Vertical distribution and radiological risk assessment of $^{137}$Cs and natural radionuclides in soil samples

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The aims of this study were to investigate the vertical distributions of natural radionuclides $^{232}$Th, $^{226}$Ra and $^{40}$K as well as anthropogenic radionuclide $^{137}$Cs in soil samples and to analyze the correlation among the radioactivity of these radionuclides and the physiochemical characteristics of soil samples namely pH, grain size, carbonate content and organic matter. Risk assessment of the radiological hazard has also been estimated. Forty-four soil samples were collected from eleven locations in Qatar at four depth levels from 0 to 16 cm. The average concentrations of $^{232}$Th, $^{226}$Ra, $^{40}$K and $^{137}$Cs in the soil depth of 16 cm were 10, 17, 201 and 4 Bq/kg, respectively, which were within the reported world mean. The external absorbed gamma dose rate, the annual effective dose, the mean radium equivalent activity, the external hazard index and the lifetime cancer risk were 22 nGy/h, 0.027 mSv/y, 47 Bq/kg, 0.125 and $0.096 \times 10^{-3}$, respectively. These values were far below the minimum recommended international values. The level of radioactivity concentrations in the soil was affected by the physiochemical characteristics of the soil. The positive correlation with highest R$^2$ value was found among the radioactivity concentrations of $^{232}$Th and $^{40}$K and the soil clay content. Total organic carbon was also positively correlated for $^{226}$Ra and $^{137}$Cs activity concentrations, whereas, carbonate content was negatively correlated with the radioactivity concentrations of $^{232}$Th and $^{40}$K. As far as soil moisture content is concerned, the positive correlation with highest R$^2$ value was obtained for $^{226}$Ra activity concentrations.

Radioactivity has become an issue of major concern over the years due to its association with human health$^{1,2}$. Natural and artificial radioactive isotopes are host in the environment. Radionuclides, with different biogeochemical processes and important movability, can influence the environment through bioaccumulation and are hazardous for the environment and human health. The radioactive isotopes in the environment cause the external radiation dose to human organisms, while the isotopes integrated by inhalation and ingestion are the origin of the internal radiation dose. Studies about dose-effect relationships of radioactive materials have helped to increase knowledge about the risks associated with radiations and have played an important role in developing radiation protection regulations.

Characteristic of soil is instantaneously concerned to the essence of life$^3$. The risk from Cs-137 varies with its diffusion rates in soil. If Cs-137 migrate slowly in soil, the internal irradiation will be higher due to higher absorption by plants roots especially from the top surface of 5 cm depth. However, if Cs-137 diffuse rapidly, the external radiation will be less as in this case, the uppermost soil surface acts as a shield against radioactivity found in deeper soil layers$^4$. Therefore, assessment of the diffusion of natural and artificial radionuclides in soil is important to protect human and environment$^5$.

Naturally Occurring Radioactive Materials (NORM) refers to the natural origins of radiations i.e. from naturally occurring radioisotopes$^6$. Natural radioactivity can result from cosmogenic radioactive isotopes which are continuously produced by the effect of cosmic radiation (e.g., $^{14}$C, $^3$H, $^{10}$Be, $^{22}$Na, $^{26}$Al, $^{32,33}$P, $^{35}$S, $^{38}$Cl, and...
39Ar) and terrestrial radioactive isotopes7. Terrestrial radionuclides contain nucleogenesis or primordial natural radioactive isotopes. This can be classified into: natural radioactive decay series (238U, 235U, and 232Th) and the daughter nuclides with relatively long half-lives and the daughter elements of these daughter nuclides, for example, 228Ra, 210Po, 210Bi, and 210Pb. The second group is the long-life nuclei, which become stable daughter nuclides with relatively long half-lives and the daughter elements of these daughter nuclides, for example, 40K and 137Cs.

In the natural state of NORM causes technologically enhanced naturally occurring radioactive material (TENORM), or enhanced levels of NORM8. The anthropogenic activities such as coal mining, oil and gas extraction, geothermal energy production, water and wastewater treatment, application of phosphate fertilizers, uranium, thorium and copper mining can increase the level of naturally occurring radioactive particles9.

Artificial concentrations are the ones that results from human activities10. There are over 1300 artificially produced radionuclides. Due to 137Cs comparatively long half-life (30.17 years), considerable amounts of 137Cs are present in soil11. The main sources of Cs-137 in the environment worldwide are from nuclear atmospheric testing and regionally from the releases from the Chernobyl accident in 1986 and the Fukushima accident in 2011. The disaster in Chernobyl nuclear power station in Ukraine (on 26 of April 1986) released huge mass of radioactive materials. This accident has released up to 3.8 × 1016 Bq of 137Cs to the environment.

In Qatar, the radiological risk may arise from accidents where the sealed radiation sources are destroyed or lost and from unmanageable radioactive waste from intensive oil industry activities in oil drilling and exploitation. On the other hand, the anthropogenic radionuclide may arise from previous weapon test, nuclear accident, Gulf war and/or from progress development of nuclear industry in neighboring countries. Evaluation of radioactivity levels of several soil samples for the State of Qatar intends to aid as a reference for future radiological investigations, bioavailability assessment and diversion future dose rates to save the human health and environment, emergency action management, and outline the regulatory control standards and recommendations.

The vertical distribution of 137Cs is tightly related to organic matter distribution. The contents of silt and clay are the second distribution factor of 137Cs9. The key factor in the fixation process is the low hydration energy of the Cs ion. The adsorption properties of organic matter and content of silt and clay minerals give the soil particles high cationic exchange capacity. 137Cs, which is highly reactive, will perform as a cation like K+ or NH4+ as well as other competitive minerals like Na+, Ca2+ or Mg2+12. Thus, the vertical migration of 137Cs can be quicker in places with large coarse soil particles holding lower cation exchange capacity than in fine soil particles13. Furthermore, the soil pH also plays a major role in determining the transport of 137Cs within soil profile. At low pH value, more 137Cs is detected. Physical soil actions caused by runoff can also affect the final distribution of 137Cs14,15.

Therefore, the aims of this study were to: (i) determine the vertical distribution of 137Cs and natural radionuclides within a 16-cm soil depth in order to quantify spatial radioactivity variations; (ii) measure the activity concentrations of some radionuclides associated of the uranium–radium (238U, 228Ra), thorium–actinium (232Th, 228Ac) decay series, and of primordial radionuclide 40K as natural sources of radioactivity, and the activity concentrations of 137Cs as anthropogenic source of radioactivity; (iii) measure the physicochemical characteristics of the collected soils namely pH, grain size, moisture content, carbonate content and total organic carbon; (iv) assess radiological risk and concentrations; and (v) statistically analyze the correlation between physicochemical characteristic of soil namely: pH, moisture content, grain size, carbonate content and total organic carbon, with the vertical distribution of radionuclides in the soil.

Materials and Methods

Description of the study region. The State of Qatar has a total area of 11,437 km2. The geological structure of Qatar consists of a sequence of limestone, chalk, clay and gypsum16. Qatar topography is a rocky desert land with spread oases shaped by 850 scatter depressions. Colluvium soils, which made up of sandy clay loam, calcareous loam and sandy loam have piled up in these depressions to depths leveling from 30 to 150 cm, mainly in northern and eastern areas, covering limestone bedrock. These depression soils, that are locally named as Rauda (means garden), were preferred in the current study for the soil sampling sites due to accumulation of washout surface soil by rainfall. Rauda soil is buildup of rather bulky sediments in shallow depressions that break the topography as the result of surface water erosional from short-living channels after heavy rain17. The average yearly rainfall in Qatar is about 100 mm per year, mostly during autumn and winter seasons and more rainfall takes place in northern and eastern areas as compare to southern areas. Therefore, the sampling sites in current study were selected mainly from northern areas of Qatar. Summer season, which start from June to October, are described by severe heat, dryness, quite changeable humidity and unusual strong wind and sandstorms18.

Sample locations. The sampling was mainly for uncultivated soils within the specified region in Qatar. The inclination of the selected regions towards the North of Qatar was due to its topography. The North of Qatar has lower altitude with scattered depressions called locally Rauda; that allows the rain to accumulate in these areas. In addition, the annual rainfall in northern of Qatar is higher than the southern, which enhanced the wet deposition in those areas. The dry deposition is also enhanced by the lower average temperature in northern areas of Qatar, whereas, in southern areas, higher temperature at the soil surface resists the dry deposition. The sampling regions were: Al-Shamal, Al-Zubara, Al-Areesh, Fwaret, Ras Lafan, Al-Guwariah, Al-Khawr, Al-Jamaliayah, Dukhan, Al-Shahaniyah and Um Al-Amad.

Soil sample collection. Forty-four samples were collected. The samples were dug and scraped using shovel and scoop by a corer 30 cm × 30 cm. The soil was removed layer by layer at approximately depth intervals of 5 cm with a total of four different depth levels (0–1), (1–6), (6–11), (11–16) cm in each location. Accumulatively, a depth of 16 cm was used for the sampling process. As the existence of foreign materials are irrelevant for the soil...
samples and may give an error in the analytical results, glass pieces, twigs, stones, or leaves were removed from the soil samples. It should also be noted that the soil was undisturbed. Each layer sample was then packed into tagged polyethylene bags and sealed. The information of each sample was documented with a waterproof marker pen on each sample bag. The labels included soil information like: the region name, geographic coordinate, date, depth and ID (identified number). Three-digit serial numbers were used for numbering the collected soil samples (for example, 01.1 means (01) digit identified location Fwaret then (0.1) digit for soil of depth level 0–1 cm). The sampling tools were always cleaned during sampling to prevent soil-to-soil contamination.

The soil samples were then mixed and homogenized well and then divided into two portions for further analysis: for the radioactivity measurements with a total of 88 samples (including replication) and for the physiochemical characteristics with a total of 44 samples. The physiochemical characteristics include pH, moisture content, grain size, carbonate content and total organic carbon. Each bag was labeled with date, sample ID, location and coordinate.

Radioactivity concentration measurements. The samples were dried in an oven at 60 °C for 24 hours in order to assure that any moisture was eliminated from the samples. To gain uniform particle sizes, a 2 mm mesh was utilized to sieve the samples. The sample was then weighed and delivered to 250 ml tagged Marinelli beakers to remove the air by tapping Marinelli beakers over a surface, then was sealed in the airtight Marinelli beakers. The samples were manually homogenized during the sieving process using the sieving pans, and then by using a stainless-steel spoon to press the soil (homogenization is blending of a soil sample to grant uniform diffusion of contaminants). Incomplete homogenization would increase the sampling error (IAEA- 2004, 2016). Marinelli beakers were then stored and kept sealed for about one month (>7 half-lives of 222Rn) to attain radioactive secular equilibrium between 222Rn and its daughters.

Radioactivity concentrations of the soil samples were measured with a high purity Germanium (HPGe) gamma-ray spectrometry system. The current system was prepared with a coaxial HPGe detector. It has a relative efficiency of 40%, an energy resolution of 1.85 keV at 1332.5 keV of 57Co and of 0.87 keV at 122 keV of 57Co, a peak-to-Compton ratio of 62:1 and operating bias voltage 4000 Vdc. After subtracting the background radionuclides contributed to the combined uncertainty of the efficiency calibration. The uncertainties were estimated utilizing the geometry composer software, a geometry file was made for 250 mL Marinelli beaker and consequently a calibration efficiency curve was created and saved. Genie 2000 Spectroscopy Software version 3.2 allows marking the efficiency calibration using the spectrum simulation. The properties of the detector include software which permits applying mathematical modeling for the efficiency calculation curves attributed to soil weight. Utilizing the geometry composer software, a geometry file was made for 250 mL Marinelli beaker and consequently a calibration efficiency curve was created and saved.

Table 1. The activity concentration of radionuclides analysis from gamma ray energies of their progenies.

| Radionuclide | Half-life (yr.) | Gamma ray energy (keV) | Progeny radionuclide |
|--------------|----------------|------------------------|----------------------|
| $^{226}$Ra | 1650 | 295.21 | $^{219}$Po |
| $^{232}$Th | 1.405 x 10$^{10}$ | 338.32 | $^{232}$Ac |
| $^{238}$U | 1.405 x 10$^{10}$ | 609.31 | $^{234}$Pa |
| $^{238}$U | 1.405 x 10$^{10}$ | 351.92 | $^{234}$Pa |
| $^{40}$K | 1.278 | 1.09 | $^{40}$Ar |
| $^{137}$Cs | 30.1 | 1460.81 | $^{137}$Ba |
| $^{137}$Cs | 30.1 | 661.66 | $^{137}$Ba |

The statistical uncertainty contributed with the total counts in the peaks, whereas the systematic uncertainty is contributed to the combined uncertainty of the efficiency calibration. The uncertainties were estimated utilizing.
error propagation, accounting for relative standard uncertainties of the sample weight, net peak area, full energy peak efficiency, half-life of the radionuclide and emission probability\textsuperscript{14}.

The software used for analysis and reduction of the gamma-ray spectra was Genie 2000 Spectroscopy Software. The radioactivity concentration of the radionuclide established in the soil samples was measured by utilizing Eq. (1) and represented in unit of Bq/kg:

\[
\text{Radioactivity} \equiv \frac{C_{\text{net}}}{\gamma} \times \varepsilon (E_{\gamma}) \times m
\]

where \(C_{\text{net}}\) is the net peak count rate, \(\gamma\) is the absolute gamma decay intensity for the specific energy photopeak, \(\varepsilon(E_{\gamma})\) the absolute photopeak efficiency of the germanium detector at this energy and \(m\) is the mass of the sample in kg.

Physiochemical characterization measurements. Various physiochemical characterization for the soils were measured; namely pH, grain size measurement by laser diffraction, moisture content measurement, carbonate measurement, and total organic carbon measurement.

Results and Discussion

Physiochemical characterizations results. The vertical distribution of radionuclides depends greatly on the soil characteristics\textsuperscript{20}. Radioactivity in soil declines by leaching of water, attenuation by enhanced porosity and by supplementary water and organic matter and rises by sorption and precipitation of radionuclides from incoming water\textsuperscript{21}. Figure 1 illustrates the vertical distribution of total organic carbon (TOC\%) for soil samples in each depth level for 11 locations in Qatar. TOC gives an indicator about the organic matters in soil samples. Figure 1 shows that the organic matter is very low for all soil samples mainly due to low vegetation cover; it varies from 0.08\% to 0.68\%. Because of the high cationic exchange capacity of the organic matter, the results of low organic matter may lead to assumption that adsorption in soil samples may be controlled mainly by an ion exchange mechanism with the clay minerals\textsuperscript{15}. Organic matter is of great importance because it tends to form soluble or insoluble complexes with radionuclides, which may then transport throughout the profile or retain within the soil\textsuperscript{22}. Generally, the top soil samples contain higher organic matter content than the deeper soil samples due to the dominance of pedogenetic processes in upper soil layer. The highest organic matter was in Fwaret soil samples in the top layer from 0–1 cm and 1–6 cm. This was due to geological structure of soil in Fwaret called Sabkha with deposits of saline and gypsiferous sand and silt flat with halophytes plants. Sabkha contained elevated radioactivity concentration of $^{226}$Ra\textsuperscript{23}.

The moisture content of the soil samples in four depth layers is also presented in Fig. 1. It shows that the moisture content of all soil samples was very low which could be due to the season in which sampling was done. It was conducted during summer season where the temperature was from 44–47 °C; the moisture content percentages varied from 0.228\% to 6.32\%. Generally, the topsoil receives direct sunshine, and therefore the topsoil samples contained lower moisture content than the deeper soil samples. The highest moisture content was in the Fwaret soil samples in the deepest layer from 11–16 cm. The lowest moisture content was in Dukhan soil samples collected from deeper depth i.e. 6–11 cm. According to literature survey, soil moisture content generally has a direct impact on redox potential and radionuclide speciation\textsuperscript{22}. Low moisture content would lead to less hydrophilic radionuclide due to decrease in solubility. In addition, radioactive gases which are not fully trapped in soil can also easily be dispersed into the atmosphere; such as: $^{222}$Rn and $^{228}$Ra that decay to $^{220}$Rn and $^{224}$Rn gases, respectively\textsuperscript{5}.

The pH values of the soil samples were in the range from 7.2 to 8.6, which means that the soil was mostly alkaline. According to literature, pH has a strong influence on the mobility of radionuclides that may precipitate some soil components such as carbonates\textsuperscript{25}. At high pH, various precipitates would be formed such as carbonate and hydroxyl, phosphate or sulfide ions complexes. These insoluble precipitates reduce the availability of radionuclides in upper soil surface. While at low pH, radionuclide cations may have displaced by H\textsuperscript{+}\textsuperscript{24}. Actinides and other redox-sensitive elements are mobile in oxidizing conditions and more strongly immobilized in reducing environments. So, any reducing environment in each site can play as an efficient radionuclide\textsuperscript{25}. In summary, Fe is precipitated as oxyhydroxide under alkaline pH, which has the high affinity to scavenge other metals\textsuperscript{26}. Cationic complexes are also abundant in slightly alkaline pH if the soil is rich in organic matter due to the high cationic exchange capacity of the organic matter. However, organic matter is not present in high quantity in Qatar’s soil\textsuperscript{27}. At high pH, the negative charge dominated the surface of the functional group of humic matter\textsuperscript{28}.

Concerning the particle size of soil, all samples were mainly comprised of sand except for Al-Guwayria soil sample, which contained silt. According to the literature, the concentration of clay minerals can affect the distribution of radionuclide bearing primary minerals and consequent radionuclide activity. Soil with sand particles will lead to lower adsorption of radionuclide to soil particles than soil with high clay and silt content\textsuperscript{29}. Sand particles have lower adsorption due to lower surface area and ion exchange capacity (between aluminum silicate (anions) that readily adsorb radionuclide cations on their surface) than clay and silt\textsuperscript{30}.

Carbonate content percentage ranged from 29.59\% to 71.52\% (Fig. 1). According to the literature, high carbonate content may influence processes such as interaction with C-bearing minerals and adsorption of radionuclide to soil particles and consequently concentration of radionuclide due to the competition for ion exchange of radionuclide ions with Ca\textsuperscript{2+} on soil particles surface\textsuperscript{1}. At alkaline environment, carbonate ligands become the major complexing species in solution\textsuperscript{24}.

Radioactivity concentration results. The radioactivity levels of the radionuclide in the soil samples were determined by Canberra Genie 2000 spectroscopy software. The weighted mean radioactivity level of $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in the four soil depth levels (0–1 cm), (1–6 cm), (6–11 cm), and (11–16 cm) for each location site are shown in Fig. 2. The Figure illustrates that the radioactivity concentration levels of the natural radionuclides $^{226}$Ra,
$^{232}\text{Th}$ and $^{40}\text{K}$ vary substantially in each location site depending primarily on the concentration of radionuclides in bedrocks from which the soil originates. Most limestones have relatively little radium concentration. Besides, the levels of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in the soil might be affected by the physiochemical characteristic of the soil. Whereas, the activity concentration of the anthropogenic radionuclide $^{137}\text{Cs}$ varied in each location site due to the variation in the metrological factors at the time of deposition, as well as to the physiochemical characteristics of the soil.

In general, it can be noticed that the radioactivity concentration levels were very low. The IAEA -suggests that it is not required to assign regulatory control for activity levels lower than 1 Bq/g. Though the activity levels at one or above can be exempt from regulatory controls if occupational exposure is established to be less than 1 mSv/y.

Figure 2 illustrates the vertical distribution of $^{40}\text{K}$ radioactivity concentration of four depth levels in different sample location sites. The highest $^{40}\text{K}$ radioactivity concentration was in Al-Zubara (327 ± 14 Bq/kg) in depth soil of (6–11 cm), whereas, the minimum $^{40}\text{K}$ radioactivity concentration was in Al-Jumayliyah (79 ± 3 Bq/kg) in depth soil of (11–16 cm). The high concentration of $^{40}\text{K}$ could be attributed to high clay content; consequently, low radioactivity concentration could be correlated to high sand content. $^{40}\text{K}$ is part of a clay minerals component rather than organic matter, and its mobility is controlled by the solubility in the soil.

Figure 2 shows the vertical distribution of $^{226}\text{Ra}$ radioactivity concentration of four depth levels in different sample location sites. Commonly, limestones have relatively low radium contents. The highest $^{226}\text{Ra}$ radioactivity concentration was in the Fwaret soil sample (32 ± 1 Bq/kg) in depth soil of (6–11 cm). The minimum $^{226}\text{Ra}$
The radioactivity concentration was in Al-Jumayliah (5 ± 2 Bq/kg) in depth soil of (11–16 cm). The result of high 226Ra radioactivity concentration in Sabkha soil from Fwarat in the current study is in line with reported elevated radioactivity concentration of 226Ra in Dukhan Sabkha in previous research. In this study, the elevated 226Ra radioactivity concentration can be attributed to radium’s co-precipitation with strontium, barium or calcium in the celestite crystal of the Sabkha. In addition, it may also be correlated to high moisture content in Sabkha soil as 226Ra is highly soluble.

Figure 2 illustrates the vertical distribution of 232Th radioactivity concentration of four depth levels in different sample location sites. The highest 232Th radioactivity concentration was in Al-Zubara soil samples (18 ± 1 Bq/kg) in depth soil of (11–16 cm), whereas, the minimum 232Th radioactivity concentration was in Al-Jumayliah (3 ± 0.1 Bq/kg) in depth soil of (11–16 cm). Al-Zubara soil samples are clayey silty sand and have large carbonate content, which could be correlated to high 232Th concentration. It was observed from Fig. 2 that the radioactivity concentration levels of 226Ra and 232Th decay products as well as 40K in the surface and deep soil samples were similarly distributed, which is consistent with results of other authors in Qatar and in the worldwide.

The minimum detectable activity (MDA) for the counting time of 20,000 s was 0.03 Bq/kg for 137Cs. 137Cs concentrations per unit mass in Bq/kg dry weight. The average value of the MDA for 226Ra, 232Th, 40K and 137Cs was established as 0.05, 0.03, 0.16 and 0.01 Bq/kg, respectively.

137Cs is detected in trace amounts. Mainly any detected values higher than MDA in this study may result from the fallout of 137Cs from severe nuclear reactor incidents and atmospheric nuclear weapons tests. Figure 2 illustrates the vertical distribution of 137Cs radioactivity concentration of four depth levels in different sample location sites. It was clear that 137Cs radioactivity concentration was high in the topsoil layers and less in the deeper layers. 137Cs radioactivity concentration in surface soil depth at (0–1 cm) and (1–6 cm) in all soil samples were found to be higher than deeper depth at (6–11 cm) and (11–16 cm). Thus, it is present as a permanent source of external gamma dose for several years until it totally decays, however, contamination of the food chain and raise of the internal dose from the human ingestion is very unlikely. The highest 137Cs radioactivity concentration was in Umm Al-Alamaad (47 ± 2 Bq/kg), while the lowest 137Cs radioactivity concentration were in AL-Khor, Ras Lafan and Al-Zubara which were in minimum detectable activity range. Some of the relatively high concentrations of 137Cs can be attributed to rain washouts in these depression sample locations that have lower height relative to the surroundings. The high 137Cs radioactivity concentration can also be correlated to high total organic carbon in these soil samples.

The average concentrations of 232Th, 226Ra, 40K and 137Cs in the 16-cm depth soil were 10, 17, 201 and 4 Bq/kg, respectively. It was noticed from Fig. 3 that in all sampling sites, the average radioactivity level was of the order 40K > 226Ra > 232Th > 137Cs. The radioactivity concentration levels of 40K were the dominant gamma radioactivity source in the soil. It is well known that 40K in the earth’s crust is of the order of percentage while 226Ra and 232Th are in ppm level. 40K is a common primary weathering product.

Comparison of activity concentrations with other studies. Table 2 represents a comparison of weighted mean radioactivity concentration of 226Ra, 232Th, 40K, and 137Cs with data of previous studies in different countries. The average radioactivity concentrations for the for the 16 cm depth in Qatar soil samples are 10, 17, 201 and 4 Bq/kg for 226Ra, 232Th, 40K and 137Cs, respectively, which were below the world averages concentration.
It was clear that the radioactivity concentrations were consistent with the previous reported studies in Qatar. The average concentrations of $^{232}$Th, $^{226}$Ra, $^{40}$K and $^{137}$Cs for the 16 cm depth in Qatar soil were 9, 17, 204 and 6 Bq/kg, respectively. The obtained results were generally comparable to the literature data acquired from other countries, and all are within the worldwide average concentration except in Nigeria, India and Yemen, which are higher than the worldwide mean concentration due to geology and bedrocks of these two countries. The mean radioactivity concentration was found to be lower than the obtained values of those reported in the USA, Turkey, Jordan and Berlin. It also was comparable to the mean concentration published in neighboring countries in Iraq, Kuwait and Saudi Arabia. The variation could mainly be due to the different geology as well as the difference in sampling depth, physiochemical soil characteristic and metrological factors at time of the deposition.

Statistical analysis and correlation of physiochemical characteristic of soil with radioactivity concentration. Statistical analysis was conducted using the analysis of variance (ANOVA) to ascertain whether there are significant differences in the variables analyzed. According to ANOVA analysis, there was no significant difference between $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs radioactivity concentration in the soil samples, which is consistent with the findings of previous studies. The examination of the relationships among radionuclides and the physio-chemical characteristics of the soil was performed through principle component analysis (PCA) and Pearson correlations. The Pearson correlation coefficient was used to measure the strength of the linear relationships between physiochemical properties; namely clay content, silt content, sand content, carbonate content, total organic carbon, and moisture content with radioactivity concentrations. The obtained results are presented in Figs 4 and 5.

Comparison of $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs radioactivity concentrations between different particle sizes of soil confirmed that the presence of clay positively correlates to radionuclides content in the soil, which is in line with the findings of previous studies. The best positive correlation was obtained between clay content, and $^{232}$Th and $^{40}$K activity concentrations with $R^2 = 0.46$ and 0.26, respectively (see Fig. 4). This was expected as natural radionuclides are adsorbed to clay surface due to larger adsorbed surface area and lattice defects, as well as the associated clay minerals, which increases the ion exchange capacity with the radionuclides. Furthermore, most $^{40}$K is adsorbed on the cation exchange sites of clay minerals. The positive correlation was also obtained between silt content, and $^{232}$Th and $^{40}$K activity concentrations with $R^2 = 0.44$ and 0.19, respectively (see Fig. 5). However, there was a...
negative correlation found between the radioactivity concentration and the sand content. The negative correlations with highest $R^2$ values were obtained between sand content and $^{232}$Th and $^{40}$K activity concentrations with $R^2 = 0.45$ and 0.21, respectively (see Supplementary Materials). Carbonate content was negatively correlated with $^{232}$Th, $^{40}$K and $^{137}$Cs, which is similar to the result obtained by\(^\text{18}\). The anions carbonate and bicarbonate can react with the radionuclides forming complexes which are either not adsorbed at all or only slightly adsorbed onto clays. The carbonate bearing cations, on the other hand, compete with the radionuclides for available adsorption sites on the sorbing mineral surfaces, and may thereby enhance nuclide migration if present in large concentrations\(^\text{34}\). The negative correlation was also obtained between carbonate content with $^{40}$K and $^{232}$Th activity concentrations with $R^2 = 0.47$ and 0.22, respectively (see Supplementary Materials). Total organic carbon as an indicator for organic matter was found to be positively correlated with $^{226}$Ra and $^{137}$Cs activity concentrations with $R^2 = 0.41$ and 0.187, respectively (see Supplementary Materials) which is similar to conclusions of\(^\text{29}\). According to the literature, the soil organic matter contains chains of carbon atoms, containing polar and/or ionized surface functional groups, like OH$^-$ and COOH$^-$, and generally, radionuclides strongly adsorbed to these functional groups and form stable complexes\(^\text{19}\). The strong correlation was not found between particle size and carbonate content of soil with $^{137}$Cs radioactivity, which is consistent with the previous studies\(^\text{4,15,35,36}\). Concerning the moisture content, the positive correlation was obtained between $^{226}$Ra activity concentrations with $R^2 = 0.44$ (see Supplementary Materials), which agreed with the study of\(^\text{32}\). The high radioactivity of $^{226}$Ra could be due to its high solubility.

Figure 6 shows the principle component analysis (PCA) for the correlation between physiochemical characteristics and radioactivity concentration of $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{137}$Cs (Bq/kg) of the soil samples. PCA explained a linear combination of the original variables. Figure 6 shows a well correlation between silt and moisture content with the radioactivity concentration of $^{226}$Ra in the first component. The second component was well correlated $^{232}$Th and $^{137}$Cs concentration to the total organic carbon, pH and clay content in soil.

**Radiological risk assessment.** By applying these factors, the total absorbed gamma dose rate ($D$) in air at 1 m above the ground level was determined as given in the Eq. (2) below provided by the UNSCEAR report\(^\text{6}\):

\[
D \ (\text{nGy/h}) = 0.462 \ A_{\text{Ra}} + 0.604 \ A_{\text{Th}} + 0.0417 \ A_{\text{K}}
\]
where $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the activity concentrations of $^{226}Ra$, $^{232}Th$, and $^{40}K$ in Bq/kg, respectively. The coefficient 0.462 is given for the complete $^{238}U$-series (thereby including gamma radiation from the $^{238}U$-daughters $^{234}$Th and $^{234}$m-Pa, although the contribution to the dose from these daughters is relatively low).

Accordingly, the calculated $D$ values from Eq. (2) are provided in Table 3. The estimated mean absorbed doses in outdoor were appeared to be in the range 15.13–32.31 nGy/h. According to UNSCEAR, the average $D$ value in the world is 51 nGy/h\(^{37}\). The result obtained by this study of average absorbed dose for the soil samples in outdoor was 22 nGy/h, which is far below the average dose value in the world.

Radium equivalent activity ($R_{eq}$) is the widely used parameter to assess the gamma ray hazards\(^{38}\), which is estimated on the assumption that $370$ Bq/kg $^{226}Ra$ or $260$ Bq/kg $^{232}Th$ or $4810$ Bq/kg $^{40}K$ yield the same gamma dose rate. The $R_{eq}$ of the sample in (Bq/kg) can be achieved utilizing the Eq. (3)\(^{39}\):

$$R_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_{K}$$  \hspace{1cm} (3)

where $A_{Ra}$, $A_{Th}$, and $A_{K}$ are the activity concentrations of $^{226}Ra$, $^{232}Th$, and $^{40}K$ in Bq/kg of the soil sample, respectively.

$R_{eq}$ values estimated from Eq. (3) are given in Table 3. The calculated range of radium equivalent activity was established to be 31–67 Bq/kg. Accordingly, the calculated mean radium equivalent activity was established to be 47 Bq/kg. The recommended maximum value of $R_{eq}$ is 370 Bq/kg\(^{40}\). All estimated values of $R_{eq}$ in this study are much lower than the recommended value.

The annual external effective dose rate (AEDE) was estimated from Eq. (4)\(^{41}\)

$$AEDE \, (\mu Sv/\text{y}) = D \, (\text{nGy/h}) \times 8760 \, (\text{h/yr}) \times 0.2 \times 0.7 \, (\text{Sv/Gy}) \times 10^{-3}$$  \hspace{1cm} (4)

Figure 5. Correlation between silt content and radioactivity concentration of $^{226}Ra$, $^{232}Th$, $^{40}K$, and $^{137}Cs$ (Bq/kg) in soil samples.
where 0.2 is the outdoor occupancy factor used for the fraction of the time spent by a person, implying that 20% of time is spent outdoors\(^{27}\); However without including spent indoors (with radiation through the building walls and radiation from the building material itself) a possible underestimation of the total external dose and the associated risk. 8760 h is the time for one year and 0.7 Sv/Gy is the conversion coefficient factor from gamma absorbed dose rate in air outdoors \(D\), which converts the absorbed dose rate in air to human effective dose to effective dose received by adults.

The average global effective dose rate to members of public from soil with weighted mean activity concentrations of 33 Bq/kg, 32 Bq/kg, 45 Bq/kg, and 420 Bq/kg for \(^{238}\)U, \(^{226}\)Ra, \(^{232}\)Th, \(^{40}\)K, respectively is 0.460 mSv/yr\(^{27}\). AEDE values estimated from Eq. (4) are shown in Table 3. The estimated range of annual mean effective dose equivalent was established to be 0.039–0.018 mSv/y. The calculated average annual mean effective dose equivalent was found to be from 0.027 mSv/yr. This value is also far below the world average value of 0.46 mSv/yr\(^{10}\).

Figure 6. Principle component analysis for the correlation between physiochemical characteristics and radioactivity concentration of \(^{226}\)Ra, \(^{232}\)Th, \(^{40}\)K, and \(^{137}\)Cs (Bq/kg) of soil samples.

### Table 3. Radium equivalent activity (Bq/kg), absorbed gamma radiation dose rate in air (nGy/h), annual effective dose (mSv/y), external radiation hazard index (\(H_{ex}\)) and lifetime cancer risk (LTCR) with the current soil samples in Qatar.

| Soil Sample Location | Dose rate (D) (nGy/h) | Radium equivalent activity (Ra\(\text{eq}\)) (Bq/kg) | External hazard index (\(H_{ex}\)) | Annual effective dose equivalent (AEDE) (mSv/y) | Lifetime Cancer Risk (LTCR) \(\times 10^{-3}\) |
|----------------------|------------------------|--------------------------------|--------------------------------|--------------------------------|----------------------------------|
| Fwarat               | 25                     | 52                            | 0.14                          | 0.03                           | 0.105                           |
| Al-Shamal            | 15                     | 32                            | 0.086                         | 0.018                          | 0.065                           |
| Al-Areesh            | 18                     | 38                            | 0.102                         | 0.022                          | 0.078                           |
| Al-Zubara            | 32                     | 68                            | 0.182                         | 0.039                          | 0.139                           |
| Al-Guwayria          | 32                     | 67                            | 0.181                         | 0.039                          | 0.138                           |
| Al-Jumayliyah        | 13                     | 28                            | 0.075                         | 0.016                          | 0.057                           |
| Dukhan               | 17                     | 34                            | 0.092                         | 0.020                          | 0.071                           |
| Al-Shahaniyah        | 19                     | 39                            | 0.106                         | 0.023                          | 0.082                           |
| Ras Lafan            | 30                     | 64                            | 0.171                         | 0.037                          | 0.137                           |
| Umm Al-Amad          | 25                     | 53                            | 0.142                         | 0.031                          | 0.108                           |
| Al-Khor              | 22                     | 46                            | 0.123                         | 0.026                          | 0.941                           |
Hex and Hin are the radiation hazards indices and are defined as the external hazard indices. These show the external exposure to gamma radiation from the studied sand and sediment samples, and internal hazard index due to internal exposure to gamma radiation from ingestion food or inhalation. Both must be less than unity and are given by Eqs (5 and 6):

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_{K}/4810$$ (5)

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_{K}/4810$$ (6)

where $A_{Ra}$, $A_{Th}$, and $A_{K}$ are the activity concentrations of $^{226}Ra$, $^{232}Th$, and $^{40}K$ in Bq/kg of the soil sample, respectively.

In this study, $H_{ex}$ was estimated only taking into consideration that the external hazard, which is caused by gamma rays, corresponds to a maximum equivalent activity of 370 Bq/kg, 259 Bq/kg and 4810 Bq/kg for $^{226}Ra$, $^{232}Th$, and $^{40}K$ respectively. $H_{ex}$ values estimated from Eq. (5) are shown in Table 3. The calculated range of $H_{ex}$ was found to be 0.083–0.181. The maximum external hazard index should be less than unity. From Table 3, it is concluded that all estimated values are lower than one.

Lifetime cancer risk (LTCR) caused by the annual effective dose rate was calculated utilizing Eq. (7):

$$LTCR = AEDE \times AL \times RF$$ (7)

where external exposure (AEDE) is the annual effective dose rate, AL is the average life time (70 years) and RF is the risk factor (0.05).

The calculated values of LTCR are presented in Table 3. The values of LTCR varied from $0.064 \times 10^{-3}$ to $0.139 \times 10^{-3}$ with an average of $0.096 \times 10^{-3}$ which is much less than the world average of $0.299 \times 10^{-3}$. From the results of radiation hazard indices in all soil samples in Qatar, we can conclude that the radiological risk for soil samples in the current study was insignificant due to the low values of calculated $D$, AEDE, $R_{aq}$, $H_{ex}$ and LTCR in all samples under investigation.

The mean values of radium equivalent activity ($R_{aq}$), the total absorbed dose rate in air 1 m above the ground (D), the annual effective dose (AEDE) and the external hazard index ($H_{ex}$) in this study were compared with the other global measurements for different countries of the world as given in Table 4. From Table 4, it can be noticed that the calculated D, AEDE, $R_{aq}$, $H_{ex}$ values were found to be far below the allowed limits, and all other countries indices are within the limits of $6$. The obtained radiological indices results were strongly agreed with previous results in Qatar. In addition, it is shown that the radiological indices in the current study were established to be lower than the calculated values of those reported in other countries.

Table 4. Comparison of the worldwide mean values of radium equivalent activity (Bq/kg), absorbed gamma radiation dose rate in air (nGy/h), annual effective dose (mSv/y), and external radiation hazard index ($H_{ex}$) with the current study.

| Country       | Dose rate (D) (nGy/h) | Radium equivalent activity ($R_{aq}$) (Bq/kg) | External hazard index ($H_{ex}$) | Annual effective dose equivalent (AEDE) (mSv/y) |
|---------------|----------------------|-----------------------------------------------|----------------------------------|-----------------------------------------------|
| worldwide average values | 55                   | 370                                           | $\leq 1$                         | 1                                             |
| Current study  | 22                   | 47                                            | 0.125                            | 0.027                                         |
| Qatar         | 24.2                 | 50.4                                          | 0.1                              | 0.029                                         |
| Saudi Arabia  | 37.2                 | 74.1                                          | 0.02                             | 0.04                                          |
| Iraq          | 23.27                | 46.82                                         | 0.116                            | 0.132                                         |
| Kuwait        | 24.65                | 50.72                                         | 0.14                             | 0.03                                          |
| Jordan        | 51.5                 | 103.1                                         | 0.28                             | —                                             |
| Turkey        | 45                   | 96                                            | 0.26                             | 0.056                                         |
| Nigeria       | 86                   | 86.32                                         | —                                | 0.147                                         |
| Yemen         | 89.45                | 191                                           | 0.52                             | —                                             |
| Berlin        | 51                   | 62                                            | —                                | —                                             |
| USA (Texas)   | 48.4                 | 102.4                                         | 0.3                              | 0.059                                         |

Conclusion

In this current study, the activity concentrations of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ of the 44 soil samples collected from Qatar (Al-Shamal, Al-Zubara, Al-Areeesh, Fwaret, Ras Laffan, Al-Guwariah, Al-Khawr, Al-Jamaliayah, Dukhan, Al-Shahaniyah, Um Al-Amad) were measured utilizing high purity germanium (HPGe) gamma ray spectrometry. The average activity concentrations of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ were lower than the average values of the earth’s crust. The average concentrations of $^{226}Th$, $^{226}Ra$, $^{40}K$ and $^{137}Cs$ in the 16-cm depth soil were 10.08, 16.6, 200.63 and 3.57 Bq/kg, respectively. The external absorbed gamma dose rate (D), the annual effective dose, the
mean radium equivalent activity ($Ra_{eq}$), the external hazard index ($H_e$) and in and Lifetime cancer risk (LTCR) were evaluated as 22.37 nGy/h, 0.027 mSv/y, 46.58 Bq/kg, 0.12 and 0.096 × 10⁻³, respectively. The results of the radioactivity concentration indices indicated that all soil samples are complying with the exemption annual dose permissible values.

Uniform vertical distributions of natural radionuclides $^{226}Ra$, $^{232}Th$, and $^{40}K$ were found in the uppermost 0–16 cm of soil. Whereas, $^{137}Cs$ radioactivity concentration was higher at 0–6 cm than deeper depth 6–16 cm, this shows that $^{137}Cs$ migrates very slowly in undisturbed soil.

Based on analytical results of Pearson correlation coefficient, the radioactivity concentrations were affected by the physiochemical characteristics of soil. The best positive correlation was observed between clay content, and $^{232}Th$ and $^{40}K$ activity concentrations and carbonate content was negatively correlated with $^{232}Th$, $^{40}K$ and $^{137}Cs$. Total organic carbon was positively correlated with $^{226}Ra$ and $^{137}Cs$ activity concentrations. Concerning the moisture content, the best positive correlation was obtained with $^{226}Ra$ activity concentrations.

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Author Contributions

Ayesha Y. Ahmad and Mohammad A. Al-Ghouti designed the experiments. Ayesha Y. Ahmad performed the experiments. Ibrahim AlSadig and Mohammed Abu-Dieyeh contributed to experimental design. Ayesha Y. Ahmad and Mohammad A. Al-Ghouti performed data analysis. Ayesha Y. Ahmad and Mohammad A. Al-Ghouti wrote the manuscript. Ibrahim AlSadig and Mohammed Abu-Dieyeh contributed to editing and reviewing the manuscript.
Additional Information

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