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Natural radioactivity assessment in soil samples from Kirkuk city of Iraq using HPGe detector

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▸ ***Original article***

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ABSTRACT

Background: Inspection of the level of radioactivity from radionuclides in soil is important for the assessment of the exposure to natural radiation. ***Materials and Methods***: Ten samples from soil were collected from different sites of Kirkuk-IRAQ and the level of natural radioactivity was measured using gamma-ray spectrometry based on a high purity germanium (HPGe) detector. ***Results:*** It was found that the specific activity ranged from 27.4 to 57.0 Bq kg^{-1} for ^{226}Ra, from 11.0 to 25.4 Bq kg^{-1} for ^{232}Th, and from 207.4 to 516.0 Bq kg^{-1} for ^{40}K. The results have been compared with the average values worldwide. The hazard indices have also been calculated. In general certain average values were found to be lower than the world average values; these included the average value of the radium equivalent activity (Ra_{eq}), the absorbed gamma dose rate (D), the external and internal hazard H_{ex} and H_{in}, the gamma radiation representative level Index (I_γ) and the outdoor and indoor annual effective dose rate (AEDE) ***Conclusion:*** In general there are no harmful radiations effects posed to the population who live in the study area; however there are some spots which have values higher than the internationally allowable values.

Keywords: *Natural radioactivity, soil contamination, activity concentrations, HPGe detector, Radiation hazard indices.*

INTRODUCTION

The soil is a major source for natural radioactivity, and it is the source for the radiation-hazard for the population and a source for migration and transfer of radionuclides into the environment. Therefore soil's natural-radioactivity is considered as a basic indicator for radiological contamination (1). From the radiological point of view, the most important primordial radionuclides are of ^{238}U-series ($t_{1/2} = 4.47 \times 10^9$ years), ^{232}Th-series ($t_{1/2} = 1.41 \times 10^9$ years) and ^{40}K ($t_{1/2} = 1.28 \times 10^9$ years) (2). In general the average annual effective dose for an individual resulting from natural background radiation is estimated to be approximately 2.4 mSv (3).

The isotopes ^{238}U and ^{235}U are the main radioisotopes of the natural uranium in earth crust with abundance 99.28 % and 0.72 %, respectively. ^{232}Th is the only primordial isotope of thorium and its abundance is 100% on earth. The non-series radionuclides such as ^{40}K can be found almost everywhere, including in human and animal tissues, soils and the oceans with various concentrations. The natural, isotopic abundance of ^{40}K on earth is around 0.012 % (4). The emitted nuclear-radiations from naturally occurring radionuclides materials; NORMs is called terrestrial background radiation (5).

Several studies report measured and evaluated values for the level of natural radioactivity in different countries. For example, in Spain (6), in Egypt (7, 8), in Saudi Arabia (9), in Brazil (10), in Cyprus (11), in Turkey (12), in Nigeria (13, 14), in Jordan (15, 16), in Japan (17), in India (18-20),

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in Qatar [21], in Iran [22], and in Iraq [23-25].

Few surveys of natural radioactivity have been conducted in the Kirkuk city of Iraq which is the host of large oil fields. Therefore, the present work aims to determine the specific activity of ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected from selected sites of Kirkuk in order to understand the occurrence and distribution of natural radionuclides in the area under investigation and to evaluate potential health hazards. The radium equivalent activity (Ra_{eq}), the absorbed gamma dose rate (D), the external hazard (H_{ex}), the internal hazard (H_{in}), the Gamma radiation representative level Index (I_γ) and the outdoor and indoor annual effective dose rate (AEDE) were also calculated and compared with the international values.

MATERIALS AND METHODS

A total of ten soil samples were collected from different locations in the city of Kirkuk-Iraq as illustrated in figure 1. Each sample was taken from a depth of 0~45 cm at the chosen point, and the Global Positioning System (GPS) were used for tracking the data recorded. The samples were mixed and sieved with 0.2 mm mesh, then placed in an oven at 100 °C for 24 hours for the purpose of drying the samples. The samples, then, were packed in a 1 kg Marinelli beaker, which were sealed and left for at least 4 weeks to ensure radioactive equilibrium between radon and its decay products [7]. The prepared soil samples filled in a Marinelli beaker and sealed with plastic tape to prevent the escape of airborne radionuclides.

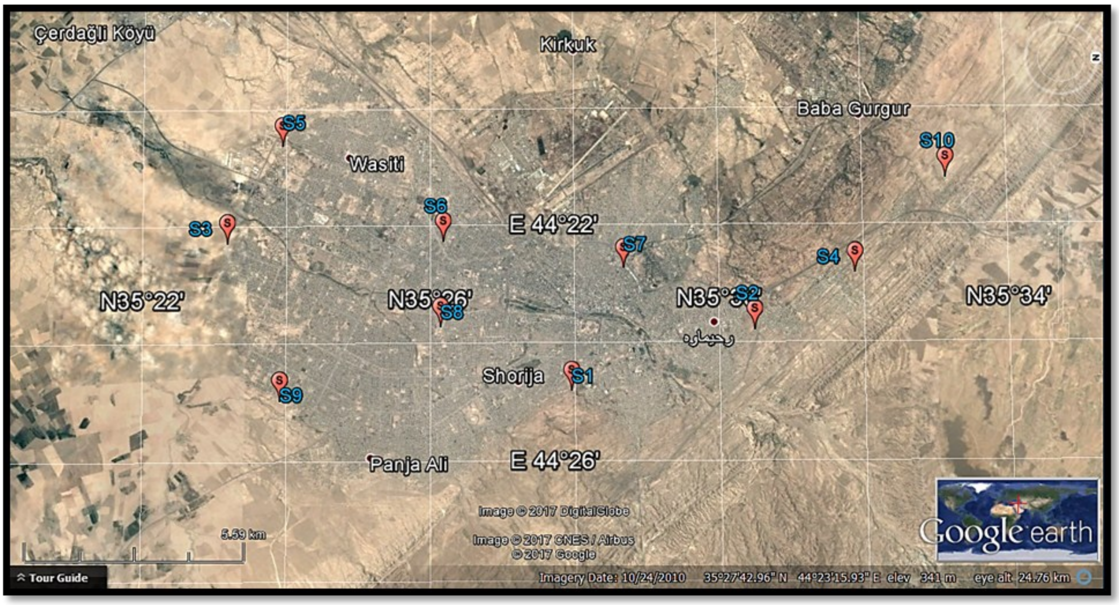

Figure 1. Map shows the locations from where the samples were taken around the city of Kirkuk-Iraq. Google-Earth is appreciated for the facility.

The gamma-ray spectrometry analysis of the samples was carried out using an HPGe coaxial detector of crystal of 50 mm diameter which was operated under a high bias voltage, of +3000V (DC). To reduce the background radiation, a cylindrical lead shield of about 10 cm thick with a fixed bottom and a movable cover shielding the detector was used.

Energy calibration and detection efficiency have been conducted each week to ensure that they were stable during the research period as part of quality control procedures. These two parameters of the detector were carried out by using the mixed radionuclide source (^{241}Am, ^{109}Cd, ^{57}Co, ^{60}Co, and ^{137}Cs); energy (59.5, 88,122, 1173, 1332 and 6616 keV, respectively) of mass 441.0 g, volume 450.0 ± 4.5 cm^3, and density 0.98± 0.01 g cm^{-3}.

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The acquisition time for each sample was 7200s (dead time range between (0.06 and 0.23%). A series of γ-ray energy transition lines ranging from ~100 keV to 2614 keV, associated with the decay products of the ^{238}U and ^{232}Th decay were analyzed independently under the assumption of secular equilibrium of the radionuclides. ^{226}Ra and ^{214}Pb specific activity were used to determine the activity of ^{238}U and the specific activity of the ^{232}Th was determined using gamma-ray transitions lines of ^{228}Ac and ^{212}Pb.

The calculated specific activity of the radionuclides was obtained using equation (1) [26],

$$A(\mathrm{Bq\,kg^{-1}}) = \frac{CPS}{\varepsilon(abs) \times I_\gamma(abs) \times m} \quad (1)$$

where A is the specific activity (Bq kg^{-1}), CPS is the net peak count per second, Ɛ(abs) is the absolute gamma peak detection efficiency, I_γ is the absolute gamma intensity of the corresponding gamma-ray energy considered and m is the mass of the measured sample (kg) [27].

The Radium Equivalent Activity (Ra_{eq}) parameter is widely used as a radiological hazard index. It is a convenient index compared with the specific activities of samples containing different concentrations of ^{226}Ra, ^{232}Th. and ^{40}K. It was calculated using equation (2) [28],

$$Ra_{eq}(\mathrm{Bq\,kg^{-1}}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

Where A_{Ra}, A_{Th} and A_K are the specific activity of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The gamma-ray dose rate (D) in air at 1 m above the ground surface can be calculated using equation (3) [5, 6],

$$D(\mathrm{nGy\,h^{-1}}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (3)$$

In natural environmental radioactivity situations, the effective dose is calculated from the absorbed dose by applying the factor 0.7 Sv Gy^{-1} [2].

The external hazard index (H_{ex}) was determined for all analyzed samples from criterion formula (4) [29, 30],

$$H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \leq 1 \quad (4)$$

Radon and its short-lived products are also hazardous to the respiratory system. The internal exposure to radon and its daughter progenies is quantified by the internal hazard index (H_{in}). It is given by equation (5) [31, 32]:

$$H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \leq 1 \quad (5)$$

The values of H_{in} must be less than unity for the radiation hazard to be negligible.

The gamma radiation representative level index (I_γ) is used to estimate the level of gamma radiation associated with different concentrations of some specific radionuclides, can be defined by equation (6) [33]:

$$I_\gamma = \left(\frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}\right) \leq 1 \quad (6)$$

The I_γ can be used to estimate the level of γ-ray radiation hazard associated with the natural radionuclide in the materials. Its value must be less than unity in order to be safe from radiation hazard.

The average annual effective dose equivalent (AEDE) can be obtained from the average outdoor conversion coefficient from absorbed gamma dose rate (D). The outdoor and indoor AEDE were estimated by equations (7) and (8) [2, 5],

$$AEDE_{out}(\mathrm{mSv\,y^{-1}}) = D(\mathrm{nGy\,h^{-1}}) \times 0.7(\mathrm{Sv\,Gy^{-1}}) \times 0.2 \times 8760(\mathrm{hy^{-1}}) \times 10^{-6} \quad (7)$$

$$AEDE_{in}(\mathrm{mSv\,y^{-1}}) = D(\mathrm{nGy\,h^{-1}}) \times 0.7(\mathrm{Sv\,Gy^{-1}}) \times 0.8 \times 8760(\mathrm{hy^{-1}}) \times 10^{-6} \quad (8)$$

Where 8760 is hours per year. The outdoor and indoor occupancy are 0.2 and 0.8, respectively. The corresponding worldwide value of AEDE is 0.08 mSv [5, 34].

Gamma spectrum analysis of the samples was performed with a computer-based gamma spectrometry system for qualitative and quantitative determination of gamma-emitting radionuclides. For signal processing, a preamplifier and shaping amplifier need a multichannel analyzer (MCA). The data were

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taken directly to the personal computer (PC) to be introduced using Canberra Genie 2000 software which perform data analysis as well as data acquisition.

RESULTS

Results of specific activity of ^{238}U-series, ^{232}Th-series and ^{40}K radionuclides in the investigated ten samples are presented in table-1 and are shown as barcodes in figures 2-4 respectively. For ^{226}Ra the minimum value observed was in sample S9 (27.4 Bq kg^{-1}) and a maximum value was for the sample S1 (57.0 Bq kg^{-1}), with an average of 40.11 Bq kg^{-1}. For ^{232}Th the minimum was 11.0 (Sample S6) and the maximum was 25.4 Bq kg^{-1} (Sample S1) with an average of 15.87 Bq kg^{-1}. For ^{40}K the minimum was 207.4 (Sample S6) and the maximum was 516.0 Bq kg^{-1} (Sample S1) with an average value of 302.82 Bq kg^{-1}. The differences are significant in all samples.

Table 2 shows the range of specific activity and its average value in a number of countries worldwide in comparison with those obtained in this research

The radiological hazard indices obtained in this study are shown in table 3. The value of Ra_{eq} for the samples varies from 19.559 Bq kg^{-1} to 114.256 Bq kg^{-1} with an average value of 81.182 Bq kg^{-1}. The absorbed gamma dose rate varies from 9.462 to 54.560 nGy h^{-1}, with an average value of 38.618 nGy h^{-1}.

The calculated external hazard indexes (H_{ex}) varies from 0.051 to 0.297 with an average value of the 0.210. The calculated average values are less than 1. The internal exposure by radon and its progeny was controlled by the internal hazard index (H_{in}). H_{in} ranges between 0.69 and 0.409 with an average value of the 0.286. The average values are less than 1.

The calculated I_γ values are also presented in Table 3. The values range from 0.147 to 0.850 with an average of 0.603. The calculated values for all samples were lower than the international values (I_γ < 1). The calculated indoor and outdoor AEDE values are also displayed in Table 3. The maximum of outdoor and indoor effective dose were obtained in sample S9: 0.069 and 0.267, respectively.

Table 1. Shows the specific activity of ^{226}Ra, ^{232}Th and ^{40}K in investigated soil samples collected.

Samples	^{226}Ra Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{40}K Bq kg^{-1}
S1	57.0	25.4	516.0
S2	40.4	15.2	353.6
S3	52.0	14.0	250.2
S4	32.1	16.2	314.2
S5	49.6	12.7	260.2
S6	31.6	11.0	207.4
S7	32.8	14.4	251.0
S8	32.0	16.0	261.4
S9	27.4	12.8	283.0
S10	46.2	21.0	331.2
Average	40.11	15.87	302.82

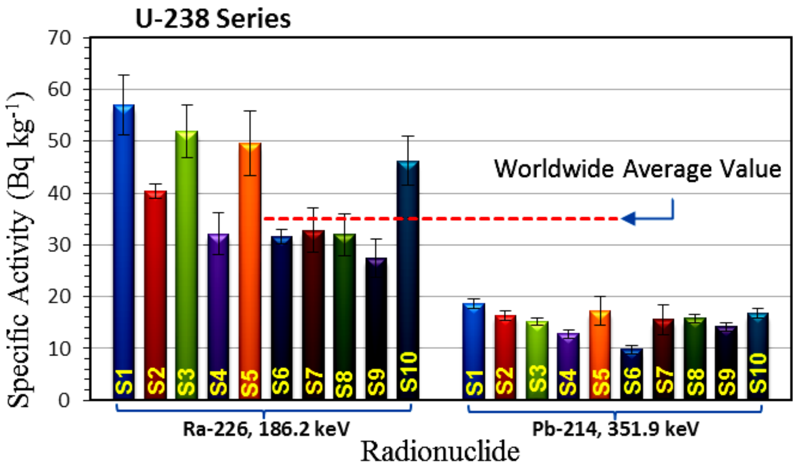

Figure 2. Shows a comparison of the specific activity of ^{238}U-series with the worldwide average value for the investigated soil samples.

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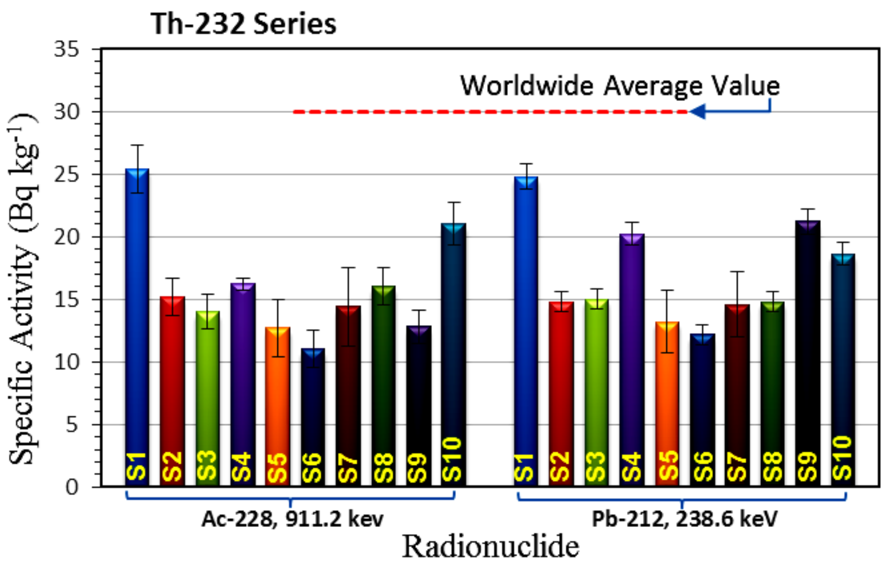

Figure 3. Clarifies a comparison of the specific activity of ^{232}Th-series with the worldwide average value for the investigated soil samples.

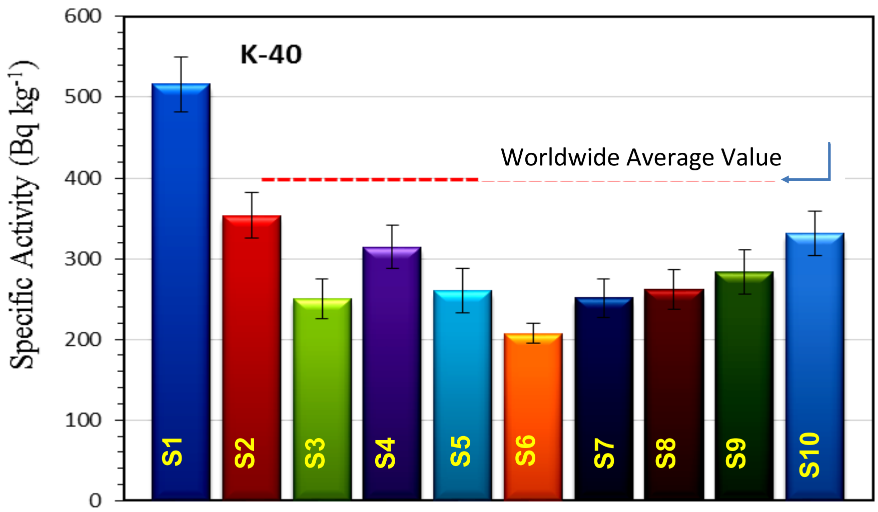

Figure 4. Shows a comparison of the specific activity of ^{40}K with the worldwide average value for the investigated soil sample.

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Table 2. Clarifies a comparison of natural radioactivity levels in the investigated soil samples of Kirkuk-IRAQ with those in other countries.

Country	Specific Activity (Bq kg^{-1})					
	^{226}Ra		^{232}Th		^{40}K	
	Range	Average	Range	Average	Range	Average
Egypt [5]	5-64	17	2-96	18	29-650	320
USA [35]	8-160	40	4-130	35	100-700	370
China [36]	2-440	32	1-360	41	9-1800	440
Japan [37]	6-98	33	2-88	28	15-990	310
Malaysia [5]	38-94	67	63-110	82	170-430	310
India [5]	7-81	29	14-160	64	38-760	400
Iran [5]	8-55	28	5-42	22	250-980	640
Syria [5]	13-32	20	10-32	20	87-780	270
Iraq [23]	16-38	32	8-28	20	261-613	378
Iraq [24]	15-41	35	8-28	19	204-568	289
Denmark [38]	9-26	17	8-30	19	240-610	460
Ireland [39]	10-200	60	3-60	26	40-800	350
Poland [40]	5-120	26	4-77	21	110-970	410
Spain [5]	6-250	33	2-210	33	25-1650	470
Worldwide Average [5]	17-60	35	11-64	30	140-850	400
Kirkuk-Iraq Our Study	27-57	40	11-25	16	207-516	303

Table 3. Radiological hazard indices obtained in this study for the city of Kirkuk-Iraq. Radium equivalent activity (Raeq), the absorbed gamma dose rate (D), the external (Hex) and internal (Hin) hazard index, the radioactivity level index (Iγ) and the annual effective dose annual equivalent (AEDE) for the investigated soil.

Soil Samples	Ra_{eq} (Bq kg^{-1})	D (nGy h^{-1})	H_{ex}	H_{in}	I_{γ}	$AEDE_{out}$ (mSv y^{-1})	$AEDE_{in}$ (mSv y^{-1})
S1	110.954	52.982	0.287	0.382	0.830	0.067	0.259
S2	86.621	41.185	0.225	0.315	0.640	0.052	0.202
S3	73.915	34.814	0.190	0.250	0.547	0.044	0.170
S4	19.559	9.462	0.051	0.069	0.147	0.011	0.046
S5	76.385	36.071	0.198	0.282	0.560	0.045	0.176
S6	87.100	40.999	0.225	0.310	0.640	0.051	0.201
S7	83.462	39.926	0.215	0.273	0.630	0.050	0.195
S8	87.271	42.177	0.227	0.293	0.663	0.053	0.206
S9	114.256	54.560	0.297	0.409	0.850	0.069	0.267
S10	72.303	34.001	0.188	0.275	0.525	0.043	0.166
Average	81.182	38.618	0.210	0.286	0.603	0.048	0.189
Recommended value	370	55	≤1	≤1	≤1	0.08	0.42

DISCUSSION

The results displayed in table 2 shows that the obtained average values fall within the range of corresponding values published for other locations in the world [5, 35-40]. The concentrations have been reported on a dry soil samples. The activity concentration of ^{40}K in soil is an order of magnitude higher than that of ^{226}Ra and ^{232}Th as it is the most abundant radioactive element under consideration. On the basis of the higher levels reported for China and the USA, the Committee revised the values for both ^{232}Th to 40 Bq kg^{-1} in the UNSCEAR 1993

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Report [2]. The results of west Asia (Iran [5], Syria [5] and Iraq [23]) were found lower than the content detected in North America (USA) [35], and East Asia (China [36], Japan [37] and India [5]).

The world average specific activity of ^{226}Ra is 35 Bq kg^{-1} within the range of 17- 60 Bq kg^{-1}, for ^{232}Th is 30 Bq kg^{-1} within the range of 11-64 Bq kg^{-1} and for ^{40}K is 400 Bq kg^{-1} within the range of 140-850 Bq kg^{-1}. In some samples the specific activity for ^{226}Ra and ^{232}Th for the investigated sites were higher than those reported in [5, 41] for radioactivity levels of ^{226}Ra and ^{232}Th internationally. The differences in values are significant. The high recorded values of the radionuclides in some soil samples attributed to the geochemical composition and origin of soil types in a particular area and may be due to the presence of radioactive rich granite, phosphate, sandstone, and quartzite.

Correlation between the radionuclide concentrations has been studied. It was found that ^{232}Th has a weak correlation with ^{226}Ra (correlation coefficient 0.319) (figure 5). However it is seen that ^{40}K and ^{232}Th are positively correlated (correlation coefficient 0.78) (figure 6).

It was inferred that for all the soil samples analyzed, the Ra_{eq} value was well within and less the permissible limits of 370 Bq kg^{-1} [6]. All the obtained D values were lower than the internationally recommended value 55 nGy h^{-1} [5]. The calculated values of H_{ex}, H_{in} and I_{γ} for all the investigated samples were lower than the international values. It can be seen that the indoor and outdoor AEDE values were lower than the corresponding worldwide values of 0.08 and 0.42 mSv, respectively [5].

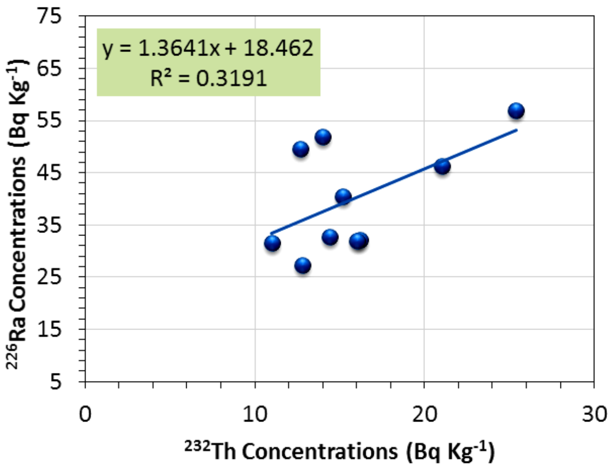

Figure 5. Correlations of activity concentration between ^{226}Ra and ^{232}Th.

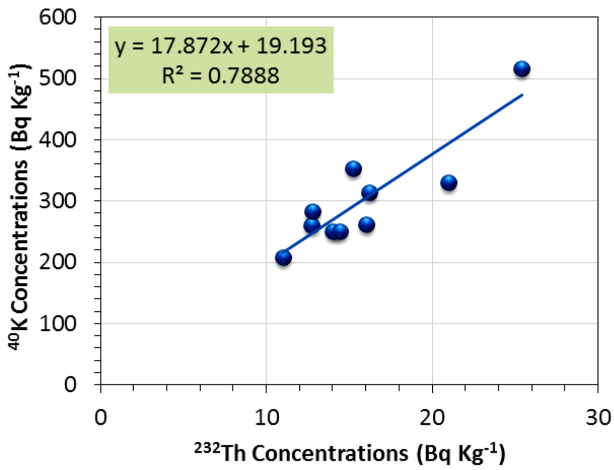

Figure 6. Correlations of activity concentration between ^{40}K and ^{232}Th.

CONCLUSION

Most of the specific activity values were lower than the world average values with some exceptions which are given in figures and tables of this study, so we should focus on these sites by collecting more samples and greater depths to give a more accurate description of the area. The high recorded values in some samples may be due to the geochemical composition and origin of soil types. The averages values of the obtained radiological effects such as: the radium equivalent activity (Ra_{eq}), the absorbed gamma dose rate (D), the external (H_{ex}) and the internal (H_{in}) hazard indices, the radioactivity level index (I_{γ}) and the annual effective dose equivalent (AEDE) were within the limit of the internationally recommended values. It was concluded that no harmful radiation effects were posed to the population who live in the study area.

Conflicts of interest: *Declared none.*

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