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The semi-annual variations of the bio-available heavy metals and natural radionuclides in Timsah Lake sediments, Egypt

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Abstract—The granulometric characteristics, the bio-available heavy metals and the natural radionuclide activities in the surface sediments were investigated semi-annually in Timsah Lake at the middle district of the Suez Canal, Egypt. The average percentages of mud and fine grained sediments recorded in summer were (4.17% and 33.89%) much higher than in winter (1.43% and 22.70%) attributed to the relatively high dispersing of the fine sediment fractions in winter by wave action and the fine sediments drifting towards Suez Canal. The average carbonate percentage was 19.72% in summer increased to 22.71% in winter, inversely, the average total organic matter (TOM) in summer was 7.52% decreased to 6.32% in winter. The highest averages of the bio-available heavy metals; Zn, Cu, Pb, Cd, Mn, Co, Ni and Fe were; 65.51, 18.06, 27.76, 0.78, 260.64, 4.10, 17.16 and 2087.71µg/g were recorded summer and the highest average activities of ^{238}U, ^{232}Th and ^{40}K were 23.79, 23.72and 221.35Bqkg^{-1} were recorded in winter. The recoded heavy metals and radionuclides were attributed to multi anthropogenic sources; untreated wastewater drains, agriculture drains, industrial runoff and shipyards. The high values of TOM and bioavailable heavy metals in summer are related to the highest fine sediment percentages, while the radionuclides may tend to associate with the coarse sediments. The significant positive correlations of TOM and Fe with heavy metals and radionuclides indicated to two essential metal phases, one with organic matter in the highly reducing conditions and the other associated and/or adsorbed by Fe-oxides and hydroxide particles in addition to the other independent metal phases. The recorded bioavailable metals are lower than the excepted because of suspended matters and water drift toward Suez Canal dilute the metal accumulation in the lake sediments.

Index Terms— Agriculture drains, sewage, bio-available heavy metals, natural radionuclides, Timsah Lake and Suez Canal.

1 INTRODUCTION

Overpopulation, industrialization, rapid urbanization, overuse of pesticides, detergent and agricultural chemicals, liquid and solid waste products and discharge of municipal wastes are the main contaminant sources of heavy metals and radionuclide in the natural water resources [1]. Because of the marine environment is a dynamic system, heavy metals and radionuclides introduced to the surface waters by liquid discharges do not stay there in steady-state conditions, but due to currents and other processes in the water column, they are transported both horizontally and vertically to different regions, as well as to bottom waters and sediments. Such pollutions can negatively impact human health and ecosystems through a range of accumulatory processes within the food chain. The environmental impacts of heavy metals and radionuclides are determined not by the total concentration but by their physical and chemical forms within the environmental system. The increased loading of heavy metals and radionuclides in the aquatic ecosystems furnished an imbalance state and threatened the health of the native biota growing under such abnormal habitat conditions, consequently, the accumulation rates of these metals are being assimilated and transferred within food chains by the processes of bioaccumulation and bio-magnification [2], [3]. The concentrations and the particular forms of the heavy metals, and their interactions with the other components of a soil, determine its potential to cause toxic effects in biological systems. Increasing numbers and quantities of radioactive materials in many different forms are being transported throughout the world, resulting in increased public concern about radiation safety in transport.

The aquatic ecosystem of Timsah Lake recorded hazardous levels of pollutants of various forms; pesticides and hydrocarbons [4], heavy metals concentrations in the seawater, bottom sediments as well as the edible marine fauna [5], [6]. The rapidly growing human activities in the last 35 years around the lake such as ship building and maintenance, municipal wastewater damping off and agricultural drainage loading have greatly increased the eutrophication and pollution status of the lake [7], [8] and consequently these conditions threatened the lake health, interfere with its recreational purpose, lake richness and diversity of indigenous fish, phytoplankton, zooplankton, plants and animal population. Subsequently, the present study aims to delineate the sources, magnitude, interactions and the geochemical cycles of heavy metals and radionuclides in the surface sediments of Timsah Lake.

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Geomorphic and environmental settings of Timsah Lake

Timsah Lake lies adjacent to Ismailia City at the middle district of the Suez Canal about 80 km south of Port Said, Egypt (Fig. 1). It plays an important role in most of the human activities in Ismailia City such as; tourism, fisheries, navigation, etc. [9]. Timsah Lake covers about 16 km² with depth variation

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between 3 and 16 m and capacity volume of about 90 million cubic meters of seawater [10]. The lake is considered one of the most productive zones along the Suez Canal [7], [11], it is the major source for fishes, crustaceans and shellfish that are largely consumed by local populations. Additionally, the northern and western boundaries shores of the lake are rapidly developed for tourism purpose, aquatic sports and recreational activities [12]. Timsah Lake represents a unique aquatic ecosystem due to the different types of water inputs. From the western side, the lake is connected to a small and shallow embayment that receive about 833,000m^3/day of treated and untreated domestic and agricultural wastewaters throughout many drains; Elmahsama, Abu-Gamouss, Abu-Attwa and El-bahtini drains [6], [13] and from the northern side, the lake receives occasional freshwater inputs from the Ismailia Channel. Despite the diminishing amounts of wastewaters, the lake is threatened from other pollutants as ships awaiting berth and the huge Timsah Shipyard [8], [14], [15] as well as the extensive human settlements whereas the domestic and industrial effluents are continuously discharged.

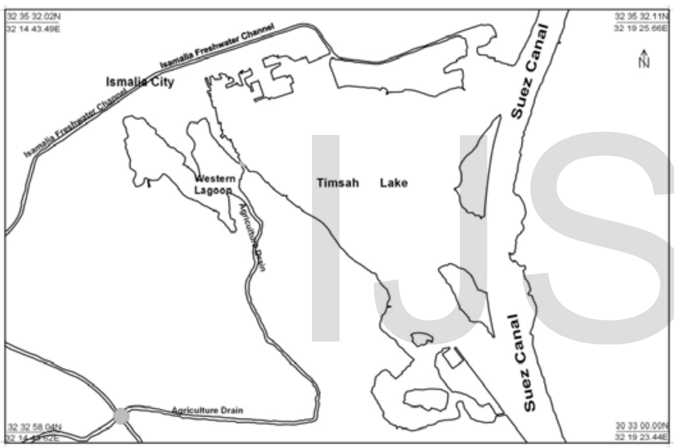

Fig. (1) Location map for Timsah Lake.

2 Materials and Methods

The sediment samples were collected semi-annually during summer and winter, 2013 from 12 sites covering the different litholgic features of the lake (12 samples at each season) using small boat and grab sampler. The samples were air dried, disaggregated then sieved through a stainless steel mesh in order to differentiate the particle-size fractions. Grain-size analyses of sediments were performed by dry method each one phi interval [16]. Seven fractions were obtained; gravel ($Ø_{-1}$>2.00mm), v. coarse sand ($Ø_0$=2.00 : 1.00mm), coarse sand ($Ø_1$=1.00 : 0.50mm), medium sand ($Ø_2$=0.50 : 0.250mm), fine sand ($Ø_3$=0.250 : 0.125mm), v. fine sand ($Ø_4$=0.125 : 0.063mm) and mud ($Ø_5$<0.063mm). To illustrate the effects of oceanographic conditions on the seafloor sediments of the lake during the study seasons, the obtained sediment fractions were categorized into three groups; coarse grain sediments ($Ø_{-1}$+$Ø_0$), medium grain sediments ($Ø_1$+$Ø_2$) and ($Ø_3$+$Ø_4$ + $Ø_5$) as fine grain sediments (Table 1).

3.1. Geochemical analyses

3.1.1. Carbonate contents and total organic matter (TOM) determination

The carbonate percentages were determined by treating one gram of powdered sample with 1 N HCl acid. The remaining insoluble residue after acid washing was determined and the carbonate percentages were calculated [17], [18] according the formula:

$$CO_3\% = \frac{\text{wt. of sample} - \text{wt. of residue}}{\text{wt. of sample}} x\, 100$$

Determination of total organic matter (TOM) was made by sequential weight loss at 550°C [19] according the formula:

$$TOM\% == \frac{\text{wt. of sample} - \text{wt. of ash}}{\text{wt. of sample}} x\, 100$$

3.1.2. Determination the bio-available heavy metals

Five grams of each homogenized sediment sample were grinded using agate mortar to less than 80mesh then used to determine the bio-available heavy metals (oxides, hydroxides, carbonates and sulphides). About 0.5g of each powdered sample was digested with a mixture of HNO_3 and $HClO_3$ to near dryness [20]. The digested samples were filtered to remove insoluble residuals then diluted with DDW to 25ml. The bio-available heavy metals of: Zn, Cu, Pb, Cd, Mn, Co, Ni and Fe were determined in the sample extracts using flame Atomic Absorption Spectrophotometer (AAS, GBC-932) at National Institute of Oceanography and Fisheries, Red Sea Branch, Hurghada. To insure that the maximum accuracies were obtained, three replicates of each measurement were applied with differences less than 3%. The resultant data were expressed in (μg/g).

3.1.3. Natural radionuclides determination

About 250gm of sample was dried in an oven at about 110ºC to ensure that moisture was completely removed. The samples were grinded using agate mortar to less than 80mesh. Weighed samples were placed in polyethylene beaker, of 350-cm^3 volumes each. The beakers were completely sealed for 4 weeks to reach secular equilibrium where the rate of decay of the progeny becomes equal to that of the parent (radium and thorium) within the volume and the progeny will also remain in the sample [21], [22].

The activity measurements of ^{238}U, ^{232}Th and ^{40}K were performed by γ-ray spectrometer at the faculty of Science, Azhar University, Assuit, Egypt, employing a scintillation detector (3" x 3"). It is hermetically sealed assembly, which includes a NaI (Tl) crystal, coupled to PC-MCA Canberra Accuspes. To reduce gamma ray background, a cylindrical lead shield (100 mm thick) with a fixed bottom and movable cover shielded the detector. The lead shield contains an inner concentric cylinder of copper (0.3 mm thick) in order to absorb X-rays generated in the lead. In order to determine the back-

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ground distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The measurement time of activity or background was 43200s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program (Genie 2000) has carried out the online analysis of each measured gamma ray spectrum. The ^{232}Th concentration was determined from the average concentrations of ^{212}Pb (238.6 keV,) and ^{228}Ac (911.1 keV) in the samples, and that of ^{238}U was determined from the average concentrations of the 214Pb (351.9 keV) and ^{214}Bi (609.3 keV and 1764.5 keV) decay products. While the gamma line for ^{40}K is (1460.6 keV). The minimum detectable activity (MDA) was 25.2 Bq/kg for ^{40}K, 6.5Bq/kg for ^{238}U and 5.7 Bq/kg for ^{232}Th as described by [23], [24].

4. Results and Discussion

4.1. Sediment characteristics

Timsah Lake sediments tend to be fine, it is composed of variable mixture of sand and mud with varying hues [25]. The formations of the bottom sediments of the lake show the presence of fluviatile sediments in the central part which is identical with species now living in the Nile. These sediments are gradually replaced in the south by typical Red Sea marine sediment and in the north by Mediterranean sediments [26]. Bedding is of alternating layers of fine to coarse grain sands. These layers are few meters thick, often silty or calcareous, sometimes, with gypsum or clay as minor constituent [26]. Gab-Alla [27] recorded that the sediments of Timsah Lake were sandy ranging from very fine sand to fine sand. He added, *C. prolgera* have a great sediment retention capacity, favoring stabilization and organic enrichment of the environment. Also, the plant density may affect the granulometric composition, shifting it to very fine sand and clay and increasing percentage of organic matter of the sediment, through slow down of water movements by the seaweed blades.

The recorded average percentages of gravel, sand and mud in summer were; 3.24%, 92.59% and 19.72% respectively. In winter, the recorded average percentage of gravel was increased to 15.89% while the average percentages of sand and mud ware decreased to 82.67% and 1.43% respectively. The coarse sediments group recorded the lowest average percentage in summer (19.05%) and the highest average recorded in winter (31.20%). The fine sediments group showed the highest average percentage in summer (33.89%) and the lowest average in winter was 22.70% (Table 1; Fig., 2). Ewais *et al.*, [28] pointed out; fine sediments usually have a much greater capacity to sorb metals and radionuclides than coarse sediments. This is normally attributed to a combination of the greater specific surface area of fine sediments and the greater exchange capacity of clay minerals, which usually have particle diameters of only a few µm. In spite of the discharged fine and particulate sediments to Timsah Lake from the different drainages are much higher than the recorded percentages in the collected sediments, it is clear that most of these fine and particulate fractions were dispersed by waves and marine currents. Fig., (2) Grain size variations between summer and winter.

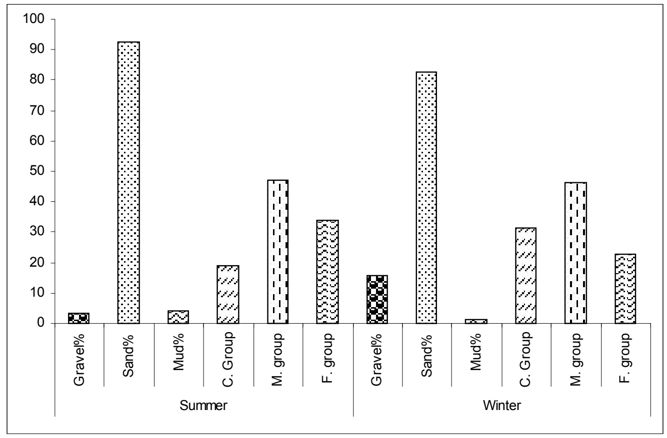

Fig., (2) Grain size variations between summer and winter

Table (1)

The variation in sediments characteristics between summer and winter seasons:

	Summer						Winter					
	Grav-el%	Sand%	Mud%	C. Group	M. group	F. group	Grav-el%	Sand%	Mud%	C. Group	M. group	F. group
1	3.54	93.51	2.95	14.87	54.41	30.72	3.62	96.22	0.16	10.97	65.08	23.95
2	3.56	95.49	0.95	14.07	69.60	16.33	3.31	91.75	4.94	12.31	57.22	30.47
3	5.21	89.57	5.22	31.12	42.89	25.99	2.93	96.78	0.29	10.68	83.31	6.01
4	0.94	95.50	3.56	4.11	63.94	31.95	3.51	94.40	2.09	18.42	52.56	29.02
5	2.76	95.22	2.02	13.47	49.91	36.62	11.07	87.75	1.18	28.95	57.37	13.68
6	6.44	87.92	5.64	27.88	47.38	24.74	42.36	56.29	1.35	65.83	23.71	10.46
7	7.86	85.29	6.85	17.78	47.14	35.08	22.60	76.53	0.87	38.78	44.81	16.41
8	0.89	88.07	11.04	15.13	48.44	36.43	33.60	64.80	1.60	61.28	26.44	12.28
9	3.92	93.08	3.00	8.69	25.78	65.53	18.37	80.35	1.28	43.02	39.89	17.09
10	0.06	91.94	8.00	1.40	15.44	83.16	0.98	98.73	0.29	5.45	58.46	36.09
11	2.49	97.01	0.50	40.61	48.39	11.00	45.38	54.10	0.52	69.18	15.04	15.78
12	1.20	98.50	0.30	39.45	51.48	9.07	2.99	94.38	2.63	9.56	29.23	61.21
Max.	7.86	98.50	11.04	40.61	69.60	83.16	45.38	98.73	4.94	69.18	83.31	61.21
Min.	0.06	85.29	0.30	1.40	15.44	9.07	0.98	54.10	0.16	5.45	15.04	6.01
Aver-age	**3.24**	**92.59**	**4.17**	**19.05**	**47.07**	**33.89**	**15.89**	**82.67**	**1.43**	**31.20**	**46.09**	**22.70**

In winter, the rate of fine and particulate sediments dispersing is much higher than in summer affecting by the wind speed, waves and the current drift towards Suez Canal. Abril and Abdel-Aal [29] reported that, in the Suez Canal, one can expect the suspended matter concentrations to be strongly dependent on the ship traffic. Extreme weather and hydraulic conditions can produce important changes in fluxes of matter and sedimentation rates. Nevertheless, most of the time, the suspended load concentrations and the instantaneous sedimentation rates show a dynamic equilibrium governed by the tidal changes in the settling and re-suspension velocities. They assumed that 25% of the material in the top layer of bottom sediments in the

Suez Canal and connected lakes can be re-suspended when the water velocity is higher than a critical velocity of 0.232 m/s. The maximum re-suspension velocity was stated as 3.4 mm/year. Particles settle down when the water velocity is lower than 0.18 m/s. The current regime in Suez Canal is northward in winter and southward in summer, whereas, the wind and the annual gradient of the mean sea level (MSL) are mostly responsible for the current regime in the canal [30]. Waves generated in Suez Canal and its associated lakes are by tides, winds and the transit ships. The tidal waves are of long wave type, while those due to wind and ship motions are of short wave type. The waves generated by wind show values of the order of 20 cm in the normal cases while it may exceed 50cm in storms while, the maximum wave heights generated by transiting ships through the Suez Canal are estimated to be 10 cm [30].

4.2. Carbonates and total organic matter (TOM) percentages in the lake sediments

In the places with slake water circulation, the bottom marine sediments contain very high proportions of organic matter, carbonates, sulfides, and chlorides [31]. The discharge of different wastes into lakes generally results in much higher concentrations of heavy metals and other contaminants in bottom sediments than in the overlying water. This reflects high sorbtive and binding rates of the substrate, particularly in areas where organic material is abundant [32].

The recorded average percentage of carbonates was 19.72±7.36% in summer increased to 22.71±8.55% in winter. The sources of carbonates in the lakes are mainly bivalve shells and their debris that mainly accumulated in the coarse grain sediments. The increasing of carbonate percentage in winter is related to the effect of waves and currents that leached the fine grain sediments from the lake. Billon *et al.*, [33] reported that Mn, Cu, Cd, Pb and Zn distributions are easily adsorbed onto the surface of carbonates followed by their incorporation into the lattice of carbonates to form a solid solution. The high proportion of carbonates contributes to the high alkalinity and buffering capacity of the sediments, and as a consequence, large amounts of acids are generally required to extract the metals.

Organic compounds in sediment, frequently existing in considerable amounts in particle form, play an important role in heavy metal transformation. In the sediment of some lakes, the heavy metal bound to OM generally takes up the largest fraction. Additionally, in sediment, the solubility of organic matters usually directly determines the mobility of heavy metals. Normally, the complexation of metal ions with insoluble organic compounds can strongly lower their mobility, whereas the formation of soluble metal complexes with dissolved organic compounds would enhance their mobility [34].

In lakes, OM is mainly composed of humic and fulvic substances. The complexation reaction between heavy metals and organic complexants is usually recognized as the most important reaction pathway, due to this reaction determining, to a large extent, the speciation and bioavailability of metal, and then influencing the mobility of trace metal in natural water environment. However, in severely polluted river, due to the complexity of organic matter, the reaction types between organic complexes and metals are difficult to predict. In most conditions, precipitation, co-precipitation or flocculation usually plays the most important role in heavy metal fixation [31].

Total organic matter (TOM) recorded 6.32±4.60% in winter increased to 7.52±5.49% in summer that may related to fine and particulate sediments increasing (Table 2). Organic matter in Timsah Lake is resulted from the untreated organic wastes and the high biological productivity of the lake. Gab-Alla [27] attributed the organic matter contents in the Timsah Lake sediments to the high density of aquatic plants. Organic matter combines with heavy metals, forming metal–organic complexes which are very stable [26]. Organic matter plays an important role not only in forming complexes but also in retaining heavy metals in an exchangeable form [35]. Hydrodynamic processes induce the accumulation of fine sediments associated with organic matter in zones characterized by lower hydrodynamic energy or a more efficient absorption of organic matter over the greater net surface area characteristic of fine sediments. Hoz *et al.,* [36] attributed the enrichment of trace metals in organic rich sediments to the fact that trace elements tend to concentrate within the surface of finer grained sediments.The nature of the organic matter is important in terms of effective binding and fixation of radionuclides to particles by active ligand groups, affecting transfer and sedimentation processes [37]. The fine grain sediments and suspended matters are the most important medium for transporting metals and may be deposited to form contaminant sinks.

4.3. The bio-available heavy metals accumulations

The discharge of the human wastes, industrial and agriculture drainages into the closed and semiclosed lakes result in much high concentrations of heavy metals and other contaminants in the bottom sediments than the overlying water. This reflects high sorbative and binding rates of the substrate, particularly in the areas where organic materials are abundant. The unstable environmental conditions may result in the large scale transport of pollutants along the bottom and in the overlying water, which in turn accounts for the occurrence of the different wastes many kilometers from the drainage sources [32]. Heavy metals are considered the most hazardous contaminant in the environment due to their persistence and accumulation in water, sediments and in tissues of the living organisms; this is by bioconcentration and biomagnification [12].

Sediments of Timsah Lake have comparatively large contents of fine particles, such as silt and clay, and exhibit high alkalinity values. These unique characteristics directly influence the mobility of heavy metals. The average contents of the bio-available heavy metals; Zn, Cu, Pb, Cd, Mn, Co, Ni and Fe that recorded in summer season were; 65.51±37.63, 18.06±12.85, 27.76±19.97, 0.78±0.80, 260.64±173.09, 4.10±4.60, 17.16±14.08 and 2087.71±326.73µg/g while the recorded averages in winter season were; 58.44±36.56, 20.26±24.46, 14.57±5.92, 0.56±0.49, 197.70±114.97, 1.98±3.04, 13.32±9.23, 2004.13±373.36µg/g respectively (Table 2). Wide variations observed between summer and winter seasons were for Zn, Pb, Mn and Co concentrations may attribute to the fine sediment particles dispersing during winter by the wind induced waves, drag and tidal currents. The other recorded metals show slight variations between summer and winter indicating to the nearly homogeneous distribution of metal occurrences between the different sediment fractions. Zeng and Wu, [38] found that Cu, Zn, Pb, Co, Mn and Fe were generally originated from anthropogenic activities and enter into the lakes through water flow. Hoz *et al.,* [36] attributed the high metals contents in Lake Chapala (Mexico) to dredge disposal, sewage and agriculture discharges. Baek and An [39] abstracted that the Elevated metal levels in urban lake sediments are associated with urban runoff, including street dust polluted by heavy metals.

Generally, the recorded bio-available contents of Cu, Co, Ni and Fe in summer and winter seasons at Timsah Lake were lower than the total heavy metals and bio-available fractions measured in the other lakes of Egypt and most of worldwide lakes (Table 3). The recorded bio-available averages of Zn were nearly equal the total Zn content at Nasser Lake [40], higher than the bio-available Zn average at Nasser and Brullus lakes [40], [41] and total Zn at Manzala Lake [44] but much lower than total Zn at Idku, Brullus and Manzala lakes [42] and Qarun Lake [43]. Winter season at Timsah Lake shows bio-available Pb average nearly equal the labile average at Nasser Lake [40] and total average at Brullus Lake [42]. In summer season the bio-available Pb was higher than the total Pb at Qarun Lake [43] and lower than the total and mobile Pb at Brullus Lake [41] and the total Pb at Edku, Manzala and Nasser lakes [40], [42], [44]. Bio-available Cd was nearly equal total Cd at Manzala and Nasser lakes [40], [44] bio-available Mn was higher than Manzala Lake [44] while both bio-available Cd and Mn were lower than the other lakes (Table 3). The recorded metals were lower than Chapala Lake, Mexico [36], [45] Taihu, Dianchi Nansi and Yangtze lakes, China [46], [47], [48], [38] Lake Dautk, Uzbekistan [49], lakes of Kumaun, India [50] and Maharlu Lake, Iran [51] and higher than Lake Karla, Greece [47] and Avsar Lake, Turkey except Fe [52].

Metal concentrations in summer follow the sequence; Fe>Mn>Zn>Pb>Cu≥Ni>Co>Cd and in winter the metal concentrations follow the order; Fe>Mn>Zn>Cu>Pb≥Ni>Co>Cd. Forghani *et al.,* [51] found that the mean metal concentration in Maharlu Lake, Iran sediments decreases in the following order: Fe >Mn >Pb ≈ Ni >Cu >Co>Zn>Cd. Özmen et al., [53] recorded the heavy metals concentration of the sediments were found decrease in sequence of; Fe > Mn > Zn > Ni > Cu > Co > Pb in sediment of Hazar Lake, Turkey. Abd El Samie *et al.,* [26] measured the heavy metal contents in the seawater of Timsah Lake, they found that the heavy metals concentration is significantly high in the north and western edges of the lake more than the middle affecting by the outflow wastewater from the western lagoon. They found that, the heavy metals increase in low salinity water toward the land from the discharging effluent. They added, the low mixing rates due to slow current of lake water led to long residence time of the pollution load enhancing accumulation and precipitation of the heavy metals to the bottom sediment near the boundaries of the lake.

4.4. The natural radionuclides occurrences

The radionuclides are deposited to the sea bottom sediments through a wide range of processes, including fixation on suspended particulate matter, direct precipitation of colloidal forms (coagulation, aggregation), direct fixation by adsorption, absorption on clay minerals and complexation with organic matter [37]. The average activities of ^{238}U, ^{232}Th and ^{40}K in the sediments of Timsah Lake that recorded in winter were 23.79±13.93, 23.72±11.20 and 221.35±143.12 $Bqkg^{-1}$ dry weight respectively and the average activities recorded in summer were; 19.53±7.65, 19.12±12.17 and 179.25±142.05 $Bqkg^{-1}$ dry weight respectively (Table 2). As shown in table (4), the average activities of ^{238}U recorded at Timsah Lake were nearly equal to that recorded at Qarun Lake [54] and Idku Lake [55] but higher than the recorded activities in Suez Canal [56], Brullus and Mariout lakes [57], [58, Nasser Lake [59] and Idku beach sediments [55]. However it was much lower than the recorded average activity at Isamlia Freshwater Canal [60]. The measured ^{232}Th average activities in Timsah Lake were nearly equal to Brullus Lake [58] and Isamlia Freshwater Canal [60], higher than Suez Canal [56], Qarun Lake [54], Brullus and Mariout lakes [57] but lower than Nasser Lake [59] and Idku Lake [55]. The recorded averages of ^{40}K in Timsah Lake sediments were nearly equal to Suez Canal [56], lower than Brullus and Mariout lakes [57], [58], Idku Lake [55], Nasser Lake [59] but much higher than Isamlia Freshwater Canal [60]. Also, the recorded averages of ^{238}U and ^{232}Th were higher than Lake Dautk, Uzbekistan [49], Deriner and Borcka lakes, Turkey [61], [62] Moticher lake, India [63] and Butrint Lagoon, Albania [64] but in the same range of Kainji Lake, Nigeria [65], Derbent nd Muratlı lakes, Turkey [61], [62] and lower than NE Tamilnadu, India [66] and the world wide averages [67], [68]. Potassium-40 shows high activities at most of the worldwide lakes.

The slightly high radionuclide averages in winter may be attributed to the high rates of untreated sewage inflow and/or active transformation from soluble phase to solid phase in winter and from solid phase to soluble phase in summer. Aguado [69] found that ^{226}Ra concentration in sediments in-

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creases with TDS due to the release of ^{226}Ra from suspended particulate matter which subsequently undergo pH induced coagulation and eventual precipitation to the sediments. Sérodes and Royb [70] inferred that ^{226}Ra and thorium are preferentially present as adsorbed forms. They added, these radionuclide concentrations should be proportional to suspended solid values which show a good correlation between suspended matter and radionuclide concentrations. Some parameters affected the sorption behaviour of radionuclides on suspended matters and bottom sediments such as; solution pH, *SM* concentration, sediment grain size, carrier concentration and competing ions [60].

4.5. Transportation, occurrences and the geochemical cycles of heavy metals and radionuclides

The environmental impacts of heavy elements and radionuclides are determined not by the total concentration but by their physical and chemical forms within the environmental system [71]. The chemical and physical behaviors of heavy metals and radionuclides in sediments depend on the constituents of the sediments (i.e. amount and type of clays, oxides/hydroxides, carbonates, organic matter and the colloidal phases), pH and redox conditions as well as water/sediments interactions that mainly controlled by adsorption/desorption between water and sediment particles [72]. They defined water/sediments interactions as the sum of sediments leaching by water and particulate sediments adsorption from water. Thus, the distribution of heavy metals and radionuclides between dissolved species, and reversibly or irreversibly bound solid fractions can affect their bioavailability [73].

Table (2):

Carbonate and TOM percentages, heavy metals (in μg/g) and the activities of the natural radionuclides (in Bq/Kg dry wt.) in summer and winter seasons at Timsah Lake:

	CO3%	TOM%	Zn*	Cu*	Pb*	Cd*	Mn*	Co*	Ni*	Fe*	U-238**	Th-232**	K-40**
	13.00	3.00	51.02	10.71	13.39	1.43	92.63	0.43	8.01	1747.15	10.21	4.28	36.83
	13.10	2.90	9.79	10.24	21.25	0.87	83.89	0.94	8.94	1808.96	22.49	16.57	16.63
	29.80	17.00	97.37	47.56	28.55	0.51	294.54	5.89	50.47	2494.53	26.92	34.82	305.68
	16.80	2.30	45.32	5.98	9.99	1.45	84.32	0.20	5.10	1649.13	17.62	14.76	102.17
	21.90	2.70	39.21	8.31	41.40	2.81	98.58	0.47	9.20	1785.27	7.03	8.79	58.72
	14.20	13.30	86.06	16.85	15.01	0.41	237.44	3.74	15.32	2310.47	19.52	21.02	167.89
	28.80	5.68	29.64	10.63	26.66	0.37	185.39	1.32	8.62	1967.14	16.04	22.97	37.89
	25.80	16.80	119.24	27.63	47.47	0.93	470.08	9.30	23.51	2412.85	33.63	43.72	290.48
	31.10	3.90	20.11	7.62	15.45	0.43	217.42	0.10	3.50	1808.14	12.82	5.17	141.43
0	10.90	10.10	84.11	28.03	20.36	0.08	521.73	11.94	33.86	2454.78	27.32	24.90	199.11
1	14.80	4.96	86.99	11.31	14.01	0.11	279.09	2.77	12.02	2145.48	17.66	7.24	327.90
2	16.40	7.58	117.28	31.81	79.60	0.01	562.58	12.07	27.33	2468.66	23.08	25.15	466.23
ax.	31.10	17.00	119.24	47.56	79.60	2.81	562.58	12.07	50.47	2494.53	33.63	43.72	466.23
in.	10.90	2.30	9.79	5.98	9.99	0.01	83.89	0.10	3.50	1649.13	7.03	4.28	16.63
v.	19.72	7.52	65.51	18.06	27.76	0.78	260.64	4.10	17.16	2087.71	19.53	19.12	179.25
D	7.36	5.49	37.63	12.85	19.97	0.80	173.09	4.60	14.08	326.73	7.65	12.17	142.05
	18.10	2.50	51.31	7.30	12.53	0.49	77.13	0.01	4.17	1672.00	33.56	29.24	399.02
	20.20	2.65	36.00	9.15	11.15	0.07	121.72	0.01	11.27	1734.90	8.05	7.32	79.83

	3	21.70	3.22	25.94	8.94	25.27	1.19	56.96	0.01	16.28	1636.34	10.99	12.68
	4	18.70	5.54	145.75	93.91	10.49	0.24	155.36	5.33	9.63	2361.12	10.58	15.25
	5	31.30	5.90	50.13	11.89	19.01	1.34	174.98	0.01	13.01	2101.37	17.76	21.23
	6	28.90	15.49	83.78	30.44	24.83	1.27	374.78	3.37	25.16	2371.46	21.34	33.89
	7	35.50	5.26	46.11	8.71	7.17	0.55	170.17	0.01	4.93	1919.22	14.69	16.53
	8	26.60	16.02	76.72	24.83	15.21	0.01	398.01	6.77	27.53	2395.15	34.41	34.95
	9	32.70	4.50	15.46	10.93	10.05	0.15	194.20	0.26	6.16	1913.51	35.25	31.18
	10	15.20	5.54	43.09	13.27	16.06	0.71	288.30	0.02	14.90	2297.95	22.18	24.66
	11	5.70	2.99	30.77	3.78	8.58	0.01	75.74	0.01	0.01	1264.96	20.19	13.23
	12	17.90	6.24	96.17	19.99	14.45	0.63	285.09	7.91	26.83	2381.54	56.46	44.54
	Max.	35.50	16.02	145.75	93.91	25.27	1.34	398.01	7.91	27.53	2395.15	56.46	44.54
	Min.	5.70	2.50	15.46	3.78	7.17	0.01	56.96	0.01	0.01	1264.96	8.05	7.32
	Av.	22.71	6.32	58.44	20.26	14.57	0.56	197.70	1.98	13.32	2004.13	23.79	23.72
	SD	8.55	4.60	36.56	24.46	5.92	0.49	114.97	3.04	9.23	373.36	13.93	11.20

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Table (3)

The comparisons of heavy metal contents in (μg/g) between Timsah Lake and other lakes in Egypt and worldwide:

Lake Name	Zn*	Cu*	Pb*	Cd*	Mn*	Co*	Ni*	Fe*	Observation	Reference
Egypt										
Timsah (Summer)	65.51	18.06	27.76	0.78	260.64	4.10	17.16	2087.71	Mobile fractions	Present Study
Timsah (Winter)	58.44	20.26	14.57	0.56	197.70	1.98	13.32	2004.13	Mobile fractions	
Edku	344.45	36.77	37.14	1.47	1390.13	-	-	6253.99	Total metals	[42]
Brullus	217.33	47.49	13.08	4.62	850.95	-	-	10999.49	Total metals	
Manzala	432.16	315.36	134.64	84.80	419.60	-	-	33386.64	Total metals	
Brullus	78.41	113.05	60.18	9.58	2069.36	44.90	78.45	25234.15	Total metals	[41]
Brullus	50.67	66.55	50.90	6.84	1703.47	35.41	58.14	5537.93	Mobile fractions	
Manzala	23.5	32.5	100	0.50	157.9	-	-	-	Total metals	[44]
Qarun	116.85	39.06	21.18	1.13	325.82	24.00	55.61	17860.00	Total metals	[43]
Nasser	56.10	42.62	49.75	0.54	914.99	-	66.21	28250	Total metals	[40]
Nasser	27.66	24.53	12.52	0.11	418.87	-	25.54	11610	Mobile fractions	
Worldwide										
Lake Chapala, Mexico	191.8	39.2	225.15	15.4	1262.0	-	61.1	22336	Total metals	[45]
	102.75	29.26	81.74	-	-	40.57	32.24	3970	Total metals	[36]
Avsar Lake, Turkey	-	26.73	3.24	0.76	-	-	29.12	24001	Total metals	[52]
Lake Taihu, China	87.32	31.12	33.05	-	-	22.49	29.81	-	Total metals	[46]
Lake Dianchi, China	153.95	90.05	65.76	-	-	33.36	45.97	-	Total metals	
Lake Karla, Greece	18.3	41.7	29.1			38.6	199		Total metals	[47]
Nansi Lake, China	110.51-235.36	38.09-78.65	24.51-53.95	0.08-1.12	-	4.12-20.14	11.30-65.4	-	Total metals	[48]
Lake Dautk, Uzbekistan	-	174	-	-	462	10.3	-	21300	Total metals	[49]
Yangtze lake, China	173	88	50	-	1303	22	46.1	48700	Total metals	[38]
lakes of Kumaun, India	40-149.2	13.4-32	88.9-167.4	11.1-14.6	90.1-197.5	-	17.7-45.9	5265-6428	Total metals	[50]
Maharlu Lake, Iran	52.1	61.3	135.4	4.7	364.5	54.9	135.2	19140.6	Total metals	[51]
	45.3	54.2	116.5	4.6	374.3	54.6	126.9	19300	Mobile fractions	

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Table (4)

Natural radionuclide activities in (Bq/Kg dry wt.) comparisons between Timsah Lake and other lakes in Egypt and worldwide

Lake Name	U-238 and daughters	Th-232 and daughters	k-40	Notes	Reference
<u>Egypt</u>					
Timsah Lake	19.53 (7.03 – 33.63)	19.12 (4.28 – 43.72)	179.75 (16.63 – 466.23)	Summer season	Present study
Timsah Lake	23.79 (8.05 – 56.46)	23.72 (7.32 – 44.54)	221.35 (33.45 – 459.89)	Winter season	
Mariout Lake	12.65 (10.52 – 15.91)	7.24 (5.44 – 8.33)	518.75 (441.64 – 582.31	Annual	[57]
Brullus Lake	17.26 (12.60 – 19.90)	10.03 (8.50 – 10.60)	299.7 (258.87 – 316.80)		
Brullus Lake	14.30 (10.30 – 21.80)	20.0 (11.90 – 34.40)	312.0 (268.0 – 401.0)	Annual	[58]
Idku Lake	20.37 (11.19 – 39.33)	26.05 (11.40 – 43.31)	329.05 (163.05 – 507.95)	Annual	[55]
Nasser Lake	14.3 – 22.0	18.4 – 24.4	222 - 326	Annual	[59]
Manzala Lake	13.78	12.53	217.74	Annual	[74]
Isamlia Canal, Egypt	89.0 (55.0 – 158.0)	19.0 (11.0 -31.0)	51.0 (34.0 – 87.0)	Annual	[60]
Industrial area, Port Said	18.03 – 398.66	5.28 – 75.7	237.88 - 583.12	Annual	[75]
Qarun Lake	20.37	14.18	244.68	Annual	[54]
Suez Canal	10.69	13.71	194.58	Annual	[56]
Worldwide					
Lake Dautk, Uzbekistan	1.8	3.58	-	Annual	[49]
Kainji Lake, Nigeria,	19.23 (4.64 -52.14)	31.59 (6.84 - 46.76)	84.12 (43.7 - 202.28)	Annual	[65]
Derbent Lake, Turkey	19.5	27.7	460	Annual	[61]
Deriner lakes, Turkey	15.8	13.9	551.5	Annual	[62]
Borcka Lake, Turkey	3.7	12.5	473.8	Annual	
Muratlı Lake, Turkey	14.4	30.0	491.7	Annual	
NE Tamilnadu, India	35.12	713.16	349.60	Annual	[66]
Moticher lake, India	6.4 (4.4-9.7)	15.6 (10.5–21.2)	160 (102–231)	Annual	[63]
Butrint Lagoon, Albania	13.0–26.6	13.1–38.1	266–675	Annual	[64]
Worldwide	35	30	400	Annual	[68]

Table (5) Correlation coefficients of the different measured parameters with each other at Timsah Lake:

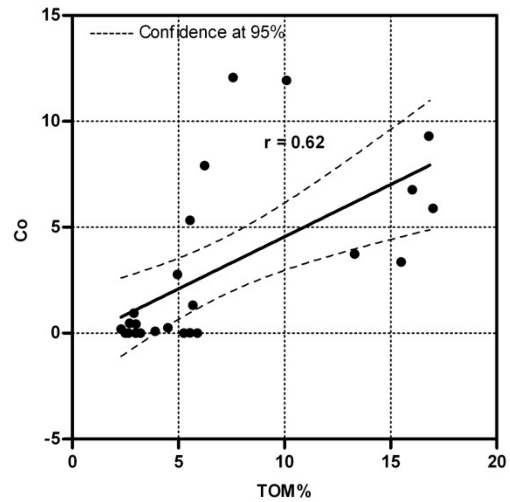
	CO3%	*TOM%*	*Zn*	*Cu*	*Pb*	*Cd*	*Mn*	*Co*	*Ni*	*Fe*	*U-238*	*Th-232*	*K-40*
CO3%	1.00												
TOM%	0.26	1.00											
Zn	-0.09	**0.75**	1.00										
Cu	0.18	**0.82**	**0.75**	1.00									
Pb	0.14	0.25	**0.51**	0.47	1.00								
Cd	0.03	-0.37	-0.38	-0.40	-0.04	1.00							
Mn	-0.03	**0.61**	**0.80**	**0.69**	**0.61**	**-0.61**	1.00						
Co	-0.16	**0.63**	**0.81**	**0.76**	**0.62**	**-0.50**	**0.96**	1.00					
Ni	0.11	**0.78**	**0.70**	**0.98**	0.36	-0.37	**0.65**	**0.74**	1.00				
Fe	0.02	**0.85**	**0.87**	**0.87**	0.47	**-0.60**	**0.87**	**0.88**	**0.85**	1.00			
U-238	0.00	**0.77**	**0.65**	**0.72**	0.30	**-0.53**	**0.70**	**0.76**	**0.69**	**0.76**	1.00		
Th-232	0.27	**0.86**	**0.64**	**0.77**	0.46	-0.30	**0.62**	**0.69**	**0.71**	**0.75**	**0.88**	1.00	
K-40	0.04	**0.54**	**0.86**	**0.69**	**0.60**	**-0.54**	**0.82**	**0.75**	**0.62**	**0.79**	**0.56**	0.47	1.00

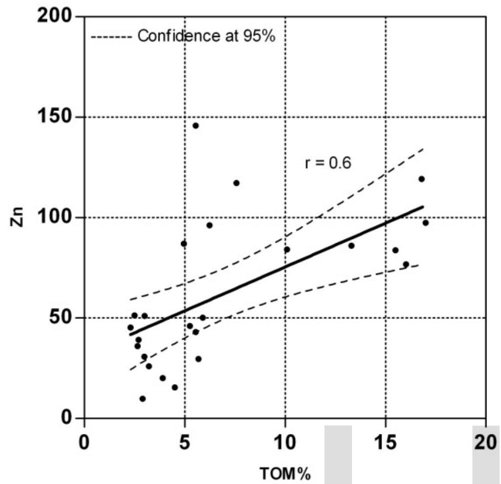
Transport models of metal migration accounted for only two forms: dissolved (mobile) form or a sorbed/precipitated (immobile) form. Metals that were not very soluble in water or readily sorbed to a solid form were considered to be largely immobile because they were effectively removed from the mobile phase. Many phases of heavy metals were found in the water column and may introduce to the geochemical cycle including; ionic form, suspended particulates, oxi-hydroxides and colloids. The radionuclide transport can take place in soluble form and in particulate form that are carried by surface runoff. The solid (particulate) form of radionuclide transport can be regarded to a flow of radionuclides sorbed on suspended sediments which were formed by surface forces and transported by overland water flow. In municipal sewage, the metallic contents are often absorbed on the sewage solids or sewage sludge. When the sludge is disposed off to lakes, the metallic contents are taken up by benthos in some amounts. These may have unpleasant effect on their tissues that may be rendered unsuitable for human consumption. In effect, the heavy metals are passed on to man through the food chain and the cumulative effects of these metals most of which are toxic. El Nemr [41] concluded that the industrial, agriculture and domestic sewage are the main sources of heavy metal contamination in Brullus Lake. Saeed and Shaker [42] attributed the high levels of Cd and Pb in sediments of Lake Manzala to the industrial and agriculture discharge as well as from spill of leaded petrol from fishing boats. At the industrial area of Port Said City – Egypt, Attia *et al.,* [75] attributed the recorded heavy metal levels especially of Cd and Zn to the phosphate fertilizers (Manmade wastes) that dumped to the site. Lerman *et al.,* [76] pointed out that the direct impact the high loads of pollutants could reach the lake margins, which controlled by diffusion, water mass movement resulting from the hydraulic flow (tidal effect) and lake circulation as induced by wind direction and velocity. Abd El Samie *et al.,* [26] added another factor, the direct effect of retention time and distribution of soluble elements that related to lake stratification.

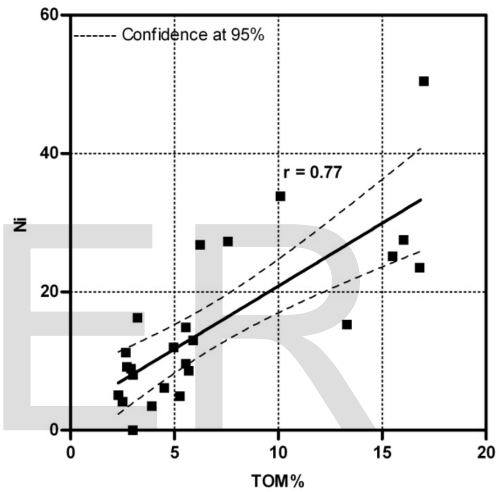
The migration of radionuclide from the water column to bottom sediments and vice-versa is a complex process involving the interaction between dissolved and solids phases of the contaminant and the sedimentation and re-suspension of particulate matter [77]. During re-suspension of benthic sediments some of the radionuclides are desorbed making them bio-available [78]. The process of interaction of dissolved radionuclides with solids particles in suspension or deposited is usually based on the notion of a reversible and rapid equilibrium between the dissolved and the adsorbed phases of the radionuclide. The equilibrium between the concentrations of the dissolved and the attached phases may be not instantaneously achieved and the adsorption–desorption processes are not always rapidly reversible [79], [80] and can occur in several stages [81]. The sinking of particulate matter in the coastal zone of the marine environment is a significant pathway for vertical transport of many anthropogenic and natural radionuclides. In the geochemical processes, beside of the sinking particles, the concentration of the radionuclides in the bottom sediments will be increased due to direct adsorption on the bottom sediments from dissolved phase of the radionuclides in the sea water [82]. Difference in the sediment mineralogy and pore-water geochemistry have a considerable effect on the potential for radionuclide remobilisation, both in the short term during active remediation and in the longer term due to passive infiltration. The fate of the released radionuclides will be strongly dependent on the chemical affinity to particulate matter in suspended loads and bottom sediments [29]. Radionuclide transfer between surface water and suspended sediments is described by the adsorption-desorption processes.

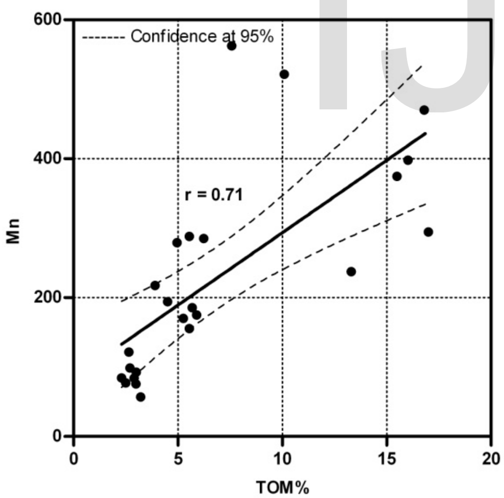
6- Statistical Analyses

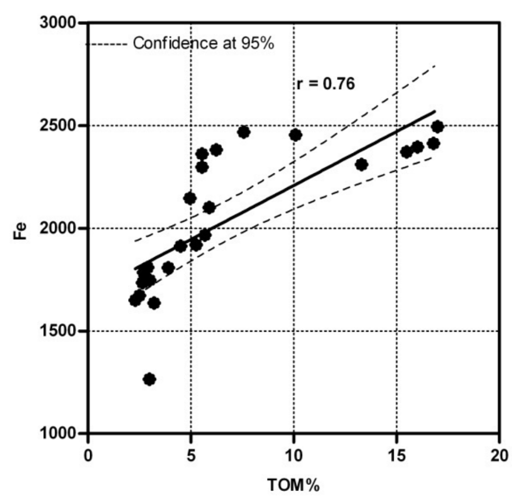
Total organic matter and Fe recorded significant positive correlations (Figs., 3 & 4) with heavy metals and radio elements (Table 5) indicating to two essential metal accumulation phases; one of them with organic matter in the highly reducing conditions as sulphides and the other associated and/or adsorbed by Fe-oxides and hydroxides. Organic matter combines with heavy metals, forming metal–organic complexes which are very stable. The deposition of fine sediments with high organic matter content produces anoxic environment with abundant sulfate which tend to become sulphides [83]. Moore and Sutherland [32] found a significant positive correlation between the heavy metals and organic matter in sediments. They added the concentrations of these metals and the radioactive elements; ^{238}U and ^{232}Th varied inconstantly in sediments and the low concentrations of radionuclides in seawater reflecting the strong chemical bonding characteristics of the sediment. An appreciation of the different forms of an element in environmental systems is therefore fundamental in assessing the availability of metals in soils to plants and animals, both as essential nutrients and as potentially toxic elements [71]. Yousry [40] pointed out that Fe-Mn oxides and organic matter seem to be the main carrier phases for the non-residual fractions of the heavy metals in the lake sediments. In the marine sediments, the non-residual forms of heavy metals may accumulate as adsorbed particulates, carbonates, sulphides, oxides and hydroxides according to the dominated oceanographic conditions. Sulphides are stable and very insoluble under reducing conditions, but oxidation takes place during re-suspension when minerals are exposed to high water maxing by winds, surge waves, atmospheric weathering or the metal sulfides oxidation is accelerated by the presence of oxidizing bacteria [84]. The slight decreasing in significance during winter season may indicate to the metal accumulations in other phases rather than the association with organic matter and Fe oxihydroxides as colloids. The positive correlation between the heavy metals and the radionuclides together indicate to the same sources of accumulations and/or they accumulated in the same phases under the same conditions (Figs., 5 & 6). The observed high uncertainties in the figures may due to the continuous active geochemical processes and the continuous changing from phase to another in Timsah Lake that may capture these radionuclides in the underlying sediments or evolve to the overlying water layer.











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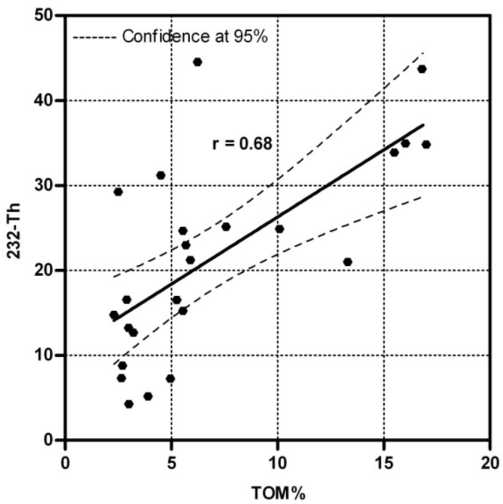


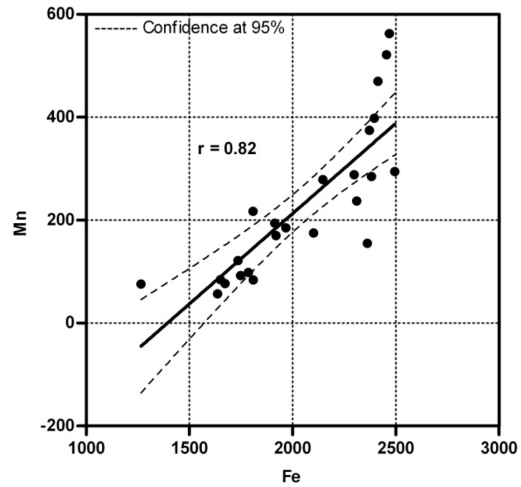
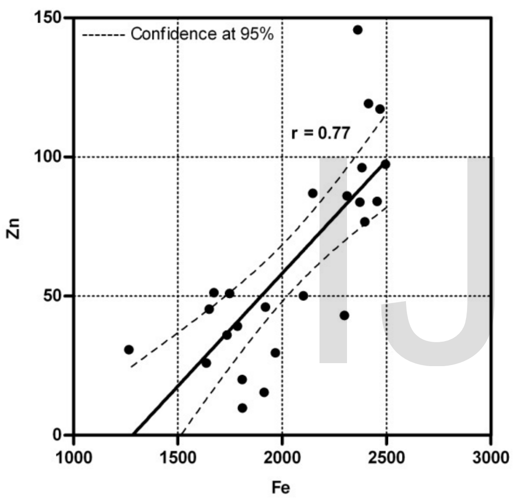
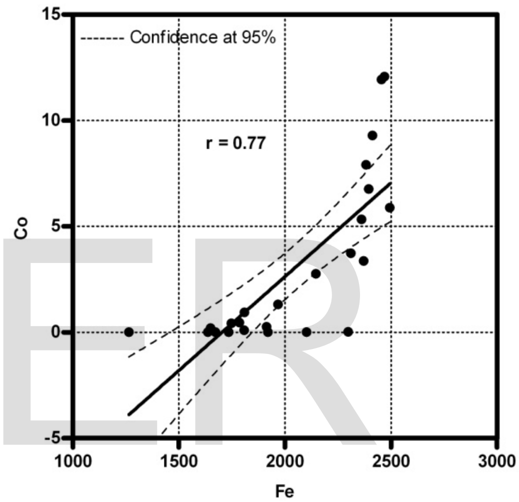
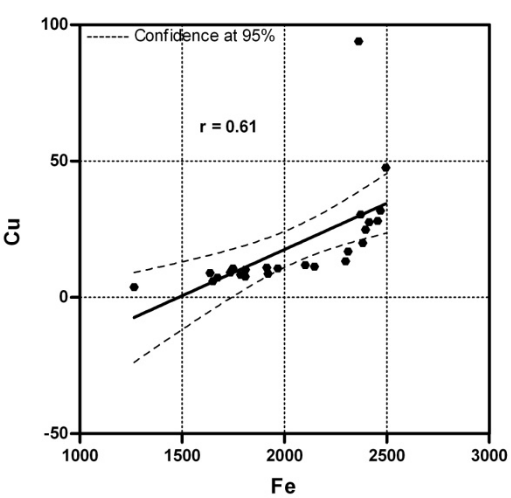
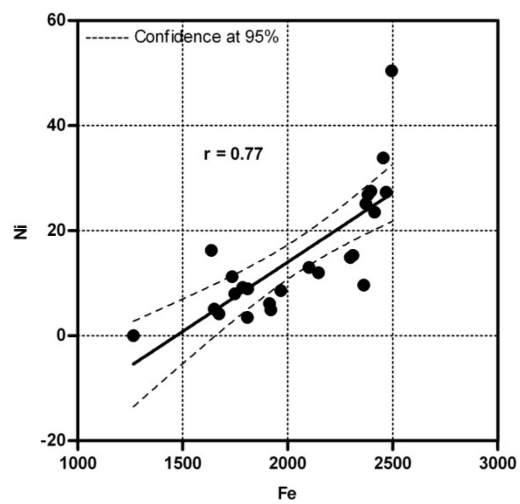


Fig., (3) The positive correlations of TOM% with; Zn, Mn, Co, Ni, Fe and ^{232}Th at 95% confidences.









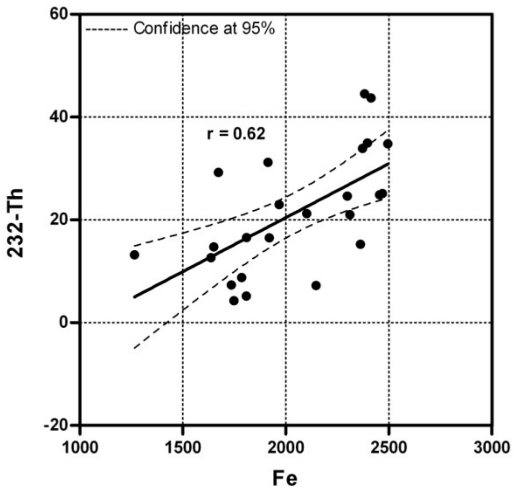
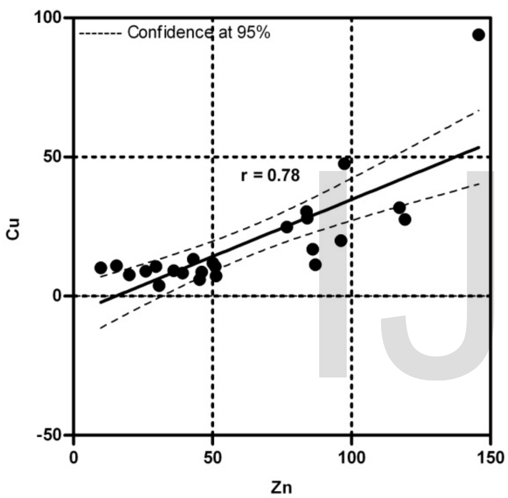


Fig., (4) The positive correlations of Fe with; Zn, Cu, Mn, Co, Ni and ^{232}Th at 95% confidences.



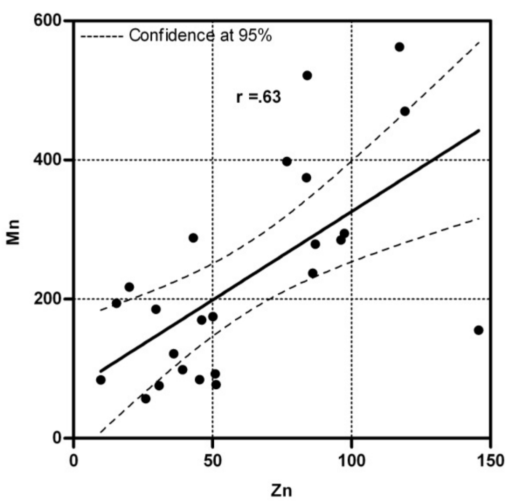
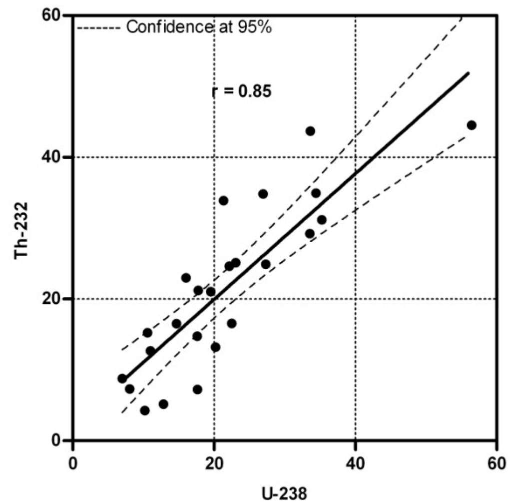


Fig., (5) The positive correlations of Zn with Cu and Mn at 95% confidences.



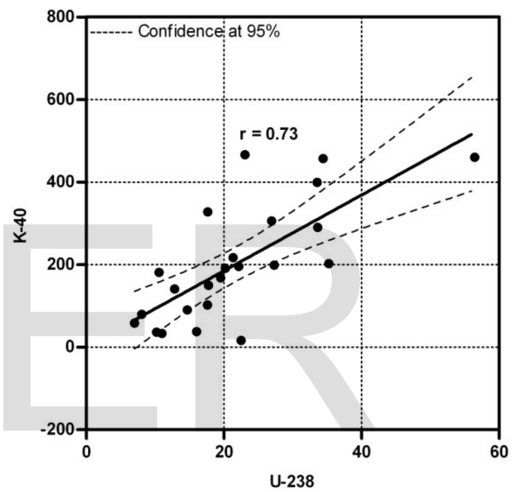


Fig., (6) The positive correlations of ^{238}U with ^{232}Th and ^{40}K at 95% confidences.

Conclusion

- Timsah Lake lies in the middle district of the Suez Canal, Egypt. It covers about 16km^2 with depth variation between 3 and 16 m and capacity volume of about 90 million cubic meters of seawater. It is also considered one of the most productive zones along the Suez Canal.
- From the western side, the lake is connected to a small and shallow embayment that receive about 833,000 m^3/day of treated and untreated domestic and agricultural wastewaters throughout many drains and from the northern side, the lake receives occasional freshwater inputs from the Ismailia Canal.
- The granulemetric analyses illustrated that the fine sediment fractions were higher in summer than in

winter affecting by the wind induced waves that disperse the fine particles in winter.

- Carbonate percentages show slight increasing in winter, while TOM was higher in summer than in winter in relation to coarse and fine sediments occurrence.
- Significant variations in Zn, Pb, Mn and Co contents between summer and winter that may attribute to the fine sediment particles dispersing during winter because of the extreme weather and hydraulic conditions can produce important changes in fluxes of matter and sedimentation rates.
- Slight variation between winter and summer in the natural radionuclides activities due to the high rates of untreated sewage inflow and/or active transformation from soluble phase to solid phase in winter and inverse process in summer.
- TOM and Fe recorded significant positive correlations with heavy metals and radio elements indicating to two essential metal accumulation phases, one of them with organic matter and the other with Fe-oxides and hydroxides in addition to many other independent phases. Carbonate percentages didn't show any significance correlations.

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