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Effect of Cement Factory Exhaust on Radiological Contents of Surrounding Soil Samples in Assuit Province, Egypt

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ABSTRACT: *In the present work, the natural radioactivity of surrounding soil samples collected from Assuit cement factory (Egypt) have been measured by using NaI(Tl) detector. Gamma analysis for each sample along with the calculated specific activities shows that the average concentrations of Ra-226, Th-232 and K-40 were 31.44 ± 3.67 Bq kg^{-1}, 39.77 ± 2.00 Bq kg^{-1} and 113.23 ± 5.66 Bq kg^{-1} respectively, where only the values of Ra-226 and K-40 are lower than the worldwide average, while the average value of radium equivalent activity, absorbed gamma dose rate, external and internal hazard indices, gamma index, alpha index, indoor and outer door effective doses for each sample were 132.02 Bq kg^{-1}, 43.27 nGy h^{-1}, 0.256, 0.346, 0.691, 0.157, 0.212 mSv y^{-1} and 0.0572 mSv y^{-1} respectively. All these values were found to be lower than the permissibility limit value.*

Keywords: Cement exhaust, natural radioactivity, radium equivalent activity, soil, annual effective dose

1. INTRODUCTION

Soil is the upper part of the earth's crust and formed as a result of rock deformation by complex physicochemical processes including weathering decomposition, water movement and organic matter addition. Soil consists of minerals, organic matter, water and air. Their percentages vary widely according to soil type, usage,

and particle size.[1] Natural radioactivity is widespread in the earth's environment. It exists in soil, plant, water, air, coal and phosphate. The natural radioactivity in soil comes mainly from ^{238}U series, ^{232}Th series and ^{40}K. One of the components of external gamma-ray exposure to which a person is exposed to regularly is the naturally occurring radioactive materials (NORMs) in soil. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on geological and geographical conditions, and exist at different levels in the soils of each region in the world. The specific levels of terrestrial environmental radiation are related to the geological composition for each lithological separated area, and to the content of uranium, thorium and potassium in the rock from which the soil originated in each area.[2]

High levels of uranium and its decay products in rock and soil and thorium in monazite sands are the main sources of high natural background areas that have been identified in several areas of the world, e.g., Yangjiang in China, Rasmar in Iran, Kerala coast of India, etc.[3] Therefore, measurements of natural radioactivity in soil are of a great interest for many researchers throughout the world, which led to worldwide national surveys in the last two decades. El-Taher et al. studied the elemental analysis of Toshki soil in south Egypt by instrumental neutron activation analysis, and also the radioactivity in soil samples from Wadi Al-Assuity protective area in Upper Egypt.[4,5]

The authors measured the ^{226}Ra, ^{232}Th and ^{40}K levels in phosphate fertiliser and its environmental implications in Assuit governorate, Upper Egypt.[6] More specifically, natural environmental radioactivity due to gamma radiation depends primarily on the geological and geographical conditions, and appears at different levels in the soils of each region in the world.[3]

Because there are no existing databases for the natural radioactivity in soil around cement factory, our results are a start to establishing a database for cement factory environment. It is hoped that the data presented here will be useful to those dealing with soil and related fields. The aim of the present work is to assess the activity concentration levels of naturally occurring radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K in some soil samples around cement factory. Additionally, the radiological parameters due to natural radionuclides in soil will also be considered. This database will be used as reference to gauge any input sand trans boundary radioactive release in future.

2. EXPERIMENTAL

2.1 Sample Collection and Preparation

A total of 21 soil samples were collected from around Assuit cement factory, Egypt. The samples were ground, homogenised and sieved to about 100 mesh by a crushing machine. The samples were dried at 110°C to ensure that moisture is completely removed. Samples weighing 1 kg each were placed in beaker of 350 cm^3 volume. The beakers were sealed completely for over one month to allow radioactive equilibrium to be reached. This is a necessary step to ensure that radon gas is adequately confined within the volume, and that the daughters will also remain in the sample.[5–7]

2.2 Samples Counting

Specific activates were performed using gamma ray spectrometer, employing a NaI(Tl) scintillation detector 3" × 3". The assembly is hermetically sealed, and includes a NaI(Tl) crystal, coupled to PC-MCA Canberra Accuspe. To reduce the gamma ray background, a cylindrical lead shield of 100 mm in thickness with a fixed bottom and movable cover shielded the detector. The lead shield contained an inner concentric cylinder of copper (0.3 mm thick) to absorb X-rays generated in the lead. To determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The γ-ray spectrometer energy was calibrated using ^{60}Co and ^{137}Cs point sources. The naturally occurring radionuclides considered in the present analysis of the measured γ-ray spectra are: ^{212}Pb (with a main gamma energy at ~239 keV and a gamma yield of ~43.1%); ^{214}Pb (~352 keV, ~37.1%); ^{214}Bi (~609 keV, ~1120 keV and ~1765 keV, ~46.1%, ~15% and ~15.9% respectively); ^{228}Ac (~911 keV, ~ 29%); ^{208}Tl (~2615 keV, ~35.9%); and ^{40}K (~1461 keV, ~10.7%). Under the assumption that secular equilibrium was reached between^{232}Th and ^{238}U and their decay products, the specific activity of ^{232}Th was determined from the average specific activities of ^{212}Pb, ^{208}Tl and ^{228}Ac in the samples, and that of ^{226}Ra was determined from the average concentrations of the ^{214}Pb and ^{214}Bi decay products. The γ-ray spectrometer energy was calibrated using ^{60}Co and ^{137}Cs point sources. The measuring time for gamma-ray spectra range was 12 h. In order to determine the background distribution due to naturally occurring radionuclides in the environment around the detector, an empty backer was counted in the same manner as the samples. After measurement and subtraction of the background, the specific activity was calculated by the equation:[8]

$$A = \text{Net count} / Ɛ * Iγ * m * t \quad (1)$$

where:

A = activity concentrations of the sample in units Bq kg^{-1}
Ɛ = energy efficiency
m = mass of sample in unit kg
t = time measurement (43200 s)
Iγ = gamma intensity

3. RESULTS AND DISCUSSION

Specific activities of ^{226}Ra, ^{232}Th and ^{40}K in 21 of the surrounding soil samples for cement factory exhaust in Assuit province were shown in Table 1. From Table 1, it is observed that the specific activity of ^{226}Ra ranged from 21.37 ± 3.5 Bq kg^{-1} in Sample 5 to 40.19 ± 2.27 Bq kg^{-1} in Sample 18 with an average value of 31.44 ± 3.67 Bq kg^{-1}. For ^{232}Th, the specific activity ranged from 1.78 ± 0.14 Bq kg^{-1} in Sample 2 to 70.82 ± 3.55 Bq kg^{-1} in Sample 11 with an average value of 39.77 ± 2.00 Bq kg^{-1}. On the other hand, for ^{40}K activity, the values ranged from 90.32 ± 4.52 Bq kg^{-1} in Sample 5 to 130.48 ± 6.25 Bq kg^{-1} in Sample 7 with an average value of 113.23 ± 5.66 Bq kg^{-1}. For the average value of specific activity of present study, value of ^{226}Ra is lower than the world average of 32 Bq kg^{-1}, activity for ^{232}Th is lower than world average value of 45 Bq kg^{-1}, and activity of ^{40}K is also lower than the world average value of 412 Bq kg^{-1}.[16] Other observation from Table 1 is the specific activity of ^{40}K is higher than the specific activity of radionuclides such as ^{226}Ra and ^{232}Th for all samples. The specific activities values of ^{226}Ra, ^{232}Th and ^{40}K in the surrounding soil samples for cement factory exhaust are shown in Figure 1.

Table 1: Specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for soil samples surrounding Assuit cement factory.

Sample locations	Values (Bq kg^{-1})		
	Ra-226	Th-232	K-40
1	33.74 ± 3.71	41.11 ± 2.07	118.40 ± 5.92
2	27.26 ± 3.47	1.78 ± 0.14	119.93 ± 6.00
3	31.58 ± 4.27	38.35 ± 1.93	121.42 ± 6.07
4	30.67 ± 3.63	37.87 ± 1.90	105.48 ± 5.27
5	21.37 ± 3.50	30.84 ± 1.55	90.32 ± 4.52
6	33.06 ± 4.01	40.45 ± 2.03	123.70 ± 6.19

(*continued on next page*)

Table 1 (*Continued*)

Sample locations	Values (Bq kg^{-1})		
	Ra-226	Th-232	K-40
7	30.44 ± 3.53	30.72 ± 1.55	130.48 ± 6.52
8	31.12 ± 4.39	2.88 ± 0.19	110.02 ± 5.50
9	31.30 ± 4.69	40.11 ± 2.01	112.45 ± 5.62
10	28.80 ± 3.89	2.45 ± 0.12	106.76 ± 5.34
11	36.74 ± 3.80	70.82 ± 3.55	118.95 ± 5.95
12	33.07 ± 3.92	42.89 ± 2.15	116.46 ± 5.82
13	31.75 ± 3.62	42.61 ± 2.14	122.16 ± 6.11
14	29.15 ± 3.93	37.10 ± 1.86	114.92 ± 5.75
15	37.18 ± 4.12	61.45 ± 3.08	117.55 ± 5.88
16	27.06 ± 3.30	36.86 ± 1.85	100.09 ± 5.01
17	21.63 ± 1.40	30.56 ± 1.54	94.16 ± 4.71
18	40.19 ± 2.27	70.31 ± 3.52	119.47 ± 5.97
19	36.43 ± 4.22	37.54 ± 1.89	111.00 ± 5.55
20	31.23 ± 3.41	69.30 ± 3.47	106.81 ± 5.34
21	36.45 ± 4.14	69.32 ± 3.48	117.37 ± 5.87
Average ± S.D.	31.44 ± 3.67	39.77 ± 2.00	113.23 ± 5.66
Worldwide average	35	30	400

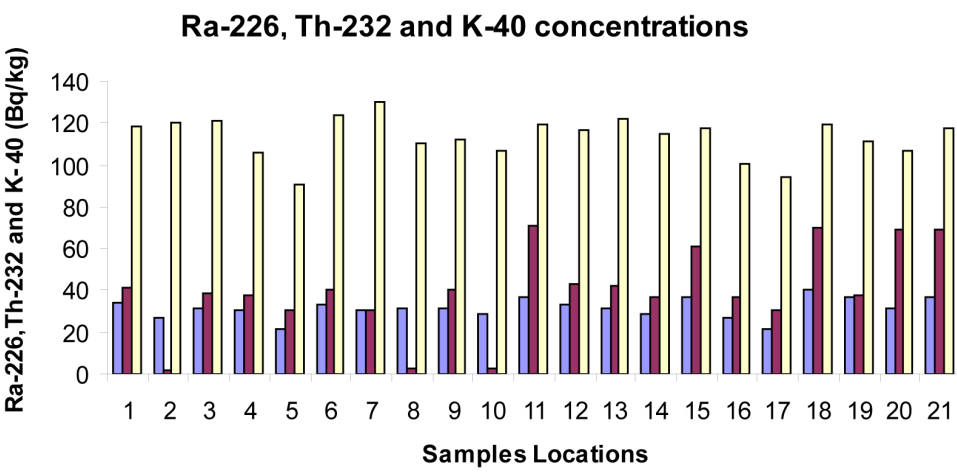

Figure 1: Specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for soil samples surrounding Assuit cement factory.

3.1 Radiological Parameters

3.1.1 Radium equivalent activity (Ra_{eq})

The natural radioactivity of cement material is usually determined from the contents of ^{226}Ra, ^{232}Th and ^{40}K. Since 98.5% of the radiological effects ^{238}U series are produced by ^{226}Ra and its daughter products, the contribution from the ^{238}U was replaced with the decay product ^{226}Ra. As sand beach minerals, rejected light sands and sea beach soils can be used in industries and building constructions, the γ-ray radiation hazards due to the specified radionuclides were assessed by three different indices. Radium equivalent activity is an index introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, taking into account the radiation hazards associated with them. The radium equivalent activity can be calculated according the following equation below:[9]

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra,^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The Ra_{eq} is related to the external γ-dose and internal dose due to radon and its daughters. From Table 2, it can be noticed that the values of Ra_{eq} ranged from 38.21 Bq kg^{-1} in Sample 2 to 149.10 Bq kg^{-1} in Sample 18 with an average value of 96.24 Bq kg^{-1}. The global limit of Ra_{eq} in building materials must be less than 370 Bq kg^{-1} for safe use.[9]

Table 2: Ra_{eq}, D, H_{ex}, H_{in}, I_γ and I_α of soil samples.

Sample locations	Raeq (Bq kg^{-1})	Dose rate (nGy h^{-1})	Hex	H_{in}	$I_{\gamma r}$	Iα
1	100.82	45.36	0.273	0.362	0.714	0.168
2	38.21	18.67	0.105	0178	0.281	0.138
3	94.91	42.81	0.258	0.343	0.674	0.158
4	92.21	41.44	0.250	0.333	0.652	0.153
5	71.79	32.27	0.195	0.253	0.510	0.1068
6	99.56	44.86	0.270	0.360	0.706	0.165
7	83.50	38.06	0.228	0.309	0.597	0.152
8	42.94	20.71	0.118	0202	0.309	0.155
9	96.54	43.38	0.262	0.347	0.684	0.156
10	39.77	19.24	0.109	0.187	0.287	0.144
11	146.34	64.71	0.305	0.496	1.032	0.183
12	102.55	46.04	0.278	0.367	0.726	0.165

(*continued on next page*)

Table 2 (*Continued*)

Sample locations	Raeq (Bq kg^{-1})	Dose rate (nGy h^{-1})	Hex	H_{in}	$I_{\gamma r}$	Iα
13	101.24	45.50	0.275	0.360	0.718	0.158
14	90.25	40.67	0.245	0.323	0.641	0.145
15	133.29	59.20	0.361	0.462	0.939	0.186
16	86.77	38.94	0.2350	0.309	0.815	0.135
17	71.92	32.38	0.196	0.254	0.512	0.1081
18	149.10	66.02	0.404	0.512	1.050	0.200
19	97.89	44.14	0.266	0.365	0.691	0.182
20	137.80	60.74	0.373	0.457	0.972	0.156
21	143.79	63.60	0.390	0.488	1.014	0.182
Average	96.24	43.27	0.256	0.346	0.691	0.157
Worldwide average	< 370	55	< 1	< 1	< 1	< 1

3.1.2 Absorbed Gamma Dose Rate (D)

The external outdoor absorbed gamma dose rate due to terrestrial gamma rays from the nuclides for ^{226}Ra, ^{232}Th and ^{40}K at 1 m above the ground level were calculated as follows outlined by:[10]

$$D\ (nGy\ h^{-1}) = 0.462A_{Ra} + 0.604\ A_{Th} + 0.0417A_K \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra,^{232}Th and ^{40}K, respectively. The highest value of dose rate was 66.02 nGy h^{-1} in Sample 18, while the lowest value was 18.67 nGy h^{-1} in Sample 2 with an average value of 43.27 nGy h^{-1}. The results of Ra_{eq} and D results are shown graphically in Figure 2.

3.1.3 External hazard indices (H_{ex}) and internal hazard indices (H_{in})

H_{ex} is obtained from R_{aeq} expression through the supposition that its maximum value allowed (equal to unity) corresponds to the upper limit of R_{aeq} 370 Bq kg^{-1}. This index value must be less than unity in order to keep the radiation hazard insignificant, i.e., the radiation exposure due to the radioactivity from construction materials is limited to 1.0 mSv y^{-1}. The H_{ex} can be defined as:[11]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{370} + \frac{A_K}{370} \leq 1 \quad (4)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra,^{232}Th and ^{40}K, respectively.

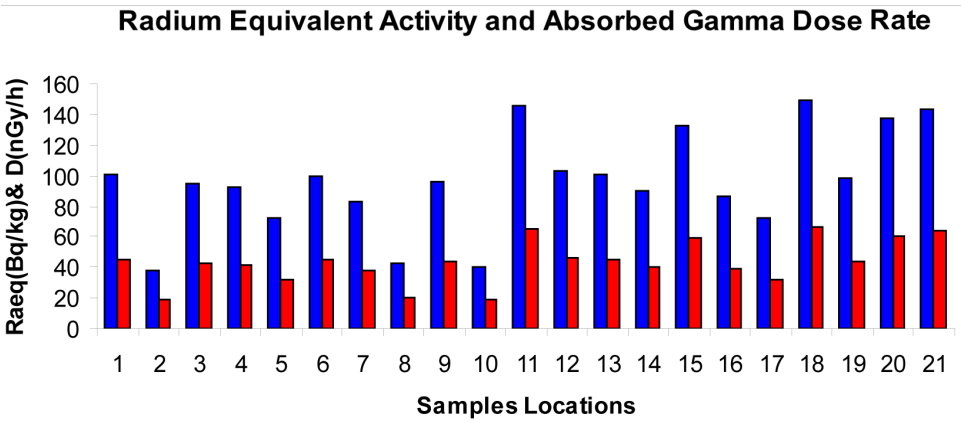

Figure 2: Radium equivalent activity, Ra_{eq} (Bq kg^{-1}) and Absorbed gamma dose rate, D (nGy h^{-1}) results for soil samples.

The internal exposure is caused by the inhalation of radon gas and its relevant examples which can be expressed in terms of H_{in} and calculated with equation below:[12]

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_k/4810 \leq 1 \quad (5)$$

The values of H_{ex} and H_{in} were varied from 0.105 in Sample 2 to 0.404 in Sample 18, 0.178 in Sample 2 to 0.512 in Sample 18 respectively, with an average value of 0.256 and 0.346. It should be added that this factor must be less than unity, so that it is placed within the allowable universal value.

3.1.4 Gamma index ($I_{\gamma r}$)

Another radiation hazard called the representative level index or gamma index ($I_{\gamma r}$), is defined as follows:[13]

$$I_{\gamma r} = A_{Ra}/150 + A_{Th}/100 + A_k/1500 \leq 1 \quad (6)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. $I_{\gamma r}$ varied from 0.281 in Sample 2 to 1.050 in Sample 18 with an average value of 0.157.

3.1.5 Alpha index (Iα)

Also, several indices dealing with assessment of the excess alpha radiation due to the radon inhalation originating from building materials called alpha index (Iα) or internal index have been developed.[15] In the present work, (Iα) was determined through the following formula:[14]

$$I\alpha = A_{Ra}/200 \text{ Bq/kg} \leq 1 \quad (7)$$

The values of Iα ranged from 0.138 in Sample 2 to 0.200 in Sample 18 with an average of 0.157.

The calculated values of Ra_{eq}, D, H_{ex}, H_{in}, Iγ and Iα are listed in Table 2.

3.1.6 The annual effective dose

To calculate the annual effective dose, the following must be taken into consideration: first, the conversion factor of absorbed dose to effective dose; and second, the internal occupation factor. The factor from observed dose in the air is used in the annual effective dose received by adult and 0.8 is put as the internal occupation (which is the ratio of time spent at home) and 0.2 put as external occupation as below:[16,17]

$$E_{in} \text{ (mSv y}^{-1}) = D \text{ (nGy h}^{-1}) \times 8.760 \text{ (h y}^{-1}) \times 0.8 \times 0.7 \text{ Sv G}^{-1} \times 10^{-6} \quad (8)$$

$$E_{out} \text{ (mSv y}^{-1}) = D \text{ (nGy h}^{-1}) \times 8.760 \text{ (h y}^{-1}) \times 0.2 \times 0.7 \text{ Sv G}^{-1} \times 10^{-6} \quad (9)$$

The values of indoor annual effective dose and outdoor annual effective dose varied from 0.0916 mSv y^{-1} in Sample 2 to 0.323 mSv y^{-1} in Sample 18, 0.0229 mSv y^{-1} in Sample 2 to 0.0809 mSv y^{-1} in Sample 18 with an average value of 0.42 mSv y^{-1} and 0.08 mSv y^{-1} respectively.

The annual effective dose rate outdoors in units of mSv y^{-1}, is also can be calculated by the following formula:[18]

$$\text{Annual Effective Dose Rate (mSv y}^{-1}) = D \times T \times F \quad (10)$$

where D is the calculated dose rate (in nGy h^{-1}), T is the outdoor occupancy time (0.2 × 24 h × 365.25, d ≈ 1753 h y^{-1}), and F is the conversion factor (0.7×10^{-6} Sv Gy^{-1}). Table 3 shows the values of the indoor and outdoor effective dose rate and the annual effective dose rate. The indoor annual effective dose and outdoor annual effective dose rate are shown graphically in Figure 3.

The specific activities measured in this study were compared with those from other countries (Table 4 and Table 5). The ^{226}Ra activity concentration for virgin soils in our study was higher than Nigeria but less compared to those values recorded in Yemen, Bangladesh, China, Turkey, Botswana, Malaysia and Iraq, as in Table 4. On the other hand, the value of the agricultural soils in the present study was higher than Pakistan, but less than those in Yugoslavia, Egypt, Algeria, Iraq and Malaysia, as in Table 5. The specific activity of ^{232}Th for virgin soils in the study area was

less than other countries except Nigeria and Iraq, while its value in agricultural soils was less than all the countries except Iraq. The ^{40}K specific activity values for both virgin and agricultural soils in this study were less than those in all other the countries.

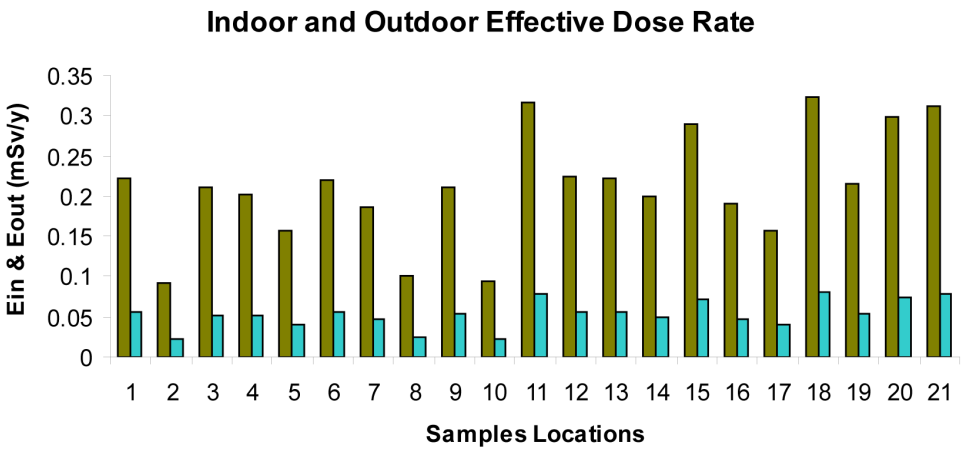

Figure 3: Annual effective dose E_{in} and annual effective dose E_{out} results.

Table 3: The indoor and outdoor effective dose rate and the annual effective dose rate of soil samples.

Sample locations	E_{in} (mSv y^{-1})	E_{out} (mSv y^{-1})	Annual effective dose rate (mSv y^{-1})
1	0.222	0.0556	0.0556
2	0.0916	0.0229	0.0229
3	0.210	0.0525	0.0525
4	0.203	0.0508	0.0508
5	0.158	0.0395	0.0395
6	0.220	0.0550	0.0550
7	0.186	0.0466	0.0467
8	0.101	0.0253	0.0254
9	0.212	0.0532	0.0532
10	0.0943	0.0235	0.0236
11	0.317	0.0793	0.0794
12	0.225	0.0564	0.0565
13	0.223	0.0558	0.0558
14	0.199	0.0498	0.0499
15	0.290	0.0726	0.0726
16	0.191	0.0477	0.0477

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Table 3 (*Continued*)

Sample locations	E_{in} (mSv y^{-1})	E_{out} (mSv y^{-1})	Annual effective dose rate (mSv y^{-1})
17	0.158	0.0397	0.0397
18	0.323	0.0809	0.0810
19	0.216	0.0541	0.0541
20	0.298	0.0745	0.0745
21	0.312	0.0790	0.0780
Average	0.212	0.0572	0.0530
Worldwide average	0.42	0.08	0.08

Table 4: Comparison of radioactivity levels in virgin soil samples with other countries.

	Virgin soil			
Country	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Reference
Yemen	44	58	822	[19]
Bangladesh	60.20	60.80	928.00	[20]
Nigeria	18	22	210	[21]
China	38	57.6	838	[22]
Turkey	115	192	1207	[23]
Botswana	34.8	41.8	432.7	[24]
Malaysia (Penang)	396	165	835	[25]
Malaysia (Pontian)	37	53	293	[26]
Malaysia	51.6	78.44	125.66	[27]
Iraq	32.52	20.30	378.93	[28]
Egypt	31.44	39.77	113.23	Present study

Table 5: Comparison of radioactivity levels in agriculture soil samples with other countries.

Country	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Reference
Yugoslavia	39.3	53	454	[29]
Pakistan	–	50.6–64	560.2–625.6	[30]
Pakistan	30	56	602	[31]
Egypt	43	54	183	[32]
Algeria	53.2	50.03	311	[33]
Malaysia	80.63	116.87	200.66	[27]
Iraq	33.55	21.52	326.74	[34]
Egypt	31.44	39.77	113.23	Present study

4. CONCLUSION

It is important to determine the background radiation level in order to evaluate the health hazards. The average specific activity values for ^{226}Ra and ^{40}K are below the worldwide average, except for ^{232}Th.[16] Additionally, the average value of the eight radiation hazard indices was found to be less than the average recommended indices given by the worldwide average.[16] The data produced in the present work can be used as baseline radiological data for future investigations.

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