Naturally Occurring Radionuclides in Sludge Samples from Some Egyptian Drinking Water Purification Stations

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Abstract

Sludge samples from nine drinking water purification stations in El-Mynia governorate namely Kedwan, Elhawarta, Demsher, Tewa, Ard Sultan, Tokh Elkhel, Abu Flow, Saft Elkhmar and Zohrt Elporgi, were analyzed for its naturally occurring radionuclides (226Ra, 232Th and 40K) by gamma- ray spectrometry system using (sodium iodide NaI (Tl) detector).The results show that the specific activity for 226Ra, 232Th and 40K in range of 6±0.7 to 113±6.7, from 5±1 to 117±5.8 and from 47±4.5 to 412±20.6 Bq Kg-1 respectively. The results indicate that sludge of drinking water purification stations in El-Mynia governorate from large scale do not pose a significant radiological risk. Additionally evaluations have been made of the absorbed gamma dose rate in air and the annual effective dose equivalent.

Keywords*: Natural radionuclides, drinking water purification stations and sediment*

1. Introduction

Radiation in the environment from natural sources is the major source of radiation exposure to man. Radiation exposure results from the naturally-occurring radionuclides in the environment (terrestrial radiation) and direct cosmic (extra-terrestrial) radiation. Some sources of natural radiation have been enhanced (concentrated) by human technological activities and include wastes from mineral ores and the petroleum industry, sludge and scale from drinking water treatment.

The water has an importance in environmental studies because of its daily use for human consumption and its ability to transport pollutants. Sludge from drinking water treatment may contain both naturally-occurring and man-made radioactive materials. Water that originates in or moves through geologic deposits containing naturally-occurring radionuclides could result in radioactivity being carried to the treatment facility with storm water runoff or infiltration entering the sewer system, and water treatment plant residuals discharged to the sewer system.

The occurrence of natural radionuclides in drinking water poses a problem of health hazard, when these radionuclides are taken to the body by ingestion. 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 [1]. In fact, the incoming water treated in these plants can contain such radionuclides as radium or uranium due to the geological media in which the waters flow. When this water is treated it passes through various filters to remove the contaminants. This

treatment may lead to the generation of radioactive wastes such as sludge samples or also to the radiological contamination of the filters used [2]. Surface water treatment processes included sedimentation, coagulation, flocculation and filtration, while the groundwater was treated using methods similar to surface water treatment [3].

Only radionuclides with half-lives comparable with the age of the earth or their corresponding decay products existing in terrestrial material such as 232Th, 238U and 40K are of great interest. Another minor source is the radioactive decay of the 235U isotope, but this is very rate in the earth's crust (only 0.72% compared with 99.27% of 238U of its total uranium content [4]. Gamma radiation from these represents the main external source of irradiation to the human body.

Natural gamma- ray spectroscopic analysis has several advantages: the gamma rays are capable of penetrating a few inches of rock or soil samples, and both NaI (TI) (with high efficiency) and HPGe detectors (with high-energy resolution) can be utilized for the analysis [5]. 238U and its daughter's rather than 226R and its daughter products are responsible for a major fraction of the internal dose received by humans from naturally occurring radionuclides [6].

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), has estimated that exposure to natural sources contributes >70% of the population radiation dose and the global average human exposure from natural sources is 2.4 mSv y-1[7]. External exposures outdoors arise mainly from terrestrial radionuclides. The specific levels are related to the types of rock from which the soils originate. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, shale and phosphate rocks have relatively high content of radionuclides.

2. Experimental Technique

2.1.Sampling and Sample Preparation

Twenty-six sludge samples were collected from (Nine Stations) in El-Mynia, governorate is shown in Figure 1. All samples, each about 300 gm in weight, were dried in an oven at about 110 C^{o} to ensure that moisture is completely removed, the samples were crushed, homogenized, and sieved through a 200 μm, which is the optimum size enriched in heavy minerals. Weighted samples were placed in polyethylene beaker, of 350 cm^3 volume each. The beakers were completely sealed for 4 weeks to reach secular equilibrium when the rate of decay of the daughters becomes equal to that the parent this step is necessary to ensure that radon gas is confined within the volume and the daughters will remain in the sample [8].

2.2.Instrumentation and Calibration

Radioactivity measurements were performed by gamma ray spectrometer, employing a scintillation detector 3″ × 3″. Its hermetically sealed assembly which includes a high-resolution NaI (Tl) crystal, photomultiplier tube, an internal magnetic/light shield, aluminum housing and a 14 pin connector coupled to PC-MCA Canberra Accuspes. It has the following

specifications: (1) resolution 7.5% specified at the 662 keV peak of ^{137}Cs, (2) window aluminum 0.5 mm thick, density147 mg/cm^2, (3) reflector oxide; 1.6 mm thick; density 88 mg/cm^2,(4) magnetic/light shield-conetic lined steel and (5) operating voltage positive 900 V (dc).

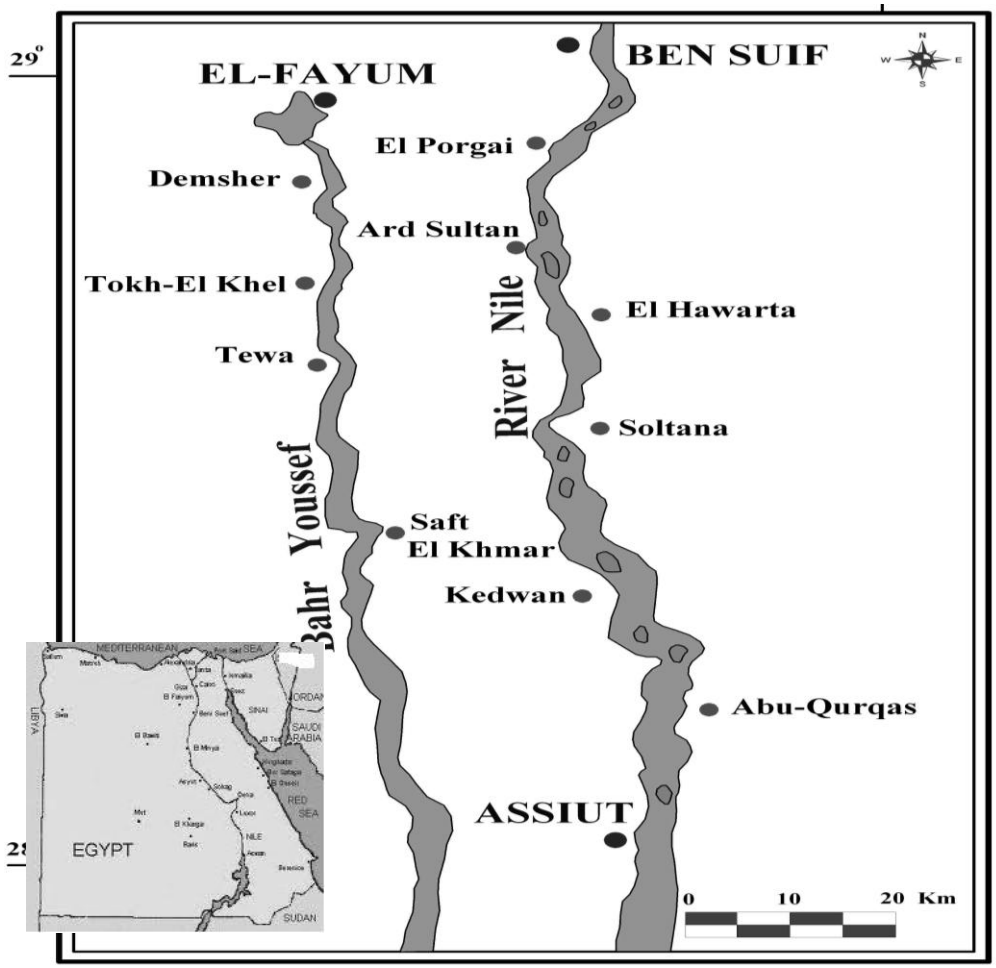

Figure 1. Location Map of the Studied Samples in El-Mynia, Governorate, Egypt

To reduce gamma ray background, a cylindrical lead shield with a fixed bottom and movable cover shielded the detector. The lead shield contained an inner concentric cylinder of copper (0.3 mm thick). 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 [8]. The detection array was energy calibrated using Co-60 (1173.2 and 1332.5 keV), Ba-133(356.1 keV), Cs-137 (661.9 keV) and Ra-226 (1764.49 keV). The efficiency calibration curve was made using different energy peaks covering the range up to 2000 keV. Daily efficiency and energy calibrations for each sample measurement were carried out to maintain the quality of the measurements. In order to determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner

and in the same geometry as the samples. The measurement time of activity or background was 43 200 s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. The offline analysis of each measured g-ray spectrum has been carried out by a dedicated software program Genie 2000 [9].

The ^{226}Ra radionuclide was estimated from the 609.3 keV (46.1%) γ-peak of ^{214}Bi, 351.9 keV (36.7%), 1120.3 keV (15%), 1728.6 keV (3.05%) and 1764 keV (15.9%) γ-peak of ^{214}Pb. The 186 keV photon peak of ^{226}Ra was not used because of the interfering peak of ^{235}U with energy of 185.7 keV. ^{232}Th radionuclide was estimated from the 911.2 keV (29%) γ -peak of ^{228}Ac and 238.6 keV (43.6%) γ -peak of ^{212}Pb. ^{40}K radionuclide was estimated using 1,461 keV (10.7%) γ -peak from ^{40}K itself. The below detectable limit (BDL) were 25.2 Bq kg^{-1} for ^{40}K, 6.5 Bq kg^{-1} for ^{226}Ra and 5.7 Bq kg^{-1} for ^{232}Th. All procedures were described in previous publication [10].

3. Chemical Analysis of the Studied Samples

Nine samples were chemically analyzed from nine drinking water purification stations (Kedwan, Elhawarta, Demsher, Tewa, Ard Sultan, Tokh Elkhel, Abu Flow, Saft Elkhmar and Zohrt Elporgi), using scanning electron microscopy (SEM) technique at electron microscope unit, Assiut university, Egypt. The chemical compositions (wt. %) of the studied samples were listed in table (1). The Underground water Saft Elhkamar station has the highest amount of iron (Fe) and manganese (Mn) element; the surface water Abu Flow station has the highest amount of zinc (Zn) and cadmium (Cd) element. Figures (2 to 4) show the results of table (1) in graphical form.

Table 1. Chemical Composition (wt. %) of the Studied Samples

Element	*Kedwan*	*Elhawarta*	*Demsher*	*Tewa*	*Ard Sultan*	*Tokh Elkhel*	*Abu Flow*	*Zohrt Elporgi*	*Saft Elkhmar*
Al	*6.31*	*6.52*	*3.09*	*13.17*	*24.02*	*3.39*	*2.45*	*1.65*	*0.09*
Si	*48.24*	*53.91*	*48.79*	*43.99*	*47.34*	*57.84*	*24.78*	*46.24*	*30.35*
P	*6.52*	*7.47*	*9.27*	*2.11*	*2.84*	*2.72*	*2.41*	*5.61*	*3.51*
S	*1.11*	*0.83*	*0.35*	*0.57*	*0.95*	*0.31*	*0.3*	*0.44*	*0.61*
K	*1.68*	*1.64*	*2.06*	*1.45*	*1*	*1.35*	*1.76*	*1.54*	*0.55*
Ca	*10.51*	*5.89*	*6.02*	*5.46*	*4.14*	*15.49*	*8.95*	*8.01*	*7.28*
Ti	*2.05*	*1.81*	*1*	*2.19*	*1.22*	*1.63*	*2.31*	*2.01*	*0.25*
Mn	*1.29*	*1.64*	*1.45*	*1.13*	*0.66*	*0.61*	*1.34*	*0.48*	*23.52*
Fe	*20.3*	*19.13*	*23.08*	*26.99*	*16.23*	*15.32*	*48.6*	*29.65*	*32.94*
Co	*0.01*	*0.34*	*2.07*	*0.03*	*0.01*	*0.01*	*1.48*	*0.41*	*0.06*
Ni	*0.24*	*0.24*	*1.72*	*0.57*	*0.57*	*0.41*	*0.29*	*0.87*	*0.05*
Zn	*0.92*	*0.14*	*0.01*	*2.14*	*0.63*	*0.97*	*4.16*	*2.59*	*0.53*
Cd	*0.82*	*0.43*	*1.08*	*0.2*	*0.39*	*0.05*	*1.18*	*0.54*	*0.28*
Total %	*100*	*100*	*100*	*100*	*100*	*100*	*100*	*100*	*100*

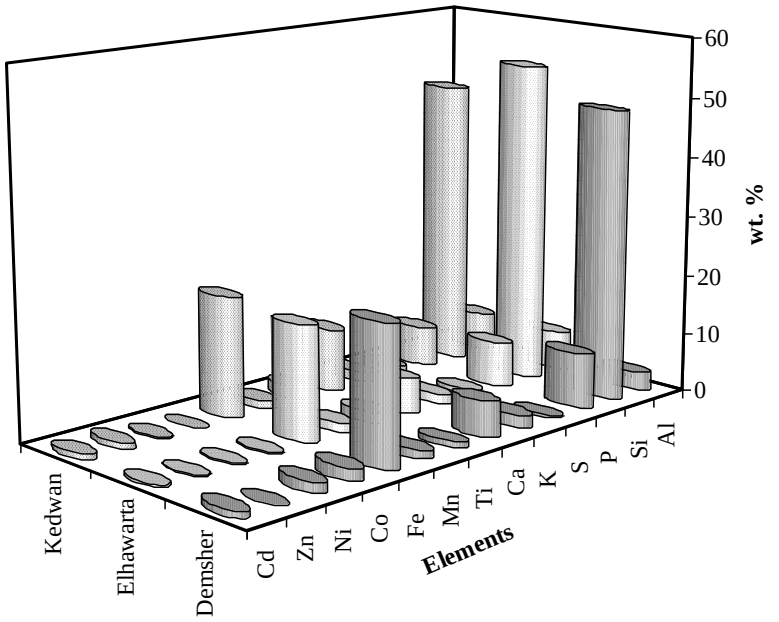

Figure 2. Chemical Analysis for Kedwan, Elhawarta and Demsher Stations

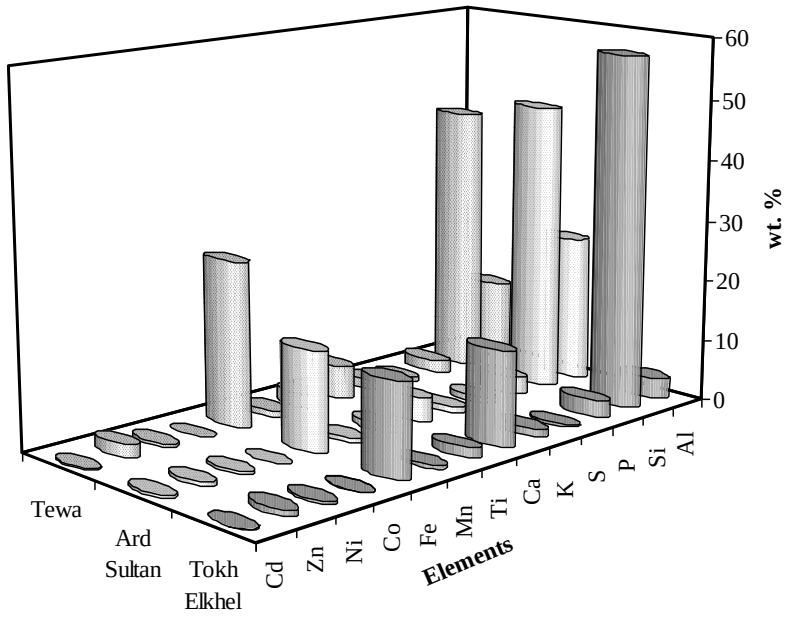

Figure 3. Chemical Analysis for Tewa, Ars Sultan and Tokh Elkhel Stations

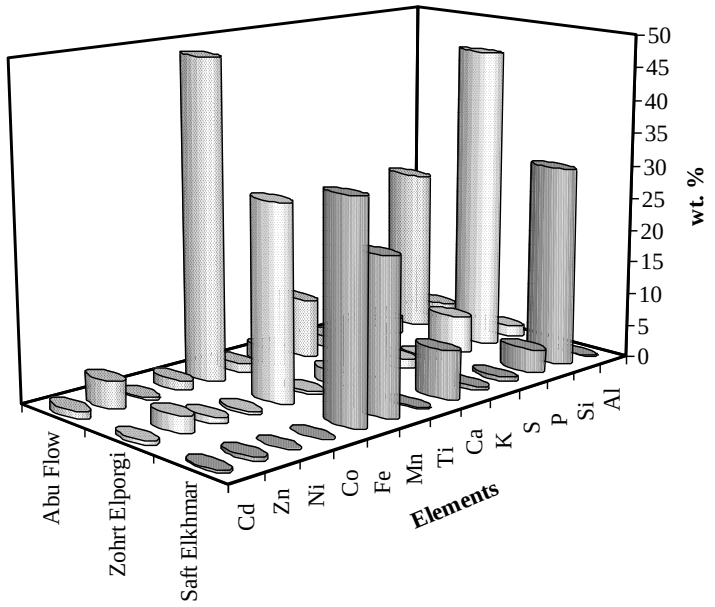

Figure 4. Chemical Analysis for Abu Flow, Zohrat Elporrgi and Saft Elkhmar Stations

4. Results and discussion

It is known that the radioactivity in sediments is similar to that in rocks, usually bed rocks, from which it derives. In this study, the concentration the various ^{226}Ra, ^{232}Th and ^{40}K activity concentrations and their corresponding total uncertainties for the 26 sediment samples under investigation were listed in Table 2. The range of specific activity for ^{226}Ra, ^{232}Th and ^{40}K were found to be from 6±0.7 to 188±9.4, from 5±1 to 117±5.8 and from 47±4.5 to 412±20.6 Bq Kg^{-1} respectively. It was clearly evident that ^{40}K always contributed to the most specific activity compared with ^{232}Th and ^{226}Ra. The Tokh Elkhel drinking water purification stations samples presented the lowest activity concentrations its averages are 13±0.7 and 119±6 Bq kg^{-1} for ^{226}Ra and ^{40}K, respectively. While the Abu Flow drinking water purification stations sediment samples presented the lowest activity concentrations are with averages 6±0.6 Bq kg^{-1} for ^{232}Th. The Kedwan drinking water purification station sediment samples presented the highest activity concentrations are with averages 76±4.2 and 86±4.25 Bq kg^{-1} for ^{226}Ra and ^{232}Th respectively. While the Zohrt Elporgi drinking water purification stations samples presented the highest activity concentrations are with averages 283±15 Bq Kg^{-1} for ^{40}K. Figure 5 shows the results in Table 2 in graphical form.

Figure 6 shows the correlation between the concentrations of the two radioactive isotopes ^{40}K and ^{226}Ra in samples under investigation (correlation coefficient =0.51). The Figure 7 indicates that there is a negative correlation (correlation coefficient =0.71) was clearly found between the ^{226}Ra concentration (Bq kg^{-1}) and Si (wt).While a positive good correlation coefficient (0.89) was found for the relation between ^{226}Ra concentration (Bq kg^{-1}) and Fe (wt) as it appears in Figure 8.

From the rest Figures 9, 10 and 11, it is clear that there were positive low correlations between (^{226}Ra and Cd), (^{40}K and Cd) and (^{226}Ra and Zn) with correlation coefficient 0.44, 0.52 and 0.69, respectively.

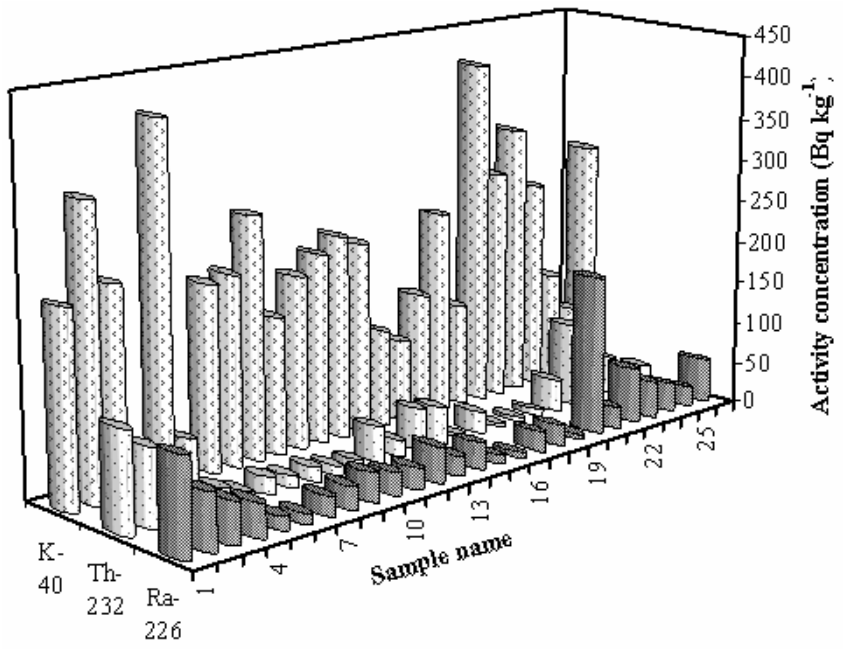

Figure 5. Activity Concentrations of 226Ra, 232Th and 40K in Different Samples

Table 2. Activity Concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Different Samples

Location	*Station*	*Sample No.*	^{226}Ra *Bq Kg^{-1}*	^{232}Th *Bq Kg^{-1}*	^{40}K *Bq Kg^{-1}*
Kedwan	Surface water	1	113±6.7	117±5.8	227±11.4
		2	67±3.4	91±4.5	337±16.8
		3	49±2.5	50±2.5	241±12.1
mean			**76±4.2**	**86±4.25**	**268±13.4**
Elhawarta	Surface water	4	40±3.1	12.5±0.6	47±6.9
		5	17±1.9	23±1.2	412±20.6
		6	12±0.8	11±0.6	47±4.5
mean			**23±1.9**	**15.5±0.8**	**168±10.6**
Demsher	Surface water	7	24±2.3	20±3	218±13
		8	27±3.5	14±3.2	223±13.3
		9	34±1.7	17±0.9	285±14.2
mean			**28±2.5**	**17±2.3**	**242±13.5**
Tewa	Surface water	10	28±2.6	11±1.4	162±9.3
		11	25±1.3	10±0.6	204±10.2
		12	42±2.1	42±2.1	224±11.2
mean			**31±2**	**21±1.3**	**196±10**
Ard Sultan	Surface water	13	24±2.7	19±2.3	239±13.5
		14	32±1.9	49±3.7	226±11.3
mean			**28±2.2**	**34±3**	**232.5±12.4**
Tokh Elkhel	Surface water	15	10±0.5	45±2.2	115±5.8
		16	6±0.7	5±1	97±4.8
		17	24±1	25±1.2	147±7.3

mean			**13±0.7**	**25±1.4**	**119±6**
Abu Flow	Surface water	18	24±2.1	5±0.3	240±14.2
		19	7±1.4	5±1.3	122±10.1
		20	188±9.4	7±0.3	412±44.5
mean			**73±4.3**	**6±0.6**	**258±22.9**
Zohrt Elporgi	Surface water	21	25±3.6	38±3.1	274±16.6
		22	66±3.3	99±4.9	324±16.2
		23	44±3.2	43±2.1	251±12.6
mean			**45±3.3**	**60±3.3**	**283±15**
Saft Elkhmar	Underground water	24	34±2.4	45±3.2	132±10.0
		25	23±1.4	26±1.3	84±4.2
		26	51±2.5	23±1.1	285±14.2
mean			**36±2.1**	**31±1.8**	**167±9.5**

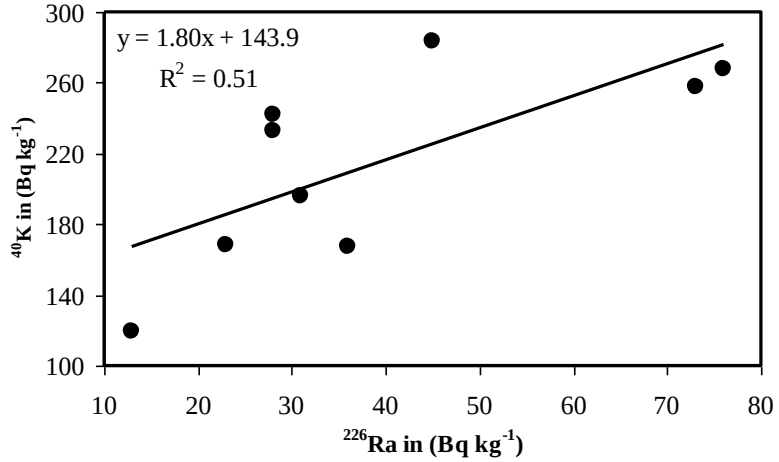

Figure 6. The Correlation between ^{40}K and ^{226}Ra Concentration in Selected Samples

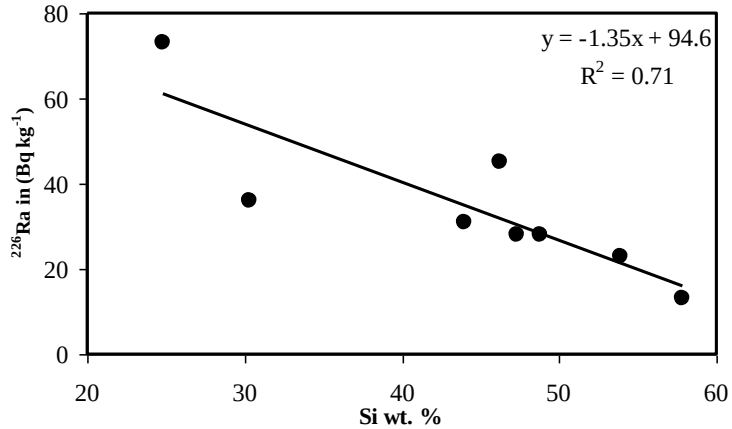

Figure 7. The Relation between ^{226}Ra concentration ($BqKg^{-1}$) and Si (wt. %)

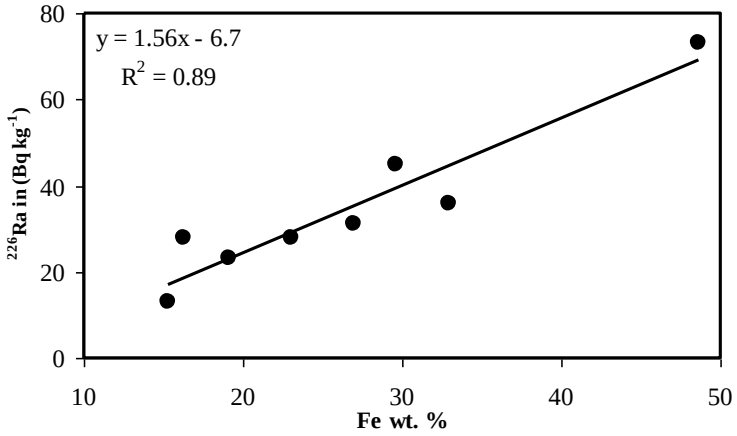

Figure 8. The Relation between ^{226}Ra Concentration (BqKg^{-1}) and Fe (wt. %)

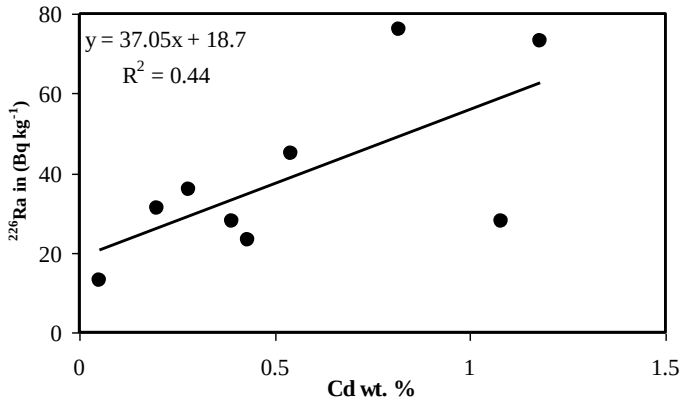

Figure 9. The Relation between ^{226}Ra Concentration (BqKg^{-1}) and Cd (wt. %)

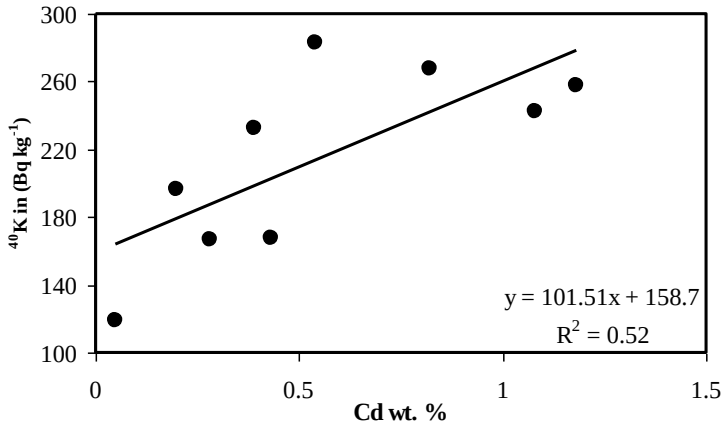

Figure 10. The Relation between ^{40}K Concentration (BqKg^{-1}) and Cd (wt. %)

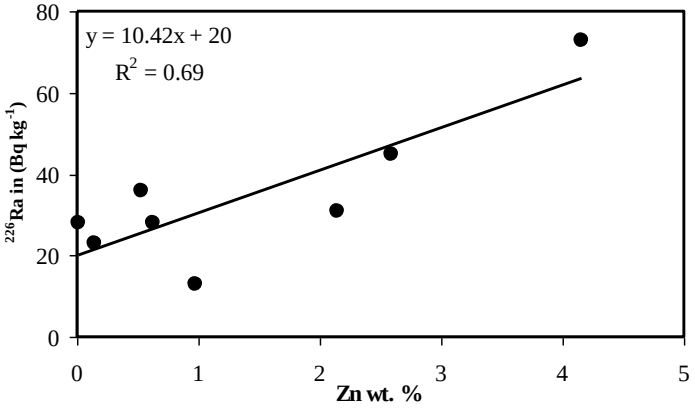

Figure 11. The Relation between ^{226}Ra Concentration (BqKg^{-1}) and Zn (wt. %)

4.1. Radium Equivalent Activity (Re_{eq})

Radium equivalent concentration (Raeq) is a common index used to compare the specific activities of materials containing ^{226}Ra, ^{232}Th, and ^{40}K. It can be expressed as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.007A_K \quad (1)$$

Where, A_{Ra}, A_{Th} and A_K are specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. Radium equivalent concentration (Raeq) was calculated based on the estimation that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4,810 Bq kg^{-1} of ^{40}K produce the same g-ray dose rate [11].

The results obtained in Table 3 showed that, the lowest Ra_{eq} was 19.4 Bq kg^{-1} in sampls from Tokh Elkhel station, while the highest value was 296.6 Bq kg^{-1} in Kedwan station. These values are less than the maximum admissible value of 370 Bqkg^{-1} [11].

4.2. Absorbed Gamma Dose Rate (D)

The absorbed dose rates due to gamma radiations in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR [12]. The conversion factors used to compute absorbed gamma dose rate (D) in air per unit activity concentration in Bq/kg (dry weight) corresponds to0.462nGy/h for ^{226}Ra, 0.604 nGy/hfor ^{232}Th and 0.042 nGy/h for ^{40}K. Therefore D can be calculated as follows [13]:

$$D = 0.462C_{Ra} + 0.604C_{Th} + 0.0417\ C_K \quad (2)$$

Where $C_{Ra,}$ C_{Th} and C_K are the concentration in *(BqKg^{-1})* of radium, thorium and potassium respectively. Column 4 of Table 3 gives the results for absorbed dose rate in air for samples under investigation. absorbed gamma dose rate for all stations ranged from 9.8 to 135.6 nGyh^{-1}, where the highest value was 135.6 nGyh^{-1} in Kedwan station while lowest value was 9.8 nGyh^{-1} in samples from Tokh Elkhel station.

4.3. Annual Effective Dose

In order to estimate the annual effective doses, one has to take into account the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor. A value of 0.7 Sv Gy^{-1} was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor, implying that 20% of time is spent outdoors, on average, around the world. The annual effective dose rate outdoors in units of μSvy-1 is calculated by the following formula [14]:

Annual Effective Dose Rate =D x T x F (3)

Where D is the calculated dose rate (in nGy h^{-1}), T is the outdoor occupancy time (0.2x24 h x365.25 d ≈1753 h y-1), and F is the conversion factor (0.7x10-6 Sv Gy-1). The experimental results of Annual Effective Dose Rate are presented in Table (3), column 6.

4.4. External Hazard Index

The external hazard index (Hex) was determined from the criterion formula as follow [15]

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (4)$$

Table 3. Equivalent Radium (Bq/kg), the Dose Rate (nGy/h), External Hazard Indices (Hex), Annual Effective Dose Rate, and Excess Lifetime Cancer Risk (ELCR)

	Sample number	*Ra_{eq} (Bq.kg^{-1})*	*Dose rate (nGy.h^{-1})*	*H_{ex} (nGy.h^{-1})*	*Ann. Eff.Dose μSv/y*	*ELCR ×10^{-4}*
Kedwan	1	296.6	135.6	0.8	164.7	5.8
	2	220.5	103.2	0.6	125.4	4.4
	3	134.8	64.6	0.4	78.4	2.7
Elhawarta	4	61.7	27.6	0.2	33.5	1.7
	5	78.9	40.3	0.2	48.9	1.4
	6	31.6	14.7	0.1	17.8	6.2
Demsher	7	67.5	32.7	0.2	39.7	1.4
	8	61.6	29.9	0.2	36.3	1.3
	9	78.4	38.1	0.2	46.3	1.6
Tewa	10	54.6	26	0.2	31.6	1.1
	11	54.7	26.6	0.2	32.3	1.1
	12	117.9	55.4	0.3	67.3	2.4
Ard Sultan	13	68.7	33.5	0.2	40.7	1.4
	14	117.4	55.6	0.3	67.5	2.4
Tokh Elkhel	15	82.3	39	0.2	47.3	1.7
	16	19.4	9.8	0.1	11.9	4.2
	17	69.8	33.6	0.2	40.1	1.4
Zohrt Elporgi	21	107.9	50.1	0.3	60.9	2.1
	22	65.8	30.5	0.2	37.1	1.3
	23	103.1	48.9	0.3	59.4	2.1
Soft Elkhmar	24	99.4	48	0.3	58.3	2.0
	25	230	107.5	0.6	130.6	4.6
	26	123.1	58.1	0.3	70.5	2.5

Where, C_{Ra}, C_{Th} and C_K are activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bq kg^{-1}. To limit the external gamma radiation dose from drinking water purification stations to 1.5 mSv $year^{-1}$ external hazard index must be less than unity in order to maintain the radiation hazard negligible [15]. The external hazard index shows in Table 3.

Excess lifetime cancer risk (ELCR) was calculated as shown in column (7) in Table 3 by using the following equation:

$$ELCR = AEDE \times DL \times RF \quad (5)$$

Where DL is duration of life (70 year) and RF is risk factor (Sv^{-1}) fatal cancer risk per Sievert. For stochastic effects, ICRP 60 [13] uses values of 0.05 for the public. The highest value (5.8E-04) for samples from Kedwan station whiles the lowest one (1.1E-05) for samples from Tewa station.

5. Conclusion

The present work shows that the natural radioactivity levels in the sediment samples which were collected from nine drinking water purification stations namely Kedwan, Elhawarta, Demsher, Tewa, Ard Sultan, Tokh Elkhel, Abu Flow, Saft Elkhmar and Zohrt Elporgi in El-Mynia, Egypt (in general) are well below the acceptable limits. The average values of all the calculated radiological indices (absorbed dose rate, annual effective dose, radium equivalent and hazard index) extracted from this activity, in all investigated samples are within the levels recommended by Radiation Protection 112 (EC, 1999) and UNSCEAR 2000 report. Therefore, the sediments do not pose any significant radiological threat to the population.

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