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# Research Article Geochemical Assessments and Distribution of Arsenic, Selenium, Tin and Antimony in the Surfacial Bottom Sediments of Brullus Lagoon and its Effects on Human Health

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## Abstract

**Background and Objective:** Brullus Lagoon is the biggest North Egyptian lake. The lagoon has an average area of 420 km<sup>2</sup>. It is sited between two Nile branches, brackish and shallower water depth. The pollution of bottom sediment of the Brullus lagoon is indicative of both water and food web quality in general. The main objective of this study was the assessment of environmental effects of arsenic, selenium, tin and antimony metals. **Methodology:** Twenty one samples were collected from bottom sediments sample among sites covering the Brullus Lagoon during summer 2014. After using the digestion technique the chemical analyses was employed to analyze Se, Sn, Sb and As using simultaneous inductively coupled plasma emission spectrometer (720 ICP-OES). **Results:** Results showed that the mean concentrations of As, Se, Sn and Sb greatly exceed (2, 156, 12 and 25 times) the average shale standard. The mean concentrations of As, Se, Sn and Sb greatly exceed (2, 156, 12 and 25 times) the average shale standard. The mean concentrations of As, Se, Sn and Sb greatly exceed to pollution index for heavy metal shows that the metals contents are above the critical level that indicate that all studied metals posed moderate to high-risk assessment to surrounding ecosystem in short or medium-term, anthropogenic activities count as the main reason for pollution in the lagoon. Such pollution is resulting from industrial activities and fertilized agricultural drains. Routine program for monitoring the abundance and distribution of sensitive and essential environmental metals in the studied lagoon should be enjoined.

Key words: Brullus lagoon, bottom sediments, pollution, arsenic, selenium, tin, antimony, Egypt

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Competing Interest: The authors have declared that no competing interest exists.

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

#### INTRODUCTION

In Egypt, there are four natural water bodies known as (Maryut, Edko, Brullus and Manzala). These lakes (Fig. 1), extend along the North Egyptian coast. This study highlights Brullus Lagoon, which is considered as a semi-closed environment of brackish water. The lagoon has an area of about 420 km<sup>2</sup> of which 370 km<sup>2</sup> are open water, the rest of the area represents several islands within the water lake. It is the shallower depth ranges from centimeters in near shore to 2.5 m. The lagoon is located between latitudes 31°25' and 31°35' N and longitudes 30°31 and 31°05 E. It is about 53 km long, with about 13 km width. The lagoon is connecting with the Mediterranean Sea through Boughaz El-Brullus at its Northeastern side<sup>1</sup>.

During the Holocene period the Nile Delta was formed. Rosetta and Damietta branches at present-day replaced instead of the seven distributaries channels that have subsequently silted up. These distributaries channels are consider as the only source of sediments, which formed the Delta. Said<sup>2</sup> discussed the evolutionary history of the River Nile and its Delta reporting five stages in the development of the Nile; with each stage being characterized by a master river system.

The Southern coastal lagoon has many entrances by which great amounts of wastewater conduct into the lagoon, which considers the main sources of pollution. The famed Drains are Brullus drain (East); El-Gharbia drain, Nasser drain, drain 7, drain 8, drain 9, drain 11, Brimbal Canal and Brullus drain (West). So, the lagoon well become as sewage reservoir, due to the dumping of wastewater, especially in the regions of El-Gharbia drain and drain 7. The poisonous metals can be introduced into lagoon from industrial and agricultural operations. Siegel<sup>3</sup> reported the various uses of toxic metals in industry. The wastes enter to aquatic environment as a result of industrial processes. El-Gharbia drain is considered as the main source for industrial wastes of the textile factories, also, electroplating, synthetic fiber production and others of El-Mahalla El-Kobra and Kafr El-Zayat cities.

The objectives of present study were as follows:

- To analyze the bottom sediments of Brullus Lagoon micro toxic nutrients (Se, Sn, Sb and As)
- To assess the distribution of arsenic, selenium, tin and antimony in the surfacial bottom sediments of Brullus Lagoon in Egypt
- To study and geochemical mapping and spatial variability of studied toxic metals across selected sampling stations of Brullus Lagoon
- To give accurate estimation of pollution level in this lagoon in order to avoid the effects of risk on human health
- To assess the spatial variability of micronutrients at the basic process level using geo-statistical analysis

#### **MATERIALS AND METHODS**

The study was carried out on Brullus Lagoon during one day in summer 2014, to monitor and evaluate the levels of arsenic, selenium, tin and antimony in sediment. The samples were collected from twenty one sites covering the sources of pollution near from the discharges of most drains which consider the main source of pollution of the Brullus Lagoon. The selected sampling stations are shown in Fig. 2.



Fig. 1: Map of the River Nile Delta showing the four coastal lagoons

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Fig. 2: Key map of Brullus Lagoon showing the location of the sampling sites

Sediment for toxic metal determination was digest using an acid method described by Pozebon *et al.*<sup>4</sup>, where 1 g powder of dried sample was digest into Teflon crucible by using mixture from HNO<sub>3</sub>, HClO<sub>4</sub> and HF. After near dryness HCl dissolved the residue and diluted to 100 mL with de-ionized 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.

#### **RESULTS AND DISCUSSION**

**Geochemical backgrounds:** The comparison between the average Earth's continental crust according to McLennan and Taylor<sup>5</sup>, average shale according to McLennan and Murray<sup>6</sup> and the geochemical evolution of the continental crust according to Taylor and McLennan<sup>7</sup> and the measurement of As, Se, Sn and Sb data is listed in Table 1. The comparison shows that the average contents of As, Se, Sn and Sb in the surfacial bottom lagoon sediments (25, 94, 74 and 38 ppm) are about 2, 156, 12 and 25 fold the average shale, respectively. According to the agency for toxic substances and disease registry, the studied sediments show high levels of pollution (Table 2).

**Toxic metals in Brullus Lagoon sediments:** The results of analyses of the toxic metals content of the investigated sediment samples are listed in Table 3. Among the 4 elements studied; concentrations of Se and Sn were higher, whereas lower concentrations of Sb and As were observed in different sampling locations. This study summarizes the environmental states of studied toxic metals that may cause risk on human health and environmental effects beside the geochemical distribution maps of toxic metals.

Arsenic: Arsenic represents extremely toxic metal, it is the 52nd out of 92 elements that is heavily find in earth's crust and has a concentration of 1.8 ppm. Arsenic encompasses two predominant forms (arsenite and arsenate) of which arsenite is more toxic. Arsenic pollution is mainly caused by natural processes as industrial activities and widespread anthropogenic use of pesticides containing arsenic lead to contamination of soil and water<sup>8</sup>. The WHO<sup>9</sup> guideline permits the maximum limit of arsenic as 10  $\mu$ g L<sup>-1</sup> in drinking water, while the maximum permissible limit of arsenic in the worldwide soils is 1.5 ppm, while a high level is 60 ppb according to the Agency for Toxic Substances and Disease Registry<sup>10</sup>. Arsenic can enter human body by drinking contaminated water and can cause cancer in skin, bladder, lung, liver and kidney IARC<sup>11</sup>. Arsenical compounds form can damage and abnormalities in chromosomes<sup>12</sup>. In the present work, arsenic content ranges from 10-44 ppm, with an

Table 1: Comparison between concentrations of As, Se, Sn and Sb metals in the present study of Brullus Lagoon and some previous studies in selected rocks

References	As	Se	Sn	Sb
McLennan and Taylor <sup>5</sup> (Earth crust)	1.5	50*	5.5	0.002
McLennan and Murray <sup>6</sup> (Shale)	13	0.6	6	1.5
McLennan and Murray <sup>6</sup> (Ocean clay)	13	0.17	1.5	1
McLennan and Murray <sup>6</sup> (Deep sea carbonate)	1	0.17	0	0.15
Taylor and McLennan <sup>7</sup> (Streams)	2*	0.06*	0.04	0.07
Agency for Toxic Substances and Disease Registry <sup>10,18,30</sup>	60*	50*	6.9	
Present study	25	94	74	38

\* Concentrations average by ppb, concentrations average by (ppm) unless otherwise noted

Table 2: Standards of pollution indicators in sediment based on the CF defined by Hakanson<sup>31</sup> and Igeo values defined by Muller<sup>32</sup>

Standards of	pollution	indicators

CF	Pollution type	lgeo	Pollution type
< 1	Low	<0	Unpolluted
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

Table 3: Concentrations of A	s, Sb, Se and Sn i	metals in the bottom	Brullus Lagoon sediments
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Sampling stations	As (ppm)	Sb (ppm)	Se (ppm)	Sn (ppm)
1	23	N.d	250	140
2	21	N.d	250	130
3	12	100	290	150
4	13	N.d	210	120
5	18	N.d	N.d	40
6	24	2	N.d	60
7	24	N.d	N.d	42
8	20	N.d	N.d	54
9	20	N.d	N.d	41
10	18	N.d	N.d	46
11	43	N.d	15	80
12	24	N.d	N.d	46
13	36	N.d	3	40
14	39	N.d	N. d	40
15	30	N.d	18	65
16	44	N.d	11	70
17	16	N.d	N.d	41
18	44	N.d	6	70
19	30	N.d	10	65
20	10	13	37	155
21	25	N.d	22	80
Average	25	38	94	74
Maximum	44	100	290	155
Minimum	10	2	3	40

N.d: Not detected

average of 25 ppm. The lowest value was recorded at the Northern area of the lagoon while the highest values were recorded adjacent to the agricultural and industrial drains toward the Southern area of the lagoon where (Fig. 3).

**Tin:** The effect of tin on human health studied by Boniewska Bernacka *et al.*<sup>13</sup> focused on the effect of tin on

selected living cells, while Clarkson<sup>14</sup> mentioned the effect of metal toxicity in the central nervous system. The effect of tin dusts on lung injury in experimental animals was observed by Jiang *et al.*<sup>15</sup>. Neurotoxic diseases and metal ions affecting the hematological system were studied by Liu *et al.*<sup>16</sup> and Roney *et al.*<sup>17</sup>. In the present study, tin concentration ranges from 40-155 ppm, with an average of 74 ppm. The lowest



Fig. 3: Geochemical map of arsenic metal in the studied bottom lagoon sediment



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

value was recorded at the Western area of the lagoon while the highest values were recorded toward industrial region toward Northern area of the lagoon (Fig. 4).

**Antimony:** In ancients' Egyptian time antimony was used in mascara and as a pigment in black form. Antimony makes up about 0.00002% of the Earth's crust. Antimony was found at very low levels in the environment. According to ATSDR<sup>18</sup>, EPA has set a limit of 145 ppb in lakes and streams to protect human health from the harmful effects of antimony taken in through water and contaminated fish and shellfish. EPA<sup>19</sup> has also set limits on the amount of antimony that industry can release. Antimony enters the environment during the production of antimony metal, alloys, antimony oxide and combinations of antimony with other substances. Most antimony ends up in the soil or sediment, where it attaches strongly to particles that contain iron, manganese, or aluminum. The concentration of antimony that dissolve in rivers and lakes is very low; usually less than 5 ppb. Antimony

does not appear to accumulate in fish and other aquatic animals. Although antimony was used as solder for water pipes, it does not seem to get into the drinking water. Exposure to antimony for a long time can irritate eyes, skin and lungs. Breathing of antimony for a long time can cause problems with the lungs, heart, stomach pain, diarrhea, vomiting and stomach ulcers.

In the present study, antimony content depleted to undetected limit at about 92% of the studied area. Its content ranges from 2-38 ppm, averaging 100 ppm. The lowest value and undetected limit was observed at most area of the lagoon while the highest values were recorded toward Northern area of the lagoon around the inlet (Fig. 5), where fossil fuel combustions, paint and pigments, addition to corrosion and cohesion processed for fishing boat.

**Selenium (Se):** The health effect of selenium as a toxic element has been discussed by many authors<sup>20-26</sup>. The protective role of selenium in cancer patients was studied by



Fig. 5: Geochemical map of antimony metal in the studied bottom lagoon sediment



Fig. 6: Geochemical map of selenium metal in the studied bottom lagoon sediment

Hu *et al.*<sup>27</sup>, also, Sieja and Talerczyk<sup>28</sup> studied the effect of selenium as an element in the treatment of cancer. A change of selenium levels and activities in blood was discussed by Vernie *et al.*<sup>29</sup>. Kesse-Guyot *et al.*<sup>26</sup> studied the daily supplementation with vitamins and minerals at nutritional doses. In the present study, selenium ranges from 3-290 ppm, averaging 94 ppm. About 45% represent undetected selenium content. The lowest value is recorded at Western area of the lagoon, while industrial area in Baltim at Eastern area of the lake represents the highest values extended toward of the inlet (Fig. 6). The MPL of Se in the worldwide soils is 50 ppb. According to the ATSDR<sup>30</sup>, a high level also, is 50 parts of arsenic per billion.

**Pollution indicators:** Contamination Factor (CF) and geoaccumulation index (Igeo) were used as pollutant indicators, reflecting a relative ranking of sampling stations. Table 4 shows the standards for these pollution indicators.

**Contamination factor:** The Contamination Factor (CF) expresses the level of contamination of sediments. The Eq. 1, by Hakanson<sup>31</sup>, classifies the contamination levels into four groups (Table 2).

$$CF = M_x / M_b \tag{1}$$

where,  $M_x$  is the measured concentration and  $M_b$  is geochemical background concentration of the element concentration of the target metal. The CF values of As, Sb, Se and Sn show moderate contamination factor (Table 4). It ranged between undetected values at most sites for Sb and Sn and very high CF at Eastern portion of the lagoon. For Sn, CF is higher than 6 (very high CF) in the all part of the Lagoon. CF of As varied from 0.77 to 3.38 (low to considerable CF).

**Geoaccumulation index (Igeo):** The geoaccumulation index (Igeo) reveals the level of contamination of sediments. The Eq. 2 according to Muller<sup>32</sup>:

	Geoaccumulation index			Contamination factors				
Sampling stations	As	Sb	Se	Sn	As	Sb	Se	Sn
1	0.07	N.d	2.44	1.19	1.77	N.d	416.67	23.33
2	0.03	N.d	2.44	1.16	1.62	N.d	416.67	21.67
3	-0.21	1.65	2.51	1.22	0.92	66.67	483.33	25.00
4	-0.18	N.d	2.37	1.12	1.00	N.d	350	20.00
5	-0.03	N.d	N.d	0.65	1.38	N.d	N.d	6.67
6	0.09	-0.05	N.d	0.82	1.85	1.33	N.d	10.00
7	0.09	N.d	N.d	0.67	1.85	N.d	N.d	7.00
8	0.01	N.d	N.d	0.78	1.54	N.d	N.d	9.00
9	0.01	N.d	N.d	0.66	1.54	N.d	N.d	6.83
10	-0.03	N.d	N.d	0.71	1.38	N.d	N.d	7.67
11	0.34	N.d	N.d	0.95	3.31	N.d	N.d	13.33
12	0.09	N.d	N.d	0.71	1.85	N.d	N.d	7.67
13	0.27	N.d	N.d	0.65	2.77	N.d	N.d	6.67
14	0.3	N.d	N.d	0.65	3.00	N.d	N.d	6.67
15	0.19	N.d	1.3	0.86	2.31	N.d	30	10.83
16	0.35	N.d	1.09	0.89	3.38	N.d	18.33	11.67
17	-0.09	N.d	0.52	0.66	1.23	N.d	N.d	6.83
18	0.35	N.d	0.82	0.89	3.38	N.d	10	11.67
19	0.19	N.d	1.05	0.86	2.31	N.d	16.67	10.83
20	-0.29	0.76	1.61	1.24	0.77	8.67	61.67	25.83
21	0.11	N.d	1.39	0.95	1.92	N.d	36.67	13.33
Average	0.12	0.39	1.78	0.92	1.96	3.67	91.19	12.50
Maximum	0.35	1.65	2.51	1.24	3.38	66.67	483.33	25.83
Minimum	-0.29	-1.95	0.52	0.65	0.77	0.02	5	6.67

Table 4: Contamination factor and geoaccumulation index of surfacial bottom sediments samples of Brullus Lagoon

N.d: Not detected

$$Igeo = log_2 C_n / 1.5 B_n$$
 (2)

where, C<sub>n</sub> is concentration of the target metal and B<sub>n</sub> is the concentration of the metal in the selected reference background value of the element n according to McLennan and Taylor<sup>5</sup>. A constant of 1.5 is used due to a given metal fluctuations in the soils as well as some very small anthropogenic influences<sup>33</sup>. The results of Igeo are shown in Table 4. The negative values of As and according to Muller<sup>32</sup> classification it indicated that the lagoon is unpolluted to moderate and moderate polluted with those metals. From the geo-accumulation index results, Igeo values of Sb showed unpolluted revealed to undetected value at most sites of the lagoon. The Igeo results of Se and Sn showed moderate to strong polluted category in all locations.

### CONCLUSION AND FUTURE RECOMMENDATIONS

The high level of pollution is investigated around the inlet and Southern area of Brullus Lagoon. According to pollution indicator contamination factors and enrichment factors, the lagoon sediments hold markedly unpolluted. The geoaccumulation index for Se and Sn showed moderate to strong polluted, contamination factors of as varied from low to considerable value. The relative order of abundance of the potentially toxic metals in the lagoon's sediment is Se> Sn> Sb> As.

From the analysis of sediment samples in the Brullus Lagoon the following most important points can be extracted:

- Brullus Lagoon receives great quantity of industrial and agricultural wastewater from the surrounding area
- The high levels of industrial pollutants investigated conclude around the inlet while agricultural pollution showed adjacent to group of drains toward Southern area of the lagoon
- Construction of special units for treatment and purification of all types of drainage and wastewater (agricultural, industrial)
- Successive analysis of lagoon water is important to assess the amount of pollutants to make suitable decisions. Take an action to prohibit throwing of wastes in the lagoon. Consumption of the lagoon water for agricultural and industrial must be under control in order to decrease water pollution

#### SIGNIFICANCE STATEMENT

This study throws more light on distribution, geochemical assessments, environmental effects and estimate the degree of risk to surrounding ecosystem through research investigated of arsenic, selenium, tin and antimony metals. This study discovers the serious effect of toxic metals on human health around the inlet of Brullus Lagoon and the area adjacent to Southern drains, where the average concentrations of arsenic, selenium, tin and antimony are 25, 94, 74 and 38 ppm, about 2, 156, 12 and 25 fold the average shale.

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