# Distribution Pattern Uranium Isotopes in Lake Sediments

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**Abstract.** Sediment samples from three Egyptian Lakes' (Qarun, Bardawill and Edku) were collected to study the environmental behavior of naturally occurring radionuclides in these lakes. The three lakes have different hydro-geological, physical and chemical features, which could affect the concentrations and the distribution pattern of uranium isotopes in the lakes' sediments. In this study, the specific activity of uranium isotopes (U-238, U-235 and U-234) were measured using alpha spectrometers after radiochemical separation and uranium alpha source electroplating on stainless steel disk. The distribution patter of uranium and the affects of sediment texture, organic matter contents, and other ecological parameters were investigated.

## Introduction

Radioactivity substances occur naturally in the environment, but can also result from human activity. It is now more than half a century since the discovery of nuclear fission, and uranium. Uranium is the fuel for about 1000 nuclear reactors of various kinds that exist in the world. These reactors produce electricity, produce plutonium for nuclear and thermonuclear weapons, and serve the search need of physical and biological scientists (Eisenbud and Paschoa 1989). Natural uranium can be detected in low concentrations in nearly all environmental samples. In radiochemical equilibrium, it consists of the isotopes $^{234}U$, $^{235}U$ and $^{238}U$ with the activity ratio of 1: 0.0462: 1, corresponding to a mass ratio of 0.0054: 0.711: 99.2836 percent. All of these three nuclides are alpha-emitters, which have a high biological effectiveness. From nuclear facilities, additional amounts of uranium are discharged into the environment. In effluents from nuclear facilities, the ratios of uranium isotopes differ very much. To be able to compare them with that from

natural background, it is necessary to determine not only the concentration of total uranium but also the ratio of the different uranium isotopes (Pimpl et al. 1992). This study is aimed at investigating the distribution pattern of uranium in three Egyptian lakes.

## Experimental work

### Sampling and samples preparation

Total of 47 bottom sediment samples were collected from Qarun, Bardawill, and Edku lakes, Fig. 1. The bottom sediment samples were collected using a grab sediment sampler according to lenz (Ekman-Birge). The collected samples were dried at 115 $^{o}$C, mechanically crushed, mixed and sieved through 2 mm mesh sieve. The organic content percentage, igination percentage, in sediment samples was determined as weight loss after ashing the samples at 450 $^{o}$C for about 8 hours (USDOE 1992; Khater et al. 2001).

### Analytical methods

#### *Radium-226 analysis*

$^{226}$Ra specific activity (Bq/kg dry weight) was measured using a well calibrated gamma ray spectrometers based on Hyper-Pure Germanium detectors. The details of $^{226}$Ra analysis are given in our previous work (Khater et al. 2001).

#### *Uranium isotopes analysis*

Uranium isotopes analysis, 1-5 g of the ashed samples were spiked, for chemical recovery and activity calculations, with $^{232}$U tracer and dissolved using mineral acids ($HNO_3$, HF and HCl). The uranium was extracted with Tri-octylphosphine oxide in cyclohexan, back-extracted with $NH_4F$/HCl solution, then co-precipitated with $La(NO_3)_3$ and purified by passing through an anion exchange column. Uranium then was electroplated on a stainless steel disk from oxalate-chloride solution. The prepared samples were measured using alpha spectrometry based on surface barrier detectors with 450 mm$^2$ surface area, about 17% efficiency and about 20 keV resolution. The chemical recovery was in the rang of 45-70 %. The lower detection limit of the procedure is in the range of 1 mBq/ sample, for a 1000 min. measuring time (Pimpl et al. 1992).

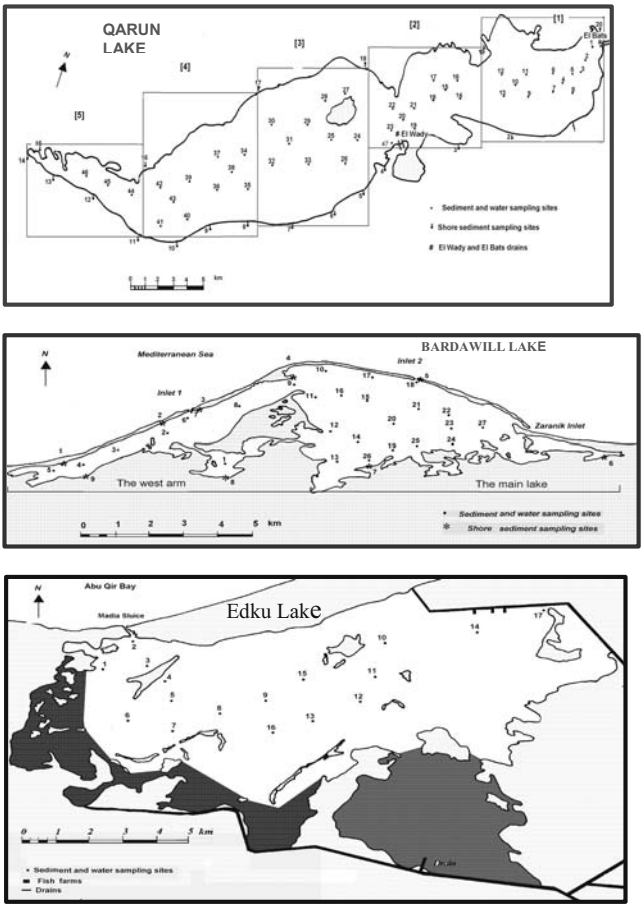


**Fig. 1.** Bottom sediment sampling locations from Qarun, Bardawill and Edku Lakes- Egypt.

## Results and discussion

The average specific activities of $^{238}U$, $^{235}U$, $^{234}U$ and total uranium (Bq/kg dry weight), and $^{234}U/^{238}U$ and $^{226}Ra/^{238}U$ ratios in Qarun, Bardawill and Edku lakes bottom sediment are represented in Table 1. Sediment texture (clay, silt and sand percentages), organic matter content, electric conductivity- EC and pH) are represented in Table 2.

The specific activities of $^{238}U$ and $^{234}U$ in Qarun and Bardawill lakes are two fold higher than that of Edku Lake. That may be explained because of water salinity and hydrological features of these lakes. Qarun and Bardawill lakes are closed lakes. They are nearly without any water's outflow except by evaporation and may

be by underground water. So, the uranium contents of inflow water (drain water in Qarun and sea water in Bardawill) are accumulating in lakes' environment. Edku lake water's is derived mainly from three drains (as Qarun Lake) and the sea (as Bardawill Lake). So, there is in and outflow water through the lake. Exchange of water between the lake and the sea occurs through a narrow inlet and most of the time in the direction of the sea. The water outflow in Edku Lake (through the inlet to the sea) is higher than that of Qarun and Bardawill lakes. The uranium contents increase greatly where the flow of water is low. It noticed that the range of uranium concentrations in Qarun Lake is larger than that of Bardawill Lake. That may be explained due to mineralogical properties of bottom sediments and the water resources of these lakes; drain water for Qarun Lake and sea water for Bardawill Lake. Although, the amount of uranium supplied to surface waters by rock and soil weathering is more or less constant. The continuous accumulation of uranium input from the drain water over many years and other parameters such as sediment texture, calcium carbonate percentage and the organic matter content could be a reason for such a big range of uranium concentration (Ivanovich and Harmon 1992).

The concentration of uranium varies widely in natural waters over four orders of magnitude. This is in part due to its varied chemical behavior in response to redox condition, from insoluble $4^+$ valence in reducing conditions to soluble 6+ valence in oxidation conditions. Most surface waters are oxidizing. The concentration of uranium in surface water is correlated with bicarbonate ion concentration or total dissolved solids (TDS) or both. The uranium concentrations increase with increasing TDS (Ivanovich and Harmon 1992; Coal and Bruland 1987; TDS: there is an empirical relationship between uranium concentration and water salinity, U-238 (dpm/l) = 0.07081 x salinity, Coal and O'Hara 1987). The average water salinity of Qarun, Bardawill and Edku lakes are 32.4, 60.5 and 2.86 g/l respectively.

The average $^{234}U/^{238}U$ activity ratios in lakes sediment are close to the equilibrium value of unity. Where leaching is less dominant, as in arid regions, these activity ratios become quit high. The reason is the relative importance of recoil fractionation over gross leaching in the less intensely weathered soils. The key factors in the development of $^{234}U/^{238}U$ activity ratios out of equilibrium are the relative rates of leaching, mechanical erosion, and daughter half-life. If uranium remains a considerable time relative to the $^{234}U$ lifetime, then the process of recoil will cause fractionation. Although the range of these activity ratios vary greatly in surface waters, often by a factor of 2 or 3, the global average value is closely constrained at 1.2–1.3 (Ivanovich and Harmon 1992).

**Table 1.** Specific activity of U-238, U-235, U-234 and total uranium (Bq/kg dry weight) and U-234/U-238 and Ra-226/U-238 ratio in Qarun, Bardawill and Edku lakes bottom sediment**.**

| | U-238 ± E | U-235 ± E | U-234 ± E | Uranium ± E | U234/ U-238 ± E | Ra226/ U-238 ± E |
|---|---|---|---|---|---|---|
| | 48.0 ± 3.66 | 1.41 ± 0.20 | 53.4 ± 4.09 | 102.9 ± 7.69 | 1.13 ± 0.04 | 0.37 ± 0.03 |
| Qarun | (21.5 - 81.7) | (0.19 - 4.25) | (22.4 - 97.7) | (45.1 - 182.5) | (0.72 - 1.59) | (0.19 - 0.79 |
| | [25] | [25] | [25] | [25] | [25] | [25] |
| | 45.4 ± 7.48 | 1.84 ± 0.68 | 46.6 ± 8.33 | 93.6 ± 16.0 | 0.99 ± 0.07 | 0.34 ± 0.21 |
| Bardawill | (9.8 - 61.6) | (0.34 – 4.9) | (7.74 - 63.3) | (17.9 - 128.8) | (0.79 - 1.27) | (0.11 - 1.16) |
| | [6] | [6] | [6] | [6] | [6] | [6] |
| | 25.5 ± 3.16 | 1.00 ± 0.20 | 29.3 ± 4.57 | 57.9 ± 8.65 | 1.11 ± 0.40 | 0.67 ± 0.1 |
| Edku | (11.6 - 55.7) | (0..25 - 3.25) | (12.8 - 84.7) | (27.0 - 142.3) | (0.9 - 1.52) | (0.17 - 1.34) |
| | [16] | [16] | [16] | [16] | [16] | [16] |

* Mean ± standard error of the mean, (Range), and [Sample number].

**Table 2.** Mechanical analysis parameters, Igination factor %, EC and pH of Qarun, Bardawill and Edku lakes.

| | Depth (m) | Organic matter % | Silt % | Clay % | Sand % | $CaCO_3$ % | E.C. | pH |
|---|---|---|---|---|---|---|---|---|
| Qarun | 3.4 | 12.4 | 15.8 | 21.0 | 53.4 | 22.7 | 14.3 | 8.1 |
| Bardawill | 1.5 | 10.4 | 10.2 | 12.3 | 68.2 | 21.8 | 18.4 | 8.2 |
| Edku | 0.8 | 12.2 | 23.0 | 25.4 | 47.1 | 18.1 | 2.1 | 7.6 |

The average $^{226}Ra/^{238}U$ activity ratios in Qarun, Bardawill and Edku lakes sediment are very low, 0.37, 0.32 (with excluding one figure corresponding to the maximum ratio, it becomes 0.16) and 0.67. That may be because of the amount of radium in surface waters is more variable than the amount of uranium. There are many chemical and physical circumstances in which radium is adsorbed on to or desorbed from associated sediments. The general condition is that sediment arrives at drain's mouth with radium adsorbed to the particulate surface, and then radium is desorbed in the more saline environment. That is clear in Qarun Lake where the activity ratio is less and the salinity is higher than that of Edku Lake. For Bardawill lake, the lowest ratio activity may be because of the low $^{226}Ra$ concentration (0.002 Bq/l) in sea water comparing to $^{238}U$ concentration (0.04 Bq/l) (Ivanovich and Harmon 1992; Maul and O'Hara 1989).

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