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ORIGINAL ARTICLE

Environmental impact assessment and statistical analysis of natural radioactivity and heavy metals of tap water in eastern Nile delta area, Egypt

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Abstract

Objectives: The present work was designed to determine radionuclide and heavy metal concentrations in tap water of the eastern Nile Delta and to investigate its validity for human consumption. **Methods:** Thirteen samples of tap water were collected from Dakahlia governorate and analyzed for selected chemical and radionuclide constituents. The concentrations of heavy metals (iron (Fe), cobalt (Co), nickel (Ni), manganese (Mn), zinc (Zn), lead (Pb), cadmium (Cd), chromium (Cr) and copper (Cu)) and radionuclides (232Th, 40K and 226Ra) were determined. Determination of heavy metal concentration was carried out by Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES). Whilst, the radionuclide measurements were conducted using Hyper Pure Germanium Detector (HPGe) Gamma-Ray Spectrometry. **Findings:** Results of this work indicated that Dakahlia tap water is slightly alkaline to alkaline water, with low salinity ranging from 233 ppm to 323.7 ppm. The heavy metal concentrations in tap water are below the permissible level for drinking. However, an anomaly of Co content (0.015 ppm) is detected in Aga site. Statistical analysis showed a strong correlation between every couple of sampling sites, but the correlation of Aga site with the rest of the sites was weak. Also, weak correlations among the heavy metals contents were detected. The maximum activity concentrations were 1.0 Bq/l, 0.9 Bq/l, and 2.6 Bq/l for ^{226}Ra, ^{232}Th, and 40K, respectively. Such values are less than the international limits recommended for water consumption. Multivariate statistical ways were also used to define the existing correlation between radionuclides and radiological health risk parameters and to pinpoint the maximum contribution of radionuclide in radioactivity. A strong correlation between every couple of sampling sites is existed, however, the correlation of Aga site with the other sites was weak. The estimated values of risk indices were below the international recommended levels. **Application:** Accordingly, the tap water of Dakahlia governorate is considered safe for human consumption except for Aga site. The exceptional water should be infiltrated before consumption to remove the excess of Cr content. The outcome of this research may be advantageous to t3.3he water database of Dakahlia governorate.

Keywords: Heavy Metals; Natural Radioactivity; Statistical Analysis; Radiation Hazard Indices; Tap Water; Eastern Nile Delta; Egypt

1 Introduction

The environment is the main element of human presence and entity. Accumulation of some elements or combinations in the environment might result in very dangerous health problems for people [1] . These health effects are essentially results of the biochemical interactions of heavy metals and radiologic interactions of energetic particles and photons with living cells [2,3]. In order to prevent possible health risks, accumulation of heavy metals and background radiation levels have to be determined. Indeed, quality of water is an essential parameter of environmental studies. The natural radionuclide and heavy element levels in tap water are significant for human health [4,5]. Heavy metal poisoning, for example, from tap-water pollution (e.g. lead pipes), high ambient air concentrations near emission sources, or absorb through the food chain by Sovioli et al. [6] and Wicke et al. [7].

Radioactivity and heavy element rates should not be more than the allowed limits for tap water. Otherwise, the likelihood of health risks will increase. The earth's crust contains heavy metals that cannot be taken down or destroyed [1]. They enter our bodies through food, tap water, and air. Some heavy metals (e.g. copper, selenium, zinc) are important to sustain the metabolism of the human body. However, at higher levels, they can be causes which lead to poisoning.

2 Materials and Methods

2.1 Site description and aim of the work

Figure 1 shows that Dakahlia Governorate is bounded from the west by El-Gharbia and KafrElshikh Governorates. El-Sharqia Governorate appears at the east and Qalubia Governorate appears at the south. The Mediterranean Sea and Damietta Governorate appears at the north. This area is approximately 3,500 km^2 and Mansoura is its capital.

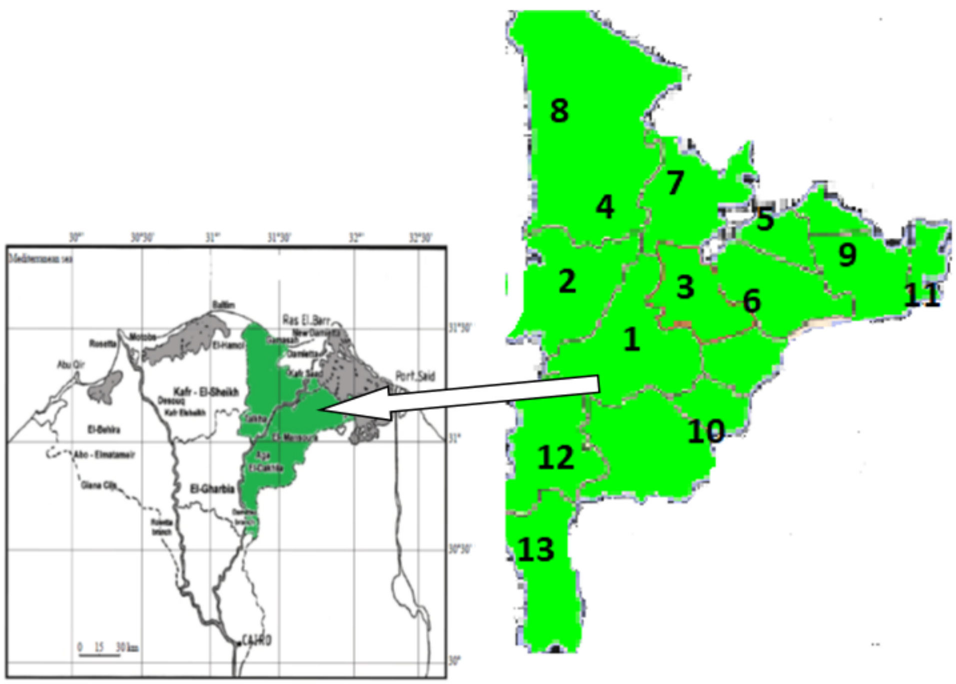

Fig 1. A map showing the study area (Dakahlia Governorate) and sampling sites

According to population rates, in 2018, the number of people living in rural areas of this governorate were estimated about 6,577,000. The urbanization rate of the study area is 28.2%. Southwest Gamasa and Asafra which contains Industrial Zone area and zone of economic activities as Food- chemicals- Textile and clothing- building materials- engineering beside Zone of infrastructure as Electricity- Natural Gas- water- Sewage stations are situated within the study area. The tap water in the study area is running from the Nile to the Damietta and its canals. Whilst, the major source of other centers of Dakahlia are water wells. At Mansoura governorate, the level of tap water in the Nile was estimated to be +1.8 m. Meanwhile, it was estimated +4 m in the nearby water wells [8]. In such sector, the groundwater mixes with the Nile to form a drain at the downstream of Mansoura governorate. The rainwater collects impurities from the air when passing through [8] as shown in (Figure 2), river streams collect surface-impurities from earth's crust, through the leaks of sewage, from agricultural and industrial effluents. These are brought to the rivers, lakes or reservoirs that supply and provide our tap water [9].

Therefore, water pollution is a serious global problem that needs continuing evaluation and revision of water resource policy at all levels. Potable water in the study area undergoes the purification process, often in public plants to make sure of its safety for consumers. The increasing

(A) (B)

Fig 2. Photographs showing the contamination of surface water A) contamination with Agricultural and household drainageB) contamination with Domestic waste

industrial activities in Dakahlia region motivated us to conduct our research. Many toxigenic heavy metals are pollutants of tap water supplies in many locations around the globe[9]. Natural radionuclides in tap water have their origin mainly from the decay series of 238U.222Rn is considered the most harmful radionuclides because its half-life is 3.8 d. Other alpha-active isotopes are those of 238U, 234U, 210Po, and 226Ra, which are long-lived radionuclides. Furthermore, long-lived beta-active 210Pb and 228Ra isotopes also occur in tap water. The 232Th undergoes series of decay to produce the isotope 228Ra[10]. Therefore, the main objective of present study is to determine radionuclide and heavy metal concentrations in tap water of Dakahlia governorate and to investigate its validity for human consumption.

2.2 Sampling and laboratory measurements

Thirteen samples of tap water were collected from the study area (Figure 2). Acid leached 2-L polyethylene bottles were used for sampling tap water. Chemicals and radiochemicals were analyzed following the standard methodologies[11] . Physicochemical parameters like pH, electrical conductivity (EC) and total dissolved solids (TDS) were determined in-situ using a portable Manta 2, Water-Quality Multiprobe device (Model Sub 3, USA). Results of radiochemical and chemical analyses were obtained by two separate laboratories for data validation. The concentrations of heavy metals: iron (Fe), cobalt (Co), nickel (Ni), manganese (Mn), zinc (Zn), lead (Pb), cadmium (Cd), chromium (Cr) and copper (Cu) and radionuclides (232Th, 40K and 226Ra) were determined (12). Determination of heavy metal concentration was carried out by Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES)at the Water and Soil Lab Unit, Desert Research Center, Cairo, Egypt. This laboratory follows the methodology of American Society for Testing and Materials (ASTM).Whilst, the radionuclide measurements were conducted using Hyper Pure Germanium Detector (HPGe) Gamma-Ray Spectrometry at the Central Laboratory for Environmental Radiation Measurement, Inter-comparison and Training (CLERMIT), Egyptian Nuclear and Radiological Regularity Authority (ENRRA).

2.3 Radiation hazard indices calculations

Fresh water should contain 226Ra activity less than 1Bq/l to be suitable for drinking (13). The following radiation hazard indices are estimated for all samples:

- **Radium equivalent activity** (*Raeq*);

$Raeq\ (Bq/kg) = A\text{Ra} + 1.43A\text{Th} + 0.077A\text{K}$ (1) ([12,13])

- **Gamma radiation hazard index** (*Irr*);

$Irr\ (Bq/kg) = 0.0067A\text{Ra} + 0.01A\text{Th} + 0.00067Ak$ (2) ([14])

- **Absorbed gamma dose rate** (*D*);

$D\ (nGy.\ h{-}1) = 0.462A\text{Ra} + 0.604A\text{Th} + 0.0417Ak$ (3) [15,16]

- **Annual gonadal equivalent dose** (*AGED*);

$AGED\ (\mu Sv.\ y{-}1) = 3.09A\text{Ra} + 4.18A\text{Th} + 0.314AK$ (4) [17,18]

- **Annual effective dose equivalent** (*AEDE*);

$AEDE\ (\mu Sv.\ y\text{–}1)$ = dose rate (in nGy/h) x 24 hx 365.25 dx 0.2(occupancy factor) x 0.7 Sv/Gy (conversion coefficient) $x10^{-3}$ (5) [18,19]

- **External hazard index** (*Hex*);

Hex =ARa370+ATh259+Ak4810$\leq$ 1 (6) [12,16]

- **Internal hazard index** (*Hin*);

Hin =ARa185+ATh259+Ak4810< 1 (7) [12,13]

- **Excess lifetime cancer risk** (*ELCR*);

$ELCR\ (\mu Sv.\ y{-}1) = AEDExDLxRF$ (8) [20]

Where ARa, ATh and Ak, are the specific activities of 226Ra, 232Th and 40K in Bq kg−1, the DL is the average duration of life (estimated to be 70 years) and RF is the risk factor (Sv), for stochastic effects, RF is used as 0.05 for the public uses[21].

2.4 Statistical analyses

SPSS version 23 statistical package and software Excel 2010 were used for data analysis. Analyses of the experimental data were carried out by using Pearson correlation matrix, Hierarchical Cluster Analysis, Principal Component Analysis (PCA) and Factor Analysis (FA) methods [22,23].

3 Results and Discussion

3.1 Hydrochemistry

pH value has a great influence on heavy metal interaction and on many other parameters. Our study confirmed that toxicity with heavy metals rises in alkaline pH values. It is obvious that the concentrations of Cu, Zn and Mn metals are under detection limits (0.006 for Cu, 0.0006 for Zn and 0.002 for Mn) of the used analytical instruments (Table 1 Table 1).

Table 1. Concentration of heavy metals (in ppm) and, Al concentrations in water

Al	Cd	Cu	Zn	Cr	Ni	Pb	Mn	Fe	Co	Sites	Site No.
0.04	0.004	<0.006	<0.0006	0.004	0.011	0.001	<0.002	<0.3	0.002	El-Mansoura	1
0.1	0.011	<0.006	<0.0006	0.008	0.017	0.033	<0.002	<0.3	<0.001	Talkha	2
0.09	0.001	<0.006	<0.0006	0.001	0.011	0.003	<0.002	<0.3	0.001	Dekernis	3
0.06	0.001	<0.006	<0.0006	0.004	0.006	0.038	<0.002	<0.3	<0.001	Nabaru	4
0.03	0.005	<0.006	<0.0006	<0.01	0.018	0.003	<0.002	0.01	0.001	Minyet el-nasr	5
0.09	0.002	<0.006	<0.0006	<0.01	0.008	0.004	<0.002	<0.3	0.002	Mit-Faris	6
0.04	0.001	<0.006	<0.0006	0.006	0.008	0.002	<0.002	<0.3	0.001	Sherbin	7
0.04	0.015	<0.006	<0.0006	0.001	0.009	<0.001	<0.002	<0.3	<0.001	Bilqas	8
0.05	0.01	<0.006	<0.0006	0.001	0.021	0.002	<0.002	<0.3	0.001	El-Gamalia	9
0.03	0.002	<0.006	<0.0006	<0.01	0.008	0.004	<0.002	<0.3	0.002	El-Sinbillawin	10
0.02	0.003	<0.006	<0.0006	0.001	0.009	0.003	<0.002	0.02	0.002	El-Manzala	11
0.01	0.001	<0.006	<0.0006	0.001	0.001	0.004	<0.002	<0.3	0.015	Aga	12
0.01	0.002	<0.006	<0.0006	0.001	0.023	0.002	<0.002	<0.3	0.003	Mit-Ghamer	13

Concentrations of Aluminum in water were in the allowed limits in all sites, the concentration is generally below 0.1 mg/l and they were equal to the allowable limits of the World Health Organization [24,25]. For all samples, concentrations of iron (Fe) were within allowable limits (0.3 mg/l) [10], and this is assured by the absence of noticeable taste; color and turbidity. Concentration of nickel (Ni) was less than allowable limits for drinking water in all samples except for MitGhmer site [26]. The exceptional sample has Ni concentration higher than the permissible limit (0.07 mg/l) recommended by WHO [25]. The high levels of Ni in tap water may cause lung disease and to affect the stomach, blood, liver, kidneys, and immune system on human health. Cobalt (Co) is an essential element for human health and may have harmful effects on human health if it is too much in concentration [27]. Concentration of Cowas within allowable limits of the WHO for all samples [10]. Lead (Pb) concentration was below allowable levels of WHO. Talkha and Nabaru showed concentrations of Pb which were higher than allowable limits of WHO (0.01mg/l) [10]. Such water may cause hazardous effects on the human central nervous system. Cadmium (Cd) concentrations from this study were within the allowable limits of WHO. On contrary, Bilqas, Talkha, El- Gamalia, El-Mansoraand Minyet el-nasr sites showed levels of Cd which were higher than allowable limits [28].

3.2 Levels of radioactivity

Activities of 40K, 232Th and 226Ra in tap-water samples collected from the study area were measured and radiation risk indices were calculated. They are presented in Tables 2 and 3. Table 2 reveals that the 40K is most abundant radionuclide followed by 226Ra and finally by 232Th.Concentrations of 232Th and 226Ra ranges from 0.1-0.9 Bq/l and 0.2-0.9Bq/l, respectively. On the other hand, 40Kactivity ranges from 1.2 to 2.6 Bq/l, with an average value that attains 1.66 Bq/l. The variation in activity concentrations in water samples may be attributed to the variation in mineral contents and other chemical parameters of tap water. Table 3 indicates that the radiation health dangers associated with radioactivity in tap water are determined to be below the limit values recommended by the WHO [25].

3.3 Statistical analyses

The statistical values corresponding to heavy metals are displayed in Table 4. Meanwhile, the activity concentrations of nuclides are displayed in Table 5. Pearson's correlation analysis was employed to correlate heavy metal concentrations of collected samples. The correlation significance between any two sets of data at 0.05 level was identified by Pearson's correlation coefficient. The results of the Pearson's correlation analysis Table 6 indicate that El- Mansoura site is significantly correlated with Talkha, Dekernis, Nabaru, Minyet El-Nasr, Mit-Faris, Sherbin, Bilqas,

Table 2. Activity Concentration of 226Ra, 232Th and 40K in Bq/l and some physicochemical parameters of water samples

Site No.	Site	pH	EC	TDS	Cl–	Alkal.	SO42–	226Ra	232Th	40K
1	El-Mansoura	7.53	486	315.9	35	148	43	0.7	0.6	2.6
2	Talkha	7.46	445	298.5	34.03	156	65	0.2	0.1	1.4
3	Dekernis	7.76	454	295.1	38	150	51	0.9	0.9	1.4
4	Nabaru	7.67	480	312	50	145	60	1	0.8	0.9
5	Minyet el-nasr	7.5	431	281.2	30	136	47	0.9	0.9	0.8
6	Mit-Faris	7.99	426	276.9	29.8	160	43.3	0.7	0.6	1.1
7	Sherbin	7.72	498	323.7	46	133	57	0.9	0.9	1.4
8	Bilqas	7.53	398	259	21	140	30	0.8	0.7	1.9
9	El-Gamalia	8.14	428	278.2	31.2	150	73.07	0.8	0.7	1.7
10	El-Sinbillawin	7.73	359	233	30	140	45	0.6	0.5	1.7
11	El-Manzala	7.79	379	246	28	145	45	0.8	0.6	1.4
12	Aga	7.53	398	259	21	140	30	0.7	0.7	1.6
13	Mit-Ghamer	7.66	388	253	23	145	38	0.6	0.5	1
	limit of WHO (2008)	6.5–8	—	1000	500	250	250	10	1	10

Table 3. Hazard indices of 226Ra, 40K and 232Th of water samples

Site No.	Sites	Dose nGy. h−1	Annual dose µSv.y-1	Ra eq Bq/kg	AGEd µSv.y−1	I^{8} Bq/kg	Ic µSv.y−1	Hin	Hex
1	El-Mansoura	0.79	0.97	1.76	5.49	0.012	0.006	0.007	0.005
2	Talkha	0.21	0.26	0.45	1.48	0.003	0.002	0.002	0.001
3	Dekernis	1.01	1.24	2.29	6.98	0.016	0.008	0.009	0.006
4	Nabaru	0.82	1.01	1.87	5.70	0.013	0.007	0.007	0.005
5	Minyet el-nasr	1.02	1.26	2.32	7.08	0.016	0.008	0.009	0.006
6	Mit-Faris	0.73	0.90	1.65	5.05	0.011	0.006	0.006	0.004
7	Sherbin	1.01	1.24	2.29	6.98	0.016	0.008	0.009	0.006
8	Bilqas	0.87	1.06	1.95	5.99	0.014	0.007	0.007	0.005
9	El-Gamalia	0.86	1.05	1.93	5.93	0.013	0.007	0.007	0.005
10	El-Sinbillawin	0.65	0.79	1.45	4.48	0.010	0.005	0.006	0.004
11	El-Manzala	0.78	0.96	1.77	5.42	0.012	0.006	0.007	0.005
12	Aga	0.81	0.99	1.82	5.59	0.013	0.006	0.007	0.005
13	Mit-Ghamer	0.62	0.77	1.41	4.32	0.010	0.005	0.005	0.004

El-Gamalia, and El-Sinbillawin sites and Talkha site is significantly correlated with Dekernis, Nabaru, Minyet El-Nasr, Mit-Faris, Sherbin, Bilqas, El-Gamalia, and El-Sinbillawin sites, etc.Statistically, a significant correlation was also found between El-Mansoura Talkha, Dekernis, Nabaru, Minyet el-nasr, Mit-Faris, Sherbin, Bilqas, El-Gamalia, and El- Sinbillawin sites were also highly correlated with each other, whereas poor correlation was found with Aga site and all sites. The results in Table (7) indicate that Cr is significantly correlated with Pb, the poor correlation was found with Al and Pb; whereas no significant correlation was found with the remaining heavy metals.

Table 4. Summary statistics of heavy metal content in water samples

Variables	Cd	Cr	Ni	Pb	Co	Al	Fe
Mean	0.0044	0.0021	0.0115	0.0076	0.0023	0.0469	0.0023
Std. Deviation	0.0045	0.00250	0.0063	0.0124	0.0039	0.0301	0.0059
Variance	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Skewness	1.429	1.376	.490	2.167	3.259	0.652	2.682
Std. Error of Skewness	0.616	0.616	0.616	0.616	0.616	0.616	0.616
Kurtosis	0.962	0.959	-0.408	3.350	11.222	-0.694	6.964
Std. Error of Kurtosis	1.191	1.191	1.191	1.191	1.191	1.191	1.191
Minimum	0.001	0.000	0.001	0.000	0.000	0.010	0.000
Maximum	0.015	0.008	0.023	0.038	0.015	0.100	0.020

Table 5. Summary statistics of radioactive elements and hazard indices in water samples

Variables	Ra 226	Th 232	K 40	Dose	Annual Dose	Radium Equivalent	AGED	Gamma Radiation	ELCR	Hin	Hex
Mean	0.738	0.654	1.454	0.783	0.962	1.766	5.422	0.012	0.007	0.007	0.005
Std. Deviation	0.202	0.218	0.477	0.214	0.262	0.492	1.474	0.004	0.002	0.002	0.001
Variance	0.041	0.048	0.228	0.046	0.069	0.242	2.173	0.000	0.000	0.000	0.000
Skewness	-1.552	-1.224	0.942	-1.577	-1.557	-1.564	-1.570	-1.519	-1.372	-1.213	-1.923
Std. Error of Skewness	0.616	0.616	0.616	0.616	0.616	0.616	0.616	0.616	0.616	0.616	0.616
Kurtosis	3.621	2.542	1.679	3.773	3.754	3.788	3.763	3.632	2.834	2.752	5.066
Std. Error of Kurtosis	1.191	1.191	1.191	1.191	1.191	1.191	1.191	1.191	1.191	1.191	1.191
Range	0.800	0.800	1.800	0.810	1.00	1.870	5.600	0.013	0.006	0.007	0.005
Minimum	0.200	0.100	0.800	0.210	0.260	0.450	1.480	0.003	0.002	0.002	0.001
Maximum	1.000	0.900	2.600	1.020	1.260	2.320	7.080	0.016	0.008	0.009	0.006

Hierarchical clustering analysis is one of the techniques that enable to partition off a site's area of water samples into subsets. Sites grouped in one cluster are considered similar and sites grouped in different clusters are considered dissimilar. Frequency distributions of cited nuclide activity are presented in Figure 3.

Skewness of 226Ra and 232Th activity exhibited negative values to indicate that their distribution area symmetric with left tail is longer than the right, while the positive value of skewness obtained for 40K activity concentrations shows that their distribution area symmetric with right tail being longer than the left as can be seen in Figure 3.

Table 6. Pearson's correlation analysis between every couple of sites area

Correlation between Cases	Vectors of Values												
	El-Mansoura	Talkha	Dekernis	Nabaru	Minyet el-nasr	Mit-Faris	Sherbin	Bilqas	El-Gamalia	El-Sinbillawin	El-Manzala	Aga	Mit-Ghamer
El-Mansoura	1												
Talkha	.939	1											
Dekernis	.984	.954	1										
Nabaru	.798	.953	.839	1									
Minyet el-nasr	.901	.827	.865	.687	1								
Mit-Faris	.978	.956	.999	.846	.851	1							
Sherbin	.992	.950	.989	.828	.866	.984	1						
Bilqas	.952	.898	.931	.732	.862	.930	.923	1					
El-Gamalia	.980	.912	.946	.760	.939	.937	.951	.962	1				
El-Sinbillawin	.987	.966	.985	.861	.904	.982	.979	.934	.973	1			
El-Manzala	.637	.591	.645	.499	.834	.639	.627	.599	.640	.644	1		
Aga	.442	.432	.457	.426	.289	.472	.439	.362	.378	.482	.187	1	
Mit-Ghamer	.516	.389	.394	.273	.694	.363	.446	.441	.628	.514	.361	.141	1

Table 7. Correlation between every couple of heavy metals

Heavy Metals	Cd	Cr	Ni	Pb	Co	Al	Fe	Cu	Zn	Mn
Cd	1									
Cr	.150	1								
Ni	.364	.015	1							
Pb	.053	.611	-.059	1						
Co	-.341	-.239	-.441	-.219	1					
Al	.186	.377	.044	.469	-.464	1				
Fe	-.072	-.244	.030	-.154	-.068-	-.327	1			
Cu	.000	.000	.000	.000	.000	.000	.000	1		
Zn	.000	.000	.000	.000	.000	.000	.000	.000	1	
Mn	.000	.000	.000	.000	.000	.000	.000	.000	.000	1

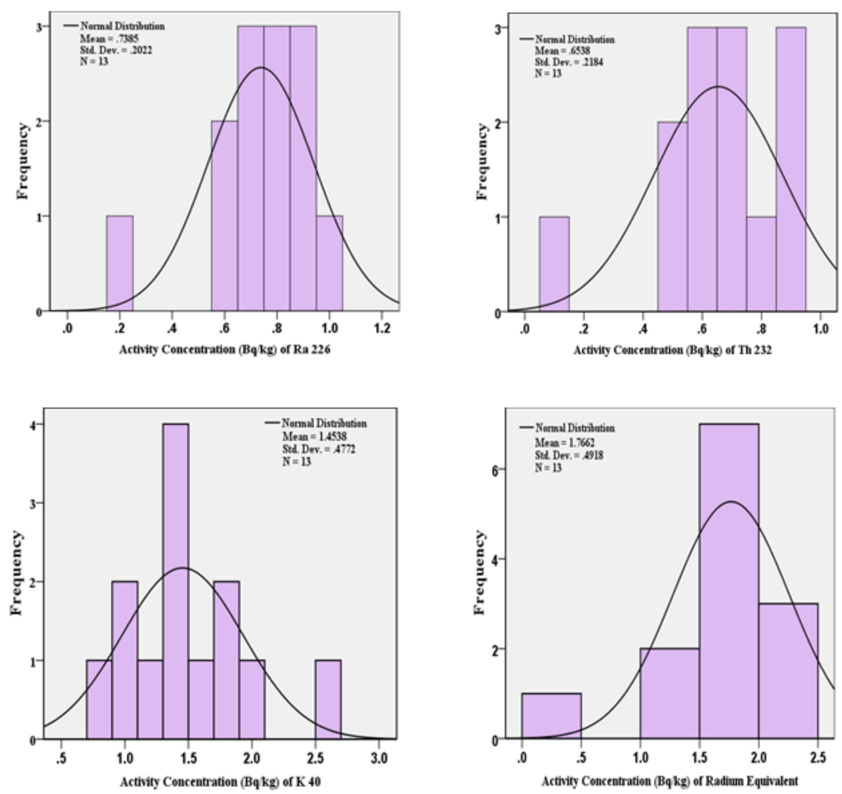

Fig 3. The frequency distribution of the activity concentrations in water samples

4 Conclusion

The present study investigated the background levels of radioactivity and heavy metal concentrations in tap water of Dakahlia governorate. Our results clarified that the determined heavy metal and radioactivity in tap water of the research area are compatible with the studies carried out in other regions in Egypt. The health dangers associated with radioactivity in tap water are determined to be below the limit values recommended by the WHO [(25)]. The slightly high value of heavy metal content (Cd and Cr above the reference values) in some parts of the region is a result of the redundant aggregation of heavy metals in water. The origin of most detected heavy metals in study area are perhaps battery, plastics and steel industries within this area. Regular monitoring strategy of the tap water in such areas is recommended. In parallel, such industries should be prohibit ed or limited to urban regions.

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