip. They represent three types that made in different countries and available for farmers in local markets. In addition to this, around eleven soil samples were collected from ten localities in the Middle Governorate of Gaza Strip, Palestine. The sample locations were identified using landmarks and global positioning satellite (GPS) technology. Table 1 details this. The samples were put in polyethylene bags and tightly closed. The field observations for soil physical properties, time day sampling, temperature, and irrigation times were noted down.

After field work the samples were directly transferred to the laboratory analysis.

Table 1, Location of soil samples in the study area

Sample no.	Location	Coordinates	
		N	E
1	El-Bureij	31º 25` 40``	34º 30` 42.3``
2	El-Bureij	31º 26` 2.0``	34º 24` 35.4``
3	El-Bureij	31º 26` 19.6``	34º 24` 2.2``
4	Wadi Gaza	31º 25` 51.7``	34º 25` 15.0``
5	El-Nusirat	31º 27` 25.8``	34º 22` 31.2``
6	El-Zwaida	31º 26` 35.4``	34º 21` 38.5``
7	Dier El-Balah	31º 23` 46.3``	34º 20` 37.6``
8-1	Dier El-Balah	31º 23` 18.5``	34º 20` 34.4``
8-2	Dier El-Balah	31º 23` 18.5``	34º 20` 34.4``
9	Dier El-Balah	31º 24` 4.1``	34º 22` 27.5``
10	El-Maghazi	31º 24` 48.2``	34º 23` 15.1``

Laboratory analysis

Mechanical and chemical analysis

Mechanical analysis for grain-size distribution, and chemical analysis for water contents (WC), and organic matter contents (OMC) of soil samples were done in laboratories of Geology Department in Al Azhar University – Gaza. Particle size of soil samples were determined using a sieve analysis method and a hydrometer method after dispersion in calgon. The data were processed using GRADISTAT software (Blott and Pye, 2001) to obtain the grain-size distribution. The textural classes of the soil samples due to grain-size distribution results were defined by using USDA soil texture triangle chart (Table 2).

Table 2, Grain-size analysis and textural classification of soil samples in the study area.

Sample no.	Clay (%)	Silt (%)	Sand (%)	Soil texture class
1	2	30.5	67.5	Sandy loam
2	1.5	36	66.5	Sandy loam
3	1	33	66	Sandy loam
4	2	31	67	Sandy loam
5	4	10	86	Loamy sand
6	2	27	71	Loamy sand
7	1.75	25.5	72.75	Loamy sand
8/1	5.25	16.5	78.25	Loamy sand

8/2	2	8	90	Loamy sand
9	3.25	26.25	70.5	Loamy sand
10	3	32	65	Sandy loam

The water contents were determined by drying soil samples in an oven up to 105 C° for 24 hours. The water contents was measured by the weight difference between the wet and dry sample.

Based on ASTM D 2974 – Standard Test Methods for Moisture, Ash, and Organic Matter of Peat and Organic Soils, the organic matter contents in the soil samples were determined. In this method the samples were heated in muffle furnace up to 440 C°. the organic matter was measured by the weight difference between of the sample before and after drying. Table 3 shows the results of chemical analysis.

Table 3, Shows the water contents (WC), organic matter contents (OMC), and radioactivity concentration values of soil samples.

Sample no.	WC (%)	OMC (%)	E (mBq/m^2h)	C_{Rn} (Bq/m^3)	C_{Ra} (Bq/Kg)	AED (mSv/y)
1	12.09	1.61	0.98	970.10	6.66	24.45
2	710.9	1.67	1.78	1767.48	10.96	44.53
3	2.79	2.52	1.86	1842.6	11.99	46.4
4	58.8	1.65	2.20	2178.05	14.57	54.88
5	2.10	1.24	1.13	1121.6	6.47	28.3
6	04.4	70.0	0.545	538.25	3.18	13.575
7	65.5	1.70	1.21	1194.78	7.75	30.13
8	3.83	0.73	0.82	812.38	5.12	20.50
9	11.64	01.6	1.84	1823.78	11.34	45.95
10	7.21	2.61	2.315	2295.73	17.13	57.85

Radon concentration measurement

Fertilizers samples like Potassium sulphate, Super phosphate and three samples of NPK from different countries were available in the local market in Gaza strip. Eleven soil samples were collected from ten different localities in the Middle Governorate in Gaza Strip, Palestine (Fig. 1B). After drying all the samples of the fertilizer and soil in an oven at 100 C° for one day, grinding and sieving through 2 mm, 600 ml

of samples were sealed in plastic containers (3liters, diameter 14 cm) with CR-39 detectors for two months (Fig. 2). The alpha particles emission form the exposed radon gas crash the plastic film of detectors, the CR-39 films were chemically etched using a 6M solution of NaOH at a temperature of 70

C° for about 6 hours. After that, the detectors were washed with distilled and allowed to dry in air. Tracks density (number of tacks per cm^2) on CR-39 with detector sensitivity 12.3 Bq/m^3 per $tracks/cm^2/day$ was calculated by scanning the numbers of tracks in 6 fields for each detector using an optical microscope.

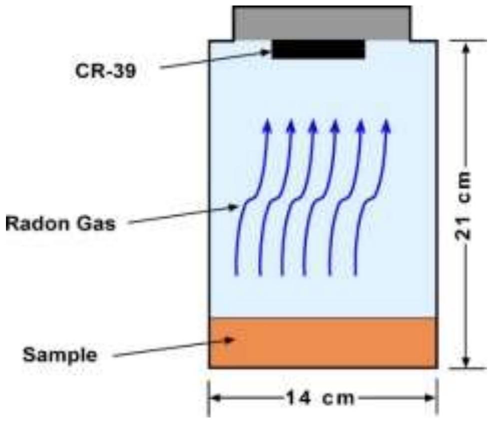


Figure 2, Schematic diagram for the sealed-cup technique used in this study.

Radon concentrations (Bq/m^3) in this study were determined by the formula: $CRn=\rho/(\eta T_{eff})$. The radon exhalation rates (Bqm-2/h) were calculated according to the following formula: $E=(\rho\lambda V)/(\eta A T_{eff})$.

Effective radium content (Bq/kg) were computed by the formula: CRa= (ρ V)/ (η M Teff).

Where, ρ: is track density (tracks/cm2), η: is the detector sensitivity (tracks cm-2h-1/ Bqm-3), λ : is the decay constant (λ=7.56 x 10-3 h-1), V: is the effective volume container (cm3), A: is the area of the sample (cm2), and M: is the sample (kg). Teff=t-τ(1-e-λt), τ: is the mean life of radon=5.5 day, t: is the time of exposure (Baykara and Dogru, 2006; Baykara, et al., 2005; Sroor et al, 2001)

The annual effective dose equivalent was calculated from radon concentrations. According to UNSCEAR (2000), the difference in radon doses and recom¬mended 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 calcu-lated using the relation $AED = \varepsilon f_{Rn} T C_{Rn}$ (Guo and Cheng, 2005).

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 discussion

Soil characteristics

The results of grain size analysis and textural classification of the soil sample in the study area show that the soils in the study area can be subdivided into two main groups, the sandy loam and loamy sand (Table 2; Fig. 3). Relatively, we can called the sandy loam soils characterized be the presence of relatively high percent of fine grains (fine-grained sands, silt and clay). The percentage of fine grains range from 33% to 38%, and sands range from 65% to 68%. While, the loamy sand soils characterized by coarse grains (sands). The percentage of fine grains range from 10% to 30%, and the sands ranges from 71% to 90%. Additionally, the sandy loam soils color ten to be brownish to reddish color. Where, the loamy sand soils tend to be light brown to yellowish color. Most of sandy loam soils were observed in the eastern and northern sides of the study area. Whereas, the loamy sand soils were observed in the western and southern sides of the study area. This means that the fine-grained soils located in eastern side, where the coarse-grained soils located in the western sides. Generally, the light brown and coarse-grained soils developed in sandy parent materials (kurkar). The darker brown and reddish color and fine-grained soils developed on fine-grained sands mixed with silty and clayey aeolian dust (Ubeid, 2011).

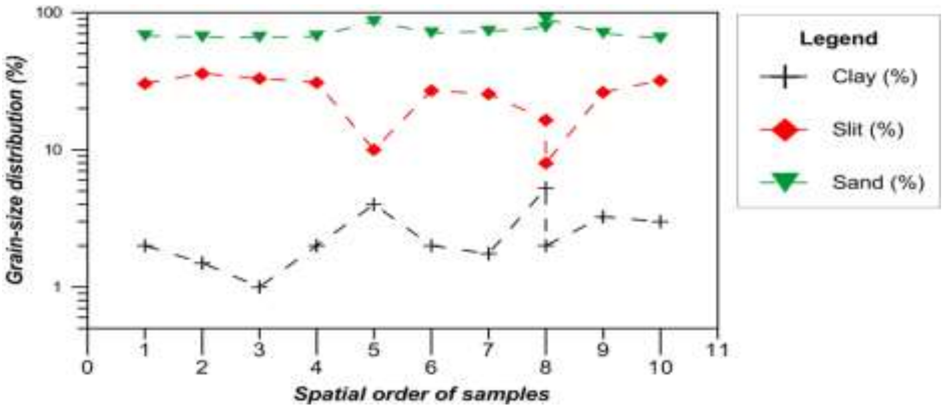


Figure 3, Correlation of grain size distribution of soil samples.

The radioactivity concentrations

The radioactivity concentrations in fertilizers

Our present investigation is based on the study of 5 fertilizer samples which available in the local market. The samples were from different kind (Triple superphosphate (TSP) (46%); Nitrogen, Phosphorus, and Potassium (NPK); Potassium sulfate (SOP)). They were made in different countries (Jordan, Israel, Spain, and China). The radon gas concentration from these samples presented in Table 4 and Figure 4.

It was observed that the NPK from Jordan has maximum radon concentration 1099 Bq/m^3 and effective radium 8.7 Bq/kg, with exhalation rate 1108 mBq/m^{2}h. While the minimum radon concentration, effective radium, and exhalation rate values were found to be 304 Bq/m3, 1.81 Bq/kg, and 308 mBq/m^{2}h respectively in SOP fertilizer made in China.

Table 4, Radioactivity concentration values of fertilizer samples.

Fertilizer Type and origin	Effective Raduim (Bq/kg)	Radon concentration (Bq/m^3)	Dose	Exhalation rate (mBq/m^{2}h)
TSP (Israel)	4.25	556.90	14.00	561.72
NPK (Jordan)	8.70	1099.00	27.70	1108.46
NPK (Israel)	6.28	735.10	18.50	741.47
NPK (Spain)	5.68	683.10	17.20	689.04
SOP (China)	1.81	304.40	7.70	307.07

TSP = Triple superphosphate (46%), NPK = Nitrogen, Phosphorus, and Potassium, SOP = Potassium sulfate

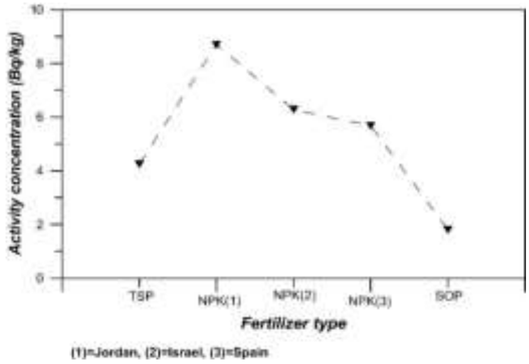


Figure 4, Activity concentration levels of fertilizer samples.

The linear correlation coefficient between radon concentration and effective radium contents (Fig. 5), and between the radon exhalation rate and effective radium concentration (Fig. 6) are equals and was found to be 0.98

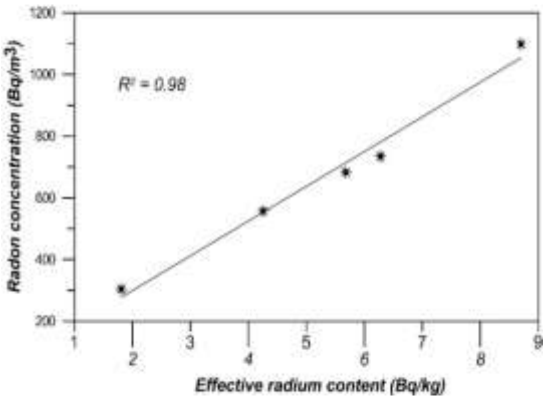


Figure 5, Relationship between effective radium content (CRa) and radon concentration (CRn) for fertilizer samples.

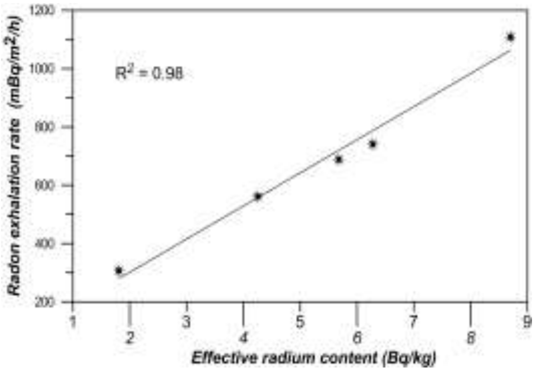


Figure 6, Relationship between effective radium content (CRa) and radon exhalation rate (E) for fertilizer samples.

Overall, the higher radioactivity level were found in NPK fertilizer from Jordan and Israel (Table 4). The Jordanian and Israeli fertilizers are resultant from mining and chemical processing of phosphate sediments particularly from Dead Sea sediments which rich in phosphate and uranium. High possibly, the phosphate sediment not well treated to reduces radioactivity concentration compared with Israeli fertilizer products which depends on the same source. This suggests that, the NPK fertilizer from Jordan has higher level in radioactivity concentration than Israeli NPK fertilizer.

However, the radon concentration in the NPK fertilizers available in the local market of Gaza Strip were above the standard limit (200-600 Bq/m^3) recommended by ICRP (International Commission on Radiological Protection) (1993). Whereas, TSP and SOP fertilizers were below the recommended limit.

The radioactivity concentrations in soils

In Table 3 and Figure 7 we present the radioactivity concentration of the radon gas from soil samples. The results show that the radioactivity values were higher in the sandy loam samples that loamy sand. The radon concentration of the sandy loam samples which mainly located to eastern side of the study area range from 970.10 to 2295.73 Bq/m^3 and effective radium 6.66 to 17.13 Bq/kg, with exhalation rate 0.98 to 2.315 mBq/m^{2}h. While the radon concentration, effective radium, and exhalation rate values in the loamy sand samples in the western side were found to be 538.25 to 1823.78 Bq/m^3, 3.18 to 11.34 Bq/kg, and 0.545 to 1.84 mBq/m^{2}h respectively.

Table 3 presents the results of water and organic matter contents. It was observed the higher values water contents and organic matter range from 2.79% to 12.09%, and 1.61% to 2.62% (except sample no. 9 has high value (11.64%) because it irrigated before two hours of sampling) respectively in sandy loam sample. Whereas, the lower values were observed in loamy sand samples, which found to be 2.10% to 5.56%, and 0.70% to 1.70% (Table 3; Figs. 8 and 9).

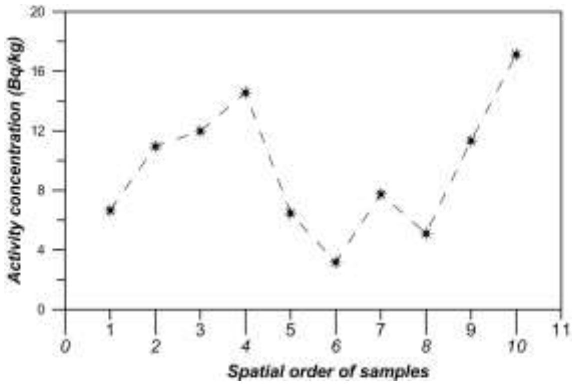


Figure 7, Activity concentration levels of soil samples.

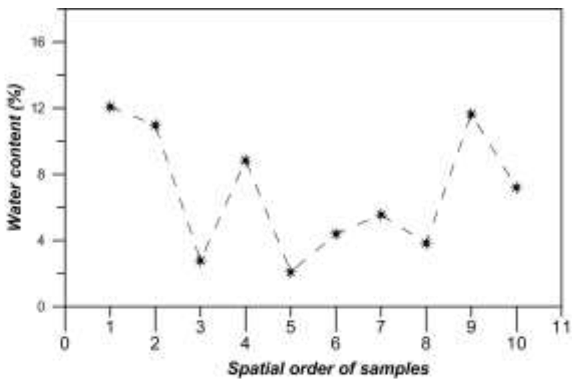


Figure 8, Water contents in soil samples.

The linear correlation coefficient between radon concentration and effective radium;

and between the radon exhalation rate and effective radium concentration were equal and about be 0.97(Figs. 10, 11). The results show the much strong linear correlation coefficient.

Overall. The higher values of radon gas concentration were associated with fine-grained sediments, high level of water and organic matter contents. This suggests the good correlation between the radioactivity levels and fine-grained sediments, water, and organic matter contents. The good correlation referred to large specific area of fine grains which able to adsorb relatively large values water and organic matter contents (Abu Saleh, 2005; El-Ghossain and Abu Saleh, 2007; Balawna and Yassin, 2018, Ubeid and Ramadan, 2020).

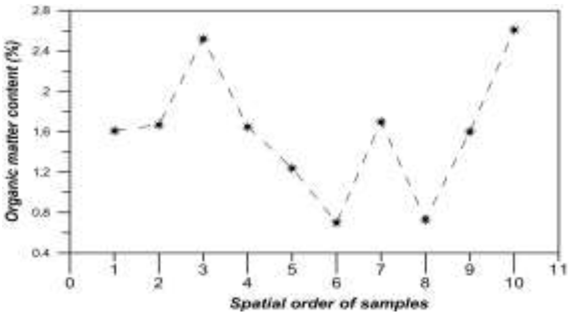


Figure 9, Organic matter contents in soil samples.

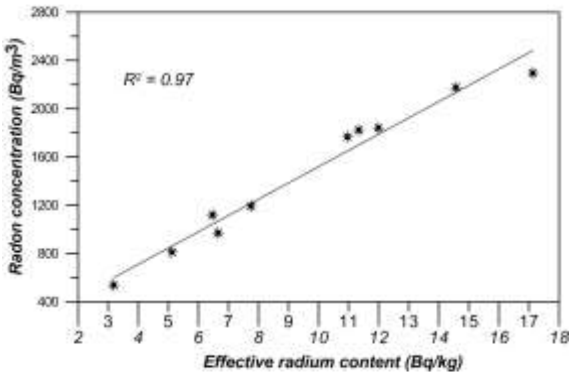


Figure 10, Relationship between effective radium content (CRa) and radon concentration (CRn) for soil samples.

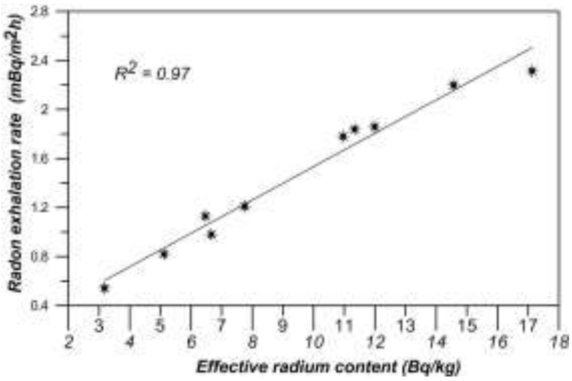


Figure 11, Relationship between effective radium content (CRa) and radon exhalation rate (E) for soil samples.

Unfortunately, the correlation between the radioactivity levels in the soils and fertilizers not be done, due to lack of the data of fertilizers that used for each location of sample. Where, during the collecting samples most of the farmers prevent to give us knowledge about the type and the quantity of fertilizer that were used during the irrigation.

Conclusions

It was found that the maximum radioactivity concentration in the tested sample of fertilizers used in agriculture field in Gaza Strip (TSP, NPK, and SOP) results from NPK which imported from Jordan, while the minimum concentration observed in SOP fertilizer made in China. The radon concentration in NPK fertilizers were above the standard limit (200-600 Bq/m^3) recommended by ICRP (International Commission on Radiological Protection) (1993), whereas, in TSP and SOP fertilizers were below the recommended limit. In the tested soil samples in Middle Governorate in Gaza Strip it was found that the high radioactivity concentrations were found in sandy loam class, which referred to fine-grained texture, organic matter and water contents.

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