

Spatial Distribution of the Natural Radioactivity of the Water and its Association with the Physico-Chemical Parameters: Abu Tig, Assiut, Egypt



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Abstract

Activity concentrations of radionuclides (^{226}Ra , ^{222}Rn , ^{232}Th and ^{40}K) and physico-chemical parameters (Temperature, pH-value and total dissolved solids (TDS)) were measured for drinking water samples collected from Abu Tig, Assiut Governorate, Egypt. The samples were measured by gamma-ray spectrometry technique using HPGe detector with specially designed shield. The average activity concentrations were $(235.71 \pm 34.04\text{mBq l}^{-1})$, $(58.26 \pm 10.29\text{mBq l}^{-1})$, $(624.08 \pm 69.29\text{mBq l}^{-1})$ and $(0.184 \pm 0.11\text{Bq l}^{-1})$ for ^{226}Ra , ^{232}Th , ^{40}K and ^{222}Rn respectively. The average annual effective doses due to ingestion of these waters were $60.79\mu\text{Sv yr}^{-1}$, $74.75\mu\text{Sv yr}^{-1}$, and $41.81\mu\text{Sv yr}^{-1}$ for different age groups adults, children and infants, respectively. There is no correlation between radionuclides and physico-chemical parameters. The obtained results were lower than the reference level of the committed effective dose recommended value (0.1mSv yr^{-1}) as reported by World Health Organization (WHO), indicating to the safe use of the examined water.

Keywords: Natural Radioactivity; Annual effective dose; Drinking water; Abu Tig

Introduction

Most environmental studies provide the best ways to preserve water and improve its quality for the necessity of human life. Water may be a factor for the transport of pollutants by human consumption. The

radionuclides cause health hazard by ingestion and through the human food chain when these radionuclides are taken to the body [1]. So that it is necessary to achieve the minimum radiation exposure to the human body and not skipped the reference dose level (RDL) of the effective annual dose of drinking water consumption of 0.1mSv yr^{-1} reported by WHO [2].

The main naturally occurring radioactive isotopes in water are uranium isotopes, radium isotopes, and radon isotopes, these radionuclides are mainly soluble in water [3]. The production of these radionuclides in groundwater is mainly from rock-water interactions. The levels of radioactivity in groundwater depends on many factors such as the concentrations of these radionuclides in the aquifer rocks, geomorphology, and lithology other ecological conditions of aquifer [4,5], depending also on chemical reactions and the physical processes of decay along the water-rock interface.

^{226}Ra is an alpha emitter with longest a half-life of 1602yr, it originates from the ^{238}U decay series and its descendant ^{222}Rn has a half-life of 3.8d. ^{228}Ra is a beta emitter with a half-life of 5.75yr and it originates from ^{232}Th series. ^{40}K is also radioactive and has a half-life of 1.3×10^9 yr. Generally, radium concentrations in surface water range from 0.01 to 0.1Bq l^{-1} [6], while its concentration in groundwater can reach values up to 38Bq l^{-1} , depending on the factors mentioned previously. ^{222}Rn is produced which is a gaseous radioisotope and diffuses into the pore water of rock formation. The ^{222}Rn concentration in groundwater reaches to 183Bq l^{-1} [7]. The recommended reference level of radon concentration in tap water is 150Bq l^{-1} as considered by [8].

The levels of detection of radon in water research are explained by the sovereignty of granite rocks as well as the rocks of aquifers under investigation. Increased risk of many types of cancer is associated with exposure to radon and its progeny [9]. In addition, the high concentrations of ^{222}Rn in water samples indicate the presence of the radon's parent nuclide ^{226}Ra in the water-rock/soil system, which is known as the health hazard when ingestion during drinking water [10].

The present study attempts to report the results of drinking water radium, thorium, potassium and radon survey in Abu Tig, Assiut Governorate, Egypt. The inhabitants of villages and rural areas in the study area are totally dependent on groundwater and on surface water of the Nile River for water supply.

The present work aims to understand the occurrence and classification of natural radionuclides ^{226}Ra , ^{232}Th , ^{40}K and ^{222}Rn in drinking water samples from study area and in a first approach to estimate natural radionuclides exposure levels for inhabitants of this region to estimate radiation doses for several ages due to water consumption by people living in this region. The results might provide some helpful information for management of drinking water use.

Materials and Methods

Description of study areas

Assiut Governorate is the most populated governorate in Upper Egypt with more than 5 million inhabitants. Abu Tig is a big district within Assiut Governorate that located on the western bank of the river Nile, between Latitudes $26^{\circ} 57' 56.8''$ and $27^{\circ} 6' 59.6''$ N and Longitudes $31^{\circ} 11' 52.9''$ and $31^{\circ} 22' 10.7''$ E. Abu Tig has a surface area of about 129km^2 . It has almost 264,087 inhabitants. Physically, it is bounded to the east by the River Nile and bounded on the western side by the Eocene limestone plateau. The drinking water samples were collected from the area under interest in the period of normal water flow rates within the Nile (Figure 1).

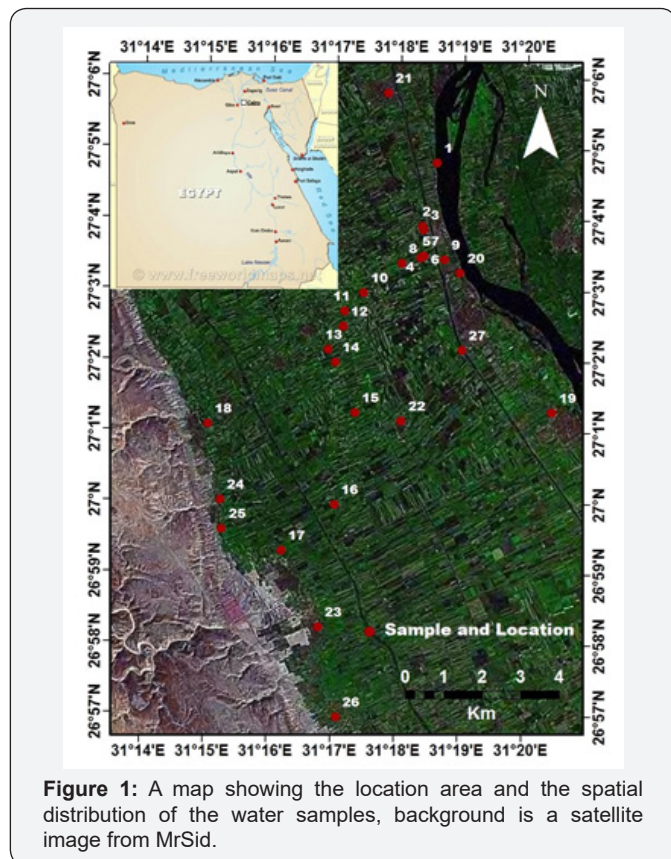


Figure 1: A map showing the location area and the spatial distribution of the water samples, background is a satellite image from MrSid.

Sampling and sample preparation

A total of twenty-seven samples were collected from various locations from the study region and classified into three types; surface water, hand-dug wells and deep wells. Standard Polyethylene Marinelli beakers (1 liter) are used as measuring containers. Before utilizing, the containers are washed with HCl and swilled with distilled water. A tad bit of nitric acid, around 0.5ml HNO_3 per liter, was added to clear solution to forbid any loss of radium isotopes around the container walls, and to avert growth of microorganisms [11]. After filling up the beaker to the brim, a tight cap is pressed on to completely remove the air from it. The samples store for over thirty days to arrive secular equilibrium before a radiometric analysis. Each sample is counted for almost 172800 second relying on the radionuclide's concentrations.

Gamma spectrometry

Water samples were subjected to a gamma ray spectrometer with a detector had closed-end coaxial Gamma-ray crystal made of high purity germanium in a vertical configuration cooled with liquid nitrogen. The energy resolution of the detector reads approximately 2.000keV and $\leq 0.925\text{keV}$ at 1.33MeV and 122keV, respectively, while the relative efficiency is 40%. The germanium crystal is existed inside a lead shield to decrease the environmental background. The shield consists of four layers with the following specifications: a low carbon steel of 9.5mm thick as an outer jacket, a bulk shield of lead of 10cm thickness, and graded linings to absorb low energy X-rays of 1.0mm tin and 1.6mm copper [12].

The spectrometer was energy-calibrated using radioactive standards of known energies such as ^{137}Cs (662keV) and ^{60}Co (1172 and 1332keV) and it was calibrated for efficiency using Canberra's ISOCS calibration utility instead of standard source [12].

^{226}Ra concentration was determined using gamma-lines of ^{214}Bi and ^{214}Pb for different energy (609.31, 1120.29, and 1764.49KeV) and (295.22 and 351.93KeV) respectively. The concentration of ^{232}Th was measured using gamma lines of ^{228}Ac , ^{212}Pb and ^{208}Tl for different energy (911.2 and 968.97KeV), (238.63KeV) and (583.19 and 2614KeV) respectively. Finally, ^{40}K concentration was determined one peak at 1460.8KeV.

Dose calculation

To calculate possibility health hazards, the effective dose radiation, DR (mSv/yr), doses arising from the ingestion of these waters were assessed using following equation [8,13],

$$DR = A \times IR \times ID \quad \text{-----(1)}$$

Where A is the activity (Bq/L), ID is effective dose equivalent conversion factor (mSv/Bq), and IR is the ingestion of water for a person in a year (L/yr). The conversion factors for infants (9.6×10^{-4} , 4.5×10^{-4} and $4.2 \times 10^{-5}\text{mSv/Bq}$), for children (8×10^{-4} , 2.9×10^{-4} and $1.3 \times 10^{-5}\text{mSv/Bq}$) and for adults (2.8×10^{-4} , 2.3×10^{-4} and $6.2 \times 10^{-6}\text{mSv/Bq}$), were utilized for ^{226}Ra , ^{232}Th and ^{40}K

respectively as reported by IAEA, ICRP, and WHO [14-16]. The dose was estimated by knowing consumption rate for adults, children and infants of 730, 350, and 150L/yr respectively.

Results

Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K for the drinking water samples collected from various locations in Abu Tig region

are listed in Table 1 and shown in Figure 2. The concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K changed from 59.04 ± 8.32 to 421.62 ± 58.11mBq/L, from 15.58 ± 2.62 to 153.07 ± 33.64mBq/L and from 103.27 ± 11.48 to 1363.72 ± 150.98mBq/L, respectively. The average measurements are 235.71 ± 34.04mBq/L, 58.26 ± 10.29mBq/L and 624.08 ± 69.29mBq/L for ²²⁶Ra, ²³²Th and ⁴⁰K, respectively.

Table 1: Activity concentrations of radionuclides and physico-chemical parameters for water samples in study area.

Sample No.	Coordinates (dms)		Physico-Chemical Parameters			Activity Concentrations			
	N	E	pH	TDS (ppt)	Temp (°c)	²²⁶ Ra(mBqL ⁻¹)	²³² Th(mBqL ⁻¹)	⁴⁰ K(mBqL ⁻¹)	²²² Rn (BqL ⁻¹)
S1	27°4' 48.16"	31° 18' 35"	8.76	0.15	26.6	257.31±37.33	65.76±8.62	419.1±46.60	0.789±0.34
S2	27° 3' 54.18"	31°18' 21.72"	7.85	0.43	26.6	243.47±35.62	43.22±5.81	538.84±59.78	0.470±0.23
S3	27° 3' 51.11"	31°18' 23.21"	8.05	0.44	24	165.36±23.60	153.07±33.64	984.54±109.29	0.289±0.14
S4	27°3' 27.6"	31°18' 20.82"	7.82	0.37	24.9	203.54±28.23	40.53±6.38	691.84±76.78	0.208±0.12
S5	27°3' 29.1"	31°18' 23.64"	7.76	0.37	26.6	234.96±34.93	104.45±22.70	765.02±84.94	0.184±0.11
S6	27°3' 29.1"	31°18' 23.64"	7.91	0.38	25	294.52±41.48	60.99±8.64	1144.19±126.65	0.146±0.10
S7	27° 3' 29.1"	31°18' 23.64"	7.71	0.45	24.3	177.39±24.80	40.20±5.62	571.74±63.59	0.141±0.06
S8	27°3' 22.8"	31°18' 2.76"	7.78	0.45	24.9	273.93±39.61	43.38±5.71	691.84±76.78	0.109±0.09
S9	27°3' 26.1"	31° 8' 43.38"	7.36	0.57	26	78.93±10.86	55.70±11.09	178.64±19.93	0.086±0.05
S10	27°2' 57.36"	31°17' 27.12"	7.49	0.95	25.5	164.44±24.35	65.01±10.04	713.51±79.20	0.165±0.11
S11	27°2' 41.46"	31°17' 9.12"	7.86	0.15	25.8	421.62±58.11	47.86±6.37	440.70±49.02	0.081±0.05
S12	27° 2' 28.86"	31°17' 8.64"	7.21	0.75	24.7	275.5±39.69	71.60±15.07	738.41±81.87	0.120±0.09
S13	27° 2' 9"	31°16' 54.54"	7.34	0.98	24.9	186.62±26.70	38.49±5.34	192.92±21.50	0.115±0.08
S14	27°1' 58.02"	31° 17' 1.2"	7.53	0.98	24.2	214.39±30.46	25.83±4.49	412.44±45.93	0.154±0.07
S15	27°1' 15.42"	31°17' 20.46"	7.78	0.67	26.5	59.04±8.32	19.88±3.08	103.27±11.48	0.146±0.10
S16	26°59'57.72"	31°17' 1.98"	7.61	0.71	25.5	134.11±18.1	15.58±2.62	327.47±36.31	0.120±0.10
S17	26°59'18.18"	31°16' 12.36"	7.3	0.38	25.1	121.61±17.64	28.40±6.05	518.33±57.33	0.187±0.12
S18	27°1' 5.3"	31°15' 1.6"	7.72	0.63	23.7	298.82±43.06	50.60±10.38	172.96±19.30	0.159±0.11
S19	27°1' 17.1"	31°20' 26"	7.98	0.32	25.6	213.29±30.02	77.04±10.84	126.39±14.08	0.130±0.10
S20	27°3' 14.9"	31°18' 57.9"	8.13	0.22	24.6	237.43±34.47	52.36±6.81	798.28±88.55	0.116±0.10
S21	27°5' 47.1"	31°17' 47.9"	7.89	0.28	25.9	273.16±39.23	39.42±4.09	312.66±34.84	0.180±0.08
S22	27°1' 8.8"	31° 18' 4"	7.95	0.28	22.9	145.92±22.27	42.73±7.06	878.1±98.37	0.204±0.12
S23	26° 58' 13.8"	31°16' 48.2"	7.89	0.38	23.1	287.57±42.52	71.35±13.89	1363.72±150.98	0.135±0.06
S24	27°0' 0.9"	31° 15' 14.2"	8.46	0.16	18.2	319.86±49.73	63.99±13.36	665.23±73.99	0.146±0.10
S25	26°59' 36.2"	31° 15' 16"	8.05	0.22	25.5	369.09±51.92	69.23±12.63	798.28±88.63	0.122±0.10
S26	26°56' 57.7"	31°17' 5.8"	7.91	0.25	26.2	298.33±44.46	36.07±4.8	1270±140.80	0.104±0.06
S27	27°2' 9.4"	31°19' 0.3"	8.39	0.09	19.9	413.98±61.69	150.18±32.63	1031.11±114.29	0.156±0.11
Min						59.04±8.32	15.58±2.62	103.27±11.48	0.081±0.05
Max						421.62±58.11	153.07±33.64	1363.72±150.98	0.789±0.34
Average value						235.71 ± 34.04	58.26 ± 10.29	624.08 ± 69.29	0.184±0.11

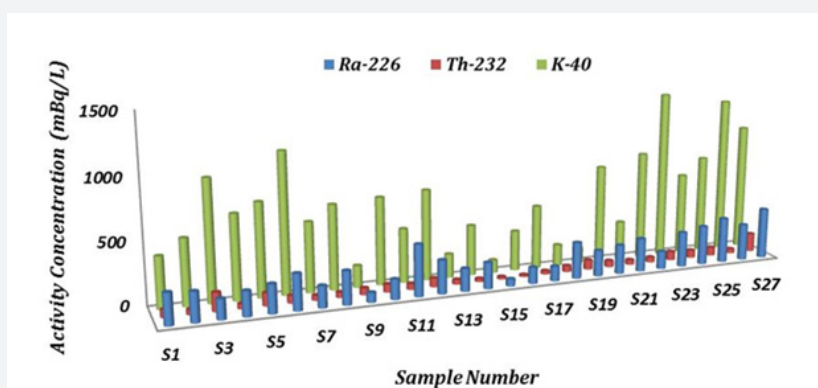


Figure 2: Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K of water samples in Abu Tig region.

According to the data, ^{226}Ra concentration in most drinking water samples is very higher than the maximum contaminant levels (MCL) of 0.185Bq/L proposed in the USA [9] for drinking water. In addition, the current work is compared to a number of researches in different regions of the world which have lower

values, such as reported in Egypt [17], Sudan [18], Malaysia [19], Jordan [20] and Ghana [21], as well as those with higher values as reported in Yemen [22] and Nigeria [23]. Table 2 includes a comparison of ^{232}Th and ^{40}K concentrations with Different values around the world.

Table 2: The activity concentration (Bq/L) of drinking water samples in the present investigation compared to other regions in the world.

Country	Type Water	Activity Concentration in Bq/L				Reference
		^{226}Ra	^{232}Th	^{40}K	^{222}Rn	
Egypt (Abu Tig)	Drinking water	0.059-0.422	0.0156-0.1531	0.103-1.363	0.081-0.789	Present work
		Mean 0.236	Mean 0.058	Mean 0.624	Mean 0.184	
Egypt (Qena)	Groundwater	Mean 0.08	Mean 0.04	—	—	(Ahmed, 2004)
Egypt (Qena)	Tap water	—	—	—	0.0061-0.118	(Abbady, et al. 1995)
		—	—	—	Mean 0.077	
Yemen (Jubani)	Groundwater	2.25-3.45	0.3-1.43	26.73-43.7	—	(El-Mageed, et al. 2013)
Sudan	Groundwater	0.007-0.014	0.001-0.039	—	—	(osman, et al.2008)
Santos region, Brazil	Surface water	—	—	—	0.4-2.4	(Marques, et al.2004)
China	Groundwater	Max 0.93	—	—	—	(Zhuo, et al. 2001)
Sweden	Groundwater	0.016-4.9	—	—	—	(Saleh, et al. 2002)
Malaysia	Drinking water	0.0182	0.0389	1.22	—	(Yussuf et al. 2012)
Ghana	Surface water	Mean 0.0137	Mean 0.0012	—	—	(Kpeglo et al.2014)
Nigeria (Lakes)	Surface water	Mean 12	Mean 12	Mean 97	—	(Agbalagba & onoja, 2011)
Malaysia (Panang)	—	—	—	—	0.580-2.540	(Muhammed, et al. 2012)
Jordan (Irbid)	Tap water	0.019-0.302	0.024-0.119	0.101-0.342	—	(Al-Shboul et al.2017)
Ghana (Adentan)	Groundwater	—	—	—	0.037-0.673	(Nguelem, et al.2013)
		—	—	—	Mean 0.392	

There is no clear correlation between ^{226}Ra , ^{232}Th and ^{40}K in this study region, where a weak correlation can be found between (^{226}Ra and ^{232}Th), (^{226}Ra and ^{40}K) and (^{232}Th and ^{40}K) with a correlation coefficient of $R^2 = 0.1156, 0.1551$ and 0.1794 , respectively.

^{222}Rn activity concentration in drinking water samples fluctuated between 0.081 ± 0.05 to 0.789 ± 0.34 Bq/L with an average value of 0.184 ± 0.11 . All the samples are below the

maximum contaminant level (MCL) of 11.1 BqL^{-1} [24] and 11 BqL^{-1} as proposed by the US Environmental Protection Agency [25] the concentration of radon changes depending on the zone resulting from its geological structure. as well as, the depth of the source of water, do not rule out climate change and geo-hydrological processes occurring in the region [26]. ^{222}Rn concentration in present work is greater than that reported in Egypt [27] and less than that reported from other countries, as Table 2.

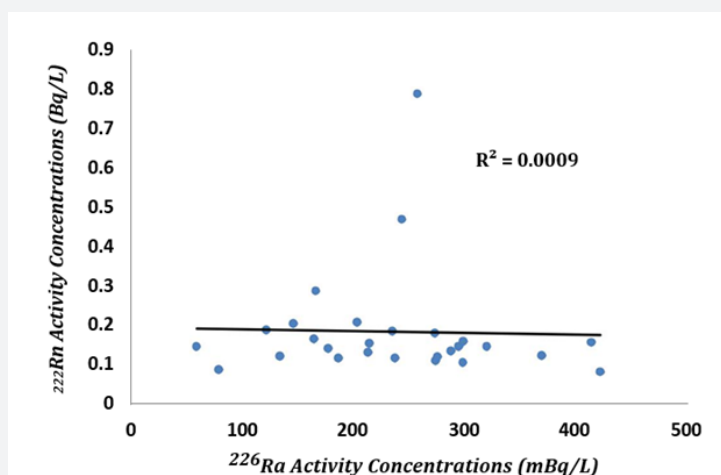


Figure 3: Correlation between the ^{222}Rn and ^{226}Ra concentrations for drinking water samples in study region.

^{222}Rn and ^{226}Ra activity concentrations are listed in Table 1. There is no correlation between them where the correlation coefficients $R^2 = 0.0009$ as shown in Figure 3.

Physico-chemical properties of drinking water samples in study region are listed in Table 1. TDS, pH-value and water temperature ranged from 0.09 to 0.98ppt, from 7.21 to 8.76 and from 18.2 to 26.6°C, respectively. There is no correlation between

physico-chemical parameters and radionuclides ^{226}Ra , ^{232}Th , ^{40}K and ^{222}Rn where the correlation coefficients R^2 in all cases are less than 0.2 as shown in Figure 4, except a moderate correlation was observed between ^{226}Ra activity concentration and pH-value and TDS with correlation coefficient $R^2 = 0.2159$ and 0.2667 respectively. Also, between ^{222}Rn activity concentration and pH-value with correlation coefficient $R^2 = 0.2527$.

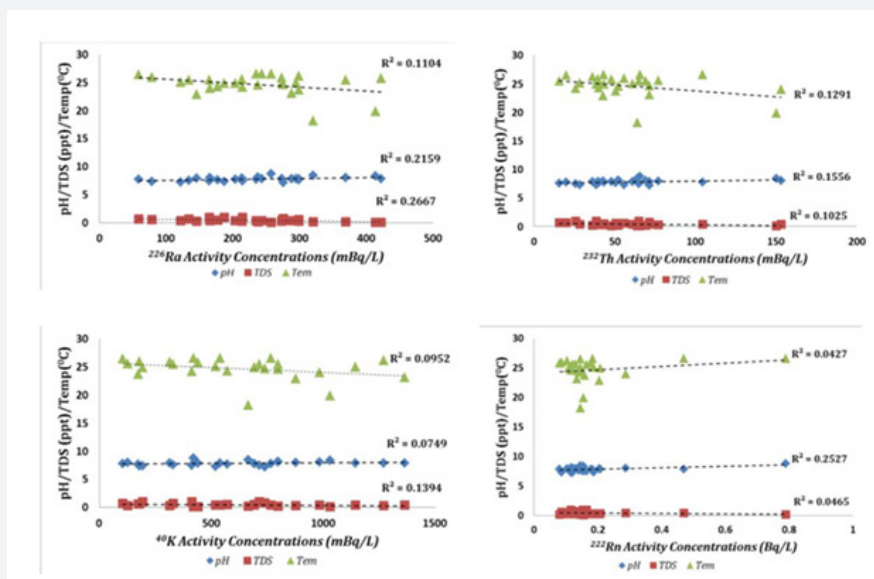


Figure 4: Correlation between ^{226}Ra , ^{232}Th , ^{40}K and ^{222}Rn Activity concentrations and the physio-chemical properties for drinking water in study region.

The Annual Effective Dose

The annual effective doses are given in Table 3 for various ages: adults, children and infants due to ingestion of ^{226}Ra , ^{232}Th and ^{40}K through drinking water from different areas in study region. The annual effective dose fluctuated between 10.49 to 76.25 μSvyr^{-1} with an average value of 41.81 μSvyr^{-1} , between 19.02 to 135.85 μSvyr^{-1} with an average value of 74.75 μSvyr^{-1} and between 15.87 to 114.50 μSvyr^{-1} with an average value of

60.79 μSvyr^{-1} for infants, children and adults respectively. Figure 5 shows that doses received by children were higher than that received by infants and adults; this is because children have small bodies. They are in the developmental stage, because their organs and bones are not yet complete. Therefore, the dose which they are exposed to is large compared to adults. In addition to being shorter than adults, they may get a higher dose of radioactive distributed in and deposited on the ground [28].

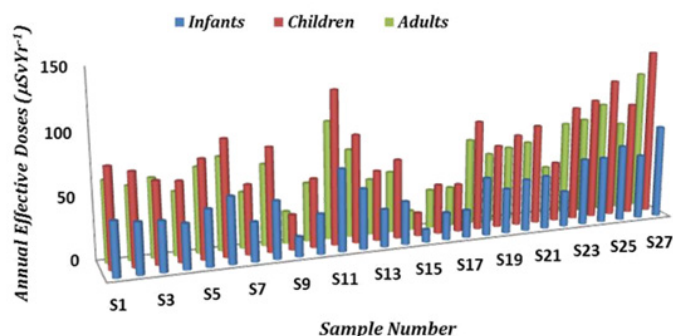


Figure 5: The annual effective doses (μSvYr^{-1}) for infants, children and adults due to intake for drinking water in study region.

Table 3: The annual effective doses ($\mu\text{Sv/yr}$) due to ingestion of ^{226}Ra , ^{232}Th and ^{40}K for various age groups in Abu Tig region.

Sample No.	Annual Effective Dose ($\mu\text{Sv/yr}$)											
	^{226}Ra			^{232}Th			^{40}K			Total Dose ($\mu\text{Sv/yr}$)		
	Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults
S1	37.053	72.047	52.594	4.439	6.6751	11.042	2.64	1.907	1.897	44.132	80.629	65.533
S2	35.06	68.173	49.766	2.918	4.3871	7.257	3.395	2.452	2.439	41.372	75.011	59.462
S3	23.812	46.3	33.799	10.332	15.537	25.7	6.203	4.48	4.456	40.346	66.317	63.956
S4	29.31	56.992	41.604	2.736	4.114	6.805	4.359	3.148	3.131	36.404	64.254	51.54
S5	33.834	65.789	48.026	7.051	10.602	17.537	4.82	3.481	3.462	45.704	79.872	69.026
S6	42.411	82.466	60.2	4.117	6.191	10.24	7.208	5.206	5.179	53.736	93.863	75.619
S7	25.544	49.67	36.259	2.713	4.08	6.749	3.602	2.601	2.588	31.86	56.351	45.596
S8	39.446	76.701	55.992	2.928	4.403	7.284	4.359	3.148	3.131	46.733	84.252	66.407
S9	11.365	22.1	16.133	3.76	5.653	9.351	1.125	0.813	0.809	16.25	28.565	26.292
S10	23.68	46.044	33.612	4.388	6.599	10.916	4.495	3.246	3.229	32.563	55.889	47.757
S11	60.713	118.05	86.178	3.23	4.857	8.035	2.776	2.005	1.995	66.719	124.92	96.208
S12	39.672	77.141	56.313	4.833	7.267	12.021	4.652	3.36	3.342	49.16	87.77	71.68
S13	26.873	52.254	38.145	2.598	3.906	6.462	1.215	0.878	0.873	30.69	57.04	45.48
S14	30.872	60.029	43.821	1.744	2.622	4.338	2.598	1.877	1.867	35.21	64.53	50.03
S15	8.5012	16.53	12.067	1.342	2.018	3.338	0.651	0.47	0.467	10.49	19.02	15.87
S16	19.313	37.553	27.414	1.051	1.581	2.615	2.063	1.489	1.482	22.43	40.62	31.51
S17	17.511	34.05	24.857	1.917	2.883	4.768	3.266	2.358	2.346	22.69	39.29	31.97

The estimated average values for the annual effective dose for adults, children, and infants approximately %60.79, %37.38 and %16 of the recommended reference values of 100, 200 and $260\mu\text{SvY}^{-1}$ respectively [14,16,29-34].

Conclusion

The present study is a contribution to study behavior and distribution of radioactivity levels of ^{226}Ra , ^{222}Rn , ^{232}Th and ^{40}K which measured in drinking water from densely populated area, which is Abu Tig city, Assiut Governorate, Egypt, using gamma-ray spectrometry technique using HPGe detector. The results clearly showed low concentrations of activity in the

studied area. The presence of ^{40}K in groundwater is attributed to the anthropogenic activities represented by the application of the agricultural fertilizers. The results of this study include the assessment of annual effective dose for adults, children and infants. The results of doses which received by children are the highest. The average annual effective doses for all radionuclides of drinking water consumption per year is much less than the recommended reference level and consequentially, therefore recommend that, the water that has been investigated be acceptable for human consumption for life. There is no clear correlation between ^{226}Ra and ^{222}Rn , as well as, no correlation between physico-chemical parameters and radionuclides.

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