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Measurement of Radioactivity Levels and Health Risks in the Surrounding Soil of Shazand Refinery Complex in Arak, Iran, Using Gamma-Ray Spectrometry Method

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ABSTRACT

Introduction: The purpose of this study was to measure the radioactivity in the agricultural soil of south-east of Shazand Refinery Complex to determine both reliable baseline data on the radiation level and the radiation dose exposure to the farmers and inhabitants of the studied area.

Material and Methods: This study was conducted on 21 soil samples collected from two different lands. Sampling spots in each land were selected for the assessment of specific activities of radionuclides of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs by using high purity germanium detector setup. Standards of International Atomic Energy Agency references material gamma ray uranium, reference gamma-ray thorium, and reference gamma-ray potassium were used for quality control and determining efficiency calibration. All samples were examined for radium equivalent, absorbed gamma dose rate, internal hazard index, external radiation hazard, annual gonadal dose equivalent, indoor and outdoor annual effective dose equivalent, and excess lifetime cancer risk.

Results: The specific activities of radionuclides ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs varied from13.12 to 33.03, 11.3 to 35.86, 257.82 to 605.5, and 1.28 to 13.36 Bq/kg, respectively. Moreover, the results of this study were compared with those reported from other countries and worldwide average.

Conclusion: Although all samples were polluted by the ^{137}Cs fission product, the measured values were within the global reported safety limits. Therefore, there is no risk for farmers and inhabitants in this region.

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Introduction

Over the course of their lives, people are exposed to ionizing radiation whether desired or not. The majority of these radiations come from the decay of radionuclides in the Earth's crust, such as the uranium, thorium series, and the ^{40}K isotope 1]. Dangerous radon gas (^{222}Rn and ^{220}Rn) arising from uranium and thorium chains can enter the human body by respiration, and consequently, induces internal exposure. In addition, some ionizing radiation has cosmic origins. Today, artificial radionuclides make up a significant proportion of the exposure of a living organism. The specific activities of radionuclides in the earth's crust estimated for uranium and thorium as 2.7 and 9.6 mg/kg, respectively [2]. Potassium as the sixth most abundant element is evaluated as 2.8% [2].

A significant amount of artificial radionuclides may also be present in the environment as a result of testing nuclear weapons in the atmosphere; accidents that occur at nuclear power plants, such as Chernobyl and Fukushima power plants; and nuclear waste from nuclear installations [3]. Additionally, the spallation of refinery waste and mazut as a product of the petroleum and coal production processes releases radionuclides into the biosphere [4]. These pollutants enter the atmosphere through the chimney of the power plant and then disperse into the atmosphere through the wind. Finally, after cooling, the radionuclides spread fallout over the surrounding soil, causing soil and vegetation pollution. These contaminants enter the human food chain via plants and animals.

Therefore, the continuous examination of the surrounding regions of these industries facilitates the adoption of measures to limit the pollutions in case of exceeding the standard limits. Knowing this, it is important to evaluate the effects of radiation exposure. Accordingly, the aim of the present study is to measure the natural radioactivity levels and to estimate the hazard indices, such as radium equivalent activities, external, and internal hazard indices in the soil surrounding of south-east of Shazand Refinery Complex by using the gamma-spectrometry method. This study may provide a reference regarding both public health and

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maintaining safe radioactivity levels in the desired soil in Arak, Iran.

Materials and Methods

Sampling and Sample preparations

In this work, two different areas of agricultural lands in south-eastern Iran, where the Shazand Refinery Complex is located, are Baghbaraftab (BB) and Ghadamgah (GH). These sites were selected for collecting the data samples. Sampling was done randomly and experimentally as a combined method. The geographical locality of the sampling map is shown in figure 1, while Table 1 gives detailed information about the samples.

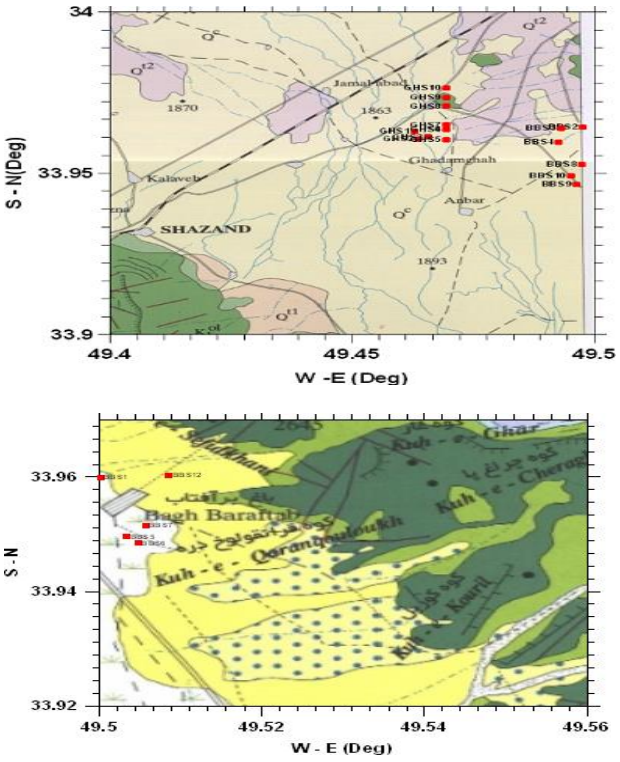

Figure 1. The geographical locality of the sampling maps above: 49.4-49.5 W-E (Deg) and below: 49.5-49.56 W-E (Deg)

For sampling, after dividing the geographical area into multiple parts, we then selected segments and designed a 1 × 1 m^2 square. In each square, we collected soil samples from five points and prepared the samples as a mixture of vertices and center points. The geographical coordinates were determined by using a

GPS apparatus with an accuracy of 1 m. Sampling was carried out from 1 July, 2017 until 20 August, 2017. Corresponding samples were cleaned from waste material, such as wood, and coded using BB and GH. Finally, each sample was packed in a plastic bag and transferred to the laboratory. Each soil sample was kept at room temperature to maintain a constant weight. Next, it was powdered through the use of an electric mill. Finally, the samples were sieved through a 50-mesh screen until a fine powder was achieved. The samples were then dried in an oven with 100 °C for 6 hours in the laboratory [5]. The samples were then packed in 300 cc Negin containers. The containers were sealed using silicon glue and kept at least 4 weeks to attain chain equilibrium, which was the equilibrium between parents and daughters for the ^{226}Ra and ^{222}Rn decay series, where after this time the decay rate of the daughter became equal to the parent in the chains [6].

Gamma-ray spectrometry

The specific activities of radioactive elements were determined using a gamma-ray spectrometry method that employed a coaxial, P-type, high purity germanium detector, a GCD30195BSI model manufactured by Baltic Scientific Instruments Ltd.(005-Lotvia) with 30% relative efficiency and its electronics units. The energy resolution (full width at half maximum) of this detector was 1.95 keV for the gamma energy line at 1332.520 keV ^{60}Co. The operating voltage was 3000 V. The detector and preamplifier were shielded in a chamber comprising three layers, including a 10 cm thick layer of lead, a 1.5 mm thick layer of cadmium, and a 2 mm layer of copper. This shield was used to reduce background radiation [7]. Standards International Atomic Energy Agency (IAEA) references material RGU, RGTh, and RGK were used for quality control and determining the efficiency calibration. These references covered an energy range from 60 to 2000 keV. A spectrum of each standard was registered using the Lsrmbsi software (00-5-Latvia) for 86400s. The Gamma vision 32 (manufacture by E&G Ortec Company, Tennessee 37831, USA) software was used for spectral analysis and to calculate the most suitable curve for each of the points. The purpose of drawing the calibration curve of efficiency was to calculate the efficiency for each of the energies whose values were not known. The detector efficiency was calculated based on equation 1 [6].

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

Where, *Ni* is the net count under the full-energy peak corresponding to the E_i energy, $P_n(E_i)$ is the photon emission probability for the particular energy E_i in percentage, Act is the activity of the radioactivity nuclei in the standard container in *Bq*, and t is the counting time.

Activity measurement

It is possible to determine radiological parameters, such as the dose rate and radium equivalent activity (Raeq), by calculating specific activities (SA) of radionuclides by means of equation 2[3]. 2 [3].

$$SA = \frac{\text{Net Area}}{\varepsilon \times BR \times t \times m} \quad (2)$$

Where Net Area is the net count under full-energy peak, *SA* (Bq/kg) is the specific activity, ε signifies the energy efficiency for gamma ray of detector-sample set up, *BR* refers to the branching ratio of gamma-ray intensity, t is the time of spectra in sec, and *m* denotes the mass of samples in kg. To research the accuracy of the net area under the full-energy peak, the manual

calculation was compared by analyzing the Gamma vision32 software. It was necessary to accurately determine the activity of the interested radionuclides to know their efficiency at the corresponding energy. The specific activity of ^{226}Ra was obtained using the gamma-ray lines 351.93 keV (^{214}Pb) and 609.31 keV (^{214}Bi). The gamma-ray lines 911.07, 969.11keV (^{228}Ac), 238.6 keV(^{212}Pb), and 583.2 keV(^{208}Tl) were used to determine the specific activity of ^{232}Th. The activity of ^{40}K and ^{137}Cs was measured directly using its own gamma ray 1460.75keV and 661.66 keV respectively [6].

Absorbed gamma dose rate

The absorbed dose rate in air (D) at one meter above the ground was used to describe the terrestrial radiation. The absorbed dose rate due to gamma radiation of naturally-occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated according to the guidelines to provide mathematical relation 3 [2].

$$D\ (nGy/h) = 0.462\ A_{Ra} + 0.604\ A_{Th} + 0.0417\ A_K \quad (3)$$

Where, 0.462, 0.621 and 0.0417 are conversion factors in nGy/h per 1Bq/kg of the specific activities of corresponding radionuclides respectively.

Radium equivalent

Almost 98.5% of the radiological hazards regarding gamma radiation are due to radium and its daughters. Therefore, to determine the environmental radiation level, the United Nations Scientific Committee on the Effects of Atomic Radiations (UNSCEAR) has defined a radiological indicator named radium equivalent, which can be calculated by equation 4 [2].

$$Ra_{(eq)} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (4)$$

Ra_{eq} was defined on the assumption that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the equal dose rate. The maximum value of this parameter for building material must be less than 370 Bq/kg [8].

Calculate the internal hazard index and external radiation hazard

The internal and external hazard indices used for estimation of risks due to respiration of Rn gas and gamma radiation in the environment. In the situation that these threats are negligible and do not require human intervention, they must be less than unity. The unity corresponds to the upper limit of Ra_{eq} activity (370 Bq/kg). The internal hazard index (H_{in}) and external radiation hazard (H_{ex}) were calculated using equation 5 and 6 [9].

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

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad (6)$$

Annual gonadal dose equivalent

Due to the importance of certain vital glands in the body, such as the breasts, thyroid gland, lymph nodes, lungs, and bone marrow, in terms of genetic and cancerous diseases, the UNSCAR determined the relation between the annual dose equivalent of the glands for the effects of nuclear radiation. Annual gonadal dose equivalent (AGDE) is defined by equation 7 [10].

$$AGDE\ (\mu Sv/y) = (3.09\ A_{Ra} + 4.18\ A_{Th} + 0.314\ A_K) \quad (7)$$

Where, A_{Ra}, A_{Th} and A_K in equations 3 to 7 are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively

Annual effective dose equivalent

People are usually exposed to 80% and 20% of radiation from the internal and external environment, respectively. An annual effective equivalent dose (AEDE) of indoor and outdoor absorption by humans can be calculated from the absorbed dose rate in the air with a conversion factor of 0.7 and the duration of human residence. Indoor and outdoor AEDE considering one year 8760 hours are computed by equation 8, 9 in mSv/y [11].

$$AEDE\ (indoor) = Dose\ rate\ (nGy/h) \times 0.8 \times 8760(h/y) \times 0.7(Sv/Gy) \times 10^{-6} \quad (8)$$

$$AEDE\ (outdoor) = Dose\ rate\ (nGy/h) \times 0.2 \times 8760(h/y) \times 0.7(Sv/Gy) \times 10^{-6} \quad (9)$$

In equations 7 and 8, the 0.8 and 0.2 coefficients are the indoor and outdoor occupancy factor, respectively.

Excess lifetime cancer risk

To calculate cancer risk in lifetime due to natural gamma radiation exposure level for resident population in this region by radionuclides content in soil, which is defined as follows:

$$ELCR = AEDE \times DL \times RF \quad (10)$$

Where, *AEDE*, *DL*, and *RF* are the annual effective dose equivalent (outdoor), duration of life (70 y), and risk factor (0.05 1/Sv), respectively [12]. The global mean excess lifetime cancer risk (ELCR) is 0.29×10^{-3} and the maximum allowed value is 10^{-3} [13].

Results

In this study, we determined the specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in 21 samples of soils. Tables 1shows the results of their analysis, ranges, and mean values. Moreover, Table 2 presents the values related to D, Ra_{eq}, Hin, Hex AEDE, AGDE, and ELCR.

Table 1. Geographical coordinates and specific activities of radionuclides ^{232}Th, ^{226}Ra, ^{40}K, and ^{137}Cs in the soil samples

Sample no	Sample ID	geographical coordinate		Ra-226 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	Cs-137 (Bq/kg)
		Longitude	Latitude				
1	BBS1	49.500038°	33.959895°	31.85±1.79	32.99±2.53	583.92±16.53	13.34±0.91
2	BBS2	49.497620°	33.964349°	29.75±1.92	30.65±2.13	560.51±20.62	10.19±0.73
3	BBS3	49.493115°	33.963944°	24.86±1.97	26.65±2.02	485.53±13.82	6.21±0.44
4	BBS4	49.492599°	33.959668°	25.95±2.15	26.5±2.09	443.98±12.85	8.83±0.47
5	BBS5	49.503340°	33.949546°	16.87±1.89	18.63±1.79	305.95±8.52	5.09±0.42
6	BBS6	49.504808°	33.948443°	28.77±1.69	30.23±2.93	524.67±12.27	8.67±0.47
7	BBS7	49.505626°	33.951517°	26.91±1.42	33.93±2.81	527.04±12.49	9.46±0.93
8	BBS8	49.497416°	33.952697°	21.68±1.81	23.28±1.84	391.73±10.17	4.09±0.6
9	BBS9	49.496305°	33.946644°	26.09±2.05	25.07±2.53	446.86±10.95	4.39±0.86
10	BBS10	49.495142°	33.949144°	24.58±2.12	26.65±1.32	509.06±16.76	6.41±0.45
11	BBS12	49.508588°	33.960345°	33.03±1.79	35.85±2.26	605.5±18.02	13.36±0.89
12	GHS1	49.462907°	33.963063°	16.75±1.39	18.86±2.57	353.89±14.77	3.12±0.4
13	GHS2	49.463713°	33.960534°	13.12±1.63	13.45±2.04	257.82±7.69	4.46±0.67
14	GHS3	49.465616°	33.961227°	20±1.75	17.85±2.43	285.2±8.22	1.28±05
15	GHS4	49.469338°	33.963595°	17.08±1.7	17.45±2.32	351.34±9.26	4.65±0.42
16	GHS5	49.473062°	33.960463°	21.45±1.98	21.02±2.67	346.85±11.31	6.46±0.73
17	GHS6	49.480261°	33.963630°	17.89±2.31	11.3±2.07	291.79±13.79	3.6±0.89
18	GHS7	49.476718°	33.964975°	30.56±2.14	26.37±2.46	435.64±16.11	4.31±0.41
19	GHS8	49.464443°	33.970749°	20.91±1.93	22.58±2.67	398.84±14.51	2.76±0.41
20	GHS9	49.473383°	33.973469°	19.69±1.97	21.08±2.22	364.71±14.74	5.17±0.43
21	GHS10	49.474818°	33.976510°	19.66±1.85	26.18±1.95	421.71±13.64	6.94±0.44
Min				33.03	35.85	605.5	13.36
Max				33.03	35.85	605.5	13.36
Mean ±SD				23.21±1.38	24.12±1.47	423.45±23.60	6.17±0.27

Table 2. Radiological parameters of the soil samples

Sample ID	Dose Rate (nGy/h)	$Ra_{(eq)}$	H_{in}	H_{ex}	AEDE indoor(mSv/y)	AEDE outdoor(mSv/y)	AGDE (μSv/y)	ELCR ($\times10^{-3}$)
BBS1	58.99	123.98	0.42	0.33	0.29	0.07	419.65	0.25
BBS2	55.63	116.75	0.4	0.32	0.27	0.07	396.07	0.24
BBS3	47.83	100.35	0.34	0.27	0.23	0.06	340.66	0.21
BBS4	46.51	98.03	0.33	0.26	0.23	0.06	330.37	0.2
BBS5	31.8	67.06	0.23	0.18	0.16	0.04	226.05	0.14
BBS6	53.43	112.39	0.38	0.3	0.26	0.07	379.99	0.23
BBS7	54.9	116.01	0.39	0.31	0.27	0.07	390.47	0.24
BBS8	40.42	85.14	0.29	0.23	0.2	0.05	287.33	0.17
BBS9	45.83	96.35	0.33	0.26	0.22	0.06	325.72	0.2
BBS10	48.68	101.89	0.34	0.28	0.24	0.06	347.19	0.21
BBS12	62.16	130.91	0.44	0.35	0.3	0.08	442.03	0.27
GHS1	33.89	70.96	0.24	0.19	0.17	0.04	241.7	0.15
GHS2	24.94	52.21	0.18	0.14	0.12	0.03	177.72	0.11
GHS3	31.91	67.48	0.24	0.18	0.16	0.04	225.96	0.14
GHS4	33.08	69.09	0.23	0.19	0.16	0.04	236.03	0.14
GHS5	37.07	78.21	0.27	0.21	0.18	0.05	263.05	0.16
GHS6	27.26	56.51	0.2	0.15	0.13	0.03	194.12	0.12
GHS7	48.21	101.82	0.36	0.27	0.24	0.06	341.47	0.21
GHS8	39.93	83.9	0.28	0.23	0.2	0.05	284.21	0.17
GHS9	37.04	77.92	0.26	0.21	0.18	0.05	263.48	0.16
GHS10	42.48	89.56	0.3	0.24	0.21	0.05	302.58	0.18
Min	24.94	52.21	0.18	0.14	0.12	0.03	177.72	0.11
Max	62.16	130.91	0.44	0.35	0.3	0.08	442.03	0.27
Mean	42.95	90.31	0.31	0.24	0.21	0.05	305.52	0.18

Discussion

The specific activities of radionuclides of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs range from 13.12±1.63 to 33.03±1.79 (mean 23.21±1.38), 11.30±2.07 to 35.85±2.26 (mean 24.12±1.47), 275.82±7.69 to 605.50±18.02 (mean 423.45±23.60), and 1.28±0.05 to 13.36±0.91 (average 6.15±0.27) in Bq/kg, respectively. As table 1 shows, the activities concentrations of ^{226} Ra and ^{232}Th for most of the samples are less than the world average value of 30 and 35 Bq/kg [7]. Moreover, the results show the specific activity mean of ^{40}K is the same as the world average (i.e., 412 Bq/kg) [2].

The impact of nuclear weapons and Chernobyl disaster can be seen in this region due to ^{137}Cs in all the soil samples. More than 0.80 % of the winds in this region blow west-east. As soon as they reach the valley of Toure in the mountain range of Sefidkhani, the winding path is almost closed and it creates a standing mass at the side of the mountain. When these winds meet the high mountains, they create a stream of stalactites, causing more suspended particles in these areas. Therefore, there is an increase in the specific

activity of radionuclide ^{137}Cs in the samples from these mountains, such as BBS2, BBS7, and BBS12. As figures 2 to 5 show, the distribution of ^{137}Cs, ^{232}Th, ^{226}Ra, and ^{40}K is increased near mountains, which is marked in red color near a geographical coordinate N34-E49.5 degree in the sampling location (Figure 1). The change of color from green, to yellow, and then red shows the increased specific activities of studied radionuclides (figures 2 to 5). Lines were drawn with the same value in Bq /kg, their values in equal lines are given in Fig. 2 to 5. These figures show the distribution of artificial radionuclide 137Cs, a fission product which is transferred under the influence of atmospheric flows, and natural radionuclides are similar. This confirms the effect of fly ash fallout from the chimney of refinery complex plant and standing air near the mountain range on the soil. Knowing this, the atmospheric processes influence the spread of fly ash in this region. In table 3, the specific activity of the radionuclide ^{137}Cs obtained from the present study is compared to studies carried out in other countries [14-20]. As can be seen, the results of the current study were in the same range as those of India, Iraq, Venezuela, and Turkey. Additionally, the obtained data in this study were much lower than those of Serbia and Poland. Furthermore, the findings of this study were in line with another research in Markazi province, Iran [20]. The mean values of D, Ra_{eq}, Hin, Hex, AEDE, AGDE, and ELCR, as well as indoor and outdoor AEDE resulting natural radioactivity content in the studied samples, were 42.95 nGy/h, 90.31 Bq/kg, 0.31, 0.24, 0.21, and 0.05 mSv/y, respectively. These values fell in the same range as the world average. The mean of AGDE was 305.52 µSv/y and ELCR as 0.18×10^{-3} which, were less than the world average and the mean of some countries (Table 4) [21-27]. Table 4 shows that the mean values of all natural radioactivity contents are lower than the UNSCEAR reported mean value and allowable limit [2].

Table 3. Comparison of specific activity of fallout ^{137}Cs (Bq/kg) obtained in this study with those reported worldwide

Region	^{137}Cs	Reference
India(Singhblum)	1.70-7.48	[14]
Serbia	48.30± 26.19	[15]
Turkey (Buyuk)	2.81-20.75	[16]
Iraq (Karbala)	1.25-10.82	[17]
Venezuela	3.5-15.0	[18]
Poland	0.00-101.61	[19]
Iran (Arak)	1.07-9.52	[20]
Iran	1.28-13.36	Present work

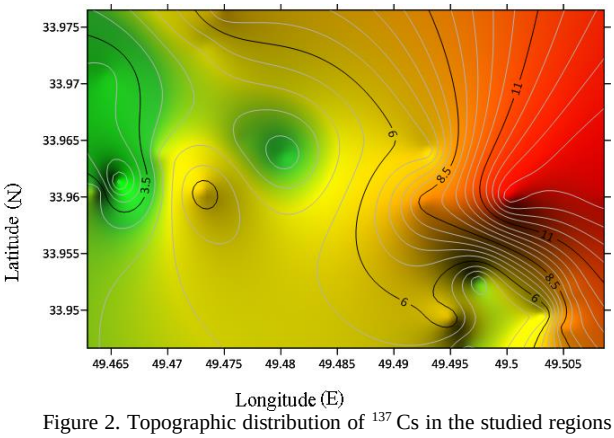

Figure 2. Topographic distribution of ^{137} Cs in the studied regions in Bq/kg.

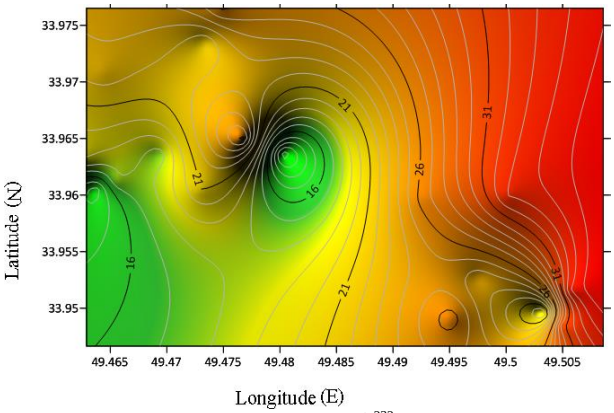

Figure 3. Topographic distribution of ^{232}Th in the studied regions in Bq/kg.

Table 4. Comparison of the mean specific activities of natural radionuclides, dose rate, and other radiological parameters in the soil samples of current study with those reported in countries.

country	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Dose Rate (nGy/h)	Ra_{eq}	AGDE (µSv/y)	ELCR	reference
Bangladesh (ship yards)	31.39	63.34	364.47	67.96	150.03	476.2	0.29	[21]
Bangladesh (Dhaka)	33	16	574	48.85	100.08	349.09	0.21	[22]
India(Kotagiri)	41	102	229	90.1	204.49	624.96	0.39	[23]
USA(California)	39.4	45.6	420	63.26	136.95	444.23	0.27	[24]
Greece	49	24	760	68.83	141.84	490.37	0.3	[25]
Taiwan	30	44	653	67.67	143.2	481.66	0.29	[26]
Iran(chabahar)	24	20	450	42.92	86.79	322.9	-	[27]
World Average	30	35	400	51.68	110.85	364.6	0.22	[7]
Iran(arak)	22.95	23.47	413.5	42.02	88.35	298.85	0.18	Present work

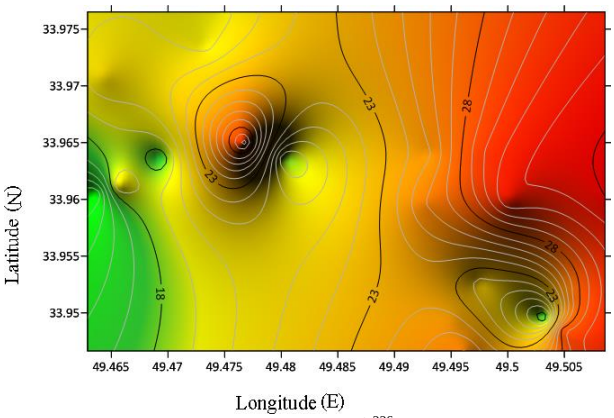

Figure 4. Topographic distribution of ^{226}Ra in the studied regions in Bq/kg.

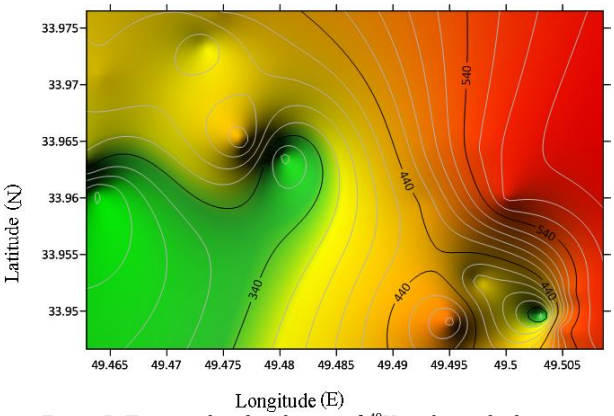

Figure 5. Topographic distribution of ^{40}K in the studied regions in Bq/kg

Conclusion

The gamma-spectroscopy method employed the high purity germanium detector to determine the natural and artificial radioactive values, including ^{226}Ra, ^{232}Th, ^{40}K, and ^{137} Cs. The obtained data showed that the mean values of radiological parameters were less than the world average and the maximum level of acceptable limits. This means that the soils of this two areas pose no radiological hazards for people and can be used safely for agriculture. The findings of this study could be used as a guideline for future investigations and natural radiation mapping.

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