

## Effect of the Grain Size of the Soil on the Measured Activity and Variation in Activity in Surface and Subsurface Soil Samples

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**Abstract.** Correlation between grain size and activity concentrations of soils and concentrations of various radionuclides in surface and subsurface soils has been measured for samples taken in the State of Qatar by gamma-spectroscopy using a high purity germanium detector. From the obtained gamma-ray spectra, the activity concentrations of the  $^{238}\text{U}$  ( $^{226}\text{Ra}$ ) and  $^{232}\text{Th}$  ( $^{228}\text{Ac}$ ) natural decay series, the long-lived naturally occurring radionuclide  $^{40}\text{K}$  and the fission product radionuclide  $^{137}\text{Cs}$  have been determined. Gamma dose rate, radium equivalent, radiation hazard index and annual effective dose rates have also been estimated from these data. In order to observe the effect of grain size on the radioactivity of soil, three grain sizes were used i.e., smaller than 0.5 mm; smaller than 1 mm and greater than 0.5 mm; and smaller than 2 mm and greater than 1 mm. The weighted activity concentrations of the  $^{238}\text{U}$  series nuclides in 0.5-2 mm grain size of sample numbers was found to vary from  $2.5 \pm 0.2$  to  $28.5 \pm 0.5$  Bq/kg, whereas, the weighted activity concentration of  $^{40}\text{K}$  varied from  $21 \pm 4$  to  $188 \pm 10$  Bq/kg. The weighted activity concentrations of  $^{238}\text{U}$  series and  $^{40}\text{K}$  have been found to be higher in the finest grain size. However, for the  $^{232}\text{Th}$  series, the activity concentrations in the 1-2 mm grain size of one sample were found to be higher than in the 0.5-1 mm grain size. In the study of surface and subsurface soil samples, the activity concentration levels of  $^{238}\text{U}$  series have been found to range from  $15.9 \pm 0.3$  to  $24.1 \pm 0.9$  Bq/kg, in the surface soil samples (0-5 cm) and  $14.5 \pm 0.3$  to  $23.6 \pm 0.5$  Bq/kg in the subsurface soil samples (5-25 cm). The activity concentrations of  $^{232}\text{Th}$  series have been found to lie in the range  $5.7 \pm 0.2$  to  $13.7 \pm 0.5$  Bq/kg, in the surface soil samples (0-5 cm) and  $4.1 \pm 0.2$  to  $15.6 \pm 0.3$  Bq/kg in the subsurface soil samples (5-25 cm). The activity concentrations of  $^{40}\text{K}$  were in the range  $150 \pm 8$  to  $290 \pm 17$  Bq/kg, in the surface soil samples (0-5 cm) and  $129 \pm 7$  to  $299 \pm 14$  Bq/kg, in the subsurface soil samples (5-25 cm). The activity concentrations of  $^{238}\text{U}$  series,  $^{232}\text{Th}$  series and  $^{40}\text{K}$  in the surface and deep soil samples are approximately same. The  $^{137}\text{Cs}$  activity concentration levels in surface soil samples in four sites were found to be higher than those observed for the soil samples that been collected at a depth of 5-25 cm. They ranged from  $1.65 \pm 0.22$  to  $19.0 \pm 0.9$  Bq/kg, in the surface soil samples (0-5 cm) and  $0.5 \pm 0.2$  to  $15.4 \pm 0.7$  Bq/kg, in the subsurface soil samples (5-25 cm).

**Keywords:** annual effective dose rate, gamma-spectroscopy, grain size, surface and subsurface soil, radiation hazard index, radium equivalent

### Introduction

Naturally occurring radioactivity can be found in certain industrial materials like ores and minerals (O'Brien, 2007). Radionuclides in naturally-occurring radioactive

material (NORM) include primordial radionuclides that are naturally present in the rocks and minerals of the earth's crust and cosmogenic radionuclides produced by interactions of cosmic nucleons with target atoms in the atmosphere and in the earth. The radionuclides of particular interest are  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{232}\text{Th}$  and their

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decay progeny, and the primordial radionuclide  $^{40}\text{K}$ . Man-made radionuclides  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are produced following nuclear fission in nuclear reactors or atomic weapons test fallout. Tremendous amounts of  $^{137}\text{Cs}$  were released into the atmosphere by open air nuclear bomb tests conducted during 1950s and 1960s and by the Chernobyl nuclear disaster occurred in 1986. Since the released  $^{137}\text{Cs}$  had spread out over the world and deposited on the ground, the most surface soils in the world generally have substantial inventory of  $^{137}\text{Cs}$  (Park *et al.*, 2013).

The elevated level of activity concentration in soil in some parts of the world may require taking action to protect humans living in dwellings built on these areas. The IAEA concluded that it is not necessary to apply regulatory control for activity concentrations below 1 Bq/g. If the activity concentrations are at this level or above, activity may still be exempted from regulatory controls if occupational exposure is found to be less than 1 mSv/y (IAEA, 2004). The average global effective dose rate to members of public from soil with weighted mean activity concentrations of 33 Bq/kg, 45 Bq/kg and 420 Bq/kg for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , respectively is 0.460 mSv/y (UNSCEAR, 2000).

External gamma dose estimation due to NORM is important because this may contribute significantly, to the total annual individual dose. The doses can vary considerably depending on the  $^{238}\text{U}$  and  $^{232}\text{Th}$  (and their daughter products) and  $^{40}\text{K}$  concentrations present in the local geology and overlying soils (Rafique *et al.*, 2011; Quindos *et al.*, 1994; Radhakrishna *et al.*, 1993).

Previous studies indicate that the radioactivity of the soil is related to the grain size i.e., soil radioactivity decreases with sand content in soil and increases with clay content. If the radium atoms are present only on the surface of the soil particles, the radium concentration is inversely proportional to the radius of the particles, whereas, if the radium is uniformly distributed throughout the soil grains, the radium concentration is independent of grain size (Kainan *et al.*, 2004).

Depth profile studies show that the  $^{137}\text{Cs}$  activity decreases with the increasing soil depth, and it is below detection limit for depths more than 25 cm (Karunakara *et al.*, 2001).

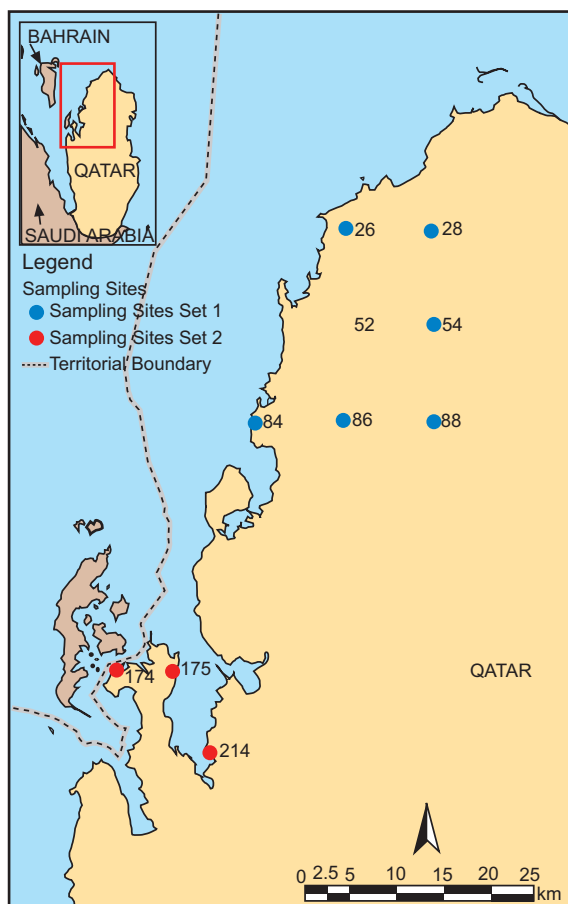
The objective of this study was to observe the effect of grain size on the radioactivity of soil and variation in radioactivity in surface and subsurface soil samples.

This will provide a baseline data for the radioactivity concentration in Qatari soil. Such data can be used to assess the biological effect of natural radiation in the environment and detect any significant future artificial release of radioactive radionuclides (Chen *et al.*, 2005). Such studies are also used to set national standards in the light of global recommendations. On the national level, this research will contribute to the establishment of Qatari national standards for the levels of natural radioactivity.

## Materials and Methods

**Sample collection and preparation.** Soil samples were collected from sampling points located at regular intervals of 1 per 5 km<sup>2</sup> square grid from the State of Qatar, using Qatar national grid (QNG) system. Preliminary reports on the determination of natural radioactivity levels in the State of Qatar and a report on the determination of the natural radioactivity levels in northwest of Dukhan (an area known for its oil fields) in the State of Qatar, have been earlier (Al-Sulaiti *et al.*, 2012; 2011; 2010; Nasir *et al.*, 2012). The State of Qatar is a peninsula with a total area of 11,437 km<sup>2</sup>, which lies over a geological formation comprising a sequence of limestone, chalk, clay and gypsum (Al-Sulaiti, 2011; MOFA, 2007).

In earlier study (Al-Sulaiti *et al.*, 2010), one sample among the six showed mean activity concentration of  $^{238}\text{U}$  as 213.9±1.4 Bq/kg, which is significantly, higher than the world average value of 33 Bq/kg. This sample was collected from an in shore oil field in the northwest of Qatar. Therefore, for the present study three more samples were collected from this area to investigate the correlation between the activity concentration and the grain size (Fig. 1, set 2). Moreover, the soil samples discussed in the preliminary report were collected from depths between 5 and 15 cm, and the activity concentration of the artificial man-made radionuclide,  $^{137}\text{Cs}$  in one sample has found to be 20.42±0.50 Bq/kg. Therefore, top soil samples (0-5 cm) and deeper soil samples (5-25 cm) were collected from 7 sampling points from the northwest of Qatar (Fig. 1, set 1) in order to investigate the correlation between the distribution of the radionuclides  $^{226}\text{Ra}$ ,  $^{228}\text{Ac}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in surface and sub surface soil samples. Approximately, 1.5-2 kg of soil was collected at each sample point. Each sample was then transferred to a 2 mm sieve fitted in a collecting pan. The sieved samples were then filled into labelled polyethylene bags, sealed, packed in a box and shipped



**Fig. 1.** The locations of the soil samples on the map of the State of Qatar. Samples of set 1 were used to study the distribution of radionuclide in surface and subsurface soil and samples of set 2, were used to study the effect of grain size on activity concentration of soil. This map was created using ArcView 9 software (ESRI, 2005).

to the UK for analysis. The final sample preparation and all the gamma-ray measurements were performed in the low-background radiation detection laboratories of the Centre for Nuclear and Radiation Physics, situated within the Department of Physics at the University of Surrey.

The samples were dried in oven at a temperature of 60 °C for 24 h to remove the moisture. In order to observe the variation in activity in surface and sub surface soil samples, a 500 µm mesh was used to sieve the samples, which were then weighed and transferred to 500 mL labelled Marinelli beakers. Similarly, to observe the correlation between the activity concentration

and the grain size, 3 soil samples collected from different locations were segregated into three grain sizes. The grain sizes used were: (i) smaller than 0.5 mm; (ii) smaller than 1 mm and greater than 0.5 mm; and (iii) smaller than 2 mm and greater than 1 mm. The samples were sealed and stored for about one month in order to reach radioactive secular equilibrium ( $> 7$  half-lives of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$ ).

**Gamma-ray spectrometry.** The soil samples were analysed using a high-resolution, low-background gamma-ray spectrometry system based on a p-type coaxial hyper-pure germanium detector (HPGe). A crystal of 50 mm diameter was operated under a high voltage, bias of +3000 V (DC). Low background 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).

The performance of HPGe detector used was characterised by the energy calibration and its absolute full energy peak detection efficiency. Further details have been reported in other papers published by our collaboration (Al-Sulaiti *et al.*, 2012; 2011; 2010; Nasir *et al.*, 2012).

The samples were placed directly over the front face of the detector. The acquisition time for each sample was 1 day (86, 400 sec). The counting geometry of the samples and the standard sources used for efficiency calibration were kept constant. A variety of different gamma-ray energy transitions ranging from ~100 keV to 2.614 MeV, associated with the decay products of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series were analysed independently. The data were analysed under the assumption of secular equilibrium of the radionuclides within these samples. Activity concentrations of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  were used to determine the activity concentration of  $^{238}\text{U}$ . The activity concentration of  $^{232}\text{Th}$  was determined using gamma-ray transitions of  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$ . Gamma-ray peaks of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  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 standard equation:

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

where:

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

The gamma dose rate (D) in the outdoor air at 1 m above ground level was calculated using the following equation (Al-Ghorabie, 2005; Singh *et al.*, 2005):

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

where:

$A_x$  (Bq/kg) is the mean activity of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  or  $^{40}\text{K}$ , 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  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  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).

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

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

The permissible limit of  $\text{Ra}_{eq}$  is 370 Bq/kg in soil samples that contain  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  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 Mathew, 1985):

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

where:

$A_U$ ,  $A_{Th}$  and  $A_K$  are the specific activities of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , 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):

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

The world average annual effective dose equivalent (AEDE) from outdoor terrestrial gamma radiation is 460  $\mu\text{Sv/y}$  (UNSCEAR, 2000).

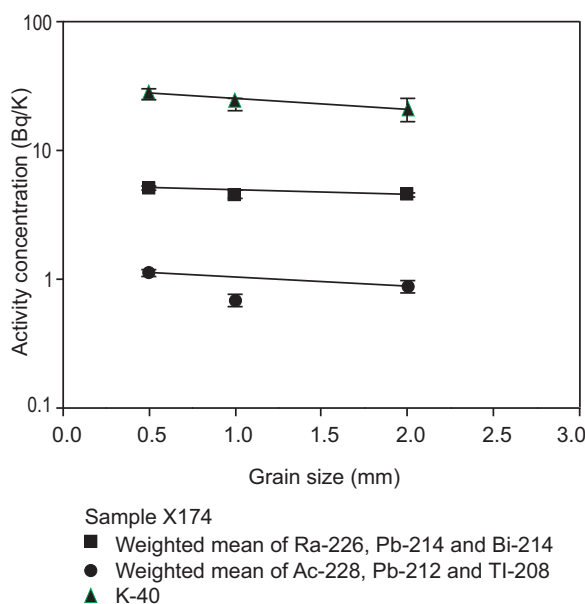
## Results and Discussion

### Effect of the grain size of the soil on the radioactivity.

The activity concentrations of soil samples can be influenced by the grain size of the samples (Hebinck *et al.*, 2007). The activity concentrations of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$  (from the  $^{238}\text{U}$  series),  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  (from the  $^{232}\text{Th}$  series),  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , along with statistical errors, in the soil samples investigated in the present study are presented in Table 1.

In Fig. 2, 3 and 4 the activity concentrations of  $^{238}\text{U}$  series  $^{232}\text{Th}$  series and  $^{40}\text{K}$  all appear to be correlated with the grain size for samples X174, X175 and 214, respectively. For all the three samples higher activities were noted for smaller grain sizes.

The weighted mean of the activity concentration of  $^{238}\text{U}$  series and  $^{232}\text{Th}$  series in the 0.5 mm grain size of sample 214 were found to be about  $28.5 \pm 0.5$  and  $8.2 \pm 0.2$  Bq/kg, respectively, higher than the values observed for 1 mm and 2 mm grain size. For the 1 mm and 2 mm grain sizes, the activity concentrations for the  $^{238}\text{U}$  series were  $19.2 \pm 0.4$  and  $16.9 \pm 0.4$  Bq/kg, while, the activity concentration of  $^{232}\text{Th}$  series were  $6.4 \pm 0.2$  and  $5.6 \pm 0.2$  Bq/kg, respectively. The activity concentrations of  $^{40}\text{K}$  were  $188 \pm 10$ ,  $154 \pm 9$  and  $139 \pm 8$  Bq/kg for grain sizes 0.5, 1 and 2 mm, respectively. Sample 214 comes from soil

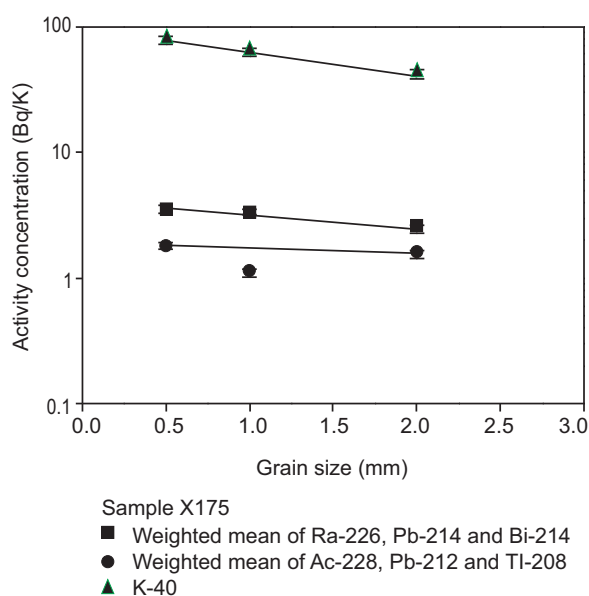
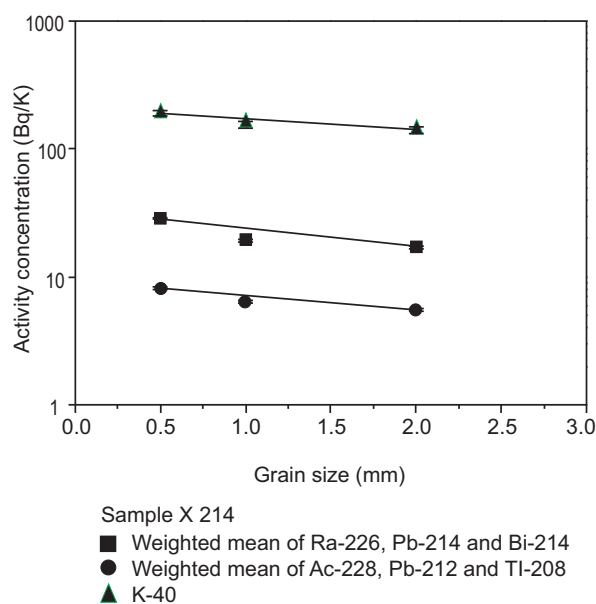


**Fig. 2.** The relationship between the weighted mean of the activity concentrations of  $^{238}\text{U}$  series  $^{232}\text{Th}$  series and  $^{40}\text{K}$  and the three different grain size for sample X174.

**Table 1.** Activity concentrations (Bq/kg) of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  ( $^{238}\text{U}$  series),  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  ( $^{232}\text{Th}$  series),  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , in the soil samples for different grain sizes

Samples	$^{238}\text{U}$ series	$^{232}\text{Th}$ series	$^{40}\text{K}$	$^{137}\text{Cs}$
	WM of $^{226}\text{Ra}$ , $^{214}\text{Pb}$ and $^{214}\text{Bi}$	WM of $^{228}\text{Ac}$ , $^{212}\text{Pb}$ and $^{208}\text{Tl}$		
Sample-X174 mesh<0.5mm	5.08±0.18	1.12±0.07	27.2±2.8	*
Sample-X174 1mm>mesh>0.5mm	4.61±0.34	0.69±0.08	22.9±2.5	*
Sample-X174 2mm>mesh>1mm	4.54±0.18	0.88±0.09	20.9±4.1	*
Sample-X175 mesh<0.5mm	3.53±0.22	1.82±0.11	77.4±5.9	*
Sample-X175 1mm>mesh>0.5mm	3.35±0.18	1.10±0.09	62.8±4.7	0.23±0.18
Sample-X175 2mm>mesh>1mm	2.46±0.17	1.55±0.12	41.9±3.3	*
Sample-214 mesh<0.5mm	28.54±0.45	8.20±0.21	188±10	0.69±0.28
Sample-214 1mm>mesh>0.5mm	19.22±0.40	6.41±0.18	154.0±8.5	*
Sample-214 2mm>mesh>1mm	16.91±0.36	5.55±0.18	139.2±8.0	0.36±0.17

\* = below minimum detectable activity MDA.

**Fig. 3.** The relationship between the weighted mean of the activity concentrations of  $^{238}\text{U}$  series  $^{232}\text{Th}$  series and  $^{40}\text{K}$  and the three different grain size for sample X175.**Fig. 4.** The relationship between the weighted mean of the activity concentrations of  $^{238}\text{U}$  series  $^{232}\text{Th}$  series and  $^{40}\text{K}$  and the three different grain size for sample 214.



overlying the dammad formation, from the middle Eocene period, which consists of shale, limestone and dolomite. It can be concluded that the finest grain size had the highest activity concentration (for sample 214 at least). The other two samples (X174 and X175) showed similar behaviour for the  $^{238}\text{U}$  series and  $^{40}\text{K}$ . However, for the  $^{232}\text{Th}$  series, the activity concentrations in the 1-2 mm grain size were found to be higher than in the 0.5-1 mm grain size. This might be related to the soil type as both of them were collected from the same geological background. The underlying geology in the area, where those samples were collected belongs to sabkha deposits which consist of sand and silt flats.

The calculated absorbed dose rate, radium equivalent activity, the external hazard index and annual effective dose equivalent along with statistical errors from soil samples having different grain sizes are given in Table 2.

In Table 2, the calculated values of absorbed dose rate, radium equivalent activity, the external hazard index and annual effective dose equivalent were found to be

higher for smaller grain sizes except for sample X174, where the absorbed dose rate, radium equivalent activity and annual effective dose equivalent were slightly higher for grain sizes <2 mm and >1 mm, compared to grain size <1 mm and >0.5 mm. This might be due to the relatively low content of  $^{232}\text{Th}$ , in this mesh size comparing with greatest mesh (2 mm > mesh > 1 mm).

**Variation in activity in surface and sub surface soil samples.** A comparison between the surface 0-5 cm soil samples and soil samples taken at depth of 5-25 cm was conducted in order to investigate the correlation between the distribution of the radionuclides  $^{226}\text{Ra}$ ,  $^{228}\text{Ac}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  and layer depth. The activity concentrations of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  (from the  $^{238}\text{U}$  series),  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  (from the  $^{232}\text{Th}$  series),  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , along with statistical errors in surface and sub surface soil samples are presented in Table 3.

Figure 5 shows the weighted mean of the activity concentration for the individual activities as deduced from the observed gamma-ray transitions associated with the direct decays of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  (from

**Table 2.** Dose rate (D) (nGy/h), radium equivalent activity ( $R_{\text{eq}}$ ) (Bq/kg), external hazard index ( $H_{\text{ex}}$ ) and annual effective dose equivalent (AEDE) ( $\mu\text{Sv/y}$ ) from soil samples for different grain sizes

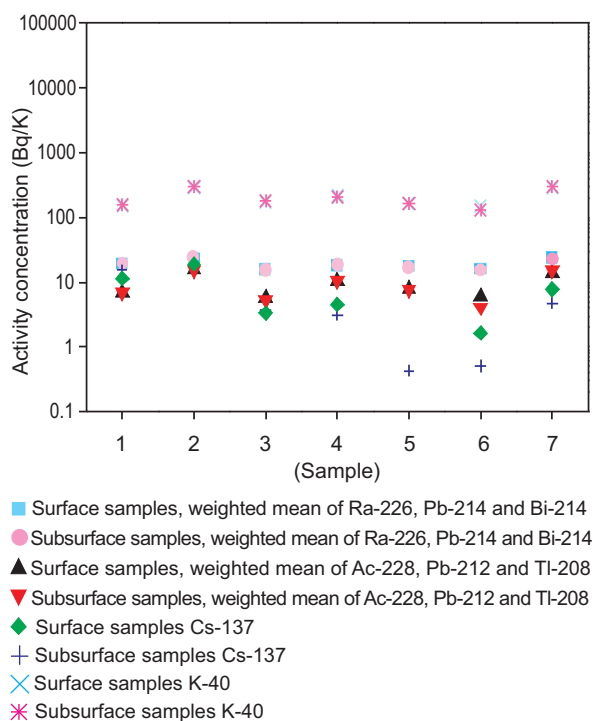
Samples	Dose rate (D)	Radium equivalent activity ( $R_{\text{eq}}$ )	External hazard index ( $H_{\text{ex}}$ )	Annual effective dose equivalent (AEDE)
Sample-X174 mesh<0.5mm	4.08±0.52	8.6±1.1	0.024±0.003	5.01±0.64
Sample-X174 1mm>mesh>0.5mm	3.41±0.62	7.2±1.3	0.020±0.004	4.19±0.76
Sample-X174 2mm>mesh>1mm	3.43±0.77	7.3±1.6	0.020±0.004	4.20±0.94
Sample-X175 mesh<0.5mm	6.06±0.70	11.6±1.3	0.033±0.004	7.43±0.85
Sample-X175 1mm>mesh>0.5mm	4.87±0.60	9.3±1.2	0.026±0.003	5.98±0.74
Sample-X175 2mm>mesh>1mm	3.89±0.51	7.6±1.0	0.021±0.003	4.77±0.62
Sample-214 mesh<0.5mm	25.8±1.6	53.4±3.3	0.148±0.009	31.57±1.9
Sample-214 1mm>mesh>0.5mm	19.1±1.3	39.2±2.6	0.109±0.007	23.4±1.5
Sample-214 2mm>mesh>1mm	16.9±1.2	34.6±2.4	0.096±0.007	20.7±1.4

**Table 3.** Activity concentrations (Bq/kg) of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  ( $^{238}\text{U}$  series),  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  ( $^{232}\text{Th}$  series),  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , in the surface and subsurface soil samples

Samples	$^{238}\text{U}$ series	$^{232}\text{Th}$ series	$^{137}\text{Cs}$	$^{40}\text{K}$
	WM of $^{226}\text{Ra}$ , $^{214}\text{Pb}$ and $^{214}\text{Bi}$	WM of $^{228}\text{Ac}$ , $^{212}\text{Pb}$ and $^{208}\text{Tl}$		
S-26 (Surface)	19.62±0.34	6.85±0.19	11.57±0.54	149.6±7.8
S-28 (Surface)	22.63±0.44	15.69±0.18	19.03±0.89	290±15
S-52 (Surface)	15.93±0.30	5.65±0.18	3.35±0.23	174.4±8.9
S-54 (Surface)	18.09±0.37	10.30±0.35	4.56±0.31	208±10
S-84 (Surface)	17.73±0.34	8.03±0.24	*	166.3±7.9
S-86 (Surface)	16.06±0.31	6.08±0.16	1.65±0.22	150.3±7.8
S-88 (Surface)	24.11±0.91	13.73±0.54	7.80±0.84	290±17
D-26 (Subsurface)	19.98±0.34	7.02±0.20	15.41±0.67	154.9±8.2
D-28 (Subsurface)	23.63±0.50	15.60±0.30	14.58±0.62	297±14
D-52 (Subsurface)	15.48±0.29	5.33±0.17	3.71±0.29	179.5±9.1
D-54 (Subsurface)	18.63±0.43	10.53±0.22	3.10±0.36	204±10
D-84 (Subsurface)	16.79±0.32	7.66±0.19	0.43±0.19	164.0±8.5
D-86 (Subsurface)	14.51±0.29	4.08±0.15	0.51±0.16	128.5±6.8
D-88 (Subsurface)	22.89±0.43	15.16±0.29	4.76±0.37	299±14

\* = below minimum detectable activity MDA.

the  $^{238}\text{U}$  series),  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  (from the  $^{232}\text{Th}$  series) and the activity concentrations for  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the surface and deep soil. The activity concentration

**Fig. 5.** Activity concentrations levels of  $^{226}\text{Ra}$ ,  $^{228}\text{Ac}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  for topsoil samples 0-5 and soil samples taken at depths of 5-25 cm.

levels of  $^{238}\text{U}$  decay products,  $^{232}\text{Th}$  decay products and  $^{40}\text{K}$  in the surface and deep soil samples were similar. Activity concentrations of  $^{137}\text{Cs}$  in surface soil samples in four sites were found to be higher than those observed for the soil samples that had been collected at a depth of 5-25 cm. This is possibly because  $^{137}\text{Cs}$  accumulates in the top soil layers and dilutes in the deeper layers (Kim *et al.*, 1997).

$^{137}\text{Cs}$  exists in soil naturally only in trace amounts following the spontaneous fission of  $^{238}\text{U}$ . Measured values in the current work are therefore due to the artificially produced fission of  $^{238}\text{U}$ . Nuclear accidents, bombs test and nuclear weapons are the origins of  $^{137}\text{Cs}$  contamination (Nasir *et al.*, 2012).

According to IAEA, the amount of  $^{137}\text{Cs}$  released during the Chernobyl accident on 26<sup>th</sup> April 1986, was  $0.09 \times 10^{18}$  Bq (IAEA, 1996). This tremendous amount of radioactive material was deposited over the entire northern hemisphere including the Middle East region. The half-life of  $^{137}\text{Cs}$  is long enough ( $\approx 30$  years) to be traced for several tens of years.

In the current study, it is expected that this contamination is mainly due to Chernobyl accident because the samples were collected before Fukushima accident in April 2011. This conclusion can not be confirmed because of the lack of data available in the area under study, before 1986.

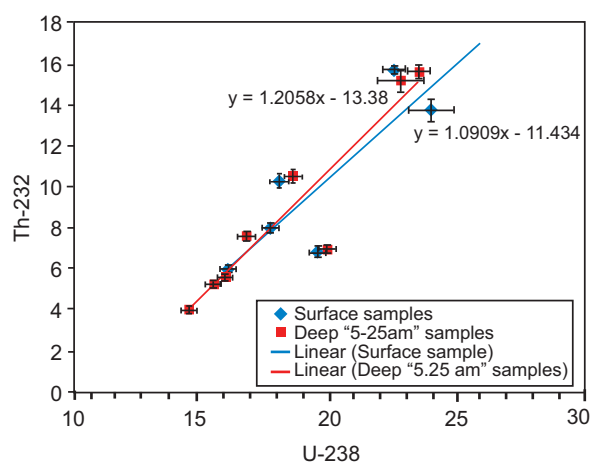
**Table 4.** Dose rate (D) (nGy/h), radium equivalent activity ( $R_{eq}$ ) (Bq/kg), external hazard index ( $H_{ex}$ ) and annual effective dose equivalent ( $\mu$ Sv/y) for surface and subsurface soil samples

Samples	Dose rate (D)	Radium equivalent activity ( $R_{eq}$ )	External hazard index ( $H_{ex}$ )	Annual effective dose equivalent (AEDE)
S-26 (Surface)	19.4±1.2	39.9±2.5	0.111±0.007	23.8±1.5
S-28 (Surface)	32.5±1.8	65.3±3.6	0.182±0.010	39.9±2.2
S-52 (Surface)	18.1±1.1	36.2±2.3	0.101±0.006	22.2±1.4
S-54 (Surface)	23.5±1.5	47.4±3.0	0.132±0.008	28.9±1.8
S-84 (Surface)	20.1±1.2	40.9±2.4	0.114±0.007	24.6±1.5
S-86 (Surface)	17.4±1.1	35.3±2.2	0.098±0.006	21.3±1.3
S-88 (Surface)	31.9±2.6	64.1±5.1	0.179±0.014	39.2±3.1
D-26 (Subsurface)	19.9±1.2	40.9±2.5	0.113±0.007	24.4±1.5
D-28 (Subsurface)	33.3±1.9	66.7±3.7	0.186±0.010	40.8±2.3
D-52 (Subsurface)	17.9±1.1	35.7±2.2	0.100±0.006	21.9±1.4
D-54 (Subsurface)	23.7±1.4	48.0±2.9	0.133±0.008	29.1±1.7
D-84 (Subsurface)	19.3±1.2	39.2±2.4	0.109±0.007	23.7±1.4
D-86 (Subsurface)	14.5±1.0	29.3±2.0	0.082±0.005	17.7±1.2
D-88 (Subsurface)	32.7±1.8	65.5±3.6	0.183±0.010	40.1±2.2

Figure 6 shows the ratio of  $^{232}\text{Th}$  to  $^{238}\text{U}$  in topsoil and deeper samples. The slope of each trend line in this plot was used to deduce the activity ratio of  $^{232}\text{Th}$  to  $^{238}\text{U}$  (assuming radioactive equilibrium). This was found to be slightly higher in the deeper soil samples than the topsoil samples. The fitted line slopes were  $1.20\pm 0.21$  and  $1.09\pm 0.26$ , respectively. This indicates that the activity concentrations of  $^{226}\text{Ra}$ , which was used to estimate the levels of  $^{238}\text{U}$  may be (slightly) affected by the dissolution reactions in the topsoil samples. Since compounds of  $^{226}\text{Ra}$  are soluble in water, radium nuclides are mobile and may be introduced along with any water, precipitate and leached out due to chemical pressure and temperature changes (Wilson, 1994).

The calculated absorbed dose rate, radium equivalent activity, the external hazard index and annual effective dose equivalent along with statistical errors for surface and subsurface soil samples are given in Table 4.

Table 4 indicates the calculated values of absorbed dose rate, radium equivalent activity, the external hazard index and annual effective dose equivalent were found to be approximately same for all the investigated surface and sub surface samples. This might be expected since these values depend on the activity concentration levels  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series and  $^{40}\text{K}$  in the soil and these were found to be almost the same in surface and subsurface soil, except for  $^{137}\text{Cs}$ , which is expected to be higher in the surface soil.

**Fig. 6.** The weighted mean of activity concentrations levels for  $^{232}\text{Th}$ , versus the weighted mean of activity concentration for  $^{238}\text{U}$  for the 7 investigated samples.

## Conclusion

It can be concluded from the weighted mean of the activity concentration of  $^{226}\text{Ra}$ ,  $^{228}\text{Ac}$  and  $^{40}\text{K}$  in soil samples having different grain sizes that the finest grain size has the highest activity concentration. However, for  $^{232}\text{Th}$  series, the activity concentrations in 2 mm grain size of two samples (X175 and 214) were found to be higher than for the 0.5-1 mm grain size.

The calculated values of absorbed dose rate, radium equivalent activity, the external hazard index and annual



effective dose equivalent have been found higher for smaller grain sizes except for one sample (X174) where absorbed dose rate, radium equivalent activity and annual effective dose equivalent were calculated to be slightly higher for grain sizes <2 mm and >1 mm as compared to grain sizes <1 mm and >0.5 mm.

The activity concentration levels of  $^{238}\text{U}$  series,  $^{232}\text{Th}$  series and  $^{40}\text{K}$  in the surface and deep soil samples are approximately same. The activity concentration levels of  $^{137}\text{Cs}$  in surface soil samples in four sites have been found to be higher than those observed for the soil samples that had been collected at a depth of 5-25 cm. As only 7 soil samples were analysed in the current work, it is suggested that a further study to be done on the radioactivity concentration of  $^{137}\text{Cs}$  in the Qatari topsoil, particularly after Fukushima accident in April 2011, in order to obtain more statistically significant picture of the radiological map of the Qatari peninsula.

The calculated values of absorbed dose rate, radium equivalent activity, the external hazard index and annual effective dose equivalent have been found approximately same for all the investigated surface and sub surface samples except for sample 86, which has slightly lower values for sub surface soil.

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