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Measurement of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs concentrations in sediment samples and determination of annual effective dose due to these radionuclides in vicinity of hot springs in Kerman Province

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► ***Original article***

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ABSTRACT

Background: Natural and artificial radioactive materials are the most important sources of radiation for human existing in all over the world, thus investigating their radioactivity is of great importance because of their ionizing properties and harmful effects on human health. Therefore, this study was conducted to measure radionuclides concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in sediment samples collected from various hot springs in Kerman province and also determining received annual effective dose. ***Materials and Methods***: Thirteen sediment samples were collected from different hot springs and concentrations of radionuclides in sediment samples were measured via spectrometry method and using HPGe detector. To evaluate radiation hazards caused by these radionuclides, radium equivalent activity, absorbed dose rate in air, and annual effective dose of samples were calculated. ***Results***: Concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in sediment samples varied from 21.01±0.71 to 193.16±1.72, 16.84±1.21 to 245.92±2.69, 90.13±3.37 to 667.81±14.59 and <MDA to 5.81±0.66 $Bqkg^{-1}$, respectively. Also, minimum and maximum values of annual effective dose were determined as 0.17±0.00 and 1.48±0.01 in $mSvy^{-1}$, respectively. ***Conclusion***: Comparison of radionuclides concentrations and annual effective dose obtained in this study with the worldṡ average values determined by UNSCEAR (2000) reports showed that concentrations of ^{226}Ra and ^{232}Th in 69% and concentration of ^{40}K in 46% of measured samples were higher than the worldṡ average values. Also, annual effective dose of 30% of samples was found to be higher than the worldṡ average value.

INTRODUCTION

Water is one of the most important and essential sources of the environment, which has very high importance in maintaining human health. Hence, investigation on presence of various contaminants including natural and artificial radioactive substances in the water is of particular importance [1, 2]. Although, it is mostly believed that ionizing rays are received from irradiations of artificial and man-made sources, but in fact, most people in the community are exposed to these irradiations via natural radioactive sources in normal conditions [3, 4]. Environmental natural radioactivity and its external radiation mainly depend on geographical and geological conditions of the site, which may vary in each region throughout the world [5].

Gamma ray is one of important sources of external radioactive produced by natural radionuclides. These radioactive materials are available in the environment and their existence time dates back to earth formation [6]. More contact of groundwater to volcanic rocks and sedimentary substrates causes the increase in the concentration of radioactive materials in these waters compared to surface waters [7, 8]. Hot springs are one of sources of groundwater, which are important for investigating probability of radioactivity. High temperatures of these hot springs are due to volcanic activity as well as nuclear reactions and vibrational movements of faults [1]. So, considering therapeutic properties as well as features regarding tourist attraction related to these hot springs, it can be said that, people who use from them are exposed to radiation and radiation damages with various intensities [9]. Therefore, measurement of radionuclides concentrations in hot springs is essential in order to offer protection instructions to users of these hot springs.

In a study performed on the Kemessie hot spring in northeastern Ethiopia, average concentrations of

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measured radionuclides of ^{238}U, ^{232}Th and ^{40}K in soil samples collected from hot spring were equal to 248.71±20.8, 60.1±3.3 and 576.46±39.86 $Bqkg^{-1}$, respectively and average annual effective dose of these samples was equal to 0.88±0.06 μSvy^{-1} [10]. In a study done on the Seberang Perai area in north Malaysia, mean radionuclides concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples measured using high-purity Germanium (HPGe) detector were equal to 36.00±1.60, 44.21±1.78 and 254.60±4.26 $Bqkg^{-1}$, respectively and also average annual effective dose of these samples was obtained as 65.31±37.31 μSvy^{-1} [5].

The aim of present study was to measure ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs concentrations in sediment samples and determine the annual effective dose in vicinity of hot springs in Kerman province. Considering that, there is no study determining concentration of radionuclides in hot springs of Kerman province so far, as well as because of risks caused by decaying of radionuclides for visitors of these hot springs, this study had novelty.

MATERIALS AND METHODS

In this study, radionuclides concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were measured in sediment samples collected from 13 hot springs in different cities of Kerman province.

Sampling and sample preparation

From each hot spring, 1 kg of sediment sample was collected in polyethylene special containers, which are chemically resistant to sediments compositions. Then these samples were dried for 5 days in open air. To achieve constant weight of the sample, each sample was placed in an oven at 105 °C for 24 hours. After that, the sample was taken out of oven, and then was cooled at room temperature. In the next step, the sample was homogenized, so that, using a large mesh, particles and stones in the sample were separated, then using a grinder, the sample was pulverized and was converted into powder. Finally, to obtain a homogeneous sample, it was passed through a 50-mesh (300 µm) according to standard protocol of IAEA-TECDOC-1360 [11]. Regarding to weight of pulverized samples, Negin container was used to measure concentrations of radionuclides. Then, 293 g of homogeneous sample was placed inside Negin container, and to prevent escape of radon gas, resulting from decaying of heavy special nuclides, the samples were shielded using thermal adhesive. These samples were coded according to numbers related to each hot spring from 1 to 13, respectively. Finally, the code for each sample with its shielding date was recorded on sample container. To maintain equilibrium between ^{226}Ra and its short-lived decayed products as well as between ^{232}Th and its daughter nuclides, shielding of the samples lasted at least 5 weeks.

Spectrometry, analysis of samples and measurement of concentration

Spectrometry method was applied to measure radionuclides concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in prepared samples. For this purpose, coaxial HPGe detector (the production of USA CANBERRA Company) of p-type and with 40% relative efficiency was used. HPGe detector is a high-resolution spectrometry system used for measuring energy spectrum of emitted gamma rays in energy range from 50 to 3000 keV.

At a counting time of 86400 s, spectroscopy was performed for each sample by this detector. Also, to correct the spectrum related to each sample, background gamma-ray spectrum around the detector was measured with an empty Negin container under the same condition and geometry, and then was subtracted from spectrum of each sample. Spectrum analysis of samples was performed using Gamma Vision-32 software, which simultaneously analyzes all photopeaks appearing in the spectrum. After analyzing the spectrum, equation 1 was used to determine radionuclides concentrations in sediment samples [12].

$$\text{Act } (Bqkg^{-1}) = \frac{\text{Net Area}}{\varepsilon.(B.R\%).t.m} \times 100 \quad (1)$$

Where, Act is the concentration in $Bqkg^{-1}$, Net Area is the net count under peak, ε is the detector efficiency for the specific gamma-ray energy, (B.R%) refers to the branching ratio of gamma-ray intensity in terms of percent, t is the counting time of spectrum in sec and m shows the mass of the sample in kg.

Calculation of radium equivalent activity, absorbed gamma dose rate and annual effective dose

Since distribution of radionuclides is not uniform in sediment samples, and also about 98.5% of radiological effects of uranium chain depend to radium and its daughters, hence a radiological index is defined as radium equivalent activity, which estimates radiation hazards of these radionuclides. This quantity is calculated by equation 2 [13].

$$Ra_{eq}\,(Bqkg^{-1}) = C_{Ra} + 1.43\,C_{Th} + 0.077\,C_K \quad (2)$$

Where, Ra_{eq} is the radium equivalent activity in $Bqkg^{-1}$, C_{Ra}, C_{Th} and C_K are the concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$, respectively.

Absorbed dose rate in air at height of 1 meter above hot spring surface caused by ^{226}Ra, ^{232}Th and ^{40}K radionuclides in sediment samples was determined by equation 3 [13].

$$D\,(nGyh^{-1}) = 0.462\,C_{Ra} + 0.604\,C_{Th} + 0.0417\,C_K \quad (3)$$

Where, D is the absorbed dose rate in air in $nGyh^{-1}$

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and the constant coefficients are conversion factor. The annual effective dose of absorbed by individuals was calculated according to equation 4 [13].

$$E_{eff}\ (mSvy^{-1}) = D\ (nGyh^{-1}) \times 0.7\ (SvGy^{-1}) \times 8760\ (hy^{-1}) \times 10^{-6} \quad (4)$$

Where, E_{eff} is the annual effective dose in $mSvy^{-1}$, D is the absorbed dose rate in air and 0.7 is conversion factor for adults.

Statistical analysis

Correlation between obtained data was analyzed using SPSS-23 statistical software and by general linear model. The significant level was considered at $p<0.05$.

RESULTS

Measured values of radionuclides concentrations in sediment samples, as well as calculated values for radium equivalent activity, absorbed dose rate in air and annual effective dose are presented in table 1.

Table 1. Concentration values of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs as well as radium equivalent activity, absorbed dose rate in air and annual effective dose in sediment samples collected from 13 hot springs in Kerman province.

Sample Code	^{226}Ra ($Bqkg^{-1}$±1SD)	^{232}Th ($Bqkg^{-1}$±1SD)	^{40}K ($Bqkg^{-1}$±1SD)	^{137}Cs ($Bqkg^{-1}$±1SD)	Ra_{eq} ($Bqkg^{-1}$±1SD)	D ($nGyh^{-1}$±1SD)	E_{eff} ($mSvy^{-1}$±1SD)
S-1	57.34±1.03	42.43±2.14	452.49±10.45	<MDA*	152.85±4.89	70.98±2.20	0.43±0.01
S-2	193.16±1.72	245.92±2.69	90.13±3.37	<MDA	551.76±5.82	241.53±2.55	1.48±0.01
S-3	74.42±1.97	46.25±2.79	600.09±16.44	4.51±1.24	186.76±7.22	87.34±3.28	0.53±0.02
S-4	30.48±1.08	30.11±1.55	494.30±10.66	2.10±0.45	111.59±4.11	52.88±1.87	0.32±0.01
S-5	27.97±1.09	19.09±1.63	343.56±9.05	5.81±0.66	81.72±4.11	38.77±1.86	0.23±0.01
S-6	75.06±2.55	56.21±5.50	520.83±19.96	<MDA	195.54±11.95	90.34±5.33	0.55±0.03
S-7	89.93±1.85	16.87±2.57	285.94±10.23	<MDA	136.07±6.31	63.66±2.83	0.39±0.01
S-8	21.01±0.71	16.84±1.21	210.50±6.32	0.98±0.33	61.29±2.92	28.65±1.32	0.17±0.00
S-9	68.15±3.45	56.03±1.57	351.49±17.53	<MDA	175.33±7.04	79.98±3.27	0.49±0.02
S-10	38.64±1.02	52.80±1.91	357.78±8.63	2.53±0.72	141.69±4.41	64.66±1.98	0.39±0.01
S-11	40.18±1.22	31.07±2.00	667.81±14.59	2.25±0.55	136.03±5.20	65.17±2.38	0.39±0.01
S-12	47.19±1.05	35.26±1.60	441.72±10.51	0.84±0.26	131.62±4.14	61.51±1.88	0.37±0.01
S-13	26.72±1.17	19.75±1.53	310.22±8.18	1.63±0.42	78.84±3.98	37.20±1.80	0.22±0.01

(*: Minimum Detectable Activity).

As illustrated in table 1, concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in sediment samples ranged from 21.01 to 193.16, 16.84 to 245.92, 90.13 to 667.81 and <MDA to 5.81 $Bqkg^{-1}$, respectively. The lowest concentrations of ^{226}Ra and ^{232}Th were measured in the sample collected from Jooshan (S-8) hot spring and the lowest concentration of ^{40}K was measured in the sample collected from Babtorsh (S-2) hot spring. Also, the lowest value of ^{137}Cs (<MDA) was determined in samples collected from Amireh Keykhosravi (S-1), Babtorsh (S-2), Ghezak (S-6), Hormak (S-7) and Khajeh (S-9) hot springs. The highest values of ^{226}Ra and ^{232}Th were in the sample collected from Babtorsh hot spring (S-2) and the highest values of ^{40}K and ^{137}Cs were in the samples collected from Maskoun (S-11) and Gevar (S-5) hot springs, respectively.

According to table 1, measured concentration of ^{40}K was relatively higher than other radionuclides in all the samples except for the sample related to Babtorsh hot spring (S-2). Concentrations of radionuclides measured in sediment samples of this study were compared with the worldś average values determined by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR (2000)) reports. According to this report, the world's average values for ^{226}Ra, ^{232}Th and ^{40}K are equal to 35, 30 and 400 $Bqkg^{-1}$, respectively.

Results presented in table 1 showed that, samples collected from Amireh Keykhosravi (S-1), Babtorsh (S-2), Chamanrang (S-3), Ghezak (S-6), Hormak (S-7), Khajeh (S-9), Khodadadi (S-10), Maskoun (S-11) and Shirinak (S-12) hot springs had ^{226}Ra concentration higher than 35 $Bqkg^{-1}$. Concentration of ^{232}Th in the samples collected from Amireh Keykhosravi (S-1), Babtorsh (S-2), Chamanrang (S-3), Dehraees (S-4), Ghezak (S-6), Khajeh (S-9), Khodadadi (S-10), Maskoun (S-11) and Shirinak (S-12) hot springs was higher than 30 $Bqkg^{-1}$. Also, ^{40}K concentration measured in the samples collected from Amireh Keykhosravi (S-1), Chamanrang (S-3), Dehraees (S-4), Ghezak (S-6), Maskoun (S-11) and Shirinak (S-12) hot springs was higher than 400 $Bqkg^{-1}$.

Statistical analysis performed between data obtained in this study showed that, there was no significant correlation between measured radionuclides concentrations with temperature and hot spring height from free sea level.

According to table 1, the lowest radium equivalent activity was found in the sample collected from Jooshan (S-8) hot spring with value of 61.29 $Bqkg^{-1}$, and the highest radium equivalent activity was found in the sample collected from Babtorsh (S-2) hot spring with value of 551.76 $Bqkg^{-1}$. Results showed that only value of the sample collected from Babtorsh (S-2) hot spring was higher than permissible maximum value Ra_{eq} (370 $Bqkg^{-1}$) determined by UNSCEAR (2008) reports.

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Range of variations in absorbed dose rate in air was between 28.65 to 241.53 in $nGyh^{-1}$. The worldś average value for absorbed dose rate is equal to 59 $nGyh^{-1}$ [11] and results of the present study showed that values of the samples collected from Amireh Keykhosravi (S-1), Babtorsh (S-2), Chamanrang (S-3), Ghezak (S-6), Hormak (S-7), Khajeh (S-9), Khodadadi (S-10), Maskoun (S-11) and Shirinak (S-12) hot springs were higher than 59 $nGyh^{-1}$.

The lowest and highest values of annual effective dose were determined as 0.17 and 1.48 in $mSvy^{-1}$, respectively, related to the samples collected from Jooshan (S-8) and Babtorsh (S-2) hot springs, respectively. Figure 1 shows histogram related to variations in annual effective dose in sediment samples collected from hot springs.

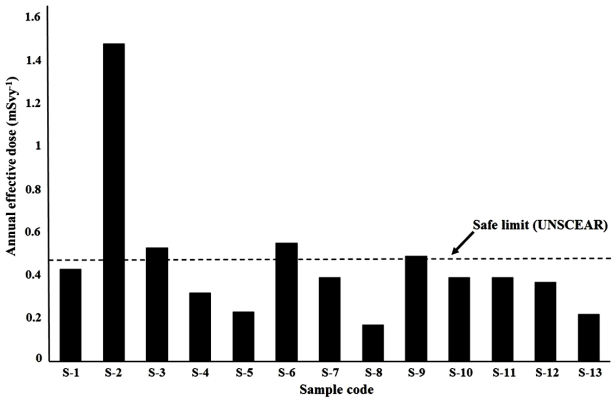


Figure 1. Variations in received annual effective dose from collected sediment samples associated with different hot springs in Kerman province and comparison of them with safe limit (0.48 $mSvy^{-1}$) proposed by UNSCEAR organization [13].

Comparison of values of annual effective dose obtained in this study with the worldś average annual effective dose (0.48 $mSvy^{-1}$) determined by UNSCEAR (2000) reports (13), as well as considering results presented in figure 1, the samples collected from Babtorsh (S-2), Chamanrang (S-3), Ghezak (S-6) and Khajeh (S-9) hot springs were found to have values higher than the worldś average value. Also, no significant correlation was observed between values of annual effective dose with temperature and hot spring height from free sea level in all investigated samples.

DISCUSSION

Investigating radioactivity condition in hot springs is of great importance, because of the absorption possibility of radioactive and mineral materials available in deep layers of earth's crust by waters of these hot springs during passing through these layers, especially under high pressure [14], thus can make them as carriers of radioactive materials into hot springs. Therefore, people who use these hot springs are at risk of radiation exposure, depending on radioactivity level of hot springs. In this study, the reason for selecting these hot springs, has been due to the high public reception as well as the accessibility possibility of visitors to them.

Comparison of radionuclides concentrations measured in the present study with the world's average values determined by the UNSCEAR (2000) reports showed that values of ^{226}Ra as well as ^{232}Th in 69% of measured samples and in case of ^{40}K in 46% of studied samples were higher than the worldś average values. In the samples collected from Amireh Keykhosravi (S-1), Chamanrang (S-3), Ghezak (S-6), Maskoun (S-11) and Shirinak (S-12) hot springs, values of all three radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were higher than the worldś average values.

^{137}Cs radionuclide was observed in 8 sediment samples with low values, which may be due to contamination from explosion accident occurred in Chernobyl, or testing of bomb and nuclear weapons carried out by several countries or any combination of these cases [15], which consequently transmitted to Iran through atmospheric processes or environmental factors [6].

In the present study, values of annual effective dose measured in sediment samples were between 0.17 to 1.48 in $mSvy^{-1}$ and these values were compared with the worldś average annual effective dose (0.48 $mSvy^{-1}$) recommended by UNSCEAR (2000) reports and results showed that 30% of samples had an annual effective dose higher than the worldś average value.

Concentrations values of radionuclides, as well as annual effective dose of the samples obtained in this study are comparable with other studies conducted in Iran and in countries around the world, which results are shown in table 2.

As shown in table 2, in a study done by Pourimani *et al.*, on radioactivity of hot springs in Mahallat (Iran) [6] as well as in a study performed by T. Bhongsuwan *et al.*, on the Khao-Than hot spring in south Thailand [16], showed that among measured values, ^{226}Ra had the highest value in comparison with other radionuclides, while in the present study, the highest concentration value was related to ^{40}K. Muslim Murat *et al.*, investigated the effects of radioactivity on soil of the Seferihisar Geothermal Region (SGR) in southwestern Turkey [14], as well as Pradeep *et al.*, in a study on the Kemessie hot spring in Ethiopia [10] and Alnassar *et al.*, in a study on the Seberang Perai region of Malaysia [5], which were similar with the present study in terms of high concentration of ^{40}K.

Alizadeh *et al.*, investigated environmental gamma in Qinarjeh, Ilando and Moiel-suyie hot springs in Meshgin Shahr (Iran) and showed that average annual effective dose of hot springs were equal to 26.3, 21.6 and 22.04 in $mSvy^{-1}$, respectively, which were very higher than the worldś average value (0.48 $mSvy^{-1}$) ,thus the area around these hot springs can be considered as a region with high natural radioactivity [19], also, these measured values showed very high radioactivity compared to the present

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Table 2. Comparison of radionuclides concentrations measured in sediment samples of the present study with other studies conducted in Iran and in countries around the world.

Country or region	^{226}Ra (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})	^{137}Cs (Bqkg^{-1})	Reference no
Mahallat (Donbe hot spring)	1405.9	26.29	137.33	3.53	6
Mahallat(Soleymani hot spring)	1630.56	39.99	125.4	<MDA	6
Thailand (Khao-Than hot spring)	From 151 to 139092	From 12 to 596	From 24 to 616	–	16
Turkey (Seferihisar geothermal region)	From 3.3 to 120.3	From 0.3 to 108.5	From 116 to 850	–	14
Malaysia (the Seberang Perai area)	36	44.21	254.60	–	5
Farasan Island	3.31	0.92	34.34	0.14	17
Northern coast of Oman Sea	From 11.83 to 22.68	From 10.7 to 25.02	From 222.89 to 535.07	From 0.14 to 2.8	18
Iran (Arak)	From 13.12 to 33.03	From 11.3 to 35.86	From 257.82 to 605.5	From 1.28 to 13.36	15
Present study	From 21.01 to 193.16	From 16.84 to 245.92	From 90.13 to 667.81	From <MDA to 5.81	–
World average values	35	30	400	–	13

study.

In a study conducted by Jomehzadeh *et al.*, on Amireh Keykhosravi, Chamanrang, Hormak, Jooshan, Khodadadi, Maskoun and Shirinak hot springs in Kerman province, in which RDS-110 survey meter device was used, values of annual effective dose in these hot springs were measured to be 1.16, 0.79, 0.97, 0.49, 0.88, 0.79 and 0.97 in mSvy^{-1}, respectively [3], while in the present study, in which HPGe detector was used, annual effective dose in these hot springs were obtained to be 0.43, 0.53, 0.39, 0.17, 0.39, 0.39 and 0.37 in mSvy^{-1}, respectively, and also comparison of these results showed that in this study, annual effective dose of all hot springs has decreased.

Comparison of all these studies with the present study revealed that, in most cases, measured concentration of ^{40}K was higher than concentration of other radionuclides. Difference in measured values in these studies depend on structure and geological characteristics, geographic conditions, soil amount and material in each area as well as environment of sampling location [5].

According to UNSCEAR (2000) reports, areas of the world such as Brazil, China, Egypt, France, India, Iran (Ramsar and Mahallat), Italy, Niue Island, and Switzerland have high levels of natural background radiation [13]. Reviewing these studies as well as other performed studies in field of radioactivity caused by radionuclides showed that, the possibility of this kind of radiation is almost everywhere on earth, and needs to be evaluated, especially in areas where radiation values are higher than safe limits, essential measures should be taken in order to prevent from the harmful effects on human health.

CONCLUSION

Results of this study showed that, concentration values of ^{226}Ra and ^{232}Th in 9 sediment samples and ^{40}K value in 6 sediment samples were higher than the worldś average values. Also, annual effective dose values of 4 sediment samples were found to be higher than the worldś average annual effective dose. So, because of presence of radioactivity amounts higher than safe limits in some hot springs, responsible organizations and institutions should take appropriate measures and necessary instructions in order to reduce hazards for people who use from these hot springs.

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