World Journal of Nuclear Science and Technology, 2015, 5, 120-128
Published Online April 2015 in SciRes. http://www.scirp.org/journal/wjnst
http://dx.doi.org/10.4236/wjnst.2015.52012

A Comparison Study of Soil Samples from Sinai Province in Egypt by Using X-Ray Diffraction and Gamma-Ray Analysis

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Received 14 March 2015; accepted 24 April 2015; published 29 April 2015

Abstract

Ten soil samples from Jabal Al Qur, Wadi Baba, and Wadi Sieh in Sinai, Egypt, were analyzed by XRD spectroscopy. The XRD spectroscopy results indicate that the major, minor and trace constituents varied from one sample to another. Samples were also analyzed by HPGe gamma spectrometer to determine the activity concentration of U-238, Th-232 series and K-40. The concentrations for ^{238}U ranged from 57.03 to 4220.41 Bq/kg with an average 1110.75 Bq/kg, for ^{232}Th, ranged from 13.55 to130.46 Bq/kg with an average 71.85 Bq/Kg. The concentrations for ^{40}K were in the range from 12.18 to 948.93 Bq/kg with an average value 457.09 Bq/kg. The average activity concentration values of ^{226}Ra, ^{232}Th, and ^{40}K, in all the collected samples were higher than the world average. The radium equivalent (R_{eq}), absorbed dose rate (D_R), the effective dose rate (D_{eff}), and hazard indices resulted due to the natural radionuclides in soil are also calculated. The Results show that the study area is not safe for human and environments.

Keywords

Sinai Soil, X-Ray Diffraction, Natural Radioactivity Concentration, Chemical Constituent, Diffract Meter

1. Introduction

Measurement of the concentrations of natural radionuclides in soil give information's on the natural radionuclide sources. Beck suggested that 50% - 80% of the total gamma flux at the earth's surface arises from ^{40}K and ^{232}Th, ^{238}U series [1]. Knowledge of the distribution of these radionuclides in the environment is essential in the sense of controlling radiation levels [2]. Further, data on natural radiation are important for designing rules and

How to cite this paper: Baz, S.S. (2015) A Comparison Study of Soil Samples from Sinai Province in Egypt by Using X-Ray Diffraction and Gamma-Ray Analysis. *World Journal of Nuclear Science and Technology*, **5**, 120-128.
http://dx.doi.org/10.4236/wjnst.2015.52012

regulations for radiation protection purposes [3]. The variation in data for various locations is due to the differences in the geology of sampling sites [3]. So several authors have studied the levels of natural background radiation by analysis of radionuclide concentration in soil samples, see e.g.: [4]-[9]. These studies pointed out that the effective gamma radiation levels were generally in the range of 10 - 200 $nGyh^{-1}$ with a mean of 60 $nGyh^{-1}$ [10].

X-Ray Diffraction (XRD) is a non-destructive analytical technique. When X-ray strikes on a crystal surface at an angle θ, a portion of the X-ray is scattered by a layer of atoms at the surface. Un-scattered X-ray penetrates to the second layer of atoms where again a fraction is scattered. The remaining X-ray passes to the third layer until the energy of the X-ray is dissipated completely. An X-ray detector measures the cumulative effect of the scattering beams. The X-ray appear to be reflected from the crystal only if the angle of incidence satisfies the condition of Bragg Equation, $n\lambda = 2d_{hkl}\sin\theta$. The X-ray pattern shows the peaks at various angles of incidence [11].

The objectives of the present study are to determine some metal concentrations and measure the natural radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K in the soil samples taken from selected places in Sinai region, Egypt, also to assess the radiological Hazard indices in air and to compare the results with international levels. Radionuclides ^{226}Ra, ^{232}Th and ^{40}K were measured with a well-type gamma-ray detector, and metal concentrations were determined by X-Ray diffraction, patterns were recorded using X'Pert PRO Powder X-Ray Diffraction with Cu Kα radiation (λ = 0.1542 nm), Ni-filter and general area detector. The data generated in this study provide baseline values of natural radioactivity in Sinai soils and may be useful for authorities in the implementation of radiation protection standards for the general population in the country as well as to plan and conduct further studies on this issue.

2. Analytical Technique

2.1. Study Area

Sinai can be divided into three areas: The northern region consists of sand dunes and fossil beaches formed by the changing levels of the Mediterranean Sea during the glacial periods two million years ago. The landscape is flat and uniform, interrupted by sand and limestone hills. The central area with limestone dating from the Tertiary Period is the highlands extend towards the south. The third area consists of granite and volcanic rocks. Limestone and sandstone sediments are replaced by granite and basalt. Both rocks are produced by volcanic activity on the bottom of the ocean. Ten soil samples were collected from different areas of Sinai; Jabal Al Qur (28˚49'46.06"N - 33˚17'45.41"E) is located inland south-east of Sinai and consists of granite and volcanic rocks, Limestone and sandstone sediments. Wadi Baba (28˚58'30.80"N - 33˚17'31.47"E) is a low altitude system located inland south-east of Wadi Gharandel. The system consists of several smaller interconnecting wadis with gravel beds and high rise sandstone cliffs either side. Wadi Sieh (29˚03'27.28"N - 33˚26'14.84"E) is located further inland north-east of Wadi Baba; the wadi has a sandy bed with sandstone canyons rising either side, as shown in **Figure 1**, (www.bedawi.com/Sinai_EN.html).

2.2. Sample Collection, Preparation and Measuring Methods

Ten soil samples were collected from different areas of Sinai; Jabal Al Qur, Wadi Baba and Wadi Sieh. These collected samples have different Descriptions as shown in **Table 1**. Samples were grounded and passed through a 1 mm sieve and dried to 95˚C for 24 hours in order not to lose the volatile polonium or cesium. Ten gm of the dried samples were analyzed by XRD model X'Pert PRO powder diffract meter equipped with Cu anode, for the chemical and mineral composition. The X-ray source used in this research has a wavelength of 1.540562 Å. The diffract grams were recorded in the 2θ range of 0.5˚ - 70˚ with step size of 0.02 Å and a step time of 0.60 s. For radiometric analysis, the dried fine grained samples were packed in polyethylene Marinelli beaker for gamma spectroscopy and then stored for four weeks to reach secular equilibrium between radium-226 and thorium-232 and their progenies. The samples were analyzed non-destructively, using gamma-ray spectrometry with Canberra high purity germanium (HPGe) coaxial detector with relative efficiency of 25% and FWHM 2.0 keV at 1332 keV of ^{60}Co. Genie 2000 basic spectroscopic software was installed in the computer for data acquisition and analysis. The system was calibrated for energy and absolute efficiency. The measurements were done for a time period of 82,800 sec. An empty polyethylene Marinelli beaker was placed in the detection system for the same time period of measurement, in order to collect the background count rates. Then, each sample was measured during the same accumulating time.

Figure 1. Location map of the collected samples.

Table 1. Description of collected samples.

Sample Code	Description
Soil 1	Sandy dolostone, medium hard to hard, grey.
Soil 2	Siltstone with ferruginous Shale, mainly red and grey.
Soil 3	Sandstone, red, medium hard with pebbles.
Soil4	Gibbsite, soft, brownish yellow.
Soil 5	Gibbsite-bearing shale, soft to medium hard, yellowish brown.
Soil 6	Siltstone, soft to medium hard, brown.
Soil 7	Gibbsite, soft, brown.
Soil 8	Ferruginous siltstone soft to medium hard.
Soil 9	Conglomeratic sandstone, pale brown, medium hard.
Soil 10	Clay stone, creamy, soft to medium hard.

3. Calculations

3.1. Activity Concentrations

The concentration of ^{226}Ra was determined from the average concentration of gamma-ray lines of energies 351.87 keV of ^{214}Pb and 609.31 keV of ^{214}Bi (since there is secular radioactivity equilibrium in ^{226}Ra series). Also, the concentration of ^{232}Th, which it is in secular radioactivity equilibrium with its short half-life daughters, was determined from the average concentrations of ^{228}Ac (with gamma-ray line 911.16 kev) and of ^{208}Tl (with gamma-ray line 583.10 keV). The analysis of ^{40}K concentrations was based on its single peak in the spectrum at energy 1460.80 kev. The activity concentrations "As" of the natural radionuclides in the measured samples were computed using the following relation [12]:

$$\mathrm{As}\left(\mathrm{Bq}/\mathrm{kg}\right) = \mathrm{Ca}/\varepsilon \mathrm{PrMs} \tag{1}$$

where: Ca is the net gamma counting rate (counts per second), ε the detector efficiency of the specific γ-ray, Pr the absolute transition probability of Gamma-decay and Ms the mass of the sample (kg).

3.2. Radiological Hazard Indices Radium Equivalent Activity Ra_{eq} (Bq/kg)

To assess the real activity level of ^{226}Ra, ^{232}Th and ^{40}K in soil, a common radiological index has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg can be used, provides a very useful guideline in regulating the safety standards in radiation protection for a human population. The index was calculated through the following formula is based on the assumption that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate [10]:

$$Ra_{eq}\left(Bq/kg\right) = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad (2)$$

where: C_{Ra}, C_{Th} and C_K are the specific activities (Bq/kg dry weight) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

3.3. Absorbed Dose Rate and Annual Effective Dose

The measured activity of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses by applying the factors 0.4551, 0.5835 and 0.0429($nGyh^{-1}/Bqkg^{-1}$) for radium, thorium and potassium, respectively. These factors were used to calculate the total absorbed gamma dose rate in air at 1 m above the ground level using the following equation [10]:

$$D_R\left(nGy/h\right) = 0.4551C_{Ra} + 0.5835C_{Th} + 0.0429C_{K} \quad (3)$$

where: C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv/Gy, which is used to convert the absorbed dose rate to annual effective dose with an outdoor occupancy of 20% [10]:

$$D_{eff}\left(mSv/y\right) = D\left(nGy/h\right)\times 8760\left(h/y\right)\times 0.7\left(Sv/Gy\right)\times 0.2\times 10^{-6} \quad (4)$$

3.4. External Hazard Index and Internal Hazard Index

In the literature a number of criterion formulae have been derived over the years to assess the radiation dose rate due to exposure to gamma radiation from the natural radionuclides contained in soil. To limit the annual external gamma-ray dose to 1.5 Gy for the samples under investigation [10], the external hazard index (H_{ex}) is given by the equation:

$$H_{ex} = \left(C_{Ra}/370 + C_{Th}/259 + C_K/4810\right) \quad (5)$$

where: C_{Ra}, C_{Th} and C_K are the activity concentrations in Bq/kg of ^{226}Ra,^{232}Th and ^{40}K Respectively. The value of this index must be less than unity for the radiation hazard to be negligible.

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index (H_{in}) [10], which is given by the equation:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \quad (6)$$

where: C denotes the respective specific activity in Bq/kg, for the safety, H_{in} should be less than unity.

3.5. Representative Level Index ($I_{\gamma r}$)

Representative level index ($I_{\gamma r}$) is used to estimate the level of γ-radiation hazard associated with the natural radionuclides in specific building materials, is defined as [4] [10]:

$$I_{\gamma r} = C_{Ra}/150 + C_{Th}/100 + C_k/1500 \quad (7)$$

where: C_{Ra}, C_{Th} and C_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/k, respectively.

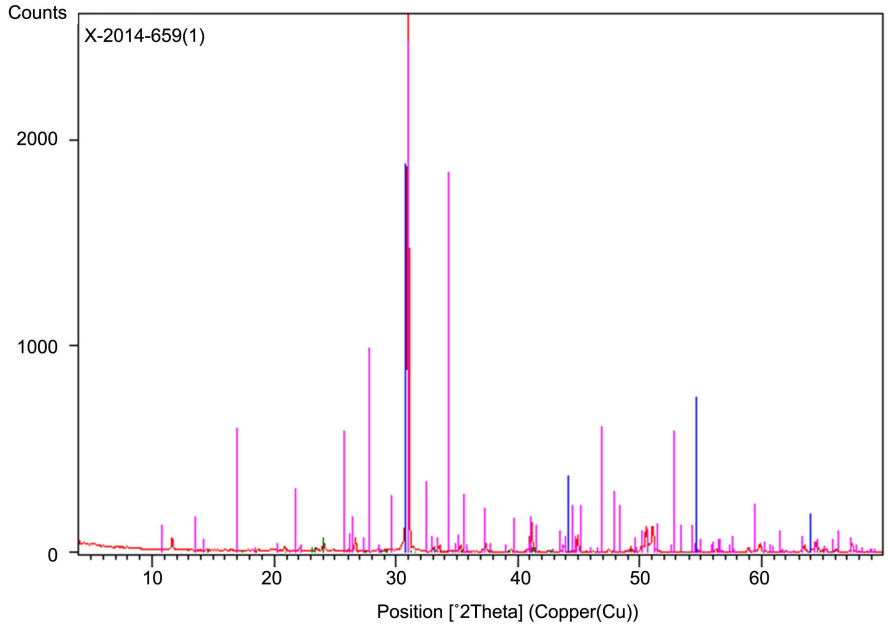
4. Results and Discussion

4.1. XRD Analysis

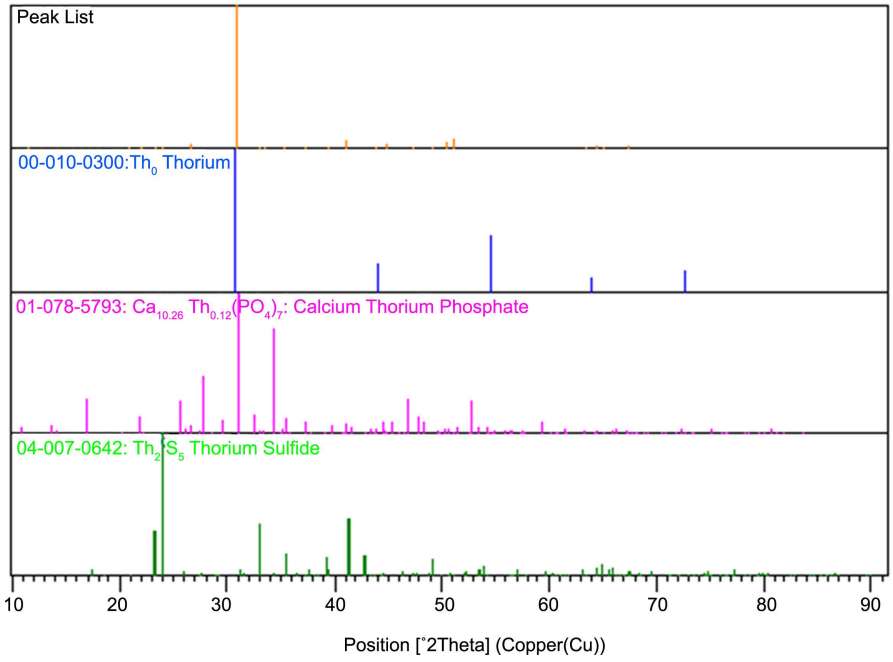
X-ray diffraction is a non-destructive analytical technique, which provides detailed information about the atomic structure of crystalline substances, chemical composition, and physical properties of materials. In the present

study, the XRD results indicate that the main major, minor and trace constituents varied from one sample to another, as shown in **Figure 2(a)**, **Figure 2(b)**, and in **Table 2**. The major element defines the samples and has the highest concentrations. In sample soil1, Calcium Thorium Phosphate is the major in XRD spectrum **Figure 2(a)**, **Figure 2(b)**. While, trace elements occur in small concentrations (usually measured in ppm). They do not change the essence of what a material is and minor elements are in between major and trace elements. XRD spectrum **Figure 2(a)**, **Figure 2(b)**, shows that Thorium (V) sulfide and Thorium are minor and trace elements in sample 1 respectively.

Table 3 is a review of the chemical composition of each mineral and its description which reflects the condition of its formation [13].



(a)



(b)

Figure 2. (a) XRD spectrum of sample soil 1; (b) XRD measurement of sample soil 1.

Table 2. The compound Name of mineral constituents of 10 samples analyzed by XRD spectrometer.

Sample code	MAJOR	MINOR	TRACE
Soil 1	Calcium Thorium Phosphate (Whitlockite-type)*	thorium(V) sulfide	Thorium
Soil 2	Thorium Oxide Phosphate Thorium germanide	Sodium Beryllium Thorium Fluoride Na Cesium Thorium Fluoride	Thorium
Soil 3	Uranium Oxide Sulfide Uranium Fluoride	Uranium Arsenic	Uranium
Soil 4	Uranium Imide	Copper Uranium Oxide	Uranium
Soil 5	Thorium Rhodium Boride	Uranium Ytterbium Sulfide Oxide Nickel Vanadium Uranium Oxide Hydrate	Thorium
Soil 6	Thorium Selenium Uranium Boron Chloride Urea Hydrate	Thorium Carbide Uranium Phosphoryl Chloride	Thorium
Soil 7	Uranium Imide Chloride	Thorium Arsenic	Uranium Uranyl Sulfate Hydroxide Hydrate (Uranopilite)
Soil 8	-----	-------	Uranium Thulium Thorium Iodide
Soil 9	Nickel Vanadium Uranium Oxide Hydrate (Meta-autunite)	Uranium Arsenide Sulfide Uranium Fluoride Hydrate	Uranium
Soil 10	Uranium Chromium Selenium (Hidalgoite)	Aluminum Uranium	Uranium Barium Uranium Gadolinium Oxide

*[14].

Table 3. The Compound name/Chemical composition and description.

Compound name/Chemical composition	Description
Calcium Thorium Phosphate (Whitlockite-type) $Ca_{10.26}Th_{0.12}(PO_4)_7$	Synthesis and new crystal structure refinement.
Thorium(V) sulfide-Th_2S_5	An inorganic chemical compound composed of two thorium atom ionically bonded to five atoms of sulfur.
Thorium-Th	Chemical element and radioactive actinide metal.
Thorium Oxide Phosphate-$Th_2(PO_4)_2O$	The compound was synthesized under wet hydrothermal conditions.
Thorium germanide-Th_3Ge_2	Compound composed of Th(rare earth), Ge(Metalloid).
Sodium Beryllium Thorium Fluoride-$NaBeTh_3F_{15}$	An inorganic chemical compound with one Na, and Be , three Th atom ionically bonded to fifteen F.
Cesium Thorium Fluoride-$CsTh_6F_{25}$	Compound composed of Cs(Alkali Earth), Th(rare earth), and F (Metalloids).
Uranium Oxide Sulfide-UOS	Compound composed of U (rare earth), O (Non-Metals), and S (Non-Metals).
Uranium Fluorid-UF_4	UF_4 is a solid composed of particles with a texture and soluble in water.
Uranium Arsenic-U_3As_4	Compound composed of U (rare earth), and As ((Metalloid).
Uranium-U	Uranium is heavy metal. In nature, uranium is found as an xide.
Copper Uranium Oxide-CuU_3O_{10}	Compound composed of Cu (Transition Metals), U(rare earth), and O (Non-metal)
Thorium Rhodium Boride Th Rh_4B_4	This compound composed of Th, Rh, B.
Uranium Ytterbium Sulfide Oxide-$U_2YbS_3O_2$	Compound composed of U, Yb, S, O
Nickel Vanadium Uranium Oxide Hydrate $Ni(VUO_6)_{24}H_2O$	This compound contains U (RARE EARTH), Ni and V (transition Metals), H, O ((Non-metal).
Thorium Selenium-Th_7Se_{12}	Seven atoms of Th and twelve atoms of Se bonded.
Uranium Boron Chloride Urea Hydrate $U(B_{12}Cl_{12})_2{\cdot}8Co(NH_2)_2{\cdot}2H_2O$	Compound composed of U, B, Cl, Co, N, H, AND O.

Continued

Thorium Carbide-ThC_2	Thorium carbide structure.
Uranium Phosphoryl Chloride-$U_3(PO)_6Cl_{32}$	Compound composed of U, P, O, Cl.
Thorium Arsenic-ThAs	Compound composed of Th (rare earth) and As (Metalloid).
Uranium Imide Chloride-U(NH)Cl	Compound composed of U and (NH)Cl (Volcanic fumaroles, burning coal seams and quano deposits).
Uranyl Sulfate Hydroxide Hydrate (Uranopilite) $(UO_2)_6(SO_4)(OH)_{10}{\cdot}12H_2O$	Secondary mineral found on altering uraninite.
Thorium Thulium Iodide-Th TmI_6	Compound composed of Th (rare earth), Tm (rare earth), and I (Metalloids).
(Meta autunite)-$Ca(UO_2)_2(PO_4)_2{\cdot}4(H_2O)$	Associated with autunite in fractures in uraniferous igneous rocks.
Uranium Arsenide Sulfide-U_2AsS	Compound composed of U (rare earth), As (Metalloid), and S (Non-Metal).
Uranium Fluoride Hydrate-$UF_4(H_2O)_{0.7}$	Uranium tetrafluoride (UF4) and water vapor.
Uranium Chromium Selenium-$UCrSe_3$	Compound composed of U (rare earth), Cr (Transition Metal), and Se (Non-Metal).
(Hidalgoite)-$PbAl_3AsO_4SO_4(OH)_6$	Secondary mineral of the oxide zone of polymetallic sulfide deposits.
Aluminum Uranium-UA_{l3}	Inorganic compound composed of U and Al.
Barium Uranium Gadolinium Oxide-$Ba_2UGdO_{5.944}$	Chemical compound composed of Be, U, Gd, and O.

4.2. Gamma Analysis

4.2.1. Radionuclide Activity Concentration

Table 4 represents the concentrations in Bq/kg of the different radionuclides in the samples. The radioactivity of the samples, as shown in **Table 4**, for Ra-226 ranged from 57.03 Bq/kg (sample 3) to 4220.41 Bq/kg (sample 2) with average value 1104.14 Bq/kg. Meanwhile; for Th-232 series the lowest value was13.55 Bq/kg (sample 5) and the highest value was 130.46 Bq/kg (sample 10) with an average value 74.21 Bq/kg, for K-40 activities ranged from 12.18 Bq/kg (sample 3) to 948.93 Bq/kg (sample 1) with average value 455.80 Bq/kg. In general, all the existed results were higher than the given values by UNSCEAR 2000 as: (35 Bq/Kg for ^{226}Ra), (30 Bq/kg for ^{232}Th) and (400 Bq/kg for ^{40}K, except samples 3, 5, and 6 of ^{232}Th and samples 2, 3, 5, and 6 of ^{40}K. The Ra-226 activity concentrations of the samples are higher than those of Th-232 and K-40. The high concentrations results of ^{238}U in these areas of Sinai are due to the presence of phosphate and granite rocks with highly enriched with this radioactive nuclide and the weathering effects [15].

4.2.2. Hazard Indices

As shown in **Table 5**, the average values of radium equivalent (Ra_{eq}), Hazard indices (H_{ex} and H_{in}), Gamma index ($I_{\gamma r}$), the total dose rate (D_R), And Annual effective Dose (D_{eff}) were 1245.37 Bq/Kg, 3.37, 6.35, 8.41, 537.30 (nGy/h), and 0.6589 (mSv/y) respectively. These average values were very high compared to the world standard values as given by UNSCEAR 2000. Based on these results of Hazard, one can deduce that the use of soil samples for construction of the dwellings is considered to be not safe for human habitation.

5. Conclusions

The results of the present work indicate that:

- XRD measurements show that the major, minor, and some of trace elements concentrations for the samples are uranium compounds and thorium compounds.
- The study area shows very high values of ^{238}U and ^{232}Th concentrations except sample 3. Also, ^{40}K has high concentrations except sample 3 and 5.
- These average values for the soil samples from these regions (Jabal Al Qur, Wadi Baba, and Wadi Sieh) are considered to be very high levels of radioactivity compared to the world standard.
- The average of absorbed dose rate, annual effective dose, radium equivalent, and the radioactivity hazard in-

Table 4. The specific radioactive concentrations in Bq/kg, dry weight for Sinai samples.

Sample code	Radioactivity concentration (Bq/kg)		
	^{226}Ra	^{232}Th	^{40}K
Soil 1	591.34 ± 0.0005	100.22 ± 0.001	948.93 ± 0.0004
Soil 2	4220.41 ± 0.0004	77.22 ± 0.001	248.65 ± 0.0006
Soil 3	57.03 ± 0.0004	15.30 ± 0.001	12.18 ± 0.0004
Soil4	704.21 ± 0.0005	82.53 ± 0.002	569.67 ± 0.0005
Soil 5	1571.29 ± 0.0004	13.55 ± 0.002	54.79 ± 0.0006
Soil 6	560.76 ± 0.0005	23.66 ± 0.002	172.72 ± 0.0005
Soil 7	779.30 ± 0.0006	87.21 ± 0.002	569.14 ± 0.0005
Soil 8	1250.81 ± 0.0005	125.57 ± 0.002	654.47 ± 0.0005
Soil 9	381.36 ± 0.0005	86.44 ± 0.002	641.97 ± 0.0004
Soil 10	924.90 ± 0.0005	130.46 ± 0.002	685.53 ± 0.0005
Range	57.03 - 4220.41	13.55 - 130.46	12.18 - 948.93
Average	1104.14	74.21	455.80

Table 5. The radium equivalent Ra_{eq} (Bq/kg), Dose rate D_R outdoor (nGy/h), annual effective Dose (mSv/y and hazard indices for the samples.

Sample code	Radium equivalent Raeq (Bq/kg)	External index H_{ex}	Internal index H_{in}	Gamma index $I_{\gamma r}$	Dose rate D_R outdoor (nGy/h)	Annual effective Dose D_{eff} (mSv/y)
Soil 1	807.74	2.18	3.78	5.58	355.75	0.4363
Soil 2	4349.98	11.76	23.16	29.07	1860.91	2.282
Soil 3	79.85	0.22	0.37	0.541	34.41	0.0422
Soil 4	866.08	2.34	4.24	5.9	376.60	0.4619
Soil 5	1594.88	4.31	8.56	10.65	681.73	0.8361
Soil 6	607.88	1.64	3.16	4.09	261.61	0.3208
Soil 7	947.83	2.56	4.67	6.45	411.57	0.5047
Soil 8	1480.77	4.00	7.38	10.03	640.47	0.7855
Soil 9	554.39	1.5	2.53	3.83	244.29	0.2996
Soil 10	1164.24	3.15	5.65	7.93	505.69	0.6202
Average	1245.37	3.37	6.35	8.41	537.30	0.6589
World standard	370	≤1	≤1	≤1	57	0.07

dices values are much higher than the corresponding world average.

- Precautions and recommendations should be taken into consideration for high levels of radioactivity concentrations in these samples especially when people may inhabit in these areas. I recommend that this study to be taken as a base line for any future studies in this area.

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