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Assessment of Radioactivity in Some Soil Samples of Qatar by Gamma-Ray Spectroscopy and the Derived Dose Rates

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Abstract. The activity concentrations of some natural and artificial radionuclides have been measured in soil samples by gamma-ray spectroscopy using a high purity germanium detector. From the obtained gamma-ray spectra, the activity concentrations of ^{238}U and ^{232}Th natural decay series and the long-lived naturally occurring radionuclide ^{40}K have been determined, in addition to the fission product ^{137}Cs. A wide range of different gamma-ray lines ranging from ~100 keV up to 2.6 MeV, associated with the decay products of ^{238}U and ^{232}Th series have been analyzed independently to obtain more statistically significant overall results. The data have been analyzed, when secular equilibrium of the radionuclides is achieved within the samples. The weighted activity concentrations of ^{238}U and ^{232}Th series vary from 4.4 to 64.4 and 0.8 to 7.6 Bq/kg, respectively. The activity concentrations of ^{40}K and ^{137}Cs have been found in the range of 13.6 to 179.5 and 0.3 to 3.7 Bq/kg, respectively. Based on the measured activity concentrations, dose rate, radium equivalent, radiation hazard index and annual effective dose rates have been estimated. The values obtained are within the recommended safety limits.

Keywords: Qatar soil, gamma-ray spectroscopy, dose rate, radiation hazard index, radium equivalent

Introduction

Human beings are primarily exposed to ionizing radiations emitted from natural and man-made sources. Natural radiations originate from the interaction of cosmic rays particles with the atmospheric elements and naturally-occurring radioactive elements found in the earth's crust (EPA, 2007). In soil, the radioactivity concentrations give information on both natural and man-made sources which is important in radiological monitoring and radiation dose assessment for public. The primary radioactive elements found in the earth's crust are uranium, thorium and their radioactive decay products and potassium (EPA, 2007). The study of these radionuclides in soil plays an important role to assess the biological effect of natural radiation in biotic environment. The global effective dose rate of public exposure from soil with weighted mean activity concentration of 33, 45 and 420 Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively is 460 μSv/y (UNSCEAR, 2000). Measurement of activity concentration level of induced fission product ^{137}Cs is also essential because 60% of the collective effective dose from external radiation due to nuclear weapons testing can be attributed to ^{137}Cs (Karunakara *et al.,* 2001).

The worldwide literature on the levels of natural radiation from soil is rich (Rahman *et al.,* 2009; Kurnaz *et al.,* 2007; Akhtar *et al.,* 2005; Ramli *et al.,* 2005; Rani and Singh, 2005; Yang *et al.,* 2005; Banzi *et al.,* 2000; Karahan and Bayulken, 2000), however, the number of studies made in the Arabian Gulf is comparatively limited (Al-Hamarneh and Awadallah 2009; Saad, 2002; Goddard, 2001).

This study is a part of a project, designed to provide a radiological map of Qatar. The present work investigated the radiation levels of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in soil samples across Qatar State, as well as to assess the dose rate for the public. Limited number of representative soil samples were collected along the Qatarian peninsula.

Many more samples have been collected from the studied area at regular intervals of 1 per 100 km^2, to a depth of 0-5 and 5-15 cm, to further investigate the depth of radioactivity across soil profile. These samples are currently under investigation and the findings would be presented in separate papers.

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Materials and Methods

Sample collection and preparation. 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 (Al-Sulaiti, 2011; MOFA, 2007).

Five soil samples were collected from different sites in the State of Qatar. One sample from coastal region, one from oil field area and three samples were collected from the rest of Qatar as shown in Fig. 1.

Approximately 1.5- 2 kg of soil was taken from 5 to 15 cm depth from each site. The samples were then filled into labelled polyethylene bags and shifted to the laboratory. Each sample was then placed in a drying oven at 60 °C for 24 h to ensure that any significant moisture was removed from the samples. Then the samples were sieved by 0.5 mm mesh. The minimum grain size was chosen to ensure a full volume filling of the Marinelli beakers. The samples were manually homogenized using a stainless steel spoon and a stainless pan then weighed and transferred to 500 mL labelled Marinelli beakers then stored and kept sealed for about one month in order to ensure secular equilibrium between ^{226}Ra and its daughters.

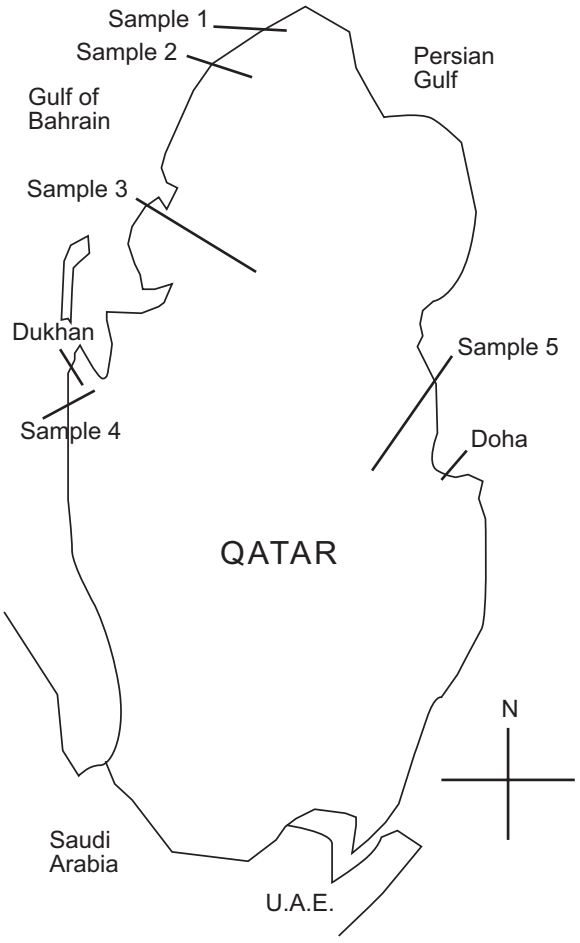

Fig. 1. Map of Qatar showing the sampling locations.

Experimental set up. Soil samples were analysed using a high-resolution, low-background gamma-ray spectroscopy system based on a p-type coaxial high-purity germanium detector (HPGe). The HPGe coaxial detector used is of 1.76 keV (FWHM) and an absolute full-energy peak gamma-ray efficiency of approximately 0.14% at 1332 keV. A crystal of 50 mm diameter was operated under a high voltage, bias of +3000 V (DC). Reducing background radioactivity was achieved by surrounding the detector with a cylindrical passive lead shield of about 10 cm thickness. The selected shaping time was 4 μs. Finally, the resultant spectral data was analysed using Canberra Genie software (Genie, 2000).

Energy calibration. Performance of HPGe detector was characterized by the energy calibration and its absolute full energy peak detection efficiency. The initial energy calibration of the detector was carried out using full-energy peaks from a standard source (^{152}Eu) with an activity of 3.02 kBq. The standard source was provided by High Technology Sources Ltd. (DKD calibration certificate, PTB, Germany, and NIST, USA). The ^{152}Eu was selected for the energy calibration because of its wide range of energies, and long half-life which is around 13.542 years (Sunyar, 1955).

The spectrum was acquired for 1200 sec. The range of energies including 121.78, 344.27, 778.90, 1112.05 and 1407.92 keV were selected to calibrate the initial energy scale of the programme. In order to avoid any drift in the later soil samples measurement, the system was recalibrated on a weekly basis during the sampling procedure.

Efficiency calibration. The absolute efficiency calibration was carried out using full energy peaks of four standard sources i.e. ^{226}Ra, ^{232}Th, ^{152}Eu and NG3 (a mixed source containing ^{241}Am, ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{137}Cs, ^{139}Ce, and ^{203}Hg). The standard sources were provided by High Technology Sources Ltd. (DKD calibration certificate, PTB, Germany, and NIST, USA). As the geometry for the standard sources used in the efficiency calibration and the Marinelli beakers (used for the measurement of soil samples) was identical, the problem of coincidence-summing effects was not significant and the correction factor was taken as unity.

The sources were placed surrounding the germanium detector with the radionuclides dispersed in gel matrices in Marinelli beakers of the same geometry used for the samples. The source activities of ^{226}Ra, ^{232}Th and ^{152}Eu were 3.10, 1.08, 3.02 kBq, respectively for active volumes of 550 mL each and were spread homogenously in gel matrices of densities 1.1, 1.1, and 1.6 g/cm^3, respectively. These were taken to be representative of the density of the samples measured and therefore, corrections for gamma-ray self-attenuation within the samples were accounted for the initial efficiency measurements using these Marinelli housed sources. The efficiency calibration spectra were obtained for 86,400 sec each (uncertainty value of counting was 5%). A range of discrete gamma-ray energies from 0.059 MeV (energy peak of ^{241}Am) up to 2.614 MeV (energy peak of ^{208}Tl) were covered using these standard sources. A Marinelli beaker with the same geometry, filled with de-ionized water, was used on a weekly basis during the measurements period to determine the background spectrum. The counting time of the ambient background spectrum was also 86,400 sec.

Samples analysis. The samples were placed directly over the front face of the detector (dead time ranged between 0.06 and 0.23%). The acquisition time for each sample was 1 day. 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 ~100 keV to 2.614 MeV, 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. ^{226}Ra, ^{214}Pb and ^{214}Bi activity concentrations were used to determine the activity of ^{238}U. The activity concentration of ^{232}Th was determined using gamma-ray transitions lines of ^{228}Ac, ^{212}Pb and ^{208}Tl. Gamma-ray 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.

The activity concentration (Bq/kg) was calculated using the following equation:

$$A = C_{net} / \gamma \times \varepsilon(E_\gamma) \times m \quad (1)$$

where, C_{net} is the net peak counts, γ is the absolute gamma ray intensity for the specific energy photo peak, $\varepsilon(E_\gamma)$ is the absolute photo peak efficiency of the germanium detector at this energy and m is the mass of the sample in kg.

Calculation formulae for dose rates assessment. Gamma dose rate (D) in the outdoor air at 1 m above ground level was calculated by the following equation (Al-Ghorabie, 2005; Singh *et al.,* 2005; Saito *et al.,* 1990):

$$D = \sum_x A_x \times C_x \quad (2)$$

where, A_x (Bq/kg) is the mean activity of ^{226}Ra, ^{232}Th or ^{40}K, and C_x (in units of nGy/h/Bq/kg) is the corresponding dose conversion factor. The dose conversion factors used in the calculation for ^{226}Ra, ^{232}Th and ^{40}K were 0.461, 0.623 and 0.0414, respectively (Singh *et al.,* 2005; UNSCEAR, 1993). The maximum permissible dose rate is 51 nGy/h (UNSCEAR, 1993).

Radium equivalent of the samples (Bq/kg) was calculated by the following equation (Rahman *et al.,* 2009):

$$Ra_{eq} = (A_K \times 0.077) + (A_U) + (A_{Th} \times 1.43) \quad (3)$$

The permissible limit of Ra_{eq} is 370 Bq/kg in soil samples that contain ^{238}U, ^{232}Th and ^{40}K measured in Bq/kg (UNSCEAR, 2000).

The external hazard index was calculated by the following equation (Kumar *et al.,* 2003; Amrani and Tahtat, 2001; Beretka and Methew,1985):

$$H_{ex} = (A_U /370) + (A_{Th} / 259) + (A_K/4810) \leq 1 \quad (4)$$

where, A_U, A_{Th} and A_K are the specific activities of ^{238}U, ^{232}Th and ^{40}K, respectively.

The external hazard index should be below unity for the radiation hazard to be negligible (Ibrahim *et al.,* 2009).

The annual effective dose equivalent was calculated by the following equation (UNSCEAR, 1988):

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

The world average annual effective dose equivalent (AEDE) from outdoor terrestrial gamma radiation is 460 μSv/y (UNSCEAR, 2000).

Results and Discussion

Activity concentrations of gamma emitters in soil samples. Five soil samples were investigated in this study. The activity concentrations of ^{226}Ra, ^{214}Pb, and ^{214}Bi (from ^{238}U series), ^{228}Ac, ^{212}Pb and ^{208}Tl (from ^{232}Th series), ^{40}K and ^{137}Cs, along with statistical errors in soil samples are presented in Table 1.

In Table 1 the activity concentration of ^{226}Ra varied from 7.5 to 63.3 Bq/kg. The worldwide average value of ^{226}Ra in soil is 33 Bq/kg. The highest value was observed in

Table 1. Activity concentrations of ^{226}Ra, ^{214}Pb, and ^{214}Bi (^{238}U series), ^{228}Ac, ^{212}Pb and ^{208}Tl (^{232}Th series), ^{40}K and ^{137}Cs, in the soil samples

Radionuclide	Activity concentration (Bq/kg)				
	Sample-1	Sample-2	Sample-3	Sample-4	Sample-5
^{226}Ra	7.5±0.5	14.7±2.0	16.3±1.7	63.3±3.2	15.1±1.7
^{214}Pb	4.0±0.2	15.3±0.4	16.4±0.5	64.5±0.8	15.3±0.4
^{214}Bi	4.2±0.2	15.7±0.5	16.5±0.5	64.4±0.9	14.7±0.5
^{228}Ac	0.9±0.3	6.5±0.5	8.8±0.6	3.3±0.5	7.0±0.5
^{212}Pb	0.7±0.1	4.9±0.2	7.2±0.2	2.8±0.2	5.8±0.2
^{208}Tl	1.5±0.3	6.4±0.4	8.2±0.5	2.9±0.3	6.2±0.6
^{40}K	13.6±1.5	179.5±9.1	179.1±8.4	119.6±6.6	143.6±7.5
^{137}Cs	*	3.7±0.3	0.6±0.2	1.3±0.2	0.3±0.2

*Below minimum detactable activity MDA

sample no. 4. It was collected from the site near an oil field area. This area is next to the first oil field in Qatar, which started to produce oil in 1947. The enhanced level of ^{226}Ra in sample no. 4 is likely due to the effects of the nearby oil-fields of which natural occurring radionuclide materials (NORM) are found in oil-production water, that could be discharged into the surrounding environment (Consequently, evaporation of oil co-produce water led to the enhanced concentration of ^{226}Ra in soil).

The activity concentration of ^{228}Ac, ^{212}Pb and ^{208}Tl varied from 0.9 to 8.8 Bq/kg, 0.7 to 7.2 Bq/kg and 1.5 to 8.2 Bq/kg, respectively.

The activity concentration of ^{40}K varied from 13.6 to 179.5 Bq/kg. The lowest value was reported for sample no. 1. The activity concentration of ^{40}K in sample no. 1 was much lower than the average worldwide value, i.e. 420 Bq/kg (UNSCEAR, 2000). Potassium is part of the clay minerals (shale) and its activity concentration can be affected by the relative solubility of potassium in the soil (El Mamoney and Khater, 2004). The activity concentration of ^{40}K in the remaining 4 samples was also below the worldwide average value (UNSCEAR, 2000).

The activity concentration of ^{137}Cs varied from 0.3 to 3.7 Bq/kg. It was not detected in sample no. 1 and the obtained values for other 4 samples were lower than the observed values reported in other countries. Reported values of ^{137}Cs in Jordan in 2002 ranged between 7.75 and 576 Bq/kg for a sampling depth of 0-2 cm (Al Hamarneh *et al.,* 2003). Concentration of ^{137}Cs in the soil samples collected from Islamabad, Pakistan was in the range of 1.6 and 9.45 Bq/kg (Rahman *et al.,* 2009). In 1991 the range of ^{137}Cs activity concentration in bottom sediments of the Eastern part of the Baltic was between 6.3 and 65.7 Bq/kg (Luksiene *et al.,* 1992). A mean value of 4.7 Bq/kg for ^{137}Cs was measured in soil samples collected in 1998 from the Nile delta and the north coast of Egypt (Shawky and El-Tahawy, 1999). Mean value of ^{137}Cs concentration in soil samples collected from different regions of the Algerian coast on the Mediterranean Sea in 1996 was 4.2 Bq/kg (Benamar *et al.,* 1997).

^{137}Cs exists in soil naturally only in trace amounts following the spontaneous fission of ^{238}U. Measured values in the present study are therefore from the artificially produced fission of ^{238}U. Nuclear accidents, bombs test and nuclear weapons are the origins of ^{137}Cs contamination.

The weighted mean values of the activity concentrations of ^{238}U and ^{232}Th series, from their decay products along with statistical error are presented in Table 2.

The weighted mean of ^{232}Th varied from 0.8 to 7.6 Bq/kg for the studied soil samples which was significantly below the worldwide average value of 45 Bq/kg (UNSCEAR, 2000). The weighted mean of ^{238}U varied from 4.4 to 64.4 Bq/kg. The highest weighted mean value of ^{238}U was noted in sample 4. It may be attributed

Table 2. Weighted means values of ^{226}Ra, ^{214}Pb, and ^{214}Bi (^{238}U series) and ^{228}Ac, ^{212}Pb and ^{208}Tl (^{232}Th series) in the soil samples

Sample no.	^{238}U series (Bq/kg)	^{232}Th series (Bq/kg)
1	4.4 ± 0.1	0.8 ± 0.1
2	15.5 ± 0.3	5.3 ± 0.2
3	16.5 ± 0.3	7.6 ± 0.2
4	64.4 ± 0.6	2.9 ± 0.2
5	15.1 ± 0.3	6.0 ± 0.2

to the elevated level of ^{226} Ra in the sample as discussed earlier.

For comparison, Table 3 shows the activity concentration levels of ^{238}U, ^{232}Th and ^{40}K in different areas of the world as well as the results of this study.

Table 3 indicates that the average activity concentrations (Bq/kg) of ^{238}U, ^{232}Th and ^{40}K in soil samples in the present study are within or below the worldwide average values except for sample 4 which has higher value of average activity concentrations of ^{238}U.

Dosimetry aspects. The calculated absorbed dose rate, radium equivalent activity, the external hazard index and annual effective dose equivalent along with statistical errors are given in Table 4.

The obtained value of gamma dose rate (D) in the outdoor air at 1 m above the ground level ranged between 3.0 to 34.6 nGy/h. The values of the radium equivalent activity (Ra_{eq}) were found between 6.4 and 76.9 Bq/ kg. The values of radiation hazard index (H_{ex}) varied from 0.018 to 0.210. The variation in annual effective dose equivalent (AEDE) was found in the range of 3.6 to 42.4 μSv/y. The maximum permissible values of gamma dose rate, radium equivalent activity and annual effective dose equivalent are 51 nGy/h, 370 Bq/kg and 460 μSv/y, respectively (UNSCEAR, 2000; 1993). The external hazard index should be below unity (Ibrahim *et al.,* 2009).

Table 3. Worldwide values of activity concentrations in soil samples

Country	Activity concentration (Bq/kg)		
	^{238}U	^{232}Th	^{40}K
USA (UNSCEAR, 2000)	35	35	370
Pakistan(Lahore, Akhtar *et al.,* 2005)	25.8	49.2	561.6
Pakistan (Islamabad, Rahman *et al.,* 2009)	73.9	152.2	325.3
India (Rani and Singh, 2005)	26.4	51.2	5.6
India (Singh *et al.,* 2003)	44.2	174.5	93.1
Japan (UNSCEAR, 2000)	29	28	310
China (Yang et al., 2005)	38.5	54.6	584
Oman (Goddard, 2001)	29.7	15.9	225
Kuwait (Bou-Rabee, 1997)	13.3	10	370
Kuwait (north region, Saad, 2002)	66.6	11.3	384.5
Kuwait (south region, Saad, 2002)	13.6	2.4	110.4
Syria (UNSCEAR, 2000)	23.0	20	270
Jordan (Al-Hamarneh, 2009)	49	27	291
Egypt (UNSCEAR, 2000)	37	18	320
Iran (UNSCEAR, 2000)	28	22	640
Worldwide value (UNSCEAR, 2000)	33	45	420
Present study	23.2	4.5	127.1

Table 4. Dose rate (D) (nGy/h), radium equivalent activity (Ra_{eq}) (Bq/kg), external hazard index (H_{ex}) and annual effective dose equivalent (AEDE) (μSv/y) due to soil irradiation in Qatar

Sample no.	Dose rate (D)	Radium equivalent activity (Ra_{eq})	External hazard index (H_{ex})	Annual effective dose equivalent (AEDE)
1	3.0 ±0.5	6.4±1.0	0.018±0.003	3.6±0.6
2	17.9±1.1	35.7±2.2	0.100±0.006	21.9±1.4
3	19.8±1.1	39.8±2.3	0.111±0.006	24.3±1.4
4	34.6±2.8	76.9±6.2	0.210±0.017	42.4±3.4
5	16.6±1.1	33.7±2.1	0.094±0.006	20.4±1.3

The values obtained for the studied parameters are below the recommended safety limits.

Conclusion

The activity concentration of ^{238}U in soil sample no. 4 is 64.4 ± 0.6 Bq/kg which is higher than the worldwide average value, i.e. 33 Bq/kg and higher than the values determined for the rest of the samples. The enhanced level of ^{226}Ra in the sample is likely due to the occurrence of the nearby oil-fields. It may be due to the mobility of ^{226}Ra in the oil-field rocks (or due to the operating process at the oil-field).

The activity concentration of ^{40}K ranged between 13.6 and 179.5 Bq/kg which is less than the worldwide average value i.e. 420 Bq/kg. The lowest values of ^{40}K might be due to the solubility of potassium in the soil along the coastal regions.

Since ^{137}Cs does not exist naturally in soil, the values reported in this study are quit low and lower than those reported in other countries.

The activity concentration of ^{232}Th ranged between 0.8 and 7.6 Bq/kg which is much lower than the world-wide average value i.e. 45 Bq/kg.

The obtained values of gamma dose rate, radium equivalent activity, radiation hazard index and annual effective dose equivalent were found below the recommended safety limits.

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