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Determination of Radioactivity Levels and Excess Lifetime Cancer Risk in Soil Samples at the Environment at Al-Mishal, Libya, using Gamma-ray Spectrometry.

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Highlights

- **Gamma-ray spectrometry analysis of eight soil samples from Al-Meshal, Libya, revealed average activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K, that all exceed international permissible safety levels.**
- **The absorbed gamma dose rate in the study area ranged from 99.91 nGy/h to 160.92 nGy/h (with an overall average of 163.07 nGy/h), significantly surpassing the UNSCEAR benchmark of 84 nGy/h.**
- **The Excess Lifetime Cancer Risk (ELCR) values across all collected soil samples ranged from 0.40×10^{-3} to 0.65×10^{-3}, well above the standard permissible threshold of 0.29×10^{-3}.**

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ABSTRACT

Eight soil samples collected from Al-Meshal, Libya, have been investigated. Concentrations of radionuclides in the samples were determined by a gamma-ray spectrometer using a NaI(Tl) detector with a specially designed shield. The contents of ^{238}U and ^{232}Th are high in the samples, and the content of ^{40}K is higher than the permissible level. The absorbed dose rate ranged from 99.91 to 160.92nGy/h. External and internal hazard indices were also determined. The Excess Lifetime Cancer Risk (ELCR) values for all samples were found to be much higher than the permissible level of 0.29

1. Introduction.

The ionizing radiation is a widespread occurrence in the environment, and natural sources account for 80% of human radiation exposure, with soil being the primary source (Saito, *et al.*, 2022). Natural radioactivity comes from ^{238}U, ^{232}Th and ^{40}K. Some other terrestrial radionuclides, like the ^{235}U series, ^{87}Rb, ^{147}Sm, and other nuclei, exist in nature but at low levels, so their contributions to the dose in humans are very small. The natural environmental radioactivity and external exposure due to gamma radiation depend on geographical and geological conditions and appear at different levels in the soils of each region in the world (UNSCEAR, 2014).

The natural radioactivity of a soil sample is usually determined from ^{226}Ra, ^{232}Th and ^{40}K. The contribution from ^{238}U and other ^{226}Ra precursors is normally ignored; 98% of the radiological effects of the uranium series are produced by radium and its daughters (Khan *et al.*, 2020). Studies of natural radioactivity are necessary because of their radiological impact, and they act as important geochemical tracers in the environment (HAZOU, 2021; Abd El-Halim *et al.*, 2017).

2.Materials and method

2.1. Study area

Al-Meshal is a rural agricultural region located in northeast Libya, with geographical coordinates of (21^0 40' 42.2''E, 32^0 28' 46.7'' N), characterized by fertile soil suitable for agriculture and livestock breeding. The main activity in this region is raising livestock, which contributes greatly to the local production of the country, so it is necessary to know the environment of this region, including the soil.

2.2. Sampling and preparation

Samples of soil were collected from various parts of the study area. These specimens were accurately labelled with their GPS coordinates. The study area was purposefully stratified into multiple sections to ensure widespread sampling as well as to obtain representative samples across the locations. The top layer of soil was scraped off at each sampling location to a depth of 25 cm to eliminate any potential contamination from human activities. Foreign materials such as pebbles and stones were cleared and removed from the soil specimens. Using a plastic hand trowel to prevent the introduction of any contamination, the collected soil samples were placed in a polythene bag properly labelled for easy identification.

After collection, the samples were securely packed in polyethylene bags and transported to the laboratory of the physics department of Omar Al-Mukhtar University, Libya. The gathered soil samples underwent air drying at 110^0 C for 4 hours (depending on the soil moisture content) until a constant weight was attained. Subsequently, a 0.25 mm mesh was employed for sieving the samples. Around 250 g of the homogenized soil samples were then packed and sealed in airtight Marinelli beakers and stored for 4 weeks to achieve secular equilibrium.

The Al-Meshal area has fourteen stores of old weapons. One of these stores was chosen to take samples from the four directions

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(East, West, North, and South) for the first kilometer of soil (Sw1), applied to all directions in the same way. After the distance of the second kilometer, soil (Sw2) was also collected, and thus we obtained 8 soil samples. Table 1 shows the details of the collected samples.

Table 1

The details of the collected samples.

Sample No	Description
SE1	Soil from the first kilometre (East).
SE2	Soil from the second kilometre (East).
SN1	Soil from the first kilometre (North).
SN2	Soil from the second kilometre (North).
SS1	Soil from the first kilometre (South).
SS2	Soil from the second kilometre (South).
SW1	Soil from the first kilometre (West).
SW2	Soil from the second kilometre (West).

2.3 Gamma spectrum analysis

A NaI (Tl) detector of "3×3" inch crystal dimension, with a digital multichannel analyzer of 4096 channels, was used with an operating voltage of 695 volts to perform this work. The measuring time used for the samples was 24 hours. Two-point sources were used to find the efficiency calibration (^{137}Cs and ^{60}Co). These sources were set at 5 cm from the top of the detector for 24 hours, and then the spectrum was recorded to obtain the net area under the peak, which can be measured from the gamma spectrum (Asma, 2023).

3. Calculations

Three natural radionuclides were detected in the samples at three gamma energy peaks: the ^{214}Bi radionuclide related to the ^{238}U series at 1764.5 keV, the ^{40}K radionuclide detected at 1460.80 keV, and the ^{208}Tl radionuclide related to the ^{232}Th series detected at 2614.6 keV. The activity concentrations in the samples were calculated by gamma-ray spectrometry using the following equation (Uosif & El-Taher, 2005; Mekuanint & Chaubey, 2022).

$$A = \frac{Net\ area\ (CPS)}{I_\gamma \times \xi\ M} \qquad (1)$$

Where A is the Activity concentration of the gamma spectral line in Bq/Kg, Net area (CPS) is per second under the peak, ξ is the Counting system efficiency of the energy, M is the Mass of the sample in Kg and I_γ is the intensity of the specific energy.

The radiation parameters of the samples: Radium Equivalent ($\boldsymbol{Ra_{eq}}$), Absorbed Dose Rate ($\mathbf{D_r}$), External and internal Hazard indices, and Excess lifetime cancer risk were calculated from the following equations (Al-Ghamdi, 2019; Khalda et al, 2021):

$$Ra_{eq} = A_{Ra} + 1.34A_{Th} + 0.77A_K \ldots\ldots\ldots \leq 370 \qquad (2)$$

$$D_r = 0.427A_U + 0.662A_{Th} + 0.043A_K \qquad (3)$$

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ldots\ldots\ldots \leq 1 \qquad (4)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ldots\ldots\ldots \leq 1 \qquad (5)$$

$$\mathrm{ELCR} = \mathrm{AEDE} \times \mathrm{DL} \times \mathrm{RF} \qquad (6)$$

Where A_{Ra}, A_{Th} andx A_K are the specific activity of (Bq/Kg) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. DL is the lifetime dose (70 years), and RF is the risk factor (0.05 Sv^{-1}).

4. Discussion and Results

For soil samples, the recorded values of radionuclides varied from [(79.09±0.13) to (119.33±0.13)] Bq.Kg^{-1} for ^{226}Ra, [(89.69±8.18) to (124.62±6.49)] Bq/Kg for ^{238}U, [(74.96±5.97) to (122.94±6.17)] Bq/Kg for ^{232}Th and [(381.55±11.69) to (750.74±10.26)] Bq/Kg for ^{40}K respectively, with average (125.10, 140.29, 129.13 and 651.24) Bq/Kg for all radionuclides ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K respectively, as show in Table 2.

The activity concentrations of all studied samples are higher than the permissible levels (33, 32, 45, and 412 Bq/Kg for radium, uranium, thorium, and potassium, respectively) (UNSEAR 2014), as shown in Fig. 1.

Table 2

The activity concentrations (Bq/kg) of the radioactive elements (^{226}Ra, ^{238}U, ^{232}Th and ^{40}K) of the soil samples.

Samples	^{226}Ra	^{238}U	^{232}Th	^{40}K
SE1	81.21±0.13	95.73±4.81	80.34±4.81	554.48±9.19
SE2	79.09±0.13*	91.14±6.76	74.96±5.97*	430.96±8.69
SN1	101.68±0.13	105.45±6.76	104.99±5.33	406.26±8.18
SN2	83.32±0.13	97.15±7.69	94.33±5.59	381.55±11.69*
SS1	119.33±0.13**	124.62±6.49**	122.94±6.17**	750.74±10.26**
SS2	81.91±0.13	89.63±8.18*	83.32±5.04	540.76±9.14
SW1	109.45±0.13	120.81±7.26	112.05±6.34	428.22±7.87
SW2	94.62±0.13	117.23±9.07	101.83±5.44	414.49±12.54
Average	125.10±0.13	140.29	129.13	651.24
P. L	33	32	45	412

** The highest value. * The lowest value.

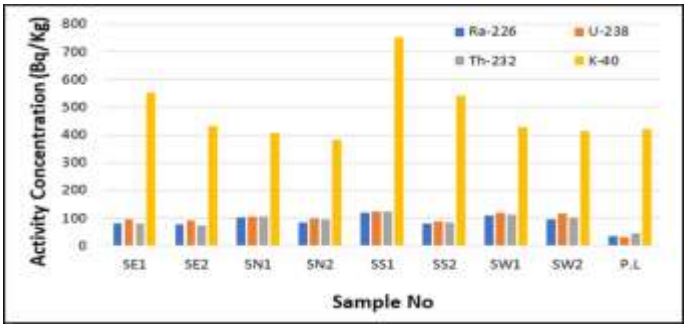

Fig. 1. The Activity Concentration of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in the soil Samples.

The radium equivalent values for all samples ranged from 219.46 to 352.95 Bq/Kg, with an average of 359.73 Bq/Kg, also the internal and external hazard results were obtained; the values of all samples ranged from 0.81 to 1.28 and 0.59 to 0.95, with averages of 1.31 and 0.97, respectively. Shown in Table 3.

Table 3

The radium equivalent activity (Ra_{eq}) and the hazards index (H_{in} and H_{ex})

Sample	Ra_{eq}(Bq.Kg)	H_{in}	H_{ex}
SE1	238.79	0.86	0.64
SE2	219.46*	0.81*	0.59*
SN1	283.10	1.04	0.76
SN2	247.59	0.89	0.67
SS1	352.95**	1.28**	0.95**
SS2	242.71	0.88	0.66
SW1	302.66	1.11	0.82
SW2	271.09	0.99	0.73
Average	359.73	1.31	0.97
P. L	370	1	1

The values of radium equivalent of the soil samples are lower than the permissible level (370 Bq/Kg) (UNSCEAR, 2014) as shown in Fig. 2.

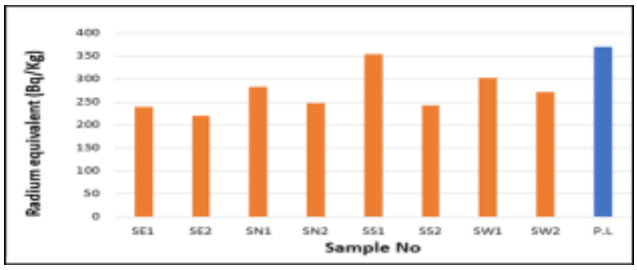

Fig. 2. Radium Equivalent (Bq/Kq) in the soil Samples.

The values of Internal Hazard of soil samples are lower than unity (UNSCEAR 2014) except for the following samples (SN1, SS1 and SW1), as shown in Fig. 3.

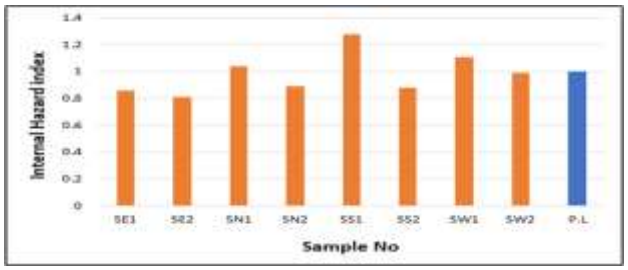

Fig. 3. Internal Hazard Index of soil Samples.

All values of external hazard for the studied soil samples are lower than unity (UNSCEAR 2014), as shown in Fig. 4.

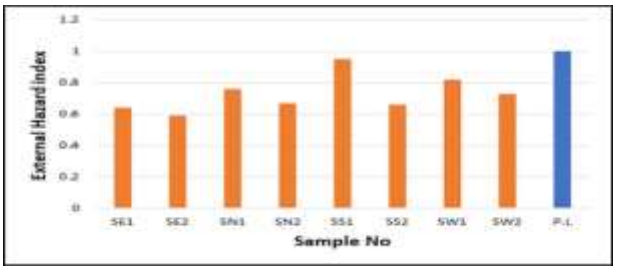

Fig. 4. External Hazard Index for soil Samples.

In Table 4, the absorbed dose rate results (Dr) of the soil samples ranged from 99.91 to 160.92 nGy/h, with an average value of 163.07nGy/h. Also, the values of the cancer risk factor (ELCR) for all samples ranged from 0.40 to 0.65, with an average value of 0.66.

Table 4

The values of absorbed dose rate and cancer risk factor for all samples

Samples	Dose rate D_r(nGy/h)	Cancer risk factor ELCR $\times 10^{-3}$
SE1	109.33	0.44
SE2	99.91*	0.40*
SN1	127.45	0.52
SN2	111.49	0.45
SS1	160.92**	0.65**
SS2	110.89	0.45
SW1	136.23	0.55
SW2	122.18	0.49
Average	163.07	0.66
P. L	84	0.29

** The highest value. * The lowest value.

All values of the absorbed dose rate for the studied soil samples are higher than the permissible level (84nGy/h) (UNSCEAR 2014) as shown in Fig. 5.

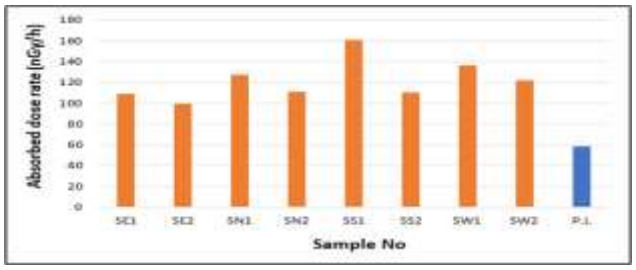

Fig. 5. Absorbed Dose Rate for Soil Samples.

The cancer risk factors for all samples are much higher than the international value of 0.29 (UNSCEAR 2014), as shown in Fig. 5.

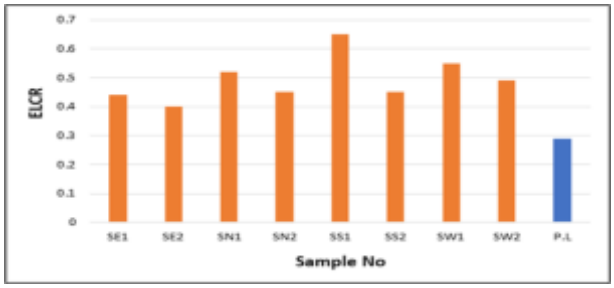

Fig. 6. The Excess Lifetime Cancer Risk for Soil Samples.

5. Conclusion

For the eight soil samples, the activity concentrations of radionuclides (^{226}Ra, ^{232}Th, ^{238}U and ^{40}K) and the radiological hazards were measured using a NaI (Tl) detector. In this work, the activity concentrations of all studied samples are higher than the permissible levels for radium, uranium, thorium, and potassium (33, 32, 45, and 412 Bq/kg, respectively).

The studied samples had radium equivalent values below the permissible level of 370 Bq/kg. The measured values of the external hazard index (H_{ex}) are lower than the recommended value for all studied samples. However, the values of the internal hazard index (H_{in}) were lower than unity for all samples except SN1, SS1, and SW1, which are higher than unity.

The Excess Lifetime Cancer Risk (ELCR) values are found to be much higher than the permissible level (0.29) (UNSCEAR, 2014) for all samples.

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