Radiological and health hazards resulting from radioactivity and elemental composition of some soil samples

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Abstract
Inspection of the radioactivity level in the soil is very important for human health and environmental protection. This study aims at evaluating the radiological hazards and pollution risks related to natural radionuclides and elements in the selected soil samples. Ten samples of soil were collected from different sites of Aurangabad-India and the level of radioactivity was measured using gamma-ray spectrometry with NaI (Tl) detector. Furthermore, the Physico-chemical properties such as pH, organic matter, electrical conductivity, moisture, soil texture, etc., and elemental composition of soils have been decided on using various standard techniques. The mean concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K were 8.178, 17.408, and 96.496 Bq/kg, respectively, which are lower than the global average values of 35, 30, and 400 Bq/kg, respectively (UNSCEAR, 2000). The radiological hazard indices such as radium equivalent, absorbed dose, annual effective dose, internal index, external index, gamma index, excess lifetime cancer risk, etc., were calculated to assess the radiation hazards and compared with internationally recommended values which found to be lower than the permissibility limits. The Pearson correlation was applied to determine the existing relationship between radionuclides and radiological health hazard parameters, as well as with the physicochemical properties of the soil samples. The major and trace elements presented in soils were measured and their mean concentration was ranked in the formed order (Mg>Na>Ca>K>N>Mn>Fe>P>Zn>Cu). The pollution risk parameters (Geo-accumulation index, contamination factor, degree of contamination, pollution load index, and potential ecological risk index) related to the elements in the samples were assessed and results shown that the soils under study are unpolluted with the measured elements. Generally, the radioactivity levels and pollution risks indices in the soils of the study area are within the permissible safety limits and do not cause any significant health threat to humans. Thus, the presented data provide a general background of the detectable radionuclides for the study area and can be helpful in the future as a reference for more extensive studies in the same field.

Key words: health hazards; radionuclides; gamma spectrometry; physico-chemical properties; absorbed dose.

Introduction
All the materials on the earth contain different amounts of natural radionuclides. Humans are exposed to natural radioactivity because of the primordial radionuclides such as $^{40}$K, $^{232}$Th, and $^{238}$U series that exist in varying amounts in soil, building materials, water, rocks, and the atmosphere. The exposure to the gamma-ray from these radionuclides on the body and the irradiation of lung tissue from inhalation of radon and its daughters cause the radiological effects. Evaluating of gamma-ray dose from natural sources is vital as natural radiation is considered the largest contributor to the external dose of the population [1,2]. Knowing the distribution of natural radioactivity in soil, rock, building materials, and food plays a vital role in protecting the environment, geoscientific research and guiding for the use and management of these materials. Most of the countries within the world implement surveys to evaluate radioactivity levels to collect data regarding possible radiological hazards and to take necessary protection. Radionuclides such as uranium exhibit a serious threat, even at small concentrations due to high chemical toxicity and
radioactivity altogether. Increasing demands have caused higher anthropogenic environmental contamination of such toxic metals along with the natural release. Radionuclides are produced artificially by different industrial activities which include “nuclear reactors, mining, nuclear power plant accidents, laboratory investigations, particle accelerators, power weapon tests, radionuclide generators, nuclear energy activities, etc.” [3-5]. These contaminants get deposited on the ground based on their weight, and later heavy rains can bring the radioactive particle to the ground [6,7]. Intensive surveys and investigations on natural radiations have received considerable attention in many countries (UNSCEAR, 2000) [1]. High radiation levels have been reported in parts of India, Italy, France, and Brazil. In Egypt, along the Nile delta studies show that clay soils have higher radiation activity than sandy soils. In Sri Lanka, a similar study on clay bricks shows low levels of hazards [8]. This subject has not been studied by researchers in Aurangabad, Maharashtra-India, and this research work is to be the first in this area. This study is beneficial to the authority and the people of Aurangabad-India as it provides information and data for the evaluation of the radiation exposure levels associated with soils use in the district. The main objective of this study is to assess health hazards to humans from the radioactivity and elements in the soil samples from the outskirts of Aurangabad, India, as this is a historic region with a documented history and having monuments of world heritage for tens of centuries. Physico-chemical parameters play a major role in the distribution and behavior of radionuclides in the soil. Therefore, the study of Physico-chemical parameters of the surface soil constitutes one of the most important aspects of the present study. It is a well-based incontrovertible fact that any change in the radionuclide content of varied components of the ecosystem depends on the interactions of various number of factors including the radionuclides itself and the location-specific characteristics, like soil type (clay, sand, silt) and its physicochemical properties, climate changes, etc. The chemical composition of the soil, particularly its metal content, is environmentally important because depending on their concentrations, levels of toxic elements can reduce soil fertility, can increase input to the food chain, which leads to accumulating toxic metals in foodstuffs, and ultimately can endanger human health. In the current work, the collected soil samples from Aurangabad, India, were analyzed using a passively shielded NaI(Tl) detector to determine the levels of activity concentrations from $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$. Moreover, Physico-chemical properties and elemental composition of the soil samples have been determined using different standard analytical techniques.

**Materials and Methods**

**Sample collection and preparation**

The soil samples were collected from ten sites of Aurangabad-Maharashtra, India (Figure 1) as this is a historic region with a documented history and having monuments of world heritage for tens of centuries. The study area located between latitudes 19.926436–20.052340°N and longitudes 75.329620–75.188994°E. Soil sampling was conducted during the period of July-September 2017 which corresponds to the rainy season in this region. The samples were collected by using the clean plastic scoop and stored in polyethylene bags. Samples were collected with the only constraint that no sampling site should be taken close to a field boundary, a road, a tree or other obstruction. About 2 kg of the samples were collected from each location. Surface soils were taken from different places within the cleared area from the ground surface up to 5 cm and mixed together thoroughly in order to obtain a representative sample of that area. Samples that are collected from various locations were brought to the laboratory. Organic materials, roots, vegetation, pebbles, etc., if present were removed and the samples were initially sun-dried by spreading them in a tray. After removing the stones, the samples were dried in the furnace at 110°C for 24 hours to remove moisture and ground it into a powder of suitable size. Then samples were sieved through 100 mesh which is the optimum size enriched in heavy mineral. These samples were filled in plastic containers with 7 cm diameter and 10 cm in height. Sample containers were filled with 500 gm of samples for uniformity depending upon the density of the sample. These sample containers were stored for a period of 1 month before gamma spectrometric analysis so as to allow the establishment of a secular equilibrium between $^{226}\text{Ra}$, $^{232}\text{Th}$, and their respective progeny products.

![Figure 1. Locations map around Aurangabad-India, wherefrom samples were collected](image-url)
Radionuclides measurements

The analysis of natural radionuclide concentrations was performed by utilizing gamma-ray spectrometry with NaI (Tl) detector coupled with a multichannel analyzer 8 K channel, with resolution full-width maximum half (FWHM) at 662 keV $^{137}\text{Cs}$ about 40 keV and about 60 keV for the photopeak of $^{60}\text{Co}$ at 1330 keV, relative efficiency at 662 keV $^{137}\text{Cs}$ is 7% and relative efficiency at 1330 keV $^{60}\text{Co}$ is 8%. The detector is shielded during a chamber of two layers starting with stainless steel. This shield work for reducing different background radioactivity. The spectrometer was calibrated for energy and efficiency over the photon energy range 123 to 1330 keV using IAEA standard reference radioactive sources $^{57}\text{Co}$, $^{133}\text{Ba}$, $^{137}\text{Cs}$, $^{54}\text{Mn}$, $^{22}\text{Na}$, and $^{60}\text{Co}$. It has been procured from the Bhabha Atomic Research Centre (BARC), Mumbai-India, which were used for calibrated energy and efficiency. The channel numbers of the photopeaks corresponding to the various gamma energies were recorded after 1200 s and therefore the obtained energy-channel linear relationship is shown in Figure 2. The standard source was put in the same geometry as that used for measured samples. For calibration of a spectrometer, the standard source is put above the detector, and therefore the measurement started. The calibration source was counted for a long time to obtain clear defined photo peaks. The background spectrum was recorded immediately before the sample counting to obtain the net count rate. Each sample was placed in plastic containers of the same size as that of the standard sources. The obtained spectrum was analyzed with the computer software program PHAST-MCS or manually with the utilization of a spreadsheet (Microsoft Excel) to calculate the specific activity of radionuclides. The activity concentration of $^{226}\text{Ra}$ in the samples was evaluated from the photopeaks of $^{214}\text{Pb}$ (295.22, 351.93 keV), $^{214}\text{Bi}$ (609.31, 1120 keV) and $^{228}\text{Ra}$ (186.1 keV). Similarly, the activity concentration of $^{232}\text{Th}$ was calculated through the photo peaks of $^{228}\text{Ac}$ (911.2, 209.25, 338.32 keV) and $^{208}\text{Tl}$ (2614, 583.19, 860.56 keV). The activity of $^{40}\text{K}$ was evaluated from its own photopeak (1461 keV).

The specific activity concentrations of radionuclides are computed using the following equation:

$$A = \frac{N}{\varepsilon \times \gamma \times t \times m}$$  \hspace{1cm} \text{Eq. 3}

where $A$ is the activity concentrations of radionuclides in the sample in Bq/kg, $N$ is the net count rate ($N = N_{\text{peak}} - N_{\text{Background}}$), $\varepsilon$ is the detector efficiency of the photopeak used, $\gamma$ is the absolute transition of gamma decay, $t$ is the counting time in seconds, and $m$ is mass of sample in kg.

Determination of Physico-chemical properties of the soil

Physico-chemical parameters such as pH, electrical conductivity, organic matter content, soil texture (sand, silt, and clay), etc., and elemental composition of the soil samples, were determined using different standard procedures [9,10]. The soil pH and electrical conductivity were measured using a pH meter and EC meter. The pH was measured during a 1:2.5 soil-water ratio. The pH was measured by dipping the pH meter into the solution until a steady value of pH was gained. The instrument measuring of pH was calibrated using the buffer solutions of pH 4, 7, and 9. Also, the same prepared for pH measurement was used for measuring the conductivity in the soil samples. The electrical conductivity of the soil sample was determined by dipping the electrode of the conductivity cell in the soil-water suspension. The EC is measured in milli-Siemens per cm (mS/cm). The organic carbon method was employed in the measurement of organic matter percentage in the soil samples [11]. The particle size distribution (sand, silt, and clay) was determined in the range of less than 2 mm fraction by hand feel method. A small amount of soil sample was taken in hand and enough water added to make a ball. This ball was moved between thumb and forefinger to determine the feel characteristics of sand, silt, and clay. The feel characteristics for sand, silt, and clay were gritty, smooth and sticky respectively. The soil ball was pushed between thumb and forefinger to make a ribbon. The longer the ribbon, the more is the clay content in the soil. The ruler was used to measure the length of the ribbon, and the data were recorded. The size fractions were defined as clay (< 0.002 mm), silt (0.002 to 0.02 mm), fine sand (0.02 to 0.2 mm), and coarse sand (0.2 to 2.0 mm).
Soil texture class was identifying by texture triangle as shown in Figure 3 which presents 12 basic soil textural classes. For example, for a soil to be classified as sand, it should present 85% or more of sand and the silt plus clay not exceeding 15%. A sand clay loam has from 20% to 35% of clay, 45% of sand or more, and less than 28% of silt. Clay soil is characterized by more than 40% of clay and less than 45% of sand and 40% of silt. The elemental composition of the dry soil samples was analyzed by an X-ray fluorescence spectrometer (XRF, SPECTRO XEPOS, AMETEK). Energy-dispersive X-ray fluorescence (EDXRF) is a nuclear analytical technique commonly used in environmental research. The principle work of the X-ray fluorescence technique is based on that when the individual atoms excited by an external energy source, the X-ray photons of a characteristic energy or wavelength will be emitted. Therefore, the elements present in the sample can be determined and quantified by counting the number of photons of each energy emitted from a sample. It requires at least 5 gm of dry fine powder sample. The instrument (SPECTRO XEPOS) was controlled by a menu based X-LAB pro software computer. This instrument characteristics included: 50 watts end-window X-ray tube, up to eight polarization, and secondary targets, automatic sample changer, SSD detection system. Standards used in the XRF peak calibration are as follows: Glass tablets FLX-SP1, Voltage/kV: 38.0, Current/µA: 15.0 for MCA peak calibration and FLX-SP2 along with the global calibration.

**Calculation of radiological hazard parameters**

**Radium equivalent activity (Raeq)**

Radiation exposure is often defined in terms of the many radiological parameters. It is known that radium equivalent activity is one of the extensively used hazard indices. It is defined based on the assumption that 10 Bq/kg of $^{226}$Ra, 7 Bq/kg of $^{232}$Th and 130 Bq/kg of $^{40}$K produce the same gamma dose rate. It was calculated through the following equation [12]:

$$\text{Raeq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_K$$  
Eq. 2

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

**Absorbed gamma dose rate ($D_{abs}$)**

The absorbed dose in air expresses the received dose in the open air from the radiation released from natural radionuclides in the environmental materials. This parameter is a very important quantity for assessment when considering radiation hazard to a biosystem. The absorbed dose rate due to gamma radiations for the uniform distribution of naturally occurring radionuclides was computed using the following relation [13].

$$D_{abs} = 0.462 A_{Ra} + 0.604 A_{Th} + 0.0417 A_K$$  
Eq.3

**Annual effective dose equivalent (AEDE)**

The absorbed dose rate was used to calculate the annual effective dose equivalent received by a member by applying the dose conversion factor of 0.7 Sv/Gy where the occupancy factor for outdoor was 0.2 [13]. It is calculated using the following equation:

$$AEDE = D_R \times 8760 \times 0.7 \times 0.2 \times 10^{-6} \text{ mSv/y}$$  
Eq.4

**External and internal hazard indices ($H_{ex} & H_{in}$)**

The external hazard index ($H_{ex}$) represents the external radiation exposure associated with gamma emission from radioisotopes in the sample. The value of $H_{ex}$ must not exceed the maximum acceptable value than unity to maintain considerable danger. This index can be evaluated using the following equation [14]:

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810) \leq 1$$  
Eq. 5

The internal hazard index ($H_{in}$) is used to control the internal exposure to $^{222}$Rn and its radioactive progeny [15]. The internal exposure to radon and its daughter products is quantified by the $H_{in}$ which is given by the following equation:

$$H_{in} = (A_{Ra}/185) + (A_{Th}/259) + (A_K/4810) \leq 1$$  
Eq. 6

where $A_{Ra}$, $A_{Th}$, and $A_K$ are the activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K, respectively.

**Gamma representative level index ($I_{\gamma}$)**

Gamma index ($I_{\gamma}$) is employed to work out the extent of gamma-ray hazards related to the natural radionuclides in the samples. It is calculated using the following equation [16]:

$$I_{\gamma} = (A_{Ra}/150) + (A_{Th}/100) + (A_K/1500) \leq 1$$  
Eq.7
Annual gonadal dose equivalent (AGDE)
According to UNSCEAR, the gonads, active bone marrow, and bone surface cells are considered the organs of interest. The annual gonadal dose equivalent (AGDE) in mSv/y due to the activity concentrations of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ was calculated using the following formula [17]:

$$\text{AGDE (mSv/y)} = 3.09 A_{\text{Ra}} + 4.18 A_{\text{Th}} + 0.31 A_{\text{K}}$$

\text{Eq. 8}

Alpha index ($I_\alpha$)
This index is used to assess the excess alpha radiation due to radon inhalation originating from building materials. In the present work, ($I_\alpha$) was determined through the following formula [18]:

$$I_\alpha = \left( \frac{A_{\text{Ra}}}{200} \right) \leq 1$$

\text{Eq. 9}

where $A_{\text{Ra}}$ is the activity concentration of $^{226}\text{Ra}$. The recommended maximum concentration of $^{226}\text{Ra}$ is 200 Bq/kg, which gives $I_\alpha = 1$.

Excess lifetime cancer risk (ELCR)
The values of the annual effective dose were used to calculate the excess lifetime cancer risk (ELCR) using the following equation [19]:

$$\text{ELCR} = \text{AEDE} \times \text{DL} \times \text{RF}$$

\text{Eq. 10}

where AEDE is the annual effective dose equivalent, DL is the duration of life (70 years), and RF (Sv$^{-1}$) is a fatal risk factor per Sievert, which is 0.05 as per ICRP-60.

Pollution risk indices

Geo-accumulation index ($I_{\text{geo}}$)
The geo-accumulation index is a common index used for assessing heavy metal pollution in soils and provides an arrangement for the degree of metal enrichment in comparison to the background. It was calculated using the following formula [20]:

$$I_{\text{geo}} = \log_2 \left[ \frac{C_n}{1.5 \times B_n} \right]$$

\text{Eq. 11}

where $C_n$ is the concentration of measured metal (n), $B_n$ is the background concentration of the same metal (n), and 1.5 is a multiplying factor of natural variability.

Contamination factor (CF) and degree of contamination (Cd)
The CF is the ratio gained by dividing the concentration of each metal in the soil by the baseline or background value. The Cd of the environment is, therefore, defined as the sum of contamination factors for all elements examined and determined using the following equation [21]:

$$C_F = C_n/B_n \text{, and } C_d = \sum_{i=1}^{n} CF_i$$

\text{Eq. 12}

where $C_F$ contamination factor, $C_n$ is the measured concentration of metal in the sample, $B_n$ is the background value for the same metal, and $C_d$ is the degree of contamination.

Pollution load index (PLI)
This index is used to evaluate the extent of metal pollution for each soil sample site. The PLI of the study area is calculated by obtaining the n-root from the n-CFs that were obtained for all the metals. The PLI provides a simple and comparative means for assessing soil quality, where a value of PLI < 1 indicates no pollution; PLI = 1 suggests that only baseline levels of pollutants and PLI > 1 indicates pollution and deterioration of soil quality. This index was developed by Tomlinson [22], and calculated with the following equation:

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n}$$

\text{Eq. 13}

where CF is the contamination factor and n is the number of metals studied in the sample.

Potential ecological risk index (RI)
The potential ecological risk index method was proposed by Hakanson [21], to assess the characteristics and environmental behavior of heavy metal contaminants in soils. RI was presented for assessing the degree of heavy metal pollution in soils, in regard with the toxicity of heavy metals and the response of the environment where (RI) is calculated as the total of all risk factors for heavy metals in the selected sample as given by the following equation:

$$E_i = T_i \times CF_i, \text{ and } RI = \sum_{i=1}^{n} E_i$$

\text{Eq. 14}

where $E_i$ is the potential ecological risk factor, CF is the contamination factor, and $T_i$ is the toxic response factor ($Mn = Zn = 1$, and $Cu = 5$).

Results and Discussions

Activity concentrations of radionuclides in the soil
The activity concentrations of primordial radionuclides in the soil samples were measured and presented in Table 1. The activity concentration of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ are in the range from 4.169 to 12.862 Bq/kg, 8.658 to 21.316 Bq/kg, and 40.088 to 130.871 Bq/kg, with a mean value of 8.178, 17.408, and 96.496 Bq/kg, respectively, which are less than that of the global average values of 35, 30, and 400 Bq/kg, respectively (UNSCEAR, 2000). The activity of $^{232}$Th series is to be greater than that of $^{238}$U series ($^{226}\text{Ra}$) in most of the studied soil samples, supportive to the very fact that the presence of thorium is 1.5 times greater than that of uranium in the earth's crust. It had been noted that the activity concentrations of $^{226}\text{Ra}$ and $^{232}\text{Th}$ in the soil samples weren't uniform, which represents the variations within the geological, geochemical and physicochemical characteristics of the soils. It had been also observed that the measured activity of $^{40}\text{K}$ exceeded the values of both $^{226}\text{Ra}$ and $^{232}\text{Th}$, are both the foremost abundant
Radiological health risk assessment

The radiological hazard parameters are calculated from the results of the activity concentrations of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in the soil samples and listed in Table 1. It was found that the radium equivalent activity varying from 29.3789 to 50.6377 Bq/kg, with the mean value of 40.5023 Bq/kg. The maximum value of Raeq is 50.6377 Bq/kg which does not exceed the recommended value of 370 Bq/kg corresponds to a dose limit of 1 mSv for the public population [23, 24]. The absorbed gamma dose rate varies from 13.6399 to 22.8246 nGy/h with a mean value of 18.3169 nGy/h. This value is lower than the recommended international levels of 59 nGy/h. The value of the annual effective dose ranges from 0.0167 to 0.0270 mSv/y with a mean value of 0.0224 mSv/y which is less than the worldwide average value of 0.07 mSv/y. The values of the gamma index were found to range from 0.0794 to 0.1368, with a mean value of 0.1094. These values are much less than the limit value of unity and below the measured values in other countries. The values of $H_{\text{in}}$ were found to range between 0.0939 and 0.1715 with a mean value of 0.1315. The obtained values of $H_{\text{ex}}$ are less than unity, as recommended for construction materials. It is observed that the values of the internal hazard index are greater than the external hazard index as shown in Figure 4.

### Table 1. The activity concentration of natural radionuclides in (Bq/kg) in the soil samples

| Sample ID | Latitude  | Longitude  | $^{226}\text{Ra}$ (Bq/kg) | $^{232}\text{Th}$ (Bq/kg) | $^{40}\text{K}$ (Bq/kg) | Ra/Th | Th/Ra | Th/K | Ra/K |
|-----------|-----------|------------|--------------------------|--------------------------|------------------------|-------|-------|------|------|
| S1        | 19.926436 | 75.329620  | 9.906                    | 20.460                   | 60.508                 | 0.484 | 2.065 | 0.338| 0.164|
| S2        | 19.94103  | 75.313918  | 12.862                   | 20.979                   | 100.983                | 0.613 | 1.631 | 0.208| 0.127|
| S3        | 19.956592 | 75.285846  | 8.280                    | 19.229                   | 120.020                | 0.431 | 2.322 | 0.160| 0.069|
| S4        | 19.966837 | 75.273615  | 7.470                    | 21.316                   | 120.142                | 0.350 | 2.854 | 0.177| 0.062|
| S5        | 19.976981 | 75.268358  | 7.425                    | 18.898                   | 70.668                 | 0.393 | 2.545 | 0.267| 0.105|
| S6        | 20.009619 | 75.261971  | 5.560                    | 19.768                   | 40.088                 | 0.281 | 3.555 | 0.493| 0.139|
| S7        | 20.019780 | 75.223154  | 4.169                    | 12.010                   | 120.061                | 0.347 | 2.881 | 0.100| 0.035|
| S8        | 20.023803 | 75.186384  | 9.992                    | 8.658                    | 90.986                 | 1.154 | 0.866 | 0.095| 0.110|
| S9        | 20.048840 | 75.196791  | 8.313                    | 14.757                   | 110.634                | 0.565 | 1.775 | 0.133| 0.075|
| S10       | 20.052340 | 75.188994  | 7.807                    | 18.007                   | 130.871                | 0.434 | 2.307 | 0.138| 0.060|
| Minimum   | –         | –          | 4.169                    | 8.658                    | 40.088                 | 0.281 | 0.866 | 0.095| 0.035|
| Maximum   | –         | –          | 12.862                   | 21.316                   | 130.871                | 1.154 | 3.555 | 0.493| 0.164|
| Mean Value| –         | –          | 8.178                    | 17.408                   | 96.496                 | 0.505 | 2.280 | 0.211| 0.095|

### Table 2. Radium equivalent activity, absorbed dose rate, annual effective dose, radiation hazard indices, annual gonadal dose equivalent, and excess lifetime cancer risk for soils

| Sample ID | RaEq (Bq/kg) | $D_{\gamma}$ (mGy/h) | AEDE (mSv/y) | $H_{\text{in}}$ (mSv/y) | $H_{\text{ex}}$ (mSv/y) | $I_{\text{g}}$ | AGDE (mSv/y) | $I_{\text{c}}$ | ELCR×10^{-3} |
|-----------|--------------|----------------------|-------------|--------------------------|------------------------|-------|---------------|-------|---------------|
| S1        | 43.8229      | 19.4576              | 0.0239      | 0.1451                   | 0.1184                 | 0.3109| 0.135         | 0.0495| 0.837         |
| S2        | 50.6377      | 22.8246              | 0.0270      | 0.1715                   | 0.1368                 | 0.3629| 0.159         | 0.0643| 0.945         |
| S3        | 45.0190      | 20.4445              | 0.0251      | 0.1439                   | 0.1216                 | 0.3275| 0.143         | 0.0414| 0.897         |
| S4        | 47.2028      | 21.3359              | 0.0262      | 0.1477                   | 0.1275                 | 0.3431| 0.149         | 0.0374| 0.917         |
| S5        | 39.8906      | 17.7916              | 0.0218      | 0.1278                   | 0.1077                 | 0.2856| 0.124         | 0.0371| 0.763         |
| S6        | 36.9150      | 16.1803              | 0.0198      | 0.1147                   | 0.0997                 | 0.2615| 0.112         | 0.0278| 0.693         |
| S7        | 30.5880      | 14.1867              | 0.0174      | 0.0939                   | 0.0826                 | 0.2279| 0.100         | 0.0208| 0.609         |
| S8        | 29.3789      | 13.6399              | 0.0167      | 0.1064                   | 0.0794                 | 0.2139| 0.095         | 0.0499| 0.585         |
| S9        | 37.9343      | 17.3673              | 0.0213      | 0.1249                   | 0.1025                 | 0.2768| 0.122         | 0.0416| 0.746         |
| S10       | 43.6341      | 19.9404              | 0.0245      | 0.1389                   | 0.1178                 | 0.3194| 0.140         | 0.0390| 0.858         |
| Minimum   | 29.3789      | 13.6399              | 0.0167      | 0.0939                   | 0.0794                 | 0.2139| 0.095         | 0.0208| 0.585         |
| Maximum   | 50.6377      | 22.8246              | 0.0270      | 0.1715                   | 0.1368                 | 0.3629| 0.159         | 0.0643| 0.945         |
| Mean      | 40.5023      | 18.3169              | 0.0224      | 0.1315                   | 0.1094                 | 0.2929| 0.128         | 0.0409| 0.7832        |
| Critical value | 370  | 59     | 0.07  | $\leq 1$  | $\leq 1$  | $\leq 1$  | 0.3  | $< 1$ | 0.290×10^{-7} |
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Figure 4. Comparison of external (H<sub>ex</sub>) and internal (H<sub>in</sub>) radiation hazard indices

Table 3. Physico-chemical properties of the investigated soil samples

| Sample ID | pH     | EC (mS/cm) | OM (%) | Moisture (%) | CaCO<sub>3</sub> (%) | Sand (%) | Silt (%) | Clay (%) | Texture class (TC) |
|-----------|--------|------------|--------|--------------|----------------------|----------|----------|----------|-------------------|
| S1        | 7.38   | 0.11       | 0.67   | 2.61         | 2.50                 | 87.63    | 6.49     | 5.8      | Sandy             |
| S2        | 7.71   | 0.14       | 1.45   | 5.49         | 5.00                 | 71.68    | 17.41    | 10.54    | Sandy Loam        |
| S3        | 7.53   | 0.01       | 0.84   | 4.36         | 3.75                 | 85.5     | 5.09     | 8.46     | Sandy             |
| S4        | 7.46   | 0.13       | 1.12   | 3.66         | 5.63                 | 79.21    | 6.81     | 13.1     | Loamy Sand        |
| S5        | 6.61   | 0.08       | 1.28   | 4.82         | 1.25                 | 83.63    | 6.36     | 9.96     | Loamy Sand        |
| S6        | 6.78   | 0.07       | 0.98   | 3.68         | 1.20                 | 85.19    | 5.34     | 9.43     | Sandy             |
| S7        | 6.69   | 0.24       | 0.72   | 10.57        | 6.25                 | 67.44    | 14.22    | 17.61    | Sandy Loam        |
| S8        | 7.17   | 0.14       | 0.47   | 4.53         | 2.63                 | 82.9     | 9.14     | 7.16     | Loamy Sand        |
| S9        | 7.00   | 0.17       | 1.22   | 4.89         | 1.22                 | 80.31    | 10.59    | 9.05     | Loamy Sand        |
| S10       | 6.57   | 0.10       | 0.93   | 3.31         | 2.25                 | 88.28    | 10.96    | 0.08     | Sand              |
| Range     | 6.57–7.71 | 0.01–0.24  | 0.47–1.45 | 2.61–10.57  | 1.20–6.25            | 67.44–88.28 | 5.09–17.41 | 0.08–17.61 | –                 |
| Mean      | 7.09   | 0.12       | 0.97   | 4.79         | 3.17                 | 81.18    | 9.24     | 9.12     | –                 |

Figure 5. Variation of pH, EC, OM, and moisture in the investigated soil samples
The alpha index has been calculated using the activity concentration of $^{226}$Ra in the samples. The estimated alpha index values for the analyzed samples ranged from 0.0208 to 0.0643 with a mean value of 0.0409 that is lower than unity hence, the soil samples are safe for construction of houses. The excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) are considered very important parameters for human health. As seen in Table 2 the AGDE values of soil samples varied between 0.095 mSv/y (S8) and 0.159 mSv/y (S2) with an average value of 0.128 mSv/y. This result is lower than the permissible safety limit of 0.3 mSv/y. The calculated of ELCR for outdoor exposure ranged from 0.585×10$^{-3}$ to 0.945×10$^{-3}$ with an average of 0.783×10$^{-3}$ which is greater than the worldwide recommended value of 0.290×10$^{-3}$ (UNSCEAR). The calculated hazard parameters of all samples are low comparing with the world average and the recommended safe limits. The values of these indices should be lower than unity to keep the radiation hazard insignificant. Building on these results of radiological hazard indices, one can indicate that the investigated soil samples are safe and can be used as building material without causing any significant radiological risk to humans.

### Physico-chemical characterization of the soil samples

The basic Physico-chemical properties like pH, conductivity, organic matter, moisture, etc., of the soil samples, were measured by employing standard methods and the results are listed in Table 3. In the current study, the pH values of soil samples varied in the ranges (6.57–7.71) with mean value 7.09 which are indicating that the analyzed soil samples were neutral. The variation of soil pH is given in Figure 5. The electrical conductivity was measured in term mS/cm and results ranged from 0.01 to 0.24 mS/cm with a mean value of 0.12 mS/cm which means the soil samples under study are non-saline. The electrical conductivity values for the selected samples are less than the other reported studies, which may be due to the absence of expected movable salts [26]. The variation of electrical conductivity in the soil samples given in Figure 5. The organic matter content in soils is medium (average 0.97) and ranges from 0.47% to 1.45%. Natural organic matter (NOM) contains functional groups that can form complexes with metals, and these complexes can affect the physical and chemical properties of the affected metal or radionuclides. Complexation affects metal and radionuclide mobility, adsorption to soils and bioavailability [27]. In sandy soils, organic matter content is low when compared to other types of soil. A calcium carbonate value ranged from 1.20% to 6.25% which reveals that these soil samples are medium calcareous. The percentage of sand ranges from 67.44% to 88.28% with mean value 81.18%, silt ranges from 5.09% to 17.41% with mean value 9.24% and clay ranges from 0.08% to 17.61% with mean value 9.12%. All soil samples of the study area show a high percentage of sand and this means that water can move easily and salinity in nature. Textural classes’ analysis of the studied soil samples has been performed to classify the studied soils into their main types. Following the USDA soil texture classification [28-30], soil samples have these types: sandy, sandy loam and loam sand. The sand is the main soil type in the study area.

### Major and trace elements concentration in the soil

The concentrations of major and trace elements were measured in the dry soil samples and results presented in Table 4. The following elements were detected in the samples: Na, K, Ca, Mg, P, N, Fe, Mn, Zn, and Cu with different concentrations. The concentration of alkali metals (Na and K) varies in the ranges of 11200–88700, and 33.80–2254.5 mg/kg with mean values of 49450, and 455.35 mg/kg, respectively. The average concentration of Na is greater than the recommended world average concentration of 25670 mg/kg, while potassium is less than the recommended worldly average concentration of 28650 mg/kg. The concentration of alkaline earth metals (Ca and Mg) varied in the ranges of 20430–43040, and 43100–82300 mg/kg, with the corresponding mean values of 33568, and 63280 mg/kg, respectively, which is higher than the permissible values of 29450, and 13510 mg/kg, respectively [31]. The concentrations of N and P varied in the ranges of 3.13–30.32, 6.87–81.14, 1.19–7.53, and 0.25–3.22 mg/kg with mean values of 13.11, 26.71, 3.79, and 1.63 mg/kg, respectively. Manganese concentrations in most samples were relatively high comparing to Fe, Zn, and Cu (Figure 6), where the maximum concentration was 81.14 mg/kg at the sample S6 and the minimum concentration was 6.87 mg/kg at the sample S3. The average concentrations of Fe, Mn, Zn, and Cu are very much lower than the recommended world average concentration of 30890, 558, 67, and 25 mg/kg, respectively [32]. This indicated that the concentration of the elements in the study area is not harmful effects on the environment and human health. Among the elements detected, Magnesium had the highest mean concentration in the selected soil samples. From the results obtained in the study, it could be concluded that the concentration of different elements in the investigated soil samples is in the following decreasing order: Mg>Na>Ca>K>N>Mn>Fe>P>Zn>Cu.
Table 4. The concentrations of the major and trace elements in the dry soil samples (mg/kg)

| Sample ID | Na   | K     | Ca | Mg | P     | N   | Trace Elements |
|-----------|------|-------|----|----|-------|-----|----------------|
| S1        | 79400| 162.12| 43040| 54800| 12.58 | 212.70| Fe | 25.53 |
| S2        | 74200| 2254.5| 62520| 70500| 29.03 | 51.25 | Mn | 6.80  |
| S3        | 88700| 98.75 | 33910| 43100| 7.62  | 14.28 | Zn | 3.13  |
| S4        | 11200| 71.29 | 30430| 66600| 6.17  | 30.71 | Cu | 7.06  |
| S5        | 15500| 527.5 | 27830| 70500| 3.39  | 265.25|    | 30.32 |
| S6        | 34350| 33.80 | 20430| 62700| 4.36  | 39.28 |    | 5.67  |
| S7        | 64950| 528   | 42170| 50900| 6.41  | 61.91 |    | 4.14  |
| S8        | 53230| 711.5 | 34030| 82300| 18.99 | 49.22 |    | 16.34 |
| S9        | 44530| 104.34| 42110| 59800| 11.36 | 52.95 |    | 11.56 |
| S10       | 28440| 61.53 | 36110| 71600| 16.27 | 52.38 |    | 21.54 |
| Minimum   | 11200| 33.80 | 20430| 43100| 3.39  | 14.28 |    | 3.13  |
| Maximum   | 88700| 2254.5| 43040| 82300| 29.03 | 265.25|    | 30.32 |
| Mean      | 49450| 455.35| 33568| 63280| 11.62 | 82.15 |    | 11.11 |
| Background values [33] | 9600 | 26600 | 22100 | 15000 | 700   | —   | —   | 47200 |

Table 5. Geo-accumulation index (I_{geo}) for the measured elements in the soil samples

| Sample ID | Na   | K     | Ca | Mg | P     | N   | Trace Elements |
|-----------|------|-------|----|----|-------|-----|----------------|
| S1        | 2.46 | -7.94 | -0.38| 1.28| -6.38 | —   | -11.49 |
| S2        | 2.37 | -4.15 | -0.32| 1.65| -5.18 | —   | -13.34 |
| S3        | 2.62 | -6.66 | -0.03| 0.94| -7.11 | —   | -14.47 |
| S4        | -0.36| -9.13 | -0.12| 1.57| -7.41 | —   | -13.29 |
| S5        | 0.11 | -6.24 | -0.25| 1.65| -8.27 | —   | -11.19 |
| S6        | 1.25 | -10.21| -0.70| 1.48| -7.91 | —   | -13.61 |
| S7        | 2.17 | -6.24 | -0.35| 1.18| -7.36 | —   | -14.06 |
| S8        | 1.89 | -5.81 | 0.04 | 1.87| -5.79 | —   | -12.08 |
| S9        | 1.63 | -8.58 | 0.31 | 1.41| -6.53 | —   | -12.58 |
| S10       | 0.98 | -9.34 | 0.12 | 1.67| -6.01 | —   | -11.68 |
| Mean      | 1.51 | -7.63 | -0.09| 1.47| -6.80 | —   | -12.78 |
| Range     | 0.11–2.62| 4.15–10.21| 0.03–0.70| 0.94–1.87| 5.18–8.27 | —   | 11.19–14.47|

Pollution risk assessment

Noteworthy that, heavy metals have great ecological importance for their toxicity and tendency to accumulate in soils. The geo-accumulation index (I_{geo}), contamination factor (CF) and degree of contamination (Cd), pollution load index (PLI) and potential ecological risk index (RI) were calculated to evaluate the pollution risks due to the elements in the soil samples. For optimal interpretation of geochemical data, an appropriate choice of background values is essential. Many studies use the average shale values or the average crustal abundance data as reference baselines. The geo-accumulation index for the selected samples was calculated and given in Table 5. The values of the geo-accumulation index are ranges as the following: (from 0.11 to 2.62) for Na, (−4.15 to −10.21)
for K, (0.03 to −0.70) for Ca, (0.94 to 1.87) for Mg, (−5.18 to −8.27) for P, (−11.19 to −14.47) for Fe, (−3.97 to −7.54) for Mn, (−4.24 to −6.90) for Zn, and (−4.39 to −8.08) for Cu. The average $I_{\text{geo}}$ values of Na (1.51) and Mg (1.47) suggesting that, the sampling locations are moderately contaminated ($1 \leq I_{\text{geo}} \leq 2$), with these elements. The average contamination degree of these elements decreases in the following order: (Na>Mg>Ca>Zn>Cu>Mn>P>K>Fe). The average values of $I_{\text{geo}}$ are generally noticed lower than unity except for Na and Mg; indicating that the investigated study area is not polluted. Variation in $I_{\text{geo}}$ indices values for the measured elements in the samples is shown in Figure 7.

The contamination factors of the elements obtained in the studied soil samples are given in Table 6. The results of CF are ranges as the following: (1.167–9.239) for Na, (0.001–0.085) for K, (0.924–1.948) for Ca, (2.873–5.487) for Mg, (0.005–0.041) for P, (0.008–0.095) for Mn, (0.013–0.079) for Zn, and (0.006–0.072) for Cu. The average contamination factors of major elements are Na (5.151), Ca (1.519), and Mg (4.219). This indicates moderate contamination and therefore the level of contamination is considerable with these elements. Furthermore, the mean values of CF for elements K, P, Mn, Zn, and Cu in the samples are less than one, therefore; the soil samples could also be classified as lowly contaminated with these elements. Generally, the contamination factors behavior like the geo-accumulation index. The contamination degree ($C_d$) values in sampling points S1 (13.927), S2 (13.918), S3 (13.744), S7 (12.21), and S8 (12.775) suggests that the selected soil samples are considered as considerable contaminate degree and moderate contamination degree for sampling (S4, S5, S6, S9, S10). The average contamination degree ($C_d$) for all elements is 11.030 indicates that the soil samples are considered of moderate contamination degree which indicating anthropogenic pollution. The results of PLI ranged from 0.079–0.257 with a mean of 0.134, thus indicating that the study area is not contaminated. The PLI values for all samples are lower than unity indicating the role of external discrete sources and agricultural activities of soil pollution. The results of pollution risk assessment in the present study indicate not probable environmental pollution, particularly with heavy metals.

Assessment of the ecological risk due to heavy metals in the investigated soils was done and presented in Table 7 and Figure 8. It had been found that the average values of ecological risk factors $E_i$ of Mn, Zn, and Cu are < 40 indicating that soils are a low potential ecological hazard. The mean values of $E_i$ ranked in the formed order (Mn<Zn<Cu). Potential ecological risk index (RI) of Mn, Zn and Cu were less than 95 indicating that these metals posed a low potential ecological risk to the environment. The values of a potential ecological risk index for the selected sample sites can be classified as low risk to the environment. Hence, soils under study area show no risk to the environment due to these metals.

**Correlation analysis**

To evaluate the correlations between activity concentrations of radionuclides and the radiological parameters, as well as with Physico-chemical properties and elemental composition of the soil samples, Pearson linear correlation coefficient was determined. Pearson product-moment correlation coefficients were calculated by the linear fit method. The obtained correlation matrices were presented in Tables 8-10. A positive correlation was observed between radium and thorium with correlation coefficients of ($r = 0.188$) due to $^{226}$Ra and $^{232}$Th occur in the same decay series. $^{40}$K shows that insignificant correlation ($r = – 0.006$), ($r = – 0.196$) with $^{226}$Ra and $^{232}$Th due to potassium occur in different decay series. There exists a very good correlation between $^{226}$Ra, $^{232}$Th with absorbed dose in air with correlation coefficients ($r = 0.525$), ($r = 0.832$). All the calculated radiological parameters have a strong correlation with $^{232}$Th and $^{226}$Ra, while there is a weak correlation with $^{40}$K. This indicates that the natural radioactivity in the soils depends on the concentration of radium and thorium.

**Table 6. Contamination factor (CF), degree of contamination ($C_d$) and pollution load index (PLI) of elements in the investigated soil samples**

| Sample ID | Na   | K    | Ca   | Mg   | P    | Mn   | Zn   | Cu   | $C_d$ | PLI  |
|-----------|------|------|------|------|------|------|------|------|------|------|
| S1        | 8.271| 0.006| 1.948| 3.653| 0.018| 0.012| 0.013| 0.006| 13.927| 0.094|
| S2        | 7.229| 0.085| 1.200| 4.700| 0.041| 0.081| 0.030| 0.052| 13.918| 0.257|
| S3        | 9.239| 0.004| 1.534| 2.873| 0.011| 0.008| 0.056| 0.019| 13.744| 0.105|
| S4        | 1.167| 0.003| 1.377| 4.440| 0.009| 0.018| 0.016| 0.027| 7.057  | 0.079 |
| S5        | 1.615| 0.020| 1.259| 4.700| 0.005| 0.010| 0.020| 0.041| 7.670  | 0.097 |
| S6        | 3.578| 0.001| 0.924| 4.180| 0.006| 0.095| 0.041| 0.038| 8.863  | 0.103 |
| S7        | 6.766| 0.020| 1.908| 3.393| 0.009| 0.015| 0.079| 0.020| 12.21  | 0.144 |
| S8        | 5.545| 0.027| 1.540| 5.487| 0.027| 0.026| 0.051| 0.072| 12.775 | 0.206 |
| S9        | 4.639| 0.004| 1.865| 3.987| 0.016| 0.034| 0.069| 0.061| 10.675 | 0.154 |
| S10       | 2.963| 0.002| 1.634| 4.773| 0.023| 0.015| 0.022| 0.025| 9.457  | 0.098 |
| **Range** | 1.167–| 0.001–| 0.924–| 2.873–| 0.005–| 0.008–| 0.013–| 0.006–| 7.057– | 0.079– |
| **Mean**  | 5.151| 0.017| 1.519| 4.219| 0.017| 0.031| 0.040| 0.036| 11.030 | 0.134 |
### Table 7. Potential ecological risk factor and potential ecological risk index in soils

| Sample ID | Ecological risk factor (E\textsubscript{ir}) | RI |
|-----------|--------------------------------------------|----|
|           | Mn     | Zn     | Cu   |     |
| S1        | 0.012  | 0.013  | 0.03 | 0.055 |
| S2        | 0.081  | 0.030  | 0.26 | 0.371 |
| S3        | 0.008  | 0.056  | 0.095| 0.159 |
| S4        | 0.018  | 0.016  | 0.135| 0.169 |
| S5        | 0.010  | 0.020  | 0.205| 0.235 |
| S6        | 0.095  | 0.041  | 0.190| 0.326 |
| S7        | 0.015  | 0.079  | 0.100| 0.194 |
| S8        | 0.026  | 0.051  | 0.360| 0.437 |
| S9        | 0.034  | 0.069  | 0.305| 0.408 |
| S10       | 0.015  | 0.022  | 0.150| 0.187 |
| Mean      | 0.031  | 0.040  | 0.183| 0.254 |

### Table 8. The Pearson correlation between radioactive variables in soil samples

| Variables | \(^{226}\text{Ra}\) | \(^{232}\text{Th}\) | \(^{40}\text{K}\) | D\textsubscript{ab} | Ra\textsubscript{eq} | H\textsubscript{a} | H\textsubscript{ex} | I\textsubscript{γ} | AEDE | ELCR | I\textsubscript{α} |
|-----------|---------------------|---------------------|-----------------|----------------|----------------|----------------|----------------|----------------|------|------|----------------|
| \(^{226}\text{Ra}\) | 1.00                | –                   | –               | –              | –              | –              | –              | –              | –    | –    | –              |
| \(^{232}\text{Th}\) | 0.188               | 1.00                | –               | –              | –              | –              | –              | –              | –    | –    | –              |
| \(^{40}\text{K}\) | -0.006              | -0.196              | 1.00            | –              | –              | –              | –              | –              | –    | –    | –              |
| D\textsubscript{ab} | 0.525               | 0.832               | 0.249           | 1.00           | –              | –              | –              | –              | –    | –    | –              |
| Ra\textsubscript{eq} | 0.512               | 0.88                | 0.163           | 0.996          | 1.00           | –              | –              | –              | –    | –    | –              |
| H\textsubscript{a} | 0.708               | 0.774               | 0.133           | 0.970          | 0.969          | 1.00           | –              | –              | –    | –    | –              |
| H\textsubscript{ex} | 0.513               | 0.875               | 0.163           | 0.999          | 1.00           | 0.973          | 1.00           | –              | –    | –    | –              |
| I\textsubscript{γ} | 0.491               | 0.850               | 0.242           | 0.999          | 0.997          | 0.960          | 1.00           | 1.00           | –    | –    | –              |
| AEDE       | 0.486               | 0.841               | 0.259           | 1.00           | 0.993          | 1.00           | 0.996          | 0.998          | 1.00 | –    | –              |
| ELCR       | 0.486               | 0.840               | 0.259           | 0.997          | 0.992          | 0.956          | 0.993          | 0.997          | 0.999 | 1.00 | –              |
| I\textsubscript{α} | 0.999               | 0.190               | -0.005          | 0.526          | 0.513          | 0.709          | 0.514          | 0.493          | 0.487 | 0.488 | 1.00 |

### Table 9. Correlation coefficient matrix of radionuclides and Physico-chemical properties

| Variables | \(^{226}\text{Ra}\) | \(^{232}\text{Th}\) | \(^{40}\text{K}\) | Ra\textsubscript{eq} |
|-----------|---------------------|---------------------|-----------------|----------------|
| pH        | 0.70                | 0.33                | 0.11            | 0.57           |
| EC        | -0.16               | -0.54               | 0.29            | -0.43          |
| Organic matter | 0.23             | 0.58                | 0.05            | 0.60           |
| Moisture  | -0.42               | -0.50               | 0.32            | -0.47          |
| Sand (%)  | 0.08                | 0.25                | -0.33           | 0.13           |
| Silt (%)  | 0.30                | -0.25               | 0.42            | 0.03           |
| Clay (%)  | -0.38               | -0.13               | 0.05            | -0.23          |
| Na        | 0.37                | -0.10               | 0.05            | 0.06           |
| K         | 0.64                | 0.02                | 0.03            | 0.25           |
| Ca        | -0.06               | -0.41               | 0.42            | -0.24          |
| Mg        | 0.37                | -0.23               | -0.12           | -0.11          |
| P         | 0.83                | -0.08               | 0.20            | 0.28           |
| N         | 0.06                | 0.18                | -0.52           | 0.01           |
| Fe        | 0.19                | 0.02                | -0.31           | -0.02          |
| Mn        | 0.17                | 0.22                | -0.46           | 0.10           |
| Zn        | -0.37               | -0.68               | 0.30            | -0.62          |
| Cu        | 0.33                | -0.49               | -0.05           | -0.33          |
The correlation between physicochemical properties and radionuclides in soils were calculated and presented in Table 9. It can be noted that all the radionuclides studies in the present study are positively correlated with the pH and organic matter content of the soil. The positive sign on all the correlation coefficient values indicates that higher concentrations of radionuclides occur at higher pH values. Where strong correlation between pH and Ra showed a positively significant correlation with Na, K, Ca, Mg, and P, respectively, were observed between Ra, Ra, and Ra activities with organic matter content in the soil samples. This suggests that higher activity concentration occurs at higher organic matter content and indicates the sequestering adsorption of these radionuclides on organic matter. Radium is positively correlated with silt (r = 0.30), sand (r = 0.08) and negatively correlated with clay (r = 0.28). Similarly, thorium is positive correlated with sand (r = 0.25), and negatively correlated with silt (r = 0.05), clay (r = 0.05), and negatively correlated with sand (r = 0.05). The best correlation obtained between silt content of the soil samples is significantly correlated with Na, K, Ca, Mg, P, Mn, and Zn which suggests that Cu is associated mostly with the mineral phase in the soils. The soil pH did not show a significant correlation with Ca, Mg, N, Mn, Zn, and Cu. This indicates that increasing pH reduces the availability of Ca, Mg, Fe, Mn, Zn, and Cu. Electrical conductivity showed a positive correlation with K, Ca, and Zn with significant correlation coefficients, however, it was a non-significant correlation with all other elements. This indicates that a decrease in soluble salts in the soils and no dilution effects of minerals. The organic matter content was a significant correlation with Na, K, Ca, and Zn at the significance level. Generally, the positive correlation analysis indicates that concentrations of radium and thorium mainly contribute to radioactive pollution in the soil, while the negative correlation among the variables indicates that physicochemical properties do not contribute in radioactive soil pollution of the study area.

The elemental concentrations were tested for any correlation with the physicochemical properties of the soils and presented in Table 10. The higher correlation coefficient (r) values suggest that chemical and physical factors control the element associations in the parent material and the soil-forming processes. Most of the elements were significantly correlated with each other, except for Nitrogen. The copper in the soil samples is significantly correlated with K, Ca, Mg, P, Mn, and Zn which suggests that Cu is associated mostly with the mineral phase in the soils. The soil pH did not show a significant correlation with Ca, Mg, N, Mn, Zn, and Cu. This indicates that increasing pH reduces the availability of Ca, Mg, Fe, Mn, Zn, and Cu. Electrical conductivity showed a positive correlation with K, Ca, and Zn with significant correlation coefficients, however, it was a non-significant correlation with all other elements. This indicates that a decrease in soluble salts in the soils and no dilution effects of minerals. The organic matter content was a significant correlation with Na, K, Ca, and Mn. This is because of the strong absorption of the transition metals by the soil organic matter. There exists strong positive correlation between silt, K (r = 0.74) and silt, P (r = 0.69).

Conclusion

The concentrations of primordial radionuclide and physicochemical properties in the soil samples have been determined using gamma-ray spectrometry and by different analytical techniques. The radioactivity level has been found

| Variables          | Na     | K      | Ca     | Mg     | P      | N      | Fe     | Mn     | Zn     | Cu     |
|--------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| Na                 | 1.00   |        |        |        |        |        |        |        |        |        |
| K                  | 0.32   | 1.00   |        |        |        |        |        |        |        |        |
| Ca                 | 0.38   | -0.27  | 1.00   |        |        |        |        |        |        |        |
| Mg                 | -0.54  | 0.34   | -0.37  | 1.00   |        |        |        |        |        |        |
| P                  | 0.39   | 0.76   | 0.02   | 0.45   | 1.00   |        |        |        |        |        |
| N                  | -0.13  | -0.01  | 0.11   | 0.07   | -0.22  | 1.00   |        |        |        |        |
| Fe                 | -0.32  | -0.12  | 0.16   | 0.42   | -0.01  | 0.82   | 1.00   |        |        |        |
| Mn                 | 0.01   | 0.44   | -0.67  | 0.22   | 0.30   | -0.31  | -0.40  | 1.00   |        |        |
| Zn                 | 0.36   | -0.04  | 0.33   | -0.38  | -0.11  | -0.46  | -0.57  | 0.02   | 1.00   |        |
| Cu                 | -0.21  | 0.40   | -0.28  | 0.66   | 0.40   | -0.20  | -0.01  | 0.39   | 0.28   | 1.00   |
| pH                 | 0.53   | 0.41   | -0.04  | -0.15  | 0.46   | -0.24  | -0.38  | 0.12   | -0.16  | -0.01  |
| EC                 | -0.02  | 0.26   | 0.47   | 0.12   | 0.19   | -0.08  | -0.15  | -0.02  | 0.43   | 0.21   |
| Organic matter     | -0.33  | 0.40   | -0.45  | 0.12   | 0.12   | 0.08   | -0.03  | 0.39   | -0.25  | 0.22   |
| Moisture           | 0.21   | 0.29   | 0.26   | -0.26  | -0.08  | -0.13  | -0.39  | -0.06  | 0.70   | 0.03   |
| Sand (%)           | -0.18  | -0.58  | -0.08  | 0.09   | -0.20  | 0.23   | 0.52   | -0.19  | -0.48  | -0.16  |
| Silt (%)           | 0.24   | 0.74   | 0.18   | 0.20   | 0.69   | -0.25  | -0.23  | 0.25   | 0.28   | 0.29   |
| Clay (%)           | 0.03   | 0.21   | -0.05  | -0.31  | -0.33  | -0.07  | -0.54  | 0.10   | 0.44   | 0.00   |
| CaCO₃              | 0.27   | 0.36   | 0.15   | -0.24  | 0.16   | -0.37  | -0.57  | -0.13  | 0.16   | -0.30  |
below the permissible limits of 35, 30, and 400 Bq/kg for $^{226}$Ra, $^{232}$Th, and $^{40}$K respectively (UNSCEAR, 2000). The difference observed in the activities from $^{226}$Ra, $^{232}$Th and $^{40}$K in the samples, maybe due to the different soil physicochemical properties, such as pH, organic matter, soil texture, geological formation, moisture, and geochemical composition for soil samples. The average value of Raeq is 40.5023 Bq kg$^{-1}$ which is below the recommended safe limit value of 370 Bq kg$^{-1}$ for the public. The absorbed dose rate in air is well below the permissible limits ($56 \text{ nGy h}^{-1}$). The calculated annual effective dose is lower than the average world recommended level of 1 mSv y$^{-1}$ for the individual member of the public. The values of the health hazard indices such as external hazard index, internal hazard index, gamma index and alpha index have been estimated and found to be less than the recommended safe levels i.e. unity., thus quite safe from radiation protection point of view and thus can be well utilized for the construction material without causing any significant radiological threat to the population. The concentration of different elements in the soils was measured as well. All elements were below the environmental background values, except Na, Ca, and Mg, while, their mean concentration was as following order: Mg>Na>Ca>K>N>Mn>Fe>P>Zn>Cu, respectively.

Correlation analysis was performed to quantify the dependence of radionuclide activity concentrations with soil variables. The results of the correlation analysis showed varying trends of linear relationships between the natural radionuclides and the elemental compositions of the soils.

The geo-accumulation index ($I_{\text{geo}}$), contamination factor (CF), degree of contamination (Cd), pollution load index (PLI), and potential ecological risk index (RI) were calculated to determine the contamination of elements in the soils. The geo-accumulation index values showed that soil samples were uncontaminated with the measured elements. Based on the contamination factor and contamination degree, the study area belongs to a moderate contamination degree for major elements and low contamination degree for elements. The pollution load index is lower than unity which indicates the status of no clear pollution. The potential ecological risk index values for the selected samples follow the classification of low contamination risk to the environment. The obtained results from the pollution indices ($I_{\text{geo}}$, CF, Cd, PLI, and RI) showed that the soils of the study area have minimal contamination by elements and therefore, do not pose significant ecological risk. Generally, the results showed minimal contamination of the soils by elements; however, community-based awareness on the issue of environmental contamination, its health impacts, and possible prevention, is advisable. The present study suggests that these indices are useful tools for the identification of anthropogenic sources of soil contamination. The soil samples used in the present study are exempted from all the restrictions concerning radioactivity thus, these soil samples are safe to be used for building construction.

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