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Radium Isotopes Levels in Drinking-Water Samples

Abstract- Water is one of the most necessary substances on earth. In order to survive, all plants, animals and humans must drink water, and there is no life on earth if there is no water. Hence, water should be uncontaminated with any type of carcinogenic materials such as heavy metals, radioactivity or other pollutants and must be clear. In this work, drinking-water samples were examined and analyzed for radionuclide levels using NaI gamma spectrometer, as well as, estimated annual effective dose (AED) has been calculated. The values of ^{226}Ra , ^{228}Ra and ^{40}K levels range from $(0.12\text{-}0.35)\text{Bq.L}^{-1}$, $(0.09\text{-}0.16)\text{Bq.L}^{-1}$ and $(31.66\text{-}49.25)\text{Bq.L}^{-1}$, with mean values 0.29, 0.12 and 43.39 Bq.L^{-1} , respectively. The total estimated annual effective dose gained from the combined ingestion of radium isotopes is found to start from 0.07 mSv.y^{-1} to 0.14 mSv.y^{-1} with an average value of 0.12 mSv.y^{-1} . Therefore, the total (AED) for all samples is below the world standard value of 1mSv.y^{-1} . Hence, drinking water is considered safe from the radiological point view and does not cause any significant health hazard for Baghdadi inhabitants.

Keywords- annual effective dose, drinking water, gamma spectroscopy, radioactivity.

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1. Introduction

Water is necessary for public life and all living things and nothing substitute water, and it is largely consumed. Hence, quality of water is an important factor of environmental studies and determination of radioactivity content in drinking water can be considered one of the main topics in both normal and emergency cases as well as it permits the estimation of radioactivity dose received by public due to everyday consumption of water [1].

The dose acquired from drinking water is, in general, a small fraction of total dose received from naturally occurring and artificial radioactive materials [2]. Radioactive isotopes in the environment come from naturally occurring and manufactured sources. Uranium, thorium, radium isotopes as the members of the uranium and thorium series as well as potassium-40 and others occur naturally everywhere in the environment which is certainly considered as the largest proportion of human exposure sources and they may cause a health risk for inhabitants. According to the drinking-water standards, the concentration of ^{226}Ra is limited [3]. Thus, the assessment of the radioactivity concentrations of radium isotopes in drinking-water samples is very significant from the radiological viewpoint and environmental protection. The two isotopes of radium (^{226}Ra & ^{228}Ra) are very considerable from a radiation protection point of view due to their comparatively long half-lives, naturally occurring elements, and high dose

conversion factors [4]. In addition, due to radium, isotopes have identical properties to calcium (group II), they can easily built-in into bones and generate short-lived radionuclides of high activity [5]. Several radioactive materials can be emitted into the environmental species, and then into drinking-water supplies. Despite of all, water consists of several amounts of radioactive materials, the amount and type of radiation depends on many parameters [6], such as concentrations of Naturally Occurring Radioactive Materials (NORM) and Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in soil and rock, in particular ^{226}Ra and ^{228}Ra , discharge radionuclides from nuclear fuel cycle facilities, and discharge of unsealed manufactured radionuclides, which might enter drinking-water supplies, in case of improper medical or industrial use .

Alpha particle emitters are more risky when enter the body by inhalation or ingestion, because they can expose human organs and tissues to radiation, causing biological damage that increases the risk of cancer. The estimated world average annual human exposure from natural sources is 2.4mSv/year [7]. Commonly, if the combined effective dose of both radionuclides ^{226}Ra and ^{228}Ra activities due to drinking-water consumption is between 0.04 and 0.1 mSv.y^{-1} , typical range stated by World Health Organization [3], or less than 1 mSv.y^{-1} to the population for long time exposure as recommended by ICRP [8], then there is no risky health effects are expected. River Tigris

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is the biggest river in Iraq and it is major source of drinking water for Baghdadi inhabitants [9]. A pollution in Tigris river cause a pollution to Euphrates River and any related water resources [10]. In this paper, drinking-water samples collected from al-Rasheed water treatment plant, located in Al-karkh side of Baghdad city, are tested to check whether the level of radioactivity in them meet world standards for radiological point of view and could not cause any significant health risk to the population. Furthermore, the results can be considered as beneficial information for Iraqi database.

2. Materials and Methods

I. Sample preparation

Drinking-water samples were collected from Al-Rasheed water treatment plant situated in Baghdad for a period of 6 months (October, November, and December 2016, January, February and March 2017). The samples were stored in clean polyethylene bottle. Then, in order to determine radium isotopes concentrations the samples were left to evaporate from a volume of 1600 ml to 800 ml. after that water samples stored, capped and kept in sealed Merinelli beakers. Then water samples were left for 28 days before counting to achieve secular equilibrium of uranium and Thorium with their decay products.

II. Gamma spectroscopy

Gamma spectrometric system is a useful non-damaging method that allows the concurrent evaluation of several radioactive nuclei in a sample [10-12], furthermore it doesn't need a sophisticated and time exhaustion radiochemical separations as for alpha spectrometry. The energy spectrum of the emitted gamma rays was measured using NaI gamma spectrometric system. The γ -spectroscopy system consists of NaI scintillation detector from ORTEC Company with an efficiency of 20%. Detector model number is 905-4 with (3"x3") inches crystal. 3-inche tube with digit BASE 14-Pin Photomultiplier Tube (PMT) Base that is connected to A35-B32 ScintiVision-32 Gamma-Ray Analysis with NaI. The detector was covered by a cylindrical shield made of 5 cm thickness of lead, which limits the background gamma radiation existing at the laboratory.

III. Instrumentation and Analysis

The concentrations of the natural radioisotopes, such as ^{226}Ra , ^{228}Ra and ^{40}K in water samples, were analyzed by γ -spectroscopy. The sample was placed into the shielded NaI detector and the concentration was counted for 18000 seconds (5 hours). Background of the environmental gamma

at the laboratory was determined using an empty Merinelli beaker under same measurement conditions. Concentrations of radium isotopes in drinking water can be determined from gamma ray photo peaks emitted by their decay products. Concentration of ^{228}Ra isotope was determined from the concentrations of its progenies ^{212}Pb (transition line 239 keV) and ^{208}Tl (transition line 583keV) gamma rays, while, concentration of ^{226}Ra isotope was determined from concentrations of its decay products ^{214}Bi (transition line 609 keV) and ^{214}Pb (transition lines 295 and 351 keV).

Equation 1 is used to calculate radionuclide concentration C_R [13]:

$$C_R = \frac{N_{E_y}}{\epsilon_{E_y} \cdot I_y \cdot V \cdot T} \quad (1)$$

Where, N_{E_y} is the net peak area of the radionuclide of interest, ϵ_{E_y} is the detector efficiency for the energy E_y , I_y is the intensity/decay for the energy E_y , V is the water sample volume and T is the total counting time in second (18000 s).

Evaluation of annual effective dose (AED) in drinking water samples were evaluated using equation 2 [8]:

$$\text{AED} = \sum_i I_i \cdot y \cdot D_i = \sum_i D_{w_i} \cdot C_R \cdot 365 \cdot D_i \quad (2)$$

Where, I_i is the daily intakes of radionuclide I (Bq.d^{-1}) = $D_{w_i} \cdot C_R$, D_{w_i} is daily consumed of radioisotope (2L.d^{-1}) and the ingestion dose coefficients D_i for ^{40}K , ^{226}Ra and ^{228}Ra for adults are given in Table 1 [8].

2. Results and Discussion

The specific activity in Bq.L^{-1} for ^{226}Ra and ^{228}Ra from their progenies and ^{40}K are shown in Figures (1&2). Mean values of the three radionuclides is also shown in Figure 1. The mean value of ^{226}Ra concentration is 0.55 Bq.L^{-1} and ranges from 0.24 to 0.70 Bq.L^{-1} . The results show that the mean concentration level of ^{228}Ra is 0.12 Bq.L^{-1} and ranges from 0.09 to 0.13 Bq.L^{-1} . The recommended level for ^{226}Ra and ^{228}Ra concentration is 5 pCi.L^{-1} (0.185 Bq.L^{-1}) [14]. Hence, the values of ^{226}Ra activity concentration are higher than recommended USEAP value, while the values of ^{228}Ra activity concentration are lower than it. As from Figure 1, the variation of radium isotopes levels during winter is almost very low and can be considered as constant. The obtained values of ^{40}K activity ranged from the minimum value of 3.51 Bq.L^{-1} to the maximum value of 6.25 Bq.L^{-1} with mean value of 4.82 Bq.L^{-1} . The fluctuations in potassium-40 levels are more obvious during the time of collecting samples.

Potassium is an important constituent in our bodies and enters the human body essentially from

ingested food. It does not pile up in the body but is preserved at a same amount independent of intake, so human body can get rid of any extra amount of this element [15], because of this ^{40}K may has no harm on the body and its effect on the body content or on the radiation dose received is small [16]. Therefore, ^{40}K is excluded in the determination of committed effective dose received by the ingestion of the sampled water. Daily intake per person, annual effective dose for ^{226}Ra , ^{228}Ra and total annual effective dose for adults (age >17) are calculated and given in Table 2. The results are calculated by assuming that 2 liter/day of drinking water is consumed per person. The computed value of AED for ingestion of radionuclides ^{226}Ra and ^{228}Ra by adults is estimated to range from 0.03 mSv.y^{-1} to 0.07 mSv.y^{-1} and from 0.05 mSv.y^{-1} to 0.08 mSv.y^{-1} , respectively. The mean values for both isotopes are 0.06 mSv.y^{-1} . The total annual effective dose is found to range from 0.07 mSv.y^{-1} to 0.14 mSv.y^{-1} with mean value of 0.12 mSv.y^{-1} . The above results show that the contributions of both radium isotopes (^{226}Ra and ^{228}Ra) activities to the total annual effective dose due to consumption of the drinking-water are less than the accepted value of 1 mSv.y^{-1} and higher than 0.1 mSv.y^{-1} as recommended by ICRP [15] and WHO's [17] for drinking-water to the inhabitants for long time exposure, respectively. Therefore, in this study the drinking water samples for Baghdad city did not exhibit considerable levels of radium activity.

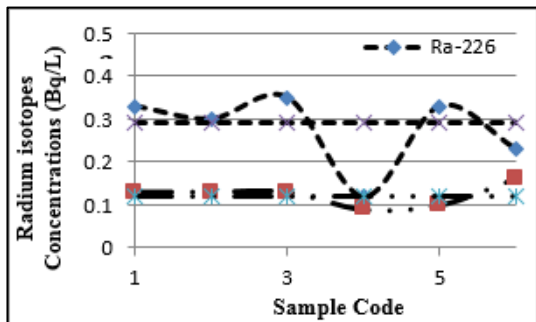


Figure1: Activity concentrations and mean values of ^{226}Ra and ^{228}Ra

Table 1: Specific dose coefficient of ^{226}Ra , ^{228}Ra and ^{40}K radioisotopes in drinking water to adults' > 17 years

Age group	Specific dose coefficient, Sv.Bq ⁻¹		
	^{226}Ra	^{228}Ra	^{40}K
Adults > 17years	2.8×10^{-07}	6.9×10^{-07}	6.2×10^{-09}

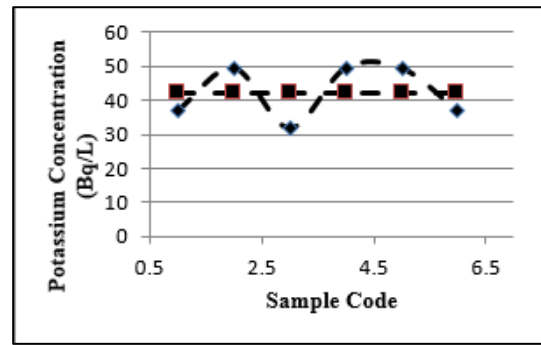


Figure 2: Activity concentrations and mean value of ^{40}K

Table 2 Assumed daily intake, AED and total AED of ^{226}Ra and ^{228}Ra

Month	Assumed daily intake		Annual Effective dose		Total AED
	^{226}Ra Bq/d	^{228}Ra Bq/d	^{226}Ra mSv/y	^{228}Ra mSv/y	mSv/y
Oct.	0.66	0.13	0.07	0.07	0.13
Nov.	0.60	0.13	0.06	0.07	0.13
Dec.	0.70	0.13	0.07	0.07	0.14
Jan.	0.24	0.09	0.03	0.05	0.07
Feb.	0.66	0.10	0.07	0.05	0.12
Mar.	0.46	0.16	0.05	0.08	0.13
Mean	0.55	0.12	0.06	0.06	0.12
Range	0.24-0.70	0.09-0.13	0.03-0.07	0.05-0.08	0.07-0.14

3. Conclusions

In the present work, determination of radioactivity concentration in drinking-water samples collected from Al-Rasheed water treatment plant in Baghdad for a period of six months started from October 2016 to March 2017 has been analyzed using γ -ray spectrometer. The results reveal that:

- 1) The concentration of radium isotopes (^{226}Ra & ^{228}Ra) are in very low proportions.
- 2) The values of the evaluated activity concentration range from 0.24 to 0.70 Bq.L^{-1} , 0.09 to 0.13 Bq.L^{-1} and 31.66 to 49.25 Bq.L^{-1} for ^{226}Ra , ^{228}Ra and ^{40}K , respectively.
- 3) The mean value of the activity concentration for ^{226}Ra is 0.29 Bq.L^{-1} , which is higher than USEPA recommended value.
- 4) The mean value of the activity concentration for ^{228}Ra is 0.12 Bq.L^{-1} , which is lower than USEPA recommended value.
- 5) The mean value of the activity concentration for ^{40}K is 43.39 Bq.L^{-1} .
- 6) Drinking-water standard recommended by USEPA for ^{226}Ra and ^{228}Ra is 5 pCi.L^{-1} , hence the values of ^{226}Ra activity is higher than recommended USEPA value, while ^{228}Ra activity is lower than recommended USEPA value.
- 7) The calculated AED received by Baghdadi inhabitants was found to range from 0.03 to 0.07 mSv.y^{-1} .

¹ and 0.05 to 0.08 mSv.y⁻¹ with mean value of 0.06 Bq.L⁻¹ as a result of ²²⁶Ra and ²²⁸Ra ingestion, respectively.

8) Hence, because of combined ingestion of both radium isotopes, the total effective dose are found to range from 0.07 to 0.14 mSv.y⁻¹ with mean value of 0.12 mSv.y⁻¹.

9) The received annual effective dose from drinking-water consumption due to contributions of both radium isotopes activities are lower than the recommended levels stated by ICRP and WHO for drinking water.

10) There is no spectacular variation in the radium concentrations during the period of collection of the samples is observed.

11) Therefore, this work reveals that inhabitants may not tolerate considerable exposure dose due to consumption of drinking water, subsequently it is safe from radiological point of view.

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