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Estimation of Radioactivity Parameters in Sibaiyaphosphate and Behaviorof Radionuclides during Acidicleachingprocesses

E. S. Abd El-Halim[1], AmanyTahaSroor[1,*], NadiaWalley El-Dine[1], Ibrahim E. El-Aassy[2], Zeinab M. Abdel- GawadShalaby[2], Naima MAl-kbashy[3].

[1] *Faculty of Women for Arts, Science and Education, Physics Department, Ain Shams University, Cairo, Egypt*
[2] *Nuclear Materials Authority, Cairo, Egypt*
[3]*Physics Department, Faculty of Science, Tripoli University, Tripoli, Libya*
Corresponding Author: E. S. Abd El-Halim

Abstract*: The activity concentrations of natural radionuclides (^{238}U,^{226}Ra , ^{235}U,^{232}Th and ^{40}K) were measured byusing gamma spectrometry in phosphate rock samples collected from phosphate mines located in El-Sibaiya,Nile Valley , Egypt . The average activity concentration of^{238}U ,^{226}Ra , ^{235}U,^{232}Th and ^{40}K (Bq/kg) inphosphate rock were 462.8, 548.08 , 21.46, 13.22 and 147.98respectively .The radiological hazards tothe occupational workers caused byγ- radiation havebeen determined. Also thisstudy aims to examine the mobility of uranium andits isotopes and daughters during the acid leaching . To study radionuclides transfer from solid material (ore) to the liquid phase (leachate) , three samplesare prepared for leaching with two different acids .Natural radionuclides were measured by HPGe detector in original samples , pregnant solution andresiduals .The results show that the transfer of radionuclides were carried out either physically through α - recoil or chemically through dissolution . The behavior of ^{226}Ra in the acidic leaching is completely different from^{238}U and its isotopes. Selection leaching studies have shown that uranium isotopes are leached to the same extent but that is not observed in ^{234}U . The lowest leachability in all acid leaching is present in ^{226}Ra ,^{214}Pb and ^{214}Bi.*

Key Words: *Phosphaterock , Natural radioactivity , leaching , hazard index.*

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I. Introduction

Phosphaterocks contain a relatively high concentration ofnaturally occurring radioactive materials (NORM) from the uranium and thorium decay series [1].The average uranium content in phosphate rock is usuallyaroundat 50-200 parts per million(ppm)[1].Thus the measurement of natural radioactivity in phosphate rocks, which is the basicindicatorof radiological contaminationin the environment, is importantin estimating the present health risks andestablishing the base line data for future monitoring of exploration and in radiation protection[2,3].The agriculture is the main outlet for phosphate ore as these minerals are important natural sources of fertilizers, in addition to other industrial uses are increasing (detergents, pharmaceutical, metals, food, oil, textiles). For this reason economic exploitation of phosphate ore becomes very important[4].El-Sibaiya cityislocated onthe Nile River ,the presence ofphosphate mines contain an excess of NORM which present a potential risk to the population . In the present work , we measured the concentration of 226Ra, 238U, 232Th, and 40K in phosphate samples collected from some differentsites of El-Sibaiyamines usinga gamma ray spectrometer and calculated the associated absorbed dose rates[2] .In the U-series , the isotope of interest is 230Th, rather than 232Th, which is the parent of another natural decay series. The concentration and occurrence of 230Th depends on its parent 234U, although, chemically, it is reported to associate with refractory elements and the resistant fraction in sediments. 230Th is a good indicator for nuclide migration in the 238U series as it is immobile. The 226Ra/238U ratio is also a good indicator for alteration processes [5] and migration of either 238U or 226Ra. [6].Uranium isotopes can dissolve from minerals and rocks and enter solution through chemical and physical processes. Physical processes involve α-recoil while chemical processes involve leaching through either acidic or alkaline reagents or any other leaching solution[7].Thus, observed fractionation between 234U and 238U is generally ascribed to selective leaching,α-recoil transfer of234Th directly into the aqueous phase, or a combination of the two processes. The principal aim of this work is tostudy radionuclide transfer from solid material (ore) to the liquid phase (leachate)[7]. Acid leaching is known to be predominant process for uranium recovery from ores, usually with sulfuric acid because its relatively low cost [8,9].

Geologic setting

The Sibaiya area is located in the Nile Valley of Egypt at the intersection of latitude25^{0}05 ' N and longitude 32^0 42 ' E .As shown in figure (1) .

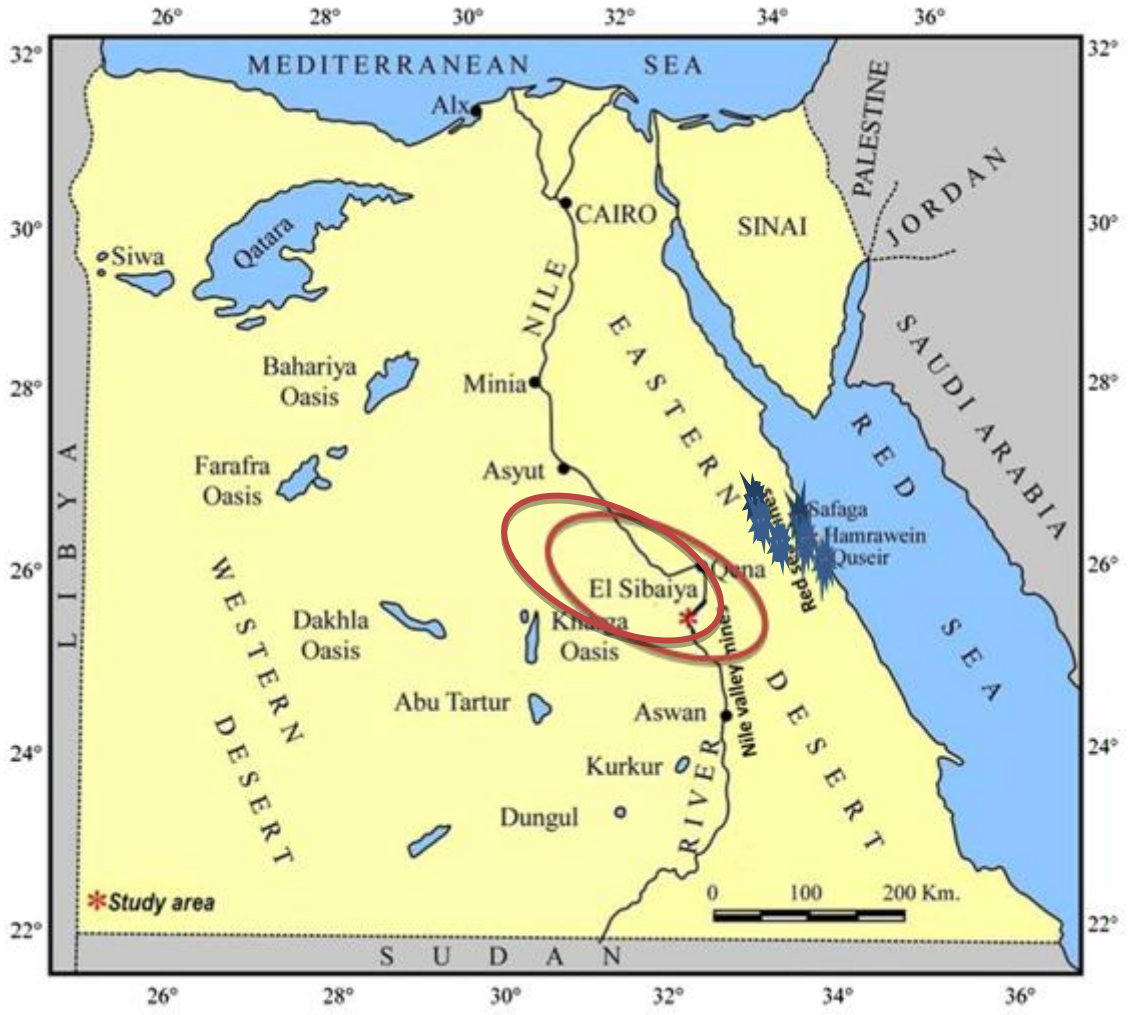


Fig (1) Location of El-Sibaiyaarea [4]

In the region of El-Sibaiya the phosphorite - bearing strata are known as the DuwiFormtion**[10]**.The DuwiFormation (Phosphate – bearing sediments) is characterized by the presence of three types of sections: a claysection , a sandy- clay section and a sandy section .Three phosphorite - bearing members : lower , middle and upper are recognized in this formation.The phosphate beds could be considered as a probable source of uranium . Where their average content reaches 130 ppm . The Egyptian phosphate beds is due to the accumulation of bones and phosphates derived from the decay of soft parts of organisms and the enrichment of the phosphorite - bearing limestone through the dissolution of calcium carbonates by carbonated waters .There is evidence in the field that processes of metasomatic replacement of calcium carbonate byphosphate took place at Sibaiya **[11]**. The Duwi Formation starts with thick yellow to yellowish grey, pebbly phosphatic bed (Bed A) with 3 m thickness (Fig2). It istopedwith hard silicified phosphate bed (10 cm) which isfollowed with hard oyster limestone, grey in colour with thin phosphateinterbed (Bed B) of 10 cm thickness and characterized by the presence of shark teeth. Up in the section, the calcareous content is represented by medium hard marl, pale yellow in color with thickness around 1.5 m, intercalated with thin interbred of chert of about 10 cm thicknesses. It overlies by yellow to yellowish grey, fissile shale with a thickness of 25 cm. The phosphatic content is returned againwith Bed C that characterized by yellowish grey to brownish grey, medium hard, coated by iron oxides andcontains shark teeth; its thickness is 20 cm. This bed forms the top of the Duwi Formation**[4]**.

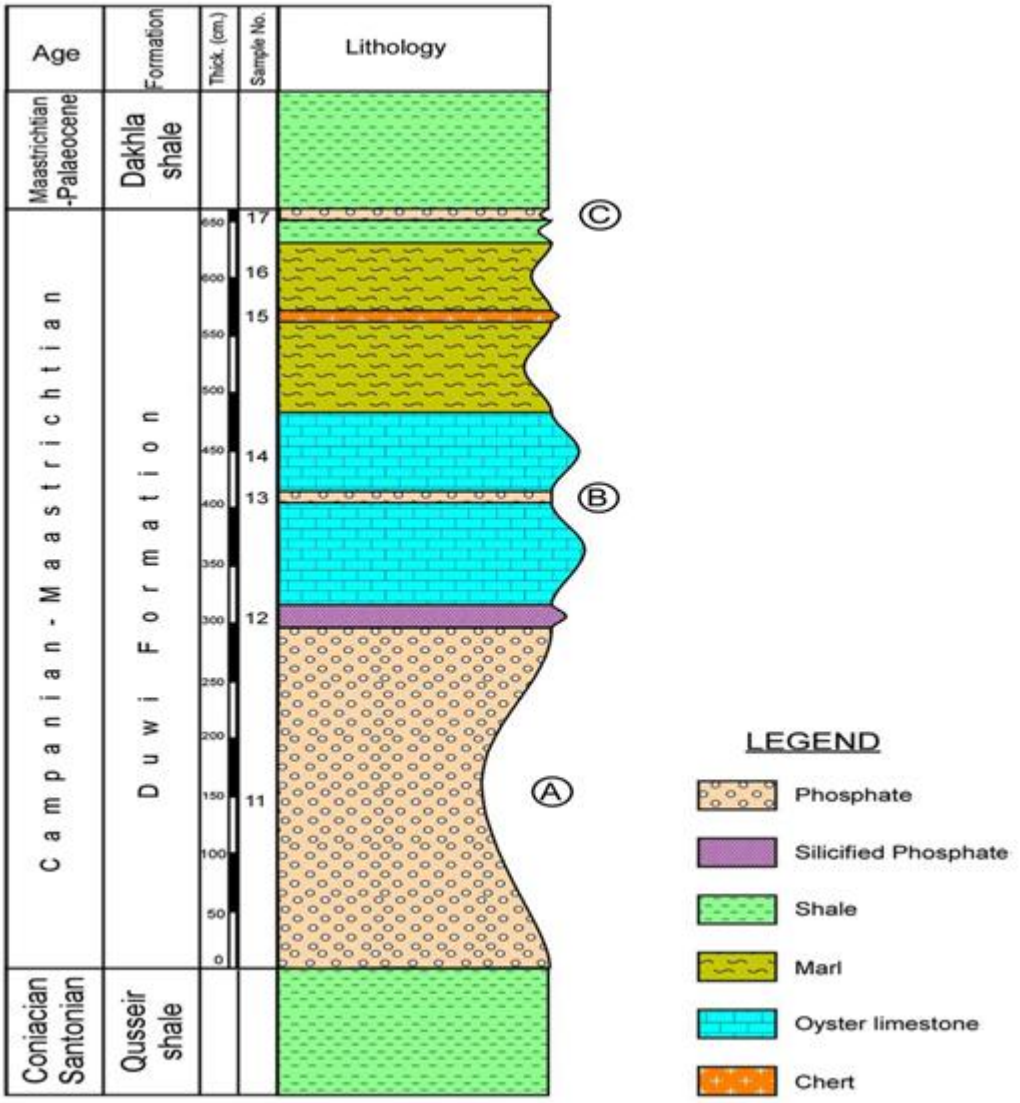


Fig (2) Lithostratigraphic succession of DuwiFormation exposed in Nile Valley locality [4].

Experimental Procedures

Sampling and samples preparation

For radioactivity measurements , a total of 12 samples each of mass 1 kg , were collected from Sibaiya area is located in the Nile Valley region , Egypt . Thecollected samples were dried at room temperature for a week crushed , homogenized and sieved through 200 mesh size . The samples were first weighted and placed in polyethylene bottles of 250cm^3 volume .The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reachedbefore measured by the gamma spectrometer . This stepwas necessary to ensure thatradongas is confinedwithinthevolume and the daughters still also remain in the sample . Three phosphate samples (Q7 , Q9 at Quseir – Safaga) and (S2 at Sibaiyaarea).Samplesare poured from the containers and prepared for leaching experiments using two acids :HCland HNO_3 . Leaching on 150g sample weight under the conditions : solid /liquid ratio 1:3 acid concentration (110 gm/L) , stirring time two hours and at room temperature .Filtration was carried out to separate leachate from residual which is dried at room temperature . The residual and leachate have been weighted and packed will in polyethylene bottles of 250ml volume and stored for more than a month for measuring by HPGe detector. The leachabilityof the radionuclides was calculated according to the following equation:

About 5 g of each sample was ground to -200 mesh for chemical analyses of major oxides and trace elements content, by the X-ray fluorescence technique (XRF).

Experiment set up

High purity vertical germanium was coupled to a personal computer with a special electronic card to make it equivalent to a multichannel analyzer. The system also contains the usual electronic components of preamplifier, amplifier and power supply. The detector has resolution (FWHM) of 1.85 keV for the 1332.5 keV γ-ray line of ^{60}Co. The γ-ray spectrometer energy calibration was performed using ^{60}Co, ^{226}Ra and ^{241}Am point sources. The detector was surrounded by a special heavy lead shield of 10 cm thickness with inside dimensions of 28 cm diameter and 40 cm height. The absolute detection efficiency of the HPGe detector was determined by using three well-known reference materials obtained from the International Atomic Energy Agency for U, Th and K activity measurements: RGU-1, RGTh-1 and RGK-1 **[12, 13].** The sample containers were placed on top of the detector for counting. The same geometry and size were used for both the samples and the reference materials **[14].** The uranium standard (RGU-1) is U-ore diluted with silica with 4940 Bq.kg^{-1} of ^{238}U, 228 Bq.kg^{-1} of ^{235}U, a negligible amount of ^{40}K (less than 0.63 Bq.kg^{-1}) and some traces of ^{232}Th (less than 4 Bq.kg^{-1}). The thorium standard (RGTh-1) is Th-ore diluted with silica having 3250 Bq.Kg^{-1} of ^{232}Th, but containing some ^{238}U (78 Bq.kg^{-1}) and ^{40}K (6.3 Bq.kg^{-1}). The potassium calibration standard (RGK-1) is produced from high purity (99.8 %) potassium sulphate with 14000 Bq.kg^{-1} of potassium with uranium and thorium contents lower than 0.001 and 0.01 ppm, respectively **[13].** The γ-ray transitions used to measure the concentration of the assigned nuclides in the series are the following. ^{238}U was determined from the gamma rays emitted by its daughter products **[15]**^{234}Th and ^{234m}Pa activities determined from the 63.3 and 1001 keV photo peaks, respectively, ^{214}Bi (609.3, 1120.3, 1238.1, 1377.7 and 1764.5 keV), ^{214}Pb (295.1 and 352.0 keV). The specific activity of ^{226}Ra was measured using the 186.1 keV from its own gamma-ray (after the subtraction of the 185.7 keV of ^{235}U). The specific activity of ^{232}Th was measured using the 338.4, 911.2 and 968.9 keV lines from ^{228}Ac and 583 keV peak from ^{208}Tl, and ^{40}K was measured using 1460.8 keV peak.In order to determine the background contribution due to naturally occurring radionuclides in the environment around the detector, an empty polyethylene beaker of the same 250 cm^3 volume was counted with the same geometrical conditions as the sample. The measurement time for both activity and background measurement was 70000 s. The background spectra were used to correct the net gamma- ray peak areas for the studied isotopes.

II. Results and discussion

Activity Concentration

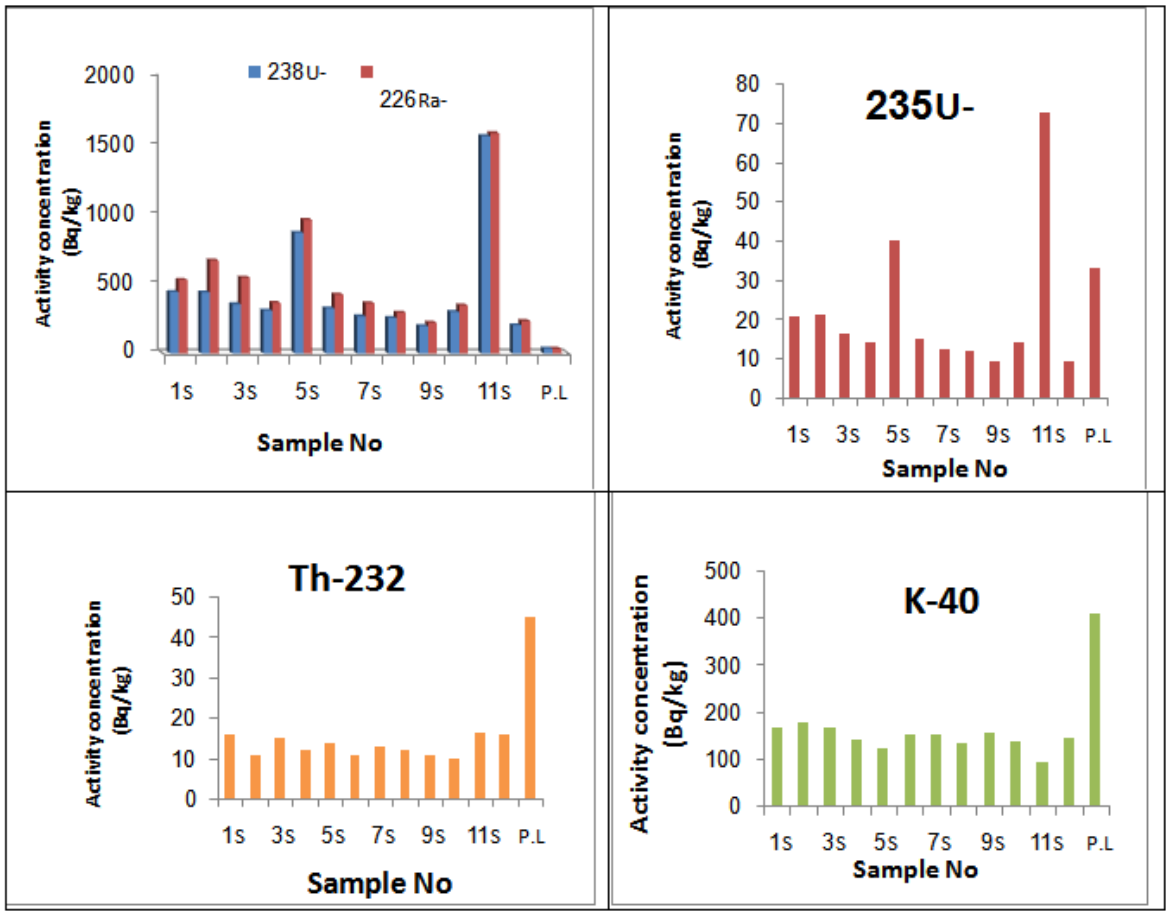
Activity concentration of ^{226}Ra , ^{238}U, ^{235}U ,^{232}Th and ^{40}K for El-Sibaiya phosphate rockssamples were calculatedas illustrated (in Table 1).The activity concentrations of ^{238}U range (from197.08 to1577.31) Bq/kg, ^{226}Ra(from 221.4to1599)Bq/kg , ^{235}Ufrom (9.21 to73.14) Bq/kg , ^{232}Th (from 10.38 to 16.44) Bq/kg and^{40}K (from96.38to 180.8)Bq/kg. The radioelements worldwide average (W.A.) values are 33 Bq/kg for ^{238}U, 32 Bq/kg for ^{226}Ra, 45 Bq/kg for ^{232}Th , 412 Bq/kg for ^{40}K and 33 Bq/kg for ^{235}U **[16]** as shown infig (3)**.** The activity concentrations of all studied samples for radium , uranium are higher than the permissible level**,** but the activity concentrations of all studied samples for thorium and potassiumare lower than the permissible level, the activity concentrations of most studied samples for^{235}Uare lower than the permissible level except samples (S5, S11)are higher than the permissible level.

Table (1) : The activity concentrations in (Bq/kg) of ^{238}U , ^{226}Ra , ^{235}U, ^{232}Th, ^{40}K and ^{226}Ra/^{238}U for phosphate samples at Sibayia area .

Samples	^{238}U	^{226}Ra	^{235}U	^{232}Th	^{40}k	^{226}Ra/^{238}U
S1	444.14	533.5	20.76	16	168.8	1.2
S2	442.76	673.82	21.03	10.97	180.8**	1.52
S3	355.52	548.18	16.32	15.35	170.3	1.54**
S4	309.57	365.7	14.12	12.11	144	1.1
S5	874.46	969.9	40.3	14.11	125.2	1.1
S6	326.1	426.9	15.05	11.12	155.5	1.3
S7	267.85	364.2	12.49	13.32	155.6	1.3
S8	256	294.2	11.78	12.08	134.3	1.1
S9	197.08*	221.4*	9.21*	10.83	158	1.1
S10	299.96	346.9	13.99	10.38*	140.1	1.1
S11	1577.31**	1599**	73.14**	16.44**	96.38*	1*

S12	202.91	233.3	9.33	16	146.8	1.1
Average	**462.8**	**548.08**	**21.46**	**13.22**	**147.98**	**1.2**
P.L	**33**	**32**	**33**	**45**	**412**	**unity**

The lowest value** * The highest value**



Fig(3) The activity concentrations for ^{238}U,^{226}Ra,^{235}U,^{232}Th, ^{40}K in the phosphate samples in comparison with the permissible level (p.l.).

In this work the activity concentration of ^{238}U and ^{232}Th in phosphate samples (in ppm) and activity ratio$^{232}Th/^{238}U$ were calculated and illustrated in (table 2).The concentrations of ^{238}U range between 15.89 and 127.2ppm, and ^{232}Th between 2.55and 4.04 ppm, while the $^{232}Th/^{238}U$ ratios range between 0.03and 0.24 which is lower than the Clark's value (3.5), which indicates that these phosohatesare enriched in uranium.

Table (2): Results of radionuclide concentrations (ppm %) inthe phosphate samples , Clark value .

Sample	^{238}U (ppm)	^{235}U (ppm)	^{232}Th (ppm)	^{40}K %	$^{232}Th/^{238}U$
S1	35.81	0.25	3.94	0.53	0.11
S2	35.7	0.26	2.7	0.57**	0.07
S3	28.67	0.2	3.78	0.54	0.13
S4	24.96	0.17	2.98	0.46	0.11
S5	70.52	0.5	3.47	0.4	0.04
S6	26.29	0.18	2.73	0.49	0.1
S7	21.6	0.15	3.28	0.49	0.15
S8	20.64	0.14	2.97	0.42	0.14
S9	15.89 *	0.11 *	2.66	0.5	0.16
S10	24.19	0.17	2.55*	0.44	0.1
S11	127.2**	0.91**	4.04 **	0.3*	0.03*
S12	16.36	0.11	3.94	0.46	0.24**

*** The lowest value** **** The highest value**

Radiologicalhazard indices

Radium Equivalent (Ra_{eq}):

Radium equivalent (Ra_{eq}) index in Bq/kg is a widely used radiological hazard index. It is a convenient index to compare the specific activities of samples containing different concentrations of ^{226}Ra, ^{232}Th and ^{40}K. It was defined on the assumption that 10Bq/kg of ^{226}Ra, 7Bq/kg of ^{232}Th and 130Bq/kg of ^{40}K produce the same gamma dose rate. It was calculated as follows.

$$Ra_{eq}=C_{Ra}+1.43\ C_{Th}+0.077C_k \quad \text{…….. (1)}$$

Where C_{Ra}, C_{Th} and C_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. Ra_{eq} was estimated for the collected samples and are given in Table (3), were estimated higher than the recommended maximum value of 370 Bq kg^{-1} except (S8 ,S9 , S12) lower than the recommended maximum value**[17].**.

External Hazard Index (Hex) :

The external hazard index (H_{ex}) represents the external radiation exposure associated with gamma irradiation from radionuclides of concern. The value of H_{ex} should not exceed the maximum acceptable value of one in order to keep the hazard insignificant. The external hazard index (H_{ex}) is defined by equation **[18]:**

$$H_{ex}=(C_{Ra}/370+C_{Th}/259+C_k/4810)\leq 1 \quad \text{…… . (2)}$$

Where C_{Ra}, C_{Th} and C_K are the concentration in(Bq Kg^{-1}) of radium, thorium and potassium respectively.The values of external hazard Indices for all samples are found to be more than 1 except samples (S8 ,S9 , S12) lower than the recommended maximum valueand sample S10 is found at unity (shown Table (3) , Fig (4).

Radiation Level Index (I_γ) :

This index can be used to estimate the level of γ-radiation hazard associated with the natural radionuclides in the samples; it is given by the equation

$$I_\gamma = C_{Ra}/150 + C_{Th}/100 + C_K/1500 \quad \text{…… (3)}$$

whereC_{Ra}, C_{Th} and C_K are the activity concentration of ^{226}Ra, ^{232}Th, ^{40}K in Bq/kg, respectively**.** The value of these indices must be less than unity in order to keep the radiation hazard insignificant , the values of Representative Level

Index (I_γ) for all samples are found to be more than 1 given in Table (3) ,Fig (4).

Absorbed Dose Rate D:

The absorbed gamma dose rates in air at 1 m above the ground surface for the uniform distribution of radionuclides (^{238}U, ^{232}Th and ^{40}K) were calculated by using equation :

$$D = 0.427{*}C_U + 0.662{*}C_{Th} + 0.043{*}C_K \text{ (nGy/h)} \quad \text{…… (4)}$$

where C_U, C_{Th} and C_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K in Bq/kg, respectively. The range of absorbed dose rate in the samples due to natural radionuclidesis (98.09–688.5) nGy h^{-1} given in Table (3) . The values of the Dose rate calculated during present study are found higher than the permissible level of 59 nGy/h**[19].**

The annual outdoor effective dose (E_{out}) is estimated from the outdoor external dose rate (D_{out}), time of stay in the outdoor or occupancy factor (OF = 20 % of 8760 h in a year) and the conversion factor (CF = 0.7 Sv**.**Gy^{-1}) to convert the absorbed dose in air to effective dose. During the present study, the E_{out} was calculated using the following equation from**[16]** :

$$E_{out} = D_{out}\ (nGy\ h^{-1}) * 0.2 * 8760\ h * 0.7\ (Sv * Gy^{-1}) \quad \text{……..(5)}$$

The outdoor effective dose rate E_{out} range from (120.26to 844.1mSv/y) , the values of the all studied samples listed (in table 3) are found higher than the world's average 0.07 mSv/y **.**

Excess lifetime cancer risk (ELCR) :

The value of annual effective dose excess lifetime cancer risk (ELCR) was calculated by using the equation :

$$ELCR_{out} = E_{out} * LE * RF \quad \text{…….(6)}$$

WhereE_{out} is the annual effective dose , LE life expectancy (66 years) and RF (Sv^{-1}) is risk factor per Sievert, which is 0.05 . The values of$ELCR_{out}$ range between ($396.87x10^{-3}$- $2785.55x10^{-3}$) in the phosphate samples Table (3). All values are higher than the permissible level $0.29 * 10^{-3}$**[20].**

Table (3): The values of radium equivalent (Bq/kg), radioactivity level index, external hazard, the outdoor absorbed rate (nGy/h), outdoor annual effective dose (mSv/y) and outdoor Excess lifetime cancer risk at Sibayia area .

Sample	Ra_{eq} (Bq/kg)	I_γ	H_{ex}	$D_{(out)}$ (nGy/h)	$E_{eff(out)}$ (mSv/y)	$ELCR_{(out)}$ $x10^{-3}$
S1	569.37	3.82	1.53	207.46	254.35	839.36
S2	703.42	4.72	1.9	204.07	250.19	825.63
S3	583.24	3.92	1.57	169.26	207.51	684.79
S4	394.1	2.65	1.06	146.37	179.45	592.18
S5	999.71	6.69	2.7	388.09	475.79	1570.13
S6	454.77	3.06	1.22	153.27	187.9	620.1
S7	395.22	2.66	1.06	129.85	159.2	525.36
S8	321.81	2.17	0.86	123.05	150.87	497.87
S9	249.05*	1.68*	0.67*	98.09*	120.26*	396.87*
S10	372.53	2.5	1.0	140.95	172.81	570.28
S11	1629.93**	10.88**	4.4**	688.5**	844.1**	2785.55**
S12	267.48	1.81	0.72	103.51	126.9	418.8
P.L	**370 Bq/kg**	**unity**	**unity**	**59 $nGyh^{-1}$**	**0.07 $mSvy^{-1}$**	**0.29*10^-3**

***The lowest value** ****The highest value**

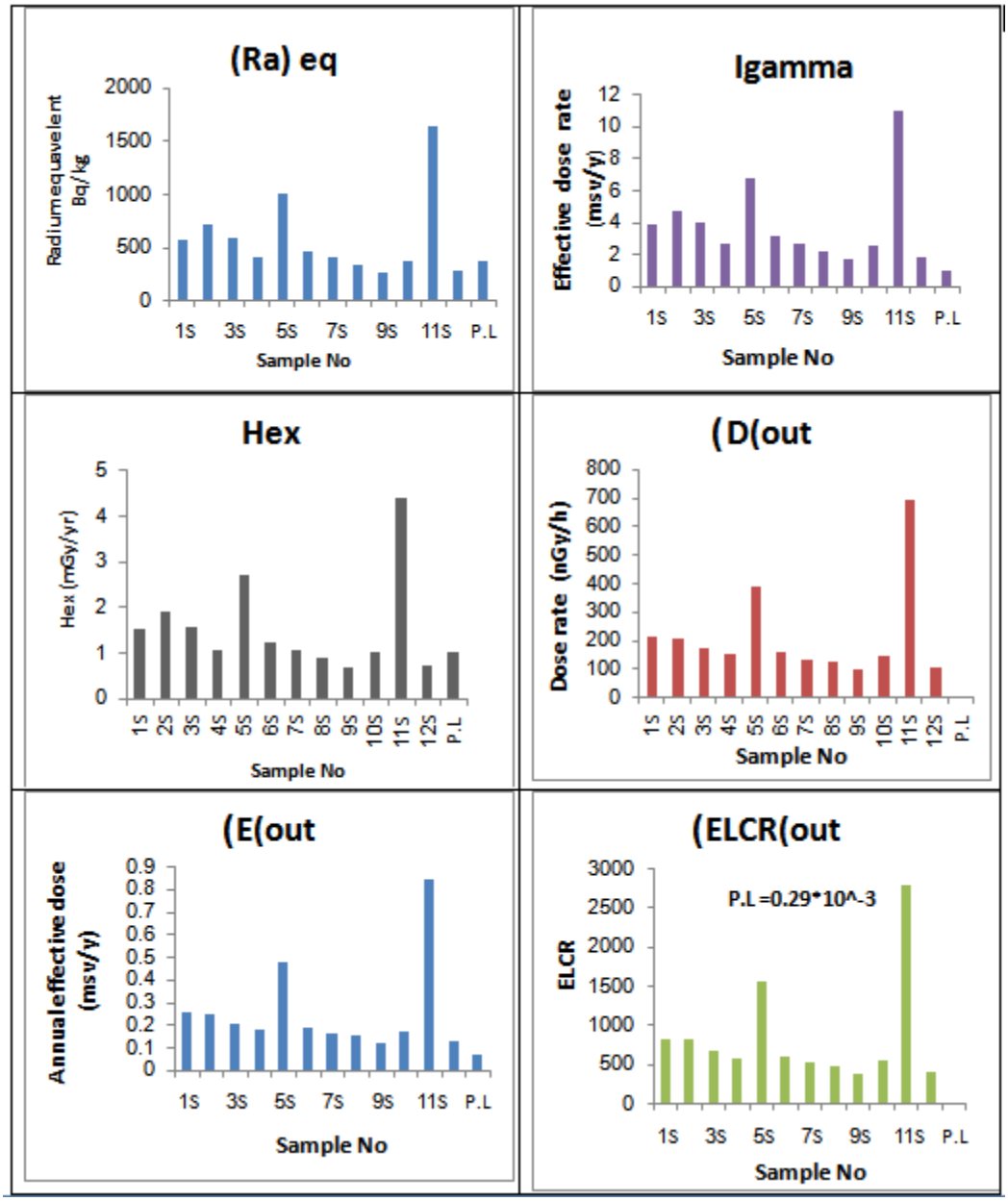


Fig (4) The values of radium equivalent (Bq/kg), radioactivity level index, external hazard, the outdoor absorbed rate (nGy/h), outdoor annual effective dose (mSv/y) and outdoor Excess lifetime cancer risk at Sibayia area .

LeachingStudies

Table (4) Activity concentrations of radionuclides in original samples, residuals and leachates (pregnant solutions).

Sample Q7 leaching by HCl					
Radionuclide	**Original (Bq/Kg)**	**Residual (Bq/Kg)**	**Solution (Bq/l)**	**Leachability %**	**Summation (Residual +solution) %**
^{238}Useries					
^{234}Th	811.29 ± 24.3	687.65±20.6	173.7 ± 5.2	21.41	106.17
^{234m}Pa	797.94± 23.9	702.4 ±21	132.96 ± 3.9	16.66	104.69
Average	804.62±24.1	695.02±20.8	153.33± 4.5	19.05	105.43
^{234}U	696.15±20.8	587.29±17.6	174.27 ±5.2	25.03	109.39
^{230}Th	657.36 ±19.7	696.57±20.8	22.62± 0.6	3.44	109.4
^{226}Ra subseries					
^{226}Ra	964.81±28.9	1035±31	101±3	10.46	117.74
^{214}Pb	682.75±20.4	457.23±13.7	21.35± 0.6	3.12	70.09
^{214}Bi	701.93± 21	616.59±18.4	78.61± 2.3	11.19	99.04
^{235}U	37.63±1.1	36.08±1	7.16± 0.2	19.05	114.93
^{232}Th series					
^{228}Ac	12.6± 0.3	9.49±0.2	3.67± 0.1		104.44
^{208}Tl	14.79±0.4	13.09± 0.3	4.42± 0.1		118.31
Average	13.69± 0.4	11.29±0.3	4.05± 0.1		112.05
^{40}K	**116.4**± 3.4	**1174± 35**	**253.16±7.5**		**1226.08**
^{238}U/^{235}U	21.38	19.26	21.38		
^{234}U/^{235}U	18.5	16.27	24.3		
^{234}U/^{238}U	0.86	0.84	1.13		
^{226}Ra/^{238}U	1.19	1.48	0.65		

Table (5) Continued

Sample Q7 leaching by HNO_3					
Radionuclide	**Original (Bq/Kg)**	**Residual (Bq/Kg)**	**Solution (Bq/l)**	**Leachability %**	**Summation (Residual +solution) %**
^{238}Useries					
^{234}Th	811.29±24.3	665.18±19.9	155.59± 4.6	19.17	101.16
^{234m}Pa	797.94±23.9	702.4 ±21	141.43±4.2	17.72	105.75
Average	804.62±24.1	683.79±20.5	148.51±4.4	18.45	103.44
^{234}U	696.15± 20.8	537.71±16.1	220.36 ±6.6	31.65	108.89
^{230}Th	657.36±19.7	409.75±12.2	76.23±2.2	11.59	73.93
^{226}Ra subseries					
^{226}Ra	964.81±28.9	931.59±27.9	197±5.9	20.41 8	116.97
^{214}Pb	682.75±20.4	637.8±19.1	17.17 ± 0.5	2.51	**95.93**
^{214}Bi	701.93±21	743±22.2	76.02±2.2	10.83	116.68
^{235}U	37.63±1.1	31.03±0.9	6.98 ± 0.2	18.56	101.02
^{232}Th series					
^{228}Ac	12.6±0.3	11.79±0.3	2.65±0.07		114.63
^{208}Tl	14.79± 0.4	13.09 ±0.3	4.06±0.1		115.89
Average	13.69±0.4	12.44 ± 0.3	3.35± 0.1		115.35
^{40}K	**116.4±3.4**	**950.59±28.5**	**358.78±10.7**		**1124.89**
^{238}U/^{235}U	21.38	22.03	21.25		
^{234}U/^{235}U	18.5	17.33	31.54		
^{234}U/^{238}U	0.86	0.78	1.48		
^{226}Ra/^{238}U	1.19	1.36	1.32		

Table (6) Continued

Sample Q9 leaching by HCl					
Radionuclide	**Original (Bq/Kg)**	**Residual (Bq/Kg)**	**Solution (Bq/l)**	**Leachability %**	**Summation (Residual +solution) %**
^{238}Useries					
^{234}Th	1135.47 ±34	1101.6 ±33	154.22±4.6	13.58	110.59
^{234m}Pa	1108.48±33.2	967.18±29	151.74±4.5	13.68	100.94
Average	1121.97±33.6	1034.39 ±31	152.98±4.5	13.63	105.83
^{234}U	1008.11±30.2	611.31±18.3	292.34±8.7	28.99	89.63
^{230}Th	544.87±16.3	677.06±20.3	40.59±1.2	7.45	131.71
^{226}Ra subseries					
^{226}Ra	2076± 62.2	2097.25±62.9	77.62 ± 2.3	3.73	104.76
^{214}Pb	968.43±29	497±14.9	72.2±2.1	7.45	58.77
^{214}Bi	1200±36	1171±35.1	105.2±3.1	8.76	106.35
^{235}U	51.39±1.5	53.97 ±1.6	7.1± 0.2	13.81	118.84
^{232}Th series					
^{228}Ac	10.29 ± 0.3	7.18±0.2	3.97±0.1		108.51
^{208}Tl	6.94 ±0.2	4.5±0.1	3.66 ±0.1		117.64
Average	8.61 ± 0.2	5.84±0.1	3.82±0.1		112.22
^{40}K	**118±3.5**	**1755.53 ±52.6**	**284±8.5**		**1728.41**
^{238}U/^{235}U	21.83	19.16	21.53		
^{234}U/^{235}U	19.61	11.32	41.14		
^{234}U/^{238}U	0.89	0.59	1.91		
^{226}Ra/^{238}U	1.85	2.02	0.5		

Table (7) Continued

Sample Q9 leaching by HNO_3					
Radionuclide	**Original (Bq/Kg)**	**Residual (Bq/Kg)**	**Solution (Bq/l)**	**Leachability %**	**Summation (Residual +solution) %**
^{238}Useries					
^{234}Th	1135.47±34	934.85± 28	144.92±4.3	12.76	95.09
^{234m}Pa	1108.48±33.2	702.4± 21	170.28±5.1	15.36	78.72
Average	1121.97±33.6	818.62±24.5	157.6 ±4.7	14.04	87.01
^{234}U	1008.11±30. 2	943.32 ± 28.2	218.16±6.5	21.64	115.21
^{230}Th	544.87±16.3	532.67±15.9	36.94±1.1	6.77	104.54
^{226}Ra subseries					
^{226}Ra	2076±62.2	2100±63	208.1±6.2	10.02	111.18
^{214}Pb	968.43±29	1030±30.9	21.87±0.6	2.25	108.61
^{214}Bi	1200±36	1370±41.1	67.29±2	5.6	119.77
^{235}U	51.39±1.5	37.95±1.1	7.21±0.2	14.02	87.89
^{232}Th series					
^{228}Ac	10.29±0.3	8.82±0.2	3.28±0.09		117.59
^{208}Tl	6.94 ±0.2	4.9±0.1	2.6±0.07		108.05
Average	8.61 ±0.2	6.86±0.2	2.94±0.08		113.8
^{40}K	**118±3.5**	**3668±110**	**508.3±15.2**		**3539.38**
^{238}U/^{235}U	21.83	21.56	21.85		
^{234}U/^{235}U	19.61	24.85	30.24		
^{234}U/^{238}U	0.89	1.15	1.38		
^{226}Ra/^{238}U	1.85	2.56	1.32		

Table (8) Continued

Sample S2 leaching by HCl					
Radionuclide	**Original (Bq/Kg)**	**Residual (Bq/Kg)**	**Solution (Bq/l)**	**Leachability %**	**Summation (Residual +solution) %**
^{238}Useries					
^{234}Th	444.31±13.3	471.92 ±14.1	143.22±4.2	32.23	138.45
^{234m}Pa	441.21±13.2	301.02 ± 9	146.72±4.4	33.51	102.27
Average	442.76±13.2	386.47±11.5	144.97 ± 4.3	32.87	120.49
^{234}U	447.35 ±13.4	410.77±12.3	213.95±6.4	47.82	139.64
^{230}Th	545.41±16.3	409.75±12.2	30.99±0.9	5.68	80.8
^{226}Ra subseries					
^{226}Ra	673.82±20.2	665.42±19.9	38.77 ±1.1	5.75	104.5
^{214}Pb	424.43±12.7	346.5±10.3	6.82±0.2	1.6	83.24
^{214}Bi	439.53±13.1	405.67±12.1	40.66±1.2	9.25	101.54
^{235}U	21.03±0.6	19.87±0.5	7.31± 0.2	34.76	129.27
^{232}Th series					
^{228}Ac	13.81 ±0.4	9.41 ±0.2	1.36 ±0.04		78.06
^{208}Tl	**8.11 ±0.2**	**14.72±0.4**	**2.39 ±0.07**		**210.86**
Average	10.96±0.3	12.07± 0.3	1.88 ±0.05		127.21
^{40}K	**180.81±5.4**	**1062.43 ±31.8**	**325.94 ± 9.7**		**767.83**
^{238}U/^{235}U	21.05	19.44	19.82		
^{234}U/^{235}U	21.26	20.76	29.25		
^{234}U/^{238}U	1.01	1.06	1.47		
^{226}Ra/^{238}U	1.52	1.72	0.26		

Table (9) Continued

Sample S2 leaching by HNO_3					
Radionuclide	**Original (Bq/Kg)**	**Residual (Bq/Kg)**	**Solution (Bq/l)**	**Leachability %**	**Summation (Residual +solution) %**
^{238}Useries					
^{234}Th	444.31±13.3	369.12±11	173.03 ± 5.1	38.94	122.02
^{234m}Pa	441.21±13.2	235.45±7	155.63±4.6	35.55	89.33
Average	442.76 ± 13.2	302.29 ±9	164.33 ±4.9	37.26	105.8
^{234}U	**447.35 ±13.4**	**573.22±17.1**	**198.87 ±5.9**	**44.45**	**172.59**
^{230}Th	**545.41± 16.3**	**961.5 ±28.8**	**38.58 ± 1.1**	**7.07**	**183.36**
^{226}Ra subseries					
^{226}Ra	673.82± 20.2	665.06 ±19.9	85.46 ±2.5	12.68	111.38
^{214}Pb	**424.43 ± 12.7**	**613.29±18.3**	**13.97 ±0.4**	**3.29**	**147.79**
^{214}Bi	439.53± 13.1	562 ±16.8	13.53±0.4	3.07	130.94
^{235}U	21.03±0.6	13.83 ±0.4	7.96 ±0.2	37.86	103.61
^{232}Th series					
^{228}Ac	13.81 ± 0.4	11.05±0.3	1.45 ±0.04		90.5
^{208}Tl	**8.11 ±0.2**	**11.51±0.3**	**2.44 ±0.07**		**172.02**
Average	10.96± 0.3	11.28±0.3	1.95±0.05		120.67
^{40}K	**180.81±5.4**	**1115.31±33.4**	**321.52±9.6**		**794.63**
^{238}U/^{235}U	21.05	21.85	20.63		
^{234}U/^{235}U	21.26	41.44	24.88		
^{234}U/^{238}U	1.01	1.89	1.2		
^{226}Ra/^{238}U	1.52	2.2	0.52		

Radiometric Measuremrnts

The results of γ-detection of radionuclides in the original samples (Bq/kg), residuals (Bq/kg) and leachates (Bq/l) are collected in tables (4-9). The activity concentration of ^{238}U in the original samples, residuals and pregnant solutions for all samplesare higher than the typical world average value of **33**Bq/kg **[16]**.The behavior of each radionuclide in the samples shows that the ^{226}Ra, ^{214}Pb and ^{214}Bi activity concentrations are higher in the residual than that in the original for all samples exceptsamples Q7, Q9 leaching by HCl.The activity concentration of ^{226}Ra in the original samples, residuals and pregnant solutions are higher than the typical world average value of **32** Bq/kg**,[16].** For ^{232}Th, all the original samples ,residuals andpregnant solutionsare lower than the permissible level **45**Bq/kg**.** For all samples^{40}K, the original samples are lower than the permissible level **412**Bq/kg whilethe residualsare higher **..**

Geochemical Characterization of Samples

Three phosphate samples (Q7 , Q9 at Quseir – Safaga) and (S2 at Sibaiyaarea)were prepared and analyzed chemically for determination of major oxides order of a (%) . The distributions of these components are of significant values of SiO_2 , Al_2O_3 , Fe_2O_3, TiO_2 , CaO , MgO , Na_2O , K_2O , P_2O_5 and L.O.I(loss on ignition), table (10) . From this table, the sample (S2) has high level of SiO_2 ,CaOandL.O.I contents are 45.57, 23.40 and 12.50 % respectively . The sample (Q7) has high level of CaO and L.O.I which contents are 44.00 , 32.02% respectively . The sample (Q9) has high level of CaO , $P_2O_5$and L.O.Iwhich contents are 50.40 , 20.50and 19.10%respectively .

Table (10) Major elements in Phosphate samples :

Locality	**Sibayia**	**Quseir – Safaga**	
Major oxides	**S2 %**	**Q7 %**	**Q9 %**
SiO_2	**45.57**	3.06	1.78
Al_2O_3	1.9	2.11	1.96
Fe_2O_3	2.69	1.59	2.47
Ti_2O	0.41	0.02	0.08
CaO	**23.40**	**44.00**	**50.40**
MgO	1.56	4.22	1.89
Na_2O	1.50	1.65	0.99
K_2O	0.79	0.26	0.04
P_2O_5	8.61	11.20	**20.50**
L.O.I	**12.50**	**32.02**	**19.10**
Total	**99.03**	**100.13**	**99.21**

The concentrations of trace elements in phosphate samples at Quseir – Safaga Area (Q6 , Q7, Q9) and phosphate samples at Sibaiya area (S2,S4,S8) were measured and listed in Table (11) .The concentrations of **Cr** (Chromium) vary from 133 to 211 ppm , **Cu** (Copper) vary from 17 to 37 ppm , **Ni** (Nickel) range from 13 to 66 ppm ,**Zn** (Zinc) range from 133 to 411 ppm , **Zr** (Zirconium) vary from 12 to 28 ppm , **Sr**(Strontium) varyfrom 570 to 1177 ppm , **Y**(Yttrium) varyfrom 15to 39 ppm and **Pb**(Lead) range from 2 to 9 ppm, while two elements **V**(Vanadium) and **Ba** (Barium) (the lowest values of phosphate samples at Quseir – Safagaarea(Q6,Q7,Q9) and the highest values of phosphate samples at Sibaiya area(S2,S4,S8) .

Table (11) The concentrations of the trace elements in (ppm) for the Phosphate samples .

Locality	**Quseir – Safaga**			**Sibayia**		
Trace Element	**Q6 (ppm)**	**Q7 (ppm)**	**Q9 (ppm)**	**S2 (ppm)**	**S4 (ppm)**	**S8 (ppm)**
Cr	188	211	162	147	133	178
Cu	37	23	17	25	23	20
Ni	22	13	32	66	18	16
Zn	261	198	311	411	155	133
Zr	18	12	15	21	17	28
Sr	972	570	828	1177	857	1084
Y	39	26	24	18	15	18
V	1.5	1.5	1.5	1996	2328	2100
Pb	6	2	4	9	4	3
Ba	1.5	1.5	1.5	852	990	917

III. Conclusions

It is noticed that, there is a difference in γ-activity between the summation of activities of residual and pregnant solution with the activity of the original sample.This phenomenon is varied in magnitude within the different radionuclides. The type of sample plays its role in these variations. But the activities of residual and pregnant solution in all samples for ^{40}K are much larger than the original , probably the reason is due tochemical reactions which hapend during leaching process .The behavior of radionuclides (^{238}U, ^{235}U, ^{234}U, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{232}Th and ^{40}K) was studied during the acid leaching process,the ^{235}U has nearly the same leachability as ^{238}U.The uranium isotopes are leached to the same extent but that the same is not observed between ^{234}U and ^{238}U due to the effect of α-recoil phenomenon.The lowestleachability in all the samples is present in ^{214}Pb between(1.6% - 7.45%) .Theleachability (leaching efficiency %) present in sample (S2 at Sibaiyain Nile Valley)by HCl and HNO_3(Tables8-9)is more than theleachabilitypresents in samples(Q7,Q9 at Quseir – Safaga)due to the action of carbonate contents in the last two samples.

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