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Evaluation of Natural Radioactivity in Selected Soil Samples from the Archaeological of Girsu City in Dhi-Qar Governorate, Iraq

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ABSTRACT

The specific activity of natural radionuclides in (18) soil samples collected from antiquities area of Girsu city in Dhi-Qar Governorate (31.0459863N, 46.2534257E) in southern Iraq have been studied and evaluated. Experimental results were obtained by using a gamma ray spectrometer analysis system consists
of a scintillation detector Sodium Iodide activated by Thallium NaI(Tl) of (3"×3") crystal dimension at the laboratory of radiation detection and measurement in Science Collage, University of Kufa. The spectrometer has been calibrated for energy by acquiring a spectrum from four standard sources of gamma radiations supplied by spectrum techniques (LLC). The measuring time of all soil samples is (18000 seconds), it was found that, for Girsu city the soil specific activity ranges from (22.68±3.94 to 14.69±2.24) Bq/kg for $^{238}_{92}U_{146}$, from (17.41±2.23 to 11.23±2.34) Bq/kg for $^{232}_{90}Th_{142}$ and from (346.49±10.68 to 266.96±10.55) Bq/kg for $^{40}_{19}K_{21}$. The results have been compared with the acceptable data of the worldwide literatures. In order to evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity (Ra_{eq}), the gamma absorbed dose rate (D), the annual effective dose rate and the both (external and internal) hazard index have been calculated and compared with the acceptable values of the worldwide average (UNSCEAR, 2000).

Keywords: Natural radioactivity; Specific activity; NaI(Tl) detector; Gamma-ray spectrometry; Radium equivalent activity.

دراسة وتقييم النشاط الإشعاعي الطبيعي في عينات تربة مختارة من أثار مدينة غيرسو في محافظة ذي قار في العراق

الخلاصة

تم دراسة وتقييم النشاط النوعي للانوية المشعة الطبيعية في (18) عينة من التربة التي تم جمعها من المنطقة الاثرية لمدينة غيرسو في محافظة ذي قار (31.0459863N و 46.2534257E) جنوب العراق. لقد تم استنتاج النتائج العملية بأستخدام منظومة تحلل طيف أشعة كاما المتكونة من الكاشف الوميضي أيوديد الصوديم المطعم بالثاليوم NaI(Tl) بقطر بلورة (3"x 3") في مختبر الكشف والقياس عن الاشعاع في كلية العلوم – جامعة الكوفة. لقد تم معايرة المطياف بالنسبة للطاقة المطلوبة للطيف من أربعة مصادر قياسية لاشعة كاما

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المجهزة بتقنية المطياف (LLC). لقد تم تحديد زمن القياس بـ 18000 ثانية، حيث وجدنا بأن النشاط النوعي لمدينة غيرسو تراوحت قيمته بين (2.68±3.94 إلى 14.69±2.24) بكرل/كغم للـ $^{238}_{92}U_{146}$ و بين (17.41±2.23 إلى 11.23±2.34) بكرل/كغم للـ $^{232}_{90}Th_{142}$ و بين (346.49±10.68 إلى 266.96±10.55) بكرل/كغم للـ $^{40}_{19}K_{21}$. تم مقارنة النتائج الحالية مع البيانات المقبولة في الادبيات العالمية. ولاجل تقييم المخاطر الخاصة بالطب الاشعاعي للنشاط الاشعاعي الطبيعي، فقد تم حساب كل من النشاط المكافئ للراديوم (Raeq) ومعدل جرعة أمتصاص كاما (D) والمعدل السنوي للجرعة الفعالة ومؤشر المخاطر (الخارجي والداخلي) ومقارنتهم بالقيم المقبولة للمعدلات العالمية (UNSCEAR, 2000).

INTRODUCTION

Natural radioactivity is present in the human environment due to the presence of Cosmogenic and primordial radionuclides in the Earth's crust [1]. Cosmogenic radionuclides are produced by the interaction of cosmic-rays with atomic nuclei in the atmosphere, while primordial ones (terrestrial background radiation) were formed by the process of nucleo-synthesis [2]. Only those radionuclides with half-lives comparable to the age of the Earth, e.g., $^{40}_{19}K_{21}$ and members of the $^{238}_{92}U_{146}$ and $^{232}_{90}Th_{142}$ series, can still be found today in different geological materials [3]. Natural Occurring Radionuclides Materials (NORM) is known to be present in rocks and soil. Specific levels of terrestrial background radiation mainly depend on the geological and geographical conditions [4,5]. Scientific motivation for this study is based on measuring concentrations and distributions of natural radionuclides $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ in soil samples from different locations of two regions of Sumer civilization antiquities in Dhi-Qar governorate, southern of Iraq, in order to identify and quantify significant gamma-emitting radionuclides in soil samples from the renowned present Sumerian city, Girsu, regardless its geo-genesis or chemical composition. Girsu city (31.5622565N, 46.1768233E) is about 7.6 km far from Al-Shatrah district. The specific activities have been investigated in the present work for $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ in (18) soil samples collected from (6) places in the Girsu archaeological city, as shown in figure (1).

Figure (1): The sampling locations of 18 soil samples in the antiquities area of Girsu city in Dhi-Qar governorate in the southern Iraq [6].

Materials and methods

Soil samples were collected from (18) locations in the antiquities area of Girsu city in Dhi-Qar governorate. Sampling locations are marked in figure (1). The samples were ground into a fine powder with a particle size less than 100 μm and then dried in a temperature-controlled furnace at (110 °C) for (24 h) to remove moisture. Each sample stored in a sealed polyethylene marinelli beaker for 30 days to achieve the secular equilibrium. This marinelli beaker was used as sampling and measuring container. Before use the containers were washed with hydrochloric acid and rinsed with distilled water [7].

The activities of natural radionuclides $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ in the samples was determined by gamma rays spectrometry NaI(Tl) of (3"×3") crystal dimensions, supplied by (Alpha Spectra, Inc.-12I12/3), coupled with a multi-channel analyzer (MCA) (ORTEC-Digit Base) with range of 4096 channel joined with analog to digital converter (ADC) unit, through interface. The spectroscopic measurements and analysis are performed via the (MAESTRO-32) software into the PC of the laboratory.

The specific activity concentration of $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ were determined using the gamma-lines (1765keV) $^{214}_{69}Bi_{145}$, (2614keV) $^{208}_{67}Tl_{141}$ and (1460keV) γ-line respectively. Counting time interval was (18000 seconds). The background spectrum was recorded immediately after or before the sample counting.

The activity concentration of each radionuclide was calculated by the following equation [8]:

$$A = \frac{C}{\varepsilon \cdot I_{\gamma} \cdot m \cdot t} \qquad ...(1)$$

Where

A is the activity concentration of the radionuclides (Bq/kg), C is the count rate, ε is the detector efficiency for the specific gamma-ray energy, I_{γ} is the absolute transition probability of gamma decay, m is the mass of sample (kg) and *t* is the counting time in second.

Exposure radiation has been defined in terms of radium equivalent activity Ra_{eq} in (Bq/kg) to compare the specific activity of materials containing different amounts of $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$. Since the distributions of $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ in soil are not uniform, The radium equivalent activity can be calculated using the following relation [9]:

$$Ra_{eq} = A_U + 1.43\ A_{Th} + 0.07\ A_K \qquad ...(2)$$

Where

A_U, A_{Th} and A_K are the activity concentrations of $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ in (Bq/kg), respectively. While defining Ra_{eq} activity according to equation (1), it has

been assumed that (370) Bq/kg for $^{238}_{92}U_{146}$ or (259) Bq/kg for $^{232}_{90}Th_{142}$ or (4810) Bq/kg for $^{40}_{19}K_{21}$ produce the same gamma dose rate [10].

The external gamma absorbed dose rate in the air at (1 m) above ground level was calculated from the measured activities of $^{238}_{92}U_{146}$, $^{232}_{90}Th_{142}$ and $^{40}_{19}K_{21}$ in

soil assuming that other radionuclides, such as $^{137}_{55}Cs_{82}$, $^{113}_{48}Cd_{65}$ and $^{238}_{92}U_{146}$ series can be neglected as they contribute very little to the total dose from environmental background[11]. The calculations were performed according to the following equation [9]:

$$D=0.462A_U+0.604A_{Th}+0.042A_K \quad \ldots(3)$$

Where

D is the dose rate in (nGy/h). The external hazard index, H_{ex}, is defined as [10]:

$$H_{ex} = A_U / 370+A_{Th}/259+A_K/4810 \quad \ldots.(4)$$

There is another hazard index called internal hazard index H_{in}, which is determined as follows [10]:

$$H_{in} = A_U/ 185+A_{Th}/259+A_K/4810 \quad \ldots.(5)$$

The value of this index must be less than unity in order to keep the radiation hazard insignificant. The maximum value of H_{ex} and H_{in} equal to unity corresponds to the upper limit of radium equivalent activity (370Bq/kg).

To estimate the annual effective dose, the following must be taken into consideration [12]:

(a) The conversion coefficient of absorbed dose in air to effective dose.

(b) The indoor occupancy factor.

Using the dose rate data obtained from the concentration values of natural radionuclides in soil, adopting the conversion factor of (0.7 Sv/Gy)[8] from absorbed dose rate in air to effective dose received by adults and considering that people in Girsu city, on the average, spent (20%) of their time outdoors, the annual effective doses are calculated[8]:

$$\text{Outdoor annual effective dose (Sv)} = D \times 24 \times 365 \times 0.7 \times 0.20 \quad \ldots(6)$$

$$\text{Indoor annual effective dose (Sv)} = D \times 24 \times 365 \times 0.7 \times 0.80 \quad \ldots.(7)$$

Also the Elemental concentrations were determined by using the relation between the specific activity and the Elemental concentrations as shown in the table (1):

Table (1) Conversion of radioelement concentration to specific activity [11].

Elemental concentrations	Specific activity (Bq/kg)
1% ($^{40}_{19}K_{21}$)	313
1ppm ($^{238}_{92}U_{146}$)	12.35
1ppm ($^{232}_{90}Th_{142}$)	4.06

Results and Discussion

The specific activity values of ($^{40}_{19}K_{21}$, $^{238}_{92}U_{146}$ and $^{232}_{90}Th_{142}$) radionuclides for (18) soil samples collected from (6) different locations in the archaeological Girsu city, were taken from depth (0-5, 5-10 and 10-15 cm) for each location are tabulated in table (2).

The values have been found to lie in the range of (266.96±10.55 Bq/kg to 364.49±10.68) Bq/kg with an average of (314.89±11.44) Bq/kg for $^{40}_{19}K_{21}$, from (14.69±2.24 Bq/kg to 22.68±3.94) Bq/kg with an average value of (18.86±3.11) Bq/kg for $^{238}_{92}U_{146}$ and (11.23±2.34 Bq/kg to 17.41±2.23) Bq/kg with an average value of (14.39±2.28) Bq/kg for $^{232}_{90}Th_{142}$.

Generally, from table (2), it can be seen that the specific activity values in all soil samples of Girsu city region are less than the worldwide average (35 Bq/kg for $^{238}_{92}U_{146}$, 30 Bq/kg for $^{232}_{90}Th_{142}$ and 400 Bq/kg for $^{40}_{19}K_{21}$)[13]. The Radium equivalent activities are calculated and listed in table (2), Ra_{eq} values vary from (53.88±6.23 to 70.89±7.75) Bq/kg with an average value of (63.66±7.26) Bq/kg. It can seen be that the Ra_{eq} values for all soil samples are lower than the recommended maximum value (370Bq/kg) [13].

The absorbed dose rate one meter above the ground, indoor and outdoor annual effective dose for Girsu city samples were calculated and given in table (3). The result shows that the highest value is (33.96 nGy/h), whereas the lowest value is (26.01 nGy/h) and the average value (30.55 nGy/h). The values of absorbed dose rate for soil sample under limit the worldwide average value (55 nGy/h) [14]. In addition, the calculated values of absorbed dose rate were given in table (3) and as well as, the values of indoor and outdoor annual effective dose were calculated ranged from (0.127 to 0.166 mSv/y) and from (0.023 to 0.042 mSv/y) respectively. The average values of annual indoor and outdoor effective dose are (0.149 and 0.037 mSv/y) respectively. All values of indoor and outdoor annual effective dose were less than (1

mSv/y) recommended by International Commission on Radiological Protection (ICRP 1993) [14].

Table (4) tabulated the results of elemental concentrations of ($^{40}_{19}K_{21}$, $^{238}_{92}U_{146}$ and $^{232}_{90}Th_{142}$) which are vary from (0.852 to 1.164 percent) with an average value (1.006 percent), from (1.201 to 1.836 ppm) with an average value (1.527 ppm) and from (2.766 to 4.287 ppm) with an average value (3.541 ppm) respectively.
The external and internal hazard indices of soil samples for Girsu city are tabulated in table (4). In this table, one can see that the values of external hazard index have been found to lie in the range of (0.145 to 0.191) with an average of (0.172). In concern with the minimum value of internal hazard index is (0.185) and the maximum value is (0.251) with an average of (0.223).

Generally the concentrations of ($^{40}_{19}K_{21}$, $^{238}_{92}U_{146}$ and $^{232}_{90}Th_{142}$) in the soil samples are relatively lower than that the world average (1.28 %), (2.83) ppm and (7.38) ppm respectively. The worldwide averages according to UNSCEAR (2008) are (400 Bq/kg, 35 Bq/kg and 30 Bq/kg) respectively.

Table (2): Specific Activity and Radium equivalent activity of soil samples form Girsu city region.

Sample code	Depth (cm)	Specific activity (Bq/kg)			Ra_{eq} (Bq/kg)
		$^{40}_{19}K_{21}$	$^{238}_{92}U_{146}$	$^{232}_{90}Th_{142}$	
G1	0-5	307.77±10.88	14.84±2.75	11.31±2.15	54.71±6.65
	5-10	266.96±10.55	20.15±2.94	15.22±2.52	62.47±7.36
	10-15	339.95±12.46	18.1±2.87	13.53±2.62	63.61±7.57
G2	0-5	315.11±10.72	16.17±3.07	13±2.21	59.03±7.06
	5-10	291.1±10.16	14.69±2.24	11.73±2.24	53.88±6.23
	10-15	278.84±12.2	17.33±2.84	13.73±2.23	58.44±6.97
G3	0-5	364.49±10.68	19.71±2.43	13.73±2.1	67.41±6.25
	5-10	312.39±11.79	18.5±3.17	15.08±2.29	64.11±7.35
	10-15	315.44±11.38	22.25±2.87	15.87±2.54	69.23±7.39
G4	0-5	357.97±11.73	17.87±3.37	13.9±2.34	65.32±7.62
	5-10	295.88±10.86	22.68±3.94	17.41±2.23	70.36±7.97
	10-15	276.2±10.4	22.31±2.95	16.95±2.13	67.81±6.79
G5	0-5	296.22±10.9	21.17±3.86	16.65±2.15	67.79±7.78
	5-10	305.19±9.48	17.07±3.21	11.23±2.34	56.63±7.28
	10-15	316.21±10.98	20.86±2.85	15.69±2.23	67.65±6.89
G6	0-5	353.96±16.02	16.67±4.13	12.92±2.25	62.41±8.57
	5-10	332.99±12.08	17.68±3.25	14.6±2.14	64.2±7.23
	10-15	341.31±12.65	21.37±3.34	16.25±2.41	70.89±7.75
Max		364.49±10.68	22.68±3.94	17.41±2.23	70.89±7.75
Min		266.96±10.55	14.69±2.24	11.23±2.34	53.88±6.23
Average		314.89±11.44	18.86±3.11	14.39±2.28	63.66±7.26
Worldwide average[15]		400	35	30	370

Table (3): Absorbed dose rate and annual effective dose of soil samples form Girsu city region.

Sample Code	Depth (cm)	Absorbed dose rate (nGy/h)	Annual effective dose (mSv/y)	
			Indoor	Outdoor
G1	0-5	26.5	0.13	0.026
	5-10	29.71	0.145	0.025
	10-15	30.7	0.15	0.023
G2	0-5	28.47	0.139	0.032
	5-10	26.01	0.127	0.036
	10-15	27.97	0.137	0.037
G3	0-5	32.58	0.159	0.04
	5-10	30.72	0.15	0.037
	10-15	33.07	0.162	0.04
G4	0-5	31.57	0.154	0.038
	5-10	33.42	0.163	0.041
	10-15	32.16	0.157	0.039
G5	0-5	32.27	0.158	0.039
	5-10	27.37	0.134	0.033
	10-15	32.35	0.158	0.039
G6	0-5	30.24	0.148	0.037
	5-10	30.89	0.151	0.037
	10-15	33.96	0.166	0.042
Max		33.96	0.166	0.042
Min		26.01	0.127	0.023
Average		30.55	0.149	0.037
Worldwide average[15]		55	≤ 1	≤ 1

Table (4): Elemental concentrations of $^{40}_{19}K_{21}$, $^{238}_{92}U_{146}$ and $^{232}_{90}Th_{142}$ and the external and internal hazard index of soil samples form Girsu city region.

Sample Code	Depth (cm)	Elemental concentrations			Hazard index	
		$^{40}_{19}K_{21}$ (%)	$^{238}_{92}U_{146}$ (ppm)	$^{232}_{90}Th_{142}$ (ppm)	H_{ex}	H_{in}
G1	0-5	0.983	1.201	2.784	0.147	0.187
	5-10	0.852	1.631	3.748	0.168	0.223
	10-15	1.086	1.465	3.331	0.171	0.22
G2	0-5	1.006	1.309	3.203	0.159	0.203
	5-10	0.93	1.189	2.889	0.145	0.185
	10-15	0.891	1.403	3.382	0.157	0.204
G3	0-5	1.164	1.595	3.382	0.182	0.235
	5-10	0.998	1.497	3.714	0.173	0.223
	10-15	1.007	1.802	3.908	0.187	0.247
G4	0-5	1.143	1.447	3.424	0.176	0.224
	5-10	0.945	1.836	4.287	0.19	0.251
	10-15	0.882	1.806	4.174	0.183	0.243
G5	0-5	0.946	1.714	4.101	0.183	0.24
	5-10	0.975	1.382	2.766	0.152	0.199
	10-15	1.01	1.689	3.864	0.182	0.239
G6	0-5	1.13	1.35	3.183	0.168	0.213
	5-10	1.06	1.431	3.596	0.173	0.221
	10-15	1.09	1.73	4.001	0.191	0.249
Max		1.164	1.836	4.287	0.191	0.251
Min		0.852	1.201	2.766	0.145	0.185
Average		1.006	1.527	3.541	0.172	0.223
Worldwide average[15]		1.28	2.83	7.38	≤ 1	≤ 1

It is evident from the calculated data, the maximum values of external and internal hazard indices were (0.249 and 0.324) and the minimum values of external and internal hazard indices were (0.158 and 0.18), the average values of external and internal hazard index are (0.201 and 0.257) respectively. It can be seen that all the soil samples of this region under the permissible limit for (H_{ex} and H_{in}) which is of value 1[14].

The activity levels due to terrestrial background radiation are related to the type of rock from which the soil originates. As regard of activity with respect to depth, no correlation was found between activity and depth. The measured values of radioactivity show that it is randomly distributed in different depth of the soil. The selected spectra of some soil samples from this region are shown in figure (2).

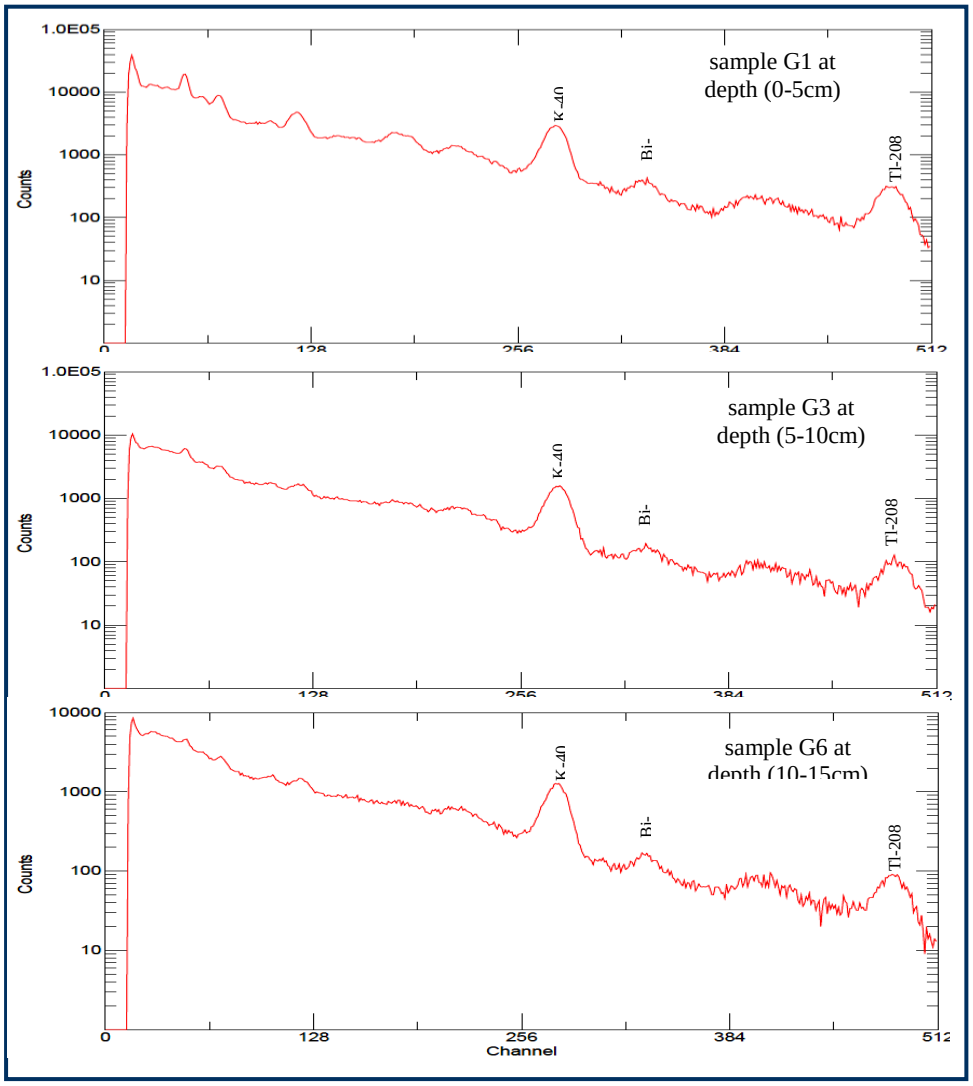

Figure (2): Gamma-ray spectra of three soil samples from Girsu city region.

CONCLUSIONS

The environmental monitoring of natural background radiation using Sodium Iodide NaI(Tl) detector revealed the distribution of the natural radiation levels in all the soil samples measured. From the obtained result, one could see that the distribution was not uniform. Also artificial radionuclide was not detected in any of the measured samples. The obtained results confirm some conclusions as below:

1- The present work has shown that the radioactivity concentration of ${}^{40}_{19}K_{21}$, ${}^{238}_{92}U_{146}$ and ${}^{232}_{90}Th_{142}$ are generally acceptable in comparison with the worldwide average.

2- Since the radiation levels were within permissible limits, thus the radioactive hazard is low for human beings (employees and tourists) in these areas.

3- Specific activity of ${}^{40}_{19}K_{21}$ is much higher than that of ${}^{238}_{92}U_{146}$ and ${}^{232}_{90}Th_{142}$.

4- The results of the present work demonstrate that the gamma absorbed dose rate of samples varies appreciably from one sample to another due to the variation of ${}^{40}_{19}K_{21}$, ${}^{238}_{92}U_{146}$ and ${}^{232}_{90}Th_{142}$ contents.

5- The estimated average absorbed dose rates in air for the studied area are below or comparable with world average (55nGy/h).

6- All average values of annual indoor and outdoor effective dose were lower than the permissible limit.

7- The gamma-ray spectrometry is a good technique for measuring natural radioactivity in soil.

8- The measured values of radioactivity show that they are randomly distributed with depth.

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