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Specific Activity and Radiation Hazard of Radionuclides in Wheat and Bean Produced Near Shazand, Iran

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ABSTRACT

Introduction: Radionuclides found in foods are harmful to human health. Wheat and bean are among the most important food ingredients in the world. Therefore, this study aimed to determine the specific activity of natural radionuclides in wheat and bean produced near the refinery complex plant.

Material and Methods: In order to determine the specific activity of radionuclides, the gamma-ray spectrometry method was used employing a high-purity germanium detector with a relative efficiency of 80%.

Results: Our findings showed that the specific activity of the ^{226}Ra isotope of radium had the ranges <1.31-5.27 and <1-5.06 Bq/kg for wheat and bean samples, respectively. Moreover, the specific activity of the ^{232}Th isotope of thorium was in the range of not detected (ND)-4.09 and ND-3.62 Bq/kg with the mean values of 2.19 and 2.69 Bq/kg for wheat and bean samples, respectively. The specific activity of the ^{40}K isotope of potassium was obtained as 103.19-168.94 and 129.22-568.98 Bq/kg with the mean values of 142.21 and 458.37 Bq/kg for wheat and bean samples, respectively. The annual effective dose for wheat and bean intake was 0.11-0.52 and 0.02-0.18 mSv, respectively. Furthermore, the mean of excess lifetime cancer risk for wheat and bean samples was calculated as 1.06×10^{-3} and 0.11×10^{-3}, respectively. The latter values are lower than the world average for bean samples.

Conclusion: According to the results of this study, the radiological parameters of wheat were higher than the global average and reference value, which may be due to ash dispersion in this area. For bean, these parameters were lower than the mean value. As a result, it could be concluded that bean is not considered as a threat to consumer health.

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Introduction

Wheat is one of the important food components in the world and contributes to 19% of the energy and 20.8% of the protein worldwide. According to the reports of the Food and Agriculture Organization (2012), wheat makes up about half of the world's diet and its annual average per capita consumption in the world is 67 kg [1]. Furthermore, beans are a rich source of protein with an average consumption per capita of 50 and 9 kg in the world and Iran, respectively [1]. Approximately 13,400,000 metric tons of wheat were produced in Iran in 2018 and per capita consumption for Iran was estimated as 167.6 kg/y.

Natural radioactive is widespread throughout the earth in various geological forms, including clay, rocks, plants, food, and water [2-4]. The specific concentration of radioactive nuclei in the soil and plants depends on soil constituents and environmental pollution resulting from chemical fertilizers, oil, coal burning, and mineral extraction, such as uranium and thorium production [5].

Nuclear radiation is harmful to human health and one of its important sources is the inhalation of radon gas and the consumption of radionuclides in food. The radiation coming from the disintegration of the ^{238}U and ^{232}Th series, as well as ^{40}K nuclei, are considered as the most important environmental radiation sources [6,7]. The average annual effective dose absorbed by people from the natural sources of ionizing radiation is estimated to be 2.4 mSv/y, of which 0.32 mSv/y comes from the ingestion of radionuclides [8].

In addition to natural nuclides, the ^{137}Cs radioactive nuclei occur in nature artificially. The source of this particular radioactive nucleus can be human activities in the field of energy production and military operations, namely nuclear weapons testing. Moreover, nuclear incidents, such as the Fukushima earthquake (2011) and the Chernobyl accident (1986) can contribute to this type of radiation [9].

Thorium, uranium, and potassium, depending on the biological effect, accumulate in certain organs of the body. The lungs and kidneys are known as the target organs for uranium storage. Thorium and potassium accumulate in the lungs, liver, bones, and muscles. This particular nucleus can cause

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biochemical changes, including chromosomal mutations or cancer when sits in the organs [10]. With this background in mind, the purpose of this study was to investigate the specific activity of natural radionuclides in the wheat and bean produced in the vicinity of a petrochemical complex. Biological parameters, such as the average annual effective dose and cancer risk were calculated for the collected samples.

Materials and Methods

Sampling and Sample Preparation

Samples were collected from villages near Shazand Petrochemical Company, Iran. The samples were cleaned and then washed with double distilled water. Drying of the samples was carried out for 24 h at room temperature and 6 h in the oven of 80^{0}C. Samples were milled and passed through a No. 40 mesh (0.635 mm) to form a fine powder (Figure 1). A total of 200 g of the prepared samples was packed in 300 cm^3 polyethylene containers and sealed with silicone glue. Samples were kept in the laboratory for at least 50 days, which was necessary for the balance of activity between ^{226}Ra and ^{222}Rn according to Equation 1 [11].

$$t_m = \frac{ln\,(\lambda_2-\lambda_1)}{\frac{\lambda_2}{\lambda_1}} \quad (1)$$

Where t_m is the activity balance time, and λ_1 and λ_2 represent the decay constants of ^{226}Ra and ^{222}Rn radionuclides, respectively.

Spectroscopy and Spectrum Analysis

In order to determine the amount and type of radioactive nuclei, the gamma spectrometry method was utilized. High-Purity Germanium (HPGe) detector (GCD30195 BSI manufactured by Baltic Scientific Instruments company, LV-1005, Latvia) was used with a relative efficiency of 80%, a working voltage of 3000 V, and an energy resolution of 1.85 keV for the gamma line of 1332.52 keV due to ^{60}Co (E&G Ortec company, Tennessee, USA). The energy and efficiency calibrations were performed by standard sources containing ^{152}Eu, ^{241}Am, and ^{137}Cs radionuclides of known activity. The absolute detector-sample efficiency was calculated using Equation 2:

$$\varepsilon = \frac{G-B}{A\times BR(\%)\times T} \times 100 \quad (2)$$

Where ε indicates the energy efficiency of the gamma radiation of the detector-sample set. G and B represent the gross and background count under the peak of full energy, respectively. A denotes the radionuclide known activity BR is the gamma branching factor - Radiation intensity emitted by the radionuclide, and T represents the time of spectra acquisition [12, 13].

Specific Activity Measurement

According to the results of gamma-ray spectra, the specific activity value was measured using Equation 3 [11]:

$$A = \frac{G-B}{\varepsilon\times Br\times M\times T} \quad (3)$$

Where A denotes the specific activity of radionuclide in the sample. G, B, BR, and T are the same as the aforementioned equation and ε represents the energy efficiency for the gamma-ray of detector-sample setup. Moreover, m denotes the mass of samples in kg.

The activity concentration of ^{226}Ra was calculated by applying the gamma-ray line of ^{214}Pb (351.93 keV) with the intensity of 37.6% and ^{214}Bi (609.31 keV) with the intensity of 46.1%. The gamma line of ^{228}Ac (911.07 keV) with the intensity of 29% was used to determine the activity of ^{232}Th. In addition, the activity concentrations of ^{40}K and ^{137}Cs were measured directly using their own gamma-rays of 1460.75 (10.7%) and 661.66 keV (85.12%), respectively [14,15]. The minimum detectable activity (MDA) of the detector set for all radionuclides was calculated using Equation 4 [14]. Values lower than MDA are unacceptable and for these MDA radionuclides listed in the tables with the sign <. and for values below the MDA, the MDA values are listed in Tables and marked with <

$$MDA = \frac{LLD\times 100}{\varepsilon(\%)\times Br\times T} \quad (4)$$

Where LLD is expressed as Equation 5:

$$LLD = 2.70 + 4.65 \times \sqrt{N} \quad (5)$$

Where LLD is low-level detection and N represents the count under the full-energy peak. The definitions of ε, Br, and t are similar to Equation 3 .

Calculating DI_r, Ra_{eq}, AED, the threshold consumption rate of wheat and bean, and ELCR

Taking into account the annual consumption of wheat and bean per capita in Iran (i.e.; 167.63 and 9.5 kg, respectively), the daily intake of radionuclides (DI_r) is calculated by Equation 6 [16]:

$$DI_r = \frac{A_r\times C_{ds}}{365} \quad (6)$$

Where DI_r, A_r, and C_{ds} are daily intake of radionuclides (Bq/d), radionuclide concentration (Bq/kg), and annual food intake (kg/y), respectively. The total radioactivity of natural radionuclides, including ^{226}Ra, ^{232}Th, and ^{40}K in the samples can be defined as the equivalent of ^{226}Ra radioactivity, which is calculated using equation 7 [8].

$$Ra_{(eq)} = A_{Ra} + 1.43\, A_{Th} + 0.077\, A_K \quad (7)$$

Radium equivalent (Ra_{eq}) was defined based on the assumption that 10 Bq/kg ^{226}Ra, 7 Bq/kg ^{232}Th, and 130 Bq/kg ^{40}K induce the same dose. The annual effective dose (AED) for an adult as a result of consuming foods containing natural radionuclides is calculated using equation 8 [8]:

$$AED = CR \times \sum_i^3 A_i \times CF_i \quad (8)$$

Where CR, A_i, and CF_i indicate annual consumption rate (kg/y), specific radionuclide activity (Bq/kg), and dose conversion factor (Sv/Bq), respectively (see Table 1). Equation 9 was used to determine the maximum annual allowable wheat and bean consumption, taking into account that the average AED due to radionuclides intake was estimated as 0.32 mSv/y [8].

$$CR = \frac{AEDE}{\sum_i^3 A_i \times CF_i} \quad (9)$$

Where A_i and CF_i represent the same factors as equation 8.

The cancer risk associated with eating food contaminated with radioactive nuclei was calculated using equation 10 [17]:

$$ELCR = ML \times AED \times RF \quad (10)$$

Where ELCR is Excess lifetime cancer risk, ML denotes average lifetime (71.15 years), AED is annual effective dose is annual radionuclide intake, and RF indicates a nominal risk coefficient for the public, which is 4.1×10^{-2} Sv^{-1} [18,19].

Table 1. Dose conversion and risk factor [20]

Dose conversion and risk factor			
Nuclide	^{226}Ra	^{232}Th	^{40}K
CF_i (Sv/Bq)	2.80E-07	2.30E-07	6.20E-09

Results

In the present study, the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides were measured in 11 wheat and 11 bean samples collected from an area near the oil industry. The corresponding values with the means and MDAs are presented in tables 2 and 3. For all samples, the specific activity of ^{137}Cs, which is also shown in tables 2 and 3, was lower than the minimum detectable activity.

Radiological parameters, such as Ra_{eq}, AED due to wheat and bean consumption, annual consumption threshold, and ELCR are summarized in tables 4 and 5. The contribution of radionuclides to total AED and the calculated correlation coefficient between ELCR and total AED for all samples are shown in figures 2 and 3.

Table 2. Specific activities of ^{226}Ra, ^{232}Th, ^{40K}, and ^{137}Cs in wheat samples

ID Sample	Specific activity and MDA value (Bq/kg)							
	^{226}Ra	MDA	^{232}Th	MDA	^{40}K	MDA	^{137}Cs	MDA
w1	<MDA	1.88	3.47±1.07	2.02	145.75±8.18	9.72	<MDA	0.42
w2	1.5±0.78	1.23	3.33±1.18	1.74	148.68±8.04	9.2	<MDA	0.42
w3	4.43±1.14	1.59	3.97±1.31	2.31	150.72±9.1	11.53	<MDA	0.52
w4	<MDA	1.31	ND		103.19±7.16	10	<MDA	0.44
w5	3.34±1.08	1.47	4.09±1.18	2.13	164.68±8.9	10.17	<MDA	0.15
w6	1.87±0.86	1.26	ND		168.94±8.55	9.06	<MDA	0.4
w7	3.23±1.06	1.38	3.14±1.08	2.03	129.22±7.64	9.4	<MDA	0.19
w8	3.31±1	1.35	3.54±1.08	1.98	150.07±8.29	9.5	<MDA	0.43
w9	4.62±1.09	1.3	2.57±1.23	1.8	134.73±7.83	9.53	<MDA	0.16
w10	5.27±1.11	1.4	<MDA	1.98	121.93±7.33	9.25	<MDA	0.16
w11	2.95±0.83	1.25	<MDA	2.12	146.42±8.09	9.45	<MDA	0.38
mean	2.77±0.81		2.19±0.74		142.21±8.1			

Table 3. Specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in bean samples

ID Sample	Specific activity (Bq/kg)							
	^{226}Ra	MDA	^{232}Th	MDA	^{40}K	MDA	^{137}Cs	MDA
B1	<MDA	1.03	<MDA	1.93	519.67±19.22	10.35	<MDA	0.36
B2	1.84±1.1	1.35	3.62±1.29	2.06	482.87±18.58	10.1	<MDA	0.2
B3	2.08±1	1.3	3.54±1.07	1.76	541.65±20.2	9.95	<MDA	0.37
B4	5.06±1.14	1.5	3.47±1.2	2.22	515.38±19.39	9.89	<MDA	0.48
B 5	<MDA	1.57	3.23±1.22	1.8	520.64±19.84	11.89	<MDA	0.47
B 6	2.84±1.08	1.5	3.15±1.16	2.14	555.4±20.63	11.12	<MDA	0.41
B 7	4.16±0.93	1.31	2.86±1.1	2.05	568.56±20.67	10.23	<MDA	0.13
B 8	<MDA	1	3.49±1.21	1.99	505.7±18.75	10.05	<MDA	0.13
B 9	<MDA	1.18	2.86±1.01	1.96	568.98±20.71	10.35	<MDA	0.24
B 10	4.94±0.98	1.3	3.4±1.09	1.8	134.03±7.6	9.13	<MDA	0.14
B 11	2.54±0.91	1.3	ND*		129.22±7.87	9.4	<MDA	0.19
mean	2.13±0.65		2.69±0.94		458.37±17.59			

- ND: not detected

Table 4. Radiological parameters due to wheat ingestion

sample	Ra_{eq} (Bq/kg)	DI_r (Bq/d)			AED (mSv/y)				Cr (kg/y)	ELCR (10^{-3})
		^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K	Total		
W1	16.18	0	1.59	66.93	0	0.13	0.15	0.28	188.04	0.83
W2	17.71	0.69	1.53	68.27	0.07	0.13	0.15	0.35	151.82	1.03
W3	21.71	2.03	1.82	69.21	0.21	0.15	0.16	0.52	103.63	1.51
W4	7.95	0	0	47.38	0	0	0.11	0.11	500.17	0.31
W5	21.87	1.53	1.88	75.62	0.16	0.16	0.17	0.48	110.46	1.42
W6	14.88	0.86	0	77.57	0.09	0	0.18	0.27	203.69	0.77
W7	17.67	1.48	1.44	59.33	0.15	0.12	0.13	0.4	131.81	1.19
W8	19.93	1.51	1.63	68.91	0.16	0.14	0.16	0.46	119.79	1.31
W9	18.67	2.11	1.18	61.87	0.22	0.1	0.14	0.46	117.65	1.33
W10	14.66	2.41	0	55.99	0.25	0	0.13	0.38	143.4	1.09
W11	14.22	1.35	0	67.23	0.14	0	0.15	0.29	184.57	0.85
Min	7.95	0	0	47.38	0	0	0.11	0.11	103.63	0.31
Max	21.87	2.41	1.88	77.57	0.25	0.16	0.18	0.52	500.17	1.51
Mean	16.86	1.27	1.01	65.3	0.13	0.08	0.15	0.36	177.73	1.06

Table 5. Radiological parameters due to bean ingestion

Sample	Ra_{eq} (Bq/kg)	DI_r			AED (mSv/y)				Cr (kg/y)	ELCR (10^{-3})
		^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K	Total		
bs1	40.01	0	0	238.62	0	0	0.03	0.03	99.32	0.09
bs2	44.2	0.84	1.66	221.72	0	0.14	0.03	0.17	73.71	0.12
bs3	48.85	0.95	1.63	248.71	0.01	0.14	0.03	0.18	67.30	0.13
bs4	49.71	2.32	1.59	236.65	0.01	0.13	0.03	0.17	59.15	0.15
bs5	44.71	0	1.48	239.07	0	0.12	0.03	0.15	80.59	0.11
bs6	50.11	1.3	1.45	255.03	0.01	0.12	0.03	0.16	64.47	0.14
bs7	52.03	1.9	1.31	261.07	0.01	0.11	0.03	0.15	59.84	0.15
bs8	43.93	0	1.6	232.21	0	0.13	0.03	0.16	81.26	0.11
bs9	47.9	0	1.31	261.26	0	0.11	0.03	0.14	76.45	0.12
bs10	20.12	2.26	1.56	61.54	0.01	0.13	0.01	0.15	106.8	0.08
bs11	12.49	1.16	0	59.33	0.01	0	0.01	0.02	211.59	0.04
min	12.49	0	0	59.33	0	0	0.01	0.02	59.15	0.04
max	52.03	2.32	1.66	261.26	0.01	0.14	0.03	0.18	211.59	0.15
mean	41.28	0.98	1.24	210.47	0.01	0.10	0.03	0.14	89.13	0.11

Discussion

The specific activity of ^{226}Ra had the ranges of <1.31-5.27 and <1-5.06 (Bq/kg) for wheat and bean samples, respectively. In addition, the values for ^{232}Th were in the ranges of ND-4.09 and ND-3.62 (Bq/kg) with the mean of 2.19 and 2.69 (Bq/kg) for wheat and bean samples, respectively. The specific activity of ^{40}K was obtained as 103.19-68.94 and 129.22-568.98 (Bq/kg) with the mean values of 142.21 and 458.37 (Bq/kg) for wheat and bean samples, respectively. The latter result shows that bean is more enriched in terms of mineral potassium content.

Cesium level in all samples was less than the minimum detectable activity indicating that cesium in the soil is associated with larger molecules and cannot be easily absorbed by the root of a plant. As demonstrated in tables 4 and 5, the calculated DI_rs due to the consumption of wheat samples were reported as 0-2.41, 0−1.88, and 47.38−77.57 Bq for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Furthermore, the DI_rs resulting from bean consumption were found as 0-2.32, 0-1.66, and 59.33-261.26 Bq for ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The Ra_{eq} value for wheat and bean samples was in the range of 7.95-21.87 with a mean of 16.84 Bq/kg and the range of 12.49-52.03 with the mean of 41.28 (Bq/kg), respectively. The mentioned finding reveals that bean has more radioactivity content, compared to wheat. The consumption-induced AEDs for wheat and bean samples were within the range of 0.11-0.52 with a mean of 0.36 and the range of 0.02-0.18 with the mean value of 0.14 mSv/y, respectively.

These results show that for the wheat cultivated in this region, the average ingestion dose is higher than the world average of 0.32 mSv/y. However, it lays within the acceptable AED of 1 mSv/y for the public [7]. Acceptable consumption rate per capita for wheat was in the range of 103.63-500.17 with the mean value of 177.73 kg/y that for most of the samples is lower than the annual consumption rate in Iran (167.63 kg/y).

Natural radionuclides in the wheat sample of this investigation were more than other countries and other parts of Iran, which could be attributed to environmental pollution due to the waste of oil burning that disperses to farms in the form of fly ash. This indicates that the wheat cultivated in this area is mostly contaminated by radioactive nuclei. Therefore, constant monitoring is required when this wheat, which is associated with health risks to the population is consumed. On the other hand, bean consumption does not threaten public health because the mean AED value of bean is definitely lower than the world average (0.05<<0.32) [8].

The contribution of each radioactive element to the AED was calculated as shown in Figure 2 for both wheat and bean. In wheat samples, ^{40}K had the main share of 41% in the dose, while ^{232}Th series had the largest share of 77% in bean seeds. The mean values for wheat and bean were 1.06×10^{-3} and 0.11×10^{-3}, respectively. Consequently, the ELCR of wheat was higher than the world average of 0.29×10^{-3} and was equal to the maximum acceptable limit of 10^{-3}, respectively [8].

Figure 3 shows a good correlation between ELCR and total AED in all samples. Correlation coefficient values (r^2) were obtained as 0.99 and 0.6 for wheat and bean samples, respectively. Table 6 lists the reported values of specific activity for natural radionuclides in wheat and bean in some countries. Comparison of the specific activity of radionuclides in this study with the data from other reports reveals a higher amount of ^{226}Ra, ^{232}Th, and Ra_{eq} in wheat and bean samples. However, in the case of potassium was in the same range except for Tanzania.

The analyzed data showed that the average radiological parameters of wheat were higher than the world average and acceptable limits, which indicates the impact of environmental pollution on the wheat produced in this region. Regarding bean, these parameters were found to be within the acceptable interval. As a result, this group of plants does not pose a radiological hazard to humans.

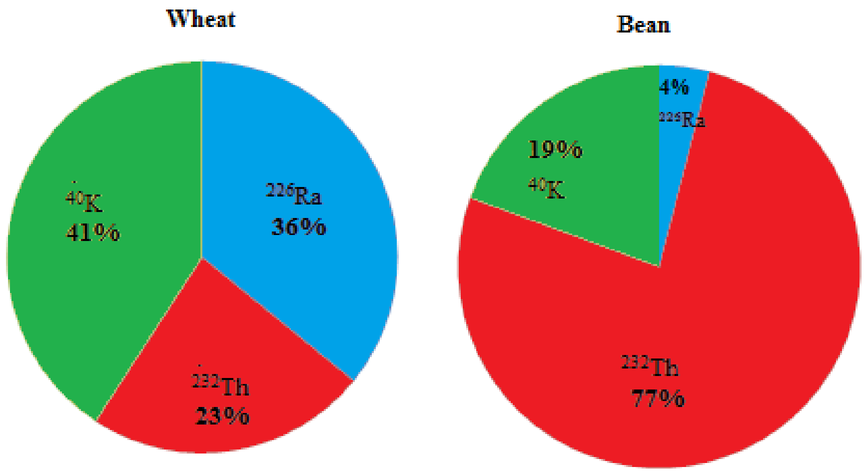

Figure 2. Share of radionuclides in the average annual effective dose in wheat and bean samples

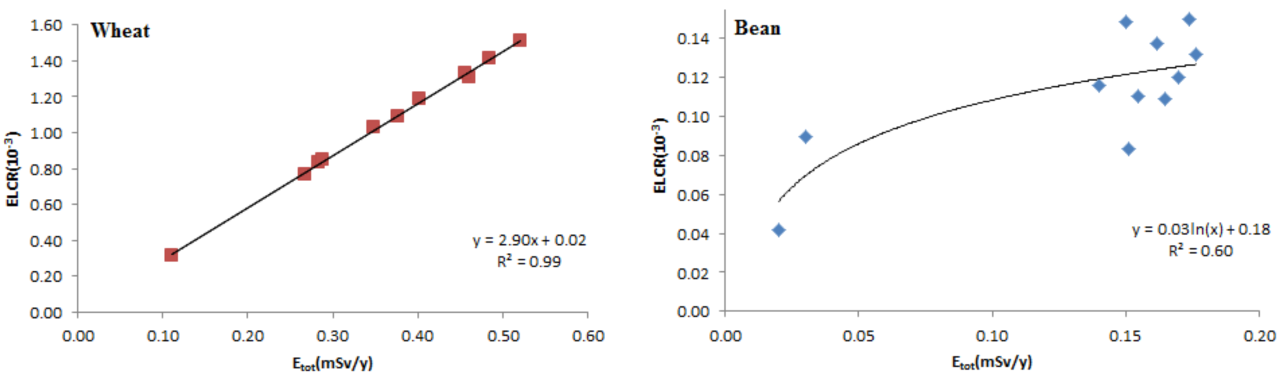

Figure 3. Correlation between AED and ELCR in wheat (left) and bean (right) samples

Table 6. Comparison of the specific radionuclide activity, average annual effective dose, and consumption rate of wheat and bean samples with other studies in the world

Country	Specific Activity (Bq/kg)			Ra_{eq} (Bq/kg)	Average Annual Effective Dose (mSv/y)				CR (kg/y)	Reference
	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K	Total		
Wheat										
France	0.57	<0.03	146.3	11.8						[23]
Iraq (Karbala)	<1.46	<1.38	180.54	13.9	*	*	*	1	*	[13]
Iran (Ilam)	1.67	0.5	91.73	8.15	*	*	*	5.28		[21]
Iran (Shazand)	2.77	2.19	142.21	16.86	0.13	0.25	0.15	0.53	177.73	Present study
Bean										
Tanzania	0.19	0.17	38.22	3.38	8.55	12.5	45.1	0.06	200	[22]
Brazil	1.43	0	434	34.85	*	*	*	*	*	[23]
Iran (Arak)					0.03	2.05	0.55	0.55	---	[24]
Iran (Shazand)	2.13	2.69	458.37	41.27	0.01	0.01	0.03	0.05	89.13	Present study
world average								290		[8]
Cereals (wheat, corn, and bean)										
Sudan (Khartoum)	0.20	0.63	230.85	18.88	*	*	*	0.57	392.2	[25]
Sudan (South Kordofan)	4.31	3.36	138.05	19.74	*	*	*	1.27	392.2	[25]
Sudan (Tokar)	0.88	1.62	323.96	28.14	*	*	*	0.33	392.2	[25]
World mean					*	*	200	290		[8]

Conclusion

Natural radioactivity values (^{226}Ra, ^{232}Th, and ^{40}K) in wheat and bean were measured by gamma-ray spectrometry method using HPGe detector. Results of the current study indicated that the ^{226}Ra and ^{232}Th contents of wheat and bean were higher than the other reports except for Sudan. The ELCR for wheat consumption was higher than the world average and was equal to the maximum acceptable value. On the other hand, the corresponding value for bean was observed to be lower than the world average.

Therefore, it seems that the emission of pollutants from the combustion chimneys of petroleum fuels led to the increased uptake of radionuclides by wheat and bean. Consequently, it is necessary to investigate these changes in the future studies. Moreover, our results demonstrated that radiological parameters did not exceed the reference values in this stage and do not pose a radiological threat to human health.

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