Radiological Assessment of the Artificial and Natural Radionuclide Concentrations of Wheat and Barley Samples in Karbala, Iraq

Reza Pourimani [1*], Seyed Mohsen Mortazavi Shahroodi[1]

1. Department of Physics, Faculty of Science, Arak University, Arak 38156, Iran

ARTICLE INFO	ABSTRACT
	Introduction: Artificial and natural radionuclides exist in the environmental materials, such as water, soil, rocks, and plant as well as in animals and human body tissues. Therefore, human being and environment are at constant radiation exposure. Regarding this, the present study aimed to determine the specific activities of radionuclides and perform the risk assessment wheat and barley samples in Karbala, Iraq. *Materials and Methods:* In this study, natural and artificial radioactivity concentrations were determined in wheat and barley samples, as well as their surrounding soil, using gamma ray spectrometry method by means of a high-purity germanium (HPGe) detector with 88% relative efficiency. According to the measured specific activities of radionuclides in food samples and according to annual consumption of wheat and barley by adult person excess lifetime cancer risk due to ingestion of radionuclides was calculated. *Results:* Specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the soil samples varied within the ranges of 29.37-38.86, 23.24-45.70, 291.15-549.78, and 1.25-10.82 Bq/kg, respectively. The excess lifetime cancer risk due to the ingestion of wheat and barley were calculated as 0.013×10^{-3} and 0.006×10^{-3}, respectively, which are lower than the maximum acceptable value (10^{-3}). *Conclusion:* As the findings of the present study indicated, the specific activities of natural radionuclides in the soil were close to that of the world average. The observation of artificial radiocaesium, which is released by nuclear accidents or weapon test, in all soil samples was indicative of the pollution of the studied regions by radioactive dust.

Introduction

Gamma radiation terrestrial is one of the main external sources of irradiation to human body, which is emitted from the natural radioisotopes. Natural environmental radioactivity mainly arises from primordial radionuclides, such as uranium (^{238}U), actinium (^{235}U), and thorium (^{232}Th) series, as well as the radioactive isotopes of potassium (^{40}K) in soil, building materials, water, rocks, and atmosphere [1]. The average amount of ^{238}U in the Earth's crust has been estimated as 2.7 mg/kg, while the concentration of this isotope sometimes has been observed up to 120 mg/kg in phosphate rocks [2].

Furthermore, ^{232}Th has the average amount of 9.6 mg/kg in the Earth's crust [3]. The amount of potassium as the eighth most abundant element in the Earth's crust is 2.8% , which depends on the type of minerals in the region [4]. Human body contains 90 μg of uranium on average, which is received through the air, water, and food. About 66%, 16%, 8%, and 10% of this element is found in the bones, liver, kidney, and other tissues, respectively [5].

Moreover, the release of artificial radioactive elements (e.g., ^{137}Cs and ^{90}Sr) to biosphere by nuclear weapon tests and nuclear power plants incidents increases the radioactivity level of the environment, and thereby enhances the radiation exposure in general populations. Therefore, humans and their environment are at constant exposure to these radiation types, 81% and 19% of which are attributed to the natural and artificial sources, respectively [6]. Food is a major source of various elements and radionuclides for the humans. Regarding this, the measurement of radioactivity in the environment and foodstuff is of paramount importance to assess the direct or indirect exposure of humans to different radiation levels.

Radiocesium pollution is considered as a major health hazard for humans since they can be transferred through food chains [7]. This study aimed to determine the specific activities of radionuclides in

Corresponding Author: Department of Physics, Faculty of Science, Arak University, Arak 38156, IranTel: 09181612224; 086- 34173402-4; Fax: 086-34173406;
Email : r-pourimani@ araku.ac.ir,

wheat and barley samples and their surrounding soils in the conflict-affected region of Najaf governorate in Iraq through gamma-ray spectrometry method using high-purity germanium (HPGe) detector. In addition, we determined the radiological hazards associated with the ingestion of wheat and barley samples and their maximum consumption threshold.

Materials and Methods

Sampling and sample preparation

This study was conducted on 22 samples of wheat (n=6) and barley (n=6) and their surrounding soil (n=10) collected from Najaf governorate. Sample collection was accomplished by random integration and experimental sampling as a combined operation. To this end, the study area was divided to many square sections; subsequently, some of the squares were randomly selected. Five samples were taken from each selected square, and then mixed. The soil samples were collected from three wheat and barley fields at 5-30 cm depth. Afterwards, the soil samples were coded as wheat or barley and according to sampling location, using the template method. The geographical coordinate values and samples' code location area are displayed in Table 1 and Figure 1, respectively.

All wheat, barley, and soil samples were dried at 200 °C for 12 h to ensure the complete removal of the moisture. In order to obtain homogeneous samples, the dried samples were pulverized by a grinder into fine powder and passed through 10-mesh and 50-mesh screens [8]. All samples were prepared in Negin containers for gamma ray spectrometry analysis. The collected samples required particular care because radon is a short-lived gaseous nuclide, which tends to escape from the samples. In this study, Negin containers with the volume of 300 cc were used to seal 200 g soil and 200 g wheat and barley samples. Gamma rays were registered after passing a minimum of 50 days from the preparation of the sealed samples because this time is necessary to reach the radioactive equilibrium [8].

Table 1. Geographical coordinates and codes of the samples

Sample code	Latitude	Longitude
S1	32.45342	44.21219
S2	32.45325	44.21245
S3	32.45360	44.21231
S4	32.45362	44.21222
S5	32.45305	44.21210
S6	32.45328	44.21210
S7	32.45311	44.21243
S8	32.45312	44.21284
S9	32.45308	44.21263
S10	32.45296	44.21287

Figure1. Geographiclocation of sampling area

Experimental Setup

Gamma ray spectrometry was performed using a p-type high purity germanium coaxial detector (HPGe) with the relative efficiency of 88%and multichannel analyzer of 8192 channel performance (manufactured by E & G Ortec Company Tennessee 37831 USA). The energy resolution of the detector was 1.85 keV for gamma ray energy line at 1332.520 keV of [60]Co. The operating voltage was set at 3,000 V, and the detector was shielded in lead, cadmium, and copper with the thickness of 10 cm, 2 mm, and 2.5 mm, respectively. This shield facilitated the reduction of the background radiation.

The soft components of cosmic rays were reduced to a very low level through shielding with a 100 mm thick lead. The X-ray (73.9 keV), emitted from the lead by its interaction with external radiation was suppressed by copper layer. Subsequently, the cadmium layer absorbed thermal neutrons produced by cosmic ray [9]. To minimize the effect of the radiation scattered from the shield, the detectors were located at the center of the chamber.

The samples were placed over the detectors for 86,400 sec using a uniform geometric setting. The background was also measured for the empty container with the same counting time and subtracted from the samples spectra. The standard sources for energy and efficiency calibration used in this study were RGU-1, RGTh-1, and RGK-1 for Ra, Th, and K, respectively, following the International Atomic Energy Agency reference material. The specific activities of Ra, Th, and K have been determined for these radiation sources by the International Atomic Energy Agency (IAEA).

The container of the standard samples was prepared similar to that of the soil samples with the same weight, and then sealed. After a minimum of 50 days, the gamma ray spectra of the standard samples were registered. Based on the registered gamma ray spectra, the absolute efficiency (ε_i) of detector configuration was calculated using Equation 1 [8].

$$\varepsilon = \frac{N_i}{Act \times P_n(E_i) \times T \times 100} \tag{1}$$

where N_i is the net count under the full energy peak corresponding to E_i, *Act* is radionuclide activity at measured date, $P_n(E_i)$ indicates the probability of E_i photon emission per decay, and *t* is counting time.

The ^{226}Ra activity of the samples was determined through the intensity of 351.9 and 609.3 keV gamma lines of ^{214}Pb and ^{214}Bi, respectively. ^{232}Th activity rate was obtained using the gamma lines of 911.21 and 968.97 keV of ^{228}Ac emission probability 26.6% and 17.4%, respectively. ^{40}K and ^{137}Cs activities were obtained using 1460.70 and 661.66 keV gamma ray lines, respectively. All of 24 registered gamma ray spectra were analyzed, and the specific activity of radionuclides in the samples was calculated using Gamma Vision 32(EG & G Ortec software, Tennessee 37831 USA). In order to calculate the activity concentration, we used Equation 2 [10].

$$Act(Bq/kg) = \frac{Net\ Area}{\varepsilon \times BR(\%) \times t \times m} \times 100 \tag{2}$$

where *Net Area* is the net count under photopeak, *Act* (Bq/kg) is the specific activity, ε_i is the energy efficiency for gamma ray by detector, *B.R.* is the branching ratio of gamma intensity (%), *t* (s) is spectral timing, and m (kg) is the mass of samples.

Transfer Factor

The soil-to-plant transfer factor (TF) indicates the transfer of radionuclides from soil to plant taken through the plant roots. Using the measured activity concentrations of the radionuclide in the plant and the surrounding soil, the TF values were calculated according to Equation 3 [11].

$$TF = \frac{activity\ of\ rdionuclides\ in\ plant\left(\frac{Bq}{kg}\right)dry\ weight}{activity\ of\ rdionuclides\ in\ soil\left(\frac{Bq}{kg}\right)dry\ wright} \tag{3}$$

Average annual committed effective dose measurement

The average annual committed effective dose (AACED) due to the ingestion of naturally occurring radioactive materials (NORMs) in medicinal plants was estimated using Equation 4 [12].

$$E_{ave} = Cr.DCF_i.A_i \tag{4}$$

where E_{ave} is the average annual committed effective dose, *Cr* is the consumption rate of radionuclides, and DCF_i is the dose conversion factor for each radionuclide (i.e., 2.8×10^{-7}, 2.3×10^{-7}, 6.2×10^{-9}, and 1.3×10^{-8} Sv/Bq for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs respectively), and A_i is the activity concentration of each radionuclide [13].

According to Equation 4, committed effective dose to an individual is directly proportional to the consumption rate of the ingredients of medicinal plants as a medicine. It should be mentioned that the AACED was calculated for 1 kg/year consumption rate of wheat and barley.

Using the same equation, the threshold consumption rate (Cr) for a medicinal plant can also be obtained from Equation 5:

$$Cr = \frac{E_{ave}}{\sum_{i=1}^{4}(DCF_i . A_i)} \tag{5}$$

where E_{ave} (0.32 mSv/y) is the threshold average annual committed effective dose due to the ingestion of NORMs in the medicinal plants, A_i is the activity concentration of radionuclide i, and DCF_i is the dose conversion factor for radionuclides [13].

The excess lifetime cancer risk (ELCR) for the consumed foodstuff contaminated with radionuclides was calculated as follows:

$$ELCR = A_{ing} \times DL \times RF \tag{6}$$

where A_{ing} denotes the annual consumption rate of radionuclides (Bq/kg), *DL* is the mean lifetime (year), and *RF* represents the risk factors of radionuclide ingestion (1/Bq).

The RF values of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined to be 4.80×10^{-8}, 2.30×10^{-7}, 5.90×10^{-9}, and 1.3×10^{-8}, respectively [14-15].

Results

In this study, the specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined in 10 samples of soil, 6 samples of wheat, and 6 samples of barley. The results of these measurements are given in tables 2-3.

Table 2. Specific activities (Bq/kg) of radionuclides in soil samples

Sample code	226Ra	232Th	40K	137Cs
S1	29.37±1.06	23.24±1.00	185.54±4.23	3.17±0.21
S2	30.79±0.78	37.35±1.04	452.07±4.41	4.13±0.24
S3	29.40±0.78	33.64±1.11	291.15±4.79	1.25±0.22
S4	31.09±0.76	39.76±1.12	447.25±4.44	7.27±0.26
S5	29.21±0.76	36.20±1.06	416.1±4.42	10.82±0.27
S6	35.54±1.01	42.85±1.11	502.44±4.71	6.22±0.25
S7	36.55±0.82	41.59±1.54	518.66±5.01	5.55±0.25
S8	36.57±1.01	42.96±1.39	508.07±4.43	7.87±0.29
S9	36.20±1.01	45.70±1.59	549.78±4.51	5.24±0.29
S10	38.76±1.04	39.55±1.13	460.67±4.52	3.64±0.32
Ave (soil)	33.35±0.90	38.28±1.21	430.27±4.55	5.52±0.26

Table 3. Specific activities of radionuclide in wheat and barley samples

Sample code	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	^{137}Cs (Bq/kg)
W1	<1.64	<1.65	165.97±8.94	<0.44
W2	<1.61	<1.58	205.57±9.71	<0.37
W3	<1.63	<1.44	177.09±8.83	<0.34
W4	<1.12	<1.08	187.76±7.76	<0.29
W5	<1.62	<1.46	180.26±8.97	<0.41
W6	<1.17	<1.04	166.59±6.93	<0.29
Ave (wheat)	<1.46	<1.38	180.54±8.52	<0.36
B1	<1.59	<1.49	232.01±10.58	<0.39
B2	<2.01	<1.88	254.04±12.06	<0.46
B3	<1.65	<1.52	241.77±10.75	<0.40
B4	1.69±1.15	3.67±1.04	262.65±11.42	<0.39
B5	3.39±1.05	1.89±1.01	233.29±10.56	<0.38
B6	<1.19	<1.17	229.57±9.18	0.34±0.19
Ave (barley)	1.92±0.36	1.94±0.34	242.22±10.76	0.39±0.03

Table 4. Transfer factor values of radionuclides from soil to wheat and barley

Soil Code	Sample code	TF (^{226}Ra)	TF (^{232}Th)	TF (^{40}K)	TF (^{137}Cs)
S1	W1	0	0	0.89	0
S2	W2	0	0	0.45	0
S3	W3	0	0	0.61	0
S4	W4	0	0	0.42	0
S5	W5	0	0	0.432	0
S1	W6	0	0	0.90	0
-	Ave (wheat)	0	0	0.55	0
S6	B1	0	0	0.46	0
S7	B2	0	0	0.49	0
S8	B3	0	0	0.47	0
S9	B4	0.04	0.08	0.48	0
S6	B5	0.09	0.04	0.46	0
S10	B6	0	0	0.50	0.09
-	Ave (barley)	0.02	0.02	0.48	0.01

Table 5. Calculated values of average annual committed effective dose (mSv/y), annual acceptable consumption rate (kg/y,) and excess lifetime cancer risk for samples

Sample code	AACED (mSv/y)	Cr (kg/y)	ELCR (in 10^{-3})
W1	0.001	291.54	0.012
W2	0.001	235.38	0.015
W3	0.001	273.23	0.013
W4	0.001	257.71	0.013
W5	0.001	268.43	0.013
W6	0.001	290.46	0.012
Ave (wheat)	0.001	268.01	0.013
B1	0.001	208.56	0.005
B2	0.001	190.47	0.005
B3	0.001	200.14	0.005
B4	0.003	101.84	0.009
B5	0.003	105.99	0.007
B6	0.001	210.12	0.005
Ave (barley)	0.002	153.63	0.006

AACED; average annual committed effective dose, Cr: consumption rate, ELCR: excess lifetime cancer risk

Table 4 presents the results of the calculated radionuclide transfer factors from soil to wheat and barley. The results of the estimated AACED for 1 kg annual consumption rate of wheat and barley, the annual acceptable criteria for the consumption of wheat and barley by an Iraqi adult person, and ELCR calculation based on annual consumption report per capita in Iraq (171.2 kg wheat and 50 kg barley) are illustrated in Table 5.

Discussion

The specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the soil samples varied from 29.21±0.76 to 38.76±1.04, 23.24±1.00 to 45.70±1.59, 185.54±4.23 to 518.66±5.01, and 3.17±0.21 to 10.82±0.27 Bq/kg, respectively. Furthermore, the average of specific activities of the mentioned radionuclides in the soil samples were obtained as 33.35, 38.28, 430.27, and 5.52 Bq/kg, respectively. These values were close to the mean global concentration for natural radionuclides (i.e., 35, 40, and 400 Bq/kg, respectively) and for ^{137}Cs as reported in a study performed in Markazi province in Iran [16-17].

The specific activities of ^{226}Ra and ^{232}Th in the wheat and barley samples, except for B4 and B5 samples, were less than the minimum detectable activity. ^{137}Cs was only observed in B6 sample in low quantity (0.34±0.19 Bq/kg). The specific activity of ^{40}K in the wheat and barley samples varied within the ranges of 165.97-205.57 and 229.57-254.04 Bq/kg, respectively. The maximum TF values of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were 0.09, 0.08, 0.90, and 0.09, respectively. The AACED for 1 kg of wheat and barley consumption varied within 0.001-0.003 μSv/y.

According to the Global Agricultural Information Network prepared by USDA foreign agricultural service in 2010, the per capita consumption of wheat in Iraq was 171.2 kg [18]. Therefore, the average AACED was 0.174μSv/y, which was lower than the maximum global acceptable value (0.32mSv/y). Considering the maximum acceptable AACED for wheat and barley, the threshold consumption rates were 268.01 and 153.63 kg/y, respectively (Table 5). The ELCR values considering the per capita consumption of 171.2 kg/y wheat and 50 kg/y barley varied within the range of $0.005×10^{-3}-0.015×10^{-3}$ (Table 5), which for all samples were significantly lower than the maximum acceptable value (10^{-3}) and average international value ($0.29×10^{-3}$) [16].

Conclusion

In this study, the specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined in 10 soil samples and 12 samples of wheat and barley using gamma ray spectrometry by means of an HPGe detector system. According to the results, the AACED for the wheat and barley was within the acceptable range. The maximum acceptable values of the annual consumption of wheat and barley were 268.01 and 153.63 kg/y. The annual consumption rate of wheat and barley in Iraq is less than these values; consequently, there is no risk threatening the health of the people living in the investigated region. Future studies are recommended to examine the trace elements in the soil and wheat samples for the investigation of toxic and heavy metals and find the answer to this question: How years of conflicts have affected the variation of trace elements?

Acknowledgment

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