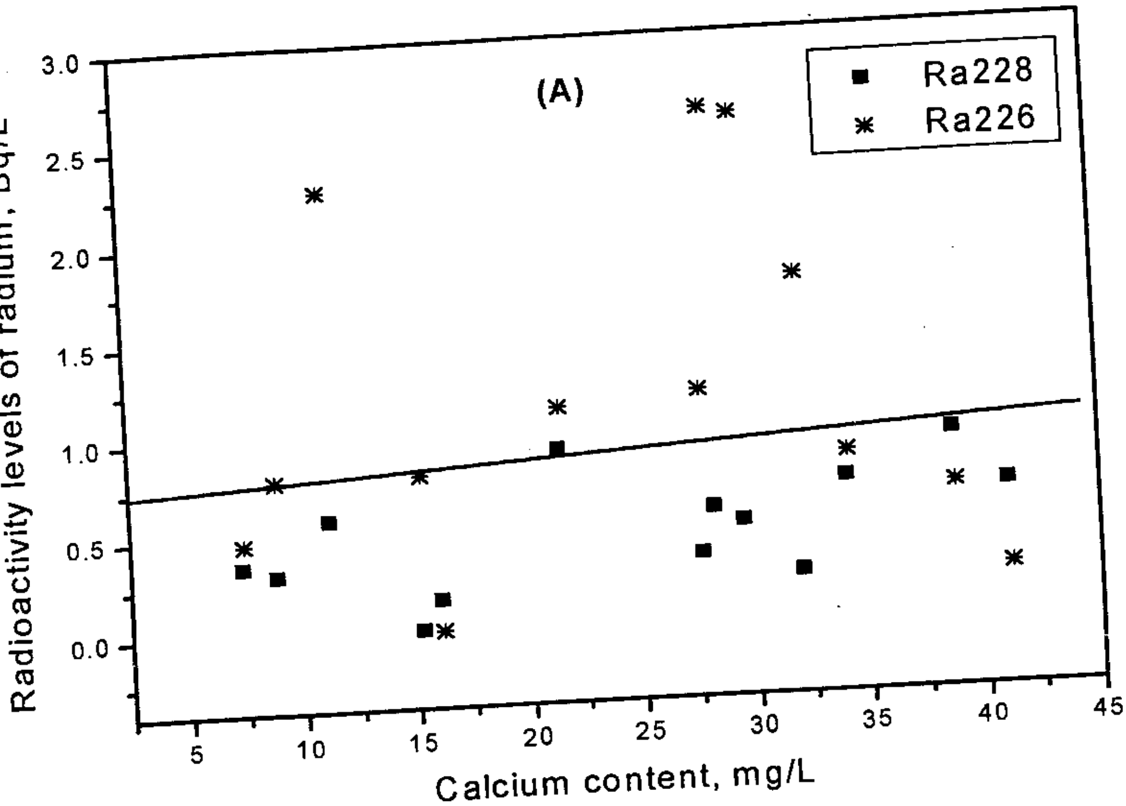

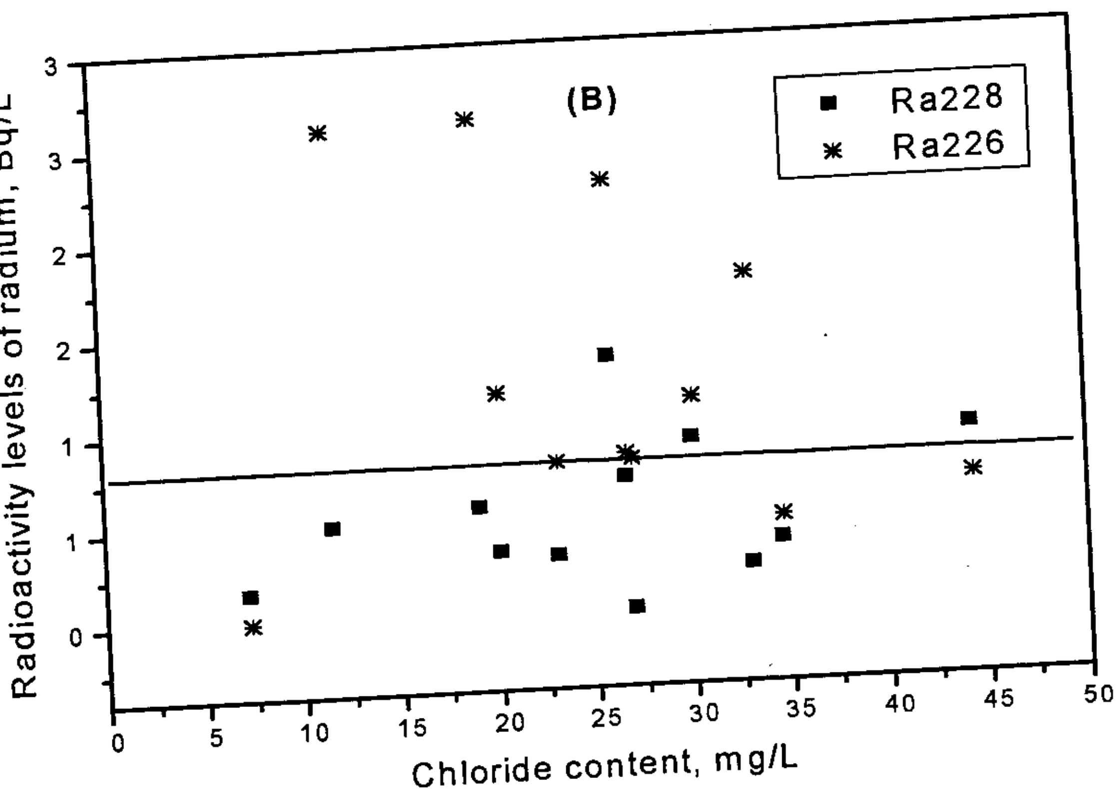

Fig.3.21 Correlation of radium radioactivity levels with both calcium (A) and chloride (B) concentrations in drinking water samples.

of water discharge, seasonal temperature changes, the nature of each aquifer and related geological formations at the time of discharge.

Table 3.19
Radium-226 and radium-228 radioactivity levels in water samples collected from different areas at different production dates

sample	production date	Ra-226, Bq/L	Ra-228, Bq/L
S04	7, 2001	2.260±0.121	1.331±0.124
S04	2. 2003	1.089±0.073	0.575±0.087
S06	11,2001	1.110±0.074	0.889±0.111
S06	3,2003	0.108±0.050	0.347±0.087
S09	6,2001	0.650±0.060	0.906±0.116
S09	2,2003	......	0.446±0.088

Further studies of several samples collected at different production dates and for specific operational conditions need still to be investigated.

Based on the data illustrating the effect of the physical characteristics and chemical composition of the tested water samples on the radioactivity level of radium, it can be concluded that:

1- The radioactivity levels of radium is found to correlate with both the TDS and pH of the tested water samples, giving a slight decrease in the radioactivity levels of radium by increasing TDS concentrations within the range 150 to 400 mg/L and by increasing the pH within a range from 7.0 to 8.5.

2- The radium radioactivity levels also give a slight positive correlation revealing an increase in the radioactivity levels by increasing both calcium and bicarbonate concentrations.

3- The effect of sulfate and chloride ions proved to give minor changes in the radioactivity levels of radium with concentrations ranging from 10 to 60 mg/L for sulfate and from 5.0 to 45 mg/L for chloride.

3.3.4 Activity ratios and secular equilibrium between related nuclides

The radioactivity levels of uranium-238, thorium-232 and their daughters radium-226 and radium-238, together with the isotopic ratios Ra-226/Ra-228, U-238/Th-232, Ra-226/U-238 and Ra-228/Th-232 are given in, Table 3.20.

- For bottled drinking water collected from **Siwa Oasis**, it is found that radioactivity levels of uranium-238 and thorium-232 range from 0.390 to 0.560 and from 0.089 to 0.138 Bq/L, respectively; while, the radioactivity levels of radium-226 and radium-228 range from 0.470 to 1.09 and from 0.0 to 0.570 Bq/L, respectively. The Ra-226/Ra-228 isotopic ratio is found to be >1.0 for all samples, and ranges from 1.382 to 2.633, with a mean value 2.603. This result is confirmed with the corresponding value related to U-238/Th-232 ratio (the parents of radium isotopes), where all samples give also high values ranging from 2.826 to 4.607, with a mean value 3.689.

Previous studies reported a wide range for Ra-226/Ra-228 activity ratios when dealing with low salinity water. Asikainen [48], found ratios in the range of 0.3 to 26 for some Finnish ground water, with a ratio around 1.0 for low radioactivity levels of radium-226; but this ratio increased rapidly with increased levels of radium-226. Other ratios, including values of 0.40-21 for water from Iowa [111] and from 0.09 to 4.41 in South Carolina [112] were also reported. The high Ra-226/Ra-228 ratio may be attributed to some processes including decay of radium-228 in water or a preferential leaching of radium-226 due to radiation damage. Some authors suggested preferential leaching of radium-226 compared to radium-228 because of three successive α-recoils in the decay series from uranium-238 to radium-226. This could cause looser bounds and damage in the crystal lattice leading to higher α-recoil rate due to the ejection of radium-226 precursors [113,114]. The α-recoil rate of radium-226 could be 1.7-2.0 times higher than that of radium-228. Also, high Ra-226/Ra-228 ratios may be due to direct decay of uranium-238 that has higher radioactivity levels relative to thorium-232 in related drinking water samples.

The high U-238/Th-232 activity ratios can be explained in the light of related chemical behaviour, where uranium species are known to have higher solubility and leachability than those for thorium compounds which depend mainly on the pH and chemical composition of each aquifer or to the local enrichment of uranium in the aquifer rocks [40].

To study the radioactive equilibrium of uranium and thorium nuclide series in drinking water, both Ra-226/U-238 and Ra-228/Th-232 activity ratios are calculated. The results show that all samples in this region have no equilibrium state between the parent uranium-238 and its daughter radium-226, except S03 (Hayat) sample, which tends to show a certain degree of equilibrium leading to Ra-226/U-238 ratio around 1.205. Also, all tested samples proved to have similar trend showing disequilibrium between the parent thorium-232 and radium-228 with a Ra-228/Th-232 activity ratios ranging from 0.0 to 4.524 with a mean value around 2.590.

The disequilibrium in these two cases may be attributed to a number of factors affecting both the parent and related progeny in different manner. These factors include:

1- The chemical composition, salinity and pH of the tested water that might affect the major processes for migration of radium from the surroundings into solution. This reveals that uranium and thorium in solution are not the only sources of radium in ground water. Instead, radium is likely to be released independently from the rocks of respective aquifers.

Table 3.20
Radioactivity levels of U-238*, Th-232*, Ra-226**, Ra-228** and related ratios in drinking water collected from different locations in Egypt

radionuclide	U-238	Ra-226	Th-232	Ra-228	U-238/Th-232	Ra-226/Ra-228	Ra-226/U-238	Ra-228/Th-232
sample code	radioactivity levels, Bq/L				ratio			
	bottled drinking water from areas within the Western Desert (Siwa Oasis)							
S01	0.560	0.800	0.135	0.000	4.148	0.000	1.429	0.000
S02	0.410	0.790	0.089	0.300	4.607	2.633	1.927	3.371
S03	0.390	0.470	0.138	0.340	2.826	1.382	1.205	2.464
S04	0.400	1.090	0.126	0.570	3.175	1.912	2.725	4.524
mean	**0.440**	**0.788**	**0.122**	**0.302**	**3.689**	**>2.603**	**1.822**	**2.590**
	bottled drinking water from areas within Western Nile Delta (El-Sadat City)							
S05	0.260	0.000.	0.135	0.150	1.926	0.000	0.000	1.111
S06	0.380	1.110	0.127	0.890	2.992	1.247	2.921	7.008
S07	0.340	0.830	0.114	0.620	2.982	1.203	2.441	6.053
S08	0.450	1.740	0.070	0.220	6.429	7.909	3.887	3.143
mean	**0.360**	**0.920**	**0.112**	**0.487**	**3.582**	**2.603**	**2.312**	**4.329**
	bottled drinking water from areas within Eastern Nile Delta (Benha and Belbeis) Cities							
S09	0.330	0.650	0.091	0.910	3.626	0.714	1.970	10.00
S10	0.230	0.220	0.156	0.640	1.474	0.344	0.957	4.103
mean	**0.280**	**0.435**	**0.124**	**0.775**	**2.550**	**0.529**	**1.464**	**7.051**
	domestic water from different Governerates							
Cairo,S11	0.430	2.610	0.122	0.490	3.525	5.327	6.070	4.016
Inshas,S12	0.500	2.580	0.136	0.560	3.676	4.607	5.160	4.118
Benha,S13	0.470	1.170	0.085	0.330	5.529	3.545	2.489	3.882
mean	**0.470**	**2.120**	**0.114**	**0.460**	**4.243**	**4.493**	**4.573**	**4.005**

*) Measured by gamma spectroscopy based on the γ-lines of related daughters, 63 keV of thorium-234, 352 keV of lead-214, 609 and 1764 keV of bismuth-214 for uranium-238, and 238 keV of lead-212 and 583 keV of thallium-208 for thorium-232.
**) Measured by LSC with α-β discrimination capability using the counting energy regions from 30.0 to 350 keV for alpha measurements and from 0.00 to 185 keV for beta measurements.

2- The distribution of radium within the surrounding formations depends on the type and size of related grains of the contact area between the ground water and the surroundings (surface to volume ratio).

3- The effect of α-recoil processes may lead to the accumulation of radium isotopes in the aquatic media.

i- The radioactivity levels of uranium-238 and thorium-232 in drinking water originating from the **Western Nile Delta (El-Sadat City)** proved to be within a range from 0.26 to 0.45 and from 0.07 to 0.135 Bq/L with mean values of 0.360 and 0.112 Bq/L, respectively. The corresponding radium-226 and radium-228 mean radioactivity levels are 0.92 and 0.487 Bq/L, respectively. Also, all Ra-226/Ra-228 activity ratios in water from the studied area have high values ranging from 0.0 for S05 (Delta) sample to 7.909 for S08 (Tiba) sample, with a mean value 2.603. This gives U-238/Th-232 activity ratios ranging from 1.0926 to 6.429 with a mean value 3.582. These data clearly show that, radium-226 and radium-228 are in excess of respective equilibrium levels of their parents uranium-238 and thorium-232. This is clearly illustrated by the mean Ra-226/U-238 activity ratio which proved to be almost around 2.312, while the Ra-228/Th-232 activity ratio is about 4.329. The only case that appeared to be in secular equilibrium between radium-228 and thorium-232 is S05 (Delta) sample that gives Ra-228/Th-232 activity ratio around 1.111.

ii- For ground water originating from the **Eastern Nile Delta**, it is found that the radioactivity levels of uranium-238 and thorium-232 range from 0.230 to 0.330 and from 0.091 to 0.156 Bq/L, respectively. In such a case, although the radioactivity levels of uranium-238 is almost double that of thorium-232, yet the radioactivity levels of radium-226 are found to be less than those for radium-228, giving Ra-226/Ra-228 radioactivity ratios <1.0. This inverse behaviour may be due to the geological formations prevailing in this region, where the ground water resources are generally within rural area; which mostly consists of clay materials from the River Nile. It is also found that U-238/Th-232 radioactivity ratios give high values following the same trend as water in the previous two areas. For S09 (Baraka) sample from Kafr Al-Arbein area, disequilibrium is shown between both radium-226 and uranium-238, and between radium-228 and thorium-232, as illustrated by the activity ratio which proved to be 1.97 for Ra-226/U-238 and almost about 10.0 for Ra-228/Th-232. For S10 (Mineral) sample secular equilibrium between uranium-238 and radium-226 is evidenced by the activity ratio for Ra-226/U-238 which is almost around 0.957. On the other hand, disequilibrium exists for thorium-232 and radium-228 giving a Ra-228/Th-232 ratio of the order 4.103.

In general, it is clearly illustrated that the Ra-226/Ra-228 activity ratios decrease from 2.603 in water resources around Siwa Oasis to 0.529 in resources from the Eastern Nile Delta regions (Belbeis and Benha Cities). Also uranium-238 and radium-226 radioactivity levels follow the same trend.

iii- Surface water samples proved to contain the highest uranium-238 and radium-226 radioactivity levels compared to the bottled ground water samples. Uranium-238, has radioactivity levels ranging from 0.430 to 0.50 (within a mean value 0.47 Bq/L); while radium-226 has radioactivity levels ranging from 1.170 for S13 (Benha) sample to 2.61 for S11 (Cairo) sample with a mean value 2.120 Bq/L. Also, surface water has the highest Ra-226/Ra-228 activity ratios ranging from 3.545 to 5.327 within a mean value of 4.493. This result is similar to the U-238/Th-232 radioactivity ratios, which give a mean value of 4.243. It is also clear that, all surface water samples show disequilibrium between radium-226 and radium-228 radionuclides and their parents. Ra-226/U-

238 activity ratios proved to be 2.489 for S13 (Benha) sample to 6.070 for S11 (a sample from Cairo area); while the Ra-228/Th-232 activity ratios range from 3.882 for S13 (Benha) sample to 4.118 for S12 (Inshas) sample, with a mean value 4.005

The results of the calculated radioactivity ratios and related secular equilibrium of studied nuclides, can be summarized in the following:

1- All tested drinking water samples have radioactivity levels for uranium-238 more than those for thorium-232, and the U-238/Th-232 ratios range from 1.474 to 6.429. This may be attributed to the chemical behaviour of uranium species, which are known to have higher solubility and leachability than those of thorium.

2- The radioactivity levels of radium-226 are almost more than those of radium-228 in all samples and the Ra-226/Ra-228 ratios range from 1.203 to 5.327. This may be due to α-recoil processes in the decay series of uranium-238 to radium-226 and/or to the high levels of uranium-238 than those of thorium-232.

3- The radioactivity levels of radium isotopes are higher than those of respective parents providing disequilibrium states in almost all tested water. This disequilibrium may be due to several factors affecting both parents and respective daughters in different manner including differences in chemical behaviour of parents and related daughters as well as the chemical composition, salinity and pH of the tested water samples.

3.3.5 Assessment of the annual effective dose from drinking water

The contribution of drinking water to the total human exposure is too small and is largely due to the naturally occurring radionuclides of uranium and thorium and related decay series. Radiation exposure from other sources is limited by regulatory control of used sources and periodic application of related practices.

On the basis of the radioactivity levels (Bq/L) of both radium-228 and radium-226 in drinking water samples, the annual ingestion of drinking water by adults is assumed to be around 730 L/y and the dose conversion factor of measured radioactivity levels for radium-228 is 6.9 x 10^{-4} mSv/Bq, and that for radium-226 is 2.8 x 10^{-4} mSv/Bq [115]. The annual intake of these isotopes and related annual effective dose are calculated from the following equation.

$$GL = (IDC/ (h_{ing} \cdot q)) \tag{3.1}$$

where:

GL	the radioactivity levels of radionuclide in drinking water, in Bq/L
IDC	the annual effective dose, in mSv/y
h_{ing}	the dose conversion factors, in mSv/Bq
q	the annual ingested volume of drinking water is assumed to be 730 L/y.

The results representing the annual intake of both radium-228 and radium-226 radionuclides and the corresponding annual effective dose compared with the annual effective dose from drinking water in certain other countries, are listed in Table 3.21.

i- annual effective dose from radium-226

The calculated annual effective dose of radium-226 in bottled drinking water samples collected from different locations show different values. Samples from the Western Desert regions within Siwa Oasis (S01-S04) have a mean value 0.159 mSv/y, ranging from 0.095 mSv/y for S03 (Hayat) sample to 0.220 mSv/y for S04 (Safi) sample. Samples from the Western Nile Delta regions located at El-Sadat City (S05-S08) proved to have 0.186 mSv/y annual effective as a mean value, but samples collected from the Eastern Nile Delta areas, S09 (Baraka) and S10 (Mineral)

Table 3.21
Annual intake of radium-226, radium-226 and the corresponding annual effective dose from bottled drinking mineral and tap water in Egypt comparable to those reported from drinking water from other countries

radionuclide	radioactivity levels, Bq/L		annual intake, Bq/person		annual dose, mSv/y	
	Ra-226	Ra-228	Ra-226	Ra-228	Ra-226	Ra-228
sample code	bottled drinking water from areas within the Western Desert (Siwa Oasis)					
S01	0.800	0.000	576.0	0.000	0.161	0.000
S02	0.790	0.300	568.8	216.0	0.159	0.149
S03	0.470	0.340	338.4	244.8	0.095	0.169
S04	1.090	0.570	784.8	416.4	0.220	0.283
mean			**567.0**	**219.3**	**0.159**	**0.150**
	bottled drinking water from areas within Western Nile Delta (El-Sadat City)					
S05	0.000	0.150	0.000	108.0	0.000	0.075
S06	1.110	0.890	799.2	640.8	0.224	0.442
S07	0.830	0.690	597.6	496.8	0.167	0.343
S08	1.740	0.220	1252.8	158.4	0.351	0.109
mean			**662.4**	**351.0**	**0.186**	**0.242**
	bottled drinking water from areas within Eastern Nile Delta (Benha and Belbeis) Cities					
S09	0.650	0.910	468.0	655.2	0.131	0.452
S10	0.220	0.640	158.4	460.8	0.044	0.318
mean			**313.2**	**558.0**	**0.087**	**0.385**
	domestic water from different Governerates					
Cairo,S11	2.610	0.490	1879.2	352.8	0.526	0.243
Inshas,S12	2.580	0.560	1857.6	403.2	0.520	0.278
Benha,S13	1.170	0.330	842.4	237.6	0.236	0.164
mean			**1526.4**	**331.2**	**0.427**	**0.228**

country	annual dose, mSv/y			
	Ra-226		Ra-228	
	min	max	min	max
U.S.A	8.5E-5	3.7E-4	0.0	2.5E-5
China	4.1E-5	0.025		
Finland	2.1E-3	10.01	9.1E-3	0.287
France	1.4E-3	0.143		
Germany	2.1E-4	0.368		
Italy	4.1E-5	0.245		
Poland	3.5E-4	9.2E-4		
Romania	1.4E-4	4.3E-3		
Switzerland	0.0	0.307	0.0	0.101
Spain	4.1E-3	0.818		
U.K.	0.0	0.037		

*) Reported data are calculated from the corresponding radioactivity levels given in table 3.17.