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Estimation of Natural Radioactivity and Radiation Exposure in Environmental Soil Samples of Golestan, Iran

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ABSTRACT

Introduction: Considering the risk of radiation, the measurement of the natural radiation sources seems to be necessary. In this study, the concentrations of the natural radionuclides, namely ^{226}Ra, ^{232}Th, and ^{40}K, were measured in the soil samples taken from different locations of Golestan, Iran. The measurement results can also be used as a baseline to evaluate the impact of non-nuclear activities and the routine releases of nuclear installations.

Materials and Methods: A total of 42 soil samples were collected. The samples were sealed for at least three weeks to ensure the secular equilibrium between ^{226}Ra and ^{232}Th and their respective radioactive progenies. The activity concentrations of natural radionuclides in soil samples were measured using a shielded HPGe detector.

Results: The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were 23, 31, and 453 Bq.kg^{-1}, respectively. To assess the radiological hazards, the Ra equivalent activity as well as the external and internal hazard indices were estimated. Radium equivalent varied within the range of 58.4-142.6 Bq.kg^{-1} with a mean value of 102.4 Bq.kg^{-1}. The estimated mean values of H_{ex} and H_{in} (0.28 and 0.34, respectively) in the area under investigation were lower than unity as desirable. Therefore, it did not pose any health risks to the population of the area.

Conclusion: The results of this study were compared with those of other studies carried out in other countries. As the findings of the present study indicated, the health-related risks causing by the naturally accruing radionuclides was very low in the investigated area.

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Introduction

The determination of the background radiation level emanated from the natural radioactivity sources of terrestrial and cosmic rays is a necessary measure [1-5]. These data are usually important in establishing baseline data for the assessment of future radiation impact as well as radiation protection and exploration [6]. The natural background radiation exposure (2.4 mSv/person/year) accounts for almost 80% of the total radiation dose [7].

According to the international standards, the natural and artificial radioactivity concentrations should be periodically determined. The investigation of these data are especially important in the border regions. Regarding this, in the current study, the concentration of such radionuclides as ^{226}Ra, ^{232}Th, and ^{40}K were measured in Golestan, Iran.

The major contributors of the terrestrial radiation are the primordial radionuclides (i.e., ^{226}Ra, ^{232}Th, and ^{40}K) with half-lives comparable to the age of the earth [8,9]. Depending on the geographical and geological factors, the concentrations of radionuclides vary from place to place [10]. ^{226}Ra and ^{232}Th can be detected indirectly through their progenies. In secular equilibrium, the activities of the progenies and their parents are equal.

The measured concentrations were compared with nominal measurements in other countries. In addition, such radiological parameters as radium equivalent activities, hazard index, absorbed dose rate, and effective dose equivalent were calculated for the sampling locations.

Materials and Methods

Geography of the study area

Golestan is located in the north-east of Iran with an area about 22,000 km^2. This province is situated between the longitude of 53° 57′ and 56° 22′ east and latitude of 36° 30′ and 38° 8′ north. Golestan province is bounded by the Caspian sea and Mazandaran province in the west, North Khorasan province in the east, and Semnan province in the south.

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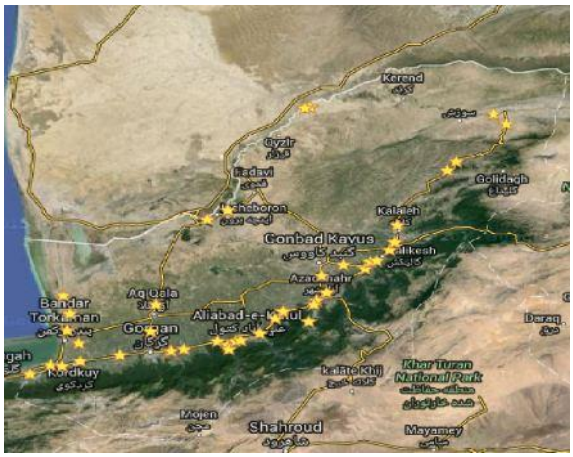

Figure 1. Sampling sites in Golestan province.

Sample collection and preparation

In total, 42 soil samples (0-5 cm in depth) were taken from the uncultivated locations of Golestan province. The majority of the samples were taken from the residential regions. The sampling sites and their geographical positions are given in Figure 1 and Table 1, respectively.

The soil samples were well mixed after removing the stones, vegetation, and roots. Subsequently, the samples were dried for 24 h at the ambient temperature, and then in an oven at 100 °C for 4 h to remove any available moisture.

The dried samples were pulverized, sieved with a 1 mm mesh, placed in 300 g polyethylene container, and sealed for at least three weeks to ensure the secular equilibrium between ^{226}Ra and ^{232}Th as well as their respective radioactive progenies. The samples were properly coded according to the sampling location; these codes are listed in Table 1.

Calculation of activity concentration

The samples and standards were measured using a shielded high-purity germanium (HPGe) detector (EGPC 5574 model, manufactured by Intertechnique, France) with an energy resolution of 2 keV at full width half of maximum (FWHM) from 1332 keV energy of ^{60}Co and relative efficiency of 20%. The counting time was between 20,000 and 60,000 sec, depending on the specific activity of the samples. The background was also measured for the same counting time and subtracted from the sample spectra. The calibration sources used in this study included RGU-1, RGTh-1, and RGK-1 for Ra, Th, and K, respectively, following the International Atomic Energy Agency's standard.

The absolute efficiency of the measurement system was calculated using the following equation:

$$\varepsilon = C_n / P_\gamma A \quad (1)$$

Where C_n is the net count rate under the corresponding photo peak; P_γ is the emission probability of gamma rays; A is the source disintegration rate.

Table 1. Coded samples based on the soil sampling location.

Sample code	Coordinates	Sample code	Coordinates	Sample code	Coordinates
S_1	N36°51.117′, E54°31.232′	S_{15}	N37°6.626′, E55°11.583′	S_{29}	N36°47.456′, E54°0.732′
S_2	N36°51.18′, E54°31.734′	S_{16}	N37°12.832′, E55°21.573′	S_{30}	N36°44.994′, E53°55.361′
S_3	N36°51.713′, E54°34.908′	S_{17}	N37°14.782′, E55°22.874′	S_{31}	N36°47.135′, E54°3.799′
S_4	N36°53.58′, E54°43.63′	S_{18}	N37°15.02′, E55°24.303′	S_{32}	N36°48.376′, E54°8.381′
S_5	N36°52.645′, E54°46.253′	S_{19}	N37°17.995′, E55°27.809′	S_{33}	N36°56.431′, E54°26.307′
S_6	N36°51.523′, E54°46.653′	S_{20}	N37°19.928′, E55°29.059′	S_{34}	N37°3.362′, E54°27.857′
S_7	N36°53.752′, E54° 48.089′	S_{21}	N37°24.548′, E55°29.633′	S_{35}	N37°26.064′, E54°40.873′
S_8	N36°53.999′, E54°49.434′	S_{22}	N37°14.004′, E55°15.793′	S_{36}	N37°28.361′, E54°46.081′
S_9	N36°55.815′, E54°54.284′	S_{23}	N37°11.051′, E55°10.134′	S_{37}	N37°55.412′, E55°7.365′
S_{10}	N36°59.805′, E54°58.523′	S_{24}	N36°50.059′, E54°18.612′	S_{38}	N37°55.14′, E55°6.091′
S_{11}	N37°1.612′, E55°0.476′	S_{25}	N36°53.207′, E54°8.075′	S_{39}	N37°53.541′, E55°54.329′
S_{12}	N37°4.533′, E55°8.753′	S_{26}	N36°56.702′, E54 4.861′	S_{40}	N37°51.142′, E55°57.633′
S_{13}	N37°3.575′, E55°8.313′	S_{27}	N37°1.058′, E54°5.516′	S_{41}	N37°41.14′, E55°44.95′
S_{14}	N36°58.893′, E55°6.97′	S_{28}	N37°5.914′, E54°4.006′	S_{42}	N37°38.721′, E55°42.463′

Table 2. The minimum detectable activity (MDA) for the radionuclides for ^{226}Ra, ^{232}Th and ^{40}K.

Radionuclide	MDA(Bq.kg^{-1})
^{232}Th	1.7
^{226}Ra	2.5
^{40}K	19.7

The Minimum Detectable Activity (MDA) is specified by gamma-ray measurement with 95% confidence [11]. For the purpose of the quality assurance, the MDA of the measurement system was calculated for the interested radionuclides. The MDA is estimated though the following equation [12]:

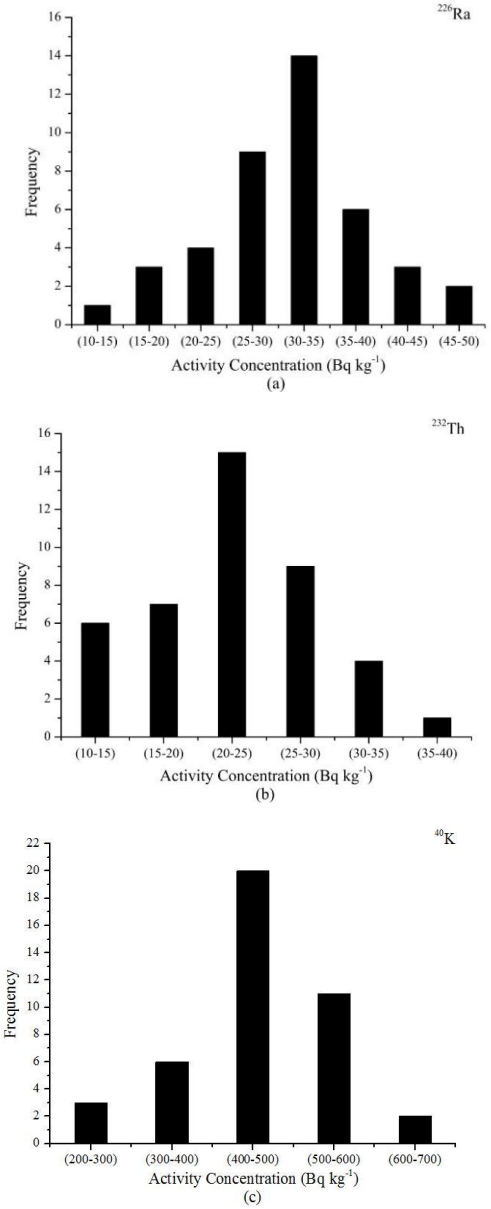

Figure 2. frequency distribution of ^{226}Ra (a), ^{232}Th (b) and ^{40}K(c) in soil samples all over the area of study.

$$MDA\ (Bq.kg^{-1}) = 4.65\sqrt{B.G}/\varepsilon t P_{\gamma} m \quad (1)$$

Where B.G is the number of counts for the interested photo peak in the background spectrum; ε is the absolute efficiency of the detector; t is the time of measurement; m is the weight of the dried sample in kg.

The calculated MDA values for measurement system are given in Table 2.

The activity concentrations of the samples were determined using the net area under the photo peaks according to the following equation (2):

$$A_c = C_n/P_{\gamma} m \varepsilon \quad (2)$$

Where A_c is the activity concentration of the radionuclides in the samples given in Bq.kg^{-1}.

The concentration of ^{40}K was measured directly by its own gamma peak at 1461 keV, whereas ^{226}Ra and ^{232}Th were estimated with the help of their gamma emitting daughter products, i.e., ^{4}Bi (609.51 keV) and ^{208}Tl (583.19 keV), respectively.

Results

The specific activities of the natural radionuclides in the soil samples taken from different locations of Golestan province are presented in Table 3. The concentration of ^{226}Ra, ^{232}Th, and ^{40}K varied within the ranges of 11.4-37.5, 14.8-49.8, and 225.5-666.9 Bq.kg^{-1} with the average values of 23, 31, and 453 Bq.kg^{-1}, respectively. The highest concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were measured in Ramian, Bandar-e Gaz, and Kordkuy cities. On the other hand, the lowest concentrations of the mentioned radionuclides were detected in Bandar-e Gaz, Kalaleh, and Khan Bebin cities, respectively.

The measured concentrations have been compared with those in other countries. Figure 2 presents the frequency distribution of ^{226}Ra, ^{232}Th, and ^{40}K in the soil samples of the investigated locations. Table 4 illustrates the mean activity concentration of the soil samples taken from Golestan, compared with similar studies performed in other countries. According to these data, the mean concentration of ^{226}Ra was higher in the sites investigated in this study than the values reported in Syria (20 Bq.kg^{-1}), Egypt (17 Bq.kg^{-1}), some parts of Turkey (i.e., Kilis [16 Bq.kg^{-1}], Sanliurfa [21 Bq.kg^{-1}], and Osmaniye [10 Bq.kg^{-1}]).

However, this value was lower than those reported for Azerbaijan (25 Bq.kg^{-1}), Bulgaria (45 Bq.kg^{-1}), Greece (29 Bq.kg^{-1}), Croatia (43 Bq.kg^{-1}), India (29 Bq.kg^{-1}), Turkey (Gaziantep, 25 Bq.kg^{-1}), and the world average (32 Bq.kg^{-1}) [13-17]. The mean level of ^{232}Th in the area of study was 31 Bq.kg^{-1}, which was less than the reported values for the world average (45 Bq.kg^{-1}), India (64 Bq.kg^{-1}), Azerbaijan (33 Bq.kg^{-1}), Croatia (37 Bq.kg^{-1}).

However, this value was higher than that of Bulgaria (30 Bq.kg^{-1}), Greece (28 Bq.kg^{-1}), Egypt (18 Bq.kg^{-1}), Syria (20 Bq.kg^{-1}), Turkey (i.e., Gaziantep [24 Bq.kg^{-1}], Sanliurfa [25 Bq.kg^{-1}], Osmaniye [12 Bq.kg^{-1}], and Kilis [15 Bq.kg^{-1}]) [13-17]. The mean level of ^{40}K in Golestan province was 453 Bq.kg^{-1} that was higher than those reported for the world average and other countries listed in Table 4 [13-17].

Table 3. The activity concentrations of ^{232}Th,^{226}Ra and ^{40}K analyzed in the soil samples

Sample code	Activity Concentration (Bq kg^{-1})			Sample code	Activity Concentration (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K
S_1	17.0±3.2	27.9±4.4	403.8±3.3	S_{23}	30.1±4.2	35.8±5.9	517.1±4.1
S_2	23.7±2.9	35.1±4.1	465.9±3.2	S_{24}	23.1±2.9	29.0±4.4	489.6±3.2
S_3	22.3±3.0	34.0±4.2	480.3±3.2	S_{25}	28.8±4.2	26.1±6.5	460.1±4.2
S_4	12.3±3.5	19.9±5.2	304.8±3.6	S_{26}	21.0±3.0	25.3±4.5	360.5±3.4
S_5	17.8±5.4	24.0±7.8	291.2±6.0	S_{27}	26.3±4.4	30.0±6.6	421.3±4.7
S_6	17.0±6.6	23.3±10.5	515.1±5.9	S_{28}	22.6±4.6	47.6±5.6	357.1±5.0
S_7	26.6±2.8	32.3±4.2	448.7±3.2	S_{29}	14.6±3.3	42.4±3.9	535.1±3.2
S_8	19.6±3.1	17.6±5.1	311.9±3.5	S_{30}	11.4±3.4	49.8±3.7	576.4±3.1
S_9	30.9±4.8	33.5±7.1	307.2±6.0	S_{31}	14.7±5.0	44.7±5.8	666.9±4.1
S_{10}	13.9±3.3	19.0±5.0	225.2±3.8	S_{32}	17.0±3.2	30.8±4.3	549.4±3.1
S_{11}	14.1±5.0	21.8±7.3	275.1±5.3	S_{33}	19.9±3.1	22.3±4.7	431.5±3.3
S_{12}	22.1±3.0	36.7±4.0	507.7±3.2	S_{34}	26.7±2.8	32.6±4.2	485.2±3.2
S_{13}	23.1±2.9	34.0±4.1	405.9±3.3	S_{35}	24.6±2.9	28.1±4.4	491.1±3.2
S_{14}	37.5±3.9	43.1±5.5	565.2±4.0	S_{36}	24.8±2.9	37.2±4.1	439.5±3.3
S_{15}	21.5±3.0	28.3±4.4	318.4±3.5	S_{37}	26.3±4.4	31.7±6.5	458.6±4.6
S_{16}	21.6±4.6	33.0±6.1	472.9±4.2	S_{38}	22.1±3.0	26.1±4.5	487.5±3.2
S_{17}	23.3±2.9	33.6±4.2	511.8±3.2	S_{39}	30.8±4.3	30.8±6.6	418.8±4.7
S_{18}	28.0±4.2	30.2±6.2	464.1±4.2	S_{40}	23.5±4.6	32.8±6.5	437.9±4.7
S_{19}	30.0±4.3	33.6±6.3	637.1±4.2	S_{41}	33.3±2.7	35.5±4.1	455.7±3.3
S_{20}	19.0±3.1	30.9±4.3	520.6±3.1	S_{42}	23.8±4.6	29.8±6.7	460.2±4.6
S_{21}	26.4±4.4	14.8±8.4	507.6±4.5	Mean	23.0	31.0	453.0
S_{22}	27.5±2.8	37.4±4.1	569.9±3.1				

Table 4. Comparison of ^{232}Th, ^{226}Ra and ^{40}K concentrations of soil samples with the available data from Iran and other countries.

Country	Activity Concentration (Bq kg^{-1})			References
	^{226}Ra	^{232}Th	^{40}K	
Azerbaijan	25	33	120	[13]
Bulgaria	45	30	400	[13]
Greece	29	28	383	[13]
Croatia	43	37	423	[13]
Egypt	17	18	320	[13]
India	29	64	400	[13]
Syria	20	20	270	[13]
Turkey (Gaziantep)	25	24	289	[14]
Turkey (Sanliurfa)	21	25	299	[15]
Turkey (Osmaniye)	10	12	243	[16]
Turkey (Kilis)	16	15	206	[17]
World average	32	45	412	[18]
Iran Golestan)	23	31	453	Present study

Table 5. Calculated values of radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), Internal hazard index (H_{in}), absorbed dose rate and annual effective dose in Golestan Province.

Sample code	Ra_{eq} (Bqkg^{-1})	H_{ex}	H_{in}	D (nGyh^{1})	E (μSvy^{1})	Sample code	Ra_{eq} (Bqkg^{-1})	H_{ex}	H_{in}	D (nGyh^{1})	E (μSvy^{1})
S_1	88.0	0.24	0.28	43.2	53.0	S_{23}	121.0	0.33	0.41	58.9	72.2
S_2	109.7	0.30	0.36	53.4	65.6	S_{24}	102.3	0.28	0.34	50.2	61.6
S_3	107.9	0.29	0.35	52.8	64.8	S_{25}	101.5	0.27	0.35	49.4	60.6
S_4	64.2	0.17	0.21	31.6	38.8	S_{26}	84.9	0.23	0.29	41.3	50.7
S_5	74.6	0.20	0.25	36.1	44.3	S_{27}	101.7	0.27	0.35	49.3	60.5
S_6	89.9	0.24	0.29	44.9	55.1	S_{28}	118.2	0.32	0.38	56.6	69.5
S_7	107.4	0.29	0.36	52.2	64.0	S_{29}	116.5	0.31	0.35	57.4	70.5
S_8	68.7	0.19	0.24	33.5	41.1	S_{30}	127.0	0.34	0.37	62.8	77.0
S_9	102.4	0.28	0.36	48.6	59.7	S_{31}	130.0	0.35	0.39	64.7	79.4
S_{10}	58.4	0.16	0.20	28.2	34.6	S_{32}	103.3	0.28	0.32	51.4	63.0
S_{11}	66.4	0.18	0.22	32.3	39.6	S_{33}	85.0	0.23	0.28	41.9	51.4
S_{12}	113.8	0.31	0.37	55.7	68.4	S_{34}	110.7	0.30	0.37	53.9	66.2
S_{13}	103.0	0.28	0.34	49.9	61.3	S_{35}	102.7	0.28	0.34	50.3	61.8
S_{14}	142.6	0.39	0.49	69.0	84.6	S_{36}	111.9	0.30	0.37	54.2	66.5
S_{15}	86.5	0.23	0.29	41.7	51.1	S_{37}	107.0	0.29	0.36	52.0	63.9
S_{16}	105.2	0.28	0.34	51.5	63.2	S_{38}	97.0	0.26	0.32	47.8	58.7
S_{17}	110.8	0.30	0.36	54.3	66.7	S_{39}	107.0	0.29	0.37	51.6	63.3
S_{18}	106.9	0.29	0.36	52.0	63.8	S_{40}	104.1	0.28	0.34	50.6	62.1
S_{19}	127.1	0.34	0.42	62.6	76.8	S_{41}	119.2	0.32	0.41	57.4	70.5
S_{20}	103.2	0.28	0.33	51.0	62.6	S_{42}	101.9	0.28	0.34	49.8	61.1
S_{21}	86.6	0.23	0.31	43.0	52.8	Mean	102.4	0.28	0.34	50.0	61.4
S_{22}	124.8	0.34	0.41	61.1	75.0						

Discussion

To assess the radiological hazard of the natural radioactivity, the Ra equivalent activity, the external and internal hazard (H_{ex} and H_{in}, respectively) indices, and the annual effective dose were calculated.

Radium equivalent activity is calculated through the following equation (3) [18,19]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3)$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the sample in Bq.kg^{-1}, respectively.

In this equation, it is assumed that 370 Bq.kg^{-1} of ^{226}Ra, 259 Bq.kg^{-1} of ^{232}Th, and 4810 Bq.kg^{-1} of ^{40}K produce the same gamma dose rate [20]. The results of the calculations are given in Table 5. The range of calculated Ra equivalent activity in the soil of study area was within 58.4-142.6 Bq.kg^{-1}, with an average of 102.4 Bq/kg, which is lower than the permissible maximum value of 370 Bq.kg^{-1} [13].

The H_{ex} and H_{in} can be calculated using the following equations (4 and 5), respectively [18]

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

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

The results of these calculations are presented in Table 5. The estimated representative level indices for all samples are less than unity indicating that the associated gamma radiation level is low [13].

The annual effective dose equivalent to be received by the public was calculated using [21]

$$E(\mu Sv/y) = D(nGy/h) \times 24h \times 365.25d \times 0.2 \times 0.7(Sv/Gy) \times 10^{-3} \quad (6)$$

Where [19]

$$D(nGy/h) = 0.427A_{Ra} + 0.662A_{Th} + 0.0432A_K \quad (7)$$

As indicated in Table 5, the maximum and minimum values of E were about 84.6 μSv y^{-1} and 34.6 μSv y^{-1}, respectively, which were related to Ramian and Khan Bebin. The mean value of E was found to be 61.4 μSvy^{-1}.

Conclusion

For the purpose of the study, 42 soil samples were taken from different locations of Golestan province. All samples were analyzed using the HPGe gamma ray detector. The concentrations of ^{226}Ra, ^{232}Th, and ^{40}K ranged within 11.4-37.5, 14.8-49.8, and 225.2-666.9 Bq.kg^{-1}, respectively. As the findings of the study revealed, the mean concentrations of ^{226}Ra (23 Bq.kg^{-1}) and ^{232}Th (31 Bq.kg^{-1}) in Golestan were lower than the world average .

The Ra equivalent activity varied between 58.4 and 142.6 Bq.kg^{-1}, and the H_{ex} and H_{in} indices were less than the acceptable limit of unity indicating that the associated gamma radiation level was low. The mean total annual effective dose was estimated to be 61.4 μSvy^{-1}, which is lower than the permissible limit. This study showed that the radiological risk due to natural radioactivity was negligible in the region under investigation.

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