Preliminary Determination of Natural Radioactivity Levels of the State of Qatar using High-Resolution Gamma-ray Spectrometry

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ABSTRACT

The State of Qatar is a peninsula with a total area of 11,437 km^2 which lies over a geological formation comprising a sequence of limestone, chalk, clay and gypsum. Establishing a baseline for the radioactivity concentration in Qatar's soil is the main purpose behind the present study. The project is focused on obtaining measurements of representative soil samples from various areas in Qatar to establish concentrations of the ^{235,8}U and ^{232}Th natural decay chains and also the long-lived naturally occurring radionuclide ^{40}K. The ^{235,8}U, ^{232}Th and ^{40}K concentrations have been measured via high-resolution gamma-ray spectrometry using a hyper-pure germanium detector situated in a low-background environment with a copper inner-plated passive lead shield. A wide range of different gamma-ray energy transitions lines arising from the multiple decay products from the ^{235,8}U and ^{232}Th decay chains have been analyzed separately to obtain more statistically significant overall results.

INTRODUCTION

Traces of radionuclides are found in water, air, soil and human bodies. We inhale and ingest radionuclides every day of our lives and radioactive material has been ubiquitous on earth since its creation. The presence of natural radioactivity in soil results in external and internal exposure to humans. Radioactive elements which can be found in nature are generally categorized in two distinct families, namely of arising from either 'Cosmogenic' or 'Terrestrial' Origin. The most commonly encountered radionuclides are ^{235,8}U and ^{232}Th and their subsequent radioactive decay products and ^{40}K [1,2]. The presence of natural occurring radioactive material (NORM) in the terrestrial composition of the natural background is dependent on the geological composition of the soil and rocks. Therefore, systematic and accurate measurements of the radioactivity level in soils are essential for understanding changes in the natural radiation background as a function of geographical location and time [3,4,5].

To our knowledge, there is no current literature on the level of environmental naturally occurring radioactivity in the State of Qatar. The main purpose behind the present study is to establish a base line value for the radioactivity concentration in Qatarian soil. Measurements of the levels of natural background level of the radioactivity from ^{235,8}U and ^{232}Th (and their decay progeny), as well as the primordial radionuclide ^{40}K are the main objective of the current study. In addition, measurements of activity arising from decays of the artificially created fission product ^{137}Cs have been made in parallel.

In soil samples it is important to monitor any change in the activity concentration with time. The soil samples measured in the current work were collected from sampling points which were located at regular intervals of a 1:5000 square grid according to Qatar National Grid (QNG) (see fig. 1). We also aim to measure the outdoor Annual Effective Dose Equivalent for Qatar.

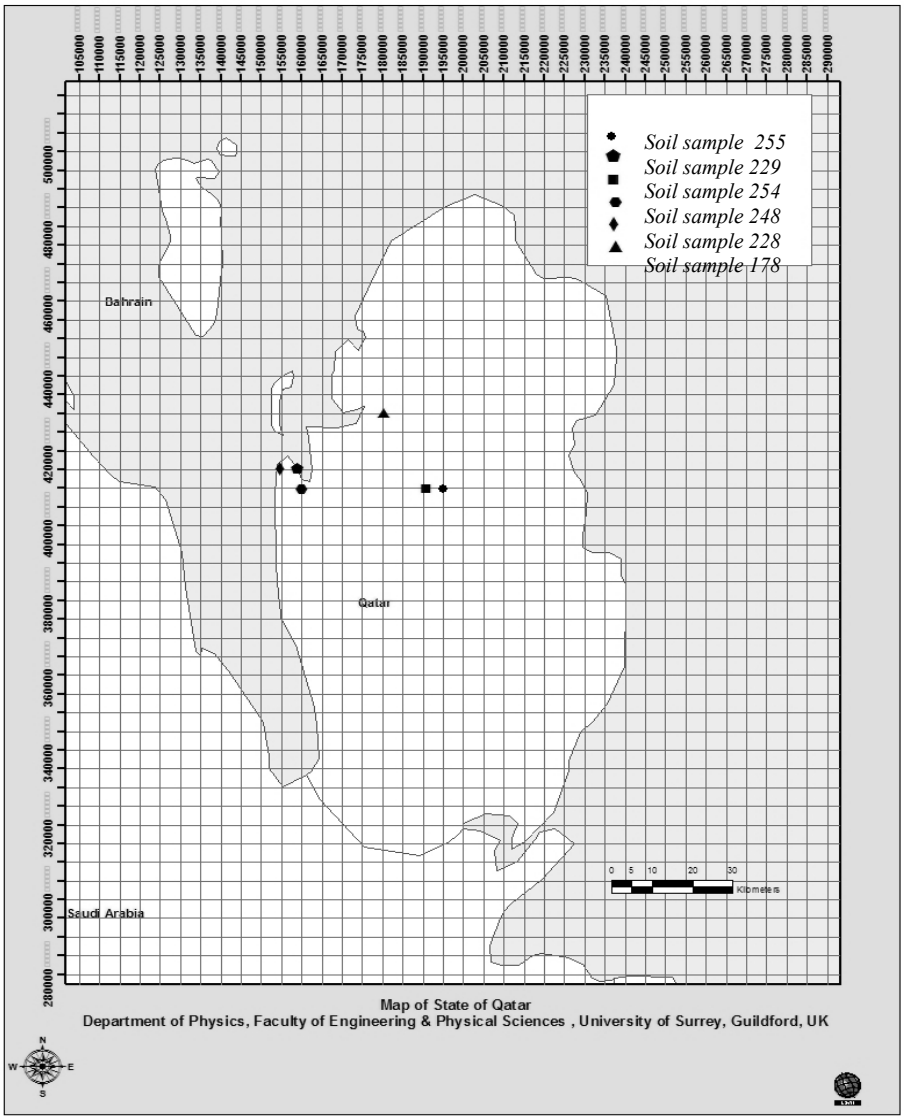

Figure 1: A map of Qatar showing the sampling points located at regular intervals on grids with a regular grid distance of 5 km. This map was created using ArcView software.

The State of Qatar occupies a total area of 11,437 km^2 extending approximately 235 km north-south, and 95 km east-west. It is situated between (25,30) north latitudes and (51,15) east longitudes. It shares a geographical border with the Kingdom of Saudi Arabia to the south. The surface of Qatar lies over a geological formation comprising a sequence of

limestone, chalk, clay and gypsum. This formation forms the largest part of Qatar's surface geology [6] (see fig. 2).

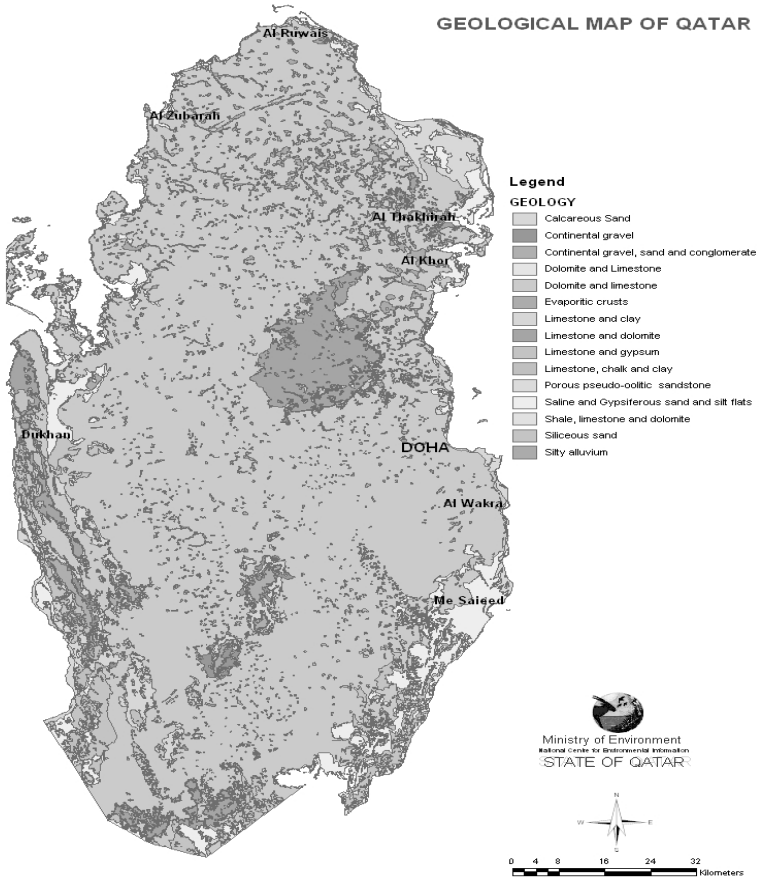

Figure 2: Geological map of Qatar, from The Ministry of Environment, Doha, Qatar.

EXPERIMENTAL PROCEDURES

2.1 Sample preparation

The soil samples measured at 5→15 cm depth level were collected from sampling points which were located at regular intervals of a 1:5000 (5 km) square grid according to Qatar National Grid (QNG). The location of the samples is shown on the map of Qatar in fig. 2. The first, preliminary results of the analysis from six samples are presented in the current work. Stones were manually eliminated from the samples, which were then sieved in their locations using a 2 mm sieve. The post-sieved samples were then filled into labeled polyethylene bags with individual sample masses in the 1→1.5 kg range. The final sample preparation and all the gamma-ray measurements to determine the sample radioactivities were performed in the radiation detection laboratories of the Centre for Nuclear and Radiation Physics, situated within the Department of Physics at University of Surrey, Guildford, UK.

Prior to final measurement in the laboratories in Guildford, the samples were placed in a drying oven at a temperature of 60^{o}C for 24 hours to ensure that any significant moisture was removed from the samples. A 500 μm mesh was then used to sieve the samples which were then weighed and transferred to 200 ml polyethylene bottles. Finally, the samples were stored

and kept sealed for about one month in order to reach radioactive secular equilibrium (> 7 half-lives of ^{222}Rn and ^{220}Rn).

2.2 Instrumentation and calibration

Sample gamma-ray energy spectra were measured using a coaxial high purity germanium (HPGe) detector. Unwanted ambient radioactivity from room background sources was reduced by surrounding the detector with a passive lead shield of about 10 cm thickness. The Pb shield incorporated an internal copper layer on the inner surface of the shield (which directly viewed the gamma-ray detector) which significantly reduced the effects from photo-fluorescence of the Pb (i.e. absorbed the Pb K_α and K_β X-rays) from gamma rays emitted from the source material. The detector output was connected to a standard spectroscopy amplifier (Canberra 2022) and the data taken directly to an ORTEC Multi-Channel Analyser for subsequent off-line analysis.

Figure 3: a coaxial high purity Ge (HPGe) detector surrounded by lead shield of about 10 cm thickness.

The energy calibration and relative efficiency calibration of the detector were carried out using photopeaks peaks from standard ^{152}Eu, ^{137}Cs, ^{241}Am and ^{133}Ba sealed point sources which were placed 14 cm (the same distance as the front face of the polyethylene bottles from the front face of the detector), 18 and 22 cm from the front face of the HpGe detector. (fig. 3). An empty polyethylene bottle of the same geometry was used to determine the background spectrum observed by the germanium detector. The counting time of each sample (and also the ambient background spectrum) was for 24 hours (i.e., 86,400 sec). The Lowest Detectable Limits (LDL) above the background level for specific transition from the decays of ^{226}Ra (186.1 keV), ^{228}Ac (911.2 keV) and ^{40}K (1461 keV) were also determined. These values were found to be 18.3 ± 4.5, 6.2 ± 2.0 and 68 ± 13 Bq/kg respectively. A reference material (IAEA soil-375) was used as a quality assurance reference sample for the detector set-up. The geometrical dimensions of the samples were kept same to that of the reference material used in measurement. Since the counting rate is proportional to the amount of the radioactivity in the samples, the Activity Concentration (*Ac*) can be calculated from the following equation:

$$Ac = \frac{C_{net}}{I_\gamma \times E_{ff}(E_\gamma) \times m} \quad \text{....(1)}$$

where C_{net} is the net peak counts,

I_γ is absolute gamma decay intensity for the specific energy photopeak (including the decay branching ratio information),
Eff (E_γ) is the Absolute Efficiency of the Germanium detector at this energy
and *m* is the mass of the sample in kg.

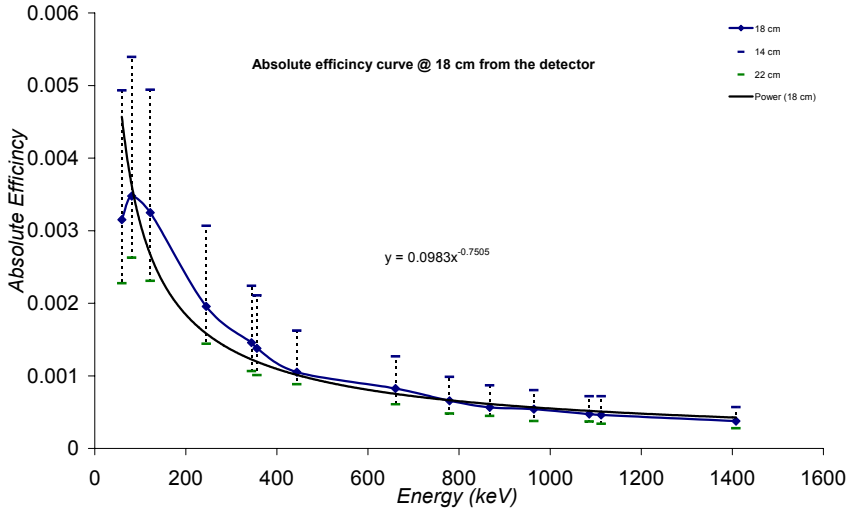

Figure 4: Absolute Efficiency calibration using peaks from standard ^{152}Eu , ^{137}Cs, ^{241}Am and ^{133}Ba sealed point sources which were placed at distances of 14, 18, 22 cm from the front face of the HpGe detector.

The preliminary activity concentration of ^{238}U present in the samples was estimated using the γ ray at 186 keV associated with the decay of ^{226}Ra. Additional γ-ray transitions from decays of shorter-lived radionuclides in the ^{238}U decay chain such as ^{214}Pb (351keV) and ^{214}Bi (609keV) were also used to estimate the preliminary measurements for the activity concentration of ^{238}U, however these results can not be compared directly to the ^{226}Ra data as the elapsed time since the samples were sealed was less than 7 half-lives of ^{222}Rn and therefore secular equilibrium between the decay products may not yet have been established. The activity concentration of ^{232}Th was determined using the ^{228}Ac decay γ-ray line at 911 keV. The gamma-ray peaks associated with decays from ^{40}K and ^{137}Cs at 1461 and 661 keV respectively, were used to determine the activity concentrations for these nuclei. The spectra were analyzed using the RADWARE gamma-ray analysis software package [8] and are expressed in Bq/kg.

RESULTS AND DISCUSSION

Fig. 5 shows γ-ray spectra of sample 228 (which had the highest observed activity among the six samples discussed in the present work), a representative background spectrum, and the resultant background subtracted spectrum for sample 228 with the background spectrum subtracted channel by channel using the GF3 program in the RADWARE analysis suite. Sample 228 clearly shows transitions associated with the decay of ^{238}U, while evidence for

the ^{228}Ac lines associated with the ^{232}Th series was sparse at best. Figure 6 shows the γ-ray spectrum of sample 228 in detail.

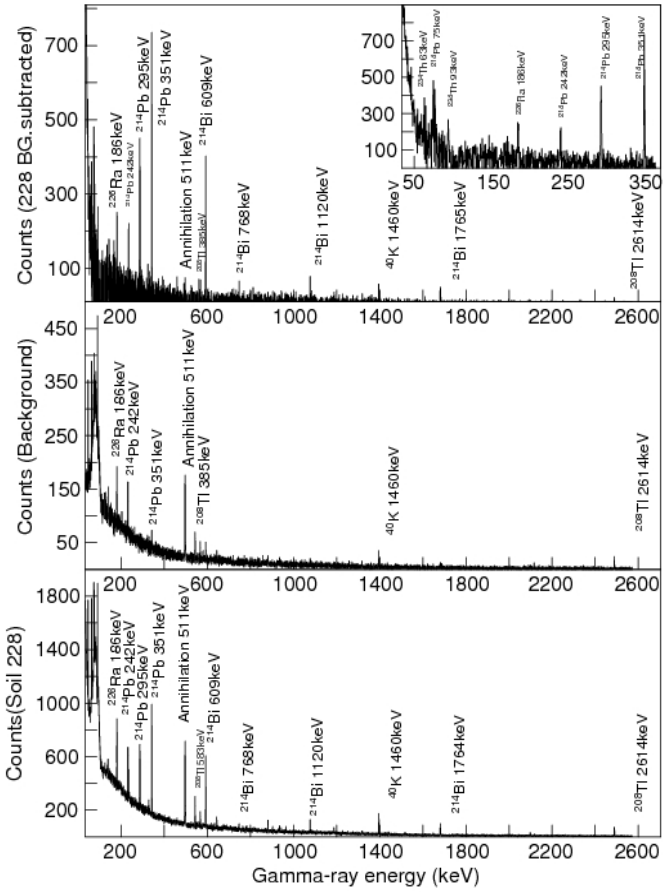

Figure 5:(Lower panel) Raw γ-ray spectra of sample 228; (Central Panel) background spectrum accumulated in the absence of any source sample; (Upper Panel) background-subtracted gamma-ray spectrum associated with soil sample 228 with lines from the ^{238}U decay series clearly identified.

The radioactivity concentrations in the six locations in west of the Qatarian peninsula are given in Table 1. As secular equilibrium was not yet achieved at the time of the measurements presented in this preliminary report, the ^{226}Ra and ^{228}Ac radionuclides from ^{238}U and ^{232}Th series have been used to calculate the activity concentrations of ^{238}U and ^{232}Th respectively in each of the six samples discussed here. Transitions associated with decays from ^{214}Pb and ^{214}Bi (from the ^{238}U series) and ^{228}Ac (from the ^{232}Th decay chain) were also measured. Table 1 gives the preliminary results for the dry weight activity concentrations of ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{40}K and ^{137}Cs in the investigated samples.

The concentrations of ^{238}U and ^{232}Th are compared with measurements of other soils by different authors. The Ra equivalent, absorbed dose rate, external hazard index and annual effective dose rates in soil samples were calculated and compared with the worldwide values. The activity concentration of the radionuclides ^{238}U, ^{232}Th and ^{40}K in sample 228 were found to be 301±119, 14±6, 160±63 Bq/kg. The worldwide concentrations of the radionuclides ^{238}U, ^{232}Th and ^{40}K have averages in soil samples of 40, 40, and 580 Bq/kg respectively [7]. The

activity concentration of ^{238}U in sample 228 appears to be higher in the current work than the worldwide average value and also significantly higher than the values determined for samples 229, 255 and 248 in the current work. The activity concentration of ^{238}U in samples 254 and 178 were found to be below the lower detectable limit (LDL). Those of ^{232}Th were slightly above the lower detectable limit, although two of the six samples were below the LDL. The activity concentrations of ^{40}K in the six samples were about one third of the average worldwide value. Since ^{137}Cs dose not exist in soil naturally and the soil samples were collected from 5 to 15 cm depth, only one sample has a reported activity concentration for ^{137}Cs (of 10±4 Bq/kg). The large uncertainties quoted in this preliminary report (typically around 30%) is directly associated with uncertainties in the photopeak efficiency response for the samples. The calibration measurements were made with simple, standard point sources at distances of 14, 18 and 22 cm from the front face of the detector. The difference between these point source efficiencies at 14 and 22 cm is taken in the current work to establish an upper limit in the uncertainties associated with the difference between the efficiency point sources and sample geometries. Note that in future work, calibration sources with similar geometries and densities to the samples will be used to determine the spectral response of the HpGe detector which will significantly reduce this part of the experimental uncertainty compared to these preliminary measurements.

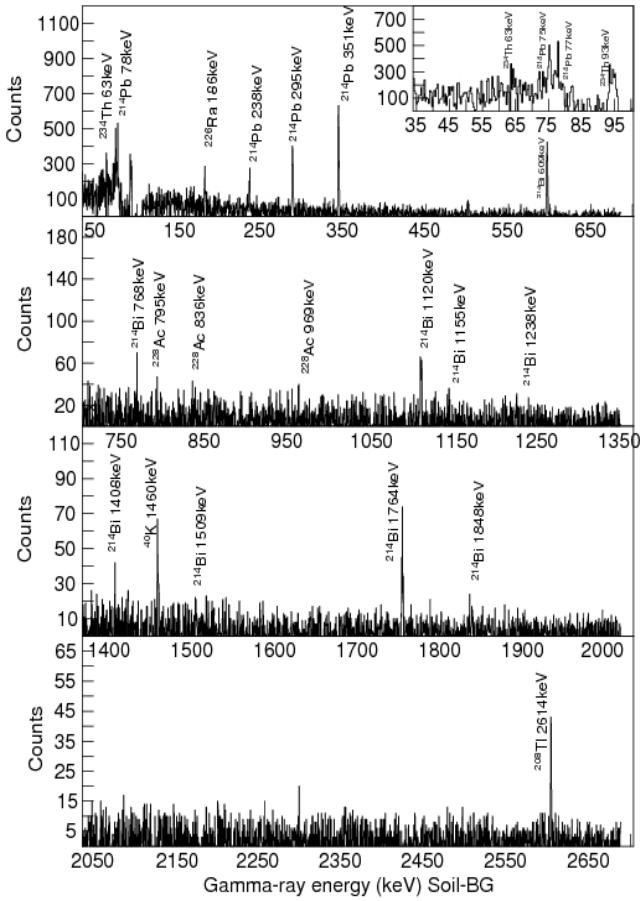

Figure 6: Background-subtracted gamma-ray spectrum associated with soil sample 228 with lines from the ^{238}U decay series clearly identified spread on four panels.

Table 1

Radioactivity concentration of ^{238}U Series, ^{232}Th series, ^{40}K and ^{137}Cs in Bq/kg in soil samples from 6 different locations in state of Qatar, compared with other area of the world. The Ra equivalent, absorbed dose rate, external hazard index, and annual effective dose rates in soil samples were calculated and compared with the worldwide values.

Sample Code	Location according to QNG*	^{238}U Series (Bq/kg)			^{232}Th Series (Bq/kg)	^{40}K (Bq/kg)	^{137}Cs (Bq/kg)	D (nGy/h)	Ra_{eq} (Bq/kg)	H_{ex}	Effective Dose Rate (μSv/year)
		^{226}Ra	^{214}Pb	^{214}Bi	^{228}Ac						
228	(155248,414314)±5m	301 ±119	178 ±70	157 ±62	14 ±6	160 ±63	**	145±102	332±234	0.9±0.6	177±125
229	(160000,415000)±5m	68 ±27	16 ±6	11 ±4	11 ±5	197 ±78	10 ±4	45±32	98±71	0.3±0.2	55±40
178	(195000,410000)±5m	**	11 ±4.5	9 ±4	25 ±10	269 ±106	**				
254	(180000,430000)±5m	**	14 ±6	11 ±4	**	296 ±116.5	**				
255	(195000,410000)±5m	97 ±38	17 ±7	15 ±6	11 ±5	234 ±92.00	**	59±42	129±93	0.4±0.3	72±52
248	(160000,410000)±5m	160 ±63	7 ±3	**	**	193 ±76	**				
Algeria [9]		2-127			2-144	35-1405	0.1-43				
Egypt (Southern Egypt) [4]		5.0-24			2.0-10	293-660					
Jordan [2]		15-60			4.0-29	99-379					
India [10]		5.0-71			15-76	20-854					
Pakistan [11]		21-48			22-59	303-945	1.0-5				
Bangladesh [12]		13-43			3.0-81	402-750	3.0-10				
China [13]		40-442			33-88	442-913					
Worldwide [7,14,15]		40			40	580		55	370		460

* Qatar National Grids

** Below Lower Detectable Limit

Gamma Dose Rate (D)

Gamma dose rate (D) in the outdoor air at 1 m above the ground level can be calculated by the equation 2 [13,16]:

$$D = (R_K \times Q_K) + (R_U \times Q_U) + (R_{Th} \times Q_{Th}) \quad \textbf{....(2)}$$

where R_K (0.042), R_U (0.430), and R_{Th} (0.666) are the conversion factors for ^{40}K, ^{238}U and ^{232}Th series respectively which are published in UNSCEAR, 2000. Q_K, Q_U and Q_{Th} are the activity concentrations of ^{40}K, ^{238}U and ^{232}Th series respectively in Bq/kg. Table 1 shows the lowest dose rate was 45 nGy/h for the soil represented by sample 229, while the highest dose rate was 145nGy/h for the soil represented by sample 228. The published maximal admissible dose rate is 55nGy/h. The value determined for Sample 228 in the current work is higher than the worldwide value.

Radium Equivalent Activity (Ra_{eq})

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 [17]. This can be achieved using equation 3:

$$Ra_{eq} = (Q_K \times 0.077) + (Q_U) + (Q_{Th} \times 1.43) \quad \textbf{.....(3)}$$

The published maximal admissible Ra_{eq} is 370 Bq/kg. It is noteworthy that all the results obtained in the present study are lower than the suggested maximal admissible value [7].

External Hazard Index (H_{ex})

The external hazard index is an evaluation of the hazard of the natural gamma radiation. It can be calculated from equation 4:

$$H_{ex} = \frac{Q_U}{370} + \frac{Q_{Th}}{259} + \frac{Q_K}{4810} \leq 1 \quad \textbf{....(4)}$$

The calculated external hazard indexes were all below the unity.

Annual Effective Dose Equivalent (AEDE)

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 had to be taken into consideration. This value is published in UNSCEAR (2000) of 0.7 Sv/Gy. The outdoor occupancy factor which is about 0.2 is given in the same reference. The annual effective dose equivalent is given by the following equation:

$$AEDE(\mu Sv/y) = D(nGy/h \times 8760(h/y) \times 0.2 \times 0.7(Sv/Gy) \times 10^{-3} \quad \textbf{... (5)}$$

The results are presented in Table 1. The highest annual effective dose rate value was found to be 177 μSv/year. While the lowest value was found to be 55 μSv/year. The world average annual effective dose equivalent (AEDE) from outdoor terrestrial gamma radiation is 460μSv/year [7]. Therefore, the obtained values from this preliminary study are all lower than the accepted, average worldwide value.

CONCLUSIONS

The measured level of the natural radiation background in the present study from the six investigated samples show that the studied areas have normal levels of background radiation with the exception of sample 228 which shows an above average value for the ^{238}U concentration. All the obtained ^{40}K values show levels within the natural background values.

Preliminary values for the Radium equivalent (Ra_{eq}), Radiation hazard index (H_{ex}) and Annual Effective dose equivalent (AEDE) were determined for each of the samples. These indicate that the areas monitored can be regarded as having normal levels of natural background radiation. The obtained value of gamma dose rate (nGy/h) in the outdoor air at 1 m above the ground level on the location where sample 228 was collected is relatively high. As just one sample represents the area with elevated level of ^{238}U this suggests in future that more samples are collected from the same area on scale of 1:2.5 km^2 to further investigate the level of ^{238}U to compare with the results of the present preliminary study. A significant uncertainty is associated with the gamma-ray detection efficiency in these preliminary results. Future work on the same samples will include the use of identical geometry mixed calibration sources to the samples which were measured using efficiency data from a point source which was not a similar geometry to the samples. The future analysis will also be extended to include data arising from higher energy gamma-ray transitions such as the 2.6 MeV line from the decay of ^{208}Tl decay in the ^{232}Th chain.

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