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Establishment of natural radioactivity baseline, mapping, and radiological hazard assessment in soils of Al-Qassim, Al-Ghat, Al-Zulfi, and Al-Majmaah

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Abstract

Radionuclide species comprising the primordial radioactive decay chains lead by 232 Th and 238 U, along with their associated daughter and granddaughter decay series, constitute a major fraction of the naturally occurring radioactive material (NORM). Gamma-ray radiation emitted by the decay of these NORMs, along with the ${}^{40}K$ in the soil, is mainly responsible for the human exposure to external gamma-rays. The radioactivity concentrations in soil samples collected from 138 sites have been determined as part of a survey to produce a radiological map of AL-Qassim, Al-Ghat, Al-Majmaah, and Al-Zulfi regions in the Kingdom of Saudi Arabia. The estimated mean values of ²³⁸U, ²³²Th, ⁴⁰K, ²²⁶Ra, and gross α and β activities in the samples were 23.8 ± 11.61, 24.33 ± 17.63, 790 ± 398, 22.83 ± 11.70, 375 ± 205, and 734 ± 344 Bq kg⁻¹, respectively. The radiological risk indices related to the natural radioactivity in the soil samples, i.e., the absorbed dose rate in air, radium equivalent activity, and annual effective dose rate, were estimated to be 58.88 ± 29.21 nGy h⁻¹, 117.1 \pm 59.65 Bq kg⁻¹, and 0.07 ± 0.04 mSv year⁻¹, respectively. The determined internal hazard index of the samples ranged from 0.07 to 1.45, with an estimated mean value of 0.4 ± 0.2 . The radon concentration in the soil gas ranged from 39 to 508 Bq m⁻³, with a mean value of 145.0 ± 52.9 Bq m⁻³.

Keywords Soil \cdot Natural radionuclides \cdot ²²²Rn soil gas \cdot Gross α and β \cdot Risk assessment

Introduction

The measurement of naturally occurring radioactive materials (NORMs) in environmental samples has received increasing attention recently. This is primarily because of the concern about human exposure resulting from the radioactive contamination of the environment. The environmental radiation background is a representation of the natural radioisotopes of ^{238}U , 232 Th, primordial 40 K, and other artificial radionuclides such as 137 Cs. Natural radiation accounts for approximately 85% of the human effective dose (UNSCEAR [2000](#page-10-0); IAEA [1996\)](#page-10-0). Its presence at certain levels can affect the human body, depending on location and geological characteristics. The activity levels of these radionuclides can be determined by a gamma

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 \boxtimes Thamer Alharbi t.alharbi@mu.edu.sa radiation hazards assessment. The natural radiation to which the human body is exposed mainly originates from the decay of radium isotope (226 Ra), which produces radon gas (222 Rn) and its decay progeny (Yuvi [1988](#page-10-0); Abdullahi et al. [2019\)](#page-9-0). Radon is a noble gas that appears in different geological formations such as soil, rocks, the earth's crust, and building materials. The local environmental radon level is mainly determined by the radon exhalation rate from the soil, and the content of 226 Ra in soil plays an important role in the accumulation of 222 Rn gas in the outdoor and indoor environments. However, 226 Ra and 222 Rn are the decay products of ²³⁸U with an estimated average of 2.7 mg kg⁻¹ (Lide [1994\)](#page-10-0). Therefore, environmental radiation background surveys are important for understanding radiological and geological characteristics, and also for identifying the biochemical and geochemical traces in the environment. Gross α and gross β activities in environmental samples (e.g., rocks, sand, soil, and water) have attracted considerable attention in recent years, because our living environment has been contaminated by natural radionuclides. Furthermore, elevated levels of β radiation may be a sign of the accidental atmospheric release of radioactivity from the Chernobyl nuclear reactor or nuclear

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test explosions. The total radioactivity of α emitters in the soil of a particular area determines the area's gross α activity, which depends on geological formations, mineral distribution, and the type of activities in the selected area.

An artificial radioactive element such as cesium is produced in nuclear power plants and disposed in wastewater. The Fukushima Daiichi nuclear power plant accident in Japan increased the activity concentrations of 134 Cs and 137 Cs, with the respective half-lives of 2.06 and 30.4 years, in the soil, air, and water to dangerous levels (Shizuma et al. [2018](#page-10-0)). Several studies have described successful methods for removing radioactive elements such as cesium $(^{134}Cs$ and ^{137}Cs) and cobalt (Co(II)) from nuclear wastewater. These studies involved the development of ligand-based nanomaterials such as mesoporous silica and conjugate adsorbent (Awual at al. [2014a](#page-10-0)), inorganic and conjugate adsorbent (Awual et al. [2014b](#page-10-0)), DSDH ligand (Shahat et al. [2015\)](#page-10-0), nano-conjugate adsorbent (Awual et al. [2015\)](#page-10-0), organic–inorganic mesoporous hybrid (Awual et al. [2016a\)](#page-10-0), crown ether-based mesoporous adsorbent (Awual [2016](#page-10-0)), Schiff base ligand (Awual et al. [2017](#page-10-0)), and macrocyclic ligand (Awual et al. [2016b](#page-10-0)).

No study has yet reported the levels of natural radioactivity in the soils of the Al-Qassim, Al-Ghat, Al-Majmaah, and Al-Zulfi regions of Saudi Arabia. The present study aimed to estimate the natural radioactivity concentrations, radon

concentrations, and gross α and β activities in these regions and to provide a radiation map as a reference for future radiological evaluations. The activity concentrations of 238 U, 226 Ra, 232 Th, and 40 K in 138 soil samples across the four regions were determined and then used to estimate the associated radiological hazards. The measurements were performed using a highpurity germanium (HPGe) gamma-ray detector. The gross α and β activities in the soil samples were estimated using lowbackground proportional detectors. The radiological hazards and risk assessment due to naturally occurring radionuclides $(^{232}Th, ^{226}Ra,$ and $^{40}K)$ in the samples were estimated and compared with available results from other countries.

Materials and methods

Geology of the study area

This study was conducted in four different regions (Al-Qassim, Al-Ghat, Al-Majmaah, and Al-Zulfi) of Saudi Arabia. The study regions (latitude 24–27° N and longitude 41–44° E) are located in the center of the Arabian Peninsula, north-west of Riyadh, and have a population of 1.9 million (Fig. 1). The geology of the study area is not uniform; it consists of sand, clay, and limestone. The

Fig. 1 The map of Saudi Arabia showing the study area

Rummah Valley or Wadi Rummah, which has various dry riverbeds, crosses the entire Al-Qassim region and is one of the longest valleys in the Arabian Peninsula, covering a distance of over 600 km starting from the west near Medina to the border of Kuwait in the north-east of the Arabian Peninsula. In the Al-Qassim province, the soil consists of loose deposits in its natural state, but it can be compacted to attain relatively high, dry densities (Al-Refeai and Al-Ghamdy [1994\)](#page-10-0).

Sample collection and preparation

There are several approaches for collection of soil samples include random, judgmental, stratified random, systematic random grid, systematic grid, search, and transect sampling (IAEA [2004](#page-10-0)). After field visit, the most suitable approach for the current study is systematic random grid due to some restrictions of sample collections in some grid points. The total area of the Al-Qassim province, Al-Ghat, Al-Majmaah, and Al-Zulfi was subdivided using square grids, and the soil samples were collected from within each cell using the random selection procedures. Soil samples were collected from 138 different sites at a depth of 0.2–0.3 m from ground level to obtain undistributed and pure soil samples from the four study regions, as shown in Fig. 2. Approximately 2–3 kg of soil from each location was collected in a polyethylene bag. The samples were sieved using a 2-mm mesh to remove unwanted materials. The samples were then dried in an oven at 90 °C for 24 h to remove any moisture. Additional sieving was performed using a 500-μm sieving machine (Octagon digital) to achieve homogeneity between samples. The dried, sieved samples were weighed and transferred into Marinelli beakers (500 mL) and sealed with adhesive tape. The beakers were then stored for 30 days to achieve secular equilibrium between radium (226 Ra) and radon (222 Rn) and radon's most abundant daughter products.

To measure the gross α and β activities, approximately 2 g of each dried sample was placed on a 5.08-cm-diameter stainless-steel planchet, and a small amount of UHU glue diluted with acetone was spread over the sample surface. The planchets were then kept under an infrared lamp for a few hours until the samples were fully glued.

Fig. 2 Location of sampling points (blue circles) and study regions: Al-Qassim (red), Al-Zulfi (green), Al-Ghat (yellow), Al-Majmaah (blue), and unmeasured areas (gray).

Experimental technique

The activity concentration of radionuclides in the soil samples was determined by using a gamma-ray spectrometry technique. The gamma-ray spectrometry system comprised a high-resolution (n-type) HPGe coaxial detector (ORTEC) with a pre-amplifier, amplifier, high-voltage power supply, and multi-channel analyzer (MCA) coupled to a PC. To reduce the thermally induced leakage current, the HPGe detector was cooled using an X-COOLER system (ORTEC). The HPGe detector was placed in a 10-cm-thick cylindrical Pb shielding that was internally lined with a sheet of Cu (1.6 mm) and Sb (0.5 mm) to reduce the external gamma-ray radiation background in the measured spectrum. The HPGe detector efficiency curve based on the experimental data was calculated from the following equation:

$$
\varepsilon_c = \frac{N_c}{t_c \times A_c \times P_\gamma},
$$

where ε_c is the photopeak efficiency corresponding to a specific radionuclide, N_c is the net count in the photopeak, t_c is the measurement time (s) of the calibration source, A_c is the present activity of the source, and P_{γ} is the probability of γ -emission of the radionuclide. The energy resolution (FWHM) of the spectrometer was 0.8 keV and 2.2 keV for the 122-keV 57 Co and 1332-keV 60 Co gamma-ray lines, respectively. The energy and efficiency calibrations of the detector were performed using a mixed radionuclide source of ²¹⁰Pb, ⁸⁸Y, ²⁴¹Am, ¹³⁷Cs, ⁵⁷Co, ¹⁰⁹Cd, ¹³⁹Ce, ²⁰³Hg, ¹¹³Sn, ⁶⁰Co, and ⁸⁵Sr. The source was kept inside a 500-ml Marinelli beaker with the same geometry as that used for sample measurements. The efficiencies and activities were determined from the counts in the photopeak of the specific nuclides using the gamma vision software. Each soil sample was counted for 60,000 s. The background spectra, which were subtracted from the sample spectra, were collected for 48 h using deionized water in the Marinelli beaker.

Activity concentration determination by gamma-ray spectroscopy

The activity concentration of $226Ra$ was calculated from the gamma-ray lines of energies 338.3, 911.1, and 969.11 keV $(2^{228}$ Ac); 609.3 keV $(2^{14}$ Bi); and 351.9 keV $(2^{14}$ Pb). The gamma-ray lines of 238.63 keV (^{212}Pb), 727.3 keV (^{212}Bi), and 583.19 keV $(^{208}$ Tl) were used to determine the activity concentration of 232 Th. The gamma-ray photopeaks at 661.6 keV and 1460.8 keV were used to determine the activity concentrations of ^{137}Cs and ^{40}K , respectively. The minimum detectable activities (MDAs) for the soil samples were 0.40 ± 0.12 Bg kg⁻¹ for ²²⁶Ra, 0.32 ± 0.08 Bg kg⁻¹ for ²³²Th, 1.68 ± 0.52 Bg kg⁻¹ for ⁴⁰K, and 0.03 ± 0.02 Bg kg^{-1} for ^{137}Cs . The activity concentrations of

specific radioisotopes were determined using the following expression (Dovlete and Povinec [2004](#page-10-0)):

where A is the activity concentration of a specific radionuclide in Bq kg^{-1} , C_n is the net count of the corresponding peak, P_{γ} is the absolute emission probability for a specific gammaray energy, ε_n is the absolute energy efficiency, m is the mass of the sample in kg, and t_s is the counting time in s.

Fig. 3 Activity concentration distributions of ²³⁸U, ²³²Th, ²²⁶Ra, and ⁴⁰K in the soil samples; red lines represent worldwide average values

Determination of radon concentration

The radon $(^{222}$ Rn) concentration of the sampling locations was measured using a RAD7 semiconductor-based detector connected to a soil gas probe. At each sampling location, the stainless-steel probe, with holes near the tip, was immersed in the soil to a depth of 1 m. The probe was connected to the RAD7 detector with a desiccant tube, followed by inert filters for absorbing the soil gas. The counting time was 30 min for each location (Durridge Co [2017](#page-10-0); Thu et al. [2019](#page-10-0)).

Gross α and β activity measurements by proportional counting system

An advanced low-background α/β proportional system (LB4200, CANBERRA) was used to determine the gross α and β activities. The multi-detector counting system was equipped with 16 gas flow proportional detectors using a mixed P10 gas (90% Ar and 10% CH₄). The system was calibrated simultaneously using disk-shaped standard sources $($ ⁹⁰Sr for β and ²⁴¹Am for α particles). The counting efficiency of the low-background proportional counting system was found to be 2.94% for α particles (based on the guaranteed

reagent U_3O_8 standard) and 28.92% for β particles (based on the high-purity reagent KCl standard) using 2 g of the standard reference sample. The background and samples were counted for 24 h. For the soil samples, the MDAs were 0.11 Bg kg^{-1} for gross α and 0.16 Bg kg⁻¹ for gross β.

Radiological risk assessment

The specific activity concentrations of the 226 Ra, 232 Th, and 40K nuclides were used to estimate the outdoor external dose rates for the measured samples. The internal hazard index (H_{int}) , external hazard index (H_{ext}) , radium equivalent activity (Ra_{eq}) , and annual effective dose rate equivalent (AEDE) were calculated using the following equations (Berekta and Mathew [1985](#page-10-0); UNSCEAR [2008](#page-10-0)):

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

\n
$$
DR = (0.462 \times A_U) + (0.604 \times A_{Th}) + (0.0417 \times A_K)
$$

\n
$$
AEDE = D \times 1.23 \times \frac{10^{-3} mS_V}{y}
$$

\n
$$
H_{int} = \left(\frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right),
$$

Fig. 4 Radiation map of activity concentration distributions of ⁴⁰K for selected regions (Al-Qassim province, Al-Ghat, Al-Zulfi, and Al-Majmaah)

where DR is the absorbed dose rate in nGy h^{-1} in the air 1 m above the ground surface; Ra_{eq} is in Bq kg⁻¹; AEDE is in mSv year⁻¹; and A_U , A_{Th} , and A_K are the activity concentrations of 238 U, 232 Th, and 40 K in Bq kg^{$^{-1}$} in the soil samples, respectively.

Results and discussion

Radionuclides concentration

Table [1](#page-3-0) lists the range of activity concentrations and average activity concentrations of the ²³⁸U, ²³²Th, ²²⁶Ra, and ⁴⁰K nuclides for all soil samples. The overall average activity concentrations of 238 U, 232 Th, and 226 Ra in the soil samples were 23.80 \pm 11.61, 24.33 \pm 17.63, and 23.83 \pm 11.70 Bq kg⁻¹, respectively. These average activity concentrations are lower than the worldwide average values of 35, 30, and 35 Bq kg^{-1} , respectively, whereas for 40 K, the mean activity concentration was found to be 790 \pm 398 Bq kg⁻¹, which exceeds the worldwide average of 400 Bq kg⁻¹ (UNSCEAR [2000](#page-10-0)). The activity concentrations of ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K at the sampling points in the Al-Qassim region were higher compared with those in the other three regions (Al-Ghat, Al-Zulfi, and Al-

Majmaah). This may be caused by the heavy mineral groups from the Paleozoic Era that are dominated by the ultra-stable minerals zircon, tourmaline, and rutile in the Al-Qassim region. Other heavy minerals, including garnet, monazite, staurolite, and kyanite, were also found in small quantities. Among light minerals, alkali feldspars were more abundant than plagioclase, owing to the significantly higher $K₂O$ concentrations, controlled by the presence of K-feldspar, K-mica, and glauconite (Bassis et al. [2016](#page-10-0)). The activity levels of ²³⁸U, ²³²Th, and ⁴⁰K in the soil samples are compared with those from other countries in Table [2](#page-4-0). The variations in the activity levels can be explained by the differences in geological and geographical conditions, as well as by the geochemical characteristics. Figure [3](#page-4-0) shows the activity concentration distributions of 238 U, 232 Th, 226 Ra, and 40 K in each soil sample compared with their worldwide average values, indicated by red lines.

Figure [4](#page-5-0) shows the distribution of the measured activity levels of 40K from the present work. The highest activities were found in the southwestern Al-Qassim region, and it is clear that its activity decreases from west to east. The maximum concentration of 40 K found in this work was below the highest worldwide concentration. The high levels of 40 K concentrations in the south-west region may be attributable to the

Fig. 5 Radiation map of activity concentration distributions of ²³⁸U for selected regions (Al-Qassim province, Al-Ghat, Al-Zulfi, and Al-Majmaah)

high content of heavy and lighter minerals in red to pink sedimentary rocks and partially micaceous silty sandstones. Moreover, there are various dry riverbeds, many crossing the entire region from the west to the north-east (Al-Refeai and Al-Gha[mdy](#page-10-0) 1994; Abu Shayeb e[t al.](#page-10-0) 2017).

The results from the activity concentration measurements were used to generate a radiological map of the selected regions (138 sampling points), as depicted in Figs. [4](#page-5-0), [5,](#page-6-0) 6, [7](#page-8-0) for 40 K, 238U, 226Ra, and 232Th, respectively. The activity distribution of 40 K, 238 U, 226 Ra, and 232 Th of the selected regions was decreased in a way west-south > north-west > north-east > eastsouth. However, radionuclide concentrations in the south-west region of Al-Qassim were higher than those in the other regions due to being highly enriched in zircon (Bassis et al. [2016\)](#page-10-0). Benaafi and Abdullatif [\(2015\)](#page-10-0) reported that the mineralogical composition of the sand dunes in the Al-Qassim region has a high percentage of $SiO₂$ (quartz) and a very low percentage of feldspars. However, in some areas, feldspars and calcite were found significant minerals. A reasonable hypothesis would be that uranium, thorium, and potassium-enriched crystallinity is the main source of heavy and lighter minerals in the sandstones or sedimentary rocks of the Al-Qassim region during the Ordovician age (Pitkin and Huffman [1986\)](#page-10-0).

The measured activity concentrations were correlated for all samples for 238 U/²²⁶Ra and 232 Th/²²⁶Ra. The ratio of 238 U/²²⁶Ra and ²³²Th/²²⁶Ra varied from 0.6 to 1.5 and 0.4 to 2.2, respectively. The average values of $^{238}U^{226}$ Ra and ²³²Th/²²⁶Ra were 1.0 \pm 0.2 and 1.1 \pm 0.3 respectively. There is a slight variation in 232 Th/ 226 Ra activity ratios across the soil samples, which may be due to the fact that uranium or thorium has been absorbed by clay minerals or organic matters in soil.

Radon concentration

The range and average value of the radon (^{222}Rn) concentration in all soil sample locations from the present work are listed in Table [1](#page-3-0). The measured 222 Rn concentration varied from 39 to 508 Bq m⁻³ with an average of 145.0 ± 52.9 Bq m⁻³. The measured values are in agreement with the typical values (UNSCEAR [2000](#page-10-0); Eisenbud and Gesell [1997\)](#page-10-0). The levels of 222Rn in the samples from the Al-Qassim region were higher than those from the other regions, which can be attributed to the different soil types and compact packing of the sandstones/bedrocks. Based on the Sweden Criteria radon hazard assessment (Lara et al. [2015](#page-10-0)), a radon activity concentration below 10 kBq m^{-3} is classified as "low risk," a

Fig. 6 Radiation map of activity concentration distributions of ²²⁶Ra for selected regions (Al-Qassim province, Al-Ghat, Al-Zulfi, and Al-Majmaah)

Fig. 7 Radiation map of activity concentration distributions of ²³²Th for selected regions (Al-Qassim province, Al-Ghat, Al-Zulfi, and Al-Majmaah)

concentration between 10 and 50 kBq m^{-3} is classified as "normal risk," and a concentration greater than 50 kBq m^{-3} is classified as "high risk." Results from the current work show that all values classified as "low risk."

Gross α and gross β activities

The gross α and β activities of the samples from the selected regions are summarized in Table [1](#page-3-0). The gross β activity in groundwater samples from the same regions (Al-Qassim, AlZulfi, and Al-Majmaah) was found to be approximately 5–100 times higher than the gross α activity, owing to the high concentrations of 228 Ra and 40 K, compared with the 226 Ra con-centration (Alharbi et al. [2018](#page-10-0)). The gross α activity depends only on the levels of natural α -emitters (²³⁸U, ²³²Th, ²³⁰Th, ²²⁸Th, ²²⁶Ra, and ²¹⁰Po), while the gross β activity depends on the levels of natural β-emitters $(^{228}Ra, ^{210}Pb,$ and 40 K). It was found that the levels of gross α and gross β activities at the sampling points of the Al-Qassim region were higher than those of the other three regions (Al-Ghat, Al-Zulfi,

Table 4 Radiological risk factors measured for all samples from the current work

a UNSCEAR (2000)

and Al-Majmaah), which may be due to the high concentrations of ²³⁸U, ²³⁵U, ²³²Th, ²²⁶Ra, and ⁴⁰K found in this study. Table [3](#page-8-0) compares the levels of gross α and gross β activities in the soil samples with those from other countries. The different ranges of gross α and gross β activities are due to the activity concentrations of uranium $(^{234}$ U and 238 U), radium $(^{226}$ Ra and 224 Ra), polonium $(^{210}$ Po), lead $(^{210}$ Pb), and potassium (^{40}K) , which normally depend on the geological and geochemical characteristics of the soil.

Radiological hazards and risk assessment

The evaluated radiological risk indices for the soil samples from the present work are summarized in Table 4. The gamma-absorbed dose rate (DR) in the air 1 m above ground level is defined as the radiation dose received by a person from gamma-rays emitted by the radionuclides $(^{232}Th, ^{226}Ra, ^{40}K,$ and $137Cs$) present in the soil. The DR values of the samples varied from 11.05 to 202.04 nGy h^{-1} , with an average value of 58.88 ± 29.21 nGy h⁻¹. Radium equivalent activity (Ra_{eq}) was used to assess the hazards associated with soil that contains 232 Th, 226 Ra, and 40 K; it is determined by assuming that 370 Bq kg⁻¹ of ²²⁶Ra, 260 Bq kg⁻¹ of ²³²Th, and 4810 Bq kg^{-1} of ⁴⁰K produce the same gamma dose rate (UNSCEAR [2000\)](#page-10-0). The range of Ra_{eq} for the samples was 23.6–147.8 Bq kg^{-1} , with an average value of 118.48 ± 59.65 Bq kg⁻¹. H_{int} was used to evaluate the internal radiation exposure to the respiratory system through the inhalation of radioactive gasses (radon and thoron) and their short-lived daughter products, which emit α particles. The H_{int} values of the samples varied from 0.07 to 1.45 with an estimated mean value of 0.4 ± 0.2 . H_{ext} was used to estimate the hazards due to exposure to gamma-ray radiation associated with the ²³²Th, ²²⁶Ra, and 40 K in soil. The H_{ext} values of the samples varied from 0.1 to 1.2, with an estimated mean value of 0.3 ± 0.2 . The AEDE values of the samples ranged from 0.01 to 0.25 mSv year⁻¹, with an estimated average of 0.07 ± 0.04 mSv year⁻¹, which is below the maximum recommended worldwide level of 1 mSv $year⁻¹ (UNSCEAR 2000).$ $year⁻¹ (UNSCEAR 2000).$ $year⁻¹ (UNSCEAR 2000).$

Conclusion

The activity concentrations of radionuclides $(^{238}$ U, 232 Th, 226 Ra, and 40 K) and a gamma radiation hazard assessment were obtained for 138 soil samples using an HPGe gamma-ray spectrometer. The average activity concentrations of 238 U, 232 Th, and 226Ra in the samples were found to be lower than the worldwide average values, whereas the average activity of 40 K was much higher than the worldwide average value. The radon concentration from the sample locations was within the typical range and can be considered "low risk." A multidetector low-background α/β counting system was used to determine the gross α and β activities in the samples, and the values obtained ranged from 29.72 \pm 1.43 to 1546.64 \pm 115.06 Bq kg⁻¹ and 146.37 \pm 24.95 to 1651.32 \pm 44.15 Bq kg−¹ , respectively. Finally, the radiological hazards associated with the natural radioactivity in the soil samples were estimated.

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Compliance with ethical standards

Conflict of interest The author declares no conflict of interest.

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