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Activity Concentration of ^{222}Rn Gas, ^{226}Ra, ^{232}Th and ^{40}K in Crops and Soil Taken from Safwan Granges Using Active, Passive and Gamma Spectroscopy Techniques

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Authors' contributions

This work was carried out in collaboration between both authors. Both authors read and approved the final manuscript.

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ABSTRACT

It is widely expected that fertilizer has great effect on the soil crops, especially when it contains a radioactive elements. The radioactive content present in crops will increase the number of deferent type of cancer cases in consumers and will explain the importance of this study. In this work we used the passive method (can technique) to measure radon concentration and gamma ray spectrometer for gamma ray concentrations measurement type NaI(Tl) 3"x3". The samples show an average radon concentration of 48.58 Bq/m^3, area radon exhalation rate of 0.091 Bq.m^{-2}.h^{-1} and mass exhalation rate of 0.007 Bq.kg^{-1}.h^{-1}. The average specific gamma activity concentration for ^{226}Ra, ^{232}Th, ^{238}U and ^{40}K were found to be 37.94, 27.88, 13.2 and 388.0 Bq/kg respectively. The gamma hazard indices R_{eq}, H_{ex}, H_{in}, D, E_{out} were found to be 105.58 Bq/kg, 0.285, 0.383, 49.35 nGy/h, 0.06 mSv/y respectively. The concentrations of radionuclides found in this study are nominal and do not pose any health hazard.

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Keywords: Radon; passive; active; radium; gamma spectroscopy.

1. INTRODUCTION

The transfer of metals and radioactive element through the planets to humans is a serious health hazard. It mostly causes lung, stomach, kidney, bladder cancers as well as leukemia [1]. To increase the crop production and to improve soil nutrient properties, farmers used different categories of fertilizers which are essential in this time. Most of the materials contain ^{238}U and ^{232}Th are radon gas emitters, which is a progeny of Radium, which in turn is derived from Uranium decay. The natural radioactivity arises mainly from ^{40}K and nuclide from the ^{238}U and ^{232}Th series and their decay product [2]. A combination of soil and fertilizer is essential for understanding changes in the background radiation [3]. The radionuclide (Uranium, Thorium, Radium and Potassium) can be transferred from soil mixed with fertilizer to roots and accumulate in various part of plant like leaves grain and stem [4,5]. These nuclei have a half- life comparable to the edge of the earth and enhanced 87% of the total radiation dose received, the rest are manmade [6,7]. The fertilized soil is considered as the main source of continuous radiation exposure to human and acts as a medium of migration of radionuclide to the biological system.

The aim of the present work is to investigate the activity concentration of NORM in fertilized soil and crops in Safwan Granges, using radon gas detection and gamma ray spectroscopy.

2. STUDY AREA

The study area falls in Safwan region near Kuwait border as shown in Fig. 1. Safwan is lying just south-western part of Basrah Governorate, southern Al-Zubiar district, is enclosed by salt water on eastern side by Khor Al-Zubiar channel between longitudes 47° 52' and 48° 30' E, and latitude circles 30° 10’ and 29° 50' N, with an area of 24 km^2 south-west, in which sediments are made up alluvial fan and sandy Dibddibba deposits. Dibddibba aquifer has acquired its name from Dibddibba formation which is composed of unconsolidated deposits, i.e. alluvial, often represented by the most common exploited aquifer. Soil of the area comprises essentially of ill-sorted gravely sand, with gypcrete top cover. The size of the gravels varies from coarse gravels (5 - 20 cm), around the apex of the fan to fine gravels and pebbles (2 - 5 cm), in the peripheral parts. The thickness of the gypcrete varies from (0.5 - 1.5) m. Geographically; the surficial fine-grained soil occupies about (50%) of Basrah area. Most cities of Basrah Governorate are located on this type of soil. These soils have a range of depth (7-15) m of depth in the area. The area contains many granges used fertilizers for crops growth [8-10].

Fig. 1. Area of study with samples marking (from Google earth)

3. MATERIALS AND METHODS

3.1 Radon Measurements

3.1.1 Passive technique

Twenty soil and crop samples were collected from different locations in granges of tomatoes in Safwan region near the border of Kuwait. The samples name and number are listed in Table 1.

Table 1. The samples name and number of crops and soil

Sample number	Sample ID
1	Soil-1
2	Tomato-1
3	Soil-2
4	Option Atrosy
5	Soil-3
6	Eggplant
7	Soil-4
8	Tomato-2
9	Soil-5
10	Tomato-3
11	Soil-6
12	Tomato-4
13	Soil-7
14	Okra
15	Soil-8
16	Qrnabit Broccoil
17	Soil-9
18	Tomato-5
19	Soil-10
20	Tomato-6

The can technique was adopted for estimation the radon concentration and exhalation rates for crops and soil samples [11]. The samples were dried in a hot air oven at a temperature 110℃ for 24 h and then crushed and sieved for uniform emanation of radon. Small masses (0.22 kg) were put inside hard plastic containers of 7.5 cm diameter and 30 cm height. In this study we used CR-39 track detectors to measure the track density. The detector 1.5 cm x 1.5 cm areas was fixed on the top inside of the containers using double side adhesive tape. A semi-permeable membrane was fitted in between the detector and sample, which allow only radon to diffuse through. The exposing time was 90 days, and then the detectors were removed and etched with 6.25 N NaOH at 70℃ during 7h. After etching, the detectors were washed in distilled water and then dried in air. The track density, due to alpha particles emitted by samples, was determined by using optical microscope (400X). The radon concentration was calculated using the following expression [12,13];

$$A_{Rn}\left(\frac{Bq}{m^3}\right) = \frac{\rho}{T\,K} \quad (1)$$

Where T is the exposure time in day, ρ is track density in T/cm^2 and K is the calibration factor, which used as K= 0.2857±0.0143 Tr/cm^2.d per Bq/m^3 measured from previous calibration experiment [11].

At the equilibrium state, the radon flux (exhalation) from each sample inside the can is written as [14,15];

$$E_A = \frac{A_{Rn}VT\lambda/S}{T+(e^{-\lambda T}-1)/\lambda} \quad (2)$$

where E_x is exhalation rate in unit Bq.m^{-2}.d^{-1}, A_{Rn} is radon concentration measured by the detector in unit Bq.m^{-3}, λ is radon decay constant, T is the exposure time, V is the volume of the can and S is the surface area of sample in the can.

The radon exhalation rate in terms of mass is calculated from the relation;

$$E_M = \frac{A_{Rn}VT\lambda/M}{T+(e^{-\lambda T}-1)/\lambda} \quad (3)$$

where E_M expressed in Bq.kg^{-1}.h^{-1} and M in kg is the mass of the sample.

The effective radium content calculated from the relation [16];

$$A_{Ra} = \frac{\rho hS}{KMT} \quad (4)$$

Where ρ is track density recorder, h distance between the detector and sample, S surface area of sample, T is exposure time and K is the calibration factor.

3.1.2 Active technique

RAD7 (Durridge Company) is a highly versatile electronic instrument used for radon detection that can measure radon concentrations in real time. For the present purposes the instrument was used to measure radon concentration emanated from soil. The soil sample was loaded into 30 cm x 7.5 cm can used as an emanation cylinder. The cylinder connected online with RAD7 system as shown in Fig. 2. The height of the active volume of the container is 25 cm, to insure ^{222}Rn detection only. The alpha RAD7

detector was operated in grab mode for one day protocol, with cycle 0.5 h and recycle 48. The removable lid was equipped with two gas-tight tubes, one used to pump air contains radon gas in RAD7 chamber (ZnS) and the other used to pump fresh air to the container. The system is a closed loop in which the air circulates continuously. The concentration of radon emanated from each sample inside the emanation chamber was allowed to build up with time and it was measured in 0.5 h cycle for an average time of 24 h. The output data are produced by computer software (Capture 1.2.0) was supplied by DURRIDGE Company.

3.2 Gamma Ray Measurements

Samples of crops and soil were heated in the oven at 110℃ for 24 h to remove moisture, grinded to fine powder, put inside Marinalli beakers and then stored for 30 days to allow the equilibrium between ^{226}Ra and ^{222}Rn. The activity concentration of ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K was estimated from the gamma spectrum using Na(Tl) detector3x3 inch with a 1024 channel computer analyzer USX supplied by Spectrum Technique Company. The detector was employed with lead shielding, 4 cm thickness, which reduced the background. The detector was calibrated using standard sources of ^{57}Co (peak 122 keV), ^{137}Cs (peak 662 keV) and ^{60}Co (peaks 1173 keV, 1333 keV). The detector resolution is about 8% at 662 keV of ^{137}Cs. The efficiency calibration was achieved using eight standard sources include the calibration sources. The system was running freely, for 12 h live time, to evaluate the background spectrum. The Marinalli beaker contains sample was placed over the detector for counting.

After measuring the net count (area under the peak) for each peak, the activity concentration for each environmental isotope calculated from [17];

$$A = \frac{Net\ count}{\varepsilon \times I_\gamma \times M \times t} \quad (5)$$

where ε is absolute gamma peak efficiency of the detector at this particular gamma-ray energy, I_γ decay intensity for the specific energy peak (including the decay branching ratio information), M the mass of the sample in kg and t is the counting time of the measurement in second.

For calculation of the specific activity concentration for each Normal Origin Radioactive Material (NORM), or Technical Enhanced Normal Origin Radioactive Materials (TENORM), one has to recognize the belongcity of each peak according to gamma decay of each isotope. The activity concentration of ^{226}Ra, is calculated from the weighted average concentration of gamma ray lines; 295 keV(19.2%), 352 keV (37.1%), 609 keV (46.1%), 1120 keV (15%) and 1760 keV (15.4%). The peak of 186 keV assumed to be from ^{235}U since it has slight effect on the total concentration after subtracting the background, 42.8% for Ra and the rest for ^{235}U. The determination of existence of ^{232}Th is achieved by the average of lines; 338 keV (12%), 969 keV (17%). The case of ^{238}U is recognized by 1001 keV (83%), 766 keV (29%) and 2204 keV (5%). For ^{40}K, this directly determined using 1460 keV (10%) peak [18]. Sample of environmental gamma ray spectrum in 512 is shown in Fig. 3.

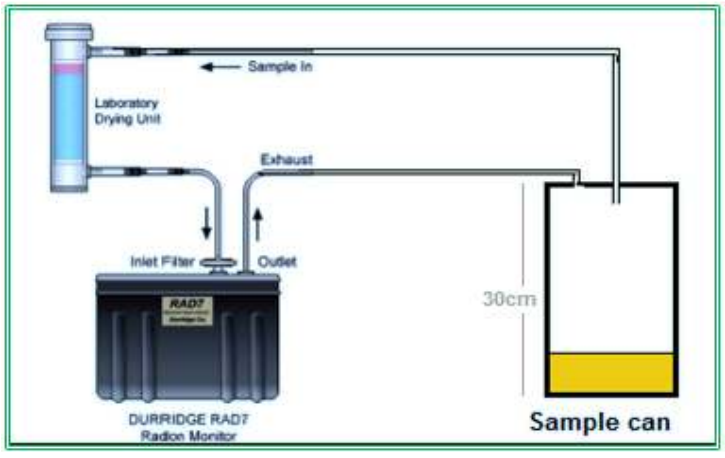

Fig. 2. Block diagram of RAD7 instrument online with sample dosimeter for active radon measurement

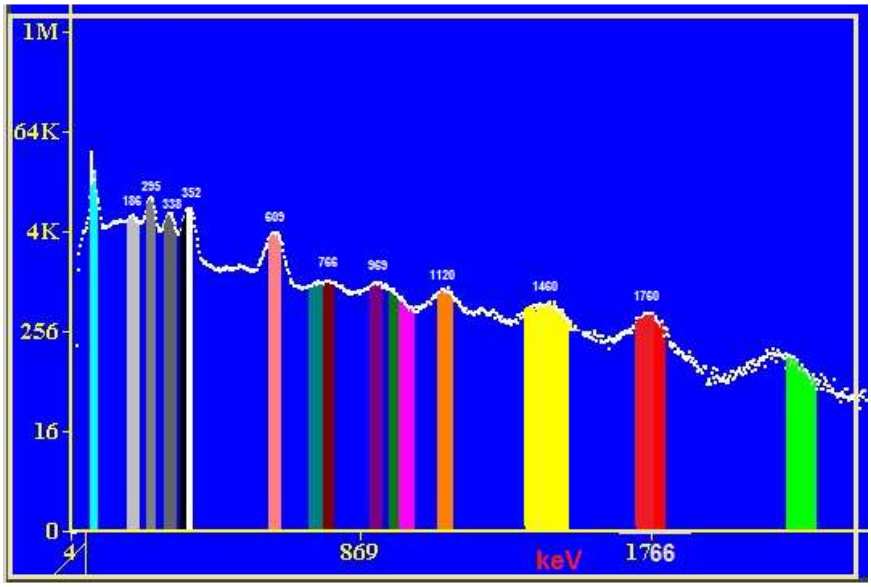

Fig. 3. Gamma ray spectrum of soil sample measured by NaI

Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, which is, determined by assuming that 370 Bq kg^{-1} of ^{226}Ra or 260 Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same γ dose rate. The $Ra_{eq}$of a sample in (Bq kg^{-1}) can be achieved using the following relation [19];

$$Ra_{eq} = (A_{Ra}) + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad (6)$$

The published maximal permissible Ra_{eq} is 370 Bq kg^{-1}.

The external and internal hazard indicesare an evaluation of the hazard of the natural gamma radiation. The prime objective of this index is to limit the radiation dose to the admissible permissible dose equivalent limit around 1mSvy^{-1}. In order to evaluate this index, one can use the following relations [20];

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_k/4810) \quad (7)$$

$$H_{in} = (A_{Ra}/185) + (A_{Th}/259) + (A_k/4810) \quad (8)$$

In order to estimate the annual effective dose rate in air, the conversion coefficient from absorbed dose in air to effective dose received by an adult must be considered. This value is published in UNSCEAR 2000 and UNSCEAR 1993, to be 0.7 SvGy^{-1} for environmental exposure to gamma rays of moderate energy. The outdoor occupancy factor is about 0.2 [18].

The annual effective dose equivalent is given by the following equation [19];

$$AEDE_{out}(mSv/y) = (nGy/h) \times 8760(h/y) \times 0.2 \times 0.7(Sv/Gy) \times 10^{-6} \quad (9)$$

Where

$$D\left(\frac{nGy}{h}\right) = 0.0417A_K + 0.462A_{Ra} + 0.606A_{Th}$$

The world average annual effective dose equivalent (AEDE) from outdoor or indoor terrestrial gamma radiation only is 0.560 mSv/year [UNSCEAR].

4. RESULTS AND DISCUSSION

4.1 Radon Results

Table 2 shows the results of radon concentrations measured by passive (using equation 1) and active technique by RAD7.

Fig. 4 shows the correlation coefficient between passive and active techniques this correlation equal (R^2=0.95), which means that, the correlation is excellent.

The maximum value of radon concentration in Table 2; is137.0±25.0 Bq/m^3 in sample number 11 (Soil-6), measured by RAD7 and minimum value is 18.3±7.4 Bq/m^3 in sample number 10

(Tomatoes-3), measured by CR39. The arithmetic mean value is 48.58 Bq/m^3, which is very low in comparison with the international recommended values. Table 3, presented the radon area exhalation rate, mass exhalation rate and effective mass concentration of radium which decays to produce such parameters by using equations 2, 3 and 4. The results show the range of E_A=0.037 to 0.214 Bq/m^2.h and the average value is 0.091 Bq/m^2. h. These results are consistence with many researches works, for example Chauhan et al. [1] worked on tobacco leaves and found that the average exhalation rate 0.115 Bq/m^2. h and the work of Mahur et al. [21] measured the average exhalation rate from soil as 0.119 Bq/m^2. h. The effective radium in the samples is in the range from 0.098 to 0.57 Bq/kg, with arithmetic average 0.244 Bq/kg.

Table 2. Radon concentrations in samples collected from safwan granges measured by passive and active methods

Sample No	Radon conc. in Bq/m^3 by RAD7	Radon conc. in Bq/m^3 by CR-39
1	94.4±7.0	106.4±35.3
2	28.5±4.2	22.0±8.6
3	60.8±5.9	66.0±22.6
4	29.9±3.6	20.8±8.3
5	45.6±4.7	51.4±17.9
6	47.5±5.2	39.1±14.1
7	46.0±6.2	57.5±19.9
8	21.8±3.6	24.5±9.4
9	37.2±5.2	25.7±9.8
10	36.0±4.8	18.3±7.4
11	137.0±25.0	97.8±32.6
12	38.5±7.2	31.8±11.7
13	38.8±5.4	36.7±13.3
14	22.4±4.2	20.8±8.3
15	52.0±6.2	50.1±17.6
16	22.0±3.8	20.8±8.3
17	54.4±5.4	51.4±17.9
18	34.3±3.7	31.8±11.7
19	54.6±5.9	80.7±27.3
20	25.8±3.4	23.2±9.1

4.2 Gamma Ray Results

Table 4 shows the specific natural activity concentrations of ^{226}Ra, ^{228}Ra, ^{232}Th, ^{238}U and ^{40}K existing in samples collected from selected areas in Safwan granges. For ^{226}Ra, the specific activity ranged from 21.39±6.31 to 68.78±6.31 Bq/kg with an average 37.94±4.92 Bq/kg. For ^{232}Th, the specific activity ranged from 0.07±0.06 to 76.53±5.33 Bq/kg. For ^{238}U, specific activity ranged from 0.04±0.01 to 64.81±17.1 Bq/kg with an average 13.28±3.17 Bq/kg and for ^{40}K it ranged from 43.8±18.0 to 1000±19 Bq/kg with an average 388.3±5.7 Bq/kg. Therefore, the results clearly indicate that all the radionuclides detected were in nominal concentration.

Fig. 5 shows the correlation between total radium concentrations measured by gamma ray spectroscopy and radon measured by CR39 track detectors or exhalation rate. There looks to be strong and positive correlation between them (R=0.98).

Fig. 6 shows the correlation between effective radium concentrations measured by CR-39 track detectors and total radium activity concentrations measured by gamma ray spectroscopy. The correlation looks positively strong, with correlation factor R=0.98.

A detailed analysis of the results in Table 4, indicates that there are no correlations between radioactive elements belong to different series. Figs. 7, 8 and 9 were introduced to check this correlation.

The radium equivalent activity, external and internal hazard index and annual effective dose calculated using equations (6) to (9) are presented in Table 5.

The results revealed that the measured activity levels from natural occurring radioactive in the investigated soil and crops samples are comparable to the corresponding worldwide level. The calculated result of radium equivalent activity ranged from 35.85±6.11 Bq/kg to 196.77±14.50 Bq/kg with a mean value of 105.58±13.60 Bq/kg, and is lower than 370 Bq/kg reported by UNCEAR2000. The average values of H_{ex}=0.282 and H_{in} =0.383 are less than 1 as recommended.

The observed gamma dose rate varies from 16.01 to 94.91 nGy/h with an average value of 49.35 nGy/h. This result is less than the reported value of UNSCEAR2000 [22] which is approximately 57nGy/h. The annual effective dose varies from 0.019 mSv/y to 0.115 mSv/y with the mean value of 0.06 mSv/y. The worldwide average effective dose, according to UNSCEAR2000is 0.07 mSv/y, this means that, our results were in agreement with the worldwide average value. In other word, the fertilized soil and the crops are safe to deal with.

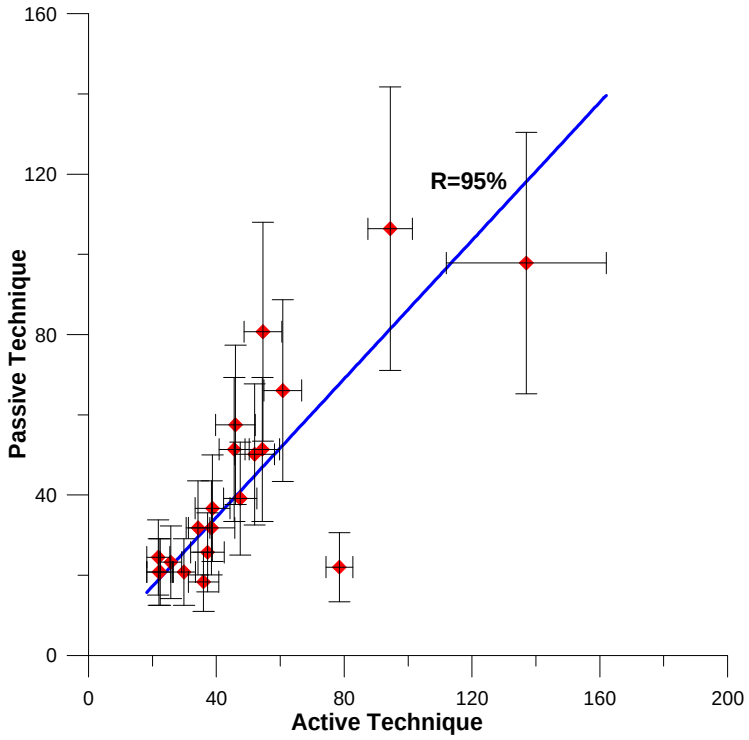

Fig. 4. Correlation between active (using RAD7) and passive (using CR-39) techniques

Table 3. Area exhalation rate, mass exhalation rate and the effective radium mass

Sample no	E_A in Bq.m^{-2}.h^{-1}	E_m in Bq.kg^{-1}.h^{-1}	A_{Ra} in Bq/kg
1	0.214±0.073	0.017±0.006	0.570±0.036
2	0.044±0.018	0.004±0.001	0.118±0.010
3	0.133±0.046	0.011±0.004	0.354±0.024
4	0.042±0.017	0.003±0.001	0.111±0.009
5	0.103±0.037	0.008±0.003	0.275±0.019
6	0.079±0.029	0.006±0.002	0.210±0.015
7	0.115±0.041	0.009±0.003	0.308±0.021
8	0.049±0.019	0.004±0.002	0.131±0.010
9	0.052±0.020	0.004±0.002	0.138±0.011
10	0.037±0.015	0.003±0.001	0.098±0.008
11	0.197±0.067	0.016±0.005	0.524±0.034
12	0.064±0.024	0.005±0.002	0.170±0.013
13	0.074±0.027	0.006±0.002	0.197±0.014
14	0.042±0.017	0.003±0.001	0.111±0.009
15	0.101±0.036	0.008±0.003	0.269±0.019
16	0.042±0.017	0.003±0.001	0.111±0.009
17	0.103±0.037	0.008±0.003	0.275±0.019
18	0.064±0.024	0.005±0.002	0.170±0.013
19	0.162±0.056	0.013±0.004	0.432±0.028
20	0.047±0.019	0.004±0.001	0.124±0.010
Max	0.214±0.073	0.017±0.006	0.570±0.036
Min	0.037±0.015	0.003±0.001	0.098±0.008
Aver.	0.091	0.007	0.244

Table 4. The specific activity concentration fornatural radionuclides

S. N	Ra-226 Bq/kg	Th-232 Bq/kg	Ra-228 Bq/kg	K-40 Bq/kg	U-238 Bq/kg
1	68.78±6.31	40.6±2.94	25.58±1.49	261.2±1.1	0.51±0.0 1
2	28.36±2.86	11.16±0.89	35.68±2.35	699.1±8.2	2.19±0. 09
3	50.49±3.49	45.85±3.29	66.82±3.90	285.0±1.3	0.45±0. 02
4	28.5±3.31	14.96±1.30	33.69±2.47	937.0±17.7	2.93±0. 14
5	39.54±2.71	24.86±2.36	26.10±1.53	272.25±1.2	0.04±0 .02
6	31.72±5.59	61.57±5.22	58.3±28.54	1000 ±19	5.74±0.2 0
7	42.37±2.85	37.28±3.11	41.85±2.44	274.4±1.2	8.56±1. 81
8	29.7±4.43	3.16±1.36	0.06±0.04	879.0±2.0	8.43±0.30
9	23.11±1.63	29.75±2.19	44.89±2.59	239.9±1.0	0.85±0. 01
10	21.39±3.65	0.60±0.88	23.67±1.56	556.0±6.5	10.03±2. 55
11	61.17±4.88	36.52±2.78	39.24±2.79	281.7±1.3	2.29±0. 55
12	30.71±4.74	0.60±0.74	18.90±1.25	58.8±0.7	6.71±1.68
13	31.54±4.89	2.87±0.21	0.02±0.01	43.9±0.2	18.01±4.42
14	25.55±6.73	9.30±0.80	10.91±0.81	229.7±4.4	64.81±17 .10
15	38.45±7.21	30.19±2.13	0.02±0.01	89.1±0.4	39.26±9.6 4
16	27.28±5.92	0.07±0.06	2.31±0.21	110.5±1.5	0.36±0.19
17	41.29±8.86	76.53±5.33	3.11±0.18	44.7±1.8	0.17±0.02
18	33.26±6.56	28.12±2.51	12.08±7.90	215.0±4.1	48.86±1 2.70
19	59.74±7.1	64.49±4.6	39.28±6.03	217.8±2.9	3.66±0.92
20	31.66±4.04	18.38±1.78	51.47±32.81	802.7±15.5	3.38± 0.15
Max.	68.78±6.31	76.53±5.33	66.82±3.28	1000±19	64.81±17. 10
Min.	21.39±3.65	0.07±0.06	0.02±0.01	43.8±18.0	0.04±0.01
Ave.	37.94±4.92	27.88±2.62	27.31±5.96	388.3±5.7	13.28±3 .1

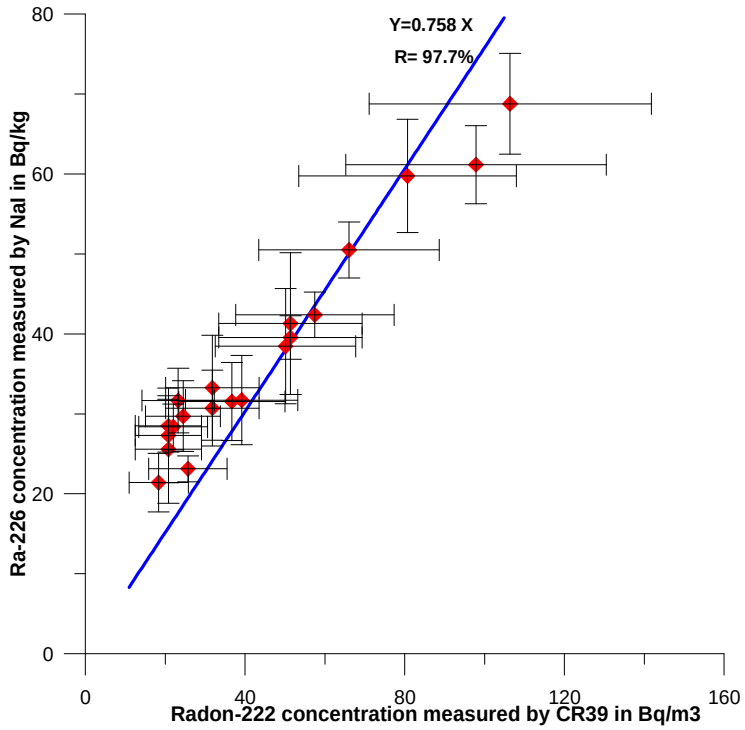

Fig. 5. Correlation between Ra-226 measured by NaI and Rn-222 measured by CR-39

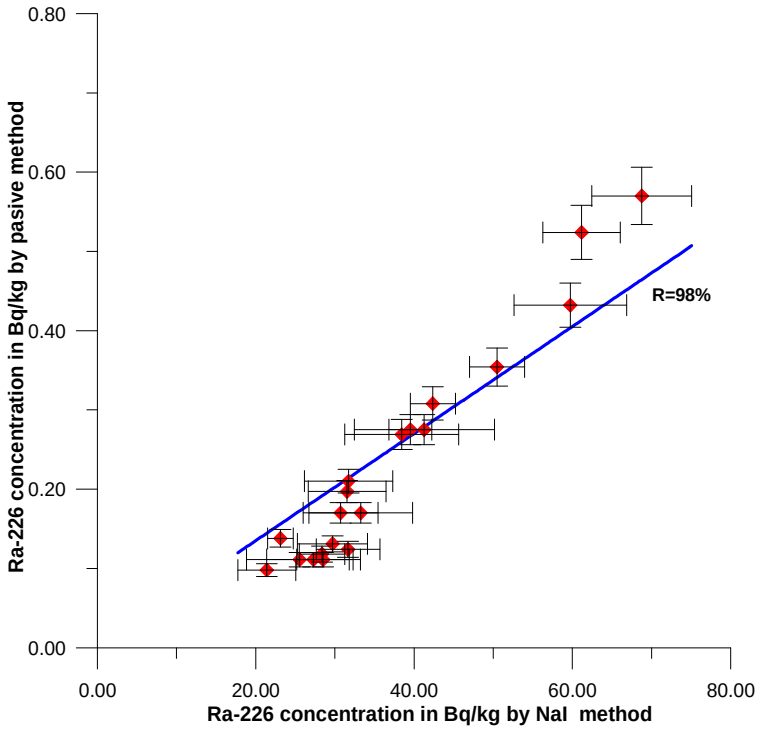

Fig. 6. Correlation between total ^{226}Ra activity concentration measured by NaI and effective ^{226}Ra measured by CR-39 track detector

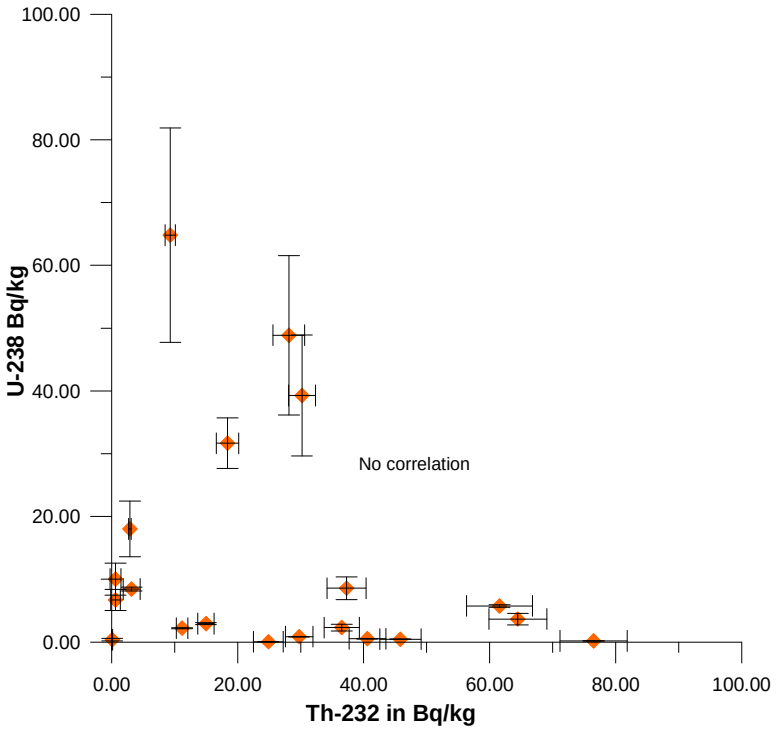

Fig. 7. Correlation between ^{232}Th and ^{238}U activity concentrations in the samples

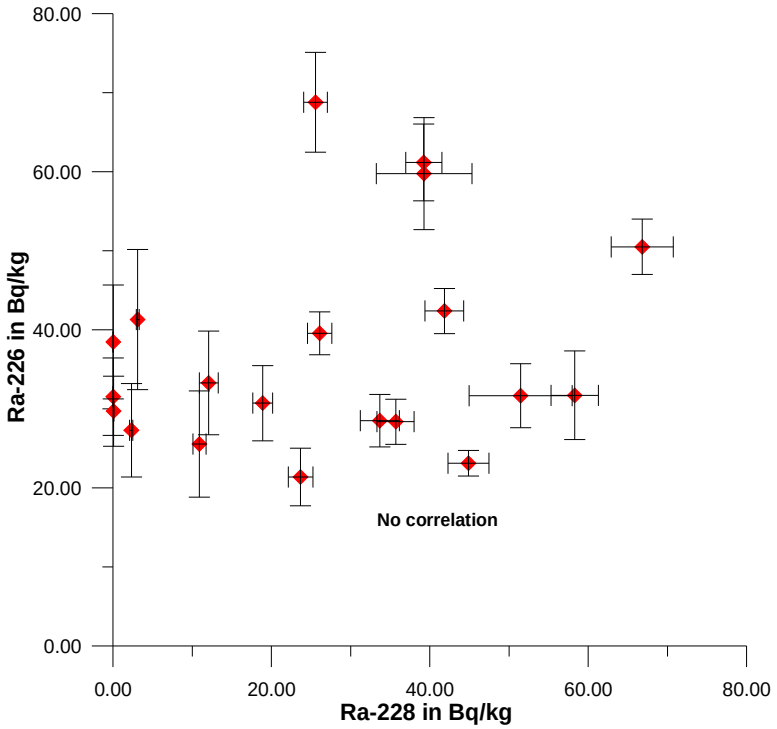

Fig. 8. Correlation between ^{228}Ra and ^{226}Ra activity concentrations in the samples

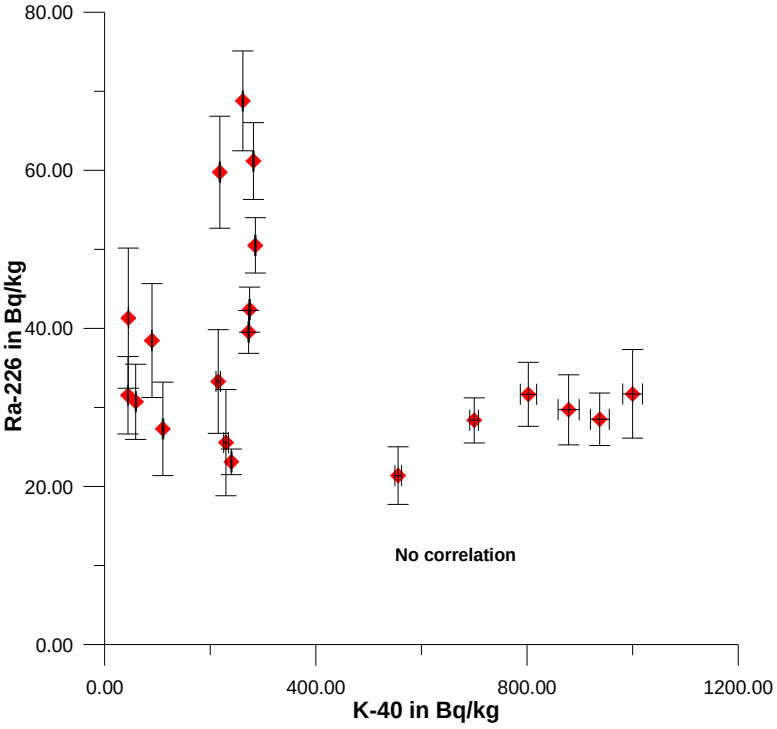

Fig. 9. Correlation between^{226}Ra and ^{40}K activity concentrations in the samples

Table 5. Values of radiation hazard parameters for samples

Sample No	Ra_{eq}Bq/kg	H_{ex}	H_{in}	DnGy/h	$AEDE_{out}$mSv/y
1	146.94±10.61	0.396	0.582	65.892	0.080
2	98.22±4.76	0.265	0.341	49.162	0.060
3	137.99±8.29	0.372	0.509	62.378	0.076
4	122.11±6.52	0.329	0.406	61.822	0.075
5	96.05±6.17	0.259	0.366	44.077	0.053
6	196.77±14.50	0.531	0.617	94.910	0.115
7	116.82±7.39	0.315	0.430	53.120	0.064
8	101.91±7.91	0.275	0.355	52.450	0.064
9	84.12±4.83	0.227	0.289	38.720	0.047
10	65.06±5.40	0.175	0.233	33.410	0.040
11	135.07±8.95	0.364	0.530	60.981	0.074
12	36.08±5.85	0.097	0.180	16.010	0.019
13	39.02±5.19	0.105	0.190	17.144	0.020
14	56.52±8.21	0.152	0.221	26.578	0.032
15	88.48±10.28	0.239	0.342	39.059	0.047
16	35.85±6.11	0.096	0.170	16.425	0.020
17	154.17±16.49	0.416	0.527	67.232	0.082
18	90.02±10.47	0.243	0.333	40.967	0.050
19	168.73±13.75	0.455	0.617	75.052	0.091
20	119.75±7.80	0.323	0.408	59.485	0.072
Max.	196.77±14.50	0.531	0.617	94.91	0.115
Min.	35.85±6.11	0.096	0.17	16.01	0.019
Ave.	105.58±13.6	0.285	0.383	49.35	0.060

5. CONCLUSIONS

The present measurements concentrate on data analysis for radon concentration and gamma ray emission from soil and crops samples collected from Safwan granges near the border of Kuwait. The samples show acceptable radon gas concentration in fertilized soil and crops. Area radon exhalation and mass exhalation have been calculated and show low values. The activity concentration of radium-226, thorium-232 and potassium-40 were measured also for the same samples. The values were below the recommended limit by the ICRP as the maximum annual dose to member of public. The radium equivalent activity and hazard indices are found to be lower than the safe limit. The values of the annual observed effective show that the radioactivity of the natural radionuclides found in the surveyed area is nominal and does not pose any potential health hazard to the farmers and occupiers.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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