

Organotin Compounds in Sediments of Northern Lakes, Egypt

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Abstract

This pioneered study is aimed to determine the levels and distributions of organotin compounds (OTCs) in sediment samples collected from five Egyptian lakes located in the southern-eastern Mediterranean Sea. The paper has given an account of and the reasons for the widespread distribution of Tributyltin (TBT) in northern lakes. The percentage of TBT ranged from 4% - 100% OTC revealing recent inputs of TBT. The highest average of TBT (2.84 µg/g Sn dry wt) was measured in lake Edku. Dibutyltin (DBT) was observed at most sampled sites. Relatively higher abundance of DBT was observed in sediments of EL-Burullus (0.69 µg/g Sn dry wt). One of most significant findings to emerge from this study is the absence of any strong and significant correlation between TBT and DBT in sediments from five northern lakes. The absence of such correlation might give a clear evidence that both compounds did not come from the same source. The observed OTC levels indicate some highly localized areas of contamination which are severe enough to cause harmful effects on marine flora and fauna. Except for El-Burllus, Triphenyltin (TPhT) was not measured in sediments of most stations. TPhT was the predominant species in El-Burullus, which indicate a source of pollution mostly originated from industrial and agricultural waste water discharge, organic wastes, commercial fertilizers, chemical wastes and pesticides. The percentages of OTC to total tin ranged from 1% to 35%. El-Brullus, Edku and El-Bardaweel exhibited the highest percentages. As a result of lack of enforcement on the control of OTC, the study has thrown up many questions need further detailed environmental assessment of OTC.

Keywords

Organotin Compounds, Sediments, Lakes, Egypt

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1. Introduction

Tin compounds are sparingly soluble in water and are likely to partition to soils and sediments. Most commercially used OTCs are relatively immobile in environment media due to their low vapor pressures, low water solubility and high affinity for soil and organic sediments [1]. Inorganic Sn cannot be degraded in the environment, but may undergo oxidation-reduction, ligand exchange and precipitation reactions [2]. Degradation of organotin compounds in sediments is much slower than in water, and half lives have been estimated to be several years [3].

In spite of legislative regulations for OTC, it is likely that OTC will continue to be produced and used as effective biocides, especially in developing countries. They continue to be used also in material and wood preservatives as effective biocides [4]. TBT has been considered the most hazardous compound to marine organisms and it is perhaps the most toxic chemical that has ever been introduced to marine environment [5] [6]. The great stability of the TBT adsorbed onto sediments shows the long-term storage capacity of this compartment [7]. It is well known that sediment is not a storage place but serves as a renewable source for OTC, especially TBT [8] [9]. Triphenyltin (TPhT) compounds metabolize to diphenyltin, mono-phenyltin and non-extractable bound residues.

DBT has mainly been related to the degradation of TBT via microbial activity and/or photochemical reactions. In recent years evidence for a direct entrance of DBT into the environment was found due to municipal wastewaters, sewage sludge and land fill leachates. For example, the major application of DBT is as additives to plastics particularly to polyvinyl chloride (PVC) to prevent degradation due to changes in temperature or lighting. PVC is used in water pipes rigid sheeting (e.g. for roofing) and bottles [10]. DBT compounds are characterized by higher water solubility than TBT, which probably gives rise to higher concentrations in groundwater and freshwater.

TPhT compounds have been used extensively as algacide and molluscicides in antifouling products since 1960s [11]. TPhT is a co-toxicant in some long-term antifouling paints [12] and is similar with TBT in toxicity [13]. The persistence of the TPhT in soil is affected by a range of soil parameters including microbial activity and sequestration, moisture content and soil type affecting adsorption and hence bioavailability [14]. It is well known that TPhT is strongly adsorbed to sediments and soil, with no desorption [15]-[17]. TPhT compounds are widely used in agriculture as fungicides. The use of TPhT has not been as strictly regulated as TBT but it is acknowledged to produce similar levels of toxicity [18].

Despite the presence of few studies that have been conducted to investigate the levels and distribution of OTC in the south eastern Mediterranean Sea [15] [19]-[21], this study is pioneered to determine the levels and distributions of OTC in sediments of Egyptian lakes.

The Egyptian Mediterranean coast exhibits five lakes (**Figure 1**), Mariout, Edku, El-Burullus, El-Manzallah and El-Bardaweel. All of them, with the exception of Lake Mariout are directly connected to the Mediterranean Sea through several straits. The northern lakes are economically the most important fishing ground. They are providing a rich and vital habitat for estuarine and marine fish and their regeneration. Northern lakes have always been major areas of fish production in the south eastern Mediterranean Sea, since more than 75% of the Egyptian lakes productions are harvesting from them. They are also important sites for wintering water birds providing valuable habitats for several hundred thousand of birds. Unfortunately many challenges are facing these lakes *i.e.* degradation and decreasing the lakes area in addition to pollution problems such as increasing the levels of heavy metals, pesticides, etc. in water, sediments and biota which are increasing due to the expansion of human activities, fish farming activities and aquatic plants.

The aim of this study is to determine the levels and distributions of organotin compounds (OTCs) in sediment samples collected from these Egyptian lakes located in the southern-eastern Mediterranean Sea.

2. Materials and Methods

Fifty four water and sediment samples were collected during summer 2009 (**Figure 1**) from Lakes Mariout (n = 10), Edku (n = 9), El-Burullus (n = 12), El-Manzallah (n = 11), and El-Bardaweel (n = 12). Water samples were collected in duplicate 10 cm below water surface at each station, using a purified PVC Niskin's bottle (5 L). Water temperature, salinity and pH were measured during the time of sampling using Hydrolab model (Mutliti Set 430i WTW) after previous calibration.

Transparency was measured using 25 cm diameter enameled Secchi disk. Total suspended matter was

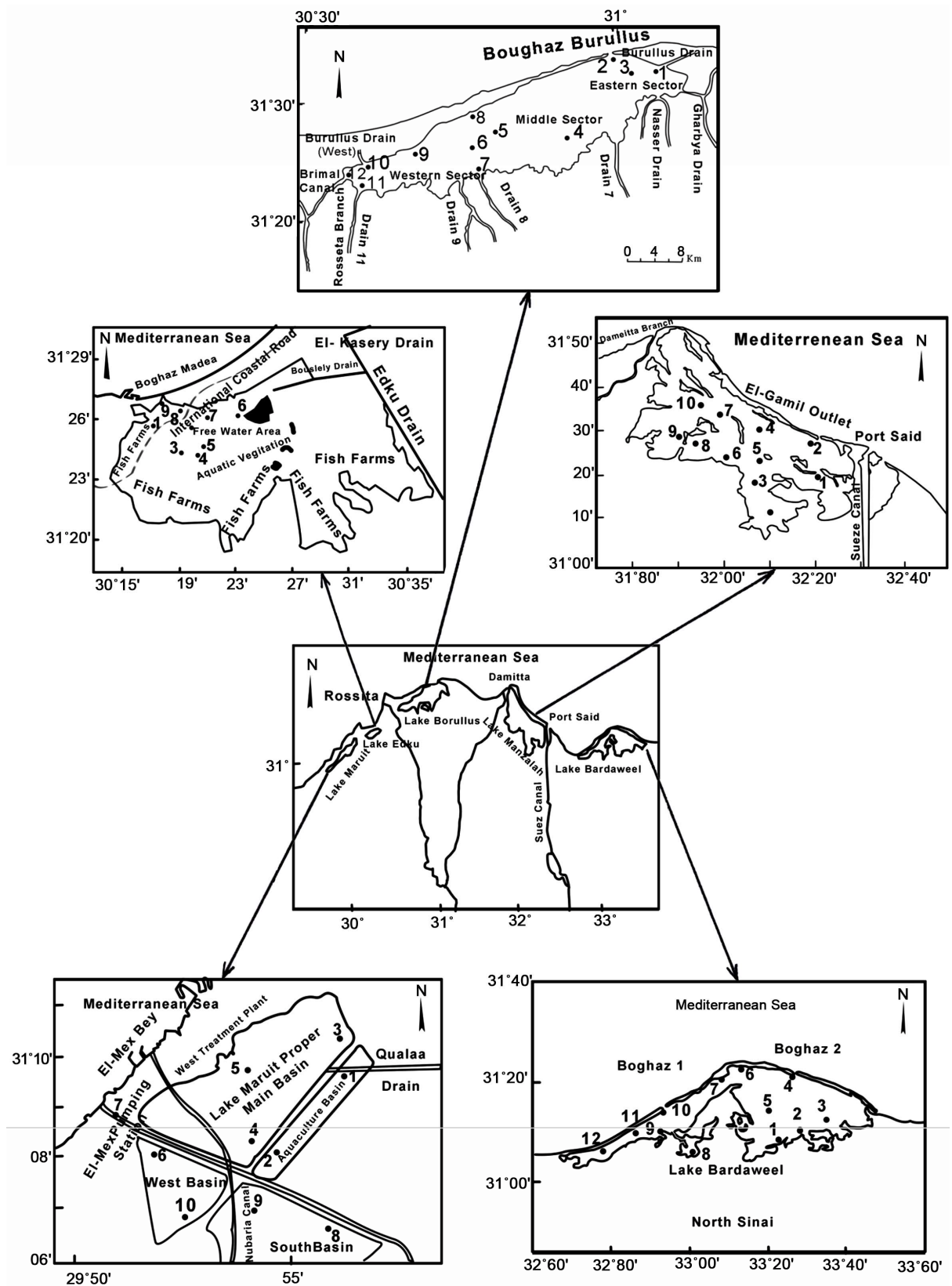


Figure 1. Map showing the positions of sampling stations of the five Egyptian northern lakes.

obtained from one liter water sample then filtered through washed, dried and weighed 0.45 µm membrane filters. The filters with its contents were kept for drying in the oven at 105°C until constant weight. The difference in the dry weight of filters before and after filtration was expressed in mg/l suspended matter [22]. Dissolved oxygen (DO) was determined at the same day of collection using the classical Winkler method [23]. Biological Oxygen Demand (BOD) was measured using 5-day method as DO and oxidizable organic matter (OOM) was measured according to FAO [24]. Sediment samples were collected using a Van-Veen grab sampler. Locations were selected taking into consideration the expected affected areas by industrial and human activities. Samples were divided into two sub-samples, one for determination of OTC, placed in polypropylene bottles and the second one for determination of total tin, placed in pre-cleaned plastic bags. Sediment samples were kept frozen at -20°C until analysis. Once in the laboratory the sediment samples were freeze-dried by using Labconco (England) freeze-drier, ground with agate mortar and stored at room temperature. For total tin analysis, an exact weight (0.5 g) of a dry sediment sample was completely digested in Teflon vessels using a mixture of HNO₃, HF and HClO₄ (3:2:1) at 70°C [25]. The final solution was diluted to 25 ml with double deionized distilled water. All digested solutions were analyzed in duplicate using an atomic absorption spectrometer (AAS Shimadzu Model AA 6800 equipped with a Graphite furnace atomizer GFA-EX7) and the results were expressed in µg/g dry wt. Conditions for GFA-EX7 obtained for tin applying AAS instrument were as follows: The absorption wavelength was 286.3 nm, Slit width was 0.5, ash temp was 400°C, Atomizing temp was 2500°C, Graphite tube was pyrolytic and detection limit was 2 ng/g. All reagents used were of analytical grade (Merck). For quality control (QC) and quality assurance (QA), replicates were analyzed under same procedures mentioned above. A standard reference material (IAEA-356, marine sediment; Analytical Quality Control Services, Austria) was also digested and analyzed similarly to ensure the quality control and accuracy of the analysis (Table 1). Sn pollution levels in the collected sediments were measured using the Index of Geo-accumulation (Igeo) which consists of six grades (Tables 2(a)-(e)). The highest grade reflects 100-fold metal concentration relative to background values: $I_{geo} = \log_2 [C_n / (1.5 \times B_n)]$ Where C_n is the measured concentration of element n in sediment sample, and B_n is the concentration of element n. A factor of 1.5 is used because of possible variations in background data due to lithogenic effects [26]. OTC were determined as follows according to Tsuda *et al.*, [27].

3. Results and Discussion

3.1. Physicochemical Characteristics

Depths, water temperature, transparency, electrical conductivity, salinity, pH, DO, BOD, and OOM for each lake are listed in Tables 2(a)-(e). Depths varied between 0.25 m measured at station 4 of lake El-Burullus and 4.5 m measured at station 9 of lake Edku, revealing the shallowness of the area. Water temperature varied in a narrow range depending on the mean of daily number of sunshine hours and the time of sampling. The minimum value of 27.0°C was observed at station 1 of Lake El-Bardaweel, while the maximum value 31.2°C was measured at station 9 of Lake El-Manzalah. The water transparency is generally low due to the shallowness of these lakes and the continuous disturbance of the mud bottom by wind actions [28]. Transparency reached maximum value (200 cm) at stations 10 & 11 of Lake El-Bardaweel, while the minimum value (12 cm) was measured at station 1 of Lake El-Bardaweel revealing its turbid water. Transparency reached maximum value (200 cm) at Lake El-Bardaweel, while the minimum value (12 cm) was measured at Lake El-Bardaweel revealing its turbid water. The discharge of drainage water from different drains into the lakes, the invasion of sea water through the lake sea connection, the rates of evaporation and the rain water during winter are the main factors that affect the salinity of different lake water [29]-[32]. Water salinity varied in a very wide range. The maximum value of 67.7‰ was measured at station 12 of Lake El-Bardaweel. Worth mentioning that the hypersaline conditions which

Table 1. Results of validation study for Tin (Sn) concentration in reference material (IAEA-356) analyzed together with sediments of the study area.

Element	LOD	Certified value	Found	SD	Recovery%	95% confidence interval
Sn	0.3 ppb	52.5 ppm	50.1 ppm	3.700	95.4	46.9 - 54.3

Mean concentration of certified value associated with the mean concentration (found) at 95% confidence interval of Sn in sediments (n = 6). SD: Standarddeviation; LOD: Limit of detection.

Table 2. (a) Physicochemical characteristics and concentrations of T-Sn, DBT, TBT and TPhT in lake Mariout during summer 2009; (b) Physicochemical characteristics and concentrations of T-Sn, DBT, TBT and TPhT in lake Edku during summer 2009; (c) Physicochemical characteristics and concentrations of T-Sn, DBT, TBT and TPhT in Lake El-Brullus during summer 2009; (d) Physicochemical characteristics and concentrations of T-Sn, DBT, TBT and TPhT in Lake El-Manzallah during summer 2009; (e) Physicochemical characteristics and concentrations of T-Sn, DBT, TBT and TPhT in lake El Bardaweel during summer 2009.

(a)

Physicochemical characteristics of water										Organotin compounds in sediments							
Stations	Depth (m) cm	Trans (cm) cm	Temp (°C) °C	EC μ S/cm μ S/cm	Sal‰	pH	DO mg/l	BOD mg/l	OOM mgO ₂ /l	T-Sn μ g/g	DBT μ g/g	TBT μ g/g	TPhT μ g/g	Σ OT	TBT/DBT	Σ OT/T-Sn%	Igeo
1	1.0	25	29.3	11.32	6.5	8.28	4.0	15.6	23.6	19.98	Nd	1.04	Nd	1.04	-	5	2.5
2	1.5	25	28.9	12.92	7.5	8.38	5.6	16.9	28.8	21.96	0.44	0.84	0.34	1.62	1.91	7	2.7
3	2.0	35	29.6	2.38	1.1	7.26	0.0	41.5	63.6	19.71	Nd	0.14	Nd	0.14	-	1	2.5
4	1.8	42	27.8	5.18	2.8	7.86	2.4	31.2	55.4	15.57	0.21	0.88	Nd	1.09	4.19	7	2.2
5	2.1	15	28.0	3.29	1.6	7.86	2.6	16.5	42.2	16.65	Nd	2.14	Nd	2.14	-	13	2.3
6	3.4	35	28.1	8.69	4.9	8.64	10.0	16.5	22.1	26.55	1.74	1.72	0.22	3.68	0.99	14	2.9
7	1.5	60	27.8	5.65	3.1	7.66	2.6	15.7	24.3	22.05	Nd	0.87	Nd	0.87	-	4	2.7
8	0.7	40	29.1	4.68	2.5	7.92	3.5	12.6	26.4	19.26	0.05	0.29	Nd	0.34	5.80	2	2.5
9	1.5	100	27.8	8.02	4.5	7.95	6.4	18.6	28.3	15.48	2.11	2.29	0.63	5.03	1.09	32	2.2
10	0.8	20	30.2	8.70	4.9	8.37	13.0	16.8	28.9	22.51	1.78	1.25	0.39	3.42	0.70	15	2.7
Max.	3.4	100	30.2	12.9	7.5	8.64	13.0	41.5	63.6	26.55	2.11	2.29	0.63	5.03	5.80	32	2.9
Min.	0.7	15	27.8	2.38	1.1	7.26	0.0	12.6	22.1	15.48	Nd	0.14	Nd	0.14	0.70	1	2.2
Average	1.6	40.0	28.7	7.8	3.9	8.02	5.01	20.2	34.4	19.97	1.06	1.15	0.40	2.61	1.08	13	2.5
SD	0.8	24.8	0.9	3.4	2.4	0.4	3.9	9.0	14.5	3.5	0.92	0.7	0.2	1.6	2.1	9.2	0.2

(b)

Stations	Depth (m) cm	Tras (cm) cm	Temp (°C) °C	EC μ S/cm μ S/cm	Sal‰	pH	DO mg/l	BOD mg/l	OOM mgO ₂ /l mg/l	T-Sn μ g/g	DBT μ g/g	TBT μ g/g	TPhT μ g/g	Σ OT	TBT/DBT	Σ OT/T-Sn%	Igeo
1	1.5	30	27.2	3.58	1.8	8.57	8.0	6.2	13.9	16.7	2.04	2.63	0.81	5.48	1.29	33	2.3
2	0.9	30	27.7	2.41	1.1	8.56	7.6	7.2	10.2	22.68	1.31	2.41	Nd	3.72	1.84	16	2.7
3	1.0	32	28.7	2.33	1.1	8.75	8.2	7.6	10.3	24.41	1.3	4.88	Nd	6.18	3.75	25	2.8
4	2.0	40	28.1	2.52	1.2	8.71	8.5	6.4	9.6	18.36	2.02	4.03	Nd	6.05	2.00	33	2.4
5	2.1	50	28.1	2.28	1.0	8.32	8.5	6.2	10.6	17.46	0.91	1.34	0.68	2.93	1.47	17	2.3
6	1.8	45	27.9	1.53	0.6	7.68	2.7	8.5	16.8	12.06	0.66	3.4	Nd	4.06	5.15	34	1.8
7	1.8	45	28.5	1.61	0.6	8.32	9.3	6.4	14.6	23.94	2.81	3.98	Nd	6.79	1.42	28	2.8
8	0.7	35	28.0	2.03	0.9	8.44	7.0	6.4	10.6	17.28	0.64	1.27	0.77	2.68	1.98	16	2.3
9	4.5	30	29.4	2.01	0.9	8.87	18.2	17.2	22.1	18.99	0.67	1.65	0.74	3.06	2.46	16	2.5
Max.	0.7	50	29.4	3.58	1.8	8.87	18.2	17.2	22.1	24.41	2.81	4.88	0.81	6.79	5.15	34	2.8
Min.	4.5	30	27.2	1.53	0.6	7.68	2.7	6.2	9.6	12.06	0.64	1.27	Nd	2.93	1.29	16	1.8
Average	2.0	38.0	28.2	2.26	1.0	8.47	8.67	8.0	13.2	19.09	1.37	2.84	0.33	4.54	2.07	24	2.4
SD	1.1	7.8	0.6	0.60	0.4	0.4	4.0	3.5	4.2	4.0	0.8	1.3	0.1	1.6	1.3	8.1	0.3

(c)

Stations	Depth (m) cm	Trans (cm) cm	Temp (°C) °C	EC μS/cm μS/cm	Sal‰	pH	DO mg/l	BOD mg/l	OOM mgO ₂ /l mg/l	T-Sn μg/g	DBT μg/g	TBT μg/g	TPhT μg/g	ΣOT	TBT/ DBT	ΣOT/T-Sn%	Igeo
1	0.50	30	29.7	4.68	2.5	8.26	7.2	5.6	25.5	22.05	1.46	1.26	Nd	2.72	0.86	12	2.7
2	3.0	40	30.5	8.36	4.7	8.35	7.6	5.4	23.4	14.4	2.21	0.21	2.07	4.49	0.10	31	2.1
3	1.3	35	29.3	5.28	2.8	8.94	8.9	7.6	20.6	21.96	2.29	0.16	2.11	4.56	0.07	21	2.7
4	2.5	20	30.3	4.60	2.4	8.77	6.6	5.0	23.5	31.59	1.64	0.48	1.29	3.41	0.29	11	3.2
5	1.5	30	28.9	3.80	1.9	8.75	7.2	6.0	20.6	22.32	0.35	0.73	1.51	2.59	2.09	12	2.7
6	1.2	18	29.0	3.30	1.6	8.44	6.6	5.6	17.6	32.22	1.49	1	1.23	3.72	0.67	12	3.2
7	1.2	30	29.9	3.57	1.8	8.30	7.6	5.6	21.8	22.5	2.85	1.35	3.33	7.53	0.47	33	2.7
8	1.0	30	29.0	2.78	1.3	8.74	7.2	6.5	22.5	25.83	2.34	0.32	3.83	6.49	0.14	25	2.9
9	1.2	35	29.3	2.02	0.9	8.70	7.2	6.5	22.5	23.49	3.32	0.39	3.96	7.67	0.12	33	2.8
10	1.3	50	29.1	1.25	0.4	8.65	7.4	4.8	22.8	21.87	2.15	0.76	1.07	3.98	0.35	18	2.7
11	1.4	60	29.3	1.07	0.3	8.54	7.5	4.5	19.6	20.16	0.11	0.23	1.62	1.96	2.09	10	2.5
12	1.10	70	30.0	1.15	0.4	8.92	8.2	6.1	17.5	25.38	0.78	1.34	0.93	3.05	1.72	12	2.9
Max.	3.0	70	30.5	8.36	4.7	8.94	8.9	7.6	23.5	32.22	3.32	1.35	3.96	7.67	2.09	33	2.9
Min.	0.25	18	28.9	1.07	0.3	8.26	6.6	4.5	17.5	14.40	0.11	0.16	Nd	1.96	0.07	11	2.1
Average	1.3	37.3	29.5	3.49	1.8	8.61	7.43	5.77	21.49	23.65	1.75	0.69	2.09	4.53	0.39	19	2.76
SD	0.7	15.52	0.5	2.1	1.3	0.2	0.6	0.85	2.4	4.8	1.0	0.5	1.1	1.9	0.8	9.13	0.3

(d)

Stations	Depth (m) cm	Trans (cm) cm	Temp (°C) °C	EC μS/cm μS/cm	Sal‰	pH	DO mg/l	BOD mg/l	OOM mgO ₂ /l mg/l	T-Sn μg/g	DBT μg/g	TBT μg/g	TPhT μg/g	ΣOT	TBT/DBT	ΣOT/ T-Sn%	Igeo
1	0.4	12	28.4	5.3	2.9	7.8	ND	24.6	34.3	18.72	1.67	1.47	1.46	4.6	0.88	25	2.4
2	1.0	33	28.7	13.5	7.9	8.5	6.4	3.2	14	19.15	1.36	1.25	Nd	2.61	0.92	14	2.5
3	1.1	32	28.0	5.1	2.7	7.9	6.8	12.8	19.3	18.72	1.48	1.75	0.83	4.06	1.18	22	2.4
4	0.9	50	30.2	17.5	10.4	8.6	7.2	6.4	22.9	24.21	1.5	3.27	1.24	6.01	2.18	25	2.8
5	0.9	44	28.3	4.1	2.1	7.9	8.6	5.2	19.8	17.91	0.52	1.45	Nd	1.97	2.79	11	2.4
6	1.3	75	30.6	3.5	1.8	8.6	7.2	4.6	21.3	19.08	0.6	0.98	Nd	1.58	1.63	8	2.5
7	1.2	50	29.7	32.2	20.2	8.6	8.6	3.6	19.8	23.67	0.77	3.93	Nd	4.7	5.10	20	2.8
8	1.2	70	31.1	6.9	3.8	8.7	7.4	4.2	16.4	26.64	1.31	4.61	Nd	5.92	3.52	22	2.9
9	1.2	52	31.2	4.0	2.1	9.2	6.2	5.2	16.6	22.59	1.23	3.7	1.68	6.61	3.01	29	2.7
10	1.5	50	31.1	17.0	10.2	8.3	8.2	4.1	17.2	19.89	0.66	3.37	Nd	4.03	5.11	20	2.5
11	0.5	25	28.0	6.8	3.7	8.2	3.2	14.8	28.3	22.68	0.63	3.83	Nd	4.46	6.08	20	2.7
Max.	0.4	75	31.2	32.2	10.4	9.2	8.6	24.6	34.3	26.64	1.67	4.61	1.68	7.96	6.08	25	2.9
Min.	1.5	12	28.0	3.5	1.8	7.8	3.2	3.2	14	17.91	0.52	0.98	Nd	1.58	0.88	11	2.4
Average	1.1	44.8	29.6	10.5	6.2	8.39	6.98	8.06	20.9	21.21	1.07	2.69	1.30	5.06	2.51	24	2.6
SD	0.3	18.58	1.32	8.9	5.7	0.42	1.6	6.7	5.9	2.9	0.4	1.3	0.4	1.6	1.8	6.3	0.2

(e)

Stations	Depth (m) cm	Trans (cm) cm	Temp (°C) °C	EC μS/cm μS/cm	Sal‰	pH	DO mg/l	BOD mg/l	OOM mgO ₂ /l mg/l	T-Sn μg/g	DBT μg/g	TBT μg/g	TPhT μg/g	ΣOT	TBT/DBT	ΣOT/T-Sn%	Igeo
1	1.3	125	27.0	81.40	57.7	8.42	8.8	3.6	4.7	16.62	0.28	3.22	0.89	4.39	11.50	26	2.3
2	0.6	125	27.2	73.40	51.0	8.56	8.8	3.2	3.7	16.81	0.38	3.72	0.08	4.18	9.79	25	2.3
3	0.9	70	28.1	81.20	57.4	8.66	9.1	3.8	4.8	15.47	0.69	2.99	0.62	4.3	4.3 3	28	2.2
4	4.2	150	28.8	67.30	46.2	8.40	9.3	3.9	3.6	13.48	0.39	3.26	0.93	4.58	8.36	34	2.0
5	1.9	60	28.6	76.00	53.2	8.50	8.5	3.3	3.2	11.97	0.47	2.97	Nd	3.44	6.32	29	1.8
6	1.9	60	29.5	69.80	48.2	8.50	8.3	3.5	3.6	7.47	0.49	1.44	0.7	2.63	2.94	35	1.1
7	0.9	60	29.3	70.40	48.7	8.46	9.1	3.2	3.3	7.38	0.48	0.55	Nd	1.03	1.15	14	1.1
8	1.5	70	27.1	77.40	54.3	8.11	7.7	2.8	2.8	13.19	0.09	1.1	Nd	1.19	12.22	9	1.9
9	0.5	120	27.7	65.00	44.5	8.24	8.5	2.9	3.0	10.06	0.64	1.12	0.48	2.24	1.75	22	1.5
10	3.2	200	27.1	58.70	39.5	8.33	8.5	3.1	2.9	9.99	0.56	1.21	0.16	1.93	2.16	19	1.5
11	1.9	200	27.2	74.20	53.2	8.33	7.5	3.3	4.0	10.16	0.71	1.11	0.61	2.43	1.56	24	1.6
12	1.4	100	28.3	93.10	67.7	8.62	8.3	3.5	4.2	10.34	0.55	0.68	0.63	1.86	1.24	18	1.6
Max.	4.2	200	29.5	39.10	67.7	8.66	9.3	3.1	4.7	16.81	0.71	3.72	0.93	5.36	12.22	35	2.3
Min.	0.5	60	27.0	58.70	39.5	8.11	7.5	2.8	2.8	7.38	0.09	0.55	Nd	1.19	1.15	9	1.1
Average	1.8	111.7	28.0	74.0	51.8	8.43	8.5	3.7	3.7	11.91	0.48	1.95	0.57	3.00	4.06	25	1.74
SD	108.4	51.5	0.9	8.9	7.3	0.2	0.5	0.3	0.7	3.3	0.2	1.2	0.3	1.3	4.4	7.7	0.4

prevailed in the area of station 12 is a result of the isolation of this area in addition to the high rate of evaporation. The minimum value (0.3‰) was measured at station 11. The water salinity at different areas revealed that the salinity decreased in the order: Bardaweel > El-Manzalah > Mariout > El-Brullus > Edku depending on the amount of drainage waters. Conductivity showed a similar trend as salinity during the study period. The maximum EC value (93.10 mS/cm) was observed at station 12 of Lake El-Bardaweel, while the lowest one (1.07 mS/cm) was measured at station 11 of Lake El-Brullus. The water of the Nile delta Lakes lies in alkaline side (Tables 2(a)-(e)). pH values ranged between 7.26 - 8.92. Station 3 of Lake Maruit sustained the lowest pH value, while station 3 of Lake El-Brullus maintained the maximum value. The average values of pH at different areas decreased in the order: El-Brullus > Edku > El-Bardaweel > El-Manzalah > Mariout. The obtained results of dissolved oxygen showed a complete depletion at station 3 of Lake Maruit, while the maximum of 18.2 mg/l was observed at station 9 of Lake Edku. The average values of DO at different areas decreased in the order: Edku > El-Bardaweel > El-Brullus > El-Manzalah > Mariout. The obtained values of biological oxygen demand (BOD) were varied in a wide range. The minimum value (2.8 mg/l) was measured at station 8 of lake El-Bardaweel, while the maximum of 41.5 mg/l was measured at station 3 of lake Maruit. The average values of BOD at different areas revealed that the BOD decreased in the order: Mariout > El-Manzalah > Edku > El-Brullus > El-Bardaweel. The observed oxidizable organic matter varied also in a wide range between different Lakes (Tables 2(a)-(e)). The maximum value (63.6 mgO₂/l) was measured at station 3 of Lake Mariout and the minimum (2.8 mgO₂/l) was observed at station 8 of Lake El-Bardaweel. The average values of OOM at different areas decreased in the order: Mariout > El-Burullus > El-Manzalah > Edku > El-Bardaweel revealing high load of anthropogenic pollutants, i.e. agricultural, sewage and industrial in the lake Mariout [29].

3.2. Total Tin Compounds

The concentrations of total Sn (T-Sn) in sediments are shown in Tables 2(a)-(e). The results show that concentrations of T-Sn are scattered in the ranges of 15.48 - 26.55 μg/g dry wt, 12.06 - 24.41 μg/g dry wt, 14.4 - 32.22

$\mu\text{g/g}$ dry wt, 17.91 - 26.64 $\mu\text{g/g}$ dry wt and 7.38 - 16.81 $\mu\text{g/g}$ dry wt for lakes Mariout, Edku, El-Brullus, El-Manzallah and El-Bardaweel, respectively. The highest average value of T-Sn was measured in sediments of Lake El-Brullus due shipping activities, sewage and industrial wastes from different drains. A negative correlation ($r = -0.67$ at $p > 0.001$) was found between T-Sn and salinity (**Table 4**) is due to freshwater of the lakes, especially Edku, Mariout, El-Brullus, and El-Manzallah contaminated with sewage and industrial wastes from different drains. To determine whether the T-Sn concentrations pose a threat to aquatic life, T-Sn concentrations in sediments of the study area were compared to threshold limit value (TLV) of 30 $\mu\text{g/g}$ dry wt. It is clear from **Tables 2(a)-(e)** that T-Sn concentrations surpassed the TLV only in lake El-Brullus at station 6 located in the western sector (**Figure 1(d)**). This station is affected mainly by fishing boats that utilizes OTC as antifouling paints as well as the discharge of drain (8) which is characterized by its high concentration of pesticides and heavy metals [27] [28]. Accumulation values (Igeo) for Sn in northern lakes (**Tables 2(a)-(e)**) cleared out that sediments of the five lakes are classified as very low to low polluted.

3.3. Tributyl Tin Compounds

Tributyl tin (TBT) concentrations in sediments (**Tables 2(a)-(e)**) varied widely depending on the location and ranged from 0.14 to 2.29 $\mu\text{g/g}$ Sn dry wt, from 1.27 to 4.88 $\mu\text{g/g}$ Sn dry wt, from 0.16 to 1.35 $\mu\text{g/g}$ Sn dry wt, from 0.98 to 4.61 $\mu\text{g/g}$ Sn dry wt; and from 0.55 to 3.72 $\mu\text{g/g}$ Sn dry wt for lakes Mariout, Edku, El-Brullus, El-Manzallah and El-Bardaweel, respectively. It is clear that TBT is the predominant species of OTC in all lakes except lake El-Brullus (**Tables 2(a)-(e)**) with percentage to total OTC ranged from 32% to 100%. The present study cleared out that concentrations of TBT in investigated sediments are higher than that of its degradation where the ratio of TBT/DBT > 1 in most samples. This indicates that TBT is still being introduced into the aquatic environment, most probably due to the use of antifouling paints.

The highest average values of TBT are measured in Lake Edku and El-Manzallah. The highest TBT concentrations measured in Lake Edku are probably due to the presence of thick layer of green phytoplankton and herbicides covering the surface of most investigated areas. The presence of such thick layer inhibits transmittance of UV and consequently decreasing photo-degradation process for TBT compounds compared to other lakes. Intensive fishing activities in the lake are leading to historical deposition of TBT compounds. Moreover, the spreading of large number of islands through the lake that helps in increasing human activities as a main anthropogenic source of TBT compounds. The three large drains El-Barseik, Edku and El-Boseily, in addition to a number of sub-drains connected with main drains are acting as additional sources of pollution with TBT compounds (**Tables 2(a)-(e)**). On the other hand, higher concentrations of TBT in sediments of lake El-Manzallah are resulted from large number of boats and intensive fishing activities, the water transparency is generally low at lake El-Manzallah (0.45 m) due to the continuous disturbance of the mud bottom by wind action (**Tables 2(a)-(e)**) revealing relatively low photo-degradation of TBT by sunlight. Impacts of different drains, *i.e.* Hadous, Ramsisand and Bahr El-Baqar that discharge large quantities of industrial, agricultural and domestic wastes are also considered.

The Australian sediment quality guidelines for TBT are 5 ng/g and 70 ng/g for low and high trigger values [33]. Sediments from different stations of the present study contain TBT concentrations higher than the highest trigger value. This confirms that these sediments may pose a threat to a benthic biota. High levels of TBT in sediment samples suggested its widespread contamination and could be indicative of the continuing usage of TBT-based antifouling paints on ship hulls. Dowson *et al.* [34] introduced a classification for TBT concentrations in sediments, characterizing concentrations below 3 ng/g as uncontaminated, 3 - 20 ng/g as light contaminated, 20 - 100 ng/g as moderately contaminated, 100 - 500 ng/g as highly contaminated and above 500 ng/g as grossly contaminated. Comparing our results with this scheme, cleared out that sediments of the Egyptian northern lakes are highly to grossly contaminated by TBT. Relatively high concentrations of TBT in some sediments of different lakes may have a potential for future TBT release from the re-suspension of fine sediments. Page *et al.* [35] and Axiak *et al.* [36] showed soluble butyltins were released from heavily contaminated sediments. Comparing these concentrations to that observed in other areas around the world (**Table 3**) revealed that sediments of northern lakes contained TBT concentrations lower than that observed in other countries such as Sanricu Coast, Japan, North East Coast, Spain, Barcelona Harbor, Spain. However, Aegean Sea, Turkey, Mediterranean Sea, Egypt and Suez Gulf, Egypt showed lower TBT than northern lakes.

Table 3. Comparison of DBT, TBT and TPhT concentrations ($\mu\text{g/g}$ Sn dry wt) in sediments of the present study to those reported in the literature for other regions of the world.

Area	DBT	TBT	TPhT	References
Egyptian Northern Lakes	0.05 - 3.32	0.16 - 4.61		Present study
Sanricu Coast, Japan	1 - 3.4	0.002 - 14	-	[38]
North East Coast, Spain	0.047 - 3.519	0.051 - 7.673	-	[39]
Barcelona Harbor, Spain	-	0.098 - 4.702	-	[40]
Aegean Sea, Turkey	-	Nd - 3.008	-	[41]
River Thames, UK	0.012 - 0.219	-	-	[42]
North West Coast, Spain	0.0005 - 0.357	-	-	[43]
Cadiz Coast, Spain	-	-	0.015 - 0.940	[44]
German North Sea and Baltic Sea Marinas	-	-	0.015 - 3.540	[32]
Scheldt Estuary, The Netherlands	-	-	0.007 - 0.012	[45]
Spanish Northeastern Coast	-	-	12.27	[46]
Mediterranean Sea, Egypt	Nd - 2.16	Nd - 8.55	-	[10]
Suez Gulf, Egypt	0.07 - 2.27	0.35 - 0.77	-	[47]

3.4. Dibutyl Tin Compounds

DBT concentrations in sediments of the northern lakes varied widely and ranged from 0.05 to 2.11 $\mu\text{g/g}$ Sn dry wt, from 0.64 to 2.81 $\mu\text{g/g}$ Sn dry wt, from 0.11 to 3.32 $\mu\text{g/g}$ Sn dry wt, from 0.52 to 1.67 $\mu\text{g/g}$ Sn dry wt and from 0.09 to 0.71 $\mu\text{g/g}$ Sn dry wt for lakes Mariout, Edku, El-Brullus, El-Manzallah, and El-Bardaweel, respectively. Presence of DBT at most sampled sites of lakes indicates that there is an important photochemical and biological degradation process on these sediments, together with its mobilization. This idea is confirmed by a relative higher abundance of DBT in sediments of lake El-Brullus (**Tables 2(a)-(e)**). TBT/DBT ratios are found >1 confirms also an active degradation process. However, this ratio may also indicate that either TBT inputs to Lake El-Brullus are older or that new inputs are absent or negligible than TBT degradation rate [37]. In spite of the positive correlation ($r = 0.45$, $p = 0.001$) between DBT and T-Sn, there was no significant correlation between TBT and DBT ($r = -0.03$, $p = 0.001$) in sediments from the five northern lakes (**Table 4**). The absence of such correlation might give clear evidence that both compounds did not come from the same source. This finding confirms the slow rate of degradation of TBT in sediments. On the other hand, a negative correlation ($r = -0.48$, $p = 0.001$) was found between DBT and water salinity (**Table 4**). The current research was not specifically designed to evaluate which factor strongly related to such variations in the distribution of these OTC.

Comparing concentrations of DBT to those reported in literature (**Table 3**) showed concentrations similar to others found in Sanricu coast, Japan and North east coast. In contrast, they were higher than the ranges reported for River Thames, UK, for North west coast Spain, for Mediterranean Sea, Egypt and for Suez Gulf, Egypt.

3.5. Triphenyltin Compounds

TPhT in sediments of the northern lakes were not observed in most sediments and its concentrations ranged from Nd to 0.63 $\mu\text{g/g}$ Sn dry wt, Nd to 0.81 $\mu\text{g/g}$ Sn dry wt, Nd to 3.96 $\mu\text{g/g}$ Sn dry wt, Nd to 1.68 $\mu\text{g/g}$ Sn dry wt, and Nd—0.93 $\mu\text{g/g}$ Sn dry wt for lakes Mariout, Edku, El-Brullus, El-Manzallah and El-Bardaweel; respectively. Variations occurring among lakes depending on changes in chemical and physical characteristics of each lake, various sources of pollutants, degradation process and drains run-off. It is clear from **Tables 2(a)-(e)** that TPhT is the predominant species in lake El-Brullus. It exhibited concentrations of TPhT range from Nd to 3.96 $\mu\text{g/g}$ Sn dry wt. This could be attributed to the presence of several numbers of drains, channels and sub-drains, *i.e.* drain west El-Burullus, El-Hoksa, Terra and Brimbal canal discharging a huge amounts of agriculture wastes containing TPhT which is used as fungicide in agriculture purposes into the lake. TPhT concentrations are decreasing in the following order: El-Burullus ($2.59 \mu\text{g/g}$ Sn) $>$ El-Manzallah ($1.30 \mu\text{g/g}$ Sn) $>$ Edku ($0.75 \mu\text{g/g}$ Sn) $>$ El-Bardaweel ($0.57 \mu\text{g/g}$ Sn) $>$ Mariout ($0.40 \mu\text{g/g}$ Sn). Although a significant positive correlation ($r = 0.67$, $p = 0.001$) was found between DBT and TPhT, a weak negative correlation ($r = -0.37$, $p = 0.001$) was found between TBT and TPhT in sediments from northern lakes revealing different sources of pollution (**Table 4**).

Table 4. Correlation coefficient (r) between physicochemical parameter of lake water and organotin compounds in sediments of all lakes.

Parameters	Depth	Trans	Temp	EC	Salinity	pH	DO	OOM	BOD	DBT	TBT	TPhT
T-Sn	-0.31	-0.55	0.43	-0.66	-0.67	0.29	-0.05	0.37	0.12	0.45	0.07	0.38
DBT	-0.15	-0.37	0.22	-0.48	-0.48	0.19	0.09	0.26	0.06		-0.03	0.67
TBT	-0.05	0.10	-0.05	0.10	0.09	0.17	0.13	-0.29	-0.21			-0.37
TPhT	0.15	0.38	0.44	-0.41	-0.41	0.22	-0.24	0.37	-0.12			

Comparing concentrations of TphT to that reported in the Literature (**Table 3**) showed higher concentrations than those reported for Cadiz coast, Spain, for German North Sea and Baltic Sea marinas, and for Scheldt estuary, The Netherlands due to the extensive shipping activities and the discharge of sewage, agricultural and industrial effluents into Egyptian lakes. On contrast they were lower than those reported for Spanish northeastern coast.

The results in **Tables 2(a)-(e)** show that the highest average concentrations of total OTC were found in sediments of lake El-Manzallah (5.06), lake Edku (4.54), Lake El-Brullus (4.53) compared to Lake El-Bardaweel (3.0) and Lake Mariout (2.61) depending on the pollution sources. A percentage of total OTC to total tin concentration were ranged from 13% to 25% in sediments of the northern lakes. Lake Edku (24%), lake El-Brullus (19%), lake El-Manzallah (24%) and Lake El-Bardaweel (25%) were remarkably higher than Lake Mariout (13%) as shown in **Tables 2(a)-(e)**. Significant levels of OTC were found in sediments of these lakes.

Results of the present investigation show that the degree of pollution in the northern lakes is influenced by industrial and sewage discharges. Concentrations found of TBT, DBT and TPhT in the sediments are a consequence of the historical use of OTC, mainly related to fishing activities in the lakes. Regular monitoring of OTC pollution to assess hazardous effects for the aquatic life is recommended.

4. Conclusion

OTCs in the five northern lakes revealed some highly localized areas of contamination that were severe enough to cause harmful effects on marine flora and fauna. Levels of contamination are depending mainly on the shipping activity, changes in chemical and physical characteristics of each lake, various sources of pollutants, degradation process and drains run-off. No correlation between TBT and DBT in sediments demonstrated that both compounds did not come from the same source (*i.e.* antifouling paints), indicating their different deposition, and confirming the slow rate of degradation in sediments. The results concluded that sediments of the Egyptian northern lakes were highly to grossly contaminate by TBT. Among the five northern lakes, Lake El-Brullus shows the maximum concentration of Σ OT. The high input of OTC particularly in sediments, overrides any removal by degradation pathway. Up to the present, no studies on biological effects have been done in this area. The present research concluded that TBT levels suggested that stringent legislative measures were needed to curb the usage of these contaminants in marine paints. Otherwise, OTC would have detrimental effect on our marine resources and ecosystem, which probably could not reverse.

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