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RESEARCH ARTICLE

Natural Radioactivity in different water samples inAl-Bayda Region Northeast of Libya

E. Salama[1,2*], S. U. El-Kameesy[1], S.A. El-Fiki[1], N. El-Faramawy[1] and Areej Hazawi[3]

1. Physics Department, Faculty of Science, Ain Shams University, Cairo, Egypt;
2. Basic Science Department, Faculty of Engineering, British University in Egypt (BUE);
3. Physics Department, Omar Elmokhtar University, Libya.

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Corresponding Author

E. Salama

Abstract

Gamma activities of 12 different water samples collected from sea, well and tap watersinAl-Baydaregion, Libyawere determined by using HPGe detector. All the observed radionuclides are belonging to the naturally occurring ^{238}U (^{226}Ra) and ^{232}Th (^{228}Ra) radioactive series in addition to ^{40}K nuclide. The obtained resultsindicated that the natural radioactivity concentrationsin both well water and tap water did not exceed the WHO recommendations. The calculation of the annual effective dose equivalent of ingestion of these waters was carried out using dose conversion factor suggested by the USA-EPA. The average annual effective dose for all the water samples are very safe and below the international recommended limits.These obtained results are essential in establishing a reference baseline map for environment radioactivity levels in the northeast of Libya.

INTRODUCTION

Measurements of natural radioactivity in ground, surface and domestic water have been performed in many parts of the world, mostly for the assessmentof the dose and risk resulting from consuming water(Gabriele et al., 2008; Al-Jaser et al., 2005; Bieise et al., 2003; Labidi et al., 2002; Jose Marcus et al., 2001; Joselene de Oliveira et al., 2001).These efforts will help in the development of guidelines for the protection of human beings.

Lately, after Chernobyl and Fukushimaaccident, considerable attention have been given to the low-level exposure arising from naturally occurring radioactive nuclides. These radionuclides highly contribute to the annual accumulated radiological doses (Korkmaz and Camgoz, 2014; Turhan et al., 2013; Sajedah et al., 2012; Forte et al., 2007; Gonzalez et al., 2001; Martin Sanchez et al., 1995, Dickson and Herczeg, 1992; Day and King, 1983).Radium and its daughter products are important contributors to environmental radiation exposure(UNSCEAR 2008).Radium has two natural isotopes which deserves concern in public water supplies. ^{226}Ra is an alpha-emitter with a half-life of 1622y generated through the decay of ^{238}U,while^{228}Ra is ashorter-lived beta-emitter (half-life 5.7y), which is directly generated by ^{232}Th decay.

Radium entersgroundwater by dissolution of aquifer solids, direct recoil across liquid-solid boundary during its formation, radioactive decay of its parent in the solid form and desorption. Alpha-particle emitters are of special concern because of their high linear energy transfer. Whenhuman ingests radium about 20% is absorbed into the blood stream. Absorbed radium is initially distributed to soft tissues and bone, but its retention is mainly in growing bone(ICRP, 1993; EPA, 1991).

Groundwater is the basic source for water supply that represents 88% of the water needs in Libya (El-Kameesy et al., 2008).Only limited investigations have been carried out in the northeast region of Libya to trace the source and nature of minerals causing enhanced levels of natural radiation in different environmental samples(Omar, 1997; El-Trriki, 2006).

The present work aims to evaluate the activity concentration and distributions of natural radionuclides ^{226}Ra, ^{228}Ra and ^{40}K in seawater, well-water and tapwaterin the northeast region of Libya. Additionally, the aim in extended toverify the possibility of exploiting the purity of well waterand/or tap water to produce some sorts of clean bottled water as a pure public drinking water.

Thephysico-chemical analysis was carried out for water samples to determine water contents and characteristics such as electrical conductivity (EC) and total dissolved salts (T.D.S) in addition to the PH value. The obtained results are essential in establishing a reference baseline map for environment radioactivity levels in the northeast of Libya.

2. Materials and Methods

2.1 Sample collection and experimental setup

This study was conducted in four locationsin Al-Bayda region, Libya (Fig. 1). These locations are: Al-Hanea, Al-Hamama, Susa and Ras Al-Helal.12 different water samples from sea, well and tap waterswerecollected.Seawater samples were collected from different locations at distancesranged from 10-15m from the shore of the Mediterranean Sea. Well water samples were collected from wells with different depths ranged from 10 to 36 m by means of a subsurface electric pump.Tapwater samples were collected directly from the tap after letting the tap wide open to about ten minute.

All the samples were collected in 20000-ml capacity linear polypropylene bottles.All the water samples were acidified with 20M of HClat the rate of 20 ml per liter of the sample as soon as possible after sampling to prevent absorption of the radionuclides in the bottles.

In order to analyze the water samples, twenty liters of it were evaporated to one liter, and put into 1000 mlMarinellibeakers. These Marinelli beakers were previously washed, rinsed with a dilute sulfuric acid and dried to avoid any contamination. Hereafter, they firmly sealed for at least four weeks to ensure that no loss of radon occurs there by ensuring a state of secular equilibrium to be reached between radium isotopes and their respective daughters.

The gamma spectrometer HPGedetectorwith relative efficiency ≃30% relative to a "3x3" NaI(Tl) detectorand resolution≃ 1.8 keV at 1.33 MeV gamma-line is used. The energy calibration of the spectrometer was performed using the 1L Marinelli calibration sources which contains well-known standard sources (^{22}Na,^{60}Co, ^{57}Co and ^{241}Am).

In this work, relative efficiency curve of the detector was carried out using standard solution of known activity of ^{226}Ra which prepared with the same geometry as the samples to be measured.

2.2 Measurement of the gamma specific activity in water samples

Eachwater sample was placed on the HPGegamma spectrometer and counted for the same time (24h). Thedetected gamma lines are belonging to the naturally occurring radionuclides and a non-series natural radionuclide (^{40}K).

The activity of ^{226}Ra and ^{228}Ra were obtained indirectly from the gamma-raysemitted by their progenies which are in secular equilibrium with them while ^{40}K was estimated directly by its gamma-line of 1460.8 keV. ^{226}Ra activities were determined using the gamma-lines 295.2 keV from ^{214}Pb, 609.3 and 1120.3 keV from ^{214}Bi. ^{228}Ra activities were determined using gamma-lines 583.1 and 2614.4 keV from ^{208}TI and 338.7 and 911.2 keV from ^{228}Ac.

Using the net area of the peak and the other parameters such as: the counting time, the absolute gamma-ray emission probability,the absolute efficiencyat close geometryand the sample volume, the activities of the radionuclides were calculated(Aksoy, 1993). The reproducibility of the results and the stability of the counting technique were checked by conducting the triplicate analyses on all the water samples.

2.3 Minimum detectable activity

The minimum detection limit (MDL) for each activity was calculated using the following equation(Palmer and Mcinerney, 1994):

$$MDL = \frac{A}{NC} \times 3.3\sqrt{B} \quad (1)$$

Where NC is the net counts for a sample of activity A in Bq/kg, and B is the background counts below the peak of interest. The MDL for ^{226}Ra, ^{228}Raand ^{40}K were0.03,0.006 and 0.02Bq/L respectively.

2.4 Determination of the annual effective dose equivalent

The annual dose received from the consumption of this water in the study area can be calculated by using the following equation (USA-EPA, 1988):

$$D_{eff} = A_w \cdot CF_w \cdot R_w \quad (2)$$

Where $D_{eff}$is annual effective dose (mSv/year), $A_w$specific activity concentration (Bq/L),$CF_w$the ingestion dose conversion factor and $R_w$is the annual water consumption intake of water for person in 1 year (730 L/year). The ingestion dose conversion factors published by the International Commission of Radiological protection are: $2.8x10^{-7}$, $6.7x10^{-7}$and $6.2x10^{-9}$Sv/Bq for ^{226}Ra, ^{228}Ra and ^{40}K respectively(ICRP, 2012).

3. Results and Discussion.

3.1 Radioactivity Concentration

The obtained results of the mean radioactivity concentrations for^{226}Ra, ^{228}Ra and ^{40}Kin seawater, well water and tap water over the four locations (Al-Hanea, Al-Hamama, Susa and Ras Al-Helal) in Al-Bayda region are presented in table 1. In all locations, the activity concentration for seawater varied from 0.04 to 0.085 Bq/L for ^{226}Ra,from 0.0035 to 0.08 Bq/L for ^{228}Ra and from 1.5 to 3.3 Bq/L for ^{40}K. For well water, the concentrations varied from 0.07 to 0.1 Bq/L, from 0.05 to 0.09 Bq/L, and from 11.2 to 11.9 Bq/L for ^{226}Ra, ^{228}Ra and ^{40}K respectively. The corresponding concentrations that obtained for tap water are ranged from 0.065 to 0.138 Bq/L, 0.055 to 0.177Bq/L, and 0.930 to 5.607Bq/L for ^{226}Ra, ^{228}Ra and ^{40}K respectively.

The difference in concentrations of ^{226}Ra and ^{228}Ra is probably due to the distinct aquifer lithologies and consequent differences in radionuclides solubility and mobility. The great difference of chemical characteristics and especially the important difference of solubility between the two elements (U,Th) implies that the equilibrium is not often achieved in water sincethe thorium is particularly insoluble element in natural water and is usually found associated with solids. Hence, it has no disposition for transition with the water. In contrary, uranium element hasnormal distribution with the water affected by some factors such as temperature and salinity.

The mean concentration for ^{226}Ra,^{228}Ra and ^{40}K that obtained in this work (Table 1) are to great extent comparable with the results of some investigators such as that obtained for Brazilian Mineral water(Jose et al., 2001), Tunisian thermo-mineral springs in particular that for AinOktor spring(Labidi et al., 2002),natural water in Morocco(Azouazi et al., 2001), the surface water of Ebro river basin northeast Spain, surface water in Istanbul(Pujol et al., 2000),the water in Canada(Kronfeld et al., 2004), the groundwater around Buraydah (Saudi Arabia)(AL-Jaser, 2004), Black sea(Alexander et al., 1998), and drinking water samples that collected from Northwest Libya(Omar, 1997).

The values obtained for the sum of the activity concentrations of both ^{226}Ra and ^{228}Ra in well and tap water are below the safety value (1Bq/L) recommended for ^{226}Ra in water which obtained by Surbeck(Surbeck, 1995),also it is below the maximum acceptable concentrations for^{226}Ra (0.6 Bq/L) (HECS, 1995).

3.2 Annual Effective Dose Equivalent

Making use of the activity concentrations (Table 1) and the ingestion dose conversion factors for ^{226}Ra,^{228}Ra and ^{40}K, the annual effective dosescalculated by means of Eq. 2 are shown in Fig. 2. The annual effective dose equivalent forwell water ranged from0.09 to 0.12mSv/y with the average 0.10 mSv/y and for tap water is0.07 to 0.14mSv/y with the average0.09 mSv/y, while, the annual effective dose equivalent for sea water ranged from the 0.04 to 0.07 mSv/y with the average 0.06 mSv/y.These results represent about 100 %, 90 % and 60% of the level recommended for both well, tap and sea water respectively by the World Health Organization (WHO)(WHO, 2011) for the effective dose due to water consumption (0.1 mSv/y).

Table 2shows the physico-chemical characteristics of different water samples.Electric conductivity (EC) depends on the temperature, the level of the water, and the type and quantity of dissolved salts. The total dissolved solids (TDS) is measurement of all constituents dissolved in water. Generally, TDS follows the same pattern as the EC. TDS in well and tapwater, largely derived from aquifer minerals that dissolve in groundwater. TDS will change significantly as a function of temperature and PH value which equal about 7.30 and 7.24 for well and tap water respectively and 8.46 for seawater.

The principle dissolved constituents in groundwater are calcium, magnesium, sulphate and chloride, they occur in the form of electrically charged ions. As rain infiltrates through the soil it dissolves carbon dioxide. The acidic solution formed reacts with carbonates in the rocks giving solutions with calcium and to a lesser extent magnesium (Table 2).

An important source of sulphate in well and tap water is the oxidation of pyrite ferrous sulphide that is widely distributed in sedimentary rocks. Most of the chloride found in groundwater that is actively circulating at relatively shallow depths is derived from rain or, near coastline, from sea spray; radium ishighly soluble as a chloride.

The physico-chemical characteristics for water samples in this work are comparable to that for the water in Canada (Kronfeld, 2004), thermo-mineral springs in Tunisian(Labidi et al., 2002) and that obtained by Nawal Al Farrah in Northwest Libya(El-Trriki, 2006).

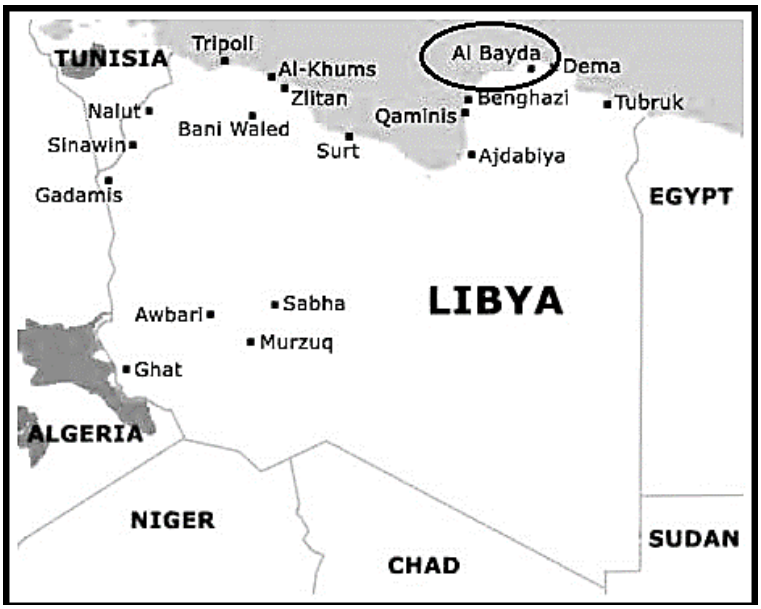

Fig. 1. Research area in Libya

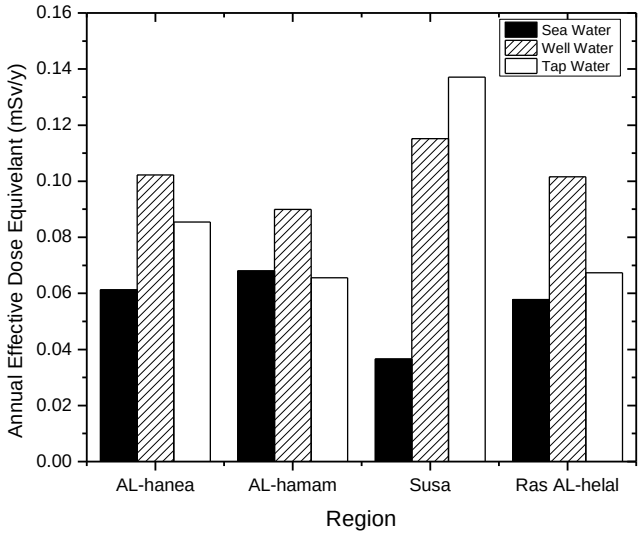

Fig. 2. The Annual Effective Dose Equivalent

Table 1. The mean activity concentration in (Bq/L) for water samples that collected from the four location in Al-Bayda region.

Location	Sea Water			Well Water			Tap Water		
	226Ra	232Th	40K	226Ra	232Th	40K	226Ra	232Th	40K
AL-hanea	0.075±0.005	0.080±0.003	1.500±0.123	0.080±0.006	0.070±0.005	11.411±0.120	0.093±0.005	0.084±0.007	5.607±0.534
AL-hamam	0.080±0.004	0.075±0.004	3.300±0.252	0.070±0.006	0.050±0.003	11.300±0.151	0.065±0.004	0.055±0.004	5.607±0.632
Susa	0.040±0.003	0.035±0.002	2.500±0.185	0.100±0.002	0.090±0.007	11.200±0.132	0.138±0.001	0.177±0.001	4.930±0.083
Ras AL-helal	0.085±0.005	0.065±0.003	1.900±0.165	0.090±0.001	0.06±0.003	11.921±0.163	0.085±0.006	0.059±0.005	4.670±0.512
Average	0.070	0.047	2.300	0.085	0.070	11.250	0.095	0.094	5.204

Table 2. Physico –Chemical characteristics of some water samples in study area.

Type of Water	PH	Ca^{++}(mg/L)	Mg^{++}(mg/L)	Cl^{-} (mg/L)	TDS (mg/L)	EC (mS/Cm)	SO_4^{--} (mg/L)
Sea water	8.46	605.00	1180.00	15332	39800.00	42.80	7120.00
Well water	7.30	13.19	1.11	96.3	200.00	40.00	4.67
Tap water	7.24	11.81	1.40	76.4	195.00	26.80	5.89

4. Conclusion

In the present work, the gamma ray spectroscopy technique has been applied to obtain the actual information on the radioactivity contained in different water samples from Al-Bayda, northeast of Libya.The results of the present work indicated that the mean radioactivity of ^{226}Ra, ^{228}Ra and ^{40}K in seawater, well water and tap water that is collected from Al-Bayda region are comparable with those reported for many other countries in the world for different types of water. The average annual effective dose from ^{226}Ra, ^{228}Ra and ^{40}K for well water, tap water and sea water samples has been obtained and found to have 0.10mSv/y, 0.09mSv/y and 0.06 mSv/y respectively. These results are below the level recommended by the World Health Organization (WHO, 2011) for effective dose due to water consumption (0.1 mSv/y).

The physico-chemical characteristics of different water samples are comparable to those obtained previously and need further investigations to draw an appropriate relation relating the radioactivity with them.

Additionally tap and well waters in the studied area could be exploited as a very clean bottled water to be a further source to improve the Libyan national income.

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