

Factors Controlling Uranium and Thorium Isotopic Composition of the Streambed Sediments of the River Nile, Egypt

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Abstract. The streambed sediments of the River Nile in Egypt have been analyzed for their long-lived uranium and thorium radionuclides (^{238}U , ^{234}U , ^{230}Th) using Alpha Spectrometric Techniques. These sediments are dominated by sand size fractions with average heavy mineral contents of 2.65%. Carbonates and organic materials constitute considerable amounts of the sediments (averages 13.82 and 3.37%, respectively). The average contents of uranium and thorium in the streambed sediments are 2.06 and 6.58 ppm, respectively, which are comparable with the corresponding values of the world rivers. The $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratios of the sediments are greater than unity indicating preferential uranium leaching. During low water flow rate, the streambed sediments probably acquired some uranium from water with high $^{234}\text{U}/^{238}\text{U}$ activity ratios. This results in abnormally high $^{234}\text{U}/^{238}\text{U}$ activity ratios in the sediments. Heavy mineral contents, sorptive uptake of uranium by sediments of relatively high organic contents, geochemical behaviour of radionuclides and flow rates of the River Nile are considered as the most important factors controlling uranium and thorium isotopic composition of the streambed sediments. The sources of the radionuclides are probably the local granitic rocks and phosphate fertilizers used in the cultivated lands adjacent to the River Nile.

Keywords: River Nile; Uranium; Thorium; Uranium-series disequilibrium; Activity ratios; Streambed sediments.

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Introduction

Uranium and thorium measurements in rivers, lakes and soils are important for monitoring environmental radioactivity and to understand the geochemical behavior of these radionuclides in the surficial water-bearing environments. Dissolved uranium in rivers is primarily a result of weathering processes on rocks and soils. The estimated worldwide averages for dissolved uranium in rivers range between 0.3-0.6 ppb (Scott, 1982) and reach to abnormal high values in some cases due to chemical weathering of uraniferous rocks such as in Platte River of the North American High Plains Region where uranium contents reach up to 31.7 ppb (Snow and Spalding, 1994). Palmer and Edmond (1993) determined the concentrations of dissolved uranium in forty major rivers from around the world with an average concentration of 0.31 ppb. They noted that this value could be biased by the very high levels observed in the Ganges-Brahmaputra and the Yellow River. Excluding these two river systems, the global average of uranium is reduced to 0.19 ppb. Compared to uranium, thorium is highly particle-reactive element and does not readily occur as a dissolved ion (Langmuir and Herman, 1980).

Eroded materials transported from the continents to the oceans are mainly carried by rivers. Rivers transport these materials both in dissolved and solid forms. The relative distribution of elements between the solute and particulate phases depends on the mobility of the chemical constituents during weathering and transport (Viers *et al.*, 2009). The naturally occurring radionuclides (U, Th and ^{40}K) present in the river streambed sediments are mostly of terrestrial origin and their concentrations are related to the type of parent rocks and to the genesis of the sediments. The river sediments generally exhibit large variation in composition. This variation can be related to the chemical and mineralogical evolution of these sediments along the river, influence of tributaries, or different properties of drained soil. The mobility of radionuclides in the aqueous system is an important factor influencing the content of radionuclides in river sediments. Surface run-off waters in the tributaries wash down a part of deposited radionuclides and finally store them in the river sediments. In view of these facts, radionuclides from the uranium and thorium series have been used for a long time as tracers and chronometers in studying the weathering processes and

environmental aspects (Porcelli *et al.*, 2001; Dosseto *et al.*, 2006a and b; and Chabaux *et al.*, 2006).

The primordial isotope ^{238}U decays, through a series of alpha and beta-emitting radionuclides, towards stable ^{206}Pb . This study is focused on the long-lived members of the ^{238}U -series: ^{238}U , ^{234}U and ^{230}Th . The activity of a radionuclide in the chain is a balance of its production by the decay of its parent and loss by its own decay. In a system which has remained closed for a sufficiently long time, activities of all radionuclides are equal. This means that the activity ratio of any two radionuclides equals unity, a condition termed as the state of secular equilibrium. Secular equilibrium between two radionuclides can be disturbed by any process that results in selective removal or addition of radionuclides, leading to the so-called disequilibrium. Disequilibrium between ^{238}U and ^{234}U in natural waters and sediments is a common phenomenon. Mechanisms responsible for such disequilibrium include α -particle recoil ejection of ^{234}Th (a precursor of ^{234}U) into solution (Kigoshi, 1971); The preferential chemical solution of ^{234}U due to the radiation damage of the crystal lattice caused by the decay of the parent ^{238}U (Rosholt *et al.*, 1963), and electron stripping during the decay process such that ^{234}U is more likely to be in the more soluble U^{6+} state, facilitating the solubility of this isotope (Jurado Vargas *et al.*, 1995). Therefore, the $^{234}\text{U}/^{238}\text{U}$ activity ratios of surface or groundwaters are usually greater than unity. U-series isotopes of the river water and sediments have been used to address different issues such as: The transport time of the river sediments, contribution of rivers to the uranium isotope budget of the oceans, behavior of U-series isotopes during weathering and transport in rivers and the sources of dissolved flux, in particular groundwater contributions (Scott, 1982; Osmond & Ivanovich, 1992; Olley *et al.*, 1997; and Chabaux *et al.*, 2003).

The radioactivity of the River Nile sediments in Egypt has been investigated by (Ibrahiem *et al.*, 1993; Ismail *et al.*; 1994; Khater *et al.*, 2005; El-Gamal *et al.*, 2006 & 2007; and Saleh *et al.*, 2006). These studies focused mainly on the radioisotope activity based on gamma spectrometric measurements without going through accurate measurements of radioactive disequilibria in these sediments. Furthermore, detailed correlations between the sedimentological characteristics of the streambed sediments and the radioactivity were not addressed in these studies. The main purposes of the present study are

describing the uranium and thorium distributions in the streambed sediments in relation to their mineralogical composition and using the activity ratios of ($^{234}\text{U}/^{238}\text{U}$), ($^{230}\text{Th}/^{234}\text{U}$) and ($^{230}\text{Th}/^{238}\text{U}$) to investigate the radioactive disequilibria and relative mobility of the long lived radionuclides. The result of this work should also contribute to the evaluation of radionuclides budget in the world rivers.

Description of the Study Area

The River Nile is the longest river in the world with a length of 6695 km and a drainage area of 3,349,000 km². The average discharge is 5,100 m³/s. The countries in the drainage basin encompass Ethiopia, Uganda, Tanzania, Congo, Kenya, Rwanda, Burundi, Sudan and Egypt. The drainage basin includes three terrains: The Ethiopian Highlands, from which the blue Nile and Atbara flow; the Central African Highlands, in which White Nile headwaters originates; and the Red Sea Hills, dissected by largely inactive wadis, which indicate past water flow and sediment discharge into the main Nile channel from eastern Egypt and northeastern Sudan (Schneiderman, 1995; and Dawood and Abd El-Naby, 2007). The Ethiopian Highlands consist of a large fault block related to the Cenozoic East African Rift System where the Blue Nile and Atbara Rivers flow across a vast plain of alkali flood basalt and tholeiitic lavas. Conversely, the Central African Highlands consist mainly of cratonic Archean basement rocks (Condie, 1981). In the region drained by White Nile tributaries, the Central African Highlands consist of the west Nile gneiss complex, amphibolite, metasediments and metavolcanics (Goodwin, 1991). The Red Sea Hills generally consist of metasediments, metavolcanics, gneisses, ophiolites, granites and pegmatites. Wadis, up to hundred kilometers long, dissect both the Red Sea Mountains and sedimentary cover of Cretaceous sandstones and Eocene marine limestones.

The River Nile flows from the highlands and forests in Uganda through tropical forests, savanna woodlands, swamps, grasslands, and finally through deserts to the Mediterranean Sea. The upriver regions are highly humid, whereas hot-hyperarid climate of the Sahara desert characterizes regions of the lower Nile (Said, 1981). About 84% of the flow in the main Nile is derived from Ethiopia during the summer floods whereas 16% is derived from the Central Africa Lake Plateau (Stanley *et*

al., 1988). The distribution of Pliocene paleo-Nile alluvial sediments in Egypt suggests that, even though currently dry, the wadis draining the Red Sea Hills must previously have been a main source of water and sediment for the Nile (Said, 1981; and Dawood and Abd El-Naby, 2007).

In Egypt, the sedimentation load and natural distribution of Nile sediments are highly affected by the hydraulic structures constructed on the River Nile. Accelerated erosion of Egypt's Nile delta coast during this century has generally been attributed to construction of Aswan High Dam, old Aswan Dam, barrages and river control structures which almost complete trapping of sediments (Stanley, 1996; and Chakrapani, 2005). Delta erosion is also accelerated by natural factors such as global warming, rising of sea level and strong coastal currents. The barrages in Upper Egypt at Isna, Assuit, and Nag Hammadi are constructed to keep the level of the Nile high enough all the year for regular irrigation. The barrier deposits form dams intercept the free flow of water. There is inverse relationship between the Nile flow and sediment flux (Krom *et al.*, 2002). The ability of a stream to move sediment depends on the water velocity, the channel slope and the size of the sediment particles. Geography, country rocks and tectonic setting of the landscape may cause a channel to bend or rise and consequently slowdown the stream flow such as at Qena and Nag Hammadi bend (Fig. 1). At the bend area, erosion is increased at the outer bank while sedimentation is increased along the inner bank where the water flow is the lowest (Bridge, 2003).

Sampling and Analytical Procedures

In summer (July-September), the water level in the River Nile is relatively high due to water inflow during the rainy season in Ethiopia. Sampling process has been carried out during winter (January-February). Sixteen streambed sediments were collected from the river; distributed as 5 samples from Rosetta and Damietta branches and 11 from the main channel (Fig. 1). The colors of these sediments are variable depending on the mineralogical composition and organic matter contents. They range from light olive gray, yellowish brown to black. The samples were collected approximately at the middle of the channel where the water depth ranges between 5-14 m, depending mainly on the topography of the channel floor and the amount of water recharge. At each sampling site, a grab sampler connected with a long rope was used from over a

fishing boat to collect about 1 kg of wet sediments from the streambed. The samples were placed in plastic packets then sealed and transported to the laboratory.

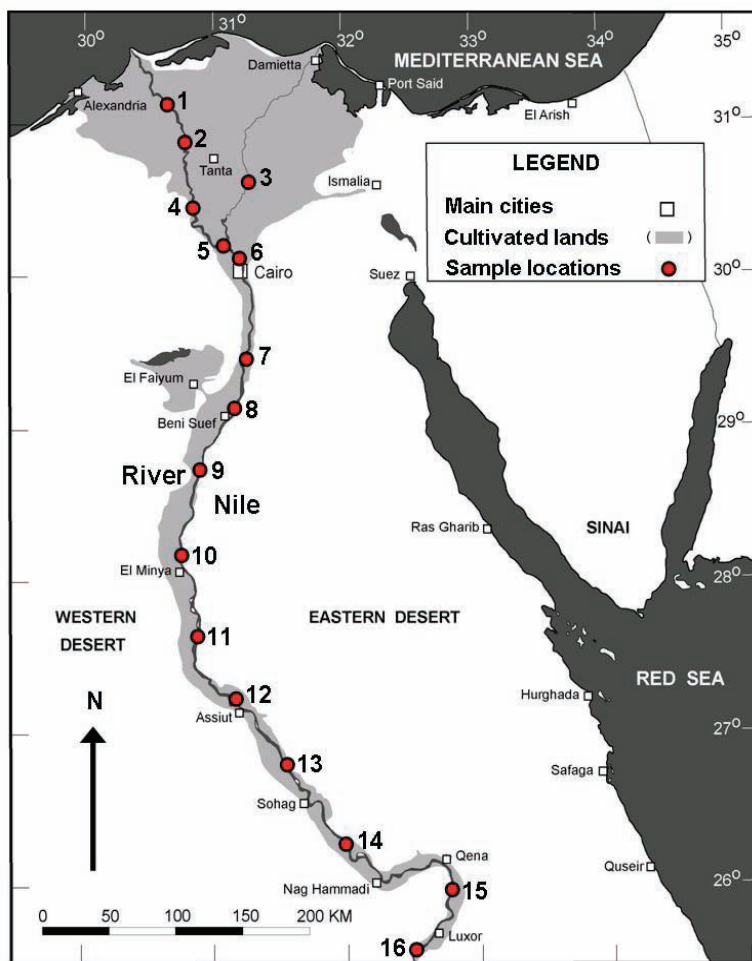


Fig. 1. Map of Egypt showing sampling sites along the River Nile.

The collected samples were dried at 100 °C in an oven for 3 days and then split into three parts for laboratory investigations. The investigations include determination of organic matter and carbonate contents and identification of clay and heavy minerals as well as whole sediment radiogenic isotope analyses.

The first portion of the sample was weighed and then treated by warm 0.2 N HCl in an Erlenmeyer flask overnight. After decantation, the sample was dried, the weight loss was measured and the carbonate percentage was calculated. The insoluble residue was later washed with distilled water and wet sieved using 63 μ m sieve size in order to separate sand size fractions for the purpose of heavy minerals separation. After dryness, the percentage of the sand fractions was calculated. The heavy minerals were separated from the sand size fractions using bromoform. Percentages of these separated constituents are given in Table 1. Microscopic investigation, Back Scattered Electron imaging (BSE), Secondary Electron Imaging (SEI) and Energy Dispersive X-ray Spectrometry (EDS) for some separated heavy mineral grains were performed using JEOL JXA-8900 Electron Probe Micro Analyzer available at the Institute of Earth Sciences, Mineralogy and Geodynamics, University of Tübingen, Germany.

Table 1. Sedimentological data of the streambed sediments of the River Nile in Egypt.

S. N.*	Organic matters %	Carbonates %	Sand %		Total sand %	Silt %	Clay %	Total
			Light minerals %	Heavy minerals %				
1	3.71	10.50	73.66	1.49	75.15	7.55	3.09	100.00
2	3.11	7.10	41.97	0.68	42.65	37.97	9.17	100.00
3	1.06	21.19	72.54	1.92	74.46	1.48	1.80	99.99
4	4.20	13.50	72.55	4.48	77.03	3.34	1.94	100.01
5	1.20	3.55	84.02	2.38	86.40	7.08	1.78	100.01
6	2.65	45.00	44.85	1.30	46.15	3.35	2.86	100.01
7	3.02	9.50	76.03	5.40	81.43	4.72	1.34	100.01
8	6.11	12.25	66.65	2.03	68.68	9.97	2.98	100.00
9	3.07	12.00	60.26	1.97	62.23	14.52	8.16	99.98
10	2.20	6.00	77.42	8.60	86.02	3.43	2.34	99.99
11	6.51	19.00	8.39	0.01	8.40	34.20	31.89	100.00
12	1.15	15.26	54.61	1.98	56.59	12.85	14.14	99.99
13	4.32	10.00	56.41	5.33	61.74	11.79	12.15	100.00
14	2.03	9.00	77.52	3.70	81.22	6.29	1.45	99.99
15	5.99	14.75	44.81	0.31	45.12	9.59	24.55	100.00
16	3.60	12.50	73.85	0.84	74.69	7.17	2.04	100.00
Avg.	3.37	13.82	61.60	2.65	64.25	10.96	7.61	100.00

* See Fig. 1 for sample locations

The clay deflocculation in the separated -63 μ m size fraction was obtained through repeated centrifugation (2500 revolutions per minute (rpm) for 3 minutes). After each centrifugation, water was eliminated and the plug was re-suspended in distilled water. Generally, three to six centrifugation–suspension cycles were necessary until deflocculation.

After transferring the suspended sediments to a graduated glass cylinder and vigorously shaking, the silt and clay percentages were determined using the pipette method. X-Ray Diffraction technique was used to identify the mineral composition of untreated clay fractions. The X-ray diffractograms were performed with Siemens diffractometer using Ni filter and Cu-K α radiation at 40 kV and 20 mA. The machine is equipped with D 500 goniometer, a graphite monochromator and a scintillation detector. The pulverized samples were scanned for about 30 minutes counting time at a rate 1 degree 2θ per half minute with step size 0.02 2θ . Peaks search and identification were performed by the DIFFRAC-AT software connected to the diffractometer.

The second portion of the sample was used to measure the percentage of organic matter contents. A clean and desiccated crucible was weighed before putting in a few grams of dried samples. The combined mass was measured and the crucible was then put in a muffle furnace at temperature of 450 °C for 8 hrs. The crucible was removed and allowed to dry in desiccator. The crucible is then re-weighed and the difference from the dry state yields the organic content.

The third portion of each sample was crushed in a jaw crusher and powdered to 200-mesh size by ball milling. The uranium and thorium isotope analyses were determined for the whole-sediments using alpha-spectrometry technique. 2 g from each pulverized fraction were dissolved using a mixture of HNO₃ and HF in a pressure container at 140°C for two weeks. ²³⁶U and ²²⁹Th spike tracers were added to the samples before digestion. The solutions obtained by complete dissolution of each fraction were left for three days to reach isotopic equilibrium. A carrier consisting of ferric nitrate was added, then the sample was heated to boiling and the pH increased to a value of about 10 by the addition of ammonium hydroxide. The actinides including uranium and thorium coprecipitate with ferric hydroxide floc. After separation of the floc by centrifugation, the iron was separated from the actinides by solvent extraction with diethyl ether. The anion exchange procedures were performed for uranium-thorium separation and purification. The resin used is Bio-Rad AG 1X8, 100–200 mesh; chloride form. Both hydrochloric and nitric acid stages of anion exchange were used. Thin sources of purified uranium and thorium fractions were prepared by electrodeposition onto stainless steel discs for subsequent isotope measurements by alpha spectrometry available at the Department of

Earth and Environmental Sciences, University of Illinois at Chicago (UIC). The uncertainty of the measurements was calculated using standard error formulae based on counting statistics (Friedlander and Kennedy, 1956). The detailed procedure for uranium and thorium isotope analyses is described in detail by Lally (1992).

Results

Composition of the Streambed Sediments

The percentages of organic matters, carbonates and sand, silt and clay size fractions in the streambed sediments are shown in Table 1. The organic contents range between 1.06 to 6.51% with an average of 3.37%. The carbonate fraction is represented mainly by shell fragments. It constitutes a significant proportion of the light minerals in the sediments with an average content of about 13.82%. The percentages of sand, silt and clay size fractions are plotted in the ternary diagram and presented in histogram (Fig. 2 and 3). The ternary diagram plot classifies the sediments basically into sand, clayey sand and silty sand (Fig. 2). The sand size fractions of the streambed sediments constitute about 64.25%, in average. Conversely, the averages of silt and clay size fractions are 10.96 and 7.61%, respectively. Figure 4 shows the distribution of the carbonate contents along the river in relation to the corresponding heavy

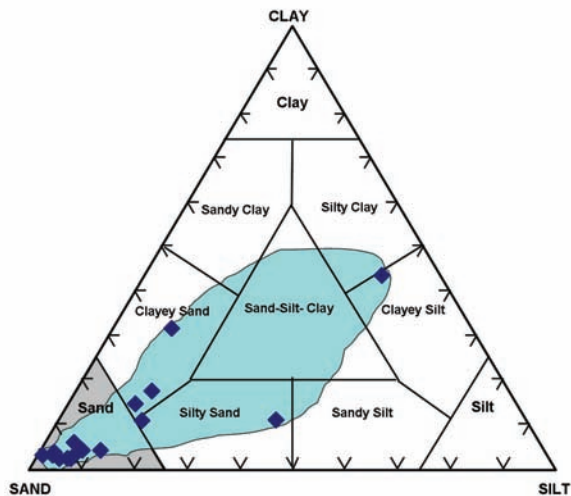


Fig. 2 Ternary diagram of sand, silt and clay fractions.

mineral contents. A correspondence between crests and troughs in both distribution curves is observed. This is anticipated since the carbonates constitute a significant component of the light mineral fraction.

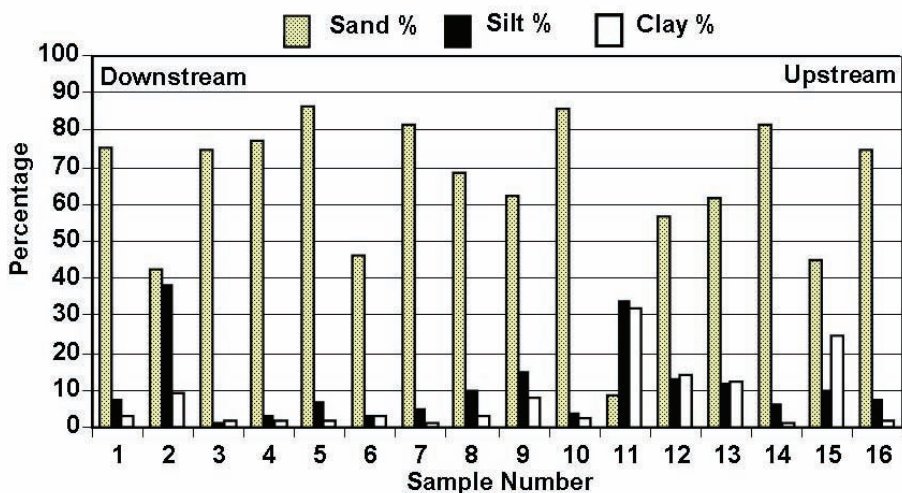


Fig. 3. Histogram of sand, silt and clay fractions of the streambed sediments.

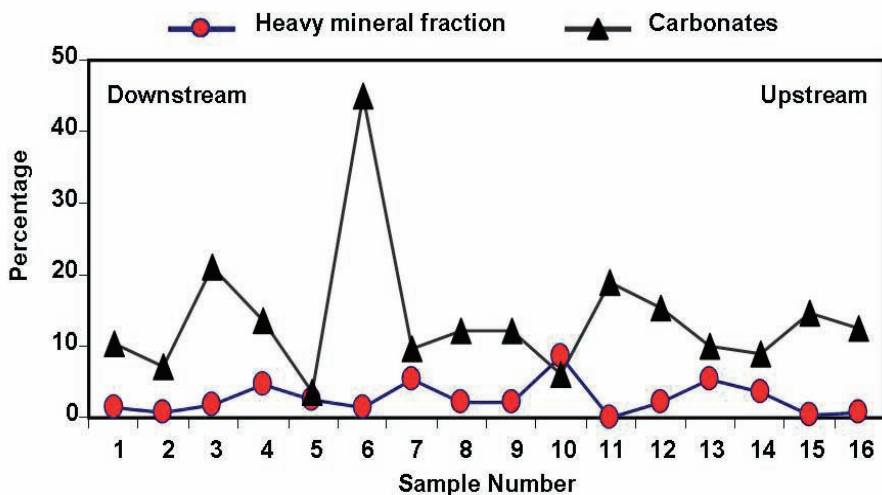


Fig. 4. Distribution of the carbonate and heavy mineral contents along the River Nile.

The composition of the clay fraction is dominated by kaolinite, illite and montmorillonite as indicated from the X-Ray diffractogram of representative untreated clay fraction (Fig. 5). The microscopic investigation of the heavy mineral fraction showed the presence of the characteristic River Nile assemblage including amphiboles, pyroxenes, biotite, muscovite, iron oxides, tourmaline, zircon, monazite, apatite, titanite, kyanite, sillimanite, andalusite, staurolite and garnet (Shukri, 1950).

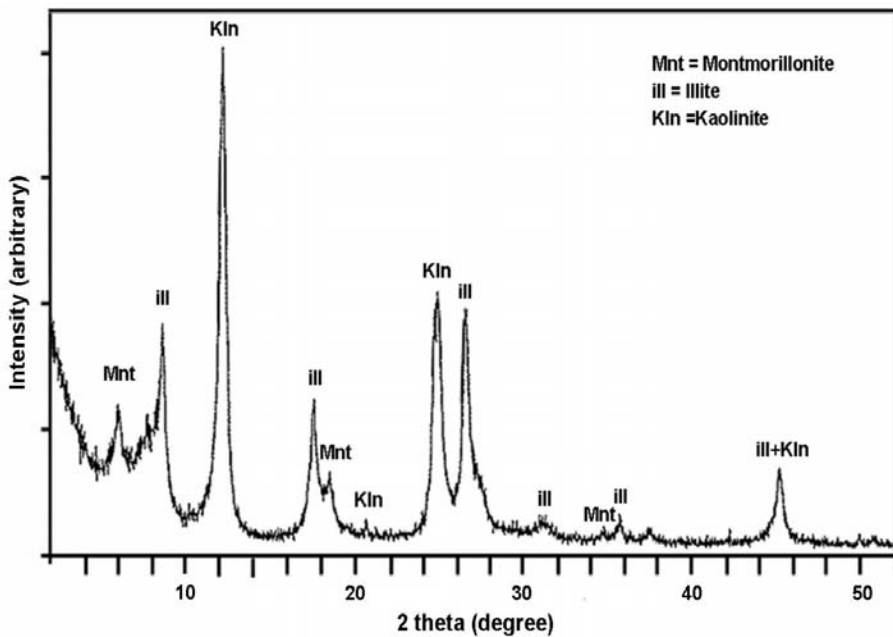


Fig. 5. X-Ray diffractogram of a representative untreated clay fraction.

Detrital zircon, apatite and monazite are considered as the major uranium and thorium bearing phases in these sediments. Uranium is associated with zirconium and hafnium in zircon and sometimes with thorium and cerium in monazite. In addition, uranium in the form of uranyl ion (UO_2)²⁺ commonly replaces Ca in the structure of apatite. Thorium is a main constituent in monazite and thorite, Th-rich monazite and thorite inclusions in monazite were reported by Dawood and Abd El-Naby (2007) in the Mediterranean beach sediments derived by the River Nile. Figure 6(a) shows a Secondary Electron Image of a separated

fractured monazite grain and its EDX spectrum. The chemical composition of this grain is dominated by Ce, Th, La, P and Si. Secondary Electron and Back Scattered Electron Images of zoned zircon grains are also shown in Fig. 6(b and c). Zircon grains display weakly fractured cores and fairly fractured rims with several growth zones. Zoning in zircon is generally associated with atomic replacements of uranium and thorium for zirconium (Mathieu *et al.*, 2001).

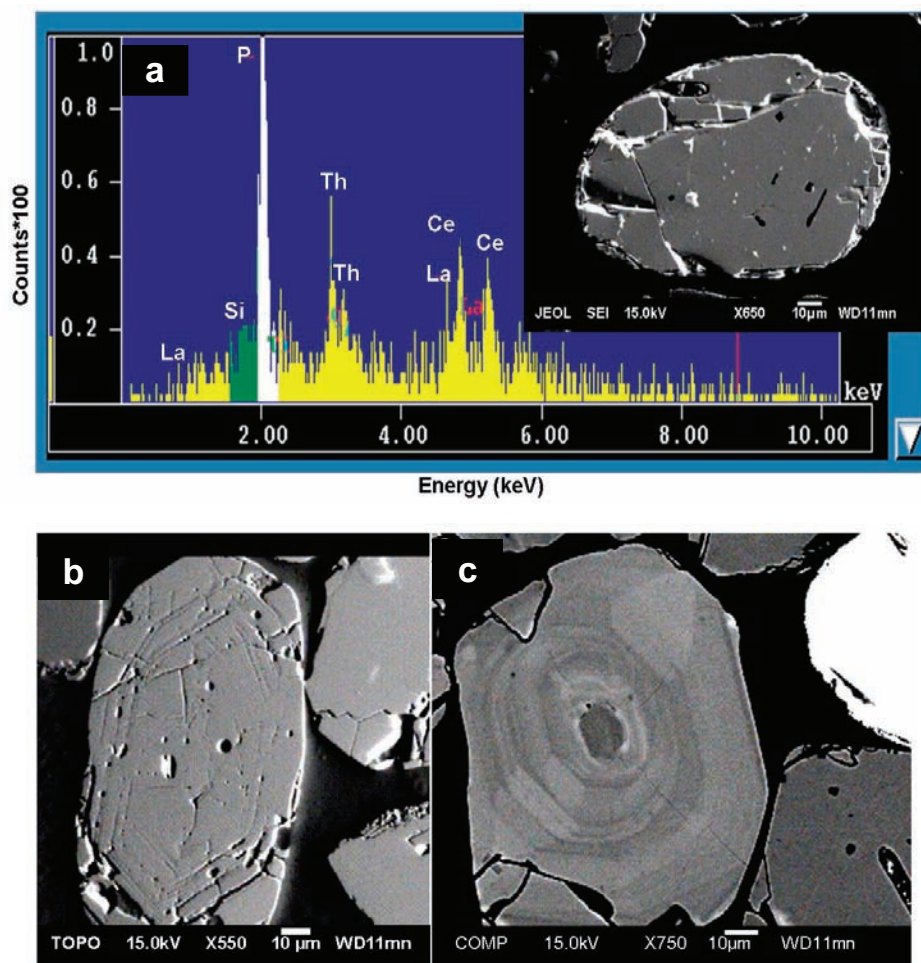


Fig. 6: a) Secondary Electron Image of a separated fractured monazite grain and its EDX spectrum. (b and c) Secondary Electron and Back Scattered Electron Images of zoned zircon grains.

U-Th Isotopic Composition of the Streambed Sediments

The concentrations of radionuclides in the River Nile sediments and their activity ratios are summarized in Table 2, the measured values are on a dry weight basis. The uranium contents of the Nile streambed sediments range between 0.3 to 4.82 ppm with an average of 2.06 ppm and that of thorium is in the range of 1.2-14.12 ppm with an average of 6.58 ppm. Generally, fractionation of uranium and thorium isotopes during chemical weathering is evident in the $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratios of river water (Scott, 1982) and also reported in the streambed sediments from different rivers in the world (Plater *et al.*, 1992; Balakrishna *et al.*, 2001; and Dosseto *et al.*, 2008). Table 2 and Fig. 7 show $^{234}\text{U}/^{238}\text{U}$ activity ratios of more than unity except for samples 3 and 12 (0.97, 0.99, respectively). The average of $^{234}\text{U}/^{238}\text{U}$ activity ratios in the streambed sediments is 1.06. The $^{230}\text{Th}/^{234}\text{U}$ activity ratios range from 1.09 to 1.44 with an average of 1.19. Moreover, the $^{230}\text{Th}/^{238}\text{U}$ activity ratios range from 1.16 to 1.58 with an average of 1.26. Figure 7 demonstrates that the $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratios deviate significantly from equilibrium compared to $^{234}\text{U}/^{238}\text{U}$.

Table 2. Uranium and thorium isotopic compositions of the streambed sediments of the River Nile in Egypt.

S.N.*	U (ppm)	Th(ppm)	U/Th	Activity ratio		
				$^{234}\text{U}/^{238}\text{U}$	$^{230}\text{Th}/^{234}\text{U}$	$^{230}\text{Th}/^{238}\text{U}$
1	1.25±0.06	4.32±0.35	0.29	1.12±0.06	1.14±0.23	1.28±0.17
2	0.93±0.05	4.06±0.30	0.23	1.06±0.07	1.13±0.15	1.20±0.22
3	4.14±0.08	5.03±0.33	0.82	0.97±0.07	1.21±0.20	1.17±0.19
4	4.61±0.11	11.23±0.42	0.41	1.05±0.08	1.26±0.22	1.33±0.24
5	1.49±0.05	4.64±0.23	0.32	1.01±0.04	1.18±0.24	1.19±0.11
6	0.78±0.06	2.87±0.19	0.27	1.08±0.05	1.11±0.19	1.20±0.12
7	1.40±0.04	9.56±0.44	0.15	1.09±0.05	1.15±0.21	1.25±0.21
8	1.30±0.08	6.70±0.45	0.19	1.14±0.08	1.17±0.18	1.33±0.29
9	1.14±0.07	7.15±0.37	0.16	1.02±0.07	1.22±0.17	1.24±0.31
10	4.82±0.18	14.12±0.56	0.34	1.03±0.09	1.30±0.30	1.34±0.16
11	0.73±0.02	2.60±0.22	0.28	1.15±0.10	1.10±0.16	1.27±0.13
12	2.96±0.12	7.67±0.37	0.39	0.99±0.08	1.17±0.13	1.16±0.14
13	1.51±0.09	12.31±0.60	0.12	1.07±0.11	1.24±0.22	1.33±0.20
14	4.34±0.2	8.05±0.33	0.54	1.06±0.05	1.09±0.19	1.16±0.10
15	0.30±0.02	1.20±0.17	0.25	1.10±0.09	1.44±0.31	1.58±0.36
16	1.30±0.14	3.80±0.16	0.34	1.04±0.07	1.12±0.17	1.17±0.23
Avg.	2.06± 0.08	6.58±0.34	0.31	1.06±0.07	1.19±0.20	1.26±0.20

See Fig. 1 for sample locations.

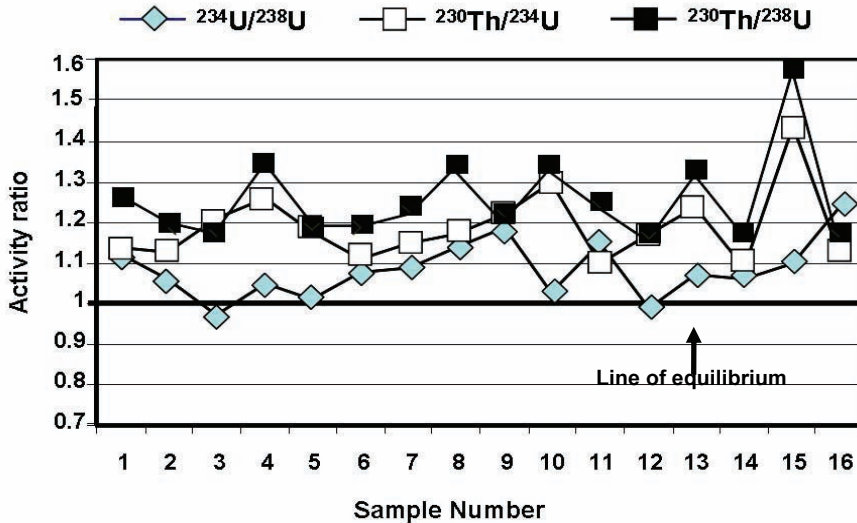


Fig. 7. Variations of the $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratios in the streambed sediments along the River Nile in Egypt.

Discussion

The uranium content of river waters depends, in general, on the distribution and mineralogical forms of uranium in the catchment area and the variation in water chemistry. Uranium and thorium in stream sediments are commonly found incorporated into minerals of detrital origin. In addition, uranium could be adsorbed directly from river water onto clay minerals or organic debris, removed from river water to sediments directly if reducing bottom condition exists (Durrance, 1986) or could be infiltrated into shallow groundwater (Lienert *et al.*, 1994). The composition of streambed sediments of the River Nile is dominated by sand size fraction followed by silt and clay sizes, respectively (Table 1 and Fig. 3). The heavy mineral varieties of these sediments reflect basically the composition of the rocks in the drainage basin. Amphiboles, pyroxene, kyanite, sillimanite, andalusite and ilmenite seem to be derived from the Ethiopian and Central African Highlands (Foucault and Stanley, 1989). Monazite and zircon are most probably of local sources, *i.e.* the Red Sea Hills and Aswan granites (Schneiderman, 1995; Dawood and Abd El Naby, 2007).

The average values of uranium and thorium in the Nile streambed sediments are 2.06 and 6.58 ppm, respectively (Table 2), which are comparable with the corresponding values in the majority of the world rivers. For example, the average uranium in the streambed sediments of the rivers draining into Gulf of Mexico is 2.48 ppm and that of thorium is 9.2 ppm (Scott, 1982). The radionuclide contents of the Nile sediments are in a continuous state of flux between the river sediments and water. Some dissolved species in the river water enhances uranium solubility while some mineral constituents, colloids and organic matters adsorb and add uranium to the sediments. Uranium is much more soluble than thorium in river water and therefore is relatively less adsorbed onto the surfaces of settling particles. Conversely, thorium is almost exclusively transported on particulate matters. This reflects the high probability of thorium to be more concentrated in the streambed sediments by the settling particulates. The main factors controlling uranium concentrations of river water are the prevailing Eh–pH condition (Scott, 1982) and availability of HCO_3^- complex, where uranyl-carbonate plays a crucial role in uranium migration (Langmuir, 1978; Mangini *et al.*, 1979; and Gorman-Lewis *et al.*, 2008). Precipitation of Fe hydroxides in some parts in the river may serve to remove additional amounts of dissolved uranium from the river water. The transport of uranium in organic-rich water can be driven by contrast in oxygen fugacity across the water-sediment interface, or uranium can be scavenged from water by organic detritus (Rosing and Frei, 2004). In addition, montmorillonite is considered as a potential sorbent of uranium. The ability of montmorillonite to sorb uranyl ions may impact the fate and transport of uranium (Borovec, 1981; Tsunashima *et al.*, 1981; Chisholm-Brause *et al.*, 2001; and Drot *et al.*, 2007). Figure 8 demonstrates the lateral distribution of uranium, thorium, heavy minerals, organic matters and clay contents in the streambed sediments of the River Nile. There are no correlations between these variables and the geographical locations; therefore, it is not possible to discern a consistent evolutionary change along the water flow path within the river basin from south to the seawards. Similar observation was reported in the River Danube (Krmr *et al.*, 2009). However, it is noticeable that the thorium concentrations are strongly correlated with heavy mineral contents (Fig. 8). Excluding samples 3, 7, 12 and 13; uranium also significantly correlates with heavy mineral contents. Samples 7 and 13 of relatively low uranium and high

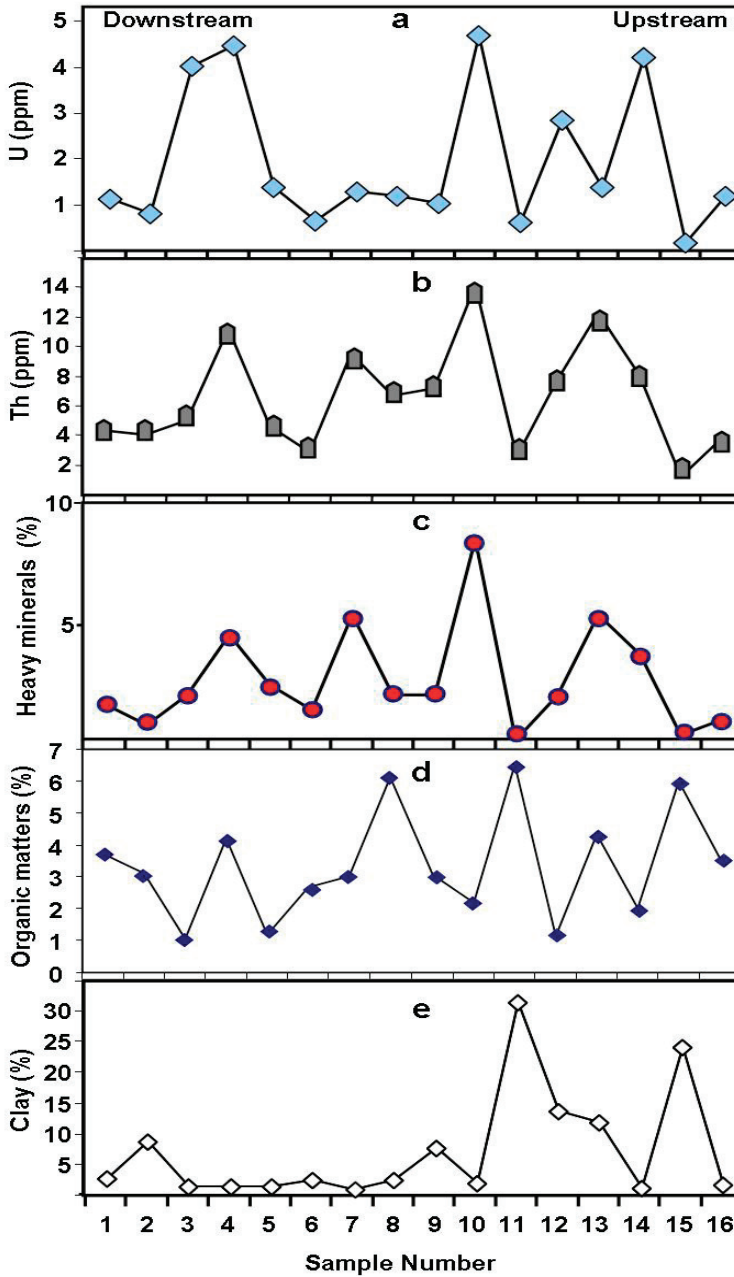


Fig. 8. Distributions of a. uranium (ppm), b. thorium (ppm), c. heavy minerals (%), d. organic matters (%) and e. clay contents (%) in the streambed sediments of the River Nile in Egypt.

heavy mineral percentages probably reflect existence of heavy mineral varieties of low uranium contents such as amphiboles, garnet and pyroxenes. The other two samples 3 and 12 show relatively high uranium contents and low heavy minerals, organic matters and clay contents (Tables 1 and 2). The microscopic investigation of the heavy mineral fractions of these particular samples revealed relative enrichment in uranium-bearing phases such as zircon and apatite which could be considered as a possible explanation for the relatively high uranium values (Fig. 8). Compared to heavy mineral contents, clays do not show any particular correlation with uranium or thorium indicating minor role for radionuclides adsorption by clays (Fig. 8). The correlations of uranium, thorium and heavy mineral contents are also indicated from the bivariant plot in Fig. 9.

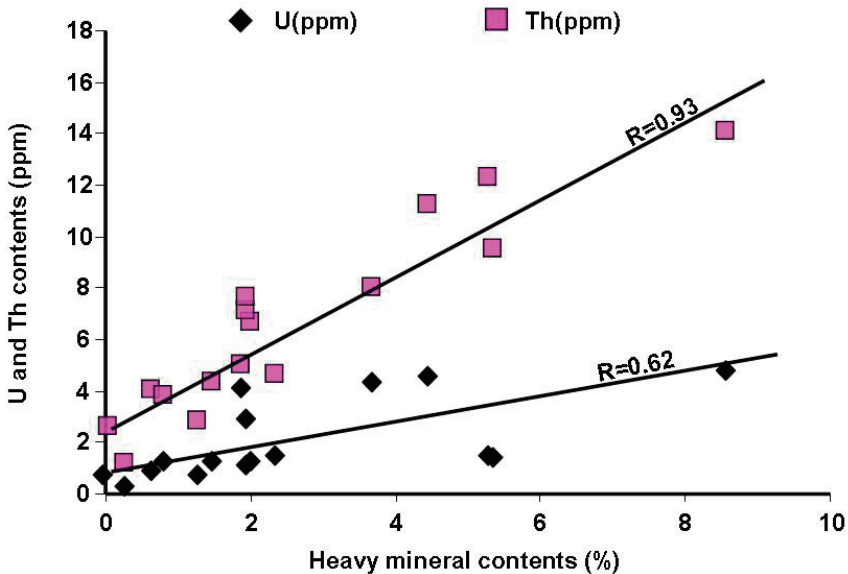


Fig. 9. Bivariant plot of uranium, thorium and heavy mineral contents.

Uranium-series isotopes are generally expressed as daughter/parent activity ratios, such as $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$. For any bedrock that has remained a closed system for more than 1 Ma, the daughter/parent activity ratios are equal to unity and the system is in "secular equilibrium". During chemical weathering, the various uranium-series isotopes have different mobility and, as a consequence,

fractionation will be induced between parent and daughter nuclides producing radioactive disequilibria. For instance, because uranium is more soluble than Th, the dissolved load in a river is expected to have a $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ ratios lower than unity, and conversely the suspended load and streambed sediments are expected to have these ratios greater than unity. The streambed sediments of the River Nile show activity ratios of $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ greater than unity (Table 2 and Fig. 7). These activity ratios are consistent with the preferential mobilization of uranium from the sediments by leaching process.

Although ^{234}U and ^{238}U are chemically the same, they also exhibit fractionation due to physical process. As a result of alpha particle emission from the parent ^{238}U , the recoiled ^{234}U is added to the water. Such fractionation is described by the equation given by Henderson *et al.* (1999):

$$^{234}\text{U}_{\text{gain}} = ^{238}\text{U}_{\text{sediment}} \cdot \lambda^{238} \cdot f$$

where $^{234}\text{U}_{\text{gain}}$ is the total ^{234}U added to the water (mol/yr); $^{238}\text{U}_{\text{sediment}}$ is the total amount of uranium contained in the sediment body which interacts with the water (mol); λ^{238} is the decay constant of ^{238}U ; and f is the fraction of ^{234}U that is ejected from the sediment into the water upon decay of ^{238}U . The ^{234}U displacement mechanism has been extensively documented in natural samples (Osmond and Cowart, 1976), and the laboratory investigations of the effects of recoiling alpha-emitting nuclei have been reported (Fleischer and Raabe, 1978). The published $^{234}\text{U}/^{238}\text{U}$ ratios of global river waters range from 1.03 to 2.59, averaging 1.17 (Chabaux *et al.*, 2001).

Uranium leaching and ^{234}U fractionation processes operate to produce the classical $^{230}\text{Th} > ^{238}\text{U} > ^{234}\text{U}$ activity sequence in the surficial sediments, so most river sediments have $^{234}\text{U}/^{238}\text{U}$ ratio between 0.9-1.0, with an average of 0.95 (Scott, 1968; and Plater *et al.*, 1992) but this trend is slightly modified by uranium sorption. Plater *et al.* (1992) concluded that if sufficient uranium is recently acquired from river water, the $^{234}\text{U}/^{238}\text{U}$ activity ratios of the sediments can mimic that of the water. During winter time, the Nile flow rate and discharge are generally low and the streambed sediments appear to have either lost more ^{238}U than ^{234}U or, more likely, acquired some uranium from water which has $^{234}\text{U}/^{238}\text{U}$ activity ratios more than unity. This results in abnormally high $^{234}\text{U}/^{238}\text{U}$ activity ratios in these sediments (1.06 in average, Table 2 and

Fig. 10). These activity ratios might be lower in summer where plenty of water is available (Plater *et al.*, 1992). This outcome is similar to the case of Himalayan Rivers (Chabaux *et al.*, 2001). Furthermore, Lewis (1976) has found $^{234}\text{U}/^{238}\text{U}$ activity ratios up to 1.8 in Susquehanna River sediments. He interpreted these abnormally high activity ratios to removal of uranium from the river water to the sediments. The importance of sorption process in controlling the uranium series isotopic characteristics of the streambed sediments is illustrated by the significant positive correlation between the organic contents of these sediments and

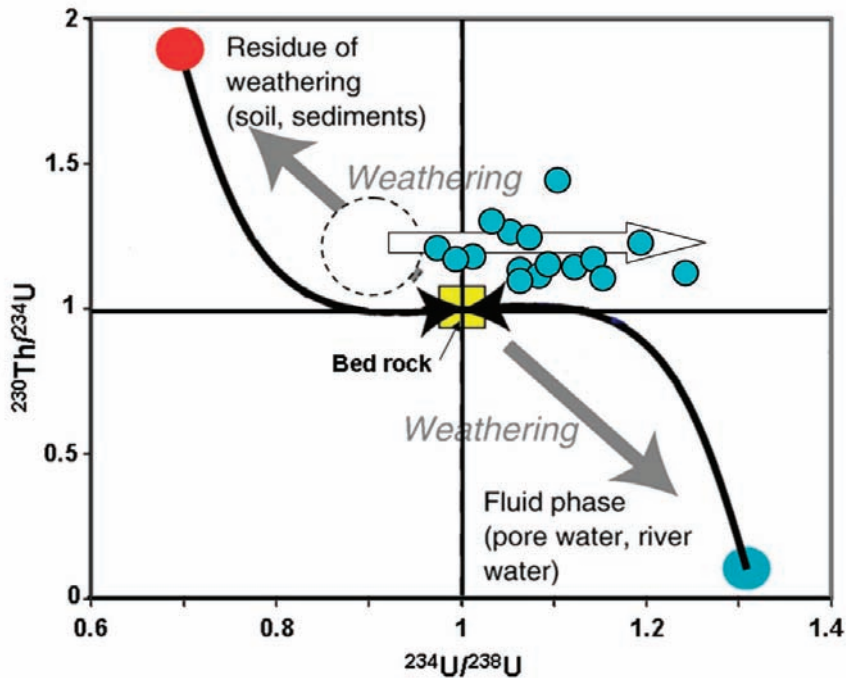


Fig. 10. Schematic diagram showing the evolution of $^{230}\text{Th}/^{234}\text{U}$ vs. $^{234}\text{U}/^{238}\text{U}$ activity ratios during weathering, modified after Dosseto *et al.* (2008). The bedrock is initially in secular equilibrium. Weathering induces radioactive disequilibrium and displaces the composition of the residues of weathering and solutes, respectively to the upper-left and the lower-right quadrants (grey arrows). Once radioactive disequilibrium is produced, each system returns to secular equilibrium by radioactive decay (black curves). The dashed line circle in the upper left quadrant was expected to be the plotting area for the River Nile sediments due to loss in ^{234}U . However, these sediments are plotted in the upper-right quadrant indicating uranium acquisition from water with high $^{234}\text{U}/^{238}\text{U}$ activity ratios.

$^{234}\text{U}/^{238}\text{U}$ activity ratios (Fig. 11). The isotopic evidence for the sorptive uptake of dissolved uranium is most apparent in sediments of high organic contents. Only samples 3 and 12 show $^{234}\text{U}/^{238}\text{U}$ activity ratios lower than unity, these samples demonstrate the lowest organic contents (Table 1 and Fig. 8). Uranium series radionuclides are adsorbed very efficiently by organic matters due to their large reactive surface. Humic acid, bacteria, fungi were reported associated with water hyacinths in the River Nile (Ghabbour *et al.*, 2004; and Ibrahim *et al.*, 2009). The humic acid is considered as an important carrier for uranium (Porcelli *et al.*, 1997), bacteria and fungi are described to be responsible for uranium reduction and precipitation (Min *et al.*, 2005).

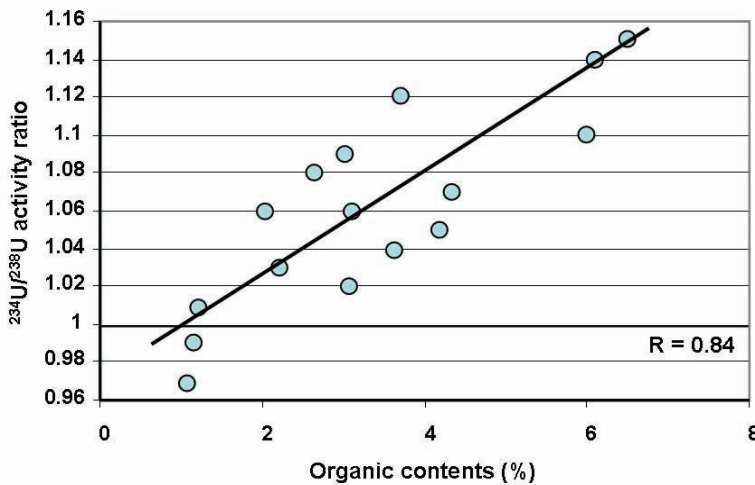


Fig. 11. Plot of organic contents vs. $^{234}\text{U}/^{238}\text{U}$ activity ratio of the streambed sediments.

Uranium and thorium in the River Nile are most probably of local sources. These sources are represented by the currently active Nile water/granitic rocks interaction at Aswan (Fig. 12), the phosphate fertilizers used in the cultivated lands along the river banks and the uranium leaching from granitic rocks of the Red Sea Hills. El-Taher (2007) reported high average uranium contents (16.64 ppm) in the granite of El-Syhail Island in the River Nile north of Aswan Dam and El-Shelal granites few kilometers to the east of the eastern bank of the Nile at Aswan. Fortunately, the rock outcrops at the upstream of the river in Ethiopia and Central Africa highlands are dominated by rocks of low uranium and thorium contents such as amphibolite, metasediments, meta-

volcanics, alkali flood basalt and tholeiitic lavas (Goodwin, 1991; and Garzanti *et al.*, 2006), that keeps uranium and thorium budget in the River Nile within the average of the worldwide rivers.

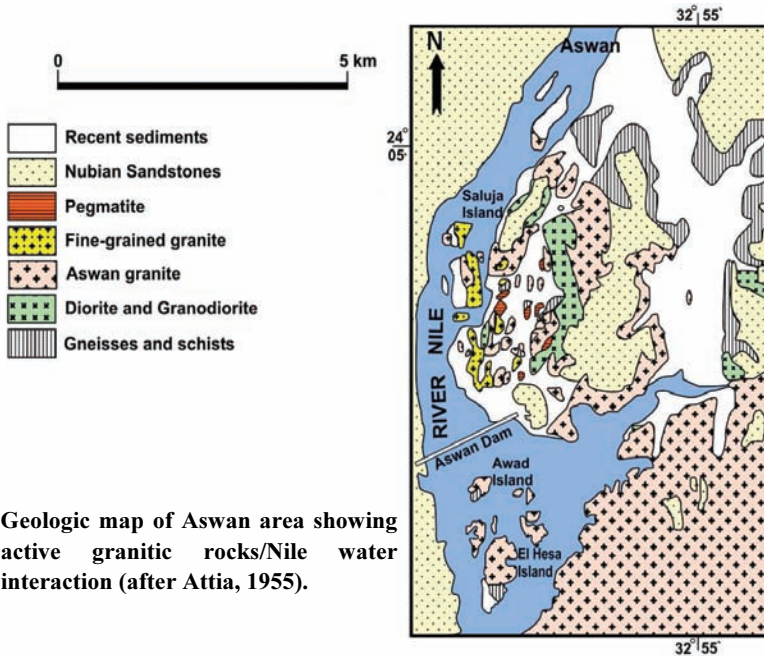


Fig. 12. Geologic map of Aswan area showing active granitic rocks/Nile water interaction (after Attia, 1955).

Conclusions

Sedimentological, mineralogical and radiogenic isotope analyses have been performed to characterize the streambed sediments of the River Nile in Egypt. These sediments exhibit large variation in mineralogical composition, so it is unfeasible to anticipate consistent values of specific activities of the radionuclides measured in a large area of the river flow. Heavy mineral contents of the streambed sediments, sorptive uptake of uranium by sediments of relatively high organic matter contents, geochemical behaviour of radionuclides and flow rates of the Nile water are considered as the most important factors controlling uranium and thorium distributions and variation of the $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratios in the streambed sediments.

The streambed sediments of the River Nile show activity ratios $^{230}\text{Th}/^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ greater than unity, which is consistent with the

preferential mobilization of uranium from the sediments by leaching process. During the low flow rate of the river, mainly in winter, the streambed sediments with relatively high organic contents acquire some uranium from water with high $^{234}\text{U}/^{238}\text{U}$ activity ratios. Consequently, the $^{234}\text{U}/^{238}\text{U}$ activity ratios of these sediments are unusually high (>1). The uranium and thorium contents of the River Nile stream sediments are comparable with the average of the World Rivers. The sources of these radionuclides are probably the local granitic rocks and phosphate fertilizers whereas the rock outcrops in the river upstream are considered as insignificant supplies.

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العوامل المتحكمة في التركيب النظائري لليورانيوم والثوريوم في رواسب قاع مجرى نهر النيل في مصر

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كلية علوم الأرض، جامعة الملك عبد العزيز

ص.ب. ٨٠٢٠٦ جدة ٢١٥٨٩ - المملكة العربية السعودية

المستخلص. تم تحليل رواسب قاع مجرى نهر النيل بمصر لمعرفة محتواها من نظائر اليورانيوم والثوريوم ذات فترة نصف العمر الطويلة (^{238}U , ^{234}U , ^{230}Th) وذلك باستخدام تقنيات المقياس الطيفي لأشعة ألفا. يغلب على هذه الرواسب المكونات الرملية الحاوية على ٢,٦٥٪ في المتوسط من المعادن الثقيلة. وتشكل الكربونات والمواد العضوية في هذه الرواسب مكونات يمكن أن تؤخذ في الاعتبار حيث يبلغ متوسطها ١٣,٨٢ و ٣,٣٧٪ على الترتيب. بينت نتائج الدراسة أن متوسط تركيزات اليورانيوم والثوريوم هي ٢,٠٦ و ٦,٥٨ جزء في المليون على الترتيب وهي قيم متقاربة مع مثيلتها في أنهار العالم. بلغت نسب النشاط الإشعاعي $^{230}\text{Th}/^{234}\text{U}$ و $^{230}\text{Th}/^{238}\text{U}$ قيمةً أكثر من الوحدة في رواسب قاع مجرى النهر دلالة على الإذابة التفاضلية لليورانيوم من الرواسب. عندما تقل سرعة تدفق المياه في النهر، تكتسب رواسب المجرى بعض اليورانيوم من مياه النهر الحاوية بطبيعتها على نسبة النشاط الإشعاعي $^{234}\text{U}/^{238}\text{U}$ بقيم أعلى من الوحدة، مما يؤدي لزيادة هذه النسبة في الرواسب بصورة غير عادية. يعتبر كل من محتوى المعادن الثقيلة واستحواذ الرواسب ذات المحتوى العضوي على اليورانيوم بالادمصاص، والسلوك الجيوكيميائي للعناصر المشعة،

ومعدل سريان مياه نهر النيل، من أهم العوامل المتحكمة في التركيب النظائري لليورانيوم والثوريوم في رواسب القاع. للعناصر المشعة في نهر النيل بمصر مصادر محلية كالصخور الجرانيتية والأسمدة الفوسفاتية المستخدمة في الزراعة بالقرب من النهر.