Estimating the natural and artificial radioactivity in soil samples from some oil sites in Kirkuk-Iraq using high resolution gamma rays spectrometry

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The natural and artificial radioactivity in soil samples from some oil sites of Kirkuk-Iraq have been estimated using a gamma spectrometry based on a high purity germanium (HPGe) detector. For this reason, soil samples have been collected from four sites; Henjera, Jabel Boor, Jambor and Qutan. It was found that the specific activity ranged from 7.31 to 63.33 Bq kg⁻¹ for 226 Ra, from 3.54 to 42.95 Bq kg⁻¹ for 232 Th, from 103.21 to 798.52 Bq kg⁻¹ for ⁴⁰ The results have been compared with the worldwide average values. The radium equivalent activity (Ra_{eq}) , the absorbed gamma dose rate (*D*), the annual effective dose rate (AEDE), the external hazard (H_{ex}), the internal hazard (H_{in}) and Gamma radiation representative level Index (I_Y) have also been calculated. The Ra_{eq} was 92.173 Bq kg⁻¹, the *D* was 45.53 nGyh⁻¹, the AEDE outdoor and indoor were 0.0959 and 0.224, respectively, the *H*ex was 0.242, the *H*in was 0.329, and *I*γ was 0.680. It has been concluded that no harmful radiation effects have posed to the population who live in the study area. Although, there are some areas where radiation effects have resulted higher than that of the international allowable radiation values.

Keywords: Natural radioactivity, Radioactive decay, Radiation detectors

1 Introduction

Natural radioactivity is ionizes radiations and has great contributions to the world population due to its presences in surrounding at a different amount because of natural presences. Soil is one of the major sources of radiation exposure to the population and of migration for the transfer of radionuclide into the environment; hence it is the basic indicator of radiological contamination¹. From a radiological point of view, the most important primordial radio nuclides are of 238Useries(half-life 4.47×10^9 years), 232 Th-series² (half-life 1.41×10^{10} years) and ⁴⁰K (half-life 1.28×10^{9} years). The average annual effective dose³ to an individual resulting from natural background radiation is approximately 2.4 mSv.

Natural uranium in earth crust is found in the form of two main radioisotopes 238U and 235U whose current natural abundance is 99.28% and 0.72%, respectively. 232 Th is the only primordial isotope for thorium and it represents 100% on earth. The non-series radionuclides such as $40K$ can be found everywhere, including in human and animal tissues, soils and the oceans contain various concentrations. The natural, isotopic abundance of K-40 on earth is around⁴ 0.012 %.

The natural radioactivity concentrations in soil determine both natural and man-made sources which are important in radiological monitoring and radiation dose assessment for public. There have been many surveys conducted to determine the background levels of radionuclides in soils, which can in turn be related to the absorbed gamma dose rates in air. All of these spectrometric measurements indicate that the three components of the external radiation field, namely from the gamma-emitting radionuclides in the 238 U and 232 Th series and 40 K, make approximately equal contributions to the externally incident gamma radiation dose to individuals in typical situations both outdoors and indoors⁵.

The present work aims to determine the specific activity of 2^{26} Ra, 2^{32} Th, 40 K and 1^{37} Cs in the soil samples collected from an oil sites in Kirkuk-Iraq in order to understand the occurrence and distribution of natural and artificial radionuclides of soil samples in the area under investigation and to evaluate potential health hazards. The radium equivalent activity (*Ra*eq), the absorbed gamma dose rate (*D*), the annual effective dose rate (AEDE), the external hazard (H_{ex}) , the internal hazard (*H*in) and Gamma radiation representative level index (I_y) were also calculated and compared with the international recommended values

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for all soil samples to assess the contribution of this radionuclide to public exposure.

2 Collections and Preparation of Samples

Soil samples were collected from four sites in the oil city Kirkuk-Iraq; Henjera, Jabel Boor, Jambor and Qutanas distributed as in Fig. 1. Each sample was taken to a depth of 0~45 cm at a chosen point, data from 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 for drying samples at 100 \degree C for 24 h. The samples were packed in a 1 kg polypropylene bottle, which were sealed and left for at least 4 weeks to ensure that radioactive equilibrium between radon and its decay products⁶. Prepared soil samples were filled in a Marinelli beaker and sealed with plastic tape to prevent the escape of airborne radionuclides.

3 Experimental Setup

The gamma ray spectrometry analysis of the samples was carried out using HPGe coaxial detector of Crystal of 50 mm diameter which was operated under a high voltage, bias of +2000 V (DC). To reduce the external gamma-ray background in the measured spectrum, a cylindrical lead shield of about 10 cm thickness with a fixed bottom and a movable cover shielded the detector was used. The view of the HPGe detector and shielding system are shown in Fig. 2. Shielding of the germanium detector was very

important for counting low-level environmental samples because it decreases the amount of background radiation which reaches the detector.

Gamma spectrometry 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 selected shaping time was 6 μs. The applied course gain and fine gain were 20 and 1.00, respectively. Finally the result and spectral data were taken directly to the personal computer (PC) to be introduced using Canberra Genie software (Canberra Industries, 2000). The configuration of HPGe detector is shown in Fig. 3.

Fig. 2 — Top view of the HPGe detector and shielding system.

Fig. 1 — Map of the investigated area showing sampling points of selected sites.

Fig. 3 — High-purity germanium detector system used in this work.

Energy calibration and detection efficiency have been conducted each week to assure 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 $(^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg,$ ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y and ^{60}Co) of mass 980 g, volume 1000 ± 10.0 cm³ and density 0.985 ± 0.01 gcm⁻³.

The acquisition time for each sample placed directly over the front of the detector was 7200 s (dead time range between 0.06 and 0.23%). The counting geometry of the samples and the standard sources used for efficiency calibration were kept constant. A range of different gamma-ray energy transition lines ranging from \sim 100 keV to 2614 keV, associated with the decay products of the 238 U and 232 Th decay series were analyzed independently. The data were analyzed under the assumption of secular equilibrium of the radionuclides within these samples. 226Ra, 214Pb and 214 Bi specific activity were used to determine the activity of $238\overline{U}$. The specific activity of the $232\overline{T}$ h was determined using gamma-rays transitions lines of ²²⁸Ac, ²¹²Pb and 208 Ti. Gamma-rays peaks of 40 K and 137 Cs were determined at 1461 and 662 KeV, respectively. Background contributions were subtracted from the peak areas for the measured samples.

4 Calculations

4.1 The specific activity

Specific activity of the radionuclides in the measured samples was obtained using the following relation⁷:

$$
A = \frac{CPS}{\varepsilon(\text{abs}) \times I_{\gamma}(\text{abs}) \times m} \quad \dots (1)
$$

where, A is the specific activity $(Bqkg^{-1})$, CPS is the net peak count per second, ε (abs) is the absolute gamma peak detection efficiency, I_{ν} (abs) is the absolute gamma intensity of the corresponding gamma ray energy considered and *m* is the mass of the measured sample⁸ (kg).

The specific activity of ²³⁸U, ²³²Th, and ⁴⁰K was determined using gamma spectrometry technique for soil sample collected from four oil sites in Kirkuk-Iraq. The specific activity of the samples under investigation in Bqkg-1 was determined from the photo-peaks of the gamma spectra corresponding to ²³⁸U, ²³²Th, and ⁴⁰K. Estimation of the count rates for each detected photopeak and radiological concentrations of the detected radionuclides depends on the establishment of secular equilibrium in the samples. Since secular equilibrium reached between 232Th and 238U and their decay products, the 232Th concentration was determined from the average concentrations of $(^{208}Tl, ^{212}Pb, ^{228}Ac)$ in the samples, and that of 238 U (226 Ra) was determined from the average concentrations of ^{214}Pb and ^{214}Bi decay products. Thus, an accurate radionuclide concentration of 232 Th and 238 U was determined, whereas a true measurement of ^{40}K (1460) concentrations was computed.

4.2 Radium equivalent activity

Radium equivalent activity is widely used as a radiological hazard index. It is a convenient index to compare the specific activities of samples containing different concentrations of 226 Ra, 232 Th. and 40 K. It was calculated as follows:

$$
Ra_{\text{eq}} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \qquad \qquad \dots (2)
$$

where A_{Ra} , A_{Th} and A_{K} are the specific activity of ²²⁶Ra, ²³²Th and ⁴⁰K in Bqkg⁻¹, respectively⁹.

4.3 Absorbed gamma dose rate

External exposure to radiation arising from naturally occurring radionuclides can be determined in terms of the absorbed gamma dose rate in air at 1 m above the ground surface. The mean specific activity of ^{238}U , 232 Th and 40 K (in Bqkg⁻¹) in soil samples was used, applying the conversion factor given $by^{5,10}$ as follows:

$$
D = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \qquad \qquad ...(3)
$$

where *D* is the absorbed gamma dose rate in $nGyh^{-1}$. In natural environmental radioactivity situations, the effective dose is calculated from the absorbed dose² by applying the factor 0.7 Sv Gy^{-1} .

4.4 External hazard index

External hazard index is used to evaluate a potential hazard which is associated with non- radiological and radiological effects. It is calculated from 226 Ra, 232 Th, and 40 K to minimize external gamma radiation dose to 1 mSvy $^{-1}$. The following equation is used to define external hazard index:

$$
H_{\text{ex}} = \left(\frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810}\right) \le 1 \tag{4}
$$

The unity for radiation hazard should be negligible compared to the index value which is less. The external hazard value is equal to unity which corresponds to the upper boundary of radium^{11,12} equivalent activity (370) $Bqkg^{-1}$).

4.5 Internal hazard index

Radon and its short lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter progenies is quantified by the internal hazard index (H_{in}) , which is given by the following equation:

$$
H_{\rm in} = \left(\frac{A_{\rm Ra}}{185} + \frac{A_{\rm Th}}{259} + \frac{A_{\rm K}}{4810}\right) < 1 \tag{5}
$$

The values of the index (H_{in}) must be less than unity for the radiation hazard to be negligible^{13,14}. The internal hazard index should be less than unity for the radiation hazard to be considered safely and also radon and its daughter product are hazardous to human health $11,12$.

4.6 Gamma radiation representative level index

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 as follows¹⁵:

$$
I_{\gamma} = \left(\frac{A_{\text{Ra}}}{150} + \frac{A_{\text{Th}}}{100} + \frac{A_{\text{K}}}{1500}\right) \dots (6)
$$

This index can be used to estimate the level of γ radiation hazard associated with the natural radionuclide in the materials. The value of these indexes must be less than unity in order to keep the radiation hazard insignificant.

4.7 Annual effective dose equivalent

The measurement of the concentrations of radionuclides in the environment due to terrestrial

gamma radiation from ²³⁸U, ²³²Th and ⁴⁰K, can be estimated by the average outdoor conversion coefficient from absorbed gamma dose rate in the air and the average annual effective dose equivalent (AEDE). The conversion factor values were established^{2,5} by UNSCEAR and its estimated value is 0.7 SvGy^{-1} for gamma ray exposure in environment. The AEDE can be calculated from Eq. (7):

AEDE
=
$$
D(nGy h^{-1}) \times 0.7 (Sv Gy^{-1}) \times 0.2 \times 8760 (h y^{-1})
$$

× 10⁻⁶ ... (7)

where *D* is the dose rate and 8760 is hours in a year. The corresponding worldwide value^{5,12} of AEDE is 0.08 mSv.

5 Results and Discussion

5.1 The specific activity

The results of analysis of specific activity of 238 Useries, 232 Th-series, 40 K and 137 Cs radionuclides in soil samples for four oil sites in Kirkuk-Iraq; Henjera, Jabel Boor, Jambor, and Qutan are presented in Figs 4-13. Table 1 shows the specific activity of ²²⁶Ra, ²³²Th, ⁴⁰K and 137Cs, respectively. The range of measured activity of 226Ra in the soil of Henjera site was 18.42 to 46.62 Bqkg⁻¹ with an average of 36.248 Bqkg⁻¹. The value in Jabel Boor site was 121.01 to 63.33 Bqkg⁻¹ with an average of 37.027 Bqkg⁻¹. The value in Jambor site was 17.87 to 52.96 Bqkg⁻¹with an average of 33.144 Bq kg ¹ and in Qutan ranged from 7.31 to 39.85 Bq kg⁻¹ with an average 27.568 Bq kg⁻¹. The minimum value was obtained in sample of code KKOSO4 $(7.31 \text{ Bq kg}^{-1})$ and a maximum value was for the sample of code KJBSO6 (63.33 Bq kg⁻¹). The high recorded values of the radionuclides in some soil samples may be due to the geochemical composition and origin of soil types.

Fig. 4 — Comparison of specific activity of ²³⁸U-series with the worldwide average value for the soil samples from Henjera site.

Fig. 5 — Comparison of specific activity of 232Th-series with the worldwide average value for the soil samples from Henjera site.

Fig. 6 — Comparison of specific activity of 238U-series with the worldwide average value for the soil samples from Jabel Boor site.

Fig. 7 — Comparison of specific activity of 232Th-series with the worldwide average value for the soil samples from Jabel Boor site.

Besides, the study area contains some old oil wells. The differences were attributable to the geochemical composition and origin of soil types in a particular area. The range of measured specific activity of ²³²Th for the soil in Henjera was 9.37 to 31.10 Bq kg⁻¹with an average of 21.691 Bq kg^{-1} , the value in Jambor site

Fig. 8 — Comparison of specific activity of 238 U-series with the worldwide average value for the soil samples from Jambor site.

Fig. 9 — Comparison of specific activity of 232 Th-series with the worldwide average value for the soil samples from Jambor site.

Fig. 10 — Comparison of specific activity of 238U-series with the worldwide average value for the soil samples from Qutan site.

was 16.33 Bq kg⁻¹to 42.95 Bq kg⁻¹ with the average of 24.054 Bq kg^{-1} , the value in Gabel Boor site was 11.34 Bq kg⁻¹ to 26.83 Bq kg⁻¹ with the average of 14.628 Bq kg⁻¹ and the value of Qutan was 13.54 Bq kg⁻¹ to 21.54 Bq kg⁻¹ with the average of 16.266 Bq kg^{-1} . The minimum value was obtained in sample code KKOSO4 $(3.54 \text{ Bq kg}^{-1})$ and a maximum for the sample code KJBSO6 (42.95 Bq kg^{-1}).

Fig. 11 — Comparison of specific activity of ²³²Th-series with the worldwide average value for the soil samples from Qutan site.

Fig. 12 — Comparison of specific activity of 40 K with the worldwide average value for the soil samples from Henjera, Jabel Boor, Jambor and Qutan sites.

Fig. 13 — Specific activity of $137Cs$ for the soil samples from Henjera, Jabel Boor, Jambor and Qutan sites.

The differences were significant in all samples. The specific activity of $40K$ in Henjera site was 181.56 to 649.75 Bq kg^{-1} , with an average value of 365.388 Bq kg^{-1} in Jabel Boor site was 245.25 to 798.52 Bq kg^{-1} with an average value of 474.52 Bq kg⁻¹ in Jambor site was 172.83 to 588.38 Bq kg^{-1} with an average 360.138 Bq kg^{-1} and the value in Qutan was 103.21 Bq kg^{-1} to 360.42 Bq kg⁻¹ with an average was 286.97 Bq kg⁻¹. These differences were also attributable to the soil type differences in the region under investigation besides the industrial oil waste. The minimum obtained value in sample code KKOSO4 $(103.21$ Bq kg⁻¹) and a maximum for the sample code KJBSO6 (798.52 Bq kg⁻¹). The differences are significant in all samples.

Moreover, our obtained average values fall within the range of corresponding world values and other published results⁵ mentioned in Table 2. The world average specific activity of 226 Ra is 35 Bq kg⁻¹ with ranges of 17- 60 Bq kg⁻¹, ²³²Th is 30 Bq kg⁻¹ with ranges of 11-64 Bq kg⁻¹ and ⁴⁰K is 400 Bq kg⁻¹ with ranges of 140-850 Bq kg^{-1} . The observed results in some samples showed that the specific activity values for 238 U and 232 Th for the investigated sites were higher than the reported international radioactivity levels^{5,16} of 226 Ra and 232 Th. The high recorded values of the radionuclides in some soil samples may be due to the presence of radioactive rich granite, phosphate, sandstone and quartzite. Notably, the study area contained some old oil wells.

Also, it can be seen from Figs 4-11 that, the specific activity of Radium is higher than thorium in all samples. It is also observed in Fig. 12 that, the measured specific activity of $40K$ exceeds markedly the values of both uranium and thorium, as it is the most abundant radioactive element under consideration.

5.2 Radiological effects

The obtained radiological effects such as the radium equivalent (*Ra*eq), the absorbed gamma dose rate (*D*), the external (H_{ex}) and the internal (H_{in}) hazard index, the radioactivity level index (I_y) and the annual effective dose equivalent (AEDE) for the soil samples collected from region under investigation are shown in Table 3.

The *Ra*eq for the soil samples in Henjera site was between 45.80 Bq kg⁻¹ to 137.713 Bq kg⁻¹ with an average value of 99.255 Bq kg^{-1} , in Jabel Boor site was from 42.05 Bq kg^{-1} to 188.23 Bq kg^{-1} with an average value of 108.247 Bq kg⁻¹, in Jambor site was from 55.52 Bq kg⁻¹ to 113.76 Bq kg⁻¹ with an average value of 108.247 Bq kg^{-1} and in Qutan site was from 20.30 Bq kg⁻¹ to 89.22 Bq kg⁻¹ with an average value

Table 1 — Specific activity of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in soil samples collected from four sites; Henjera, Jabel Boor,

Table 2 — Comparison of natural radioactivity levels in soil samples collected from four sites; Henjera, Jabel Boor, Jambor and Qutanof Kirkuk-Iraq with those in other countries (UNSCEAR, 2000)

of 73.25 Bq kg-1. It was inferred that for all the soil samples analyzed, the radium equivalent activity value was well within and less the permissible limits¹⁰ of $370 Bq kg⁻¹$.

The obtained absorbed gamma dose rate varied from 23.026 to 67.56 nGyh⁻¹, with an average value of 49.241 nGyh⁻¹ in Henjera site. The value was 35.65 to

92.60 nGyh⁻¹ with an average of 53.437 nGyh⁻¹ in Jabel Boor site. The value was 28.55 to 61.52 nGyh⁻¹ with an average of 43.694 nGyh⁻¹ in Jambor site. The value in Qutan site was 10.35 to 43.93 nGyh⁻¹ with an average value of 35.522 nGyh⁻¹. Some values were higher than the international recommended⁵ value 55 nGyh⁻¹, but the averages values were less than it.

The calculated external hazard indexes (*H*ex) varied from 0.12 to 0.38 with an average value of the 0.269 in Henjera site, the range in Jabel Boor site was 0.186 to 0.488 with an average value of the 0.281, the range in Jambor site was 0.145 to 0.319 with an average value of the 0.228, and the values in Qutan site varied from 0.053 to 0.251 with an average value of the 0.191. The calculated average values were less than 1. The internal exposure by radon and its progeny was controlled by the internal hazard index (H_{in}) . H_{in} ranged between 0.17 and 0.475 with an average value of the 0.356 in Henjera site, the range in Jabel Boor site was 0.269 to 0.664 with an average value 0.381, the range in Jambor site was 0.201 to 0.462 with an average value 0.317and in Qutan site was 0.073 to 0.355 within an average value 0.263. The average values were less than 1.

The calculated I_{γ} values for all the samples were presented in Table 3. The values in Henjera site ranged from 0.34 to1.03 with an average of 0.734, in Jabel Boor site, it varied from 0.525 to 1.397 with an average 0.80, in Jambor site, it varied from 0.402 to 0.895 with an average 0.648, the range in Qutan site was 0.153 to

0.685 with an average value 0.537. The calculated values for all samples were lower than the international values $(I_1 < 1)$ while samples KHNSO5 and KJBSO6 were higher than the international values $(I_1 > 1)$ as shown in Table 3.

The calculated indoor and outdoor AEDE values were quoted in Table 3. The results of outdoor and indoor effective dose were 0.0621, 0.076, 0.0552 and 0.045, and 0.262, 0.269, 0.241 and 0.175 for Henjera, Jabel Boor, Jambor and Qutan sites, respectively. It can be seen that the above-mentioned values were lower than the corresponding worldwide values of 0.08 and 0.42 mSv, respectively⁵.

6 Conclusions

Gamma ray spectrometry was employed to determine specific activity due to naturally and artificially occurring 226 Ra, 232 Th, 40 K and 137 Cs radioisotopes and the associated radiation hazard levels in 23 of soil samples from four different oil locations in Kirkuk-Iraq. Most of the specific activity values were lower than the world average values with some

exceptions which were described in figures and tables of this study, so we should focus on these areas by collecting more samples and greater depths to give more accurate description of the areas. The high recorded values in some samples may be due to the geochemical composition and origin of soil types. Besides, the study area contained some old oil wells. The averages values of the obtained radiological effects such as the radium equivalent (*Ra*eq), the absorbed gamma dose rate (D) , the external (H_{ex}) and the internal (H_{in}) hazard index, the radioactivity level index (I_y) and the annual effective dose equivalent (AEDE) were within the limit of the international recommended values except that of samples KHNSO5 and KJBSO6. It was concluded that no harmful radiation effects were posed to the population who live in the study area.

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