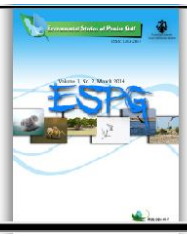
Volume 1, No.2, March 2014, Pages 74-80

Environmental Studies of Persian Gulf

journal home page: espg.pgu.ac.ir



Natural Radioactivity of Drinking Water Resources in Arak, Iran

Reza Pourimani*, Zahra Nemati

Department of Physics, Faculty of Science, Arak University, Arak 38156, Iran

ARTICLE INFO

Article History:
Received
April 14, 2013
Accepted
July 11, 2013

Keywords:
Natural radioactivity
Drinking water
Arak city
HPGe
^{226}Ra radionuclide

ABSTRACT

The activity concentrations of ^{235}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs in ground and surface of drinking water resources in Iranian city of Arak were determined by gamma ray spectrometry with a HPGe detector setup, coaxial p type and 8192 channels MCA. The water samples differed in radioactivity content depending on their origin and place. In the drinking water samples from Aman Abad, Mobarak Abad, and Taramazd wells, the mean values of the ^{226}Ra activity concentrations were 8.43±1.96, 1.56 ±1.04, 1.29±1.07 and 2.20±0.10 Bq/l and the mean values of ^{232}Th were 2.70± 0.18, 0.41±0.16 and 1.27±0.44 Bq/l, respectively. The maximum and minimum activity concentrations of ^{232}Th belong to Amman Abad area wells No. 7 and Mobarak Abad area wells No. 3, respectively. In Kamal Saleh water, the results of activity concentrations show 0.33 ± 0.14 and 0.08±0.03 Bq/l for ^{232}Th and ^{137}Cs, respectively. The other radionuclide activities were lower than the MDA. A mean annual effective dose for ^{232}Th and ^{226}Ra taken into the body by population drinking water for Kamal Saleh dam water and Aman Abad well No. 14 was calculated to be 0.01 ±0.00 and 1.84±0.17 mSv/year respectively, which is lower than the limit recommended by WHO.

1 INTRODUCTION

Water is very important in environmental studies due to daily use by human and also its potential infection (Malanca et al., 1998, Deglier and Karahan, 2010). The ground waters contain natural radionuclides ^{40}K and natural decay chains of ^{238}U, ^{235}U and ^{232}Th (Fort et al., 2007). The dissolution of the salts of uranium and thorium enters the rock and sediment layers in the soil and pollutes the ground water (Mokhtari, 2004). The average ^{238}U content in the Earth`s crust has been estimated to be 2.7 mg/kg and its concentration may be as high as 120 mg/kg in phosphate rocks. Meanwhile, the average $^{232}$Th contents of the Earth`s crust is about 9.6 mg/kg. The presence of these elements in the ground water depends on the three main factors geological, hydraulic and chemical properties of water (Azarvand, 2011). The Uranium isotopes (^{238}U, ^{234}U and ^{235}U) have non-negligible radio toxicity (WHO, 1978). The most radiotoxic and the most important among them are the Radium isotopes and the Radon gas. Among the radionuclides ^{226}Ra, ^{222}Rn from ^{238}U decay series and ^{228}Ra from ^{232}Th decay series have the greatest impact on human health (Piroozfar, 2007). Considering the high radio toxicity of radium isotopes, their presence in water and the associated health risks require particular attention. It is known that even small amounts of the radioactive substance may produce damaging biological

* *Corresponding author: r-pourimani@araku.ac.ir*

effects; moreover, ingested and inhaled radiation can be a serious health risk (Rowland et al., 1994). When Radium is taken into the body, its metabolic behavior is similar to Ca and an appreciable fraction is deposited in the bone, the remaining fraction being distributed almost uniformly in soft tissues. If radionuclides be taken into the body through drinking water, inhalation or absorption through the skin, their distribution in the body over time and consequently energy of the emitted radiation will create irreparable damages such as bone cancer, leukemia, genetic effects, damage to blood vessels, eye lenses cataract induced infertility (Wallner, 2008; Nollet, 2007). For this reason, recently there has been a surging interest in the study of radioactivity in drinking water in many countries. The Radium concentration in surface water ranges from 0.01 to 0.1 Bq/l normally (Iyengar, 1990), from which the highest values have been found in waters close to Uranium mining and milling sites (Paschoa et al., 1979; Benes, 1990). In the ground water, the Radium concentration can reach values up to 38 Bq/l, depending on factors such as type of aquifer rock and chemical and physical characteristics. The present study attempts to understand the occurrence and distribution of natural radio nuclides ^{235}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs in drinking water resources in Arak city.

2 MATERIALS AND METHODS

2.1 Sampling and sample preparation

Arak city has 25 active wells with 120 m depth that are used to supply drinking water. In this work, due to the proximity of the wells in an area about 500 meters away and the status of considering the topography of the area, 11 active wells were sampled. These include 5 samples from Amman Abad region, 3 samples from Mobarak Abad region, 3 samples from Taramazd region and one sample from Kamal Saleh dam. Samples were taken in 1.5 liter volume polyethylene containers from special valves that were built on wells for taking water samples.

Immediately after filling bottles, samples' pH decreased by adding nitric acid and was adjusted to 2 that is necessary for preventing radionuclide absorption by walls of containers (Perez-moreno et al., 2001). All samples were prepared in Marinelli Beaker containers for gamma spectroscopic analysis. The collection of water samples requires particular care because radon is a short-lived gaseous nuclide and tends to escape from water during sampling. In this study, Marinelli beaker with volume 800 cc was used and sealed. After the minimum 210 days for preparing samples, gamma ray was registered because this time is higher than that is necessary to take radioactive chain equilibrium (IAEA, 2003).The sampling locations are shown in Fig. 1.

Fig. 1. Location map of the area under consideration.

2.2 Experimental setup

This measurement has been done using gamma ray spectrometer HPGe coaxial detector p type with 80% relative efficiency setup and multichannel analyzer of 8192 channels. The energy resolution of the detector is 1.85 keV for 1332.52 keV energy gamma ray due ^{60}Co. In this measurement, the operating voltage was 3000 V and the Marinelli standard containers were used. The detector was shielded in a chamber of two layers 10 cm thick lead and 3mm thick by copper. The soft components of cosmic ray, consisting of photons and electrons, reduce to a very low level by 100 mm of lead

shielding. The X-ray (73.9 keV) emitted from lead by its interaction with external radiation is suppressed by the copper layer (Aziz, 1981). In order 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 86400 s. The Marinelli container has 800 cc volume with an outside diameter of 14 cm and 11 cm height and the internal diameter of 10 cm and the 7.5 cm of internal height. The system was calibrated for the energy and the efficiency. The energy calibration was carried out by using radioactive standards source. Regarding the efficiency calibration, we used the Marinelli Beaker standard source including radioisotopes ^{241}Am, ^{152}Eu, and ^{137}Cs with exact activities. According to the registered gamma ray spectrum, the absolute efficiency of detector configuration was calculated by Eq. (1):

$$\varepsilon(\%) = \frac{Netarea}{Act \times (DN\%_i) \times t} \times 100 \qquad (1)$$

Where Netarea is the net counts under the full-energy peak corresponding to the energy of E_i. Act denotes the radioisotope activity. $P_n(E_i)$ shows the photon emission probability, and T being the counting time. One can find the plot of efficiency versus gamma ray energy in Fig. 2.

The function fitted to experimental data by polynomial curve is y= a +b Lnx + c/lnx+d(lnx)^2+e(lnx)^2+f(lnx)^3+g/(lnx)^3+h(lnx)^4. Here, y denotes the efficiency, a, b, c, d, e, g and h are constants (see Fig. 3). x (keV) is the energy of gamma ray. The ^{226}Ra activities of the samples were determined through the intensity of 351.9 keV and 609.3 keV gamma lines of ^{214}Pb and ^{214}Bi, respectively. The ^{232}Th activity was obtained using gamma line of ^{228}Acat 911.21 keV with an intensity 26.6% and 968.97 keV with an emission percentage of 17.4%. In order to determine ^{235}U activity concentration, gamma ray line 143.78 and 205.03 keV and 185.715 keV was used after separation from 186 keV of ^{226}Ra. ^{40}K and ^{137}Cs were obtained using gamma ray line 1460.7 and 661.6 keV, respectively. All 12 registered gamma ray spectra were analyzed and activity concentrations were calculated with Gamma Vision Ortec Software. In all of the analyzed spectra, the corrections were done for background gamma ray that was registered using empty Marinelli container in same condition.

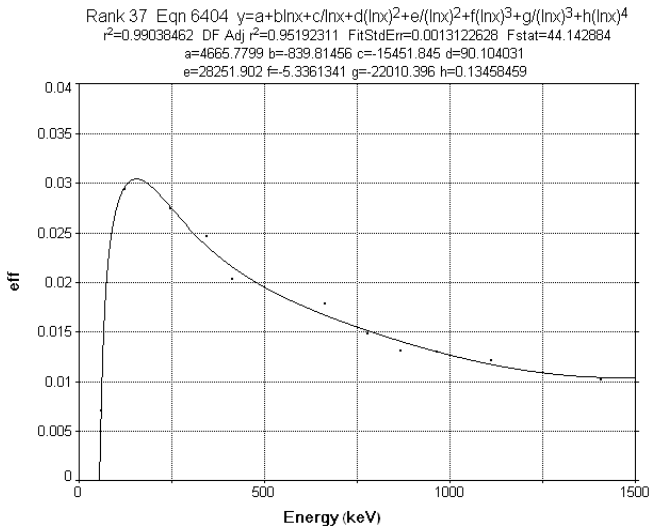


Fig. 2. Detector efficiency curve of HPGe with 80% relative efficiency for the standard Marinelli Beaker source.

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

Sample code		^{235}U	^{232}Th	^{226}Ra	^{40}K	Cs^{137}
	AMW01	< MDA	1.78±0.20	1.26±1.03	1.77±0.68	< MDA
	AMW04	< MDA	< MDA	< MDA	< MDA	0.06±0.02
	AMW07	< MDA	1.42±0.19	11.08±0.80	1.26±0.69	< MDA
	AMW11	4.13±0.92	1.61±0.16	< MDA	4.23±0.65	< MDA
	AMW14	< MDA	7.45±0.23	10.62±1.00	2.78±0.67	< MDA
Ground water	MBW01	< MDA	< MDA	1.16±1.02	< MDA	< MDA
	MBW02	< MDA	< MDA	1.96±0.96	< MDA	0.07±0.03
	MBW03	< MDA	0.41±0.16	< MDA	< MDA	< MDA
	TRW01	< MDA	1.55±0.20	1.75±1.07	2.34±0.68	< MDA
	TRW02	0.64±0.05	1.64±0.20	< MDA	1.82±0.66	< MDA
	TRW03	< MDA	0.42±0.11	1.15±0.89	< MDA	< MDA
Surface water	KSDS01	< MDA	0.33±0.14	< MDA	< MDA	0.08±0.03

In order to calculate the activity concentration, the following relation was used:

$$Act = \frac{\text{Net Area}}{\varepsilon(\text{B.R})tv} \times 100 \qquad (2)$$

where Net Area is the net count under peak, Act (Bq/l) is the activity concentration, ε is the energy efficiency for gamma ray by detector, B.R. is the branching ratio of gamma intensity (%), t (s) is the time of spectra, and v (liter) is the volume of sample.

The annual effective dose was calculated with the intake of individual radionuclides and ingestion dose coefficients (SvBq−1) reported by the International Commission on Radiological Protection (ICRP, 1991). The equation for calculating the annual effective dose per person is:

$$\text{Annual effective dose} = \sum_i I_i \times 365 \times D_i \qquad (3)$$

where I_i is the daily intakes of radionuclide i (Bqd^{-1}), and D_i is the ingestion dose coefficient ($Sv\ Bq^{-1}$). Considering the intake volume of drinking ground water by adult males in Arak to be 1 l/d (WHO, 1978).

3 RESULTS AND DISCUSSION

Table 1 shows the activity concentrations of ^{235}U, ^{232}Th, ^{226}R, ^{40}K and ^{137}Cs for sample codes AMW01, AMW04, AMW07, AMW11 and AMW14 from wells No. 1, 4, 7, 11 and 14 of Amman Abad region, MBW01,MBW02 and MBW03 from wells No. 1, 2 and 3 of Mobarak Abad region, TRW01, TRW02 and TRW03 from wells No. 1, 2, 3 of Taramazd region and KSDS01 is from Kamal Saleh Dam surface water. Table 2 shows the average activity concentrations for three regions of ground water and top water of Kamal Saleh dam not considering values lower than the MDA. Table 3 shows the measured annual effective doses for different regions in Arak.

This measurement shows that the ^{226}R radioactivity concentration in Aman Abad ground water is higher than another regions and the minimum amount belongs to Taramazd wells.

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

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

Table 3
Measured annual effective doses for different regions in Arak.

Radionuclide	Region	Intake, Bq d-1	Ingestion dose Coefficient (Di), Sv Bq-1 ICRP 68 (1994)	Annual effective dose, mSvy-1
^{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	-	-	-
^{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

The maximum value of the ^{232}Th radioactivity concentration was obtained for well No. 14 in Mobarak Abad region. This study shows that the top water of Kamal Saleh dam contains ^{232}Th and ^{137}Cs in low amounts.

The ground water samples AMW07 and AMW14 contain the highest radioactivity concentration for ^{226}Ra. It also contains ^{232}Th and ^{40}K.

The present study suggests that these should be exited from drinking water resources. The top water of Kamal Saleh dam contains ^{232}Th radioisotope chain, which indicates that the river- with the same name- originates from Zagros mountain chains in its road to dam passes from enrich of this isotope mine or rocks. Comparing radio nuclide specific activity concentration of ground and top water in drinking water of Arak with the results of other countries shows higher levels of radiation in consuming water of this city. This could be due to changes in rocks and soil structure in lower layers of rocks eroded by water entering to top and ground water.

Table 4 shows our results of radionuclide activities in Arak water resources in comparison with the data measured in Italy, Austria, Brazil, Jordan, Egypt and North Korea, which reveals the higher levels of radioactivity of drinking water in Arak.

4 CONCLUSION

We have measured the activity concentrations of radionuclides in some regions in Arak city of Iran.

Table 4
Measured specific activities of radionuclides in Arak water resources and their comparison with those of some countries.

City		Type of water	^{235}U (Bq/l)	^{232}Th (Bq/l)	^{226}Ra (Bq/l)	Reference
Arak		Ground water	Up to 4.13	Up to 7.45	Up to 11.08	
		Surface water	-	0.33	-	
Italy		Tap water	0.02	0.00	0.00	Fort et al., 2007
Austria		Ground water	0.00	-	0.05	Wallner, 2008
China		Ground water	-	0.02	Up to 0.77	Ziqiang et al., 1988
Denmark		Wells	-	-	1.08	Ulbak and Klinder, 1984
Tunisia		Springs	-	-	0.06-7.68	Labidi et al., 2002
Egypt	Qena	Ground water	-	0.00	1.75	Nourkhalifa, 2004
	Sfaga	Ground water	-	0.22	0.00	
Korea		Hot spring	0.37	-	-	Lee, 2001
		Cold spring	0.14	-	-	

Aman Abad zone has higher levels of radiation containing ^{235}U, ^{232}Th, ^{226}Ra and ^{40}K. In Aman Abad well No. 4 artificial radiocesium has also been observed that indicates ^{137}Cs radionuclide has penetrated to the ground water in 120 m depth. The ^{137}Cs radionuclide has also been observed in ground water of Mobarak Abad well No. 2 and Kamal Saleh dam top water.

Consuming water of Arak has been evaluated to be about 1900 liter per second that 1000 ls^{-1} supplied from Kamal Saleh dam and 900 ls^{-1} from the three regions under consideration. The average annual effective dose has been calculated with the intake of the individual radionuclides ^{226}Ra and ^{232}Th for mixed consuming water as 0.2 mSv/year.

Our results of radionuclides activity concentrations have been compared with measured data in Italy, Austria, Brazil, Jordan, Egypt and North Korea are of higher levels in drinking water of Arak. This could be due to changes in cropping patterns, water, rocks and soil structure probably in lower layers of rocks and soil in this region. According to ICRP recommendations (ICRP60, 1991), the limit for public exposure should be expressed as an effective dose of 1 mSv/year. The doses obtained in this work are significantly below that recommended for all categories of water.

REFERENCES

Azarvand, B. (2011). Natural radioactivity in water and its environmental impact. Fourth Conference on Environmental Engineering.

Aziz, A. (1981). "Methods of Low-Level Counting and Spectrometry Symposium", Berlin, 221.

Benes, P. (1990). Radium in (continental) surface water. The Environmental Behavior of Radium, Technical Reports Series No. 310, International Atomic Energy Agency, Vienna, 373-418.

Deglier, M., Karahan, G. (2010). Natural radioactivity in various surface water in Adana, Turkey. Desalination 261, 126-130.

Fort, M., Rusconi, R., Cazzaniga, M. T., Sgorbati, G. (2007). The measurement of radioactivity in Italian drinking waters. Micro. Chem. J. 85, 98-102.

Gruber, V., Maringer, F. J., Landstetter, C. (2009). Radon and other natural radionuclides in drinking water in Austria. Appl. Radiat. Isot.67, 913-917.

ICRP Publication 20. (1993). Retention equation. Health Phys. 65, 507-513.

ICRP60. (1991). Recommendations of the International Commission on Radiological Protection. Pergamon Press, Oxford, ICRP Publication 60.

International Atomic Energy Agency. (2003). Collection and preparation of bottom sediment samples for analysis of radio nuclides and trace elements. IAEA-TECDOC-360, IAEA, Vienna.

Iyengar, M. A. R. (1990). The natural distribution of Radium and the environmental behavior of Radium. Technical Reports Series No. 310, IAEA, 1, 9-128.

Labidi, S., Dochraoui, M., Mahjoubi, H., Lemaitre, N., Ben Salah, R., and Mtimet, S. (2002). Natural radioactive nuclides in some Tunisian Thermo-Mineral Springs. J. Environ. Radio.62, 87-96.

Lee, M. H., Choi, G. S., Cho, Y. H., Lee, C. W. and Shin, H. S. (2001). Concentration and activity ratios of uranium isotopes in the ground water of the Okchun Belt in Korea. J. Environ. Act. 57, 105-116.

Malanca, G. A., Repetti, M., Macedo, H. R. (1998). Gross alpha and beta activates in surface and ground water of Rio grand do Norte. Brazil. Appl. Radiat. Isot. 499, 893-898.

Mokhtari, M. (2004). Mechanisms of elimination of natural radioactivity of drinking water. Sixth National Conference on Health.

Nollet, Leo M. L. (2007). Handbook of water analysis. CRC Press, 77-80.

Nourkhalifa, A. Natural radioactivity of ground water and drinking water in some Areas of upper Egypt. Torkish J. Eng. Env. Sci. 28, 345-354, 2004.

Paschoa, A. S., Baptista, G. B., Montenegro E. C., Miranda, A. C. and Sigaud, G. M. (1979). Radium-226 concentrations in the hydrographic basins near Uranium mining and milling in Brazil. Proceedings of the Mid Year Topical Symposium, Low Level Radioactive Waste Management, Williamsburg, VA, 337-343.

Perez-moreno, Bolivar, J. P., Gaicia-Tenerio, San Miguel, E., Agudao, J. L., Mas, J. L. and Vaca, F. (2001). A simple method for efficiency calibration of HPGe in γ-spectrometric measurements. Radiat. Phys. Chem. 61, 437-438.

Piroozfar, P. (2007). Study of natural radionuclides of water and sediment in Tehran 1:100000 map. Twenty-fifth assembly of Geological Sciences Geological.

Rowland, R. E., Wrenn, M. E, Durbin, P. W., Howard, B., Lipsztein, J., Rundo, J., Still, E. T. and Willis, D. I. (1994). Low-level Radium retention by the human body a modification of the metabolism of ingested U and Ra. Health Phys.48, 601-633.

Ulbak, K. and Klinder, O. (1984). Radium and Radon in Danish drinking water. Radiat.Prot. Dosim.7, 87-89.

Wallner, G. (2008). Natural radionuclides in Austrian mineral water and the sequential measurement by fast methods. J. Environ. Radio.99, 1090-1094.

WHO. (1978). Radiological examination of drinking water. World Health Organization, Copenhagen.

Ziqiang, P., Yang, Y. and Mingqiang, G. (1988). Natural radiation and radioactivity in China. Radiat. Prot. Dosim. 24, 29-38.