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The Natural Background Activity Concentration of (^{226}Ra, ^{232}Th and ^{40}K) and the Annual Effective Dose from Different Water Sources Consumption in Phosphate Polluted Area

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THE present study is a part of series studies carried out on Nile valley phosphate mining areas, Egyptin order to gather information about radionuclides mobility and distributionand access the associated radiological health risk. This study was carried out on different water sourcessamples collected from Al-Mahamid, Upper Egypt where the study showedthat the activity concentrations of ^{226}Ra and ^{232}Th are close to each other for all water sources samples and the activity concentration of ^{40}K is higher than that of ^{226}Ra and ^{232}Th for all water sources samples.These data are compared with those obtained for different countries.The estimated annual effective dose (D_{eff}) ranged from infants to adults were also examined. The total annual effective dose obtained for infant, children and adults are lower than that obtained by WHO and UNSCEAR. The health risks to members of the public due to exposure to low dose radiation from the various drinking water sources which is regarded as chronic risk of somatic or hereditary effects were estimated.The result shows that 56 out of 1,000,000 may suffer some form of cancer fatality and 20 out of 100,000 may suffer some hereditary effects.

Keywords: Phosphate / Radionuclides /Annual effective dose / WHO /UNSCEAR.

Introduction

Radionuclides are known to be associated with Phosphate rocks which contain relatively high concentrations of naturally occurring radioactive materials from the uranium and thorium decay series[1].In April 2015, the International Atomic Energy Agency (IAEA) World Distribution of Uranium Deposits (UDEPO) estimated there were 13.8 million tons of uranium in phosphate rock deposits [2, 3 and 4]. There are no radiological controls on the operation of these industries or restrictions on how waste is discharged which relate to its radionuclide content [5]. When radioactivity released in the air, they can travel some distance depending upon many factors as wind speed and direction and altitude of the release. The products of airborne releases can be transported to humans by direct ingestion, inhalation or that deposited on the ground will find their way into plant and animal life and thereby into the food chain, or through deposition of airborne into water which can reach humans either by direct ingestion or via the food chain. The radionuclide contributing significantly to the ingestion dose via consumption of water is radium [6]. Many salts of radium are soluble in water and therefore surface water may be enriched in radium and its progenies. ^{226}Ra is an earth alkaline element sharing the metabolic pathways of calcium in the human body [6 and 7]. Ingestion of natural radionuclides depends on the consumption rates of water and on the radionuclide concentrations. Ingested radionuclides are absorbed into the blood [8] and accumulates in specific tissues that they may damage. Of absorbed uranium, 66% is rapidly eliminated via urine while the rest is distributed and stored in the kidney (12-15%), bone (10-15%) and soft tissues. The rate of clearance of such radionuclide from the tissue or organ is dependent on the biological half-life.Higher concentration of radioactivity in environmental media can cause exposure risk to the general populace which may lead to radiation related sickness such as leukemia,

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cancer of bladder, kidney, testis and lungs [9, and 10]**.** World Health Organization (WHO) [11] recommended reference dose level of 0.1 *m*Sv from one-year consumption of drinking water. This study was carried out on different water sources samples from phosphate polluted area in Al-Mahamid, Upper-Egypt in order to estimate the activity concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K and access the associated radiological health risk due to ingestion of such water.

Geographical aspects

Phosphate deposits in Egypt are part of the Middle East to the North African Phosphogenic Province of the Late Cretaceous-Palaeogene age. The occurrences are divided into three east-west trending facies belts, phosphorite of the northern facies belt spreading from Bahariya oasis to Sinai, phosphorite of the central facies belt which confined to three localities: (i) The Red Sea Coast from Safaga to the Quseir land-stretch. (ii) The Nile Valley between Idfu and Qena. (iii) The Western Desert on the Abu Tartur Plateau (New Valley area) and phosphorite of the southern facies belt [12]. The Nile Valley phosphate deposits extend between latitudes 25^{o} 30^{-} – 26^{o} 30^{-} and longitudes 32^{o}30^{-} – 33^{o} 30^{-} on both sides of the Nile Valley [13]. Our study area is a part of the Nile Valley phosphate area in the area of Al-Mahamid,Upper Egypt (25° 6’18.81”N, 32°49’15.53”E and 25° 6’4.11”N, 32°49’31.91”E), where mining of phosphate ore is mostly by surface mining. The overburden is removed either by scraping or by drilling and blasting, depending on the nature of the rock. The phosphate bed is drilled, blasted and removed by trucks to the crushing plant, where it is crushed to less than 5 cm and screened, then attrition washed to remove the clayey fine fraction and the hard-siliceous coarse fraction the control of air pollution under these circumstances is very challenging [12 and 13]. During these operations, the dust spreads in varying degrees to coverneighboring areas [14].

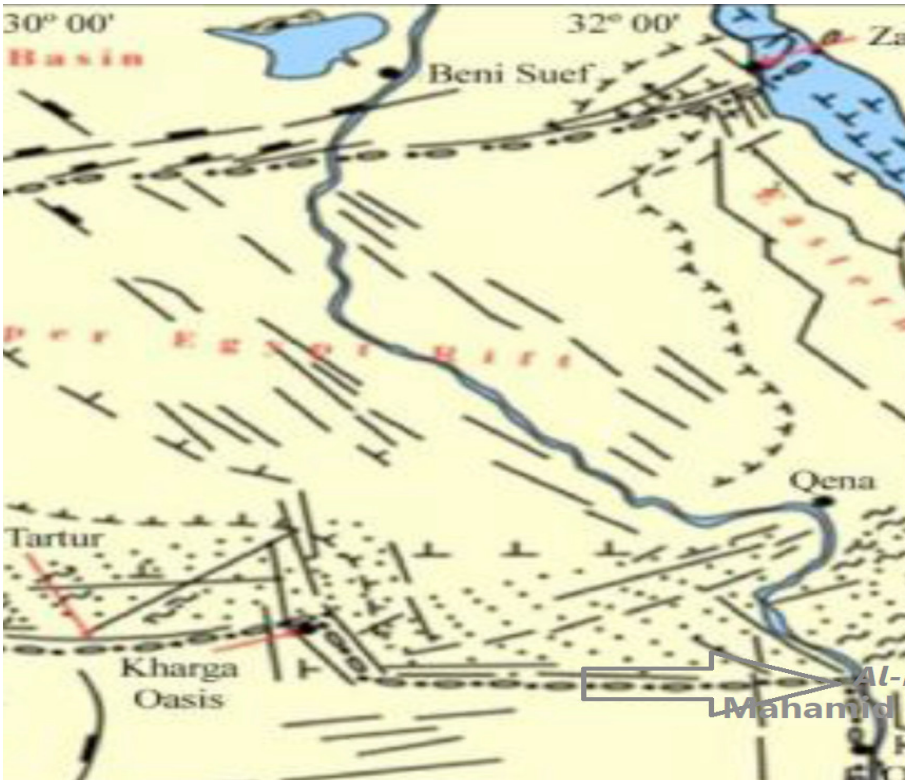
Sampling and measurements

For the purposes of this study a number of different samples of water were collected from different sources and points to cover the total area under investigation (three water samples from three wells distributed randomly throughout the area, a canal water, Nile water as well as tap water from some houses around the study area). Samples were collected using polyethylene bottles 5-liters volume. Two samples were taken from each position. The position was recorded using the global positioning system (GPS). The 5-liters volume of solution was concentrated to 1-liter volume [15]. Marinelli beakers (1-liters volume) were used as a measuring container. Each beaker was filled up to brim and a tight cap was pressed on so that the air was completely removed from it. The samples were stored for over 4 weeks to reach secular equilibrium before radiometric analysis. Low-level background gamma-ray spectrometer consists basically of 3× 3-inch NaI(Tl), S-1212-I model, with a 1024 microcomputer multichannel analyzer, 5510 ORTEC Norland was used to measurethe specific activity of the samples. The applied detectorhas a peak gamma-ray efficiency of 1.2 × 10^{-5} at 1332 KeV, energy resolution of 7.5% at 662 KeV [16] and operation bias voltage 800-1000 V D.C. The detector was housed inside a massive lead shield to reduce the background radiation. The energy calibration was carried out by acquiring a spectrum from radioactive standards of known energies (^{137}Cs and ^{60}Co). For the efficiency calibration a multi-element standard of known activities was used. By assuming secular equilibrium in the uranium and thorium decay series, the ^{238}U and the ^{232}Th activities were determined indirectly via activities of their daughter. The nuclides chosen were ^{214}Bi (609.3, 1120.3 and 1764 KeV) and ^{214}Pb (351 KeV) for ^{238}U, ^{208}Tl (2614 KeV), ^{212}Pb (238 KeV) and ^{228}Ac (911 KeV) for ^{232}Th [17 and 18]. The specific activity of ^{40}K was determined directly by 1461 KeV photopeak. The background was measured frequently and subtracted from the net count for all measured samples.The activity concentrations of the natural radionuclides in the measured samples were computed using the following equation [19]:

$$A_s = \frac{N}{(\varepsilon.M.Pr)} \quad (Bq/Kg) \qquad (1)$$

Where N is the net counting rate of γ-ray (counts per second) corrected for background, ε the detector efficiency of the specific γ-ray, P_r the absolute transition probability of γ-decay and M the mass of the sample (Kg). The total annual effective dose due to intake of drinking water sources sampled for 3 different age groups (infants, children and adults) was estimated using the following formula [7, 10 and 20]: (2)

Where: D_{eff} is the annual effective dose equivalent from consumption of drinking water, $A_R$is theconcentration of natural radionuclide in water (Bq/L),$I_L$isthe consumption rate of water (L/y).



Map of the study area, Al-Mahamid, Upper Egypt.

TABLE 1. Dose conversion factors (mSv/Bq)for ingestion of radionuclides for members of the public [21].

S/N	Radioisotopes	Infant < 1year	Children 10 years	Adult > 17 years
1	^{226}Ra	5.7 E-06	8.0 E-07	2.8 E-07
2	^{232}Th	1.6 E-06	2.9 E-07	2.3 E-07
3	^{40}K	5.2 E-05	1.3 E-08	6.2 E-09

DC_R is the dose conversion factor (*m*Sv/Bq) table (1). The water intake rates were 0.5 L/d and 1.0 L/d for infants (0-1 yrs) and children (10 yrs) respectively and 2 L/d for adults (≥ 17 yrs) [20]. The health risks to members of the public due to exposure to low dose radiation from the various drinking water sources which is regarded as chronic risk of somatic or hereditary effects were estimated [7].

$$\text{Fatalitycancerrisk} = D_{eff} \times C_{RF} \quad (3)$$

where C_{RF} is the cancer risk factor = 5.5×10^{-2}
Lifetime fatality cancer risk to adult = fatality cancer risk × 70 yrs (4)

$$\text{Severehereditaryeffects} = D_{eff} \times H_{EF} \quad (5)$$

where H_{EF} is thehereditary effect factor = 0.2×10^{-2}

Lifetime hereditary effect in adult = Severe hereditary effects × 70 yrs (6)

Results and Discussion

From table (2) and fig (1)the average activity concentrations of ^{226}Ra,^{232}Th and ^{40}K were 1.35 ± 0.06, 0.87 ± 0.04, and 5.40 ± 0.20 Bq/L respectively for tap water(W.T) samples. The values of ^{226}Ra ranged from 0.98 ± 0.05 to 3.04 ± 0.16, ^{232}Th ranged from 0.81 ± 0.04 to 1.84 ± 0.08 and ^{40}K ranged from 3.72 ± 0.13 to 10.47 ± 0.40Bq/L for canal water(W.C) samples**.** For ground water (W.G) samples the values of ^{226}Ra ranged from 1.99 ± 0.10 to 3.80 ± 0.18, ^{232}Th ranged from 1.30 ± 0.06 to 1.44 ± 0.07 and ^{40}K ranged from 5.07 ± 0.19 to 12.40 ± 0.45Bq/L**.** The values of ^{226}Ra ranged from 1.09 ± 0.05 to 2.56 ± 0.11, ^{232}Th 1.29 ± 0.06 to 1.46 ± 0.06 and ^{40}K from 8.01 ± 0.29 to 9.16 ± 0.33Bq/L for Nile water(W.N) samples. The specific activity concertation of ^{40}K is greater than that of ^{226}Ra and ^{232}Th for all water

sources samples and the activity concentration of ^{226}Ra is more than that of ^{232}Th for almost water sources samples as ^{226}Ra is more soluble than ^{232}Th. Table 3 summarizes the concentration values of ^{226}Ra, ^{232}Th and ^{40}K which were obtained from the current study and other studies for other countries. As can be seen from the table 3 ^{226}Ra values for ground water of present study matches with the values reported by Isam, et al.[28]in Sweden and Godoy, et al.[31]in brazil and higher than the values reported by Ahmed [29] in Egypt (Qena) and (Safaga - Quseir) and lower than those reported by Salonen [24]in Finland. ^{232}Th values for ground, Nile, canal and tap water of present study close to each other and are matches with the values reported by Saqan, et al.[27] in Jordan.^{40}K values of ground, Nile, canal and tap water for present study are lower than the values reported by Saqan, et al.[27]in Jordan.From table 4 and fig. 2 the annual effective dose for different age groups ranges from 0.037 to 0.103 *m*Sv/y for infants, 0.0004 to 0.001 *m*Sv/y for children and from 0.0004 to 0.0008 *m*Sv/y for adult for canal water samples. In ground water, it ranges from 0.052 *m*Sv/y to 0.120 *m*Sv/y for infants, 0.0008 to 0.001 *m*Sv/y for children and 0.0007 to 0.001 *m*Sv/y in adult. For Nile water, the total effective dose ranges from 0.078 to 0.090 *m*Sv/y for infant, 0.0005 to 0.0009 for children and 0.0005 to 0.0008 *m*Sv/y for adult. It can be observed that the radiation dose received by infants is higher than that received by children and adults for all water sources samples. The total annual effective dose obtained for infant, children and adults are lower than that obtained by WHO [10] and UNSCEAR [20]. The result showed that cancer risk for adults varies from 20×10^{-6} to 56×10^{-6} for all water sources samples and Lifetime fatality cancerrisk varies between 1×10^{-3} and 4×10^{-3}. Whereas the hereditary effect to adult per year varied from 0.728×10^{-6} to 2.036×10^{-6} and the lifetime hereditary effects in adult varies from 51×10^{-6} to 142×10^{-6}. the result shows that 56 out of 1,000,000 may suffer some form of cancer fatality and 20 out of 100,000 may suffer some hereditary effects. The United States Environmental protection Agency (USEPA) recommended acceptable cancer fatality risk limit of 1.0×10^{-6} to 1.0×10^{-4} (i.e. 1 person out of 1,000,000 to 10,000 persons suffering from some form of cancer fatality) [7, 10 and 32].

TABLE 2. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (Bq/L) for different water sources (W.T= Tap water, W.C= Canal water, W.G= Ground water and W.N= Nile water)samples.

Sample ID	Latitude	Longitude	±A(^{226}Ra)	±A(^{232}Th)	±A(^{40}K)
$W.T_1$	25° 7’56.72”N	32°47’11.17”E	1.30 ± 0.05	0.85 ± 0.03	5.61 ± 0.20
$W.T_2$	25° 7’56.99”N	32°47’1.42”E	1.40 ± 0.07	0.90 ± 0.04	5.20 ± 0.19
$W.C_1$	25° 7’58.67”N	32°46’46.05”E	2.77 ± 0.12	1.40 ± 0.06	6.06 ± 0.22
$W.C_2$	25° 7’56.09”N	32°46’43.24”E	2.11 ± 0.09	1.53 ± 0.07	4.92 ± 0.18
$W.C_3$	25° 7’52.99”N	32°46’38.36”E	1.46 ± 0.07	1.84 ± 0.08	10.28 ± 0.37
$W.C_4$	25° 7’41.22”N	32°46’25.59”E	0.98 ± 0.05	0.81 ± 0.04	6.01 ± 0.22
$W.C_5$	25° 7’47.56”N	32°47’23.60”E	3.04 ± 0.16	1.03 ± 0.06	10.47 ± 0.40
$W.C_6$	25° 7’24.45”N	32°47’57.89”E	2.54 ± 0.11	1.20 ± 0.05	8.84 ± 0.32
$W.C_7$	25° 6’22.25”N	32°49’3.40”E	1.06 ± 0.05	0.87 ± 0.04	3.72 ± 0.13
$W.G_1$	25° 7’48.67”N	32°46’34.69”E	3.80 ± 0.18	1.30 ± 0.07	5.07 ± 0.19
$W.G_2$	25° 7’44.12”N	32°46’29.44”E	1.99 ± 0.10	1.31 ± 0.06	12.40 ± 0.45
$W.G_3$	25° 7’33.74”N	32°48’43.29”E	2.59 ± 0.11	1.44 ± 0.06	8.49 ± 0.31
$W.N_1$	25° 7’31.13”N	32°46’17.90”E	2.56 ± 0.11	1.46 ± 0.06	9.16 ± 0.33
$W.N_2$	25° 6’58.46”N	32°48’4.38”E	1.63 ± 0.08	1.29 ± 0.06	8.01 ± 0.29
$W.N_3$	25° 6’20.28”N	32°48’59.67”E	1.09 ± 0.05	1.30 ± 0.06	8.21 ± 0.30

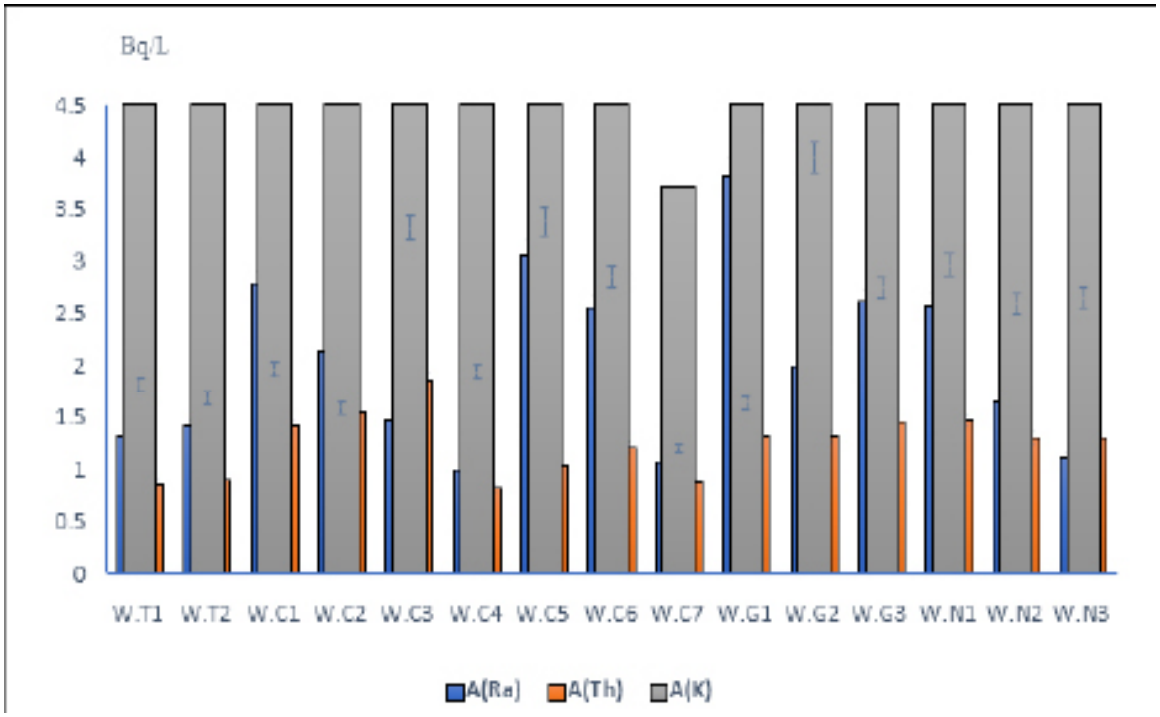


Fig. 1. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (Bq/L) for different water sources (W.T= Tap water, W.C= Canal water, W.G= Ground water and W.N= Nile water)samples.

TABLE 3. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (Bq/L) for water samples (Bq/L) in present study in comparison with other countries.

Country	Water type	Activity concentration Bq/L			Ref.	
		Ra-226	Th-232	K-40		
Current study	ground water	1.99-3.8	1.3-1.44	5.07-12.4	present study	
	Nile water	1.09-2.56	1.29-1.46	8.01-8.16	present study	
	canal water	0.98-3.04	0.81-1.84	3.72-10.5	present study	
	tap water	1.30-1.4	0.85-0.9	5.2-5.61	present study	
Egypt	natural water	1.6-11.1	0.21-0.79	9.7-23	El-Arabi, et al., 2006	[22]
Spain	natural water	0.02-4			Soto, et al., 1988	[23]
Finland	ground water	0.01-49			Salonen, 1994	[24]
Italy	drinking water	0.0002-1.2			Sgorbati and Forte.1997	[25]
China	ground water	Max 0.93			Weihai, Z., et al., 2001	[26]
Jordan	hot springs	3.8-6.8	1.42-2.37	23.2-34.8	Saqan, et al., 2001	[27]
Sweden	ground water	0.016-4.9			Isam., et al., 2002	[28]
Qena , Egypt	ground water	Mean 0.08	Mean 0.04		Ahmed, 2004	[29]
Egypt (Safaga-Quseir)	ground water	Mean 0.1	Mean 0.05			
U.S.A	mineral water	Max. 20			Kittto, et al., 2005	[30]
brazil	ground water	0.01 - 3.79			Godoy, et al., 2006	[31]

TABLE 4. The total annual effective dose, Cancerrisk to Adult, LifetimeFatality, Heredity effects to adult, hereditary effectsfor different water (W.T= Tap water, W.C= Canal water, W.G= Ground water and W.N= Nile water) samples.

Sample ID	Total annual effective dose (mSv/y)			Fatality Cancerrisk to Adult per year × 10^{-6}	Lifetime Fatality cancerrisk	Severe Heredity effects to adult per year × 10^{-6}	Estimated lifetime hereditary effects
	Infant ≤ 1	Children = 10	Adult ≥ 17				
$W.T_1$	0.055	0.0005	0.0004	23.88	0.002	0.868	0.000061
$W.T_2$	0.051	0.0005	0.0005	25.36	0.002	0.922	0.000065
$W.C_1$	0.061	0.0010	0.0008	45.62	0.003	1.659	0.000116
$W.C_2$	0.049	0.0008	0.0007	39.13	0.003	1.423	0.000100
$W.C_3$	0.100	0.0007	0.0007	35.97	0.003	1.308	0.000092
$W.C_4$	0.058	0.0004	0.0004	20.01	0.001	0.728	0.000051
$W.C_5$	0.103	0.0010	0.0008	46.35	0.003	1.685	0.000118
$W.C_6$	0.087	0.0009	0.0008	41.84	0.003	1.522	0.000107
$W.C_7$	0.037	0.0004	0.0004	20.82	0.001	0.757	0.000053
$W.G_1$	0.052	0.0013	0.0010	56.00	0.004	2.036	0.000143
$W._G2$	0.120	0.0008	0.0007	37.62	0.003	1.368	0.000096
$W.G_3$	0.084	0.0010	0.0008	44.56	0.003	1.620	0.000113
$W.N_1$	0.090	0.0009	0.0008	44.54	0.003	1.620	0.000113
$W.N_2$	0.078	0.0007	0.0006	32.22	0.002	1.172	0.000082
$W.N_3$	0.079	0.0005	0.0005	26.36	0.002	0.958	0.000067

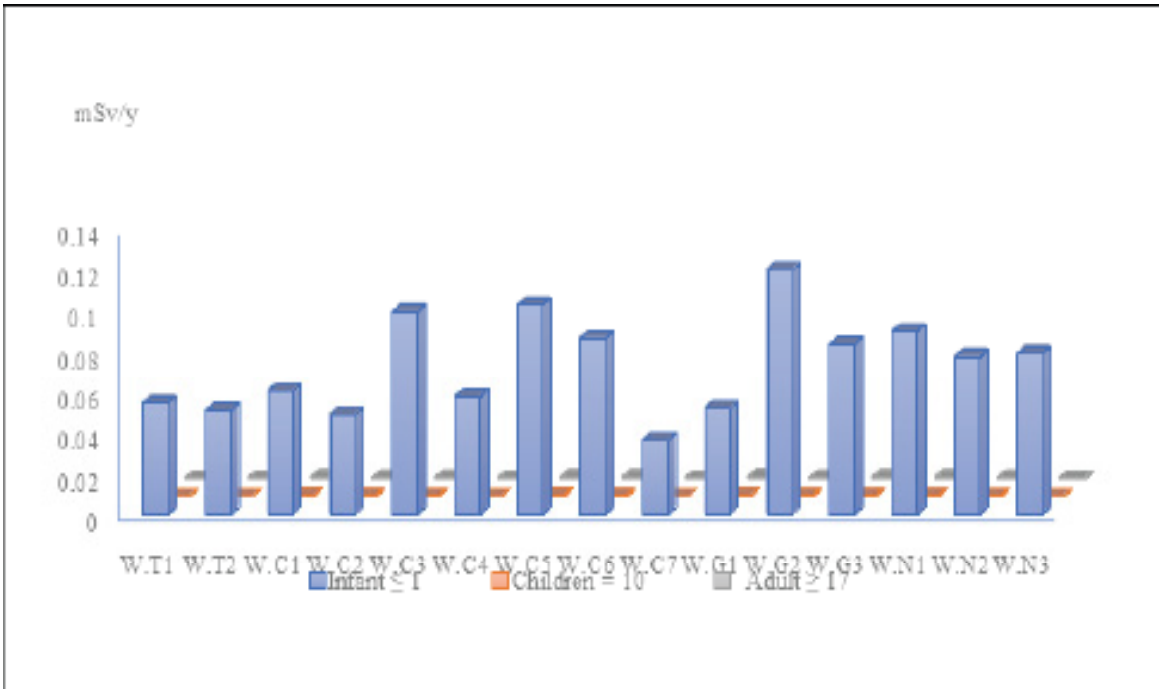


Fig. 2. The total annual effective dose (*m*Sv/y) for different water sources (W.T= Tap water, W.C= Canal water, W.G= Ground water and W.N= Nile water)samples.

Conclusion

The distribution of natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K were measured by gamma-ray spectrometry with NaI (Tl) detectors for water sources samples in phosphate polluted area in Al-Mahamid, Upper Egypt. The results from this study indicate that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the studied water sources samples are found to be normal. The results of this study showed that the total annual effective dose obtained for infant, children and adults are below the limit set by WHO.

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تعتبر الدراسة الحالية واحدة من العديد من الدراسات التى تم اجرائها علي مناطق تعدين خامات الفوسفات في مصر وغيرها من دول العلم وذلك لتقدير المخاطر الاشعاعية الناتجة من تعدين خامات الفوسفات نظرا لاحتوائها علي تركيزات عالية من عنصر اليورانيوم المشع. حيث تم جمع عدد من عينات المياة من مصادر مختلفة (مياة جوفية' مياة النيل' مياة الترع ومياة الحنفية) والتي يتم تناولها فى البيئة المحيطة بمنطقة تعدين خام الفوسفات في منطقة المحاميد أدفو أسوان. حيث تم تقدير تركيز النوويات المشعة طبيعيا (الراديوم-22' الثوريوم-232 والبوتاسيوم-04) لهذة العينات وذلك باستخدام مطياف يوديد الصوديوم وتم حساب الجرعة الثانوية الناتجة من تناول المياة من المصادر المختلفة والمخاطر الإشعاعية الناتجة لعدد من الفئات العمرية المختلفة (الرضع أقل من عام' الأطفال أقل من عشرة أعوام والشباب) وذلك طبقا للمعايير العيارية المحددة لمنظمة الصحة العالمية.