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NATURAL RADIOACTIVITY MEASUREMENTS OF SOIL SAMPLES FROM SORAN DISTRICT IN KURDISTAN REGION-IRAQ

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ABSTRACT

The amount of radioactivity in soil samples, thirty six samples at different depths (surface, 15cm, 30cm), were collected from twelve different locations at Soran district in the northeastern of Iraqi Kurdistan region. The samples have been studied by using gamma-ray spectrometry, NaI(Tl) Scintillation detector and CR-39 Solid State Nuclear Track Detector (SSNTD) . Activity concentrations of each of K-40, Cs-137, Ra-226 (U-238) , Th-232 (Ac-228) and Radon radionuclides were measured; Radon activity concentration was ranged from (135.301 to 770.727) Bq.m^{-3}. Within the studied area the value of radium equivalent activities Ra_{eq} have been calculated and ranged from (0.1130-0.3292) Bq.g^{-1}, these values are below the internationally accepted value of 0.370 Bq.g^{-1}. The value of absorbed dose rate D has been calculated and ranged from (0.0528 to 0.1510) nGyh^{-1}, these calculated values were through the estimate of average global terrestrial radiation of 0.055 nGyh^{-1}. Indoor annual effective dose rate E_{ied}, outdoor annual effective dose rate E_{oed}, external hazard indices H_{ex} and internal hazard indices H_{in} have been computed and found to be below the permissible values.

Keywords: Natural Radioactivity; Gamma-ray Spectroscopy; CR-39; Absorbed Dose; Hazard Indices;

INTRODUCTION

Soils are naturally radioactive, because of their mineral content. The natural radioactivity may vary considerably from one type of soil to another. The sources of radioactivity in soils other than those of natural origin are mainly due to extensive use of fertilizers rich in phosphates for agricultural purposes. routine authorized low level radioactive effluent discharges or accidentally release into the environment from nuclear fuel cycle installations, mineral extraction industries, industries working with mineral materials enriched in naturally radioactive elements [1].

Although the influence of radioactivity to the human health was discovered about hundred years ago, not earlier than last 20 years are provided more intensive measurements of radiation levels and recommendations for their limitation. From all the natural ionizing radiation, the greatest influence on people health has radiation that arise from everywhere presented radioactive gas radon and its natural short lived progenies. All over the world, there is a lot of necessary attention that is dedicated to radon levels measurements, especially in the living and occupation indoor environment. The considerable interest for radon and its healthy effects comes from very big part that radon has in the total human dose that men receive from all the natural radioactive sources [2].

Among all the radon isotopes only two of them are important, namely Rn-222, the immediate decay product of Ra-226, deriving from the uranium series of natural radionuclide's, which has a half-life of 3.825 days, and Rn-220 (thoron; Tn), the immediate decay product of Ra-224, deriving from the thorium series [3].

In this work the radioactive measurements associated with the ^{238}U, ^{226}Ra, , ^{222}Rn ^{40}K, and ^{137}Cs radionuclide's has been performed for the soil of Soran district using the gamma-ray spectroscopy and solid-state nuclear track detector techniques to determine the radionuclide concentration levels in the agricultural soil and the associated radioactive dose measurements.

EXPERIMENTAL

MATERIALS, METHOD AND INSTRUMENTS

A total of thirty six (36) soil samples at three depths: surface, 15cm, and 30cm for 12 locations were collected from Bapshtean, Badlean, Kawren, Hawdean, Balakean, Rwandz, Zozk, Berowan, Azadi (kany), Saruchawa, Shawrawa and harem, located in the northeastern of Kurdistan region, Iraq, Erbil Governorate, Soran district. Table (1) illustrates the place and code of each sample. All the samples were taken under the supervision of geological expert from the study area and brought to the laboratory (Post Graduate Nuclear Laboratory).

Table 1: Samples location and their codes.

No.	Location	Samples Code		
		Surface	15cm	30cm
1	Bapshtean	S1A	S1B	S1C
2	Badlean	S2A	S2B	S2C
3	Kawren	S3A	S3B	S3C
4	Hawdean	S4A	S4B	S4C
5	Balakean	S5A	S5B	S5C
6	Rwandz	S6A	S6B	S6C
7	Zozk	S7A	S7B	S7C
8	Berowan	S8A	S8B	S8C
9	Azadi(kany)	S9A	S9B	S9C
10	Saruchawa	S10A	S10B	S10C
11	Shawrawa	S11A	S11B	S11C
12	Shahedan(harem)	S12A	S12B	S12C

Soil samples were dried in an oven at 110 ^{o}C for 24 hours to evaporate all the moisture content and to maintain the actual weight; the dried sample have been grounded by using Grinding Machine, then equal weights [1000 gm for NaI(Tl) and 150 gm for SSNT detectors] of the powdered samples were packed in plastic bags (Marinelli beaker) and sealed for about 4 weeks to reach secular equilibrium between radium and thorium and their progenies. After, the sealed samples then mounted on the detector for counting [21600 seconds for NaI(Tl)], Figure (1).

Figure 1: The NaI(Tl) Scintillation Detector

The γ-ray spectrum of each studied sample was taken by the planar NaI(Tl) detector, the area under the peaks (net area) have been calculated and from the efficiency curve the specific activity (activity concentration) A_s were determined using the formula [4].

$$A_s(Bq.g^{-1}) = \frac{A}{W} \qquad (1)$$

where A the activity of the isotope and W is the Weight of the sample. The expression for the calculation of the activity of each isotope was [4].

$$A(Bq) = \frac{A_{net}}{\varepsilon \times I_\gamma \times t} \qquad (2)$$

where A_{net} is the net area of the total absorption line, I_γ the absolute intensity of the transition, t the sample measurement time, and ε the gamma efficiency evaluated in function of the transition energy.

The distribution of Ra-226, Th-232 and K-40 in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/g to compare the specific activity of materials containing different amounts of Ra-226, Th-232 and K-40 [5]. It is calculated using the following relation [6]:

$$Ra_{eq}(Bq.g^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \qquad (3)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of Ra-226, Th-232 and K-40 respectively [6]. The absorbed dose rates (D) due to gamma radiations in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (Ra-226, Th-232 and K-40) were calculated based on guidelines provided by UNSCEAR 2000 [7]. We assumed that the contributions from other naturally occurring radionuclides were insignificant. Therefore, D can be calculated according to UNSCEAR 2000 [7]:

$$D(nGy.h^{-1}) = 0.462A_{Ra} + 0.621A_{Th} + 0.0417A_K \qquad (4)$$

where D is the dose rate in nGy/h and A_{Ra}, A_{Th} and A_K are the activity concentrations of Ra-226, Th-232 and K-40 respectively. In the above equation, it is assumed that all decay products of Ra-226 and Th-232 are in radioactive equilibrium with their precursors.

A widely used hazard index (reflecting the external exposure) called the external hazard index H_{ex} is defined as follows [7]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (5)$$

In addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter product is quantified by the internal hazard index H_{in}, which is given by [7]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (6)$$

The values of the indices (H_{ex}, H_{in}) must be less than unity (≤ 0.001) for the radiation hazard to be negligible [7].

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose (D) in air to effective dose, 0.7 $SvGy^{-1}$ was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor and implying that 20% of time is spent outdoors, outdoor occupancy factor of 0.2 proposed by UNSCEAR 2000 were used. The effective dose rate (E) in units of $mSvy^{-1}$ was calculated by the following formulae [4]:

Indoor effective dose:

$$E_{ied}(mSvy^{-1}) = D(nGyh^{-1}) \times 8760h \times 0.8 \times 0.7SvGy^{-1} \times 10^{-6} \qquad (7)$$

Outdoor effective dose:

$$E_{oed}(mSvy^{-1}) = D(nGyh^{-1}) \times 8760h \times 0.2 \times 0.7SvGy^{-1} \times 10^{-6} \qquad (8)$$

The representative level index, $I_{\gamma r}$, used to estimate the level of γ-radiation hazard associated with the natural radionuclides in specific investigated samples, is defined as [8]:

$$I_{\gamma r} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \qquad (9)$$

The SSNTD technique which uses a plastic detector CR-39 depending on the extend time measurement method has been depended for detecting emitting α-particle from the Rn gas which produces from the natural decay of U-238 of the used soil samples. The CR-39 detectors with width 500µm and dimensions of (1x1.5) cm were placed at the uppermost part of the tube, Figure (2), and leaved for 60 days of exposure after a perfect sealing of the dosimeter.

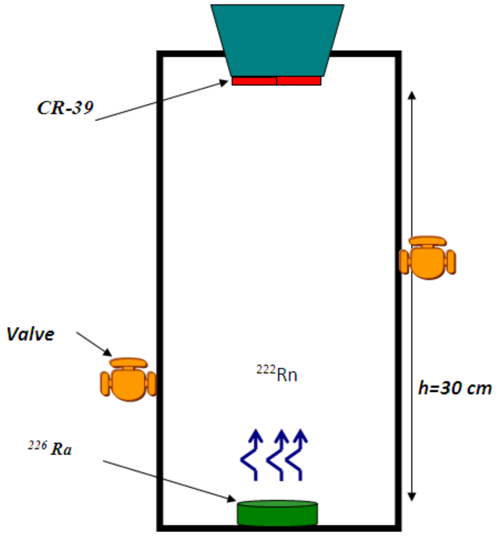

Figure 2: Experimental arrangement of the configuration of exposure of CR-39 detector in the calibration chamber

Chemical etching (CE) of plastic detectors is straight forward; that of mineral crystals, nearly so. The etching is usually carried out in thermostatically controlled baths (kept constant). For plastics, the most frequently used etchant is the aqueous solution of NaOH (or KOH), with concentrations ranging from a molarities of 1–12 (~6 M being the most popular). The temperatures usually employed range from ~ 25 to 70 ^{o}C .
After several hours of chemical etching, the etched tracks have diameters of several micrometers. In this study a digital system were used for track counting and track diameter measurement. The tracks produced by alpha-particles were observed and counted under the digital microscope at total magnification(400X), The field area was equal to 0.00395196 cm^{2}. The number of α-tracks per field was averaged over twenty fields counted for each detector.
The measurement of radon alpha-particles track density on upper detector (ρ_u) (tracks.cm^{-2}) was converted in to the radon activity concentrations in (Bq.m^{-3}) using Eq.(10), the measured calibration factor (K) for the given CR-39 plastic track detector which was equal to (0.237 tracks.cm^{-2}. d^{-1}/Bq.m^{-3}) Radon activity concentration in (Bq.m^{-3}) could be calculated from formula [9].

$$C_{Rn} = \frac{\rho}{T_e K_{Rn}} \qquad (10)$$

Where (ρ) is the track density observed in the upper CR-39 plastic track detector, T_e is the effective exposure time, and K is the calibration factor for radon detection.

Where $T_e = T - \frac{1}{\lambda}(1 - e^{-\lambda T})$

$\lambda=2.1 \times 10^{-6}\ S^{-1}$, T=60 and 70 day.

Referring to Figure (2), it is clear that the effective radium content (i.e. radium in radioactive equilibrium with the measured radon released from the sample) could be calculated from the formula [10]:

$$(C_{Ra})_{eff.} = \frac{\rho h A}{K M T_e} \qquad (11)$$

Where (ρ) is the track density observed in the upper CR-39 plastic track detector, M is the mass of the sample (M=150g), h is the distance between the detector and the top of sample surface (0.30m), A is the cross section area of the chamber (0.2464 m^2) and K is the calibration factor for radon detection (0.237 tracks.$cm^{-2}.d^{-1}$ per $Bq.m^{-3}$).

RESULTS AND DISCUSSION

NaI(Tl) detector

The γ-ray spectrum of each studied sample was taken by the gamma-ray spectroscopy including the planar NaI(Tl) scintillation detector. The spectroscopy used for detection to multi-channel analyzer (MCA) [512 channels NaI(Tl) detectors], The net sample γ-ray spectra (Figure 3 as a sample spectrum) were obtained by subtracting the background spectrum, from (sample + background) spectra.

The activity concentrations in (Bq/g) for the studied samples were measured for each of the K-40, Cs-137, Ra-226, and Th-232 radionuclide's. The results are presented in Tables (2 through 4) and shown in Figs (4 through 6).

The comparison of activity concentration between K-40, Cs-137, Ra-226 and Th-232 for the surface soil samples has been shown in Table (2) and Figure (4). In this figure it has been noticed that K-40 radionuclide has a higher activity and also Cs -137 shows a somewhat lower activity than that of K-40, while Ra-226 and Th-232 shows low activities in compare to K-40 and Cs-137. These results are in agreement to the facts that Cs-137 has a fallout origin and the existence of Ra-226 and Th-232 (members of U and Th natural decay serieses, respectively) within the rocks rather than soil. The high activity of K-40 may attribute to its light weight which assists its transition and attachment to soil particles within the earth's cluster.

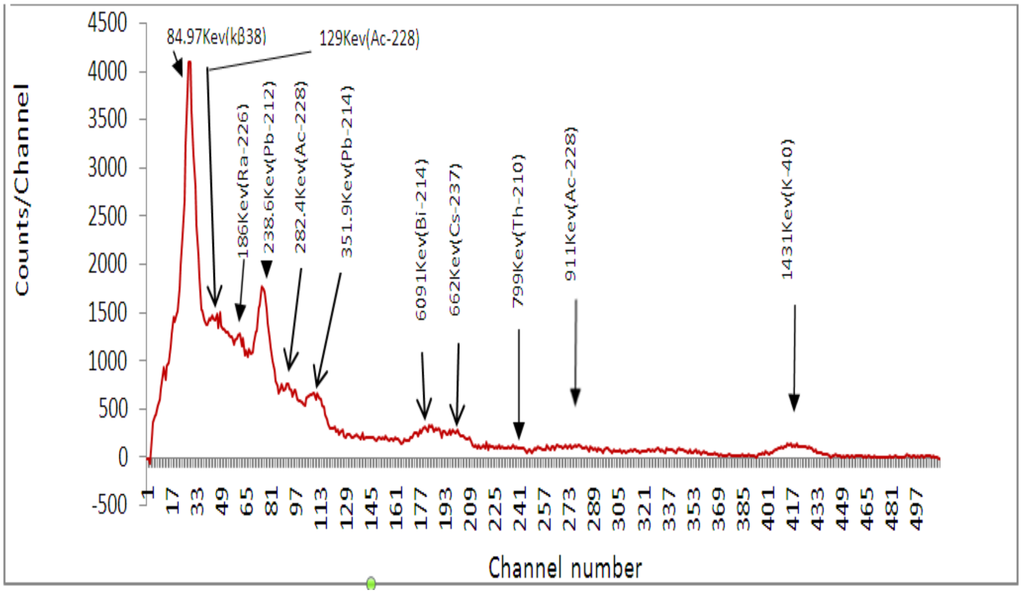

Figure 3: The net sample γ-ray spectrum of sample (S12B)

Table 2: Activity concentrations in (Bq.g^{-1}) of K-40, Cs-137, Ra-226 and Th-232 (Ac-228)

No.	Sample code	Activity Concentration (Bq/g)			
		K-40	Cs-137	Ra-226	Th-232
1	S1	0.235	0.274	0.052	0.063
2	S2	0.433	0.298	0.098	0.070
3	S3	0.492	0.298	0.112	0.045
4	S4	0.378	0.296	0.065	0.055
5	S5	0.415	0.452	0.134	0.078
6	S6	0.514	0.537	0.141	0.052
7	S7	0.475	0.361	0.086	0.070
8	S8	0.448	0.457	0.122	0.143
9	S9	0.265	0.270	0.075	0.025
10	S10	0.574	0.291	0.100	0.091
11	S11	0.406	0.210	0.087	0.094
12	S12	0.515	0.445	0.132	0.136

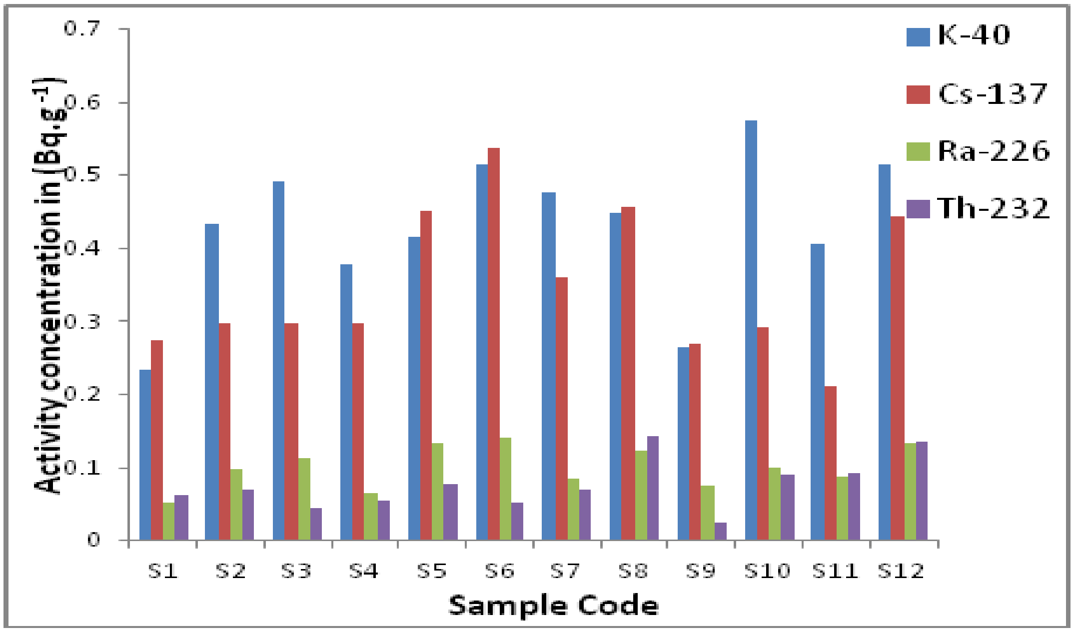

Figure 4: Activity concentrations in (Bq.g^{-1}) of K-40, Cs-137 , Ra-226 and Th-232 (Ac-228)

Table 3: Activity concentration in (Bq.g^{-1}) of K-40, Cs-137 , Ra-226 and Th-232 in Soran soil samples at depth of 15cm

No.	**Sample code**	**Activity Concentration (Bq/g)**			
		K-40	**Cs-137**	**Ra-226**	**Th-232**
1	S1	0.204	0.290	0.052	0.014
2	S2	0.349	0.230	0.093	0.049
3	S3	0.378	0.334	0.081	0.043
4	S4	0.430	0.305	0.073	0.041
5	S5	0.375	0.373	0.100	0.073
6	S6	0.507	0.412	0.127	0.101
7	S7	0.430	0.219	0.088	0.072
8	S8	0.507	0.391	0.120	0.043
9	S9	0.232	0.223	0.058	0.020
10	S10	0.619	0.234	0.106	0.058
11	S11	0.410	0.244	0.080	0.059
12	S12	0.548	0.431	0.127	0.103

For the samples taken at depth of 15 cm from the surface the comparison of activity concentrations of K-40, Cs-137, Ra-226 and Th-232 radionuclides have been presented in Table (3) and shown in Figure (5) which reveals approximately the same systematic variation of radionuclide's activity concentration as existed in the surface samples. The decrease in the Cs-137 activity concentration as compared to the surface samples becomes obvious.

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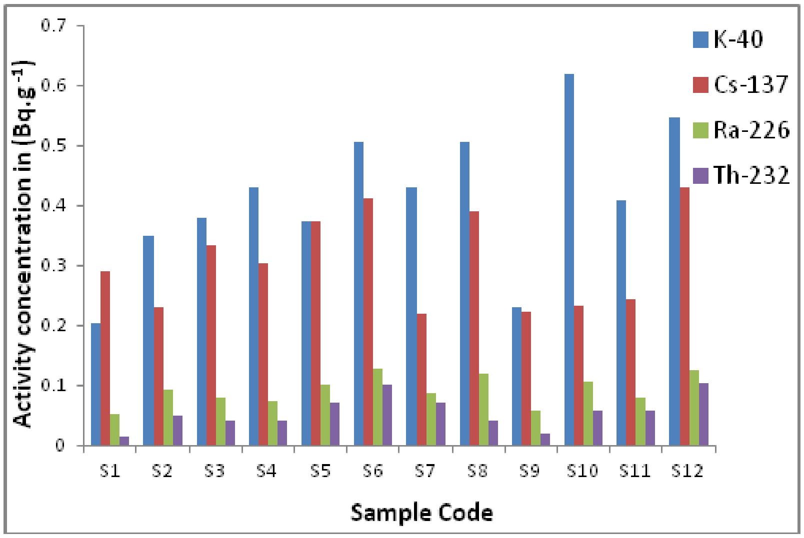

Figure 5: Activity concentrations in (Bq.g^{-1}) of K-40, Cs-137, Ra-226 and Th-232 (Ac-228) in Soran soil samples at depth of 15cm

Table 4: Activity concentration in (Bq. g^{-1}) of K-40, Cs-137, Ra-226 and Th-232 in Soran soil samples at depth of 30cm

No.	**Sample code**	**Activity Concentration (Bq/g)**			
		K-40	**Cs-137**	**Ra-226**	**Th-232**
1	S1	0.296	0.181	0.065	0.005
2	S2	0.437	0.221	0.084	0.053
3	S3	0.328	0.308	0.104	0.042
4	S4	0.378	0.165	0.06	0.023
5	S5	0.380	0.278	0.087	0.047
6	S6	0.496	0.359	0.157	0.111
7	S7	0.541	0.227	0.115	0.074
8	S8	0.480	0.359	0.119	0.052
9	S9	0.241	0.113	0.053	0.023
10	S10	0.531	0.245	0.126	0.089
11	S11	0.434	0.175	0.080	0.044
12	S12	0.554	0.218	0.143	0.084

Similarly, the comparison between activity concentration of K-40, Cs-137, Ra-226 and Th-232 radionuclide's for the 30 cm depth soil samples have been performed and illustrated in Table (4) and Figure (6).Within these samples a similar variation in the activity concentration of the mentioned radionuclide's as found for the depths 0 and 15cm have been observed.

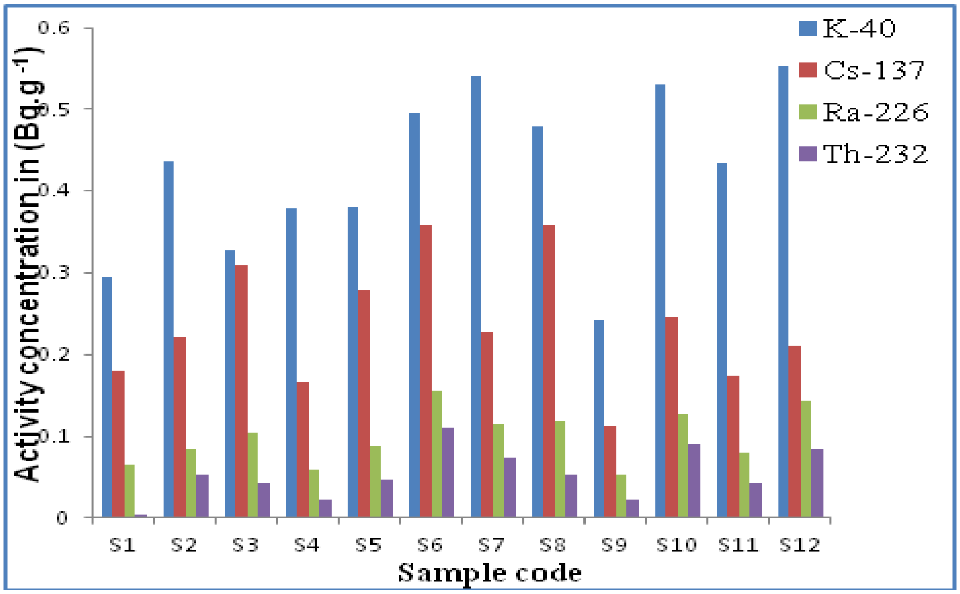

Figure 6: Activity concentrations in (Bq.g^{-1}) of K-40, Cs-137, Ra-226 and Th-232 (Ac-228) in Soran soil samples at depth of 30cm

In general, for Cs-137 at every location and every depth, several factors influence the actual range or variation. Transport of the cesium in the soil, once it is deposited, is affected by many site specific factors including soil type, rainfall, drainage, terrain, vegetation and local activities and conditions. Furthermore, local meteorological conditions can have significant impact on the fallout deposition and transport in a given area. Finally, sampling protocol significantly effects the reported soil concentrations [11].

CR-39 SSNTD

Table (5) show that the variation in the activity concentration of Ra-226 with depth from the surface to a depth of 15 cm and 30 cm shows the decline and rise according to the site, and can tell the difference between the method of solid state nuclear track detector (SSNTD) and the method of γ-ray spectroscopy to determine the activity concentration of Ra-226. Through the comparison of two methods it is deduced that the Ra-226 activity concentration in each case for any one sample almost equal in fluctuation, but differs in amount. Generally, the two methods give a comparable result for the Ra-226 concentrations, a manner which confirms the work reliability.

Table 5**:** Activity concentrations in (Bq. g^{-1}) of Ra-226 in Soran soil samples measured by the SSNTD

No.	Sample code	Activity Concentration (Bq/g)			Other works
		A (surfaces)	B (15	C (30	
1	S1	0.034	0.027	0.051	0.013±0.001 Bq/g[16], 0.0051 to 0.128 Bq/g[12], 0.0079±0.0028 to 0.0961±0.0098 Bq/g[4], 0.0257 to 0.156 Bq/g [17], 0.1482 Bq/g to 0.2656 Bq/g [5] 0.128 ± 0.0 85 Bq/g- 0.115 ± 0.0 86 Bq/g[15]
2	S2	0.072	0.067	0.102	
3	S3	0.061	0.078	0.058	
4	S4	0.042	0.042	0.024	
5	S5	0.026	0.047	0.074	
6	S6	0.078	0.093	0.109	
7	S7	0.048	0.056	0.103	
8	S8	0.073	0.079	0.082	
9	S9	0.050	0.037	0.046	
10	S10	0.065	0.084	0.063	
11	S11	0.039	0.064	0.042	
12	S12	0.139	0.114	0.050	

The measured radon activity concentration shown in Table (6) reveals that variation in the radon activity concentration increases with the increase of depth in most places, while in the others it do not obey a systematic trend due to the physical nature of the surface areas which contain cracks that disturbs the natural trapping of radon gas within inner soil layers.

Table 6 Radon activity concentration in (Bq. m^{-3}) in Soran soil samples measured by the SSNTD

No.	Sample code	Radon content (Bq. m^{-3})			Other works
		A (surfaces)	B (15 cm)	C(30m)	
1	S1	188.749	150.141	279.882	11000 Bq/m3 - 75000 Bq/m3 [83], 3794.3 Bq/m3-4891.4 Bq/m3[84], 1 Bq/m3-1000 Bq/m3[85], 25±9 Bq/m3- 500±100 Bq/m3[86], 2.878 Bq/m3- 9.305 Bq/m3[21], 29 Bq/m3 - 422 Bq/m3[79].
2	S2	402.284	371.0793	561.641	
3	S3	338.408	437.868	322.144	
4	S4	230.929	236.413	135.301	
5	S5	143.468	259.133	414.349	
6	S6	428.922	517.895	610.1	
7	S7	271.997	312.517	569.791	
8	S8	409.910	434.855	455.603	
9	S9	277.657	210.347	255.414	
10	S10	357.744	473.452	350.330	
11	S11	216.871	354.779	234.507	
12	S12	770.727	632.0778	280.264	

RADIATION HAZARD INDICES

It is important to assess the gamma radiation hazards to human; which can be done by calculating the different radiation hazard indices. Table(7) presents the average range of Radium Equivalent Activity Ra_{eq}(Bq.g^{-1}) of samples under investigation which was calculated on the basis of equation (3) (0.1130-0.3292) Bq.g^{-1}, and are below the internationally accepted value of 0.370 Bq.g^{-1} [12]. Absorbed Gamma Dose rate D(nGyh^{-1}) expressed in equation (4) are found to be in the range (0.0528 - 0.1510) nGyh^{-1}, these calculated values were through the estimate of average global terrestrial radiation of 0.055nGyh^{-1} [12]**.** Annual Indoor effective Dose rate were determined from equation (7) to be in the range (0.259 - 0.741)10^{-3} mSvy^{-1} and Annual Outdoor effective Dose rate calculated from equation (8) was in the range (0.0647 - 0.1853)10^{-3} mSvy^{-1}, these values are below the internationally accepted value [12].

Table 7: Average Radium Equivalent Activity Ra_{eq} (Bq.g^{-1}), Absorbed Gamma Dose rate D(nGyh^{-1}), Annual Indoor effective Dose Rate (mSvy^{-1}), and Annual Outdoor effective Dose rate (mSvy^{-1}) within the studied samples

No.	Sample code	Ra_{eq} (Bq.g^{-1})	D (nGyh^{-1})	E_{ied} (mSvy^{-1})*10^{-3}	E_{oed} (mSvy^{-1})*10^{-3}
1	S1	0.1143	0.0532	0.260	0.0652
2	S2	0.2046	0.0947	0.464	0.1161
3	S3	0.1917	0.0892	0.437	0.1095
4	S4	0.1532	0.0716	0.350	0.0878
5	S5	0.2313	0.1066	0.522	0.1307
6	S6	0.3057	0.1408	0.690	0.1727
7	S7	0.2361	0.1091	0.535	0.1339
8	S8	0.2705	0.1247	0.611	0.153
9	S9	0.1130	0.0528	0.259	0.0647
10	S10	0.2685	0.1244	0.61	0.1526
11	S11	0.2074	0.0957	0.469	0.1175
12	S12	0.3292	0.1510	0.741	0.1853

Table (8) present the average range of External Hazard index H_{ex}**,** Internal Hazard index H_{in}, and Representative Level Index $I\gamma_r$ of samples under investigation. External Hazard index H_{ex} was calculated on the basis of equation (5) (0.305 - 0.889)10^{-3}, which are below the internationally accepted value of 10^{-3} [12]. Internal Hazard index H_{in} was calculated on the basis of equation (6) (0.473 - 1.250)10^{-3}, which exceeds the internationally accepted value of 0.001 for the S6, S8, S10 and S12 soil samples. Representative Level Index $I_{\gamma r}$ was calculated on the basis of equation (9) (0.801-2.327)10^{-3}, for all the locations shows higher values in comparison to the internationally accepted value of 0.001 except for the (S1 and S9) samples.

Table 8**:** Average External Hazard index H_{ex}, Internal Hazard index H_{in}, and Representative Level Index $I\gamma_r$ within the studied samples

No.	Sample Code	$H_{ex}*10^{-3}$	$H_{in}*10^{-3}$	$I\gamma_r*10^{-3}$
1	S1	0.308	0.460	0.812
2	S2	0.552	0.8	1.453
3	S3	0.517	0.785	1.359
4	S4	0.413	0.592	1.1006
5	S5	0.624	0.914	1.632
6	S6	0.825	1.207	2.156
7	S7	0.637	0.898	1.681
8	S8	0.730	1.056	1.914
9	S9	0.305	0.473	0.801
10	S10	0.725	1.024	1.915
11	S11	0.56	0.782	1.477
12	S12	0.889	1.250	2.327

CONCLUSION

The studied soils of Soran district higher values of activity concentration of (K-40, Cs-137,Ra-226, Ac-228 and radon) showed in most deeper soil sample. The physical, chemical, and environmental factors cause the variations in the regional radioactivity. The values of radium equivalent activities Ra_{eq}, absorbed dose rate, indoor annual effective dose rate, outdoor annual effective dose rate, and external hazard indices H_{ex} have been found to be below the internationally accepted values.

The internal hazard indices H_{in} (for the S6, S8, S10 and S12 soil samples) and the Representative level index $I_{\gamma r}$ for the (S1 and S9) samples have exceeded the internationally accepted values. Thus it is better to avoid the usage of these regional soils in dwelling activities.

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