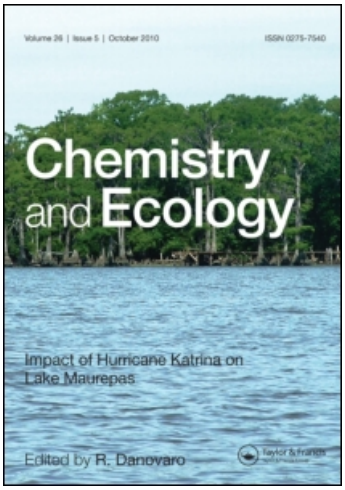


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# Speciation of organotin compounds in sediments of semi-closed areas along the Mediterranean coast of Alexandria

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# Speciation of organotin compounds in sediments of semi-closed areas along the Mediterranean coast of Alexandria

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Organotin compounds were measured in sediments of four different semi-closed areas of the Mediterranean coast of Alexandria: the Eastern Harbour, Western Harbour, El-Max Bay, and Abu-Qir Bay. Due to the commercial trade activity inside the Western Harbour, in addition to the effect of wastes discharged from El Noubaria canal, it shows the highest concentrations of total tin ($6.34\,\mu g\,g^{-1}$ dry weight), dibutyl tin ($1.63\,\mu g\,g^{-1}$ wet weight), tributyl tin ($0.33\,\mu g\,g^{-1}$ wet weight) and diphenyl tin ($1.06\,\mu g\,g^{-1}$ wet weight) compared with other locations. The concentrations of TBT species showed the highest contents compared with DBT and DPhT compounds in all sampling areas. This trend might be due to the worldwide use of TBT not only as biocides in antifouling paints but also as preserving agents for wood, fungicides in agricultural activities, and heat and UV stabilizers of PVC, which results in a direct release of TBT into the water body, accumulation in aquatic fauna. There is also precipitation into sediments and a decrease in degradation rate into its derivatives. Variations, types, concentrations, and distribution of different organotin compounds are discussed in the areas under investigation. The study reveals a new record of organotin compounds along the Alexandria coast and makes comparisons with other surrounding areas of interest.

*Keywords*: Organotin; Sediments; Alexandria coast; Egypt

## 1. Introduction

The occurrence of organotin compounds in aquatic environment is of great concern because of their different toxicological behaviours. Thus, speciation of these compounds becomes necessary to evaluate the risk associated with their occurrence [1]. They generally occur at very low concentrations (ng–μg level) in highly polar media such as natural waters or exceedingly complex biological matrices [2]. Environmental monitoring and toxicological studies dealing with mussels and fish [3–6] have indicated that organotin compounds continue to pose a major ecotoxicological threat in the aquatic environment. Several years after regulation, organotin

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compounds are still detected in marine organisms worldwide. A tributyltin (TBT) level of about 0.8 ng $l^{-1}$ causes pathogenic symptoms in the dog whelk *Nucella lapillus* [7]. The harmful effects of coastal environment contamination by TBT were first observed in the Bay of Arcachon oyster. In addition, significant and repeated disturbances were observed to occur in the *Crassostrea giges* oyster farms of the Arcachon Bay along the French Atlantic coastline [8]. They proved that organotin compounds are extremely toxic to aquatic organisms in general and to bivalves, molluscs, and gastropods, in particular, for which lower effective levels were $<1$ ng $l^{-1}$.

The Alexandria region was one of the most important marine fishery grounds in Egypt. Many fishes of economic importance migrate to coastal waters for breeding, penetrate into brackish water lakes connected to the sea to feed, and then attain sexual maturity and migrate back to the sea. Nevertheless, with the presence of areas with high shipment and pleasure-boating activities, incorporating numerous harbours and marines in Alexandria, the available data on organotin levels are scarce.

The present investigation aims to study the speciation of different forms of organotin compounds residue in sediment of the semi-closed areas along the Mediterranean coast of Alexandria. The study also includes a new record of organotin compounds on the Alexandria coast and their comparison with other surrounding areas of interest. Types, concentrations, and distributions of different organotin compounds as well as factors affecting their distribution patterns are discussed in detail.



## 2. Materials and methods

### 2.1 *Study area*

Alexandria occupies a T-shaped peninsula and strip of land separating the Mediterranean from Lake Mariout. The Mahmudiya Canal, connecting Alexandria with the River Nile, runs to the south of the city and, by a series of locks, enters the Western Harbour. Some degree of variation in water colour seen on El Max Bay and the Western Harbour is due to municipal and industrial waste water entering the Mediterranean. The total cumulative volume of waste water disposed of into the sea from all point sources along this stretch of coast is roughly equal to the Nile outflow from the Rosetta outlet: roughly 9 million $m^3$ $day^{-1}$, that is 3.33 $km^3$ $yr^{-1}$. Two-thirds of the city waste water is released into Lake Mariout and subsequently pumped into El Max Bay, along with agricultural run-off from the northwestern delta. A few hundred metres from the foundered Pharos lighthouse, 200 000 $m^3$ of waste water enters the sea each day at Kayet Bay.

### 2.2 *Sampling and analyses*

Sediment samples were collected from 28 stations by means of a hydro-Bios stainless steel grab sampler (0.2 m capacity) from depths ranging between 1.5 and 25 m and the surface layer (about 2 cm) collected with a Teflon spatula (figure 1). The sediments were divided into three sub-samples; one for determination of the organic carbon content after air drying in a clean-laminar flow branch, a second for the determination of organotin compounds, and a third stored frozen until analysis for the determination of the total tin compounds. The water content of each sample was determined by drying a representative sample in an oven at 105 °C over night to a constant weight [9].

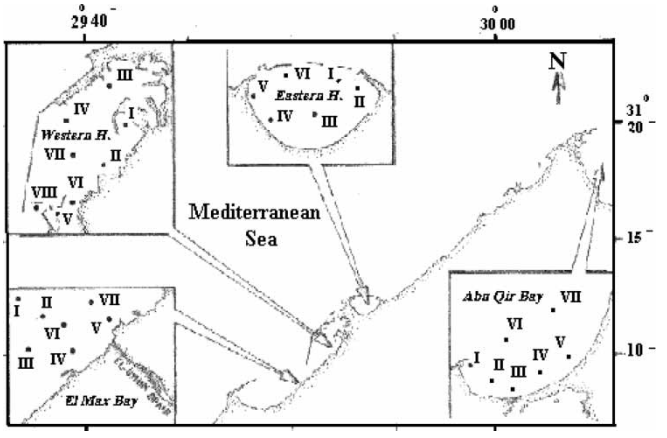


Figure 1. Sampling stations of the area of investigation along the Alexandria coast Suez Canal during 2004.



Organic carbon was determined using acid/dichromate titration method [9]. Total tin ($\sum$Sn) compounds in sediment were determined according to [10]. Samples (0.5 g) of freeze-dried sediment were weighed into 30 ml Teflon containers with screw caps and digested with $HNO_3$, $HClO_4$, and HF-HCl, the sediment ends up in 5 ml of an equal volume mixture of 6 N HCl and 2 N $HNO_3$ resulting from the total destruction procedure. When digestion was completed, the sample was transferred into a measuring flask (25 ml), diluted with deionized distilled water up to 25 ml, and reacted with $NaBH_4$. Then, the concentration of total tin was measured by hydride-AAS (Perkin Elmer, Model 2380). Organotin compounds, TBT, DPhT, and DBT, were determined according to [11]. Twenty grams of sediment samples (wet weight) was placed into a 300 ml separating funnel and extracted for 30 min with 50 ml ethyl acetate after adding 50 ml of water and 5 ml HCl. The mixture was centrifuged (2500 rpm, 5 min), and 30 ml of the organic layer was transferred to a 50 ml round-bottom flask. The organic layer was evaporated nearly to dryness (0.1 ml) under a rotary-vacuum at 40 °C. The residue was dissolved in 1 ml of ethanol, 2 ml of hydrogenation reagent was added while shaking, and the mixture left to stand for 10 min at room temperature. Five millilitres of water was added to the reaction mixture, shaken slightly, and transferred to a 50 ml separating funnel. The flask was then rinsed with two 5 ml quantities of water, transferred to the separating funnel and extracted for 5 min with 5 ml of hexane, extracted through a silica-gel column clean-up after adding 5 g of NaCl. The hexane layer was poured onto a silica gel column and passed through the column. The first 20 ml was collected in a round-bottom flask and evaporated to 2 ml under a rotary vacuum at 40 °C. The concentrate was transferred to a 5 ml gradual test tube, the flask was rinsed with hexane, and the mixture was adjusted to 1 ml under nitrogen stream in a 40 °C dry bath. The sample solution was then injected into a gas chromatograph (GC/ECD, Model HP 5890 II).

### 2.3 *Method validation and quality-control studies*

Method validation and quality control of the samples were carried out using standard solutions and applying the computerized 4.3 quality system program provided by DANIDA from VKI. Two natural samples were analysed in duplicate in each of six batches of samples after spiking with a known concentration from the standard solution. The same two natural samples were analysed without spiking. The highest and lowest percentages of recovery for spiked samples were used to determine the accuracy, which ranged between 90 and 105%, while precision was agreed to be within 10%.

## 3. Results and discussion

### 3.1 *Water content*

Water contents varied between 14.3 and 75.3%, with an average value of 37.7% in the area of study. The maximum values of 75.3, 70.9, 38.2, and 35.0% were observed at Abu Qir Bay (in front of the El Tabia pumping station), the western Harbour (in front of shipyard floating dock), the Eastern Harbour (station VI), and El Max Bay (station VII), respectively, suggesting that the sediments of these areas are mainly muddy. The muddy nature of sediment reflects the effect of wastes discharged in these areas. The minimum % of water contents of 14.3, 15.7, 16.9, and 17.8% were observed at the Western Harbour (station IV), El Max Bay (station II), Eastern Harbour (station III), and Abu Qir Bay (station I), respectively, suggesting that these areas were sandy in nature.

Figure 2 shows that the average percentages of water contents decreased in the order Western Harbour > Abu-Qir Bay > Eastern Harbour > El-Max Bay, with values of from 14.3–70.9%, 17.8–75.3%, 16.9–38.2%, and 15.7–35.0%, respectively. These values are closely related to the particle size of the sediments in these areas and definitely will affect the rate of reactions, particularly the redox processes, the pH, and the overall amount of organotin compounds that can be trapped due to the hydrophobic character.



### 3.2 *Total organic carbon (TOC)*

Organic carbon contents varied between 0.03 and 4.8% with an average value of 1.32%. The maximum values of 4.8, 3.85, 0.65, and 3.25 were observed at Abu Qir Bay (in front of El Tabia pumping station), the western Harbour (in front of shipyard floating dock), the Eastern Harbour (station VI) and El Max Bay (station VII), respectively, suggesting that sediments of these areas are affected by the impacts of different land-based sources especially the effect

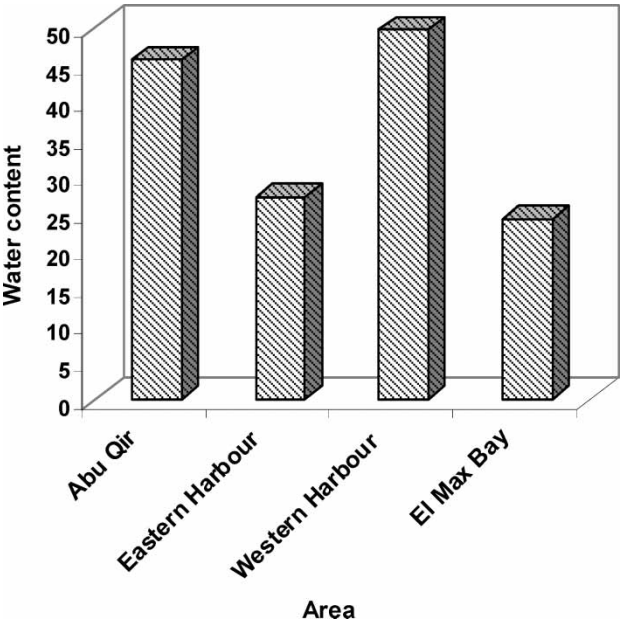


Figure 2. Average percentages of water contents in the investigated area.

of sewage wastes discharged in these areas. Going away from the shore area, minimum percentage organic carbon contents of 0.25, 0.03, 0.35, and 0.03 were observed at the Western Harbour (station VIII), El Max Bay (station I), Eastern Harbour (station II) and Abu Qir Bay (station VII), respectively. The average percentage of organic carbon decreases in the order Western Harbour > Abu-Qir Bay > El-Max Bay > Eastern Harbour, with values of 0.25–3.85, 0.03–4.8, 0.03–3.25, and 0.35–0.65%, respectively. This clearly reflects the effect of agricultural and industrial waste waters which are discharged from El-Nubaria Canal to the area of study. Positive and significant correlations ($p < 0.01$) between organic carbon and water contents in sediments of Abu Qir Bay ($r = 0.86$), Eastern Harbour ($r = 0.69$), western Habour ($r = 0.44$), and El Max Bay ($r = 0.42$) were observed.

### 3.3 *Total tin compounds*

Concentrations of total tin ranged from 0.47 to 6.34 ($\mu g\, g^{-1}$, dry wt.) in the study area (table 1). The relatively high values observed in sediments of different areas are mainly due to the

Table 1. Concentrations of TSn ($\mu g\, g^{-1}$; dry weight), TBT, DBT, and DPhT ($\mu g\, g^{-1}$; wet weight) during 2004.

| | | Area | | | | | |
|---|---|---|---|---|---|---|---|
| Stations | Species | Abu Qir Bay | Eastern Harbour | Western Harbour | El Max Bay | Average | ±SD |
| I | TSn | 4.21 | 4.63 | 6.34 | 1.11 | 4.07 | 2.18 |
| | TBT | 1.09 | 0.65 | 1.63 | 0.08 | 0.86 | 0.66 |
| | DBT | 0.02 | 0.15 | 0.12 | 0.01 | 0.07 | 0.07 |
| | DPhT | 0.60 | 0.24 | 0.27 | 0.02 | 0.28 | 0.24 |
| II | TSn | 1.94 | 3.80 | 4.72 | 2.63 | 3.27 | 1.23 |
| | TBT | 0.27 | 0.36 | 0.87 | 0.21 | 0.43 | 0.30 |
| | DBT | 0.04 | 0.02 | 0.03 | 0.01 | 0.03 | 0.01 |
| | DPhT | 0.06 | 0.20 | 0.32 | 0.03 | 0.15 | 0.13 |
| III | TSn | 1.72 | 4.21 | 6.23 | 3.03 | 3.80 | 1.91 |
| | TBT | 0.47 | 0.40 | 1.28 | 0.60 | 0.69 | 0.40 |
| | DBT | 0.11 | 0.02 | 0.21 | 0.01 | 0.09 | 0.10 |
| | DPhT | 0.12 | 0.21 | 0.46 | 0.21 | 0.25 | 0.15 |
| IV | TSn | 5.47 | 3.63 | 3.45 | 4.16 | 4.18 | 0.91 |
| | TBT | 1.22 | 0.30 | 0.40 | 0.99 | 0.73 | 0.45 |
| | DBT | 0.16 | 0.07 | 0.13 | 0.03 | 0.10 | 0.06 |
| | DPhT | 0.39 | 0.15 | 0.08 | 0.77 | 0.35 | 0.31 |
| V | TSn | 3.68 | 3.92 | 4.95 | 0.47 | 3.26 | 1.94 |
| | TBT | 0.62 | 0.52 | 0.93 | 0.03 | 0.53 | 0.37 |
| | DBT | 0.14 | 0.02 | 0.03 | 0.01 | 0.05 | 0.06 |
| | DPhT | 0.48 | 0.17 | 1.06 | 0.15 | 0.47 | 0.42 |
| VI | TSn | 2.36 | 2.31 | 5.39 | 2.66 | 3.18 | 1.48 |
| | TBT | 0.70 | 0.21 | 0.86 | 0.40 | 0.54 | 0.29 |
| | DBT | 0.03 | 0.01 | 0.16 | 0.00 | 0.05 | 0.07 |
| | DPhT | 0.48 | 0.07 | 0.35 | 0.31 | 0.30 | 0.17 |
| VII | TSn | 1.18 | – | 4.69 | 0.76 | 1.66 | 2.16 |
| | TBT | 0.18 | – | 1.00 | 0.06 | 0.31 | 0.51 |
| | DBT | 0.08 | – | 0.33 | 0.01 | 0.10 | 0.17 |
| | DPhT | – | – | 0.56 | 0.01 | 0.14 | 0.39 |
| VIII | TSn | – | – | 5.23 | – | 1.31 | |
| | TBT | – | – | 0.91 | – | 0.23 | |
| | DBT | – | – | 0.07 | – | 0.02 | |
| | DPhT | – | – | 0.66 | – | 0.17 | |

*Note*: TSn: total tin concentration; TBT: tributyl tin; DBT: dibutyl tin; DPhT: diphenyl tin.

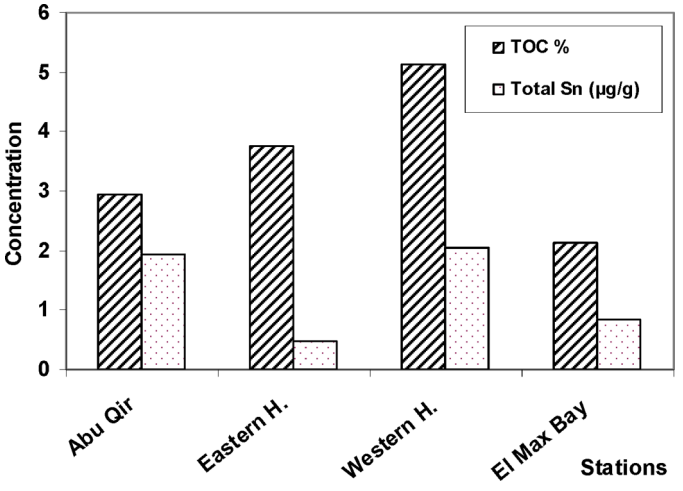


Figure 3. Relationship between total Sn and TOC% concentrations during 2004.



increasing use of organotin compounds as antifouling paint in boats, its use as biocide for cooling waters inside power stations, in addition to the effect of huge amounts of industrial wastes discharged into the area as in the case of station IV at Abu Qir Bay. This is in accordance with a previous study [12] which stated that the increasing numbers of commercial ships in the Western Harbour that use organotin compounds as antifouling paints has resulted in increasing concentrations of tin compounds in the harbour.

Investigations of the correlation between total Sn contents and TOC% (figure 3) revealed no significant correlations ($r = 0.42$), which is in accordance with a previous study [13] which stated that no significant correlation was found between tin compounds and organic carbon contents.

Comparing concentrations measured in the present study (1.31–4.18 $\mu g\, g^{-1}$, dry wt.) with those observed by previous workers, one can easily find that they were lower than those measured in Spain for Riade Arouda (5.0–20.8 $\mu g\, g^{-1}$) [14], Gipuzkoa (11.0–113 $\mu g\, g^{-1}$) [15], and the Gulf of Codiz (8.1–24 $\mu g\, g^{-1}$) [16]. However, they were higher than those recorded in bivalves (*Donex trunculus*), 0.37–0.58 $\mu g\, g^{-1}$, and fish (*Trigloporus lastoviza* and *Diplodus sargus*), 0.37–0.53 and 0.13–0.24 $\mu g\, g^{-1}$, respectively, recorded along the Alexandria coast [17].

### 3.4 *Tributyltin compounds (TBT)*

Concentrations of tributyltin in the investigated area ranged between 0.03 and 1.63 $\mu g\, g^{-1}$ wet wt. (table 1). The relatively high concentration of 1.63 $\mu g\, g^{-1}$ measured at station (I) situated at the shipyard dry dock of the Western Harbour is mainly due to the activity of commercial boats inside the harbour (the major commercial harbour in Egypt). TBT compounds are used in painting the hulls of new vessels and on older vessels during dry docking. Ian *et al.* [18] stated that the average TBT content of marine paints in the North Sea was about 4%, and therefore approximately 1200 tonnes of TBT was applied annually to ships hulls. Loss of TBT during dry docking will arise from hull washing/scrapings and the overspray of new paint. In the meantime, the high concentrations observed at station (IV) in front of the power station near Petrojet Company at Abu-Qir Bay, were a result of the TBT species used in antifouling

paints for ships and fishing nets; different wastes discharged from the power station near Petrojet Company in which TBT compounds are used as a biocide for cooling waters inside the power station; and the effect of huge amounts of industrial wastes discharged from El-Tabia pumping station. The highest concentrations measured at station (I) at the Eastern Harbour were also dependent on the amount of sewage sludge discharged into the harbour. This is in accordance with a previous study [19], which revealed that the major contribution of tin compounds resulted from sewage sludge discharged into the Eastern Harbour. On the other hand, station (IV) in front of the petrochemical platform at El-Max Bay exhibited the highest concentration of TBT due to the high concentration of total tin and high amount of organic carbon (3.25%) and organic matter at this station. The organic carbon content of sediments influences the bioavailability and toxicity of TBT, and sediments with high concentrations of organic matter may act as a sink for TBT [20, 21].

The average concentrations of TBT (0.23–0.86 $\mu g\, g^{-1}$, wet wt.) measured in the study area were lower than those observed [22] in Goleborg Harbour at Sweden (10.94 $\mu g\, g^{-1}$) and in Vancouver Harbour in Germany (10.8 $\mu g\, g^{-1}$) [23]. However, the concentrations were comparable with those measured in Toronto, Canada (0.5 $\mu g\, g^{-1}$) [24], Mainz, Germany (0.18 $\mu g\, g^{-1}$) [25], and Osaka, Japan (0.28 $\mu g\, g^{-1}$) [26]. However, the concentrations were higher than those recorded in bivalves (*Donex trunculus*), 0.16–0.25 $\mu g\, g^{-1}$, and fish (*Trigloporus lastoviza* and *Diplodus sargus*), 0.15–0.21 and 0.04–0.08 $\mu g\, g^{-1}$, respectively, recorded along the Alexandria coast [17].



### 3.5 *Dibutyltin species (DBT)*

DBT in Abu-Qir Bay varied from 0.019 to 0.16 $\mu g\, g^{-1}$, with an average of 0.075 $\mu g\, g^{-1}$, wet wt. (figure 4). The maximum value of DBT observed at station (IV) in front of the power station near the Petrojet Company is due to the high concentration of TBT and total tin at this station. According to previous studies [27, 28], butyltin species concentrations decrease in the sequence tribulyltin > dibutyltin > monobutyltin. A distribution like this is considered to be typical for the degradation of tributyltin in surface waters. Therefore, dibutyltins are probably broken-down products of tributyltin. In the Eastern Harbour, DBT varied from 0.008 to 0.153 $\mu g\, g^{-1}$, wet wt., with an average of 0.046 $\mu g\, g^{-1}$, wet wt. The maximum value of DBT observed at station (I) located at Al-Boughaz is due to the increase in total tin (4.63 $\mu g\, g^{-1}$, dry wt.) and TBT (0.65 $\mu g\, g^{-1}$, wet wt.) at this station. The increase in DBT could be explained by the effect of sewage sludge discharging into the Eastern Harbour. DBT concentrations in sediments of the Western Harbour varied from 0.03 to 0.33 $\mu g\, g^{-1}$, wet wt., with an average value of 0.135 $\mu g\, g^{-1}$, wet wt. The maximum DBT concentrations were observed at station (VII) located at the centre of the harbour. The high concentrations of DBT at this station are due to the direct emission or degradation of TBT to DBT in the surface water [27] and microbial degradation of TBT to DBT and MBT in oxic and anoxic sediments [28]. DBT concentrations in the sediments of El-Max Bay varied from 0.001 to 0.03 $\mu g\, g^{-1}$, wet wt., with an average value of 0.010 $\mu g\, g^{-1}$, wet wt. The maximum value of DBT concentrations observed at station (IV) in front of the petrochemical platform is due to the high organic carbon content. In addition, the effect of huge amounts of waste waters discharged from El-Umum drain is also a contributing factor in increasing organotin concentrations in the sediments of El-Max Bay.

The average concentrations of DBT (0.02–0.1 $\mu g\, g^{-1}$) were lower than those recorded in Toronto, Canada (0.25 $\mu g\, g^{-1}$) [22], Vancouver Harbour, Germany (8.51 $\mu g\, g^{-1}$) [24], Mainz, Germany (0.47 $\mu g\, g^{-1}$) [23], and Osaka, Japan (0.14 $\mu g\, g^{-1}$) [25]. However, the concentrations were higher those recorded in bivalves (*Donex trunculus*), 0.01–0.03 $\mu g\, g^{-1}$, and fish

(*Trigloporus lastoviza* and *Diplodus sargus*), 0.01–0.02 and 0.01–0.016 $\mu g\, g^{-1}$, respectively recorded along the Alexandria coast [17].

### 3.6 *Diphenyltin species (DPhT)*

DPhT concentrations in Abu-Qir Bay varied from 0.06 to 0.59 $\mu g\, g^{-1}$, wet wt., with an average concentration of 0.32 $\mu g\, g^{-1}$, wet wt. (table 1). The maximum value of DPhT observed at station (I) located in front of the fishing harbour is due to the increase in number of fishing boats inside the Bay and the use of triphenyltin as an antifouling agent in boat paints [29]. The highest concentration of DPhT in Abu-Qir Bay may also be due to the effect of huge amounts of brackish waters discharged in agriculture from different sources. According to a previous study [30], high concentrations of butyltin and phenyltin derivatives are possibly due to their use in agricultural and industrial activities or their high sorption affinity onto soils. DPhT in the Eastern Harbour was varied from 0.07 to 0.24 $\mu g\, g^{-1}$, wet wt., with an average concentration of 0.17 $\mu g\, g^{-1}$, wet wt. The maximum value of DPhT observed at station (III) resulted mainly from sewage waters discharged into the Harbour through the main pipeline (12.000 $m^3\, d^{-1}$). According to another study [19], the occurrence of butyltin and phenyltin derivatives in an aquatic environment could be a result of sewage sludge and high degradation of triphenyltin to phenyltin derivatives (diphenyltin and monophenyltin), in addition to the increasing number of fishing boats and time spent inside the harbour. The DPhT concentration in the Western Harbour varied from 0.08 to 1.061 $\mu g\, g^{-1}$, wet wt., with an average of 0.47 $\mu g\, g^{-1}$, wet wt. The maximum concentration of DPhT observed at station (V) located at the oil dock is due to the type of activity for this dock and the time commercial ships spend inside the dock as well as the degradation of TPhT to phenyltin derivatives (DPhT and MPhT) and the effect of huge amounts of waste waters (agriculture and industrial waste) discharged into the harbour from El-Noubaria canal. According to a previous study [4], the occurrence of butyltin and phenyltin derivatives in the aquatic environment could be a result of the use of these compounds in agriculture as fungicides. At El-Max Bay, DPhT concentrations varied from 0.013 to 0.768 $\mu g\, g^{-1}$, wet wt., with an average concentration of 0.21 $\mu g\, g^{-1}$, wet wt. The maximum value of DPhT observed at station (IV) located in front of the petrochemical platform is due to high concentrations of total tin at this station, high amounts of organic

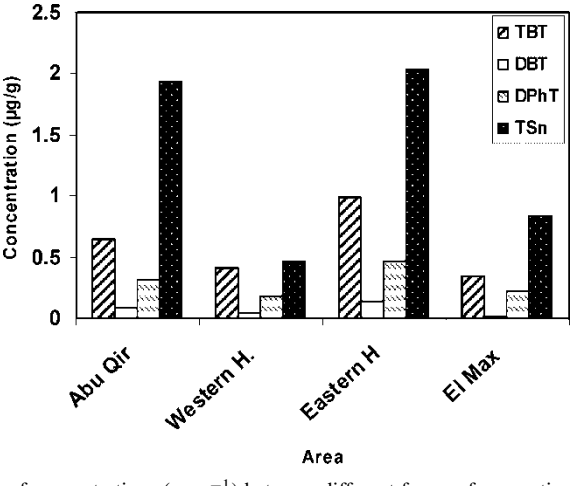


Figure 4. Comparison of concentrations ($\mu g\, g^{-1}$) between different forms of organotin compounds measured in the area of investigation during 2004.

carbon (3.25%), and the use of triphenyltin as an antifouling agent in fishing-boat paints in the bay, in addition to huge amounts of wastewaters (agricultural and/or industrial waste waters) discharged from El-Umum drain.

The average concentrations of DPhT in the study area (0.14–0.47 $\mu g\, g^{-1}$) were lower than those recorded in Spain (the western Mediterranean Sea): 2.55 $\mu g\, g^{-1}$ [31]. However, the concentrations were higher than those recorded in bivalves (*Donex trunculus*), 0.04–0.07 $\mu g\, g^{-1}$, and fish (*Trigloporus lastoviza* and *Diplodus sargus*), 0.03–0.06 and 0.01–0.04 $\mu g\, g^{-1}$, respectively, recorded along the Alexandria coast [17]. Figure 4 shows that the order of decreasing organotin compounds was: TSn > TBT > DBT > DPhT, with the predominance of TBT compound in all areas of study, especially in the eastern harbour.

Results from various laboratory studies have demonstrated that TBT is toxic to aquatic biota at concentrations of $>2\, ng\, l^{-1}$ [32]. Regarding the production, usage, accumulation, and toxicity of organotin compounds, and based on the results obtained in the present study, we conclude that the prohibition of the use of organotin in antifouling paints was an effective action for both the protection and conservation of marine life. Furthermore, detecting the concentration ($\mu g\, g^{-1}$) of TBT, DBT, and DPhT in our investigated areas proves that there is a real need for improvement in the enforcement of existing regulations.



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