MEASURMENT OF RADIO ACTIVITY LEVELS OF RADIONULIDES IN SOIL SAMPLES AROUND LEPTIS CEMENT FACTORY USING GAMMA RAY SPECTROSCOPY

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الملخص

تستعرض هذه الورقة قياس تراكيز الفاعلية الإشعاعية للنويدات المشعة ذات المنشأ الأرضي (نواتج انحلال سلسلتي اليورانيوم–238 والثوريوم–232 وعنصر البوتاسيوم–40) وكذلك عنصر السيزيوم–137 في عدد 21 عينة ترابية تم تجميعها على أبعاد مختلفة حول مصنع لبدة للإسمنت. أجريت القياسات الإشعاعية باستعمال منظومة مطيافية جاما المبنية على كاشف الجرمانيوم عالي النقاوة. النتائج أظهرت تراكيز سلسلة الثوريوم ما بين (21.38 Bq/kg - 34.51 Bq/kg) وبمتوسط 25.89 Bq/kg ولسلسة اليورانيوم–238 ما بين (21.26 Bq/kg - 34.59 Bq/kg) وبمتوسط 25.34 Bq/kg بينما تركيز عنصر البوتاسيوم يتراوح بين (399.58 Bq/kg - 599.65 Bq/kg) وبمتوسط 458.37 Bq/kg. أما تراكيز نويدة السيزيوم–137 فكانت تتراوح من 0 إلى 14.93 Bq/kg وبمتوسط 3.91 Bq/kg. نتيجة لهذه التراكيز فقد تم حساب معايير الأخطار الإشعاعية مكافئ الراديوم، معايير الخطر الخارجي والداخلي ومعيار مستوى الفاعلية للقاطنين حول المصنع والتي تراوحت ما بين (88.72 - 122.31 Bq/kg)، (0.24-0.33)، (0.30-0.42)، (0.67-0.92) على التوالي وكانت هذه النتائج ضمن الحدود الدولية المسموح بها.

ABSTRACT

This paper presents a study of activity concentration of the terrestrial radionuclides ^{238}U, ^{232}Th, ^{40}K and artificial radionuclide^{137}Csin 21 soil samples around Leptis Cement factory. Samples were taken at different directions, depths and distances around the factory. A gamma-ray spectroscopy system based on high purity germanium (HPGe) detector was used for these measurements. The activity concentrations were found to range from 21.38 to 34.51 Bq/kg with an average value of 25.89 Bq/kg for ^{232}Th, from 21.26 to 34.59 Bq/kg with an average value of 25.34 Bq/kg for ^{238}U, and from 399.58 to 599.65 Bq/kg with an average value of 458.37 Bq/kg for ^{40}K. The activity concentrations of the artificial radionuclide^{137}Cs were found between below detection limit and 14.93Bq/kg, with an average value of 3.91 Bq/kg. The radiological hazard indices; Ra_{eq}, H_{ex}, H_{in} and I_{γ} of the natural radionuclides were calculated and found to be as follow: 88.72-122.31 Bq/kg, 0.24-0.33, 0.30-0.42 and 0.67-0.92 respectively. The results were found to be within the acceptable international limits.

KEYWORDS: Gamma Spectroscopy; Leptis Cement Factory; Soil Samples; Cesium.

INTRODUCTION

Many radionuclides occur naturally in terrestrial soils and rocks and in building materials derived from them. Upon decay, these radionuclides produce an external radiation field to which all human beings are exposed. In terms of dose, the principal

primordial (half-life comparable to the age of the earth) radionuclides are ^{40}K, ^{232}Th, and ^{238}U. Both ^{232}Th and ^{238}U head series of radionuclides that produce significant human exposures. The decay of naturally occurring radionuclides in soil produces a gamma-beta radiation field in soil that also crosses the soil-air interface to produce exposures to humans. The main factors that determine the exposure rate to a particular individual are the concentrations of radionuclides in the soil, the time spent outdoors, and the shielding by buildings. However, as the materials of which most buildings are built also contain radionuclides, the shielding by buildings of the outdoor radiation field is often more than offset by the presence of additional radionuclides in the building materials [1].

External exposures outdoors arise from terrestrial radionuclides present at trace levels in all soils. The specific levels are related to the types of rock from which the soils originate. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides. There have been many surveys to determine the background levels of radionuclides in soils, which can in turn be related to the absorbed dose rates in air. The latter can easily be measured directly, and these results provide an even more extensive evaluation of the background exposure levels indifferent countries. All of these spectrometric measurements indicate that the three components of the external radiation field, namely from the gamma-emitting radionuclides in the^{238}U and ^{232}Th series and ^{40}K, make approximately equal contributions to the externally incident gamma radiation dose to individuals in typical situations both outdoors and indoors [1].

This study is devoted to reporting the activity concentration levels of both natural radionuclides (^{238}U, ^{232}Th, ^{40}K) and artificial radionuclides (^{137}Cs) in soil samples collected from different sites of lept is area. Such data are essential in establishing baseline values of radionuclides in the soil of Libyan territories. Furthermore, the study is aimed at estimating a group of health hazard indices such as the radium equivalent activity (*Raeq*), the absorbed dose rate (*Dr*),the annual effective dose equivalent (*AEDE*), the external radiation hazard index (*Hex*) , the radioactivity level index (*I*) , and the excess lifetime cancer risk (*ELCR*). This type of measurement is of great importance in providing us with a clear picture about the radiation health hazards due to the presence of radionuclides in Libyan soils.

MATERIALS AND METHODS

Soil Sampling

In the present study a twenty-one soil samples were taken during field studies in 2016 by means of a steel sampler of 8cm diameter and 25cm height, which made it possible to obtain core of soil samples. Soil samples were collected from an area of four kilometers radius in all four directions around the factory. The samples were collected at different depths (from surface, at 20-30 cm, and at 50-60 cm). At each location, one circle with one-meter radius made, four to five core soil samples were collected, these samples were mixed into one sample [2].

Laboratory Sample Processing

The soil samples were first dried in a temperature-controlled oven at 105 ^{o}C. The dried samples were grinded, powdered and passed through a sieve of mesh size less than 2 mm, one liter Marinelli beakers were filled and packed with soil samples. The containers were thick enough for permeation of radon. The net weight of the soil was

noted. The containers were closed by caps and tightly sealed by plastic tapes were wrapped over the caps [2].

Radiometric analysis

The technique of gamma-ray spectroscopy was applied for determination of radioactivity of the samples under investigation. The spectroscopy system consisted of an HPGe detector with relative efficiency 30% and operating voltage +4000 V. The resolution of the system was 1.85 keV at 1332.5 keV peaks of ^{60}Co.

To minimize the uncertainty, the spectrum of every soil sample was collected for 86400 seconds (24 hr.). Spectrum analysis was done with help of the computer software Gamma Vision. The energy and the efficiency calibration were performed using ^{152}Eu energy peaks covering the range up to≈1408keV. The ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs activity concentrations were determined from gamma lines as shown in Table (1).

Table 1: Gamma-ray energies used for measurement of activity concentration of the radionuclides of interest.

Parent Nuclide	Daughter Nuclide	Energy keV
^{232}Th	Pb-212	238.63
		300.09
	Bi-212	727.33
	Ac-228	327.99
		338.32
		463.01
		794.95
		911.21
^{238}U	Pb-214	295.21
		351.92
	Bi-214	609.31
		768.36
		934.06
		1238.11
		2204.21
^{40}K	-	1461.00
^{137}Cs	-	661.66

Specific Activity (A_{SP})

The following equation represents the general formula used to calculate the specific activity of radioactive nuclides for a given sample [3]:

$$A_{sp} = \frac{\Sigma_{\gamma}(\mathrm{E}_i) - \Sigma_{BG}(\mathrm{E}_i)}{t} \cdot \frac{1}{m.f.\varepsilon_a(\mathrm{E}_i)} \qquad (1)$$

Where: $\sum_{\gamma}(E_i), \sum_{BG}(E_i)$ represents the total counts and the radiation background respectively, t: counting time (sec), m: the sample mass in kg. While, ε_a : the absolute detector efficiency and f: the gamma-ray intensity [3].

Radiological hazards

Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain ^{226}Ra, ^{232}Th and ^{40}K in the sample. The published maximum permissible Ra_{eq} is 370 Bq/kg [4-5]. It is given by the following equation.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.0774A_K \quad (2)$$

Where: A_{Ra}, A_{Th} and A_K represent the activities in Bq/kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K respectively.

External Hazard Index (H_{ex})

The external hazard index (given by equation 3) is an evaluation of the hazard of the natural gamma radiation. The prime objective of this index is to be certain that the radiation dose is within the permissible dose equivalent limit of 1 mSv/y [6].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

This model takes into consideration that the external hazard, which is caused by gamma rays, corresponds to a maximum radium-equivalent activity of $370\ Bqkg^{-1}$ for the material.

Internal Hazard Index (H_{in})

The internal hazard index should be less than unity for the radiation hazard to be considered negligible. Inhalation of alpha particles emitted from the short-lived radionuclides radon (^{222}Rn, the daughter product of ^{226}Ra) and thoron (^{220}Rn, the daughter product of ^{224}Ra) are also hazardous to the respiratory tract. This hazard can be quantified by the internal hazard index (H_{in}) [6]. This is given by the following equation:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

The internal hazard index should also be less than unity to provide safe levels of radon and its short-lived daughters for the respiratory organs of individuals living in the dwellings.

Annual Effective Dose Rate

In order to estimate the annual effective dose rate in air, the conversion coefficient from absorbed dose in air to effective dose received by an adult must be considered. This value is published in UNSCEAR 2000 and UNSCEAR 1993, to be $0.7SvGy^{-1}$ for environmental exposure to gamma rays of moderate energy [7-8].Equations (5) and (6) can be used to determine the annual effective dose rate.

In which the occupancy factor T is about 0.2 [1] for outdoor studies the annual effective dose equivalent is given by the equation (5):

$$D_{out}(mSv/y) = D(nGy/h) \times 8700(h/y) \times T \times 0.7(Sv/Gy) \times 10^3 \quad (5)$$

For indoor measurements (as the case in building materials) the occupancy factor T is approximately 0.8 [1] and the equation becomes:

$$D_{in}(mSv/y) = D(nGy/h) \times 8700(h/y) \times T \times 0.7(Sv/Gy) \times 10^3 \quad (6)$$

$$D_{tot}(mSv\ y^{-1}) = D_{out} + D_{in} \quad (7)$$

.

Absorbed Dose rate (D_r)

Conversion factors to transform specific activities A_{Ra}, A_{Th} and A_K of Ra, Th and K, respectively, into absorbed dose rate at 1m above the ground in ($nGyh^{-1}/Bqkg^{-1}$) can be given by the following equation [9]:

$$\dot{D}_r\,(nGyh^{-1}) = 0.461A_{Ra} + 0.623A_{Th} + 0.0414A_K \tag{8}$$

Eq. (8) was modified to include the contributions of artificial radionuclides of cesium, ^{137}Cs, as well as cosmic radiation via the following equation [9].

$$\dot{D}_\gamma\,(nGyh^{-1}) = 0.461A_{Ra} + 0.623A_{Th} + 0.0414A_K + 0.03 \times A_{Cs} + 34 \tag{9}$$

Where: 0.03 is the dose rate conversion factor to convert the activity concentration of ^{137}Cs radionuclide into absorbed dose rate.

Radioactivity level index (I_γ)

The radioactivity level index, I_γ, is generally used to assess the hazardous level of radionuclides in the human body when exposed to an amount of external (indoor or outdoor) annual effective doses of γ-radiations resulting from radioactive nuclides in soils. This index is very important for quality control of γ-radiation annual effective doses and in monitoring radiation inside human body, to ensure that such radiation does not exceed the worldwide permissible high dose values [9]. Values of I_γ can be calculated according to the following semi empirical formula [9].

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \le 1 \tag{10}$$

Excess Lifetime Cancer Risk

The excess life time cancer risk (ELCR) values are calculated using the below equation [9].

$$ELCR = D_{tot} \times D_L \times R_F \tag{11}$$

D_L is the duration of life (approximately 70 years), and R_F is the risk factor (Sv^{-1}), which reflects the fatal cancer risk per Sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public [9].

Relative Activity Concentration (q_i)

The relative activity concentration is defined as the activity concentration in each depth (layer), A_i divided by the total activity concentration at the site, A_{total} and can be calculated by equation (12) [10].

$$q_i = \frac{A_i}{A_{total}} \tag{12}$$

Exposure Rate ($\dot{X}$)

The exposure rate (R/h) can be calculated by the following equation [11].

$$\dot{X} = \Gamma \frac{\alpha}{d^2} \quad (13)$$

where:

Γ : Exposure rate constant (R.cm^2/h.mCi)

α : Activity (mCi)

d : distance (cm)

RESULTS AND DISCUSSION

From analyzing the spectra collected for soil samples, the concentrations of the radionuclides found in the samples were calculated using equation (1). Table (2) shows radiation concentrations of ^{137}Cs, ^{40}K and total activity A_{total} obtained form (^{40}K, ^{238}U and ^{232}Th) at different depths and directions around the cement factory. The highest concentration of ^{137}Cs was found in NS/20 sample. The results show that there is vertical migration in uncultivated soil samples such as WS1.3, WS2.9.While in cultivated soil samples such as SS0.8 the concentrations at different layers are almost equal due to plowing.

Table 2: Activity concentrations of^{137}Cs, ^{40}K and total activity A_{total} in each layer and ^{137}Csratios.

Sample code	$A_{^{137}Cs}$ Bq/kg	$A_{^{40}K}$ Bq/kg	A_{total} Bq/kg	$(A_{total})_{Avg}$ Bq/kg	$A_{^{137}Cs} / (A_{total})_{Avg}$
NS/0*	5.152±0.09	444.46±2.8	494.17	498.36	1%
NS/20	14.93±0.1	473.30±2.9	524.63		3%
NS/50	1.483±0.09	431.64±3.1	476.29		0.3%
WS/0	1.2108±0.07	439.51±2.7	487.36	485.50	0.2%
WS/20	2.524±0.08	445.82±2.8	489.03		0.5%
WS/50	12.06±0.1	436.49±2.7	480.10		2.5%
SS/0	2.507±0.08	456.52±2.8	506.18	510.46	0.5%
SS/20	1.851±0.08	455.79±2.7	507.78		0.4%
SS/50	0	465.87±2.9	517.42		0%
ES/0	2.166±0.08	399.58±2.6	445.59	479.77	0.5%
ES/20	4.782±0.09	452.76±2.9	504.14		0.9%
ES/50	0	440.15±2.7	489.59		0%
SS0.8/0	3.104±0.08	414.78±2.6	459.52	478.43	0.6%
SS0.8/20	4.739±0.09	433.52±2.7	482.83		0.9%
SS0.8 /50	2.108±0.08	441.70±2.8	492.93		0.4%
WS1.3/0	3.698±0.09	509.87±3	569.57	627.13	0.59%
WS1.3/20	0	599.65±4	668.75		0%
WS1.3/50	0	576.95±4.2	643.08		0%
WS/2.9/0	2.268±0.08	414.46±2.6	463.08	487.58	0.46%
WS/2.9/20	0	438.24±2.8	490.81		0%
WS/2.9/50	0	454.64±2.9	508.85		0%

*(Direction, Soil/Depth)

Figure (1) shows the average activity concentration of ^{137}Cs in soil samples. The average activity concentration of ^{137}Cs as well as its ratio to the total average activity in the site values are presented in Table (3). The highest average concentration was found

in sample NS at north direction while the lowest average concentration was found in sample SS at south direction.

Table 3: The Ratio between average activity concentration of ^{137}Cs and total average activity in the site.

Sample code	$(A_{^{137}Cs})_{Avg}$ Bq/kg	$(A_{^{137}Cs})_{Avg} / (A_{total})_{Avg}$ Bq/kg
NS	7.188±0.09	1.4%
WS	5.265±0.08	1.1%
SS	2.18±0.08	0.43%
ES	3.474±0.09	0.72%
SS0.8	3.317±0.08	0.7%
WS1.3	3.698±0.09	0.59%
WS2.9	2.268±0.08	0.46%

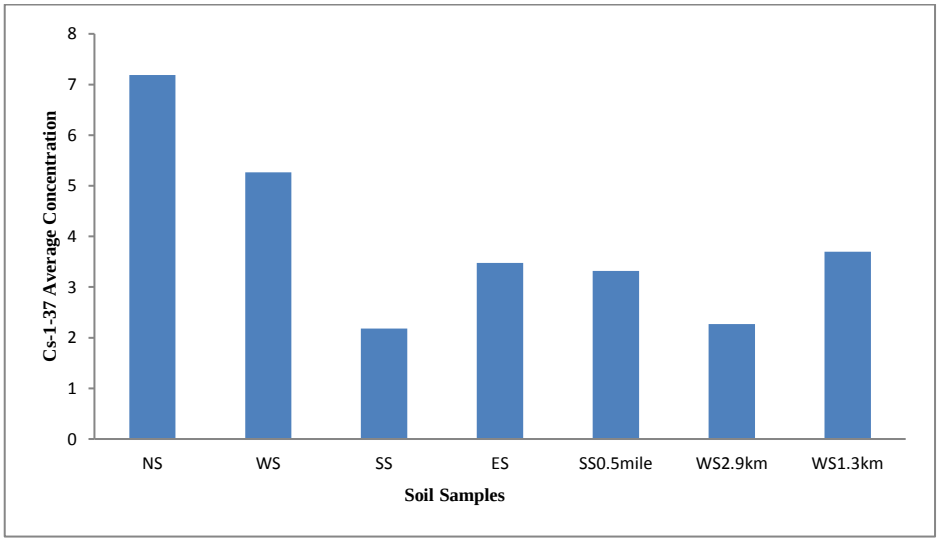

Figure 1: The average activity concentration of ^{137}Cs distribution for Leptis cement factory.

As it can be seen Table (4) shows the range of measured activity concentration of ^{232}Th varies from 21.38 to 34.51Bq/kg, with an average value of 25.89 Bq/kg. The maximum value of the activity concentration of ^{232}Thradionuclide was found in soil sample WS1.3 from area west of Leptis cement factory, whereas the minimum concentration value was in sample ES collected from area east of Leptis cement factory. This difference may be attributed to the geochemical composition and soil types in that particular area. The measured activity concentrations of^{238}U were found to range from 21.26 to 34.59 Bq/kg, with an average value of 25.34 Bq/kg. Moreover, the range of activity concentration of ^{40}K was found to be from 399.58 to 599.65 Bq/kg, with an average value of 458.37 Bq/kg. When comparing the average activity concentrations of ^{238}U and ^{232}Th, the results show that they were very similar and fall within the acceptable international limits. The activity concentration of ^{40}K was found to be higher than the reported international levels. This is due to the abundance of this radioactive element in the soil and the effect of marl and clay quarrying activities near the factory.

Table 4: Activity concentrations for Leptis soil samples.

Sample code	Activity concentrations Bq/kg								
	No. of samples	^{232}Th			^{238}U			^{40}K	
		Min.	Max.	Avg	Min.	Max.	Avg	Min.	Max.	Avg
NS	3	23.39	26.99	24.97	21.26	25.18	23.59	431.64	473.30	449.80
WS	3	21.49	24.42	22.63	21.64	23.43	22.26	436.49	445.82	440.61
SS	3	24.81	27.17	26.17	24.38	25.53	24.90	455.79	465.87	459.39
ES	3	21.38	26.59	24.39	24.23	24.79	24.55	399.58	452.76	430.83
SS0.8	3	22.53	27.18	24.97	22.21	24.12	23.46	414.78	441.70	430.00
WS1.3	3	29.22	34.51	32.17	30.48	34.59	32.81	509.87	599.65	562.16
WS2.9	3	24.84	26.65	25.96	23.78	27.56	25.84	414.46	454.64	435.78
Total	21	21.38	34.51	25.89	21.26	34.59	25.34	399.58	599.65	458.37

The calculated values of Ra_{eq} for the same soil samples (see Table 5) were found to range from 88.72 to 122.31 Bq/kg, with an average value of 97.84 Bq kg^{-1} For most inspected soil samples, Ra_{eq} values are lower than the international acceptable limit of 370 Bq/kg. The calculated values of H_{ex} were found to range from 0.24 to 0.33, with an average value of 0.26, all these values are much less than unity. The values of H_{in} were found to range between 0.30 and 0.42 with an average value of 0.33. The obtained values of H_{in} are less than 1. The calculated values of I_{γ} were found to range from 0.67 to 0.92 with an average of 0.74, they all values were lower than 1.

Table 5: The radium equivalent (Ra_{eq}), the external (H_{ex}) and the internal (H_{in}) hazard indices, and radioactivity level index (I_{γ}) of soil samples collected from Leptis soil samples.

Sample code	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	I_{γ}
NS	94.11±2.9	0.25	0.32	0.71
WS	88.72±2.7	0.24	0.30	0.67
SS	97.85±2.8	0.26	0.33	0.74
ES	92.77±2.7	0.25	0.32	0.70
SS0.8	92.45±2.7	0.25	0.31	0.69
WS1.3	122.31±3.7	0.33	0.42	0.92
WS2.9	96.69±2.8	0.26	0.33	0.72
World-wide value	370	<1	<1	<1

Table (6) shows the average activity concentration values of ^{137}Cs in all collected soil samples, they were found to range from 2.18 to 7.188 Bq/kg with an average value of 3.91 Bq/kg, which is within the permissible limit of 20 Bq/kg [12-13]. The minimum activity concentration value of ^{137}Cs was obtained for a soil sample collected from south area of Leptis cement factory, whereas the maximum value was measured in a soil sample collected from North area. The calculated values of D_r were found to vary from 42.60 to 58.44 nGy/h, with an average value of 46.79 nGy/h. The measured average absorbed dose rate in the air is lower than the recommended international limits of (57 nGy/h published in UNSCEAR 2000). The weighted mean value of 46.79 nGy/h represents 82% of the world average outdoor exposure due to terrestrial gamma radiation. The measured values of D_{γ} range from 76.76 to 92.55 nGy/h and have an average value of 80.91 nGy/h.

Table 6: The activity concentrations, the absorbed dose rate (D_r), and the total dose rate (D_γ) of ^{137}Cs of the soil samples collected from Leptis Soil Samples.

Sample code	Average activity concentration of ^{137}Cs	D_r (nGy/h)	D_γ (nGy/h)
NS	7.188	45.05	79.27
WS	5.265	42.60	76.76
SS	2.18	46.79	80.85
ES	3.474	44.35	78.45
SS0.8	3.317	44.17	78.27
WS1.3	3.698	58.44	92.55
WS2.9	2.268	46.13	80.19

Table (7) represents the calculated values for D_{out}, D_{in}, and D_{tot} as well as the excess lifetime cancer risk (ELCR). The values for D_{out}, D_{in}, and D_{tot} averages were 0.057, 0.23, and 0.29 mSv/year respectively. In comparison to global measured values, these values were all below the assigned worldwide values of (0.07, 0.4, and 0.47 mSv/year published in UNSCEAR 2000), respectively. The *ELCR* values are ranged between 0.91×10^{-3} and $1.26\times10^{-3}$and the average value is about 1.01×10^{-3}. The calculated average value of *ELCR* for all samples is higher than the world average of 0.29×10^{-3} [9]. This indication of the possibility of developing cancer cases among individuals cannot be neglected.

Table 7: The outdoor ($D_{outdoor}$), indoor (D_{indoor}), and total annual effective dose equivalent (D_{total}) and excess lifetime cancer risk (ELCR) of the soil samples collected from Leptis soil samples.

Sample code	D_{out} (mSv/yr)	D_{in} (mSv/yr)	D_{total} (mSv/yr)	ELCR ($\times10^{-3}$)
NS	0.05	0.22	0.27	0.95
WS	0.052	0.21	0.26	0.91
SS	0.057	0.23	0.29	1.02
ES	0.054	0.22	0.27	0.95
SS0.8	0.054	0.22	0.27	0.95
WS1.3	0.072	0.29	0.36	1.26
WS2.9	0.057	0.23	0.29	1.02

CONCLUSION

Analysis of gamma spectra collected for soil samples around Leptis cement factory showed the presence of natural radioactivity and Cs^{137} contaminations. The contamination values are within the acceptable international limits. Radiological hazards indices associated with radioactivity concentrations were calculated and found to be below the international limits.

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