

**ASSAY OF URANIUM ISOTOPIC RATIOS
234U/238U, 235U/238U IN BOTTOM SEDIMENT
SAMPLES USING DESTRUCTIVE AND NON
DESTRUCTIVE TECHNIQUES (NASSER LAKE)**

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Nasser Lake is the greatest man-made lake in the World. It is considered as the main source of water where the Nile water is impounded behind the Aswan high dam.. Uranium has three naturally occurring isotopes ^{234}U , ^{235}U and ^{238}U with isotopic abundance 0.00548, 0.7200 and 99.2745 atom percent. Dissolved uranium in the lake is primary due to weathering process. Monitoring of the isotopic ratios of uranium is used as a good indicator to trace and evaluate the origin and activities associated with any variation of uranium in the lake environment. The main objective of the present study is to clarify any potential variation of natural uranium $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ ratios in sediment samples of Nasser Lake by using destructive alpha and non destructive gamma- techniques. The results show that the uranium isotopic activity ratios are very close to the natural values. This study can also be used for radiological protection and safety evaluation purposes.

INTRODUCTION

The Egyptian government has constructed a very huge dam known as High Dam on 1964. This action led to forming a big industrial lake on the front of the high dam known as the lake of Nasser. It extends to about 500 km south Aswan. Actually it consists of two lakes: Lake Nasser and Lake An-Nuba. Nasser Lake, object of study, extends to 300 km within the Egyptian borders [1].

Out of the strategic importance of the River Nile and Nasser Lake for the national security, it is necessary to conduct a continuous upgrading for studying the levels and signature of radioactivity of the lake for the sake of radiological safety and radiation control.

The purpose of the present study is to clarify natural variation of $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ ratios in bottom sediment samples of Nasser Lake, and

to examine the possibility of applying uranium isotopic ratios to detect any potential discharge of artificially altered uranium from surrounding external nuclear activities to the environment.

SCOPE OF THE WORK

1- Updating of the previous baseline γ radioactivity data to detect and trace any variation in the activity levels and isotopes using different destructive (α) and non-destructive (γ) techniques.

2- Determination of the signature isotopes (^{234}U , ^{235}U , ^{238}U) and calculation of uranium isotopic ratios, ($^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{234}\text{U}$, $^{235}\text{U}/^{238}\text{U}$). These signature isotopes for bottom sediment pathway of Nasser Lake are indicative and good indicator for evaluation of any potential sabotage on the Nile (Nasser Lake).

EXPERIMENTAL WORK

During sampling trip, 16 bottom sediment samples were collected along 350 km of Lake Nasser. The sample locations are given in the following map (fig 1).

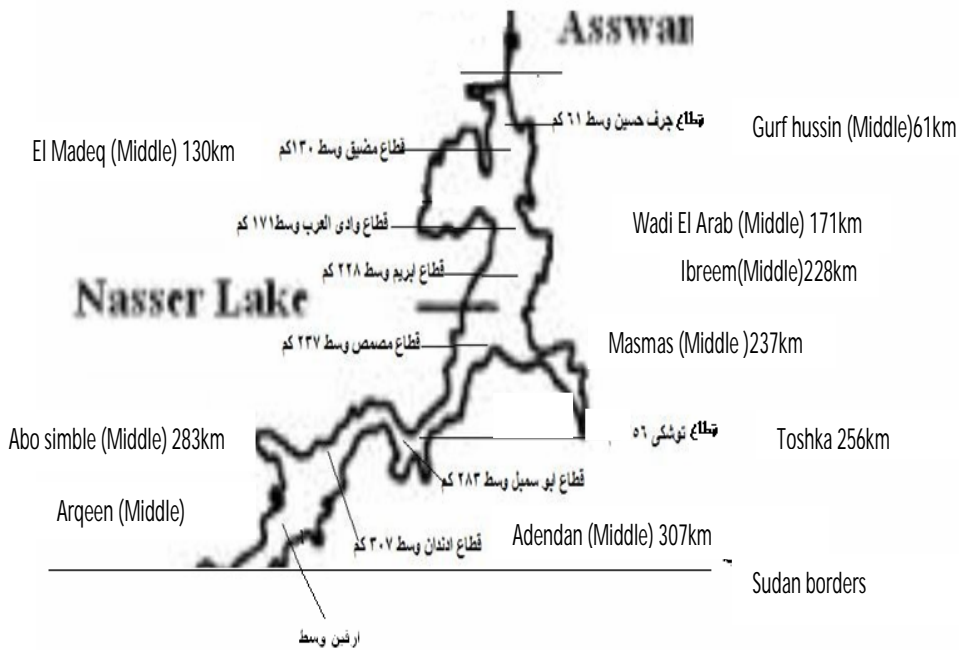


Fig (1): Map of the Nasser Lake and sample locations

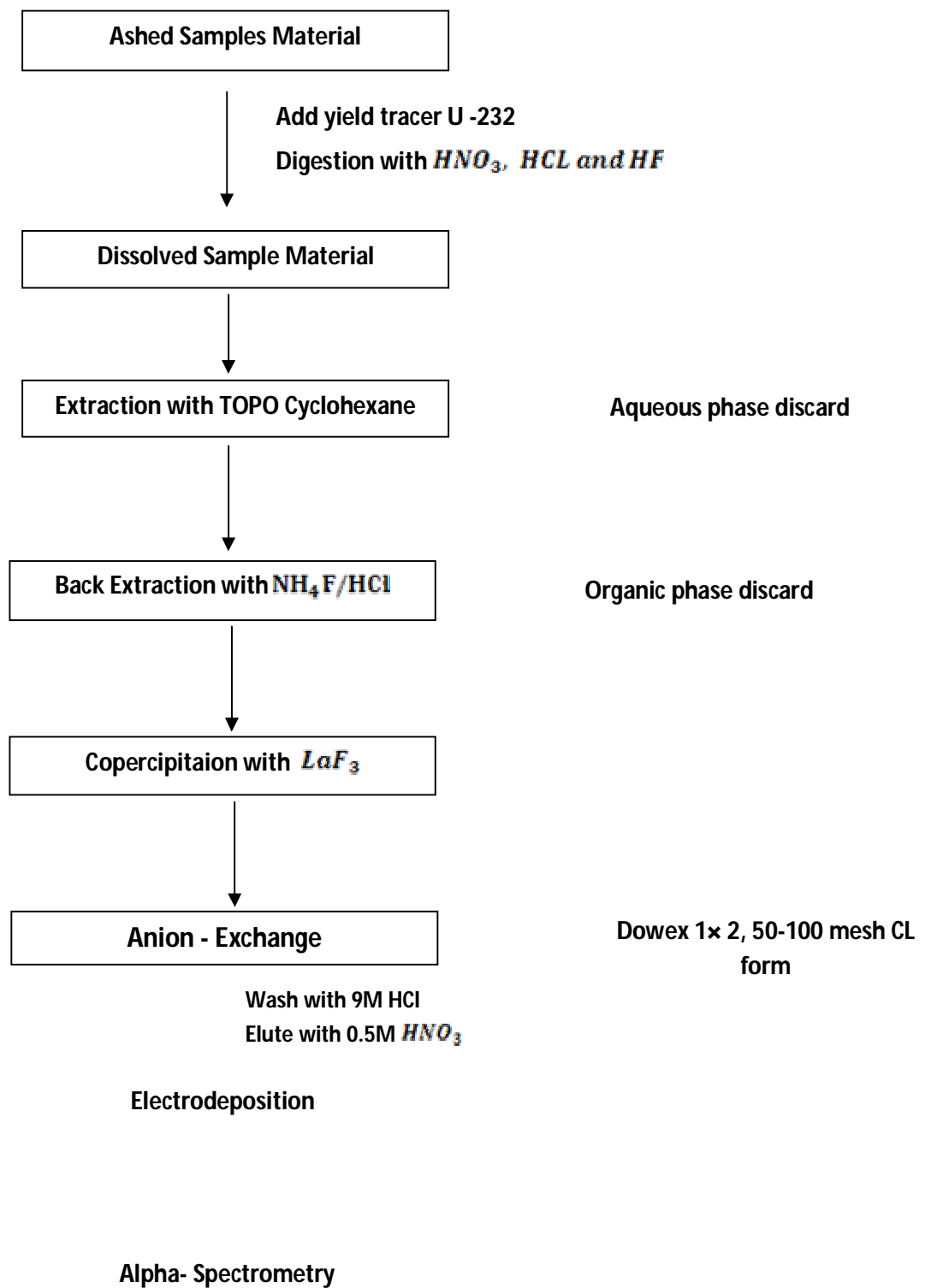


Fig (2): Schematic representation of the radiochemical separation of uranium

The collected sediment samples were dried, sieved, packed and shelved for 28 days to be measured by gamma ray spectrometer that is based on hyper pure germanium detector of 45% efficiency and 1.95 keV resolution at 1332 keV of Co-60 as non destructive technique. The γ - lines 185.7 keV of ^{235}U and 186 keV of ^{226}Ra were used to determine the $^{235}\text{U}/^{238}\text{U}$ isotopic ratios of the samples.

Radiochemical separation of uranium is based on drying, ashing, digestion, purification and electroplating. The separation procedure is given in (fig.2). Surface barrier detector of α - spectrometer technique was used for the determination of uranium isotopic ratios. The α -transitions 4.39 MeV of ^{235}U and 4.19 MeV of ^{238}U with the tracer ^{232}U of 5.32 MeV where used to determine the uranium isotopic ratios [2]

RESULTS AND DISCUSSION

Uranium has three naturally occurring isotopes, ^{234}U (progeny of ^{238}U , half-life 2.45×10^5 yrs), ^{235}U (half-life 7.04×10^8 yrs) and ^{238}U (half-life 4.47×10^9 yrs). IUPAC has reported isotopic abundance of U-234, 235 and 238 in natural uranium to be 0.00548, 0.7200 and 99.2745 atom percent as “Best measurement from a single terrestrial source” [3], While Richter et al. have proposed slightly different values, 0.005420, 0.72041 and 99.27417 atom percent for U-234, 235 and 238 [4]. The “original” $^{235}\text{U}/^{238}\text{U}$ abundance ratio should have been established by nucleon synthesis in a premolar environment and have been decreased due to radioactive decay since then. ^{234}U and ^{238}U are in radiological equilibrium, namely activity ratio of ^{234}U to ^{238}U being 1.0 [5], if the uranium was placed in closed system. Isotopic fractionation of heavy elements such as uranium usually is less significant compared with that of light elements such as hydrogen and oxygen. $^{235}\text{U}/^{238}\text{U}$ ratio has not been found to deviate from 137.5 ± 0.5 in most environmental samples [6], except for urbanite deposit where natural nuclear reactor existed 1.8×10^9 years ago and therefore ^{235}U was significantly depleted relative to ^{238}U [7].

Table 1 shows the calculated ^{235}U and ^{238}U in the analyzed samples in Bq/kg using gamma ray spectrometry. It could be noticed that, the obtained isotopic uranium activity ratios are in accordance with the natural certified values. The % difference ranged from – 0.64 % to 1.15% maximum.

Table (1): Uranium isotopic activity ratios in sediment samples by gamma assay

Sample name	U-235 (Bq/kg)	U-238 (Bq/kg)	U-235 / U-238 Ratio			
			Reference value	Measured	% difference	Measured / Reference
Gurf hussin (middle)	1.391	29.981	0.0462	0.0464	0.43	1.0043
El madeq (middle)	1.441	30.926	0.0462	0.0466	0.86	1.0086
Wadi el arab (middle)	1.251	27.034	0.0462	0.0463	0.21	1.0021

Wadi el arab (west)	1.247	30.926	0.0462	0.0468	1.15	1.0129
Ibreem (east)	1.435	30.997	0.0462	0.0463	0.21	1.0021
Masmas (west)	1.749	37.622	0.0462	0.0465	0.64	1.0064
Masmas (middle)	1.382	29.883	0.0462	0.0462	0.00	1.000
Adendan (west)	1.306	28.164	0.0462	0.0464	0.43	1.0043
Adendan (middle)	1.441	30.926	0.0462	0.0466	0.86	1.0086
Abo simble (middle)	1.606	34.407	0.0462	0.0467	1.08	1.0108
Abo simble (west)	1.487	31.711	0.0462	0.0469	1.29	1.0151
Sara (east)	1.395	30.207	0.0462	0.0461	-0.21	0.9978
Arqeen (middle)	1.297	28.273	0.0462	0.0459	-0.64	0.9935
Arqeen (east)	1.468	31.818	0.0462	0.0461	-0.21	0.9978
Arqeen (west)	1.698	36.764	0.0462	0.0462	0.00	1.000

Fried et al found increase in $^{235}\text{U} / ^{238}\text{U}$ ratio (0.007267) by 0.18 % to normally observed value of 0.007254 in some pre-historic volcanic materials by using Thermal Ionization Mass Spectrometry (TIMS) [8].

Table 2 shows the calculated ^{235}U and ^{238}U in the studied samples in Bq/kg using alpha spectrometer. The isotopic uranium activity ratios are also in agreement with the natural certified values (0.0462). The % difference ranged from - 4.32 % to 4.76% maximum.

In general the slight difference might be attributed to the recovery percentage of radiochemical. Emission from anthropogenic nuclear sources can also cause deviation in $^{235}\text{U} / ^{238}\text{U}$ ratios in environmental samples. Because soils and sediments can dilute out uranium with unnatural isotopic abundance because of their high natural uranium contents, low-uranium materials such as biological and water samples are usually favorable for sensitive detection of anthropogenic altered uranium [9]. In fact, depleted uranium (i.e. low $^{235}\text{U} / ^{238}\text{U}$ ratio) of anthropogenic origin was detected in groundwater with low uranium content [10]. Uranium from ancient atmospheric nuclear explosion was found in rainwater using isotopic ratio as the fingerprint [10-12]. If the amount of the uranium discharged from anthropogenic source was relatively large and isotopic composition of the discharged uranium was significantly different from that of natural uranium, however, "fingerprint" of the anthropogenic uranium could be detected even in environmental samples of high uranium content. For example, Taylor et al. used TIMS to measure soil

collected near the former United States Air Base in UK where enriched uranium was discharged into the environment in 1958, and found $^{238}\text{U} / ^{235}\text{U}$ atom ratio in the range 111-174 for some soil samples while natural $^{238}\text{U} / ^{235}\text{U}$ atom ratio is 137.9 [11].

Table (2): Uranium isotopic activity ratios in sediment samples by alpha analysis

Sample name	U-235 (Bq/kg)	U-238 (Bq/kg)	U-235 / U-238 Ratio			
			Reference value	Measured	% differenc e	Calculate d/ Reference
Gurf hussin (middle)	1.513	31.214	0.0462	0.0484	4.76	1.0476
El madeq (middle)	1.714	36.656	0.0462	0.0471	1.95	1.0195
Wadi el arab (middle)	1.351	30.034	0.0462	0.0451	-2.38	0.976
Abo simble (middle)	1.213	27.461	0.0462	0.0442	-4.32	0.956
Arqeen (middle)	1.487	31.687	0.0462	0.0468	1.29	1.00129
Adendan (middle)	1.672	27.431	0.0462	0.0465	0.64	1.0064

The correlation between the $^{235}\text{U} / ^{238}\text{U}$ isotopic ratios as assayed by non-destructive (γ) and destructive α -assay is given in figure (3).

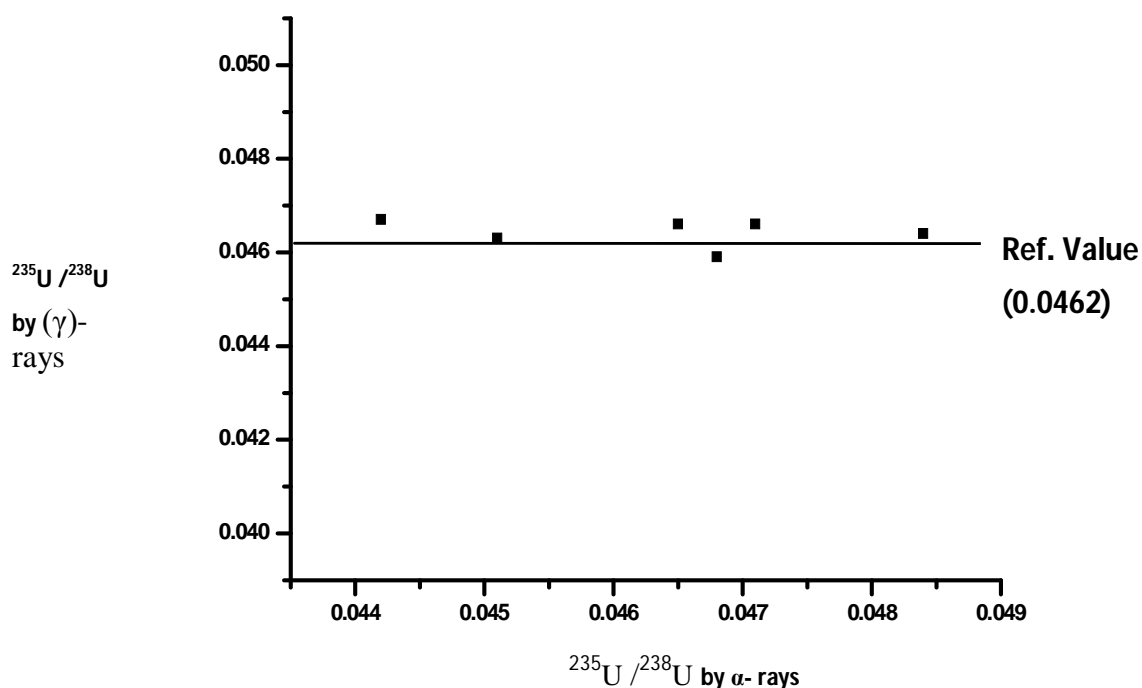


Fig (3): Correlation of $^{235}\text{U} / ^{238}\text{U}$ as measured by (γ) - and (α) - techniques

It can be shown that the $^{235}\text{U} / ^{238}\text{U}$ ratios as measured by γ - and α - techniques are very correlated to each other and very close to the natural target value (0.0462).

The results of $^{234}\text{U} / ^{238}\text{U}$ analysis by α - technique in the samples are given in table (3).

Table (3): U-234 / U-238 activity ratio in sediment samples by alpha analysis

Sample name	U-234 (Bq/kg)	U-238 (Bq/kg)	U-234 / U-238 Ratio			
			Reference value	Measured	% difference	Measured / Reference
Gurf hussin (middle)	30.861	31.214	1.00	0.989	1.1	0.989
El madeq (middle)	33.123	36.656	1.00	0.903	9.7	0.903
Wadi el arab (middle)	28.346	30.034	1.00	0.944	5.6	0.944
Abo simble (middle)	29.864	27.461	1.00	1.088	-8.8	1.088
Arqeen (middle)	31.231	31.687	1.00	0.986	1.4	0.986
Adendan (middle)	28.893	27.431	1.00	1.053	-5.3	1.053

It can be observed that the measured $^{234}\text{U} / ^{238}\text{U}$ values are in good agreement with the (certified / reference) values. The % difference ranged from -8.8 % to 9.7%. This result reflects the accuracy of radiochemical separation of uranium isotopes and α - spectrometry analysis.

The $^{234}\text{U} / ^{238}\text{U}$ ratios also vary due to anthropogenic discharge. Fertilizer-derived uranium, for example, could be distinguished from indigenous uranium using $^{234}\text{U} / ^{238}\text{U}$ as an indicator and could be used to trace the migration of fertilizer in the environment [13]. Goldstein et al. found depleted $^{234}\text{U} / ^{238}\text{U}$ ratios in soil samples of elevated uranium concentrations and considered it to be a contribution from anthropogenic uranium [14].

$^{234}\text{U} / ^{238}\text{U}$ ratio has been also found to vary considerably due to natural cause in many sediment and uranium ores of different geographical areas. The mechanism of such variation is preferential leaching of ^{234}U compared with ^{238}U from solid phase, caused by radiation damage of crystal lattice upon alpha decay of ^{238}U and oxidation of insoluble tetravalent ^{234}U to soluble hexavalent ^{234}U during decay solution phase [15].

CONCLUSION

Based on the results obtained by γ - and α - techniques, the $^{235}\text{U} / ^{238}\text{U}$ activity ratios were precisely determined and found to be very close to the natural ratio (0.0462). The maximum % difference does not exceed 1.9 and 4.76% for γ - and α - assay respectively. The determined $^{234}\text{U} / ^{238}\text{U}$ ratios were

also found to be very close to unity with a maximum difference of 9.7 % .The results obtained by (α) and (γ) analysis were correlated and found to be around the natural uranium activity ratio (0.0462).

It can be concluded that α - as a non destructive and α - spectrometry destructive techniques are recommended tools to assay uranium activity ratios in environmental samples. This is to investigate their status regarding being natural, enriched or depleted (origin tracking).

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