

Original Article

Measurement of Radionuclide Concentration in Some Water Resources in Markazi Province, Iran

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Abstract

Introduction

Natural and artificial radionuclides are the main sources of human radiation exposure. These radionuclides, which are present in the environment, can be dissolved into water. Evidence suggests that radionuclides being entered the human body through drinking or hot spring water can be harmful for human health.

Materials and Methods

In this study, 10 samples were collected from ground water resources of Arak, one sample from the surface water of Kamal-Saleh Dam, and four samples from the hot springs of Mahallat region. The specific activities of ²²⁶Ra, ²³²Th, ⁴⁰K, and ¹³⁷Cs were determined in the samples, using gamma ray spectrometry and a high-purity germanium (HPGe) detector.

Results

Specific activities of ²²⁶Ra, ²³²Th, ⁴⁰K, and ¹³⁷Cs were determined in the water samples. The mean ²²⁶Ra activity concentrations in drinking water samples from Aman Abad, Mobarak Abad, and Taramazd wells were 7.65 ± 1.64 , 1.56 ± 1.04 , and 1.45 ± 1.39 Bq/l, while the corresponding values for ²³²Th were 2.70 ± 0.18 , 0.41 ± 0.16 , and 1.27 ± 0.44 Bq/l, respectively. The annual effective dose due to drinking water varied from 0.01 to 0.78 mSv/y. Moreover, the specific activity of ²²⁶Ra in the water samples from the orifice of Donbe, Shafa, Soleymani, and Souda hot springs varied from 0.47 ± 0.16 to 1.90 ± 0.21 .

Conclusion

The calculated annual effective dose due to water consumption by Iranians was within the average annual global range. Therefore, based on the present results, radionuclide intake due to water consumption had no consequences for public health; however, it is recommended that hot spring baths use air conditioning devices.

Keywords: Effective Dose, Hot Spring, Radionuclide, Water resource

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1. Introduction

Water is a very important element in environmental studies due to its daily use by humans and the possibility of water-related infections [1,2]. Ground water contains natural radionuclides, such as ^{40}K , as well as natural decay chains of ^{235}U , ^{238}U , and ^{232}Th [3]. The dissolution of uranium and thorium salt enters rocks and sediment layers in the soil and pollutes the ground water [4].

The average ^{238}U content in the Earth's crust has been estimated at 2.7 mg/kg, and its concentration may be as high as 120 mg/kg in phosphate rocks [5]. Meanwhile, the average ^{232}Th content in the Earth's crust is nearly 9.6 mg/kg [6]. The presence of such elements in the ground water depends on three main factors, i.e., geological, hydraulic, and chemical properties of water [7].

The isotopes of uranium (^{234}U , ^{235}U , and ^{238}U) exhibit non-negligible radiotoxicity [8]. Overall, the most important radiotoxic agents include radium isotopes and radon gas. Among different radionuclides, ^{226}Ra and ^{222}Rn from ^{238}U decay series and ^{228}Ra from ^{232}Th decay series have shown the greatest impact on human health [9]. Considering the high radiotoxicity of radium isotopes, their presence in water, and the associated health risks, particular attention should be paid to these isotopes.

It is well established that even small amounts of radioactive substances may produce damaging biological effects; moreover, ingested and inhaled radiation can be regarded as serious health risks. The metabolic behavior of radium is similar to calcium, as it enters the human body. An appreciable fraction of radium is deposited in the bones, while the remaining is distributed almost uniformly among soft tissues [10].

If radionuclides enter the body through drinking water, inhalation, or skin absorption, they gradually distribute in the body, and consequently, the energy of the emitted radiation causes irreparable damages, such as bone cancer, leukemia, genetic problems, blood vessel damage, eye cataract, and

infertility [11,12]. Consequently, in recent years, there has been a surging interest in the study of radioactivity in drinking water in many countries.

The radium concentration in surface water normally ranges from 0.01 to 0.1 Bq/l [13]. The highest concentrations have been detected in water resources close to uranium mining and milling sites [14, 15]. In ground water, the radium concentration can reach up to 38 Bq/l, depending on factors such as the type of aquifer and its chemical and physical characteristics [16].

With this background in mind, the present study aimed to determine the presence and distribution of natural and artificial radionuclides, i.e., ^{235}U , ^{232}Th , ^{226}Ra , ^{40}K , and ^{137}Cs in drinking water resources of Arak, located in the northwest of Iran. ^{137}Cs radionuclide is not naturally found in the environment and is normally distributed due to nuclear weapon tests and accidents in nuclear power plants. This radionuclide distributes in the environment with radioactive dust through the atmospheric process and can penetrate into the soil, thus causing pollution in surface and ground water.

In Mahallat region, there are four hot springs, known as Shafa, Soleymani, Donbe, and Souda, visited by people every year, with a total yield of 35.5 l/s and temperature of 18.2-47.3°C [17]. Hot spring regions show high levels of radioactivity, and the hot water is used for hydrotherapy by tourists and for irrigation by farmers; therefore, study of this region in terms of radionuclide distribution is essential.

2. Materials and Methods

2.1. Sampling and sample preparation

At present, Arak has 25 active wells (depth of 120 m), used to supply drinking water for the region. In this study, due to the proximity of the wells (about 500 meter distance) and topographic features of the area, 11 active wells were sampled, including five samples from Aman Abad region, three from Mobarak Abad region, three from Taramazd, and one

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from the surface water of Kamal-Saleh Dam. Also, four samples were obtained from the orifices of Donbe, Shafa, Soleymani, and Souda hot springs, respectively.

The samples were transferred to 1.5 L polyethylene containers from special valves, which were built on the wells for collecting drinking water samples. Immediately after filling the bottles, pH of the samples decreased by adding nitric acid, which was consequently adjusted to 2; this pH value was required for preventing radionuclide absorption by the container walls [18]. All the samples were

prepared in Marinelli beakers for gamma spectroscopic analysis.

The collection of water samples requires particular care, since radon is a short-lived gaseous nuclide and tends to escape from water during sampling. In this study, a Marinelli beaker with the volume of 800 cc was used and sealed. The gamma ray was registered at 210 days (minimum duration) following the preparation of the samples, since this duration is longer than what is necessary for reaching radioactive chain equilibrium [19]. The sampling locations are shown in Figures 1 and 2.



Figure 1. Sampling Location around Arak

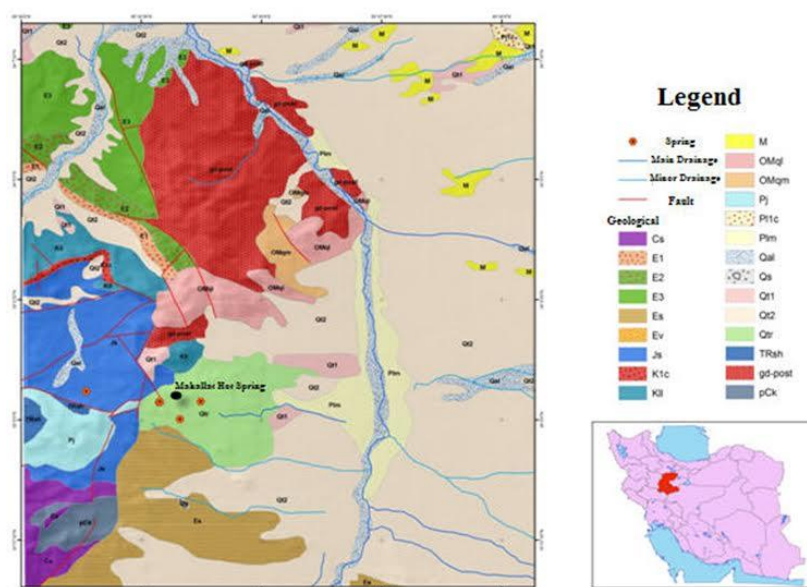


Figure 2. Geological map of hot springs zone 1: 250000

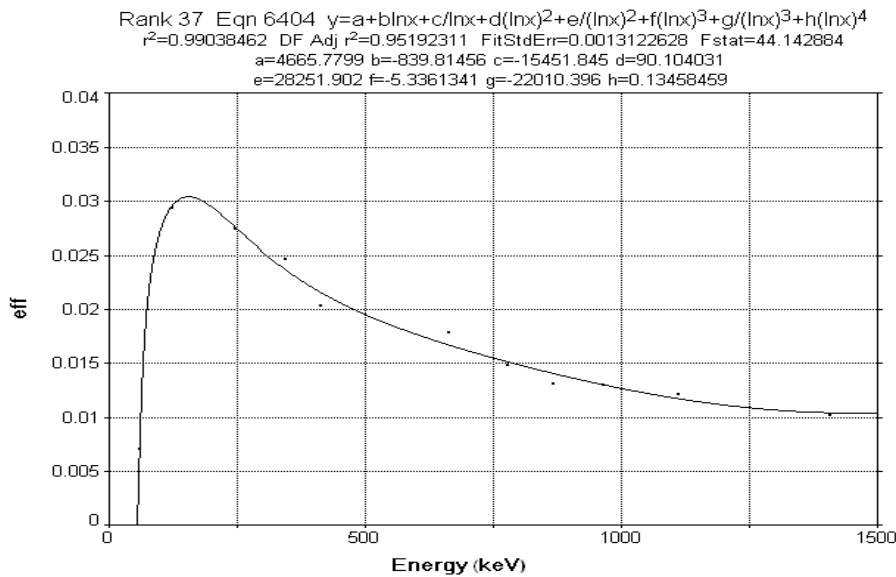


Figure3. Detector efficiency curve for standard Marinelli Beaker source

2.2. Experimental setup

The measurements were performed, using a gamma-ray spectrometer, P-type coaxial HPGe detector with 80% relative efficiency, and a multichannel analyzer (with 8192 channels). The energy resolution of the detector was 1.85 keV for a 1332.52 keV energy gamma ray produced by ^{60}Co . The operating voltage was 3000 V, and standard Marinelli containers were used in the measurements.

The graded shield detector in the chamber included two lead and copper layers, respectively (thickness of 10 cm and 3 mm, respectively); this shield served to reduce the background radiation. The soft components of cosmic ray, consisting of photons and electrons, were reduced to a very low level by 100 mm of lead shielding. The X-ray (73.9 keV), emitted from the lead by its interaction with external radiation, was suppressed by the copper layer [20].

To minimize the effect of scattering radiation from the shield, the detector was located in the center of the chamber. Then, the sample was placed in a face-to-face geometry over the detector for 86,400 sec. The features of Marinelli containers were as follows: 800 cc volume, outer diameter of 14 cm, height of 11

cm, inner diameter of 10 cm, and internal height of 7.5 cm.

The system was calibrated in terms of energy and efficiency. The energy calibration was carried out by using the radioactive standard source. Regarding efficiency calibration, we used the Marinelli beaker standard source, including radioisotopes with exact activities of ^{241}Am , ^{152}Eu , and ^{137}Cs . According to the registered gamma ray spectrum, the absolute efficiency of the detector configuration was calculated, using Equation (1):

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

where N_i is the net count under the full-energy peak corresponding to E_i energy, Act indicates radioisotope activity, $P_n(E_i)$ shows the probability of E_i photon emission, and T denotes the counting time [19]. The plot of efficiency versus gamma ray energy is depicted in Figure 3. Furthermore, the function fitted to the experimental data by the polynomial curve was as follows:

$$y = a + blnx + c/lnx + d(\lnx)^2 + e/(\lnx)^2 + f(\lnx)^3 + g/(\lnx)^3 + h(\lnx)^4 \quad (2)$$

where y refers to efficiency, a, b, c, d, e, g, and h are constants as shown on top of Figure 3, and x is the gamma ray energy, expressed in keV.

The ^{226}Ra activity of the samples was determined, considering the intensities of

351.9 and 609.3 keV gamma lines of ^{214}Pb and ^{214}Bi , respectively. ^{232}Th activity was measured, using the gamma line of 911.21 keV ^{228}Ac with an intensity of 26.6% and 968.97 keV ^{228}Ac with an emission percentage of 17.4%. In order to determine ^{235}U activity concentration, 143.78 and 205.03 keV gamma ray lines of ^{226}Ra were used. Moreover, ^{40}K and ^{137}Cs were obtained using 1460.7 and 661.6 keV gamma ray lines, respectively.

All 15 registered gamma ray spectra were analyzed, and the activity concentration was calculated with ORTEC GammaVision-32 Software. In all the analyzed spectra, correction was performed for the background gamma ray, which was registered using an empty Marinelli container. In order to calculate the activity concentration, Equation 3 was applied [19]:

$$\text{Act} = \frac{\text{Net Area}}{\epsilon \times (\text{BR}) \times T \times v} \quad (3)$$

where net area is the net count under the peak, Act (Bq/l) is the activity concentration, ϵ is the energy efficiency for the gamma ray by the detector, BR is the branching ratio of gamma ray intensity (%), T (s) denotes the time of spectra, and v (L) is the volume of the sample.

The annual effective dose for drinking water was calculated, based on the intake of individual radionuclides and ingestion dose coefficients (Sv/Bq), reported by the International Commission on Radiological Protection (ICRP) [21]. The annual effective dose per person was calculated as follows:

$$\text{Annual effective dose} = \sum_i I_i \times 365 \times D_i \quad (4)$$

Where I_i is the daily intake of radionuclide i (Bq/d), and D_i denotes the ingestion dose coefficient (Sv/Bq). It should be noted that the volume of drinking water intake by adult males in Arak is reported to be 1 l/d [8].

3. Results

Table 1 shows the activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs in AMW01, AMW04, AMW07, AMW11, and AMW14 samples from well No. 1, 4, 7, 11, and 14 in Aman Abad region, respectively. Moreover, the activity concentrations in MBW01, MBW02, and MBW03 samples from well No. 1, 2, and 3 in

Mobarak Abad region, TRW01, TRW02, and TRW03 samples from well No. 1, 2, and 3 in Taramazd, and KSDS01 sample from the surface water of Kamal-Saleh Dam are presented in this table, respectively.

Table 2 shows the average activity concentrations in the ground water of the three studied regions (Aman Abad, Mobarak Abad, and Taramazd) and the tap water from Kamal-Saleh Dam, irrespective of values lower than the minimum detectable activity. Also, Table 3 shows the annual effective dose received by the population, resulting from the ingestion of ^{226}Ra and ^{232}Th in water. The measurements showed that ^{226}Ra radioactivity concentration in Aman Abad ground water was higher than other regions, with the minimum value attributed to Taramazd wells.

The maximum ^{232}Th radioactivity concentration was obtained for well No. 14 in Mobarak Abad region. Based on the present results, the surface water of Kamal-Saleh Dam contained low amounts of ^{232}Th and ^{137}Cs . In fact, ^{137}Cs radionuclide was observed in some ground water and surface water samples. This radionuclide originated from nuclear accidents or nuclear weapon tests in other countries. In such cases, the radionuclide distributed in the environment with radioactive dust through the atmospheric process, and in some cases, it penetrated into the ground water.

The AMW07 and AMW14 ground water samples contained the highest ^{226}Ra concentration; also, ^{232}Th and ^{40}K were detected in the samples. According to the findings, these elements should be removed from drinking water resources. Additionally, the surface water from Kamal-Saleh Dam contained ^{232}Th radioisotope chain. It is speculated that the river (with the same name), which originates from Zagros mountains, passes through rocks containing this isotope on its path to the dam.

Comparison of radionuclide specific activity concentrations of Arak ground and surface water with water resources in other countries showed higher levels of radiation in the consuming

water of Arak city. This could be due to changes in rocks and soil structure in lower layers of

rocks, eroded by the ground water entering from the surface.

Table 1. Specific activities of ^{238}U , ^{232}Th , ^{226}Ra , ^{40}K and ^{137}Cs (Bq / l) in water resources of Arak.

Sample code	^{226}Ra (Bq/l)	^{232}Th (Bq/l)	^{40}K	Cs^{137}	
Ground water	AMW01	1.26±1.03	1.78±0.20	1.77±0.68	< MDA
	AMW04	< MDA	< MDA	< MDA	0.06±0.02
	AMW07	11.08±0.80	1.42±0.19	1.26±0.69	< MDA
	AMW11	< MDA	1.61±0.16	4.23±0.65	< MDA
	AMW14	10.62±1.00	7.45±0.23	2.78±0.67	< MDA
	MBW01	1.16±1.02	< MDA	< MDA	< MDA
	MBW02	1.96±0.96	< MDA	< MDA	0.07±0.03
	MBW03	< MDA	0.41±0.16	< MDA	< MDA
	TRW01	1.75±1.07	1.55±0.20	2.34±0.68	< MDA
	TRW02	< MDA	1.64±0.20	1.82±0.66	< MDA
	TRW03	1.15±0.89	0.42±0.11	< MDA	< MDA
	Surface water	KSDS01	< MDA	0.33±0.14	< MDA
Hot Springs	Donbe	0.90±0.19	<MDA	< MDA	< MDA
	Shafa	1.62±0.24	<MDA	< MDA	< MDA
	Soleimani	1.09±0.15	0.64±0.20	< MDA	< MDA
	Souda	0.47 ± 0.16	< MDA	< MDA	< MDA

Table 2. Mean radio activity concentration of ^{232}Th , ^{226}Ra and ^{40}K (Bq/l) in drinking water of Arak

Sample code	^{226}Ra (Bq/l)	^{232}Th (Bq/l)	^{40}K	
Ground water	A	7.65± 1.64	3.06 ±0.33	2.51±1.34
	M	1.56±1.41	0.41±0.16	< MDA
	T	1.45±1.39	1.20 ± 0.30	2.08±0.94
Surface water	KSDS01	< MDA	0.33±0.14	5.32±0.99

Table 3. Estimated annual effective doses for drinking water ingestion

Nuclide	Region	Intake, Bq d ⁻¹	Ingestion dose	
			Coefficient (Di), Sv Bq ⁻¹	Annual effective dose, mSv y ⁻¹
ICRP 68 (1994)				
^{226}Ra	Aman Abad	7.65± 1.64	2.8×10^{-7}	0.78
	Mobarak Abad	1.56±1.41	2.8×10^{-7}	0.16
	Taramazd	1.45±1.39	2.8×10^{-7}	0.15
	Kamal Saleh	-----	2.8×10^{-7}	-----
^{232}Th	Aman Abad	3.06 ±0.33	9.2×10^{-8}	0.10
	Mobarak Abad	0.41±0.16	9.2×10^{-8}	0.01
	Taramazd	1.20 ± 0.30	9.2×10^{-8}	0.04
	Kamal Saleh	0.33±0.14	9.2×10^{-8}	0.01

Table 4. Comparison of results of this research with results of some countries

Country	Type of water	²²⁶ Ra (Bq/l)	²³² Th (Bq/l)	⁴⁰ K	Reference
Iran	Ground water	<MDA to 11.08	<MDA to 7.45	<MDA to 4.23	This research
	Surface water	<MDA	0.33	5.32±0.99	
	Hot spring Donbe	0.90±0.19	<MDA	5.72±1.20	
	Hot spring Shafa	1.62±0.24	<MDA	12.39±1.66	
	Hot spring soleimani	1.09±0.15	0.64±0.20	13.57±1.07	
Italy	Tap water	0.49	0.60	-----	3
Austria	Ground water	0.008- 1	-		22
Slovenia	Tap water	1.0±0.2	8.5±0.8		23
China	Ground water	Up to 0.77	0.02		24
Denmark	Wells	1.08	-		25
Tunisia	Springs	0.06-7.68	-		26
Egypt	Qena Ground water	1.75	0.00		27
	Sfaga Ground water	0.00	0.22		
Korea	Hot spring	0.13- 3.27	-		28
	Cold spring	0.11- 0.61	-		

The maximum ²³²Th radioactivity concentration was obtained for well No. 14 in Mobarak Abad region. Based on the present results, the surface water of Kamal-Saleh Dam contained low amounts of ²³²Th and ¹³⁷Cs. In fact, ¹³⁷Cs radionuclide was observed in some ground water and surface water samples. This radionuclide originated from nuclear accidents or nuclear weapon tests in other countries. In such cases, the radionuclide distributed in the environment with radioactive dust through the atmospheric process, and in some cases, it penetrated into the ground water.

The AMW07 and AMW14 ground water samples contained the highest ²²⁶Ra concentration; also, ²³²Th and ⁴⁰K were detected in the samples. According to the findings, these elements should be removed from drinking water resources. Additionally, the surface water from Kamal-Saleh Dam contained ²³²Th radioisotope chain. It is speculated that the river (with the same name), which originates from Zagros mountains, passes through rocks containing this isotope on its path to the dam.

Comparison of radionuclide specific activity concentrations of Arak ground and surface water with water resources in other countries showed higher levels of radiation in the consuming water of Arak city. This could be due to changes in rocks and soil structure in lower layers of rocks, eroded by the ground water entering from the surface.

Table 4 shows the comparison of present findings with the results reported in Italy, Austria, Brazil, Jordan, Egypt, Slovenia, and North Korea, revealing the higher levels of radioactivity in the drinking water of Arak, Iran. In hot springs of Mahallat, ²²⁶Ra and ²³²Th concentrations were similar to other countries, while ⁴⁰K radionuclide showed more specific activity, which might be attributed to feldspars in igneous rocks of this region.

4. Discussion

According to the results of the present study, Aman Abad region had higher levels of ²²⁶Ra, ²³²Th, and ⁴⁰K radiation. In this region, artificial radiocesium was observed in well

No. 4, indicating that this radionuclide could penetrate to the ground water in a depth of 120 meter. ^{137}Cs was also observed in the ground water of Mobarak Abad well No. 2 and Kamal-Saleh Dam surface water.

In the present study, 1900 l/s of the consumed water in Arak was evaluated, 1000 l/s of which was supplied by Kamal Saleh Dam and 900 l/s by water resources in the three evaluated regions. The average annual effective dose was calculated with the intake of individual radionuclide ^{226}Ra and ^{232}Th for mixed consuming water and obtained 0.2 mSv/y. The activity concentrations of radionuclides in this study in comparison with the findings in Italy, Austria, Brazil, Jordan, Egypt, and North Korea showed higher levels of radionuclides in the drinking water of Arak [3, 22-28].

Based on the available information, it is assumed that any radiation exposure imposes a degree of risk to the public. The United States Environmental Protection Agency (USEPA) has established the maximum contaminant level (MCL) for radium in public water supplies to be 0.185 Bq/l (5 picocuries per liter) [29].

Accordingly, the ground water in Arak mostly exceeded the mentioned criterion for drinking

water, which could be due to changes in cropping patterns, water, rocks, and soil structure (in lower layers of rocks) in this region. According to ICRP recommendations, an effective dose of 1 mSv/y is considered as the limit for public exposure [30]. Therefore, the doses measured in this study were significantly lower than the recommended value for all water types.

5. Conclusion

The calculated annual effective dose due to water consumption by Iranians was within the average annual global range. Therefore, based on the findings, radionuclide intake due to the consumption of drinking water had no consequences for public health in the evaluated regions. However, hot spring bathrooms should employ air conditioning devices.

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