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Effect of Ground Water Chemistry and Surrounding Rocks on Radionuclides Distributions and their Environmental Hazard in Southwestern Sinai, Egypt

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Abstract

Ground water samples from four drilled wells with different depths were collected in May 2011 from Southwestern Sinai to study the distribution of radionuclides and their hazards effect. The depths are ranging between 30 and 150 m. The collected water has very low salinity as the total dissolved salt (TDS) is ranging between 1226 and 1836 ppm. The chemistry is mainly chloride as the anions distribution are in the order Cl^- > SO_4^{2-}> HCO_3^-> CO_3^{2-} and the cations are in the order Na^+ > Ca^{+2}> K^+> Mg^{+2}. Results shown that the correlation coefficient between Na^+ and Cl^- was strongly positive (0.99), while it was 0.82 between Ca^{2+} and SO_4^{2-}. The activity concentration of ^{238}U is ranging between 8.2 and 14.0 Bq/L and it is slightly correlated with Mg^+. ^{232}Th activity concentration is very low and ranges between 0.316 and 0.683 Bq/L, while ^{40}K ranges between 0.868 and 2.3 Bq/L. The activity concentrations of ^{222}Rn are ranging between 540.5 and 1163.3 Bq/L and its progenies (^{214}Pb, ^{214}Bi) did not reflect its concentration. Radon exhalation rate was calculated using a-track detector. The annual effective dose was calculated for the different personal ages. The babies (< one year) are the most annually dosed.

Keywords: Radioisotopes; Ground water; Radium-Radon; Exhalation rate; Hazards parameters

Introduction

Ground water naturally contains several chemical components which can lead to different kinds of health problems. According to Reiman and Banks [1], a groundwater source can potentially contain several naturally occurring chemical elements with their toxicity. Uranium and its daughter product radon are two naturally occurring elements that can lead to health problems if present in high concentrations in groundwater. Uranium is more harmful due to its toxic nature rather than its radioactivity. Ground water can either be extracted from bed rock (drilled well) or from soil aquifer (dug well). Radon is principally a problem in well drilled in bed rock that contains average or high concentrations of uranium. Regularly limits for ^{238}U and ^{222}Rn vary in different countries over the years [2], the standards and guide lines that are commonly adopted are shown in Table 1, together with the results of Ost Ostergren et al. [3].

It is worth to mention that person consuming water with a uranium content of 100 μg/L of a daily for basis, will receive a dose of 0.1 mSv/y [3]. Also a daily consumption of water with an activity concentration of 0.5 Bq/L of ^{226}Ra leads to a dose of 0.1mSv/y. The reference of 0.5 Bq/L is used as regularity limit for ^{226}Ra in Europe, an even lower value of 0.185 Bq/L (5pCi/L) is adopted for total radium (^{226}Ra and ^{228}Ra) in the US.

In groundwater, uranium (VI) as UO^{2+} ions, can form complexes with commonly existing ions in ground water such as OH^-, CO_3^{2-}, F^-, PO_4^{3-} and SO_4^{2-} [4], and may also be strongly complexed by dissolved humic substances [5]. U(VI) is also strongly bounded by Fe oxides at pH>5 [6].

The present study has been carried out to calculate the level of natural radioactivity in wells water Southwestern Sinai, Egypt. These wells are mainly the drinking water source for the people populated this area. Measurement of the activity due to ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in samples was performed by means of gamma spectrometry, using a hyper-pure germanium detector, and to measure radon and thoron concentrations by CR39- detector. Finally calculation of the hazards at different personal ages has been take place.

Geological setting

Three main rock units are exposed in the area connecting the studied four water wells their depths in meters are 150, 90, 75, and 30 named 1, 2, 3 and 4 as shown in Figure 1. The Cambrian unit (450 my) is consisted of three formations known as Sarabit El Khadim, Abu Hamata and Adediya [7]. The Adediya Formation (131 m) is composed mainly of weakly cemented sandstone and represents the main aquifer of the ground water. The Adediya Formation overlies the Abu Hamata Formation which is consisted from siltstone and shale. Sarabit El Khadim Formation is underlying Abu Hamata Formation and is consisted of sandstone and conglomerate. This unit is nonconformably overlying the granite. The second rock unit is the Um Bogma Formation

Source	Radon Bq/L	Uranium(μg/L)
WHO	----	15
USEPA	~150	30
Ostergren et al	100# 1000##	15

More than 100 Bq/L is a compulsory action level for public water plants
More than 1000 Bq/L is a compulsory action level for all kinds of water

Table 1: Standards and guide lines for radon and uranium [5,21,22].

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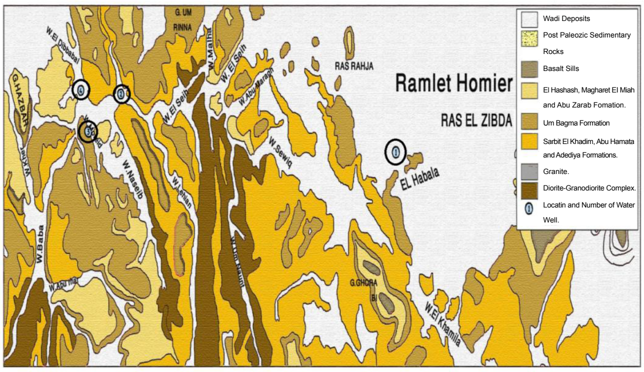

Figure 1: Geological map with locations of studied water well [23].

(0.0 – 40 m) which is unconformably overlying the Adediya Formation; the unconformity surface is poly metallic with anomalous uranium and thorium contents and shows gypsum ($CaSO_4 - 2H_2O$). The third rock unit is consisted of three formations named El Hashash Formation (60 m) mainly of sandstone, Magharet El Miah Formation (33m) mainly of carbonaceous shale and Abu Zarab Formation (100m) mainly of sandstones.

Samples collection and preparation

A total of four samples were collected from four drilled wells of different depths. Water samples are transformed to Marinelli beakers having (250 ml) volume, and sealed for a period of about four weeks before counting by gamma spectrometry then sealed and stored for four to eight weeks to prevent the escape of the radiogenic gases (^{222}Rn and ^{220}Rn), and to allow the attainment of radioactive equilibrium in the decay chain in order to reach secular equilibrium and to ensure that radiogenic gases are confined within the volume. Gamma spectrum has been collected for 72 hours for each sample.

Methodology

Chemical analyses

The total dissolved salts (TDS) are determined by evaporation of a certain volume of water sample till dryness. The difference in weight between empty container and container with precipitate is equal to the amount of the dissolved salts. SO_4^{2-} anions are determined by evaporation after precipitation with $BaCl_2$. Na^+ and K^+ are determined by flame photometric technique, while the other cations and anions are determined by titration.

HP-Ge detector and γ-spectrometry

The samples have been analyzed non-destructively, using gamma-ray spectrometry with high purity germanium (HP- Ge) detector. This detector has a relative efficiency of about 50% of the 3″ × 3″ NaI(Tl) crystal efficiency with resolution of 1.90 keV and peak/Compton ratio of 69.9:1 at the 1.33 MeV gamma transition of ^{60}Co. It is coupled to conventional electronics and connected to a multi- channel analyzer card (MCA) installed in a PC computer. The detector is shielded from the background radiation, using a 10 cm thick lead, internally lined with a 2mm copper foil. The software program (MAESTRO-32) was used to accumulate and analyze the data. The system is calibrated for energy to display gamma photo-peaks between 63 and 3000 keV.

The efficiency calibration was performed by using three well-known reference materials obtained from the International Atomic Energy Agency for U, Th and K activity measurements: RGU-1, RGTh-1andRGK-1 [8].

Uranium-238 activity was determined indirectly from the gamma rays emitted by its daughter products (^{234m}Pa) whose activities are determined from the 1001 keV photo-peaks, respectively [9]. Activity of ^{235}U was determined by its gamma ray photo-peaks; 143.8, 163.4, 185.7, and 205.3 keV [10]. The specific activity of ^{40}K was measured by its own gamma- ray at 1460.8 keV, while activities of ^{226}Ra and ^{232}Th were calculated based on the weighed mean value of the irrespective decay products in secular equilibrium. The specific activity of ^{226}Ra was measured using the 186.1 keV from its own gamma-ray (after the subtraction of the185.7 keV of ^{235}U). The specific activity of ^{214}Pb was measured using the 295.2 keV and 351.9 keV while the specific activity of ^{214}Bi was measured using the 609.3 keV. The specific activity of ^{232}Th was measured using the 338.4 keV and 911.2 keV for ^{228}Ac and 583 keV and 2614.4 keV for ^{208}Tl.

Gamma measurements

Samples are measured by γ-spectrometry using hyper pure germanium detector to determine the activity concentrations in Bq/kg for ^{238}U, ^{235}U, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{232}Th and ^{40}K.

For radiometric analysis, each sample was transferred to 250 ml capacity polyethylene Marinelli beakers after the attainment of secular equilibrium between ^{238}U, ^{232}Th and their progenies, the samples were subjected to gamma- ray spectrometric analysis. After each sample counting, an empty cylindrical plastic container (polyethylene Marinelli beaker) was placed in the detection system, for a counting period of 48 h, in order to collect the background count rates.

Radioactivity counting

The net area count after background corrections in each photo-peak was used in the computation of the activity concentration (C) in Bq kg^{-1} for each of the radionuclides in the samples using the following expression after Jibiri et al. [11]: $C(BqKg^{-1}) = \dfrac{C_n}{\varepsilon P_\lambda M_s}$ (1)

Where C_n is the count rate under each photo- peak due to each radionuclide, ε is the detector efficiency for the specific γ-ray, P_λ is the absolute transition probability of the specific γ-ray and M_s is the mass of the sample (Kg). The lowest limits of detection (LLD) were obtained from the relation [12,13]:

$$\text{LLD} = \frac{4.66 S_b}{\varepsilon X I_\gamma} \qquad (2)$$

Where S_b is the estimated standard error of the net background count rate in the spectrum of the radionuclide and Iγ is the abundance of gamma emissions per radioactive decay. The LLD values obtained were 9.347, 1.307, 1.344 and 0.025 Bq kg^{-1} for ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs, respectively [8].

Alpha particles detections

To determine the concentrations of ^{220}Rn (Thoron) and ^{222}Rn (Radon), CR39 SSNTD (Solid State Nuclear Track Detector) is used to record the produced α-particles activity. The irradiation was performed in air tied closed chamber designed such that the produced track only represents ^{220}Rn and ^{222}Rn. Figure 2 shows used chamber, A represents the CR39 detector which is used to record α-track of particles produced by ^{222}Rn gas that travel a distance more than the alpha particles range and ^{220}Rn diffusion length, B represents another

CR39 detector provided for the whole ^{220}Rn + ^{222}Rn activities rather than other α-emitter isotopes activity present in the sample, F is a filter paper that allows ^{220}Rn and ^{222}Rn to pass.

Results and Discussions

Chemical characterization of ground water

The results of chemical analyses of the studied ground water are shown in Table 2. From these results, it can be concluded that there is good correlation between Na^+ and Cl^- as shown in Figure 3. This relation defines these waters as chloride type. Ca^{2+} is more related to SO_4^{-2} than CO_3^{-2} Figure 4 and 5 it is noticed that HCO_3^- is more dominating than CO_3^{2-}. The domination of anions is in the order Cl^- >SO_4^{2-}> HCO_3^-> CO_3^{2-}, while cations are in the order Na^+ >Ca^{2+}> K^+> Mg^{2+}.

Activity concentrations

The activity concentration values in Bq/L of ^{232}Th, ^{238}U and ^{40}K for the measured samples are listed in Table 3.

The results show that the lowest ^{238}U and ^{232}Th activity concentrations are noticed in the shallowest well GW4 located in Wadi El Dibbabat in which the surrounding surface rocks are low in radioactivity. The highest ^{238}U activities are noticed in the two wells GW2 and GW3 which are located at the downstream of wadies Moried,

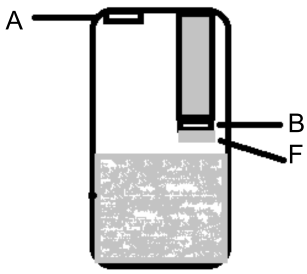

Figure 2: Air tied closed chamber for Radon & Thoron measurements.

Sample Code	pH	Ca^{2+}	Mg^{+2}	Na^+	K^+	Cl^-	CO_3^{2-}	HCO_3^-	SO_4^{2-}	TDS
GW1	7.6	86	36	232	42	376	8	82	352	1226
GW2	8.2	98	42	422	56	632	16	102	456	1836
GW3	8	84	31	242	44	391	11	70	382	1268
GW4	7.9	92	38	398	48	602	18	88	402	1696

Table 2: Results of chemical analyses (ppm) of the studied ground water.

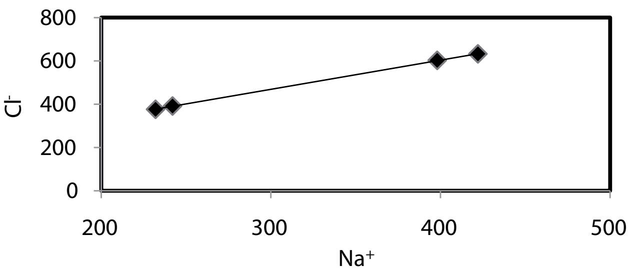

Figure 3: Cl^- vs Na^+ presented in tested ground water.

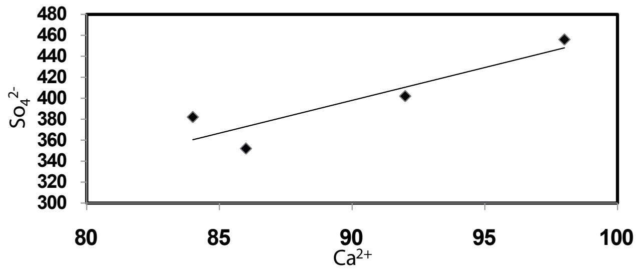

Figure 4: Correlation between Ca^{2+} and SO_4^{2-} in the studied waters.

Sample code	^{232}Th	^{238}U	^{40}K
GW1	0.656 ± 0.12	10.103 ± 0.73	2.28 ± 0.17
GW2	0.525 ± 0.08	14.004 ± 0.52	1.156 ± 0.33
GW3	0.683 ± 0.11	13.424± 1.02	0.868 ± 0.41
GW4	0.316 ± 0.02	8.214 ± 1.14	1.050 ± 0.06

Table 3: Activity concentrations of ^{232}Th, ^{238}U and ^{40}K (Bq/L) in the studied samples.

Naseib and Seih in which the uraniferous Um Bogma Formation is exposed. The highest ^{40}K content is noticed in well GW1 which is the deepest one (150m). In this depth, thin clay beds are known to occur in the section below the reservoir sandstone of Adediya Formation which may increase the K content of the groundwater in this drilled well. The relation between uranium activity concentrations vs HCO_3^-, CO_3^{2-}, Cl^- and SO_4^{2-} ; are clear in Figure 6. It is noticed that ^{238}U is more related to SO_4^{2-}, in the form of urinal sulphate $[UO_2 (SO_4)_2]^{2-}$ complexes, than the other anions. Radium is easily removed from solution by adsorption on clays and silicates or by co-precipitation with insoluble sulphate. ^{226}Ra was not detected in the studied water as it does not dissolve in water, while high concentrations of Ca^{2+}, Mg^{2+} and Cl^- are present in the studied water; this could be because these ions compete for adsorption site. Adsorption of ^{226}Ra on clays and silicates could have occurred. ^{232}Th and ^{40}K activities are very low as listed in Table 3, which may be produced due to mechanical washing during flashfloods as they are of low mobility. Activity concentrations of ^{214}Pb and ^{214}Bi have been gamma-measured and listed Table 4.

Alpha track measurements

After 4 weeks of irradiation for the exposed and unexposed CR39 detectors, they were collected and immediately etched chemically in NaOH with optimum conditions which are 6.25 N of for 6 h at constant temperature 70°C with an accuracy of + 0.1°C [14]. During the etching process the solution was constantly stirred. The detectors were then washed under running tap water for about 5 min and dried using tissue paper.

An optical microscope (400 x magnifications) was used to count the number of tracks per cm² on each detector; Figure 7 shows the counting system. The track density was converted into radon concentration Bq/L using the calibration factor of CR39 where integrating radon's concentration is known [15].

The concentrations were determined according to the equation given by Tanner [16]: $C_{Rn} = \frac{(N-B)}{tC_F}$ (3)

Where, C_{Rn} is the mean Rn-222 concentration (in Bqm^{-3}), N is the track density ($Track.cm^{-2}$), B is the background track density ($Track.cm^{-2}$), C_F is the calibration factor in terms of α- tracks. ($cm^{-2}d^{-1}$per Bqm^{-3}) and t is the exposure time (hours). Radon exhalation rate E, has been calculated using the following equation.4 [15]:

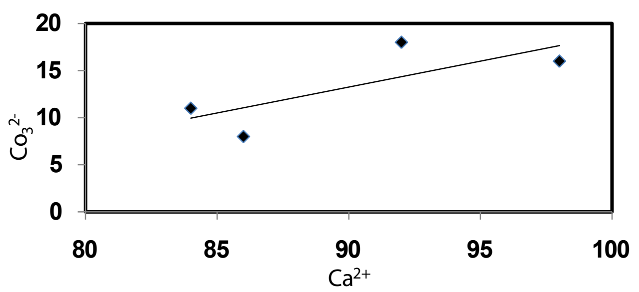

Figure 5: Correlation between Ca^{+2} and CO_3^{-2} in the studied waters.

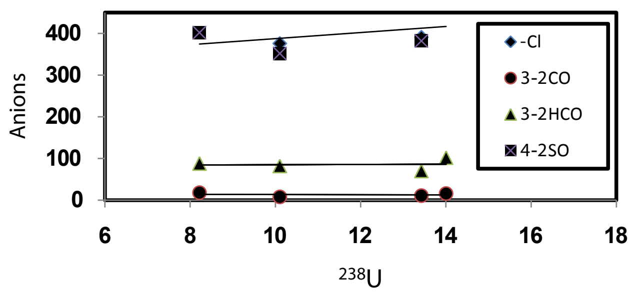

Figure 6: Uranium concentrations vs different anions.

$$E = \frac{CV\lambda}{A\left[T + \frac{(e^{-\lambda T} - 1)}{\lambda}\right]} \quad (4)$$

where A, V, λ and T are the area of the chamber in m^2, effective volume of the chamber in m^3, decay constant for radon in h^{-1} (0.00756 h^{-1}), and the exposure time in hours, respectively. The results of alpha track showed that thoron concentration was within the background level in the laboratory. Results have been listed in Table 4, radon concentrations range between 540 and 1163 Bq/L. Although the ^{214}Pb and ^{214}Bi activities measured using gamma spectroscopy show no relation with radon concentrations as shown in Figure 8, yet this may be explained as a result of radon being transmitted to water by bed rock interaction of radium migrated from other areas as previously suggested. This explains that the studied water samples did not contain radium content.

Calculation of annual effective dose

Equation.5 is used to calculate the annual effective dose due to the intake of natural radionuclides from drinking water: D = CIE (5)

Where D is the annual effective dose (Sv/ y) to an individual due to the ingestion of radionuclides from drinking water, C is the activity concentration of radionuclides in the ingested drinking water (Bq/ l), I the annual intake of drinking water (l /y) and E the ingested dose conversion factor for radionuclides (Sv/Bq) [17,18]. E values used in the calculations are listed in Table 5.

The annual effective dose has been calculated for different age groups as listed in Table 6 for babies (age below 1y), children (age from 2 to 7 y) and adults (age from 17 y and above), with annual water consumption per year, as daily drinking volume of water equal two litters. Contribution of each radionuclide depends on its ingested dose

^{222}Rn (Bq/L)	^{214}Pb (Bq/L)	^{214}Bi (Bq/L)	Radon Exhalation rate (Bq/h)
1019.16	1.36	1.24	1.40 $\times 10^{-3}$
827.824	1.46	1.55	1.12 $\times 10^{-3}$
1163.28	0.495	0.48	2.24 $\times 10^{-3}$
540.46	0.94	0.97	1.04 $\times 10^{-3}$

Table 4: Activity concentrations of Radon (Bq/L) and its progenies (Bq/L) and exhalation rate (Bq/h).

Radioisotope	Dose conversion factors (mSv Bq^{-1})		
	<1 y	2–7 y	≥ 17 y
^{214}Bi	1.4×10^{-9}	3.6×10^{-10}	1.1×10^{-10} (Ajayi and Owolabi 2007)
^{226}Ra	4.7×10^{-6}	6.2×10^{-7}	2.8×10^{-7} (Ajayi and Owolabi 2007)
^{228}Ac	7.4×10^{-9}	1.4×10^{-9}	4.3×10^{-10} (Ajayi and Owolabi 2007)
^{212}Pb	1.5×10^{-7}	3.3×10^{-8}	6.0×10^{-9} (Ajayi and Owolabi 2007)
^{235}U	3.5×10^{-7}	8.5×10^{-8}	4.7×10^{-8} (Ajayi and Owolabi 2007)
^{40}K	6.2×10^{-8}	2.1×10^{-8}	6.2×10^{-9} (Ajayi and Owolabi 2007)
^{238}U	3.4×10^{-7}	8×10^{-8}	4.5×10^{-8} IRPA 2010

Table 5: Dose conversion factors for ingestion of radionuclides in water [17,18].

Sample code	Annual effective dose		
	Age		
	≤1y	2-7y	≥17y
GW1	0.90	0.30	0.35
GW2	1.25	0.41	0.48
GW3	1.20	0.39	0.46
GW4	0.73	0.24	0.28

Table 6: Net annual effective (mSv/y) dose has been calculated for different age groups.

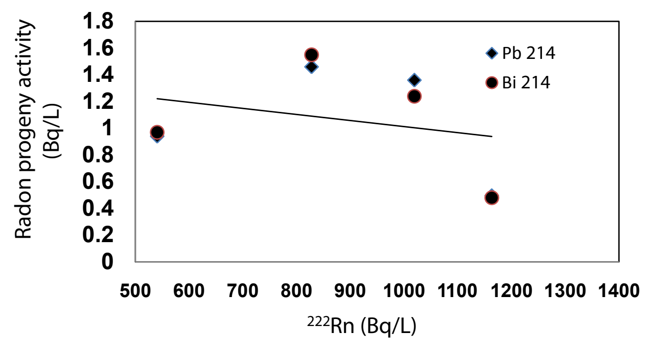

Figure 7: activity concentration or radon progenies vs radon activity concentration.

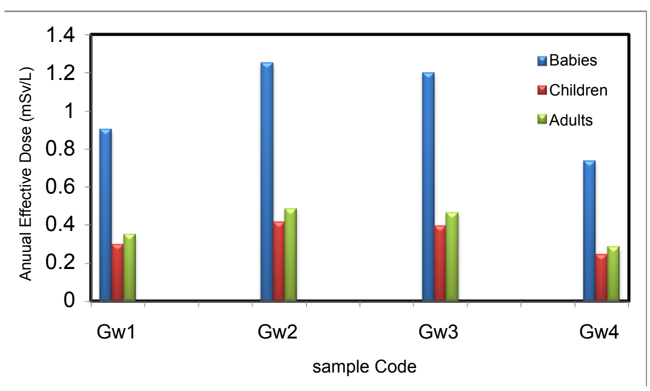

Figure 8: Annual effective dose for different age groups.

conversion factor E and its present activity. Babies are more annually dosed from these wells water intake, and the adults are the less affected by drinking water as shown in Figure 8. Similar results have been found in some areas in Yemen [19], and some aquifer systems in Brazil [20].

Conclusions

In the present study the levels of radionuclides are determined through four selected water from well water using Gamma-spectrometry with high purity germanium detectors which is a very efficient analysis technique for measuring radionuclides in environment. Also solid state nuclear track detectors are used, for radon and thoron measurements.

The mean activity concentration of the radionuclides in different samples is presented and indicates that ^{238}U concentration was significant in all measured samples. The results obtained show undetectable radium while its parent was present ^{238}U from the source rocks to water as a result of bed rock interaction. The general conclusions of this document may be listed in the following points:

1. There is a good relation between uranium in the studied samples and the uraniferous rock exposures around the wells where ^{238}U activity concentration present in the studied sample reflect the higher uranium activity of the bed rock around the wells.
2. Results of radon and its progenies measurements prove undetectable radium content; this could explain the origin of collected water as seasonally affected by young transmitted uranium.
3. Ratio of radionuclides in the studied water is completely different from normal ratio this reflect the effect of chemical properties of the water wells and their lithologies on the distribution of radionuclides.
4. Presence of significant activity ^{238}U without ^{226}Ra shows that the decay path is affected by water processing on bed rock of the studied wells.
5. Babies are more exposed to hazards due to studied water intake, so this document recommends another drinking water source for babies' stage.

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