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Measurement of naturally occurring radioactive materials concentration in Tehran's water using Gamma spectrometry

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HIGHLIGHTS

- Natural radioactivity of water resources in city of Tehran is measured.
- A precisely calibrated HPGe detector has been employed.
- Radioactivity concentration is higher at south part of Tehran.
- Activity is mostly due to Th-232 and K-40 but little from Ra-226.
- Results show that the whole activity is more than many countries but comparable with standards.

ABSTRACT

The concentration of naturally occurring radioactive materials (NORM) in surface water and irrigation wells is measured using gamma ray spectrometry by HPGe detector. Measurement was carried out for samples that were collected over seventeen points in Tehran city and its suburbs. The samples were prepared in compliance with the principles from irrigation wells of city. The specific radioactivity of Ra-226, Th-232 and K-40 were measured and the results from different locations covered a range with the minimum being below "minimum detectable activity" up to maximum of 4.04, and 6.85 and 4.7 Bq per liter of water, respectively. The accumulation of radioactive materials in the samples from southern areas of Tehran was more than that of central areas. Also, concentration of Ra-226 in all the samples was less than the Derived Release Limit of Canada and Environmental Protection Agency standard threshold.

KEYWORDS

Naturally radioactive materials
Gamma ray spectrometry
HPGe detector
Tehran's water

HISTORY

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1 Introduction

Naturally Occurring Radioactive Materials (NORMs) are substances that are found naturally in the environment and are radioactive. These materials are found in water, soil, plants, coal, oil, ore, geothermal waste, waste water and etc. in low and variable quantities (Bou-Rabee et al., 2009). These materials are usually related to U-238 and Th-232 decay chains and the daughter of these nuclides as well as K-40 radioisotope.

The average concentration of uranium and thorium in the earths crust was measured 4.2 and 12.5 ppm, respectively (IAEA, 1989). According to the expansion of industrial activities, the concentration of NORMs has increases proportionally. These materials are dispersed as a result of human activities or some industrial processes. In areas with high natural radiation, the probability of accumulation of uranium and thorium in surface and underground water is remarkable. While groundwater moves through the gaps that contain uranium and thorium chemical compounds, these materials are dissolved in water and will be transferred to the surface and water resources. Concentration of K-40 stems from water salinity. More salt in water leads to higher concentration of K-40. Therefore, in such cases water qualification tests will show radiation contamination. Uranium isotopes have high radiation toxicity. Due to the Ra-226 and Ra-228 radiation toxicity and their presence in water, it is necessary to control their concentration in the environment (Agbalagba and Onoja, 2011; UNSCEAR, 2000; Diab et al., 2008; Ahmed, 2004; Godoy and Godoy, 2006).

Tehran is located in Northern half of Iran near Alborz mountains. According to geology of the area, active volcanoes might release lava which can contain uranium progeny. In addition, radium and its daughters have more chance to dissolve in underground water and appear in sur-

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face. Damavand is highest point in Iran and once it was an active volcano. On one hand, there is possibility of existing uranium progeny around it and hence theoretically such radioisotopes appear in surface water. It is noteworthy that part of drinking water in Tehran is supplied from underground. Thus it justifiable to analyze water supplies for natural radioactivity.

2 Materials and Methods

2.1 Sampling

Since Tehran is located on foothills, generally slopping from north downward to the south, underground water tends to accumulate in southern part of this city. Hence, the selected sites for sampling were located in the central and the southern parts of Tehran including surface water resources and irrigation wells. Sampling procedures were conducted according to IAEA method (IAEA, 1989). As sample container, 1.5 liters polyethylene bottles, previously washed with distilled water and acid, were utilized. Before collecting the original samples, the containers were filled and emptied twice. To prevent the adhesion of radioactive material to the container wall, 3 mL hydrochloric acid was added to samples. Then samples were transferred to the Marinelli containers (800 cc) and leached for 30 days to reach radioactive equilibrium between the radium and its daughters.

2.2 Gamma ray spectroscopy system

The specific activity of the K-40 in the U-238 and Th-232 decay chains were calculated after measurements with the use of HPGe coaxial detector (ORTEC 10180 GMX) and multichannel analyzer (MCA) with 4096 channels. Experimental setup of detection system is shown in Fig. 1.

The Maestro software was used to analyze the collected gamma ray spectrum (Zare et al., 2015). It is common to report energy resolution of detectors in 1332.5 keV. Therefore, a Co-60 source was place 10 cm away from the detector window. After counting for 60 seconds, Full Width at Half Maximum (FWHM) was obtained and divided by energy (1332.5 keV). FWHM for this detection system is equal to 3.1 keV rendering energy resolution equal to 0.23%. The shield of detection system consisted of 5 cm of lead, 2 mm of iron, and 2 mm of copper for eliminating environmental radiation. Energy calibration was carried out using point sources of Cs-137 and Am-241.

For obtaining the efficiency calibration curve of the HPGe detector, liquid standard sample in the same geometry of water samples was used. Standard sample was a Liquid 800 cc sample consisted of Eu-152 with known activity (0.081 ± 0.0007 μCi) and emission gamma rays in a wide energy range which makes it suitable for efficiency calibration. For Eu-152 gamma lines and their energy and emission probabilities refer to Table 1.

Energy efficiency curve of detector was obtained using MATLAB software. Efficiency calibration curve is shown in Fig. 2. Statistical and systemic errors in efficiency calibration points amount to 4% which is not shown in Fig. 2. Before analyzing unknown samples, it is essential to find Minimum Detectable Activity (MDA) of the detection system. It is only affected by background radiation. MDA for working energies are shown it Table 2.

Table 2: MDA for corresponding energies of NORMs.

Energy (keV)	MDA (Bq/L)
295	1.3
351	0.87
661	0.43
911	2
964	0.74
1460	2.3

We calculated MDA using Eq. (1) (Zare et al., 2015)

$$\mathrm{MDA(Bq/L)} = \frac{2.71 + 4.66\sqrt{N}}{T\,\varepsilon\,p_\gamma\,V} \qquad (1)$$

where N is net count under the peak, T is time (in seconds), ε is the detector efficiency, p_γ is gamma emission probability and V is sample volume.

2.3 Analysis method of gamma ray spectrum

After energy and efficiency calibration, the samples were placed on the detector in a coaxial direction and the count-

Table 1: Properties of liquid standard sample for calculating efficiency.

Radioactive nuclei	Radioactivity (μCi)	Emission energy (keV)	Emission probability (%)	Efficiency (%)
Eu-152	0.081 ± 0.0007	121.78	28.58	2.2917
Eu-152	0.081 ± 0.0007	244.69	7.58	1.3881
Eu-152	0.081 ± 0.0007	344.27	26.5	1.0115
Eu-152	0.081 ± 0.0007	411.234	2.234	0.7966
Eu-152	0.081 ± 0.0007	778.9	12.94	0.4439
Eu-152	0.081 ± 0.0007	864.37	4.245	0.3874
Eu-152	0.081 ± 0.0007	964.07	14.6	0.3367
Eu-152	0.081 ± 0.0007	1112.069	13.64	0.3074
Eu-152	0.081 ± 0.0007	1408	21	0.2404

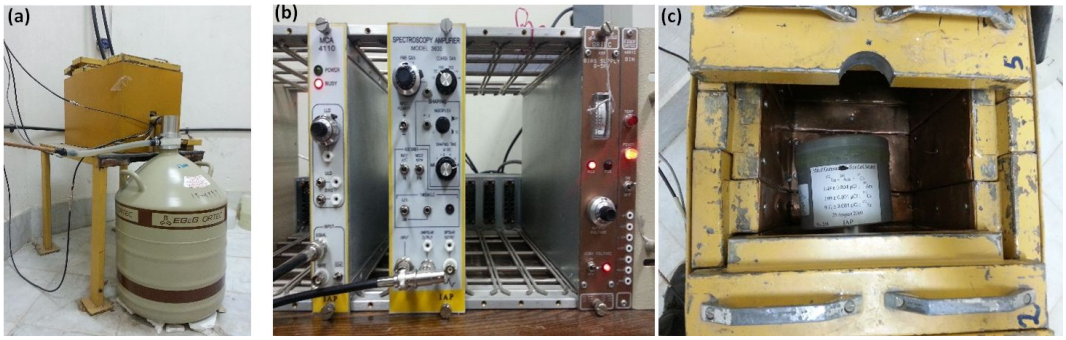

Figure 1: (a) Detector Dewar, (b) Electronic modules, and (c) Shield of samples.

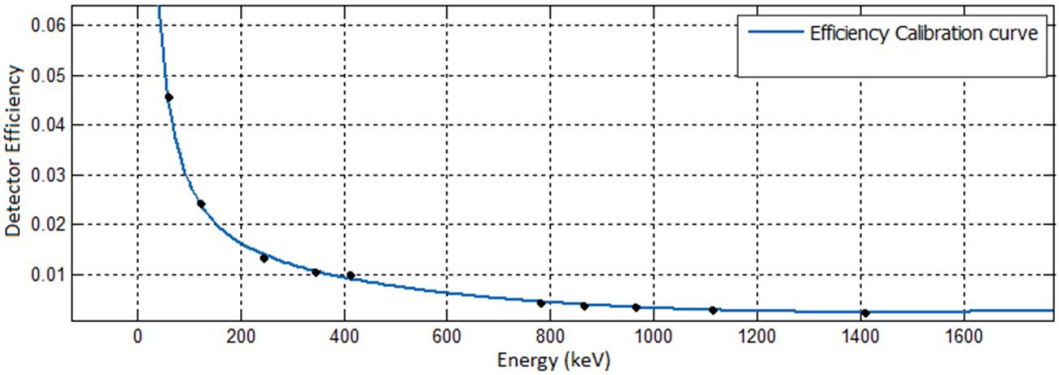

Figure 2: HPGe efficiency calibration curve.

ing time was set to 86400 seconds (24 hours). For correcting the net area under a full energy peak, the background was measured using blank sample (double distilled water) in Marinelli beaker (800 cc). Absolute radiation was calculated using Eq. (2) (Zare et al., 2015):

$$\mathrm{Act(Bq/L)} = \frac{C_{net}}{\varepsilon(\%)\,\lambda\,t\,V} \times 100 \qquad (2)$$

where C_{net} is the net count under the photopeak, ε is the full energy peak efficiency in the desired energy, λ is the probability of gamma emission at the photopeak energy, t is the live time of counting and V is the sample volume. Net peak area calculated using OMNOGAM software (Zare et al., 2015).

Pb-214 and Bi-214 gamma lines were selected to measure the concentration of Ra-226, and Ac-228 was selected for determining Th-232 concentration. The probability of each emitting gamma is given in Table 3.

3 Results and discussion

Natural radioactivity of Ra-226, Th-232 and K-40 were measured in the samples from underground water from 17 districts of Tehran. The measurements were performed using high resolution HPGe detector. Tables 4 and 5 show the results. The highest concentration of Ra-226 was observed in "Bazzar Gol" area southern region of Tehran. The concentration of Ra-226 in all samples was less than DRL (5 Bq/L) and EPA standard (1 Bq/L). Meanwhile, the highest concentration of Th-232 was in the area of "13$^{\mathrm{th}}$ Aban". The highest K-40 activity was measured in the sample of "Gharchak" south-east of Tehran as shown in Table 4. The results indicate that concentration of NORM is greater near the southern region of Tehran. In addition, the results show that radioactive accumulation depends on various factors and can be due to the different underground layers as well as to the chemical compounds existing in the soil of those areas, which causes the most

Table 3: Gamma lines used to measure the sample activity.

Radioactive nuclei	Emission energy (keV)	Emission probability (%)	Uncertainty (%)
K-40	1460.83	10.67	0.56
Pb-214	295.220	18.70	2.140
	351.930	35.80	1.960
Bi-214	609.320	45.00	0.890
	1120.28	14.90	1.340
Ac-228	338.400	11.51	20.07
	968.900	16.74	9.980

Table 4: Radioactive concentration of Ra-226, Th-232 and K-40 in 17 samples.

Sample code	Sampling location	Ra-226 (Bq/L)	Th-232 (Bq/L)	K-40 (Bq/L)
1	Gharchak	0.31	1.29	4.7
2	Eslam Shahr	2.02	MDA>	4.7
3	13th Aban	2.13	6.85	MDA>
4	Amir Abad-College's Dormitory	0.63	4.57	MDA>
5	Amir Abad-Technical College	0.317	1.29	MDA>
6	Institute of Geophysics	MDA>	1.48	MDA>
7	Fatemi Aghaghiya Park	MDA>	MDA>	MDA>
8	Ghezelghale Park	MDA>	MDA>	MDA>
9	Goftgoo Park	0.87	MDA>	MDA>
10	The End of Hashemi Highway	2.77	MDA>	MDA>
11	Raah Ahan Square	MDA>	MDA>	MDA>
12	Shahr Rey-Modarres Square	2.02	1.293	MDA>
13	Bazzar Gol	4.04	MDA>	4.6
14	Parek-e Shahr	MDA>	MDA>	MDA>
15	Zanjan Avenue-Recycling Center	2.02	2.58	MDA>
16	Shariati Hospital	MDA>	MDA>	MDA>
17	Razi Park	MDA>	MDA>	MDA>

Table 5: Radioactivity comparison of water samples in different parts of the world.

Country	Sample type	Ra-226 (Bq/L)	Th-232 (Bq/L)	K-40 (Bq/L)	Reference
Nigeria	Lake water	8.4 – 20	8 – 21	7 – 130	(Agbalagba and Onoja, 2011)
Egypt	Lake water	0.021	0.006	0.119	(Ahmed, 2004)
Brazil	Underground water	0.01 – 3.79	-	-	(Godoy and Godoy, 2006)
Saudi Arabia	Well water	1.28 – 3.61	-	-	(Aksoy et al., 2002)
Serbia	Drinkable water	0.26	-	-	(Todorovic et al., 2012)
Japan	Sea water	0.0014 – 0.0048	-	-	(Inoue et al., 2012)
Malaysia	Mineral water	0.0266	0.0645	1.28	(Yussuf et al., 2012)
Turkey	Reservoir water	0.003 – 0.045	-	0.009 – 0.29	(Cevik et al., 2005)
Pakistan	Drinkable water	0.011	0.005	0.14	(Fatima et al., 2006)
Iran (Oman Sea)	Sea water	2.19 – 2.82	1.66 – 2.17	132.6 – 148.87	(Zare et al., 2015)
Iran	Water	MDA>-4.04	MDA>-6.85	MDA>-6.85	This study

of these materials to be dissolved in water and move to the surface.

4 Conclusion

In this study, the radiation concentrations in 17 samples of Tehran underground water were measured. Activities of Ra-226, Th-232 and K-40 nuclides vary from MDA to 4.04, from below MDA to 6.85 and from below MDA to 4.7 Bq/L, respectively. The concentration of natural radioactive material in water, as reported by researchers from different countries, is presented in Table 5.

According to Table 5, radioactivity of Ra-226 and Th-232 in Tehran water is more than water samples of Egypt, Brazil, Saudi Arabia, Serbia, Japan, Turkey, Pakistan and southern shores of Iran upon Oman Sea. Concentration of K-40 in surface water samples is more than samples of Egypt, Malaysia, Turkey, and Pakistan and yet it is lower than Nigeria and Oman Sea (in Iran). In addition, concentration of NORMs in Tehran water is almost near the natural environmental activity.

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