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Research Paper

Natural Radioactivity in Water Samples from Assiut City, Egypt

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Abstract: *The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were determined in water samples. The radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K showed large variations, so the results were classified into two groups (A and B) to facilitate the interpretation of the results. Group A represents samples collected from different locations in Assiut and characterized by low activity concentrations with average values of 0.20, 0.13 and 5.29 Bql^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Group B represents samples mainly collected from the area around Assiut Thermal Power Plant and characterized by very high activity concentrations with average values of 7.68±3.91, 8.07±0.74, and 27.20±1.79 Bql^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. As far as the measured gamma radionuclides is concerned, the mean annual effective doses for analyzed samples of group A are in the range of 0.02–0.08, 0.03-0.17 and 0.03-0.10 mSv yr^{-1} for infants, children and adults, respectively, all being lower than the reference level of the committed effective dose recommended by the WHO, while the mean annual effective doses calculated for samples of group B, all being higher than the reference level of the committed effective dose recommended by the WHO.*

Keywords: Natural radioactivity, Middle Egypt, Ground water, annual effective dose.

1. Introduction

Measurements of natural radioactivity in drinking water have been performed in many parts of the world [1-4], mostly for assessment of the doses and risk resulting from consuming water. The occurrence of natural radionuclides in drinking water poses a problem of health hazard, when these radionuclides are taken to the body by ingestion [5]. Radionuclides in drinking water causes human

internal exposure, caused by the decay of radionuclides taken into the body through ingestion and inhalation indirectly when they are incorporated as part of the human food chain [6].

The radionuclides contributing significantly to the ingestion dose via consumption of water are radium. Radium is a naturally occurring isotope, found in the earth's crust, a member of the uranium ^{238}U decay series. 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 [7-9]. Many salts of radium are soluble in water, and therefore surface, drinking and mineral waters may be enriched in radium and its descendant radon. ^{226}Ra is an earth alkaline element sharing the metabolic pathways of calcium in the human body. Thus, an appreciable fraction of radium is preferentially deposited in bone, the remaining fraction being distributed almost uniformly in soft tissues [10]. Due to their radio-toxicity, especially those of ^{226}Ra, a contamination hazard for human beings exists even at low concentration levels [11].

The levels of concentrations of radionuclides according to nature in ground waters are mainly depend on uranium and thorium-bearing soil and rock mineral or with uranium, thorium and radium deposits. Therefore, the happening and dispensation of natural radioactivity in water depend on the local geological characteristic of the source, soil or rock [12-13].

Potassium is a major element widely distributed in crustal rocks [14]. Thus, potassium occurs in various minerals and clays, from which it may be dissolved through weathering processes and transferred into the liquid phase. ^{40}K decays directly to ^{40}Ca beta emission; it also decays through electron capture to ^{40}Ar [15] followed by a prompt 1.46 MeV gamma emission. As a consequence of water/rock–soil interactions, ^{40}K is released to water bodies, contributing to the presence of radioactive constituents of drinking water.

The present work is the first one aims to suppose the radiological assessment program of Assiut city, with the ultimate aim to establish a baseline map of radioactivity background levels in this area. This study attempts to understand the occurrence and distribution of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in water samples from Assiut city and to assess the concentration of ^{226}Ra, and to estimate the radiation doses received by humans living in this area. The calculated radiation doses are compared to the limits proposed by united nation scientific committee on the effect of atomic radiation.

2. Materials and Methods

2.1 Study Area

Assiut is the largest town in Middle Egypt and lies about 234 miles south of Cairo. The city of Assiut is located at 27°11′00″N, 31°10′00″E and spread across 26,000 km2. It is located between two mountain ranges of about 600 m in height. There is also a rise in altitude from the Mediterranean and the Red Sea towards Middle Egypt. This gives the city and nearby towns and villages the typical properties of a continental climate meaning that the city has harsh and chilly winter weather and very hot but non humid summers.

2.2 Sample Collection and Preparation Techniques

The water samples were collected in standard (1 liter) polyethylene Marinelli beakers, which also used as a measuring container. Before use, the containers were washed with dilute hydrochloric acid and rinsed with distilled water. Each beaker was filled up to brim and a tight cap was pressed on, so that the air was completely removed from it. Samples were acidified by adding 0.5 ml of conc. HNO_3 per liter, to prevent any loss of radium isotopes around the container walls, and to avoid growth of micro- organisms [16]. The samples were stored in the laboratory for a minimum of 1 month to allow daughter products to come into radioactive equilibrium with their parents ^{226}Ra and ^{232}Th before radiometric analysis.

2.3 Gamma-Ray Detection System

Each sample was measured with a gamma-ray spectrometer consisting of a NaI(Tl) detector and multichannel analyzer of 8192 channel, with the following specifications: Resolution (FWHM) at 1.33 MeV ^{60}Co is 60 keV – relative efficiency at 1.33 MeV ^{60}Co is 7.5 %. The detector is shielded in a chamber of two layers starting with stainless steel (10 mm thick) and lead (30 mm thick). This shield serves to reduce different background radioactivity.

The system was calibrated for energy and efficiency. The spectrometer was calibrated for efficiency and energy using multinuclide standard solution (QCY48) PTB (Germany). The standard source peaked in the same geometry as that used for measured samples. For calibration, the standard source is placed above the detector, and the measurement started. The dependence of the efficiency on the radiation energy was determined at 0.0 mm sample detector distance. The absolute efficiency of the NaI(Tl) detector was determined using the standard solution QCY48 PTB (Germany) The detector efficiency decreases continuously with energy. The dependence of the efficiency on the volume of the sample was determined by aMarinelli beaker (1 liter). It can be noticed that the detector efficiency decreases with the volume of the sample in the energy range of interest [17].

To minimize the effect of scattered radiation from the shield, the detector is located in the center of the chamber. The sample was placed above the detector and measured for at least 24 h. The spectra were either evaluated with computer software program Maestro (EG&G ORTEC), or manually using a spread sheet (Microsoft Excel) to calculate the natural radioactivity. ^{226}Ra activity of the samples was determined via its daughters (^{214}Pb and ^{214}Bi) through the intensity of the 295.22, 351.93 keV, for ^{214}Pb Gamma-lines and 609.31, 1120, 1764.49 keV, for ^{214}Bi Gamma-lines. ^{232}Th activity was determined from the daughters (^{228}Ac), (^{212}Pb) and (^{208}Ti) through the intensity of 209.25, 338.32, 911.2 keV Gamma-lines for (^{228}Ac), (^{212}Pb) emissions at 238.63 keV and (^{208}Ti) emissions at 583.19, 2614 keV Gamma-lines. ^{40}K activity determined from the 1460.7 keV Gamma-line.

3. Results and Discussion

3.1 Activity Concentrations

The measured activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K show much variation ranging from small values to high values, such that the results were classified into two group (A and B) to facilitate the interpretation of the results. The samples of group A were collected from different locations in Assiut and were characterized by low activity concentrations (Table 1). The samples of group B were collected from the vicinity of Assiut Thermal Power Plant (ATPP) and were characterized by high activity concentrations (Table 2). Table 1 shows that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K ranged from 0.07±0.05 to 0.54±0.35 Bql^{-1}with an average of 0.20±0.11 Bql^{-1}, from 0.05±0.04 to 0.5±0.28 Bql^{-1}with an average of 0.13±0.08 Bql^{-1}, and from 3.25±2.09 to 8.72±4.32 Bql^{-1}with an average of 5.29±2.41Bql^{-1}, respectively.

Table 1 shows that in all samples except (Sa1 and Sa6), the concentrations of ^{226}Ra is higher than that of ^{232}Th and this reflects the fact that radium is more soluble in groundwater than its thorium and uranium precursors, and its solubility is enhanced by: the common-ion effect (when dissolved solids are high), an oxygen-poor environment, and the fragmentation of uranium-bearing minerals [17]. Samples Sa1and Sa6 have not the same symmetry like the rest of samples. This may be explained by the different origins of these water, these waters come from different depths and pass through different geological layers.

Table 1: The activity concentration in Bql^{-1} of water samples of group A

Sample No.	Activity concentration in Bql^{-1}		
	^{226}Ra	^{232}Th	^{40}K
Sa1	0.17±0.09	0.37±0.23	5.06±3.42
Sa2	0.31±0.12	0.13±0.07	4.19±2.33
Sa3	0.23±0.15	0.08±0.07	5.73±1.97
Sa4	0.14±0.11	0.07±0.05	5.42±2.50
Sa5	0.54±0.35	0.10±0.08	6.55±1.93
Sa6	0.29±0.16	0.50±0.28	8.72±4.32
Sa7	0.22±0.13	0.12±0.09	3.25±2.09
Sa8	0.13±0.06	0.12±0.06	3.51±1.09
Sa9	0.32±0.12	0.11±0.05	3.71±1.39
Sa10	0.13±0.10	0.07±0.03	7.21±2.32
Sa11	0.13±0.07	0.06±0.03	6.02±1.90
Sa12	0.10±0.04	0.08±0.06	4.43±2.36
Sa13	0.07±0.05	0.05±0.04	5.20±2.35
Sa14	0.09±0.02	0.07±0.03	7.07±4.06
Sa15	0.19±0.12	0.08±0.07	3.39±2.13
Average	0.20±0.11	0.13±0.08	5.29±2.41

Table 1 shows variations in radionuclide concentrations in ground waters from one sample to another; these variations depend on the minerals derived from aquifer rocks. The abundance of ^{40}K activity observed in all samples, may be due to agricultural activities going on in the area that involve the use of potassium fertilizers which may have been transported to the groundwater, given that ^{40}K is a highly soluble element [17].

The highest activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in water samples are presented in table 2 (group B). These samples were collected from the pond (1 & 2) and from groundwater wells (3 - 6) which are quite close to ATPP and are used for agricultural purposes, the pond is mainly used for collecting washing water (discharge water) from ATPP where ash from the boiler and ash removed by the precipitations are flushed with water to the pond [18]. As seen, the concentrations of ^{226}Ra, ^{232}Th and ^{40}K vary from 1.05 to 12.68 Bq l^{-1}, from 1.47 to 9.91 Bq l^{-1} and from 8.09 to 55.16 Bq l^{-1} respectively. The values of ^{226}Ra in groundwater samples are much higher than the maximum contaminant levels of 1.85 mBq l^{-1} proposed in the EPA [19]. The measured concentrations of ^{226}Ra and ^{232}Th evidently exceed the values of the River Nile water and groundwater in Upper Egypt which are 0.05, 0.03 Bq l^{-1} for the River Nile water and 0.12, 0.05 Bq l^{-1} for groundwater [20], respectively. These results apparently confirm the presence of actual harmful impacts of the plant on the surrounding aquatic environment [18].

Table 2: Activity concentrations (Bq l^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K of water samples of group B

Sample No.	^{226}Ra	^{232}Th	^{40}K
1	12.68± 6.79	5.00 ± 0.19	52.45± 4.15
2	7.37 ± 5.70	1.47 ± 0.26	29.83 ±1.72
3	6.24± 2.90	9.91 ± 0.78	19.82± 1.91
4	5.05± 4.31	3.08 ± 0.42	55.16± 4.00
5	4.43 ±1.59	6.37 ± 0.79	8.09 ± 7.71
6	1.05±0.91	2.99 ± 0.52	47.17 3.82

3.2 Radiation Dose Estimation

The annual effective doses have been calculated according to the equation introduced by EPA [19] and by Meltem and Gursel [6].

$$DR_W = A_W \times IR_W \times ID_F$$

where DR_W is the effective dose (mSv year^{-1}), A_W is the activity (Bql^{-1}), IR_W is the intake of water for person in one year and ID_F is the effective dose equivalent conversion factor (mSv Bq^{-1}).

Several factors must be addressed for proper dose evaluation due to the radionuclides in drinking water, these factors are: the weighted average activity concentration in (Bq l^{-1}), the amount of water intake by person in one year, the dose coefficient for radionuclides ingestion by humans (mSv Bq^{-1}). Doses were estimated by considering a consumption rate (150, 350 and 500 l yr^{-1}) for infants, children and adults, respectively. The conversion factors for ^{226}Ra, ^{232}Th and ^{40}K as reported by ICRP [20], IAEA [21] and WHO [22] are (9.6×10^{-7}, 4.5×10^{-7} and 5×10^{-9} Sv Bq^{-1}) for infants, (8×10^{-7} , 2.9×10^{-7} and 5×10^{-9} Sv Bq^{-1}) for children and (2.8×10^{-7}, 2.3×10^{-7} and 5×10^{-9} Sv Bq^{-1}) for adults.

Table 2: Estimates of annual effective doses in mSv yr^{-1} due to ingestion of ^{226}Ra, ^{232}Th and ^{40}K for different age groups and due to the total ingestions for group A

Sample code	^{226}Ra			^{232}Th			^{40}K			Total doses		
	Infants	Children	adults	Infants	children	adults	Infants	children	adults	Infants	children	adults
Sa1	0.024	0.047	0.042	0.024	0.037	0.042	0.003	0.008	0.012	0.053	0.094	0.079
Sa2	0.045	0.087	0.014	0.008	0.013	0.014	0.003	0.007	0.010	0.057	0.108	0.069
Sa3	0.033	0.064	0.009	0.005	0.008	0.009	0.004	0.010	0.014	0.042	0.082	0.055
Sa4	0.020	0.039	0.008	0.004	0.007	0.008	0.004	0.009	0.013	0.028	0.055	0.041
Sa5	0.077	0.151	0.011	0.006	0.010	0.011	0.004	0.011	0.016	0.089	0.172	0.103
Sa6	0.041	0.081	0.057	0.033	0.050	0.057	0.006	0.015	0.021	0.082	0.147	0.119
Sa7	0.031	0.061	0.013	0.008	0.012	0.013	0.002	0.005	0.008	0.042	0.079	0.052
Sa8	0.018	0.036	0.013	0.008	0.012	0.013	0.002	0.006	0.008	0.029	0.054	0.040
Sa9	0.046	0.089	0.012	0.007	0.011	0.012	0.002	0.006	0.009	0.056	0.107	0.066
Sa10	0.018	0.036	0.008	0.004	0.007	0.008	0.005	0.012	0.018	0.028	0.056	0.044
Sa11	0.018	0.035	0.006	0.004	0.006	0.006	0.004	0.010	0.015	0.026	0.051	0.039
Sa12	0.014	0.028	0.009	0.005	0.008	0.009	0.003	0.007	0.011	0.023	0.043	0.034
Sa13	0.010	0.0196	0.005	0.003	0.005	0.005	0.003	0.009	0.013	0.017	0.033	0.028
Sa14	0.012	0.025	0.008	0.004	0.007	0.008	0.005	0.012	0.017	0.022	0.044	0.038
Sa15	0.027	0.053	0.009	0.005	0.008	0.009	0.002	0.005	0.008	0.035	0.067	0.044

The calculated effective doses for different age groups infants, children and adults are presented in table 2. It should be noted that doses were ranged from 0.02–0.08 mSv yr $^{-1}$ for infants, 0.03-0.17 mSv yr $^{-1}$ for children and 0.03-0.10 mSv yr $^{-1}$ for adults. Fig. 1 shows that doses received by children are higher than that received by infants and adults and doses received by adults are higher than that received by infants.

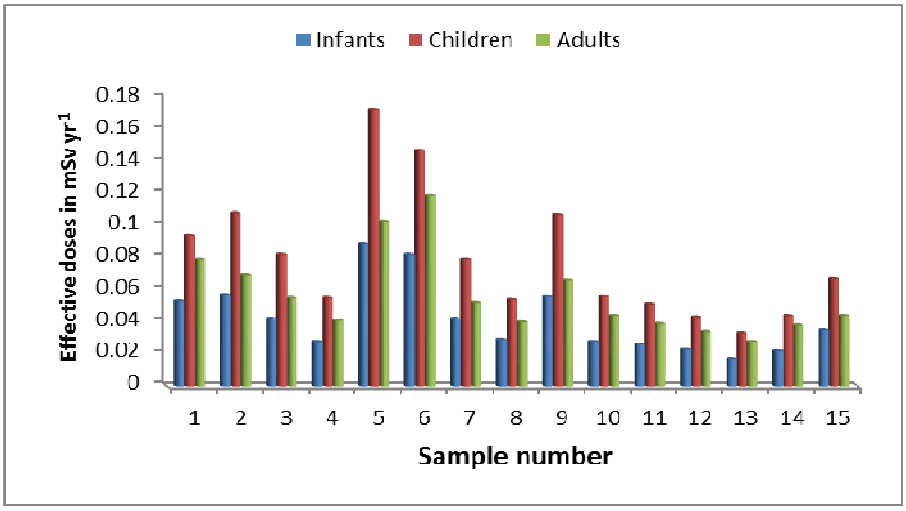

Fig. 1: The mean annual effective doses for infants, children and adults in mSv yr^{-1} due to the total ingestions for group A

The doses obtained in our study for group A are much lower than the recommended reference level of 0.26, 0.2 and 0.1 mSv yr^{-1} for effective doses for infants, children and adults, respectively which published by IAEA [22], WHO [23] and UNSCEAR [24], from one year consumption of drinking water, and consequently, it can be recommended that, the investigated waters are suitable for life-long human consumption.

The calculated effective doses for different age groups: infants, children and adults for group B are shown in Fig. 2. It should be noted that the doses ranged from 0.54 to 3.57 mSv yr^{-1} for infants, 1.03 to 6.59 mSv yr^{-1} for children and 0.88 to 4.22 mSv yr^{-1} for adults. According to the recommended reference level of 0.26, 0.2 and 0.1 mSv yr^{-1} for effective dose for infants, children and adults respectively, published by IAEA [22] and UNSCEAR [24] doses obtained from one year of consumption of drinking water are much higher than the recommended reference level. Consequently, it can be recommended that the investigated water is not acceptable for healthy life-long human consumption and a reduction in consumption or radionuclide concentration is essential.

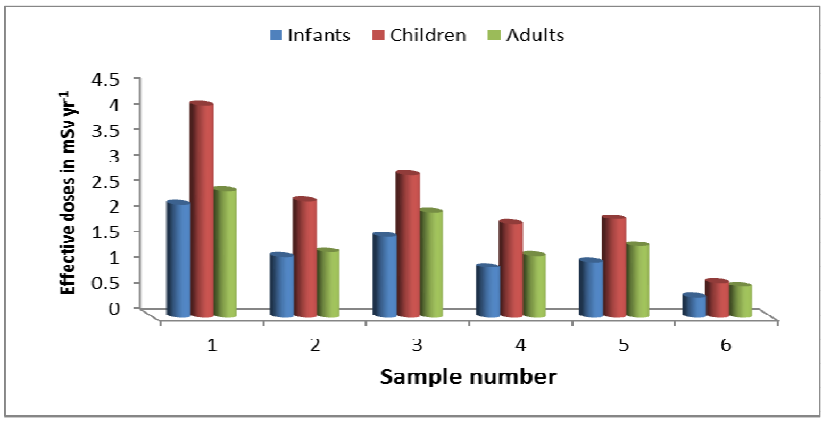

Fig. 2: The effective doses for infants, children and adults in mSv yr^{-1} due to the total ingestions of water samples of group B

4. Comparison of Results with Similar in Other Countries

The values of ^{226}Ra, ^{232}Th and ^{40}K concentrations from the present work (group A) are compared with those from other countries and they listed in table 3. ^{226}Ra values from the present work are higher

than that reported in Egypt [20] and in Sudan [25], they are lower than that reported in Yemen [17], Brazil [26], China [27], Sweden [28] and Finland [29]. The activity concentrations of ^{232}Th are lower than that reported in Yemen [17] and higher than the concentrations reported from other countries. The values of ^{40}K concentrations presented in this work seem to be much higher than that reported from other countries.

The values of ^{226}Ra, ^{232}Th and ^{40} K concentrations of group B are much higher than that in other countries by several orders of magnitude (Table 3).

Table 3: The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bql^{-1} of the investigated samples in comparison with other countries

Country	Activity concentration in Bql^{-1}			Reference
	^{226}Ra	^{232}Th	^{40}K	
Egypt(A)	Mean 0.20	Mean 0.13	Mean 5.29	Present work
Egypt(B)	1.83 - 18.53	1.47 -12.78	8.09 - 64.48	Present work
Yemen	2.01-6.55	1.07-2.93		[17]
Egypt(Qena)	Mean 0.08	Mean 0.04		[20]
Sudan	0.007-0.014	0.001-0.039		[25]
Brazil	0.01-3.79			[26]
China	Max 0.93			[27]
Sweden	0.016-4.9			[28]
Finland	0.01-49			[29]
Denmark	0.55			[30]
Poland	0.001-0.049			[31]

5. Conclusions

The natural radioactivity levels of ^{226}Ra, ^{232}Th and ^{40}K have been measured in water samples from Assiut, Egypt using gamma ray spectroscopy. The obtained results were classified into two groups according to the large variation in the activity concentrations. The first group showed low activity concentrations, and the total effective doses due to all radionuclides are 0.04, 0.07 and 0.06 for infants, children and adults respectively, which are 15%, 35% and 60% of the values of 0.26, 0.2 and 0.1 mSv for the recommended reference level of committed effective dose from 1 year's consumption of drinking water for infants, children and adults, respectively. From this one can recommend that, the investigated water of the first group (A) is acceptable for life-long human consumption.

The second group describes the high activity concentrations of water samples collected from the area around Assiut thermal power plant. All calculated annual effective doses of this group are exceeded the international recommended values by several orders of magnitude leading to significant health hazards in these areas.

The high activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K observed in water samples explain the high impact of water infiltration from the pond to the underground water in the surrounding areas and cautionary measures must be considered when using this water in agricultural irrigation.

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