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Natural Radioactivity of Ground and Drinking Water in Some Areas of Upper Egypt

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Abstract

Concentrations of natural radionuclides (^{226}Ra and ^{232}Th) in ground and drinking waters of some areas in Upper Egypt were determined by gamma ray spectrometry with a HPGe detector setup, coaxial type and 8192 channels MCA. The investigated waters differed in radioactivity content depending on their origin and place. In drinking water in Qena, Upper Egypt, the mean ^{226}Ra concentration was 1.32 ± 0.7 pCi/l. In ground water in Safaga and Quseir, the Red Sea region, where there are phosphate mines, ^{226}Ra and ^{232}Th mean values were 3.05 ± 0.9 and 1.39 ± 0.6 pCi/l. A mean annual effective dose taken into the body by the population drinking this tap water may account for 0.008 mSv, which is lower than the limit recommended by the WHO.

Key words: Natural radioactivity, Upper Egypt, Ground water, Drinking water.

Introduction

There are different forms (isotopes) of uranium but ^{238}U is the predominant contributor to natural radioactivity. The average ^{238}U content in the Earth's crust has been estimated to be 2.7 mg/kg and concentrations may be as high as 120 mg/kg in phosphate rocks (Padam *et al.*, 1996). Meanwhile, the average ^{232}Th content of the Earth's crust is about 9.6 mg/kg (Firestone *et al.*, 1996). Enhanced levels of uranium, thorium and their daughter products might be present in water in areas that are rich in natural radioactivity. As groundwater moves through fractures in the bedrock that contain these deposits radioactive minerals can leach out into the groundwater system. Wells constricted in bedrock within such areas could show levels of natural radioactivity in water quality tests. The analysis of drinking and groundwater shows that the natural radioactivity in water varies over a wide range, mainly depending on the geological characteristics of the soil.

Uranium isotopes (^{238}U, ^{234}U and ^{235}U) have a non-negligible radiotoxicity (WHO, 1978; Malcome-Lawes, 1979). In addition, several radionuclides in the radioactive decay chain starting from ^{238}U and ^{235}U are highly radiotoxic. The most radiotoxic and most important among them is radium, which is a known carcinogen and exists in several isotopic forms. The predominant radium isotopes in groundwater are ^{226}Ra, an alpha emitter with a half-life of 1600 years, and ^{228}Ra, a beta emitter with a half-life of 5.8 years (Iyenger, 1990; Marovic *et al.*, 1996; Sidhu and Breithart, 1998).

Considering the high radiotoxicity of ^{226}Ra and ^{228}Ra, their presence in water and the associated health risks require particular attention. It is known that even small amounts of a radioactive substance may produce a damaging biological effect and that ingested and inhaled radiation can be a serious health risk (Rowland, 1993). When radium is taken into the body, its metabolic behavior is similar to that of calcium and an appreciable fraction is deposited in bone, the remaining fraction being distributed almost uniformly in soft tissues (Wrenn *et al.*, 1985). When people are exposed to very high levels of radium for a long period of time, cancer of

the bone and nasal cavity may result.

The radium concentration in surface waters normally ranges from 0.01 to 0.1 Bq/l (Iyenger, 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 groundwater the radium concentrations can reach values up to 38 Bq/l, depending on factors such as type of aquifer rock and chemical and physical water characteristics (Gascoyne, 1989). On the other hand, these radionuclides may contribute appreciably to the dose received by humans through internal exposure due to their ingestion (UNSCEAR, 1993; Eisenbud and Gesell, 1997). Experimental works suggested that the number of fatal cancers due to ^{222}Rn, a daughter of ^{226}Ra, ingested from drinking water may equal the fatal lung cancers due to the inhalation of indoor Rn (Correia *et al.*, 1987). Thus, high quantities of these nuclides in water necessitate the use of appropriate methods of elimination like those summarized by Sorg (Sorg, 1988, 1991). An important aspect of radium protection is the prevention of its entry into the human body, the critical pathway being ingestion through the food chain or drinking water (Kahlos and Asikainen, 1980). The present study attempts to understand the occurrence and distribution of natural radionuclides ^{226}Ra and ^{232}Th in ground and drinking waters in Qena, a city in Upper Egypt, and the Red Sea regions, to assess the concentrations of ^{226}Ra that exceed the standard for ^{226}Ra in these areas, and to estimate the radiation doses received by adults living in these areas.

Experimental Methods

Sampling and sample preparation

A total of 89 ground and drinking water samples were collected and prepared for gamma spectroscopic analysis from Qena, Upper Egypt, and the Red Sea regions as follows:

- 25 drinking water samples taken from the tap water system in Qena, Upper Egypt.
- 15 drinking water samples taken from the tap water system in Safaga and Quseir regions, Red Sea.
- 24 water samples collected from artesian wells distributed in Qena governorate (used for drinking water).
- 25 water samples collected from open wells – in Safaga and Quseir, Red Sea regions, (not used for drinking).

The sampling locations are shown in Figure 1.

The collection of water samples requires particular care because radon is a short lived gaseous nuclide that tends to escape from water during sampling. In this study, a 1.4 -l polyethylene Marinelli beaker was used as a sampling and measuring container. Before use, the containers were washed with dilute hydrochloric acid and rinsed with distilled water. Each beaker was filled up to the brim and a tight cap was pressed on so that the air was completely removed from it. The samples were taken to the laboratory and stored for a minimum of 1 month to allow daughter products to come into radioactive equilibrium with their parents ^{226}Ra and ^{232}Th. Every sample was counted for 600 to 900 min depending on the concentrations of the radionuclides. This method of sampling and sample preparation has been discussed before in earlier publications (Abbady *et al.*, 1995; Ahmed *et al.*, 1995).

Experimental setup

Each sample was subjected to a gamma ray spectrometer with a HPGe setup and multichannel analyzer of 8192 channels. The detector had coaxial closed facing window geometry with a vertical dipstick (500-800 μ). The HPGe detector was p-type with the following specifications:

Resolution (FWHM) at 122 keV ^{57}Co is 1100 eV and at 1.33 MeV ^{60}Co is 2.00 keV.

Relative efficiency at 1.33 MeV ^{60}Co is 30% .

The detector is shielded in a chamber of 4 layers starting with Plexiglas (10 mm thick), followed by copper (30 mm thick), lead (100 mm thick) and finally cadmium (3 mm thick). This shield serves to reduce different radiation hazards. The soft component of cosmic rays, consisting of photons and electrons, is reduced 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).

The emitted X-rays from lead, which contains radioactive impurities due to the presence of antimony, can be absorbed by lining the inside of the shield with a graded layer of 0.05 inch cadmium and 0.25 inch perspex (Aziz, 1981). To minimize the effect of the scattered radiation from the shield, the detector was located in the center of the chamber. Then the sample was placed in face to face geometry over the detector for at least 10 h.

The system was calibrated for energy and efficiency. The energy calibration was carried out by acquiring a spectrum from radioactive standards of

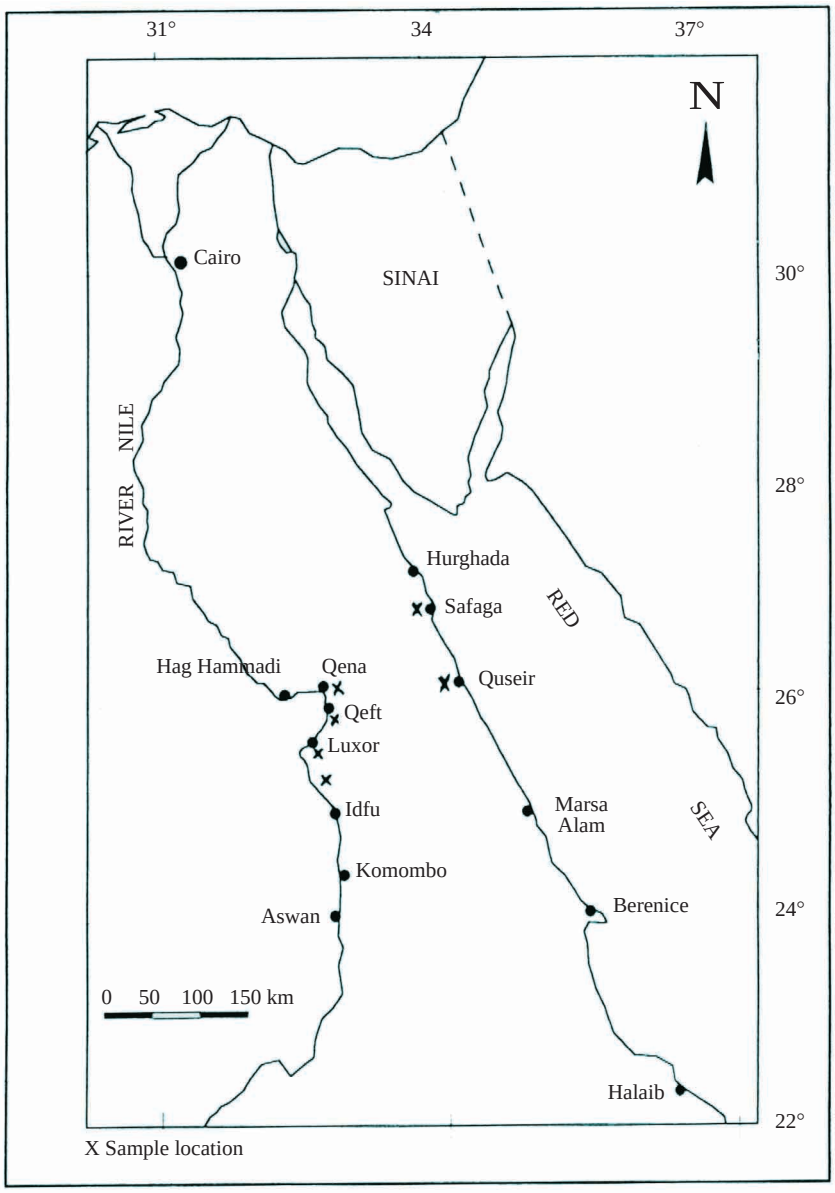

Figure 1. Location map of the area under study.

known energies like ^{137}Cs E_γ= 662 keV and ^{60}Co E_γ = 1332 keV. For the efficiency calibration we used point sources ^{133}Ba, ^{137}Cs and ^{60}Co. For background a 1.4 - l Marinelli beaker filled with distilled water was used. The background was measured frequently usually every week under the same conditions of sample measurement. The spectra were evaluated either with the computer software Maestro (EG & G ORTIC), or manually with the use of a spread sheet (Microsoft Excel).

The ^{226}Ra activity of the samples was determined through the intensity of 351.9 keV and 609.3 keV γ-lines of ^{214}Pb and ^{214}Bi respectively. Several gamma lines of ^{214}Pb and ^{214}Bi were also monitored. ^{232}Th activity was obtained through the gamma line of ^{228}Ac at 911.21 keV with an emission percentage of 26.6% . In addition, the 968.97 keV line with an emission percentage of 17.4% was also used and the results were averaged.

Results and Discussion

Table 1 shows the radionuclide activities of ^{226}Ra and ^{232}Th in different waters in different locations in

Qena and the Red Sea regions. The range of ^{226}Ra and ^{232}Th nuclides in tap water in Qena are 0.27-2.45 and 0.27-1.35 pCi/l, with mean values of 1.32 ± 0.7 and 0.74 ± 0.44 for the 2 radionuclides respectively. Furthermore the minimum concentration values of ^{226}Ra and ^{232}Th of drinking water (tap water) in Safaga and Quseir are 0.27 pCi/l for both ^{226}Ra and ^{232}Th, and the maximum concentration values are 2.43 and 1.35 pCi/l for these radionuclides respectively. Their mean values are 1.52 ± 0.77 and 0.79 ± 0.45 pCi/l. The concentrations of ^{226}Ra in the samples are higher than the ^{232}Th concentrations in the 2 locations.

The values of ^{226}Ra and ^{232}Th in drinking waters of Safaga and Quseir are nearly the same as that of Qena. This is because the origin of the raw water of the 3 stations, Qena, Safaga and Quseir, is the same, being supplied by intake from the River Nile. The drinking water of Safaga and Quseir cities comes from a water purification plant on the River Nile and then through pipe lines from Qena to Safaga and from Quseir across the Eastern Desert. The natural radioactivity of the subsoil in Qena area is generally low. The range of ^{222}Rn concentration of tap water in Qena area is 2 to 63 pCi/l (Abbady *et al.*, 1995). The range of radium values in this investigation is much smaller than that of radon, indicating the lower variability of the radium of drinking water due to changing environmental influences.

Table 1. Activity concentrations of ^{226}Ra and ^{232}Th (pCi/l) in waters of different locations.

Sample No.	Drinking water in Qena (Tap water)		Drinking water in Safaga & Quiser (Tap water)		Artesian wells in Qena Governorate (used for drinking)		Open wells in Safaga & Quseir (not used for drinking)	
	^{226}Ra (pCi/l)	^{232}Th (pCi/l)	^{226}Ra (pCi/l)	^{232}Th (pCi/l)	^{226}Ra (pCi/l)	^{232}Th (pCi/l)	^{226}Ra (pCi/l)	^{232}Th (pCi/l)
1	0.43	0.27	1.08	0.27	3.51	2.16	2.7	1.35
2	1.08	0.2	1.62	1.08	2.43	0.54	2.7	1.89
3	1.62	1.08	0.27	0.27	0.43	0.27	2.97	1.35
4	0.43	0.54	1.35	0.54	2.7	2.16	2.97	1.89
5	2.43	1.35	1.35	0.27	2.16	1.08	1.35	1.35
6	1.62	1.08	2.43	1.08	1.62	1.08	1.62	2.43
7	1.35	1.08	0.43	0.27	2.7	1.35	2.97	2.16
8	0.43	0.27	2.16	1.08	1.89	2.16	2.7	1.35
9	0.27	0.27	2.16	0.54	1.62	1.62	1.35	1.35
10	1.62	0.54	1.89	1.35	2.43	1.35	2.7	1.89
11	1.08	0.27	1.08	1.08	2.97	1.62	2.97	1.35
12	1.35	1.08	2.43	1.35	1.62	0.27	3.51	0.54
13	1.62	0.54	0.27	0.27	2.16	1.62	3.51	0.27
14	0.43	0.27	2.43	1.08	2.16	0.54	4.05	2.43
15	1.62	1.35	1.89	1.35	1.89	1.62	2.43	1.62
16	2.16	1.08	-	-	2.97	1.35	3.51	0.81
17	2.16	0.54	-	-	3.51	-	3.78	1.35
18	1.62	1.08	-	-	2.7	0.81	2.97	1.08
19	1.89	1.35	-	-	1.08	0.27	1.08	1.62
20	0.27	0.43	-	-	1.35	1.08	4.59	2.43
21	0.43	0.27	-	-	1.08	1.35	4.59	0.54
22	1.35	0.27	-	-	2.97	1.08	4.05	0.81
23	2.16	1.35	-	-	1.62	1.08	3.78	0.27
24	1.08	0.54	-	-	1.89	1.62	3.78	1.62
25	2.43	1.35	-	-	-	-	3.51	0.89
average	1.32 ± 0.7	0.74 ± 0.44	1.52 ± 0.77	0.79 ± 0.5	2.14 ± 0.8	1.1 ± 0.6	3.05 ± 0.9	1.39 ± 0.6

A large number of drinking water samples were analyzed for ^{226}Ra content in Salzburg with ionization chamber techniques (Steinhauser *et al.*, 1982). The results indicated that the range of ^{226}Ra concentrations was 0.1 to 8.2 pCi/l with a mean value of 1.5 pCi/l. Results similar to those found in Salzburg were also found in Finland (Asikainen and Hahlos, 1980), where nearly 700 groundwater samples were analyzed and showed a mean concentration of 0.2 pCi/l, with a maximum of 8.5 pCi/l. Domestic bottled water had a ^{226}Ra concentration less than an average of 2.2 pCi/l, while imported bottled water had concentrations as high as 13.5 pCi/l (McCurdy and Mellor, 1981). These investigations are in agreement with the results obtained for the drinking water in this investigation. Gans *et al.* (1982) investigated the ^{226}Ra content of drinking water (tap water and bottled water) in Germany. The results show that the average for tap water was 0.1 pCi/l and that of bottled water was about 1 pCi/l ^{226}Ra. The maximum value found was 3.7 pCi/l. The average values are somewhat lower than the results of our investigation.

Table 1 shows the activity concentrations of ^{226}Ra and ^{232}Th in artesian wells in Qena governorate. The minimum contents are 0.43 and 0.24 pCi/l and the maximum contents are 3.51 and 2.16 pCi/l for the radionuclides, respectively. The mean values are 2.14 ± 0.78 and 1.17 ± 0.62 pCi/l. The activity concentrations of ^{226}Ra and ^{232}Th in open wells in Safaga and Quseir are given in Table 1. The average values of the concentrations of these radionuclides are 3.05 ± 0.9 and 1.39 ± 0.6 pCi/l. The minimum concentrations are 1.08 and 0.27 pCi/l, while the maximum values are 4.59 and 2.43 pCi/l for the same radionuclides, respectively. Table 2 presents the activity concentration of ^{226}Ra and ^{232}Th pCi/l in all areas under investigation.

The activity of groundwater in Qena is less than that in Safaga and Quseir for ^{226}Ra and ^{232}Th contents. No correlation between the concentrations of the radionuclides and the depth of the wells was found in the whole area. Specific activities of ^{226}Ra and ^{232}Th by location are shown in Figure 2. ^{226}Ra

Table 2. Activity concentrations of ^{226}Ra and ^{232}Th (pCi/l) in all areas under investigation.

Type of water and locations	^{226}Ra	^{232}Th
Drinking water in Qena (tap water)	1.32 ± 0.7	0.74 ± 0.44
Drinking water in Safaga and Quseir (tap water)	1.52 ± 0.77	0.79 ± 0.45
Artesian wells in Qena governorate (used for drinking)	2.14 ± 0.78	1.1 ± 0.62
Open wells in Safaga and Quseir (not used for drinking)	3.05 ± 0.9	1.39 ± 0.6

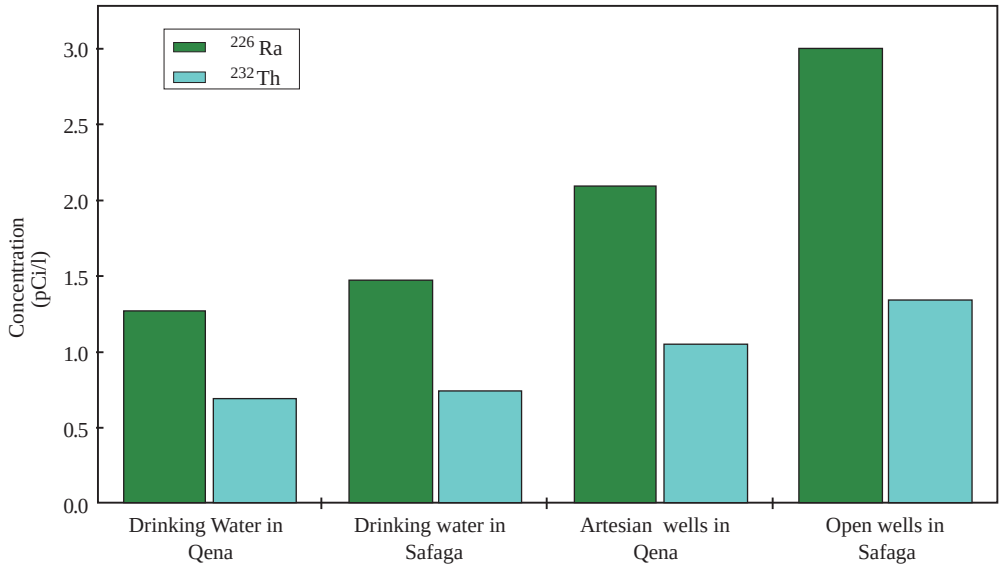

Figure 2. Mean activity concentration of radium and thorium in ground and drinking water in studied samples.

concentrations in the drinking water in Qena and both Safaga and Quseir varied between 0.27 and 2.47 pCi/l, and ^{232}Th concentrations varied between 0.2 and 1.35 pCi/l. These values show small ranges of ^{226}Ra and ^{232}Th activities in the investigated waters, which suggests that a valid and reliable conclusion on the observed activities could be inferred by investigating them over a longer period of time and on a more regular basis.

In the groundwater, ^{226}Ra concentrations in Qena, Safaga and Quseir varied between 0.43 and 4.59 pCi/l and ^{232}Th concentrations varied between 0.27 and 2.43 pCi/l. Although a regional variation in ^{226}Ra concentrations was found, the range of this variation was not high because the geological structure of the subsoil in the Qena area is generally limestone with a low content of natural radionuclides. On the other hand, the radioactivity in open wells in Safaga and Quseir was high, compared with the wells in Qena, because these wells (Safaga and Quseir) are near phosphate mines. Phosphate rock makes the water sources rich in ^{238}U and its daughters, which may be found in dissolved or suspended form in water. The increase in uranium concentration may be due to leaching. It has been proposed that some factors are likely to influence the uranium concentration in natural surface water and ground water (Langmuir, 1978). These factors are summarized as follows:

1. The uranium content of the source rocks, sediments or soils, and the processes through which uranium may be leached.

2. The proximity of the water to the uranium source.

3. The degree of hydraulic isolation of the water by fresher surface or groundwater.

4. The oxidation state of the water and,

5. The concentration of suitable complexing agents, which can increase the solubility of uranium.

In addition, no correlation was found between the results and the concentrations of ^{226}Ra and ^{232}Th. In our investigation, in the majority of cases the concentration of ^{226}Ra exceeded that of ^{232}Th. The reason is that the geological and solubility properties of ^{226}Ra and ^{232}Th are different. Their occurrences in water are determined by several factors such as the geology and their geochemistry (Molinari and Snodgrass, 1990). Thorium is essentially insoluble in normal surface waters, and so thorium transport is within particulate matter rather than in solutions. The natural radionuclides in drinking water and hot springs in Jordan were investigated by Saqan *et al.*(2001). They found that the ^{232}Th mean value concentration was 0.29 mg/l in drinking water, while the concentrations of ^{232}Th in hot springs were 0.59, 0.44, 0.45 and 0.51 mg/l for different sites. Table 3 summarizes the average values of ^{226}Ra and ^{232}Th concentrations in other countries and those from the present investigation.

The maximum allowed value for radium concentrations in drinking water is 5 pCi/l (USEPA, 1976). When this limit is taken into consideration, the drinking water in Qena, Safaga and Quseir as well as the groundwater in Qena (which is used for drinking) are below this limit. Generally, measures should be taken if the activity of the radioactive contaminates detected in water exceeds the permissible levels.

Daily intake of ^{226}Ra and ^{232}Th from water

When analyzing the total annual effective dose to the human population from natural sources, the dose received by ingestion of long-lived natural radionuclides must be considered. Effective doses resulting from the intake of ^{226}Ra and ^{232}Th may be determined directly from external measurements of their concentrations in the body or estimated from concentrations in intake materials such as air, food and water. Intakes of the natural radionuclides ^{226}Ra and ^{232}Th through water in Qena, Safaga and Quseir tap and groundwaters were calculated. Assuming the volume of drinking water for adult males to be 1 l/d (WHO, 1993), these intakes of ^{226}Ra and ^{232}Th through ground - and drinking water (tap water) in all locations are presented in Table 4.

Total annual effective dose

The annual effective dose was calculated with the intake of individual radionuclides and ingestion dose coefficients (Sv Bq^{-1}) reported by the International Commission on Radiological Protection (ICRP, 1994). The equation for calculating the annual effective dose per person is

$$\text{The annual effective dose} = \sum_i \mathrm{I}_i \times 365 \times \mathrm{D}_i$$

where $\mathbf{I}_i$ is the daily intakes of radionuclide **i** (Bq d^{-1}), and $\mathbf{D}_i$ is the ingestion dose coefficient (Sv Bq^{-1}). Table 4 is a summary of the estimates of annual effective doses for Qena, Safaga and Quseir males for tap and groundwater. In the area under investigation, the doses received by the ingestion of ^{226}Ra and ^{232}Th are shown in Figure 3.

Considering the volume of intake of drinking or groundwater by the adult males in Qena, Safaga and Quseir to be 1 l/d (WHO, 1993), the annual effective dose received by the population as a result of the ingestion of both ^{226}Ra and ^{232}Th in water is estimated to be 0.0072 and 0.0080 mSv for drinking water and 0.0114 and 0.0156 mSv for groundwater, respectively. These results are within the range observed in other countries (Ulback and Klinder, 1984; Jankowiski *et al.*,2000; Kralik *et al.*,2003). According to ICRP recommendations (ICRP, 1991) the limit for public exposure should be expressed as an effective dose of 1 mSv y^{-1}. The doses obtained in our study are significantly below that recommended for all categories of water.

Table 3. Average values of ^{226}Ra and ^{232}Th concentrations in units of pCi/l in water samples of the present investigation in comparison with other countries.

Country	Type of water	^{226}Ra (pCi/l)	^{232}Th (pCi/l)	Reference
China	Ground water	Min. 0.03 Max. 25.4 Mean 0.34	- - -	Weihai, Zhuo et al.(2001)
Finland	Ground water in bedrock (Drilled wells) Ground water in soil (Dug wells and springs)	Mean 2.97 Mean 0.27	- -	(Salonen, 1988)
China	Ground water	Up to 10.6	0.3	Ziqiang *et al.*(1988)
Denmark (Island of Bornholm)	Wells Tap water	14.9 -	-	(Ulback and Klinder, 1984)
India (Bombay)	Tap water	-	8×10^{-4}	(Fisenne, 2000)
Inner Mongolia	Tap water Wells Springs	- - -	54×10^{-4} 24×10^{-4} 81×10^{-4}	(Fisenne, 2000)
Germany (Erzgebirge)	Tap water	-	14×10^{-3}	(Fisenne, 2000)
U.S. (New York)	Tap water	-	14×10^{-4}	(Fisenne, 2000)
Austria	Domestic bottle water	1.12	-	Kralik *et al.*(2003)
Tunisia	Springs	0.92–105.3	-	Labidi *et al.*(2002)
Hungary	Bottled mineral water	2.7-81	-	Somlai *et al.*(2002)
Portugal	Mineral water	0.73	-	Bettencourt *et al.*(1988)
Poland (Lodz)	Underground water	0.27–1.35	-	Jankowski *et al.* (2000)
Qena (Egypt)	Drinking water Ground water	1.32 ± 0.7 2.14 ± 0.78	0.74 ± 0.44 1.1 ± 0.62	Present work
Safaga-Quseir (Egypt)	Drinking water Ground water	1.52 ± 0.77 3.05 ± 0.9	0.79 ± 0.45 1.39 ± 0.6	Present work

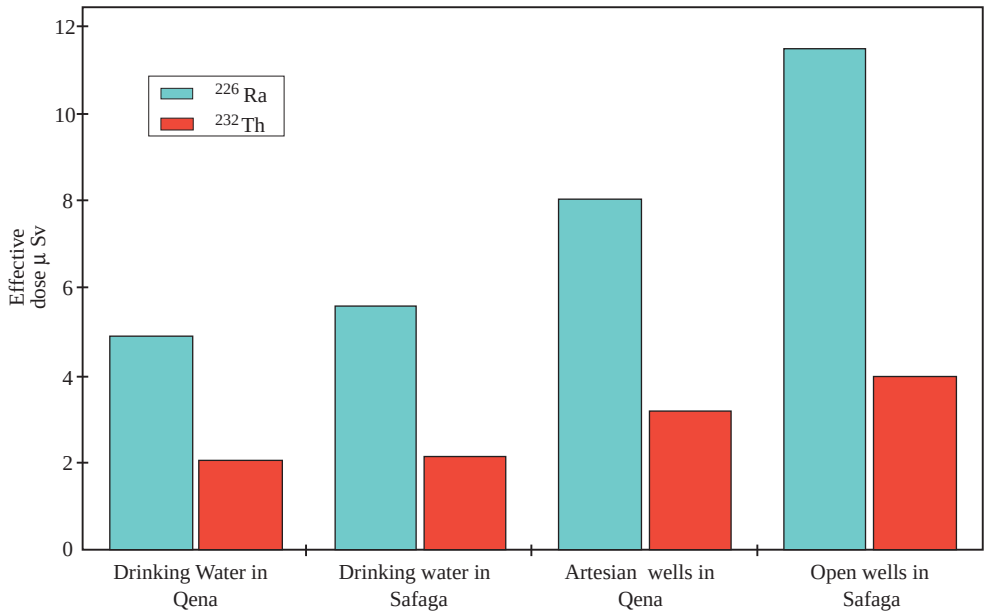

Figure 3. The estimated annual effective dose received by Qena, Safaga and Qusier males as a result of the ^{226}Ra and ^{232}Th ingestion.

Table 4. Daily intake of ^{226}Ra and ^{232}Th of water in Qena and Red Sea regions and the estimated annual effective doses for the present investigation.

Nuclide	Region	Intake (Bq d^{-1}) per person	Ingestion dose coefficient (Sv Bq^{-1})	Annual effective dose mSv y^{-1}
			ICRP 68 (1994)	
^{226}Ra	Qena (tap water)	0.049		0.005
	(groundwater)	0.079		0.0081
	Red Sea (tap water)	0.056	2.8×10^{-7} $(0.2)^a$	0.0057
	(groundwater)	0.113		0.0115
^{232}Th	Qena (tap water)	0.027		0.0022
	(groundwater)	0.041	2.2×10^{-7} $(0.0005)^a$	0.0033
	Red Sea (tap water)	0.029		0.0023
	(groundwater)	0.051		0.0041

a Figures in parentheses are fractional transfer to blood.

Conclusion

The average values of activity concentrations of ^{226}Ra and ^{232}Th radionuclides in drinking water (tap water) in Qena (Upper Egypt) were 1.32 ± 0.7 pCi/l and 0.74 ± 0.44 pCi/l, while those of Safaga and Quseir (Red Sea) were 1.52 ± 0.77 pCi/l and 0.79 ± 0.45 pCi/l, respectively.

The average values of activity concentrations of ^{226}Ra and ^{232}Th radionuclides in groundwater in Qena governorate (artesian wells used for drinking) were 2.14 ± 0.78 pCi/l and 1.1 ± 0.62 pCi/l, while the concentration of these nuclides in water from open wells in Safaga and Quseir (Red Sea, not used for drinking) were 3.05 ± 0.9 pCi/l and 1.39 ± 0.6 pCi/l, respectively.

All types of water (drinking or groundwater) in Qena, Safaga and Quseir are below the maximum allowed values for ^{226}Ra.

The annual effective dose received by Qena and

Safaga and Quseir males as a result of ingestion of this drinking water (tap water) are 0.0072 and 0.0080 mSv and for groundwater are 0.0114 and 0.0156 mSv, respectively.

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