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Research Article
Geochemical Assessment of Pollution at Manzala Lake, Egypt: Special Mention to Environmental and Health Effects of Arsenic, Selenium, Tin and Antimony

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Abstract
Background and Objective: Manzala Lake is the largest shallower lakes which located in the Northern shoreline of the Nile delta. The water quality has a brackish, mixed with Mediterranean Sea water through Boughaz El-Gamil opening at Eastern rim of the lake and strait of Sheikh Ali at Western rim East of Damietta Governorates. It link with Suez Canal through a navigation aqueduct. A few study were carried related to study of arsenic, selenium, tin and antimony elements, so, the main objective of this study was assessment of environmental effects for these toxic elements. **Methodology:** Twenty five samples were collected to carry out this study covering the sources of pollution near discharging drains which consider the main source of pollution. Sediment was digested and the measurement was performed using simultaneous inductively coupled plasma emission spectrometer. **Results:** Arsenic content ranges from 4.6-22 ppm, averaging 12 ppm, about 8 folds the average earth's crust. Selenium concentrations range from 3-5 ppm averaging 4 ppm, about 80 fold the average earth's crust. Tin in studied lake ranged from 25-90 ppm with an average 46 ppm, about 9 fold the average earth's crust. The highest values for arsenic selenium and tin are extended toward the industrial area in Port Said Governorate. **Conclusion:** Statistically, pollution indicators were reflects high level of pollutants in the vicinity of the industrial activity around Port Said and Damietta governorates while agricultural pollution focused at El-Serw and Bahr El-Baqar drains. The unsupervised anthropogenic activities are the main causes of pollution in the lake sediments. Routine program for monitoring the concentrations and distribution of arsenic, selenium and tin in the lake water, fish, sediments and aqueous organisms should be imposed. The relative order of abundance of the toxic metals in the lake's water is As>Sn>Se.

Key words: El-Manzala Lake, bottom sediments, pollution, arsenic, selenium, tin, antimony

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Manzala Lake is existed in the North Eastern border of the Nile delta. It is connected by the Mediterranean Sea to the North, Port Said and Suez Canal to the East, El-Dakahlia and El-Sharkia Governorates on the South while Damietta Governorate on the West (Fig. 1). It is the largest shallower lakes which located in the Northern shoreline of the Nile delta and it located between latitudes (31°10" to 31°40" N) and longitudes (31°50" to 32°25" E). It has a depth ranging between 20 and 200 cm, it is covered with silty clay and sandy silty clay[1]. The Nile delta was formed during the Holocene period[2,3] discussed the evolutionary history of the Nile river and its valley and delta. The geology and origin of the study area, as a part of the Nile delta has been treated by Sneh and Weissbrod[4], Lotfy[5], El Askary and Frihy[6], Coutellier and Stanley[7], Stanley[8] and Hasaneen[9].

The water quality has a brackish quality, mixed with Mediterranean Sea water through Boughaz El-Gamil opening-West of Port Said-at the North Eastern rim of the Lake and strait of Sheikh Ali about 25 km East of Damietta governorates. It link with Suez Canal through a navigation aqueduct known as El-kabooty Canal[10]. These openings are equipping the lake with sea water and fishes. The Southern and Western Lake coast has many entrances by which great amounts of wastewater conduct into the lake. The famed drains are Bahr El-Baqar, Hadous, Ramsis, Al-Serw, Abu-Garida and Faraskur. Generally, the lake turned into a sewage reservoir due to the dumping of sewage where, especially in the regions of Hadous and Bahr El-Baqar. The pollution of Manzala Lake has been the objective of Abdel-Mouti[11], Abdel-Mout and Dowidar[12], El-Sabrouti[13], Hussein[14], Abdel-Satar[15], Dewidar and Khedr[16], Lotfy[17,18], Abdalla[19] and El-Badry[20]. The environmental health and the functioning of ecosystems are widely recognized at the Manzala Lake discussed for the first time by Orabi and Osman[21].

A few studies were carried related to arsenic, selenium, tin and antimony elements, so, the main objective of this study is assessment of environmental and health effects for that toxic elements through the Egyptian standards and the world-wide organizations. Attention for pollution risk and face the rapid deterioration of this important limnology ecosystem.

MATERIALS AND METHODS

The study was carried out on Manzala Lake through summer, 2014 to monitor and evaluate the levels of arsenic, selenium, tin and antimony in sediment. Twenty five sediment samples were collected by Ekman grab. The stations were selected mainly to covering the sources of pollution near from the discharges of most drains which consider the main source of pollution of the Manzala Lake. The selected sampling stations are presented in Fig. 1.

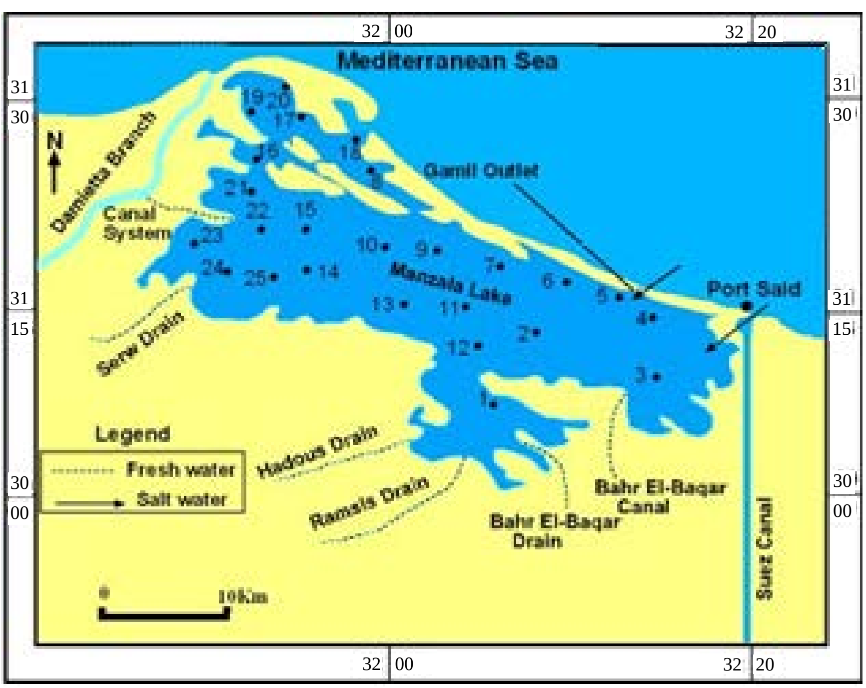

Fig. 1: Location map of Manzala Lake showing the sampled sites

For determination trace metal, the sediment was digested using an acid method[22]. Where 1 g powder of dried sample is weighed into teflon crucible and digested by using mixture from (HNO_3, $HClO_4$ and HF), after near dryness the residue was dissolved with HCl and diluted to 100 mL with distilled water. The measurement of As, Se, Sn and Sb was performed using simultaneous inductively coupled plasma emission spectrometer (720 ICP-OES), Agilent Technologies. Samples were introduced via glass concentric nebulizer fitted to glass cyclonic spray chamber (single pass). An independent three-channel peristaltic pump was used for pumping the sample. High solid torch standard (axial 2.4 mm id injector) was used. Statistical analysis was done for all samples to compute the pollution indicators by using SPSS program.

Pollution sources: Nine major drains and channels drained Agricultural wastewater to Manzala Lake. Bahr El-Baqar drain considers mainly and badly polluted drain influx into the lake. Main pollutants in agricultural drains derived from industrial sources, fertilizer, pesticide materials and domestic wastewater.

The industrial pollution influenced among by two sources, firstly multiple industrial activities in Port Said region (East side of the lake) in several areas, most notably the field of textile manufacturing and garment manufacturing as well as chemical manufacturing and the manufacture of leather and leather products. This is unlike the areas of food and food products, manufacture of industrial detergents, electrical appliances and household items manufacturing. Also, in the areas of petroleum service activities, scrapping, maintenance of ships, marine equipment, marine services in addition to the conservation, cooling, mobilize food, the area of screening and grading of agricultural crops as well as the storage of goods re-exported out of the country. The second industrial pollution sources derived from the areas of activity in Damietta Governorate (West side of the lake) is petrochemical, oil services, gas liquefaction, medical supplies, a collection of computers and electronic appliances, textile and garment, food, activity maritime transport, marine services, refrigerators for keeping food intended for export outside the country, furniture and wood products industry, processing and sorting of grains. Also, it is famous for its fishing and textile industry, shoes, candy, canned sardines and shrimp also famous for its dairy industry.

RESULTS AND DISCUSSION

Geochemical backgrounds: The comparison between the measurement of As, Se, Sn and Sb data and the average earth's crust according to McLennan and Taylor[23] is set in Table 1. The comparison suggests that the average concentrations of As, Se, Sn and Sb in the studied sediments (12, 4 and 46 ppm and none detected for antimony) are about 8, 80 and 9 fold the average earth's crust, respectively, while antimony content is depleted. Comparing with the agency for toxic substances and disease registry, which setup the sediment quality guidelines in the form of level of probable effects, the studied sediments seem to be polluted at high levels.

Heavy metals in lake sediments: The following is a summarized study on the environmental status of studied metals that may cause some risk or endanger on human health and environmental effects beside the geochemical distribution maps of toxic metals. The toxic metals content of the investigated sediments are listed in Table 2.

Arsenic: According to Tantry *et al.*[28] arsenic is consider as one of the most toxic elements that can be found. In spite of their toxic effect, inorganic arsenic bonds occur on earth naturally in small amounts. Humans risk to arsenic through food, water and air. Exposure may also occur by means of skin contact with soil or water that contains arsenic. Arsenic concentration in food is low, but it may be high in fish and seafood due to absorbing arsenic from the water they live in. The fish that comprise considerable levels of inorganic arsenic possibly jeopardy to human health. Arsenic exposure may be higher for people who live on farmlands where arsenic-containing pesticides have been applied in the past. Also, the exposure to inorganic arsenic can cause different health effects, such as irritation of the stomach and intestines, decreased production of red and white blood cells, skin changes and lung irritation.

Table 1: Comparison between concentrations of As, Se and Sn metals in the present study and some previous studies of Manzala Lake

References	As	Se	Sn
Threshold limit value	15	1	30.0
Earth crust McLennan and Taylor[23]	1.5	50 ppb	5.5
Ghani[24]	1.9	1.5	173.2
Agency for toxic substances and disease registry[25-27]	60 ppb	50 ppb	6.9
Present study	12	4	46.0

Average concentration (ppm)

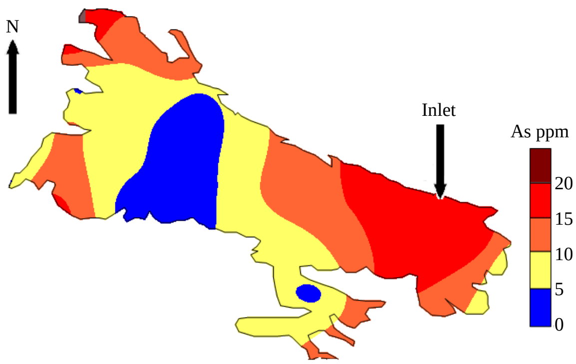

Fig. 2: Geochemical map of arsenic in the studied bottom lake sediment

Table 2: Concentrations of As, Sb, Se and Sn elements in bottom sediments of Manzala Lake

Stations No.	As	Sb	Se	Sn
1	15	Nd	Nd	33
2	16	Nd	Nd	34
3	4	Nd	Nd	32
4	5	Nd	3	50
5	20	Nd	180	45
6	18	Nd	Nd	41
7	15	Nd	2	35
8	2	Nd	Nd	31
9	12	Nd	4	30
10	5	Nd	Nd	27
11	8	Nd	Nd	37
12	4	Nd	2	40
13	Nd	Nd	10	40
14	6	Nd	Nd	32
15	4	Nd	Nd	25
16	12	Nd	Nd	90
17	14	Nd	Nd	51
18	22	Nd	3	55
19	17	Nd	5	50
20	11	Nd	Nd	31
21	10	Nd	Nd	44
22	4	Nd	Nd	45
23	22	Nd	3	55
24	10	Nd	Nd	34
25	8	Nd	Nd	35
Average	12	Nd	4	46
Maximum	22	Nd	5	90
Minimum	4	Nd	3	25
Detection limit	15 ppb	25 ppb	25 ppb	20 ppb

Nd: Undetected

It is suggested that the uptake of considerable amounts of inorganic arsenic can exalt the probability of cancer development, especially the chances of development of skin, lung, liver and lymphatic cancer. A very high exposure to inorganic arsenic can cause infertility and miscarriages with women and it can cause skin disturbances, declined resistance to infections, heart disruptions and brain damage with both men and women. Finally, inorganic arsenic can damage DNA. About 100 mg from arsenic oxide consider as A deadly dosage.

Arsenic is mainly released by the copper industries, lead and zinc production and in agriculture. It cannot be get rid of the moment that it has step inside the environment, so the amounts that drained can spread and cause health effects to humans and animals on many locations around studied Lake. Plants soak up arsenic easily, so that high-dose supposedly present in food. The concentrations in Lake water increase the chances of alteration of genetic character of fish. This is due to accumulation of arsenic in the cells of phytoplankton-feeding freshwater organisms. Arsenic content predominantly derived from geological sources and its consumption can cause chronic health. Arsenic found in groundwater is considered as the largest environmental health disaster that goal to threaten people in the lake suburb. Consumption and exposure of arsenic has been affecting human health with carcinoma disease and other related sickness[29]. Arsenic is found in earth's crust with concentration of 1.8 ppm. In soils, mostly the inorganic forms of arsenic are found such as arsenite (Arsenic trivalent) and arsenate (Arsenic pentavalent)[30,31].

In the present study, arsenic content ranges from 4.6-22 ppm, with an average of 12 ppm. The lowest value is recorded at middle section of the lake while the highest values are recorded near El-Serw agricultural drain, industrial area in Damietta region and also, at South Eastern area of the Lake where the group of drains Bahr El-Baqar, Hadous and Ramsis drain, extended toward the industrial area in Port Said Governorate (Fig. 2). The MPL of As in the worldwide soils is 1.5 ppm. According to the agency for toxic substances and disease registry, a high level is 60 parts of arsenic per billion.

Selenium (Se): The highest concentrations of Se in the bottom sediments of the Manzala Lake were observed in the Eastern portion of the lake where industrial zone of Port Said Governorate. The extensive uses of selenium which consider as good photovoltaic and photoconductive properties, in electronics, glass industry and use for animal feeds and food supplements. Other uses of selenium are in metal alloys such as the lead plates used in storage batteries and in rectifiers to convert AC current in DC current. Selenium is used to improve the abrasion resistance in vulcanized rubbers. Some selenium compounds are added to anti-dandruff shampoos.

Selenium can reached to soils or an aquatic environment through weathering of rocks, aeolian deposition, fluvial input, suspension load and biological effect. It is getting in the air by means of coal and oil combustion as selenium dioxide. There is evidence selenium can accumulate in the body tissues of

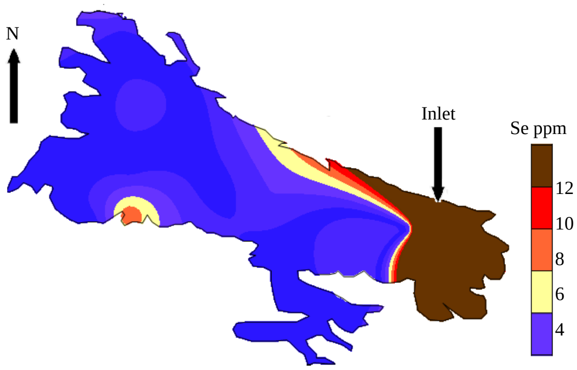

Fig. 3: Geochemical map of selenium in the studied bottom lake sediment

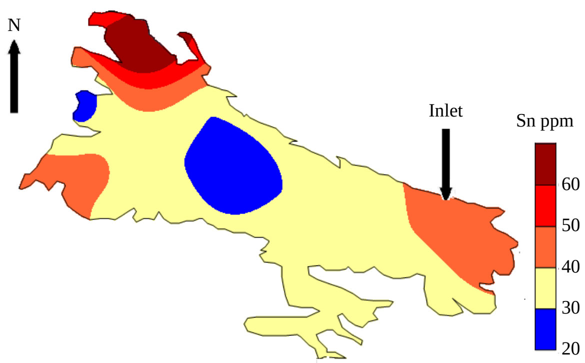

Fig. 4: Geochemical map of tin in the studied bottom lake sediment

organisms and can then be passed up through the food chain, when animals eat a plants which irrigated by a high concentrations of selenium content. Concentrations of selenium tend to be very high in aquatic organisms in many areas.

The health effects of selenium have been the objective of Hu *et al.*[32], Duffield-Lillico *et al.*[33], Stranges *et al.*[34], Loef *et al.*[35] and Davis[36]. Humans undergo to it through food or water or by contact with soil or air. The exposure to selenium mainly happens by feeding, because it is surely existent in grains and meat. Humans need to a little amounts of selenium every day, food generally include suitable contents enough to prevent disease caused by shortages. In the present study, selenium ranges from 3-5 ppm, averaging 4 ppm. The lowest value is recorded at middle and Western area of the lake, the median values are recorded near El-Serw agricultural drain, while industrial area in Port Said Governorate and Eastern area of the lake represent the highest values where Bahr El-Baqar and Hadous drain the highest content extended toward the West of inlet (Fig. 3). Selenium may be higher than usual in agricultural land, because fertilizers have been applied on farmland. It will drained to the lake water through irrigation from hazardous waste-drains and from surrounding cultivated land. People that live near suburbs of Manzala Lake will undergo to a higher exposure through soil and air, also by eating a lot of grains that grow near hazardous sites may experience a higher exposure through food. The MPL of Se in the worldwide soils is 50 ppb. According to the Agency for Toxic Substances and Disease Registry, a high level also, is 50 parts of arsenic per billion.

Tin: The depress concentration of tin in soils is due to insoluble of tin oxide and strongly resistant for weathering process, also it is protected by an oxide film. Tin is used in coating of plated steel containers which is used for food conservation. Tin alloys are used in welder for gathering metallic pipes and dental amalgams. The tin oxide is used for ceramics and in gas sensors.

Although, the organic tin is dangerous forms for humans, we used it in a great number of industries, such as the paint, plastic industries, also, in agricultural pesticides. The health effects of tin have been the objective of Alessio and Dell'Orto[37], Abou-Arab *et al.*[38], Amouroux *et al.*[39], Graf[40], Arambarri *et al.*[41], Batt[42] and Tabrez *et al.*[43]. The effects of organic tin substances can vary. They depend upon the kind of substance that is present and the organism that is exposed to it. Triethyltin is the most dangerous organic tin substance for humans. Food and breathing through the skin considered as the main way for humans absorption tin.

Tin is hard absorbed by animals and humans. The huge use of tin in canned food tends to be the low toxicity[40]. Nausea, vomiting and diarrhea have been reported after ingesting tinned food containing 200 ppm of tin while un-lacquered tin cans with food, fruits and vegetables can contain high concentrations of tin[44]. The Food Standards Agency in the United Kingdom propose upper limits of 200 ppm Food Standards Agency[27]. Organic tins can propagate through the water systems when adsorbed on sediment particles. They are known to cause a great degree of harm to aquatic ecosystems, as they are very toxic to fungus, algae and phytoplankton which is a very important relate with the aquatic ecosystem, where it supply water organisms with oxygen. It is consider also an important part of the aquatic food chain. The accumulation of organic tin compounds exposure lay in the top layer of the water. Tin (in surficial bottom sediment of Manzala Lake) ranged from 25-90 ppm with an average 46 ppm. Tin was seen least at middle portion of the lake and was seen highest in the three sites near Damietta industrial region, near El-Serw drain and near of industrial area at Port Said Governorate (Fig. 4).

Table 3: Standards of pollution indicators in sediment

Enrichment Factor (EF)	Pollution type	Contamination Factor (CF)	Pollution type	Geoaccumulation index CF Igeo	Pollution type
<1	Without anthropogenic sources	<1	Low	<0	Unpolluted
>1	With anthropogenic sources	1-3	Moderate	0-1	Unpolluted to moderate
		3-6	Considerable	1-2	Moderate
		>6	Very high	2-3	Moderate to strong
				3-4	Strong
				4-5	Strong to extremely strong
				>5	Extreme

Standard based on the EF defined by Rule[45], Rubio *et al.*[46], CF defined by Hokanson[47] and Igeo values defined by Muller[48]

Antimony: Antimony and its compounds were known to the ancient's time. In Egypt antimony sulfide (Sb_2S_3) which occurs naturally as the mineral stibnite is used in black form as a pigment and in mascara user. Antimony makes up about 0.00002% of the earth's crust. In the present study, antimony content depleted to undetected limit.

Pollution indicators: Enrichment Factor (EF), Regional Pollution Index (RPI), Contamination Factor (CF) and geoaccumulation index (Igeo) were used as pollutant indicators, reflecting a relative ranking of sampling stations. Table 3 shows the standards for these pollution indicators.

Enrichment Factor (EF): The EF is computed using the following relationship:

$$\text{Enrichment Factor (EF)} = \frac{(\text{M/Fe})_{\text{sample}}}{(\text{M/ Fe})_{\text{crust}}}$$

where, $(\text{M/Fe})_{\text{sample}}$ is the ratio of metal and iron concentration of the sample (Fe values measured according to El-Badry[20] and $(\text{M/Fe})_{\text{crust}}$ is the ratio of metal and iron in crust[49,50].

The enrichment factors of studied toxic metals in the Lake sediments were listed in Table 4. The values of enrichment factors for arsenic, selenium and tin detected that the pollution type sidetracked with anthropogenic sources, while the minimum and undetected value was observed for selenium in 16 stations in the Western area of the lake.

Regional Pollution Index (RPI): The Maximum Permissible Limits (MPL) of an element is considered as the "Pollution standard level or goal". The index of an individual pollutant is calculated as follows:

$$\text{Regional Pollution Index (RPI)} = \frac{\text{Pollution concentration}}{\text{Pollutant standard level}} \times 50$$

The Maximum Permissible Limits (MPL) used for the worldwide soil used as pollutant standard level. For each region, the RPI is classified into low, medium and high pollution, as follows: From 0-24 low pollution from 25-49 medium pollution-greater than 50 of high pollution.

The RPI of the studied lake bottom sediments is listed in Table 4. It can be concluded that the studied area is exposed to high pollution levels by Se, Sn and As.

Contamination factor: Individual contamination factor is an efficient tool for estimation of pollution during a period of time and determination of the pollution status of single substances:

$$CF = M_x/M_b$$

where, M_x is the measured concentration and M_b is geochemical background concentration of the element concentration of the target metal.

The calculated of contamination factor are listed in Table 4. Very high contamination was recorded at all stations for tin except stations 9, 10 and 15 which showed moderate contamination factor. Selenium showed variable contamination factor in lake varied from not detected to high and very high contamination especially which recorded at stations 5. For arsenic, moderate to considerable contaminations factor at 10 stations 3, 4, 8, 10, 11, 12, 14, 15, 22 and station number 25, while high contamination factor was recorded at other stations of the lake.

Geoaccumulation index (Igeo): With a view to identify the grade of pollution in each sample, the geoaccumulation index (Igeo) values were calculated using the following arithmetical formularization:

$$\text{Igeo} = \log_2 C_n/1.5\ B_n$$

where, C_n is concentration of the target metal and B_n is the concentration of the metal in the selected reference background[51-53].

The calculated geoaccumulation Table 4 indicates that this has to be considered as unpolluted to moderately with respect to arsenic, selenium and tin for the lake sediments.

Table 4: Calculated Enrichment Factor (EF), Contamination Factor (CF) and geoaccumulation index (I_{geo}) of bottom sediments samples of Manzala Lake

	EF			RPI			CF			Igeo		
Stations	AS	SE	SN	AS	SE	SN	AS	SE	SN	AS	SE	SN
1	7.7	Nd	4.6	500	Nd	300	10.0	Nd	6	0.82	Nd	0.60
2	8.1	Nd	4.7	533	Nd	309	10.7	Nd	6	0.85	Nd	0.62
3	2.0	Nd	4.3	133	Nd	291	2.7	Nd	6	0.25	Nd	0.59
4	2.3	4.1	6.2	167	300	455	3.3	6	9	0.35	0.60	0.78
5	8.0	215.2	4.9	667	18000	409	13.3	360	8	0.95	2.38	0.74
6	6.2	Nd	3.9	600	Nd	373	12.0	Nd	7	0.90	Nd	0.70
7	5.2	2.1	3.3	500	200	318	10.0	4	6	0.82	0.43	0.63
8	0.7	Nd	2.9	67	Nd	282	1.3	Nd	6	0.05	Nd	0.57
9	4.0	4.0	2.7	400	400	273	8.0	8	5	0.73	0.73	0.56
10	1.5	Nd	2.2	167	Nd	245	3.3	Nd	5	0.35	Nd	0.51
11	2.4	Nd	3.0	267	Nd	336	5.3	Nd	7	0.55	Nd	0.65
12	1.2	1.8	3.2	133	200	364	2.7	4	7	0.25	0.43	0.69
13	Nd	8.7	3.2	Nd	1000	364	Nd	20	7	Nd	1.12	0.69
14	1.7	Nd	2.5	200	Nd	291	4.0	Nd	6	0.43	Nd	0.59
15	1.1	Nd	1.8	133	Nd	227	2.7	Nd	5	0.25	Nd	0.48
16	3.1	Nd	6.4	400	Nd	818	8.0	Nd	16	0.73	Nd	1.04
17	3.6	Nd	3.6	467	Nd	464	9.3	Nd	9	0.79	Nd	0.79
18	5.6	2.3	3.8	733	300	500	14.7	6	10	0.99	0.60	0.82
19	4.2	3.7	3.4	567	500	455	11.3	10	9	0.88	0.82	0.78
20	2.7	Nd	2.1	367	Nd	282	7.3	Nd	6	0.69	Nd	0.57
21	2.4	Nd	2.9	333	Nd	400	6.7	Nd	8	0.65	Nd	0.73
22	0.9	Nd	2.9	133	Nd	409	2.7	Nd	8	0.25	Nd	0.74
23	5.1	2.1	3.5	733	300	500	14.7	6	10	0.99	0.60	0.82
24	2.3	Nd	2.1	333	Nd	309	6.7	Nd	6	0.65	Nd	0.62
25	1.8	Nd	2.2	267	Nd	318	5.3	Nd	6	0.55	Nd	0.63
Average	2.9	2.7	3.1	389	367	414	7.8	7	8	0.71	0.69	0.74
Maximum	5.0	3.4	5.6	733	500	818	14.7	10	16	0.99	0.82	1.04
Minimum	1.1	2.5	1.9	133	300	227	2.7	6	5	0.25	0.60	0.48

Nd: Undetected

CONCLUSION

Manzala Lake located at Northern of Egyptian delta receives a greet quantity of wastewater (Agricultural, industrial, municipal and domestic), in addition to navigation and fisheries activities. The highest concentrations of As, Se and Sn in the bottom sediments of the Manzala Lake were observed in the Eastern portion of the lake where industrial zone of Port Said country and at the Western portion of the lake where industrial zone of Damietta region, while the lowest values were recorded at the middle portion of the Lake. The relative order of abundance of the potentially toxic metals in the lake's water is As>Sn>Se. The present study is based on chemical analysis data on the bottom sediments of the Manzala Lake, relative to average earth's crust, the lake sediments are markedly enriched in As, Sn and Se. The calculated pollution index reflects high level of pollutants. Generally, the distribution of heavy metals in Manzala Lake revealed that the industrial districts represented by Port Said and Damietta Governorates, agricultural drains represented by El-Serw, Hadous and Bahr El-Baqar drains beside the fisheries activity considered the main pollution sources.

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