Determination of naturally occurring radionuclides in Disi aquifer water of Jordan

M B H Al-Bedri¹, T A J Younis², I J Abdulghani² and W O Hameed³

¹ Department of Medical Physics, Madenet Al-Elem University College, Baghdad, Iraq
² Physics Department, Education College for Pure Sciences/Ibn Alhaithem, Baghdad, Iraq
³ Physics Department, Faculty of Science, University of Jordan, Amman, Jordan

E-mail: mohd.albedri@gmail.com

Abstract. The Disi water samples were collected from different Disi aquifer wells in Jordan using a clean polyethylene container of 10-liter size. A hyper-pure germanium (HPGe) detector with high-resolution gamma-ray spectroscopy and a low background counting system was used for the identification of unknown gamma-rays emitting from radionuclides in the environmental samples. The ranges of specific activity concentrations of 226Ra and 228Ra in the Disi aquifer water were found to be from 0.302 ± 0.085 to 0.723 ± 0.207 and from 0.047 ± 0.010 to 0.525 ± 0.138 Bq L⁻¹, with average values of 0.516 ± 0.090 and 0.287 ± 0.091 Bq L⁻¹, respectively. The average combined radium (226Ra + 228Ra) activity and radium activity ratio (228Ra/226Ra) in Disi groundwater were found to be 0.803 ± 0.187 and 0.550 ± 0.178, respectively. A comparison was made between the present results and the international minimum recommended limits and previously published data from different countries.

Keywords: HPGe detector; Disi aquifer water; Specific activity concentrations of 226Ra and 228Ra; Combined radium activity, 228Ra/226Ra activity ratio

1. Introduction

Natural background radiation comes from cosmic rays, terrestrial radiation, and radioactivity in the body. People are exposed to natural background radiation everywhere [1]. Naturally-occurring radionuclides (NORs) are present in soil and rocks, they can also be found in groundwater and surface water [1, 2]. Typical radionuclides found in drinking water are the isotopes of uranium (238U and 235U) and thorium (232Th) series and their decay products, and potassium (40K). The concentration of 228Ra tends to exceed that of 226Ra reflecting the higher levels of 232Th in the earth's crust compared to that of 238U.

The Disi aquifer water lies beneath the desert in southern Jordan and northwestern Saudi Arabia. The aquifer system in south of Jordan is known as the Rum Group. The Disi groundwater pumped from Dubaydib wells in the aquifer's Rum Group to the main Disi reservoir, and then it is piped to the capital, Amman, and other cities to meet increased demand for water. Underground rocks containing natural uranium and thorium, and continuously releases radon into water in contact with groundwater. Radioactive radon gas is readily released from surface water; consequently, groundwater has potentially much higher concentrations of radon (222Rn and 220Rn) than surface water [1, 3]. 222Rn and 220Rn are the decay products of the radium isotopes 226Ra and 224Ra respectively, which are present in water, rocks and soil [2]. The decay products of radon—222 (222Rn) are: 214Pb (77.11, 87.30 and 351.62 keV) and 214Bi (609.31 keV), while the decay products of radon—220 (220Rn) are: 228Ac (338.32, 911.21 and 968.97 keV) and 208Tl (583.12 keV). 222Rn and 220Rn are found to be in secular equilibrium with radium—226 (226Ra) and radium—224 (224Ra) respectively, after four weeks or more. The water samples were sealed to stop radon (220Rn and 222Rn) from escaping from Marinelli beakers. From these decay products the specific activity concentration in Bq L⁻¹ of 238U and 232Th can be obtained.
Hyper-pure germanium (HPGe) detector was used to determine the activity concentration of naturally occurring radionuclides (NORs) in Disi aquifer water samples. It is a non-destructive technique, simple analytical methods, rapidly and high sensitivity [4]. Applications of high-resolution gamma-ray spectrometry system have been used for determination of specific activity concentrations of $^{238}$U, $^{232}$Th series and $^{40}$K in soil and sand [4 – 6], phosphate rocks [7], and ground water [8 – 11].

The specific activity concentrations of $^{226}$Ra and $^{228}$Ra, radium activity ratio ($^{228}$Ra/$^{226}$Ra) and combined radium activity of $^{226}$Ra and $^{228}$Ra were measured in Disi aquifer water samples. Comparison was made between the present results and international minimum recommended limits reported by (EU, 1998 [12]; ICRP, 2000 [13]; WHO, 2008 [14]) and the previous published data [3, 8 – 11, 15 – 27].

2. Materials and methods

2.1. Water sample collection and preparation

The layout of the Disi ground water samples from Dubaydib wells field of the aquifer's Rum Group in the south of Jordan are shown in figure 1, consists of 55 wells: 46 of them are operated wells by pumps and 9 are used in emergencies only (modified from Disi-Mudawaraa conveyor ESA Addendum 2, 2008 [30]). The Disi water samples were collected from nine different Dubaydib wells (marked with black solid circle as shown in figure 1), using a clean polyethylene container of 10 – liter size. Water samples were properly marked and then sent to the laboratory at the University of Jordan to be analyzed. The volume of each water sample was measured precisely. Marinelli beakers of 600 mL were filled with water samples, closed by caps, and firmly sealed with plastic tapes to prevent the escape of radon ($^{220}$Rn and $^{222}$Rn) from the Marinelli beakers. The sealed water samples were stored for at least four weeks or more before measurement to achieve radioactive secular equilibrium between $^{226}$Ra, $^{228}$Ra and their short-lived $^{222}$Rn and $^{220}$Rn, and their decay products, respectively.

![Figure 1. Shows the geographical locations of the Disi aquifer water samples collected from nine Dubaydib wells field in the south of Jordan as marked with black solid circle (modified from Disi-Mudawarrra conveyor ESA Addendum 2, 2008 [30]).](image)

2.2. Instrumentation and calibration

A hyper-pure germanium (HPGe) detector with high-resolution gamma-ray spectroscopy low background counting system at the University of Jordan was used to determine the activity concentration of naturally occurring radioactive materials (NORMs) in Disi water samples. The spectrometry system consists of HPGe coaxial detector with active volume of 180 cm$^3$, an operating voltage of ~4000 V, and the
associated electronics were installed in a personal computer for data analysis [7]. The energy calibration was carried out by using multi-standard radioactive sources emitting γ-rays of precisely known energy, and then identifying the peak position in channels for each energy. The detention efficiency of the HPGe detector was determined by using standard radioactive sources. The energy resolution (FWHM) of the HPGe detector at 1.332 MeV from $^{60}$Co is 1.9 keV, and the relative efficiency at 1.332 MeV is 30%.

Spectrum for each sample was collected for 79200 seconds counting time to reduce statistical errors. The area under each identified peak of the spectra was calculated individually. The background carefully measured by filling the Marinelli beaker with inactive de-ionized water, then placed on the top of the detector to get the same geometry. The net number of counts for each photopeak was obtained by subtracting the background counts from the total counts in the same photopeak, for the same counting time of 79200 seconds.

2.3. Determination of the specific activity concentration

The activity concentrations of γ-rays emitting from radionuclides in water samples have been determined by employing a gamma-ray spectrometry system using an HPGe detector in a low background configuration. The majority of NORMs belongs to the radionuclides in the $^{238}$U and $^{232}$Th series [28] was determined in each water sample. In the $^{238}$U and $^{232}$Th decay series, $^{222}$Rn and $^{220}$Rn are the only radionuclides found in the gaseous state and they can emanate naturally from Disi aquifer water.

The area under each photopeak of the spectrum was calculated individually and was subtracted from the natural background radiation. The net specific activity concentrations of individual radionuclides $i$ for photopeak at energy $E_i$ in water samples can be calculated by using the following expression:

$$ A (\text{Bq/L}) = \frac{N}{(C_{E_i} I_{E_i} t V)} $$

where

- $A$ = specific activity concentrations of individual radionuclides $i$ for a peak at energy $E_i$ in the water samples (Bq/L),
- $N = (N_{E_i})_S - (N_{E_i})_B$ is the net peak area at energy $E_i$ of radionuclide $i$,
- $(N_{E_i})_S = $ peak total counts at energy $E_i$ of radionuclide $i$ (counts),
- $(N_{E_i})_B = $ background total counts at energy $E_i$ of radionuclide $i$,
- $C_{E_i} = $ peak detection efficiency of a particular γ-ray energy ($E_i$),
- $I_{E_i} = $ absolute intensity corresponding to the peak at energy $E_i$,
- $V = $ volume of the measured water sample (L) and $t = $ counting time of the water sample (s)

The activity concentration of $^{226}$Ra was determined by measuring the average activity of γ-ray lines of its daughter products: $^{214}$Pb (77.11, 87.30 and 351.62 keV) and $^{214}$Bi (609.31 keV), while the activity concentration of $^{228}$Ra was determined by evaluating the average activity of γ-ray lines of its daughter products: $^{228}$Ac (338.32, 911.21 and 968.97 keV) and $^{208}$Tl (583.12 keV).

3. Results and discussion

3.1 Activity concentrations of $^{226}$Ra and $^{228}$Ra in Disi aquifer water

The range of total dissolved solids (TDS) per liter in Disi aquifer water was found between 125 and 250 mg/L, with low salinity and is usually acceptable for drinking water [29]. Table 1 shows the specific activity concentration values ± standard deviation (S.D.) in Bq/L for $^{226}$Ra and $^{228}$Ra, combined radium ($^{226}$Ra + $^{228}$Ra) activities, and radium activity ratio ($^{228}$Ra/$^{226}$Ra) in groundwater from nine different Disi aquifer wells compared with the international minimum recommended limits (IMRL) reported by (EU 1998 [12], ICRP 2000 [13] and WHO 2008 [14]). The ranges of specific activity concentrations of $^{226}$Ra and $^{228}$Ra in the Disi aquifer water were found to be from 0.302±0.085 to 0.723±0.207 with an average value of 0.516±0.090 Bq/L and from 0.047±0.010 to 0.525±0.138 Bq/L with average values of
The activity concentrations of 226Ra and 228Ra, and radium activity ratio for Disi groundwater samples of the present study compared with previously published data in groundwater in some other countries are shown in Table 2. The range and mean values of the activity concentrations of 226Ra and 228Ra, and radium (228Ra/226Ra) activity ratio in Disi water of the present work were found to be less than the results obtained in groundwater in some other countries such as Brazil [16]; Japan [23]; Jordan [8, 27]; Nigeria [22]; Palestine [11]; Poland [9]; Saudi Arabia [17]; Spain [24]; Sweden [25]; Tanzania [10]; Thailand [21]; UK [15]; USA [19]; Yemen [26], except those results obtained in Egypt [18], Romania [20] and Finland [3]. This variations of the concentrations of 226Ra and 228Ra, 228Ra/226Ra activity ratios...
and combined radium activity of the present study and some previous published data over the world as shown in tables 1 and 2 are due to the variations of the activity concentrations of the parent’s series of $^{238}\text{U}$ and $^{232}\text{Th}$, which depends on type of rocks and their geographical locations as reported by (Grabowski et al., 2015 [9] and Kodcharin et al., 2018 [21].

A study in Jordan by Vengosh, et al., 2009 [27] has revealed that high levels of radium activity in ground water from the Rum Group of the Disi sandstone aquifer and they found that the average combined radium ($^{228}\text{Ra} + ^{226}\text{Ra}$) activities in the unconfined and confined zones of the Rum Group are respectively 9 and 18 times higher than international standard limits of drinking water as shown in table 2. They found that the radium ($^{228}\text{Ra}/^{226}\text{Ra}$) activity ratios are 19 to 30 times higher than the minimum recommended limits reported by WHO 2008 [14]. Vengosh and his coauthors may be collected the water samples from all wells in the Disi area used for agriculture activities, which are not considered as resources of drinking water in Jordan. Present results of the determination of radium concentrations, combined radium activity and radium activity ratios in the groundwater from Rum Group of Disi wells are usually used for daily consumption in Jordan are 3 to 7 times lower than the results obtained by Vengosh et al., 2009 [27], this disagreement may be due to that their measurements of radium activity concentrations of both $^{228}\text{Ra}$ and $^{226}\text{Ra}$ in groundwater wells which are used for agriculture activities.

### Table 2. The activity concentrations of $^{226}\text{Ra}$ and $^{228}\text{Ra}$, and radium activity ratio in the Disi aquifer water of present work compared with previously published data from other countries.

| Country                  | Specific activity concentration (Bq/L) | Radium activity ratio ($^{228}\text{Ra}/^{226}\text{Ra}$) |
|--------------------------|----------------------------------------|----------------------------------------------------------|
|                          | $^{226}\text{Ra}$ | $^{228}\text{Ra}$ |                                              |
| Brazil, 2004 [16]        | < 0.002 ± 0.492 | < 0.01 ± 1.5 | – |
| Egypt, 2004 [18]: Qena   | 0.079 ± 0.029 | – | – |
|                          | Safaga-Quseir  | 0.113 ± 0.033 | – |
| Finland, 2007 [3]        | 0.05 | 0.034 | – |
| Japan, 2012 [23]         | – | – | 0.80 – 1.1 |
| Jordan, 2009 [16]: Unconfined Rum | 0.534 | 0.906 | 1.90 |
|                          | Confined Rum group | 0.678 | 1.991 | 2.93 |
| Jordan, 2016 [8]         | 0.37 | 1.35 | – |
| Present study (Range)    | 0.302 – 0.723 | 0.047 – 0.525 | 0.084 – 0.905 |
| Present study (Mean value ±S.D.) | 0.516 ± 0.090 | 0.287 ± 0.091 | 0.550 ± 0.178 |
| Nigeria, 2013 [22]       | 3.7 ± 2.2 | 3.6 ± 3.3 | 0.973 |
| Palestine, 2012 [11]    | 1.62 – 9.11 | 0.45 – 4.4 | – |
| Poland, 2015 [9]        | – | – | 0.396 – 4.96 |
| Romanian, 2019 [20]     | 0.28 ± 0.03 | – | – |
| Saudi Arabia, 2015 [17] | 0.29 ± 0.025 | 0.86 ± 0.029 | – |
| Spain, 1996 [24]        | 0.96 | 1.130 | – |
| Sweden, 2002 [25]       | 0.26 ± 4.9 | – | – |
| Tanzania, 2013 [10]     | 2.35 | 1.85 | 0.787 |
| Thailand, 2018 [21]     | 0.77 ± 0.13 | 1.03± 0.13 | – |
| UK, 1993 [15]           | 3 – 70 | – | – |
| USA, 2005 [19]          | < MDC | < MDC | – |
| Yemen, 2015 [26]        | 0.86 – 3.09 | – | – |

* Minimum detectable concentration

The average annual effective dose (AED) for adults is 0.250±0.067 mSv/y who consumed an average of 730 liter of Disi groundwater per year which is 2.5 times higher than that of WHO, 2008 [14] recommended limits of 0.1 mSv/y. The mean annual effective dose for adults is more than the recommended limits reported by WHO, 2008 this difference is due to the people consume high concentrations of $^{238}\text{U}$ and $^{232}\text{Th}$ in Disi drinking water.
3.2 Treatment method of Disi ground water
To bring Disi water in Jordan up to the international standards of drinking water (WHO 2008 [14]), groundwater from Disi wells must be treated to reduce radium (\(^{228}\text{Ra}\)) levels to be below 0.1 Bq/liter. From the results obtained in the present study; Disi water must be diluted to reduce the level of \(^{228}\text{Ra}\) activity down to acceptable recommended limits, by mixing it in Dabouk reservoir and Abu Alanda reservoir with non–Disi water from other sources with 3:1 ratio of non-Disi water: Disi water ratio. In this suggested procedure the water dilution would be satisfactory enough to reduce the activity concentration of \(^{228}\text{Ra}\) down to 0.095 Bq/L. This dilution revealed that the radiological quality of the Disi drinking water to be satisfied for human consumption and to bring the Disi water up to the international standards.

4. Conclusions
Hyper-pure germanium detector with high-resolution gamma-ray spectroscopy system was used to determine the specific activity concentrations of natural radionuclides in Disi aquifer water samples. The average activity concentration of \(^{226}\text{Ra}\) in Disi groundwater was found to agree with the minimum recommended limits reported by (EU 1998 [12] and WHO 2008 [14]), while the average activity concentration of \(^{228}\text{Ra}\) was about 2 to 3 times higher than that of MRL as suggested by WHO 2008 [14]. The average radium (\(^{226}\text{Ra}/^{228}\text{Ra}\)) activity ratio was about 5.5 times higher than MRL of 0.1 reported by WHO 2008.

The average annual effective dose (AED) for adults is 0.250±0.067 mSv/y which is 2.5 times higher than that of WHO, 2008 [14] recommended limits of 0.1 mSv/y.

From the present investigations; Disi water must be diluted to reduce the level of \(^{228}\text{Ra}\) activity down to be less than 0.1 Bq/L, by mixing it with non-Disi water from other sources with 3:1 ratio of non-Disi water: Disi water ratio. In this procedure, the water dilution would be satisfied enough for human consumption to reduce the activity concentration of \(^{228}\text{Ra}\) down to 0.095 Bq/L.

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