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Accumulation and rate of degradation of organotin compounds in coastal sediments along the Red Sea, Egypt

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ABSTRACT

Coastal sediments' quality may become impaired due to anthropogenic pressures, risking the well-being of humans and the ecosystem concerned. In this study, butyltin contamination in Egyptian Red Sea Coast's sediments was assessed for the first time, following the ten-year ban on antifouling paints' harmful organotin. We collected sediment samples from ten stations based on the proximity of anthropogenic activities. TBT, MBT, DBT, ΣBT, and TSn's concentrations were within the following ranges 7–220, 4.2–78, 14.7–339.8, 3.5–48.8 ng Sn g^{-1}, and 0.09–1.47µg Sn g^{-1}, respectively. The highest of butyltin concentrations were recorded in summer. TBT was the most abundant (52.79%–53.13%) while the averages of percentages of DBT were 18.45% and 19.33%, whereas MBT values were 27.88% and 28.42% in winter and summer, respectively. A significant positive correlation was found between butyltin derivatives and geochemical characteristics such as grain size of investigated sediments and organic matter. Contamination patterns of butyltin derivatives in the sediments of the area of study were TBT > MBT > DBT and most of the investigated samples had BDI values around one, which proves TBT has no recent input and degraded immensely. Meanwhile, a portion of 80% of the samples was established to be highly tributyltin contaminated.

INTRODUCTION

Coastal areas' marine pollution is a paramount environmental challenge resulting from industrial activities during the past decades, leading to the discharge of plethora of substances of critical toxicity **(Libralato *et al.*,2010; Prato *et al.*, 2015, Younis, 2018; El-Naggar *et al.*, 2019)** and accumulating of these pollutants in coastal sediments which considered as reservoir of pollution **(Nikolaou *et al.*, 2009; Rzetala, 2015; Younis *et al.*, 2018)**.

Since the 1940s, Organotin Compounds (OTCs) became one of the commonly used pollutants following the expansion of the plastic manufacturing, especially the manufacturing of Polyvinyl Chloride (PVC) **(Blunden *et al.* 1984)**. Mono-

and diorganotin compounds were utilized as stabilizing ingredients in plastic materials **(Hoch, 2001)**. Triphenyl- (TPT) and tributyltin (TBT) were utilized as biocide agents since the late 1950s because of their extensive toxicity traits **(Said *et al.*, 2006; Yi *et al.*, 2012)**.

By the 1960s, Tributyltin (TBT) incepted its role as an antifouling agent, acting as a protective coating in paints applied to aquaculture nets, hulls of ships, offshore structures, long-term seawater-exposed stationary structures, and ducts to diminish situation resulting from fouling adhesion.

Corrosion and fuel consumption resulted from biofouling settlement and influenced the navigating boats and ships **(Kotrikla, 2009)**. Formulations of antifouling paints consist of chemical biocides; these biocides could be steadfastly leaked into the marine environment. Chemical biocides are of extreme toxicity affecting a broad range of aquatic ecosystems with intense damages to the environment.

Formulating TBT paints evolved from simple contact by surface leaching to a polymer base where the rate of discharging biocide is regulated by its reaction with water. Paints with tributyltin formula were quite long lasting and effective. By the end of the 1970s, tributyltin-based paints were widely applied on recreational and commercial sea ships **(Hartwell *et al.*, 2016)**. By the 1980s, it was established that they were found to cause endocrine disorders in marine organisms, flagging them for a threat to the marine environment **(Alzieu *et al.*, 1986)**. Because of these effects, female organisms superimposed male sex organs, causing pseudohermaphroditism or imposex, a reproductive disturbance. This issue was shown to dominate when TBT concentrations where greater than 1 ng/l **(Alzieu, 1998; Hoch, 2001)**. Therefore, the decline of affected species, e.g., *Nucella* and *Hinia*, declined in terms of presence as a result. Additionally, TBT showed extensive accumulation in marine organisms, mainly because of its lipophilic chemical nature, sometimes reaching up to 5 μg/g in molluscs. **(Iwata *et al.*, 1995)** as well as in birds **(Hoch, 2001)**.

Hence, France established the first legislative and regulatory actions to manage tributyltin in 1982, followed by the UK in 1985 **(Terlizzi *et al.*, 2001)**. However, the advent of the International Convention on the Control of Harmful Anti-fouling Systems on Ships (the AFS Convention) (IMO, 2001) which was signed in 2003 but effected since January 2008 where initiated the ban of any OTC entirely. The food and Agriculture Organization of the United Nations (FAO) and United Nations Environment Program (UNEP) enlisted the pesticide TBT on a global trade "watch list" following the Rotterdam Convention in November 2008. And yet extensive OTCs' concentrations persist in marine sediment **(Lofrano *et al.*, 2016)**. This extensive use extremely contaminated the

environment, raising concerns about their toxicity and influence on marine communities for the long run.

The AFS Convention was a reaction to a multitude of concerning accounts on the disruption of endocrine and toxic influence of Organotins (OTs), mainly tributyltin (TBT), on aquatic life **(Hoch, 2001)**. In 2008, The Convention concluded with a total ban on applying organotin compounds based antifouling paints was applied.

Recent literature reports illustrate that the problem with tributyltin persists in various marine areas **(Filipkowska *et al.*, 2016; Egardt *et al.*, 2017; Abraham *et al.*, 2017; Castro *et al.*, 2018)** despite the cease of its release by paints from vessels. Following the total ban, it is surmised that TBT's main source in aquatic life is from harmful OTC-deposited sediments. Due to TBT's hydrophobic characteristics and its tenure as an active ingredient in antifouling coatings, substantial levels of it accumulated throughout the world in bottom sediments **(Berto *et al.*, 2007; Filipkowska *et al.*, 2011)**. Concentrated spots are mostly harbors, ports, shipyards, and regions related to extensive marine traffic.

It is problematic to gauge the extent of tributyltin degradation rate in the aquatic environment since it could be effected by a multitude of chemical, biological, or photochemical processes **(Hoch, 2001; Said *et al.*, 2006)**. Moreover, environmental conditions play an important factor in assessing TBT degradation besides its initial concentration in the medium **(Hoch, 2001; Shreadah *et al.*, 2006; Cruz *et al.*, 2007; Filipkowska *et al.*, 2014)**. In sediments, TBT's half-life roughly ranges from months to several years while it could extend to decades in anaerobic sediments **(Matthiessien 2013)**.

Therefore, high concentrations may remain in older buried sediments that caused spontaneous releases in the past. Whether this material becomes bioavailable depends on the deposition rates of local sediment and following disturbance; this is sometimes known as dredging.

Therefore, we aim to gauge the remaining TBT contamination and products of its degradation products, Dibutyltin (DBT), and Monobutyltin (MBT) that endure in the Egyptian Red Sea Coast sediments after implementation of AFS Convention to confirm the load of organotin compounds remains deposited in the area of study's sediments.

MATERIALS AND METHODS

2.1. The Area of Study

The Red Sea is a narrow-shaped, elongated sea, spanning from the north to south over approximately 2,000 km. It extends from 22^{O} 15 to 27^{O} 15 N and 33^{O} 50 to 37^{O} 50 E. The sea's average width is about 280 km, maxing to 306 km in the

south. Bal-el Mandeb strait's width is roughly 26 km and separates the Red Sea from the outer Indian Ocean through the Gulf of Aden. The Red Sea's northern tip diverges into two gulfs: Aqaba Gulf, and the Gulf of Suez, interconnected with the Suez Canal. The Sea is adjacent to a mostly arid landmass, of desert or semi-desert regions lacking any dominant river inflow. Moreover inland, the desert regions are extensively bordered by mountain ranges **(UNEP, 1997 and PERSGA, 2000)**. The Red Sea is situated in an extremely arid region. In summer, its sever hot weather is considered to be among the hottest in the world, especially the southern region. Unlike the northern region, the air temperature is scarcely lower than the southern region. The deepest region lies between 14 N and 28 N, with 2,920 m as the foremost depth in the areas. These deep areas remain active geologically and with numerous volcanic vents exuding heated, metal-rich, and salty seawater. The two gulfs in the northern region maintain the same shape but differ in their topography. On the other hand, the Gulf of Aqaba consists of a narrow shelf and a deep basin, the latter is divided into two by a submarine sill; both basins are over 1,000 m deep **(UNEP, 1997)**. The Red Sea's Egyptian side seems to be affected by the swift and surging extents of civilization because of industrial (broadly shipping and phosphate), recreational (swimming and diving), and fishing activities. Over the last 35 years, the coasts of the Egyptian Red Sea witnessed extensive touristic developments**.**

Therefore, the substantial threats to the environment of the Egyptian Red Sea coast are urbanization and coastal development, industries activities and tourism activities, shipyards, marine traffic, wastewater treatment facilities, quarrying activities, coastal mining, oil bunkering, and the offshore drilling process.

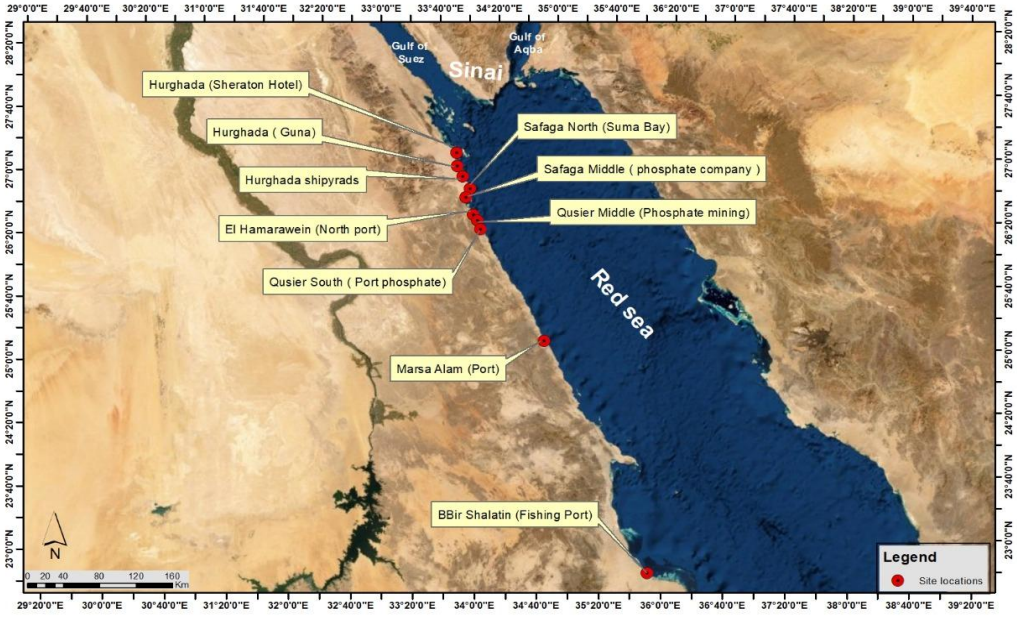

Fig.1. A map of Egyptian Red Sea coast showing sampling locations of sediment

2.2. Sediment Samples

Investigated sediment samples were collected from ten sites according to the proximity of anthropogenic activities verified by a Hydro-Bios stainless-steel grab sampler during winter and summer 2018 (Figure 1).

At each station, three subsamples were taken and were mixed, yielding a composite sample. This sample was kept frozen at -20^{o}C until analysis.

The content of Organic Matter (OM) in sediments was measured by the wet oxidation method with potassium dichromate and concentrated H_2SO_4 of the sample, followed by the titration of excess dichromate with 0.5 N ferrous ammonium sulfate solution to a sharp, 1 drop, end point. Oxidation of C1- can be prevented by the use of Ag_2SO_4 in the digestion mixture according to Loring and Rantala (1992). Total carbonate was determined using the titration method according to Black (1965). Sediments' total tin (Sn) were measured according to APHA (1992) and the concentrations of total tin were measured via an atomic absorption spectrophotometer (AAS, Model AA-6800 Shimadzu). Butyltin derivatives MBT, DBT, and TBT were measured as per Hodge, *et al.,* (1979). Finally, the solution of the sample was administrated into an electron capture detector gas chromatograph, Model HP 5890 II.

Validating the Method and samples' quality control were applied via standard solutions and the measurements were performed based on ISO 17025. Two natural samples were evaluated in pairs in each of the six batches of samples after being spiked by a specific concentration from the standard solution. The samples were also evaluated without spiking. The highest and lowest percentages of recovery for spiked samples were the benchmark for accuracy, ranging between 90 and 105%. The precision parameter was set to be within 10%.

2.3. Degradation Index Calculation

By using the Butyltin Degradation Index (BDI) organotin compounds can be estimated to be old or fresh **(Burton, *et al.*, 2005)**. These following equations were estimated according to the ratios of concentration between two main degradation products (DBT and MBT) from the parent compound; it was assumed that the rates of degradation between the two compounds similar **(Diez, *et al.*, 2002)**. BDI values exceeding one denotes that the input of TBT in the sediment is old, while BDI values below one indicates fresh TBT input **(Filipkowska, *et al.*, 2011).**

$$BDI = \frac{[MBT] + [DBT]}{[TBT]} \quad (1)$$

RESULTS AND DISCUSSION

3.1 Characteristics of Sediments

Physicochemical parameters of sediments such as organic matter (OM), particle size, and $CaCO_3$ are the primary factors affecting the spatial distribution of metals and organometal compounds **(Soliman *et al.*, 2019)**. The organic matter and grain size have been pointed out as to significantly affect mobility, destination, and availability of organotin compounds in coastal marine ecosystems. **(Pinochet *et al.*, 2009; Quintas *et al.*, 2016**).

The results of surface sediments' geochemical characteristics from the investigated area are illustrated in Table 1. The amount of carbonate in the sediments ranged from 27.5% (site VI in summer) to 68.0% (site IX in summer) with an average value of 48.19 and 47.85% in summer and winter respectively. Following Maxwell classification in 1968, the majority of sediment sampling sites contain low terrigenous materials of a carbonate range of > 40% (around 70% of all investigated sediments) (Table 1), except sites II, VI, and X. The lowest percentage of carbonate was found in station VI in summer.

The distribution of grain size (sand, clay, and silt) in the samples of sediment is presented in Table 1. Concerning the grain size fractions in this study, most of the stations were classified as sandy sediments with no significant difference between the seasons**.** The particles of sand were found to be a dominant fraction (81.4–100%) followed by silt (0–13.2%), and clay (0-5.4%). Table 1 denotes that high OM content matches the presence of clay content for all investigated sediments. The contents of organic matter were from 0.15 (site IX in winter) to 1.33% (site VI in summer), with a mean value of 0.55% in summer and 0.48% in winter, the highest percentage of sediments was obtained from site VI.

According to Pearson's correlation, OM and grain size were directly proportional to OTC ($P < 0.05$), denoting that distribution of OTC was affected by the size of the grain and content of organic matter in the study area.

3.2 Butyltin Compounds (BTS) and Total Sn (TSn)

The distribution of BTs and total Sn (TSn) of the area of study during 2018 is shown in Table 2. The concentrations of TBT, DBT, MBT, ΣBT, and TSn varied between 7–220, 3.5–48.8, 4.2–78, 14.7–339.8 ng/g, and 0.09–1.15µg/g, respectively (Table 2). The concentrations of BTs and TSn exhibited a distinct temporal distribution. The highest of their concentrations were recorded in summer. TBT was the most abundant (52.79%–53.13% in winter and summer, respectively) while the averages of the percentage of DBT were 18.45% and 19.33% in winter and summer, respectively. Whereas MBT has the averages of percentages in winter and summer to be 27.88% and 28.42%, respectively (Figure 2).

The butyltin derivatives' concentrations (DBT, TBT, MBT) were present in all collected samples of sediments from the area of study and the lowest levels were recorded in site IX (Marsa Alam) because of their quite low concentrations implying the absence of fresh inputs of the derivatives into the area, unlike the higher butyltin derivatives' concentrations (BT, including DBT, TBT, and MBT) and total tin were found at station VI (El Hamarawein, North Port) during summer season compared with winter (Table 2). This may be due to the exponential surge in tourism activities and heavy boating traffic in this port, especially during summer. The lower levels of BTs in all sites during winter could be because of the contaminated sediments' resuspension phenomena.

A significant positive correlation was found between butyltin derivatives and organic matter ($p < 0.05$). From a spatial perspective, concentrations of OM and TBT in the area of study's surface sediment showed peculiar and similar trends. Distribution, transport, and contaminants' presence or lack of it in coastal ecosystems are majorly affected by OM (**Rubio *et al.*, 2000; Younis, 2018**).

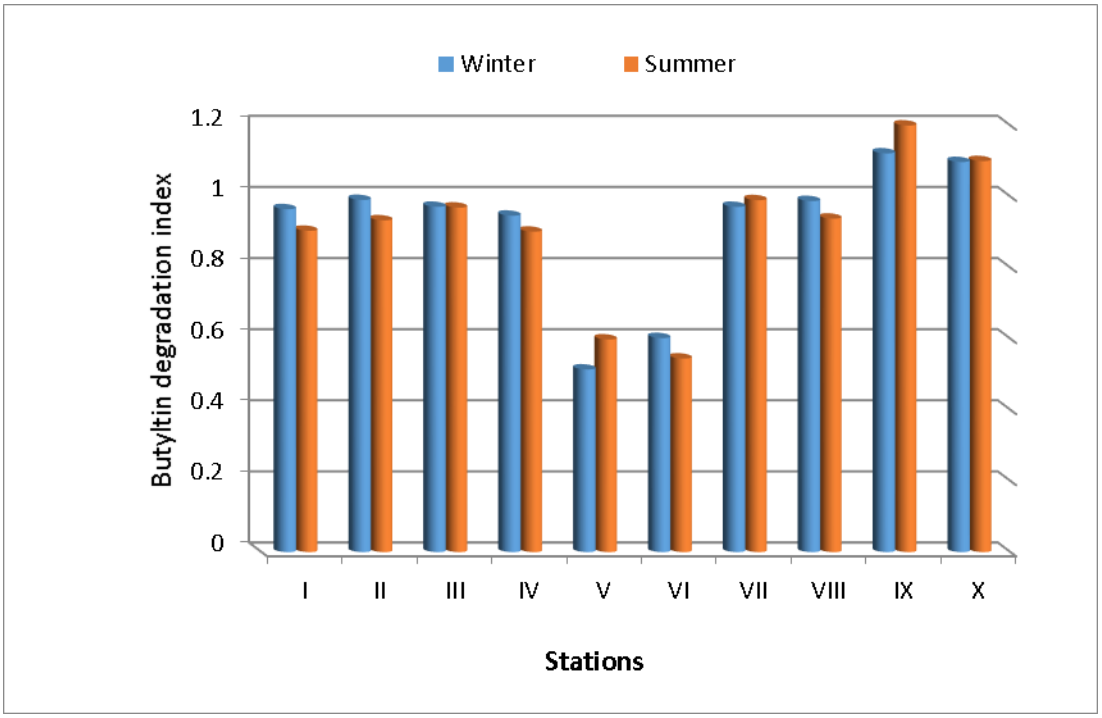

Fig. 2. Butyltin degradation index (BDI) values of the investigated sediments

Table 1. Description of the sampling stations, grain size, $CaCO_3$, WC and TOM (%) in sediments of Red Sea coast of Egypt during winter and summer 2018.

Station No.	Station name	Latitude and longitude	Summer						Winter						Sediment type
			Sand	Silt	Clay	$CaCO_3$	WC	TOM	Sand	Silt	Clay	$CaCO_3$	WC	TOM	
			%						%						
I	Hurghada (Sheraton Hotel)	27° 1.0' 37.5'' 33°50' 48.4''	100	0.0	0.0	66.5	9.5	0.16	100	0.0	0	64.5	10.2	0.17	Sand (very coarse sand)
II	Hurgada (Guna)	26° 55' 30.0'' 33°54' 20.0''	90	6.7	3.3	33.2	17.3	0.75	92	7.2	0.8	30.2	15.4	0.72	Sand (medium sand)
III	Hurghada shipyards	27° 50' 47.0'' 33°59' 35.0''	92.1	6.8	1.1	46.3	16.5	0.66	90.1	5.8	4.1	48.3	17.3	0.60	Sand (medium sand)
IV	Safaga North (Suma Bay)	26° 47' 34.9'' 33° 56' 12.5''	100	0.00	0.0	58.5	8.4	0.21	98.0	2.0	0	59.4	9.5	0.19	Sand (very coarse sand)
V	Safaga Middle (Phosphate Company)	26° 30' 20'' 34° 0.0' 20''	99.2	0.8	0.0	50.8	11.2	0.38	97.3	0.9	1.8	52.7	10.2	0.23	Sand (fine sand)
VI	El Hamarawein (North Port)	26° 15' 9.0'' 34° 12' 5.0''	85	11.3	4.2	27.5	22.5	1.33	81.4	13.2	5.4	28.3	20.1	1.2	Silty sand (medium sand)
VII	Qusier Middle (Phosphate mining)	26° 8.0' 30'' 34° 14' 30''	98.2	1.8	0.0	46.6	10.5	0.55	97.3	1.5	1.2	45.6	12.5	0.45	Sand (fine sand)
VIII	Qusier South (Port phosphate)	25° 55' 48'' 34° 36' 36''	98	1.7	0.3	45.8	13.5	0.60	95.3	2.1	2.6	47.1	11.1	0.54	Sand (fine sand)
IX	Marsa Alam (Port)	25° 4.0 ' 6.1'' 34° 45' 0 .4''	100	0.00	0.0	68.0	8.9	0.15	100	0.00	0	65.2	9.5	0.15	Sand (very coarse sand)
X	BBir Shalatin (Fishing Port)	23° 9.0' 9.9'' 35° 36' 48.3''	97.3	2.7	0.0	38.7	12.0	0.68	98.2	2.5	0.7	37.2	10.7	0.55	Sand (medium sand)

In broad literature, results of contradictory implications were reported about the relationship between organotin level and the contents of organic matter. For example, Pinochet *et al.* (2009) showed an optimal correlation between organic matter content, DBT, and TBT levels in marine sediments from San Vicente Bay in Chile. Even though this barely anticipated behavior can be related to the inert complexity of the physicochemical sediment, e.g., the varying organic matter composition, the existence of biological activity (bioturbation) in the upper sediment layers, and relative adsorbability on inorganic particles (Braga Castro et al. 2012). Furthermore, multiple sources affecting the same ecosystem may impose a "masking effect", which means biasing a potential relationship.

Organometals are linked with organic matter in the fine-grained sediment fraction, e.g., colloids, organic-material coating particles, and detritus **(Bryan *et al.*, 1989**) because of the high specific surface area of organic matter content. This enrichment is a result of surface adsorption and ionic-attraction binding to organic matter **(Younis *et al.*, 2014; Soliman *et al.*, 2018)**.

Therefore, sediments containing high organic matter content could be traps for organotin more than to sediments without organic matter. Adsorption of TBT by OM depends on the mineral constituents of the sediment and the pH value.

As illustrated above, organotin deposition depends on the content of the organic matter of the sediment. As a result, sampling site IX showed low organic matter content (0.15%); lower organotin was found (Table 2). On the other hand, site VI maintained higher organic matter content (1.2%) and higher organotin load was observed as a result.

Also, a significant positive correlation was found between butyltin derivatives and grain size ($p < 0.05$). At station VI (El Hamarawein, North Port) where the sediment sample at this station has the lowest percentage of sand and the highest percentage of silt among all investigated stations of the area of study, the concentrations of butyltin derivatives were of the highest values at this station. The homogeneity of sediment is an important factor in the occurrence of coastal features.

Fine-grained sediments provide a stable environment for retaining contaminants. Contaminants' loss and remobilization from fine-grained sediment correlated with in situ physical traits, e.g., marine influx with tides and turbulence related to adverse weather conditions **(Miles and Tome, 1997)**.

Table 2. Levels of TBT, DBT, MBT, ΣBTs (ng Sn g-1 d.w.) and total Tin (µg Sn g-1 d.w.) in surface sediments of Red Sea coast of Egypt during winter and summer 2018.

Seasons	Summer					Winter				
stations	TBT	DBT	MBT	ΣBTs	Total Tin µg/g	TBT	DBT	MBT	ΣBTs	Total Tin µg/g
	ng/g					ng/g				
I	52	18	29	99	0.61	50.88	19.6	29.48	99.96	0.52
II	45	18	24	87	0.53	42.5	14.7	27.4	84.6	0.48
III	65	30	33	128	0.64	60.8	28.22	30.84	119.86	0.62
IV	140	48.2	78	266.2	0.84	101	45.88	49.7	196.58	0.72
V	160	32.7	63	255.7	0.85	120	29.3	32.35	181.65	0.72
VI	220	48.8	71	339.8	1.15	180	39.73	68.65	288.38	0.90
VII	60	19.4	40	119.4	0.58	44	15.4	27.33	86.73	0.43
VIII	71	24.6	42	137.6	0.74	52	19.9	31.45	103.35	0.64
IX	8	3.7	5.9	17.6	0.09	7.4	3.5	4.8	15.7	0.09
X	7	3.5	4.2	14.7	0.11	7.2	3.5	4.4	15.1	0.09
Min.	7	3.5	4.2	14.7	0.09	7.2	3.5	4.4	15.1	0.09
Max.	220	48.8	78	339.8	1.15	180	45.88	68.65	288.38	0.90
Average	85.8	25.59	40.51	151.9	0.761	67.478	21.973	31.44	120.891	0.536

Contamination patterns of butyltin derivatives in the sediments of the area of study were TBT > MBT > DBT (Table 2). Hajisamoh (2013) showed that the extensive contamination caused by TBT alongside Southern Thailand coastal areas is likely caused by antifouling paints applied on seafaring vessels' hulls and medium to large fishing boats. On the contrary, water transportation contamination is not majorly affecting the coasts. The TBT ban in antifouling paints intensely inhibited the contamination by butyltins in seawater, decreasing the already small amounts of butyltins in the sediment, yet continuous monitoring of butyltins is needed for surveillance of other sources.

MBT was ranked second after TBT in the investigated sampling sites of the area of study. Hence, following the intense local anthropogenic effect the contamination caused by MBT on those sites might be incepted from their usage in industrial materials of biocides, e.g., large-scale use of PVC, containing organotin derivatives as stabilizers **(De Carvalho and Santelli, 2010)**.

The Butyltin Degradation Index (BDI) values of organotin compounds were recorded in Figure 3. All investigated samples had BDI values around one except sites V and VI (BDI value < 1). This implies that TBT degradation (debutylation) to dibutyltin and monobutyltin and finally tin is perpetual for all compounds. This would imply that no new TBT input besides high-degree TBT degradation in most of the investigated sediments. TBT compounds in surface sediments have a long half-life that ranges between 160 to 775 days prior to the compound's production of other butyltin products: DBT and MBT**(Amouroux *et al.*, 2000).** Both butyltin products are created by the aerobic biological process but with less oxygen conditioning, mostly in deeper sediments, where the degradation slows.

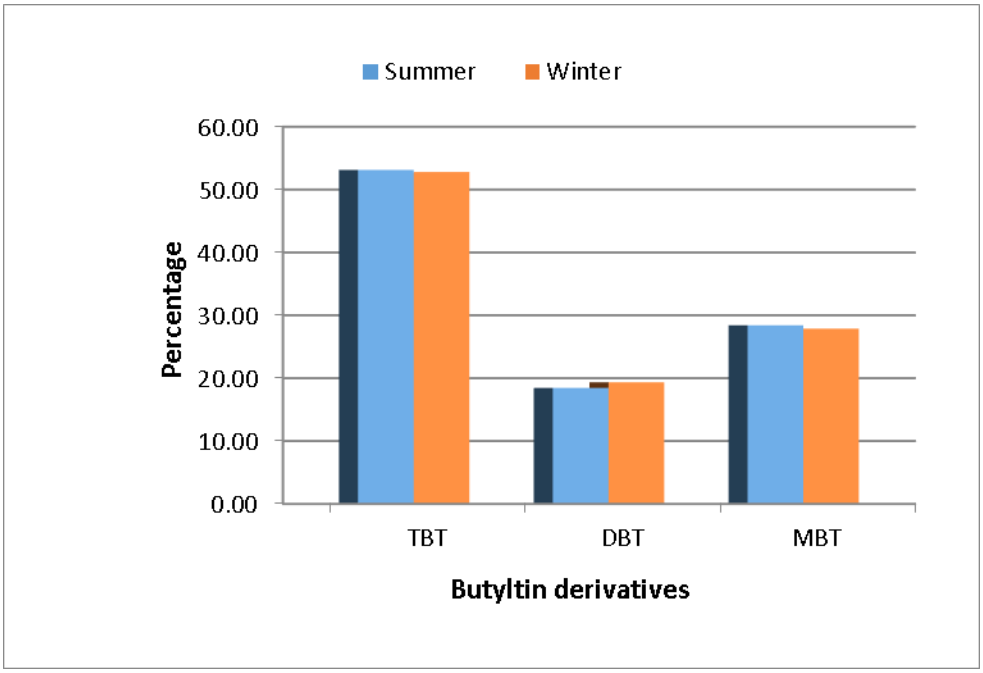

Fig. 3. Mean percentage of butyltin derivatives of the sediments from the Red Sea coast of Egypt

3.3 Comparison the levels of butyltins with the global scales

Globally, after being banned for ten years, a major reduction of TBT concentration was found in seawater and biota, but sediments remain with no significant variance. Hence, the concentration of TBT continues to exceed global environmental quality control **(Kim *et al.*, 2014)**.

Because of the great importance of the Red Sea at the global level in terms of biological diversity, protected areas, tourism, and maritime transport between countries of the world, Egypt finds great interest in implementing the

International Convention on the Control of Harmful Anti-fouling Systems in ships to protect and conserve biological diversity in the Red Sea.

Although data on sediment's organotin compounds contamination are relatively limited in Egypt, a slight increase in the levels of butyltins, measured before the restriction on butyltins antifouling paints in Egypt, is found when comparing our results with those reported for collected sediments in 1988 from lake Mariut, Egypt **(Aboul Dahab *et al.*, 1990)** and the Egyptian Mediterranean coast of Alexandria **(Shreadah *et al.*, 2006)** as shown in Table (3). In countries where tri-organotin antifouling paints have been regulated, a similar trend was found. In Canada, St. Lawrence Estuary's sediments with levels of butyltins ranged from 3 to 6 ng Sn g^{-1} d.w. **(Michaud and Pelletier, 2006)**, San Francisco Bay 's sediments in USA with the levels of tri-organotin between 0.3 and 3.6 ng Sn g^{-1} d.w. **(Pereira *et al.*, 1999)** and Townsville and Cairns marina's sediments within the Great Barrier Reef World Heritage Area, Australia with the concentration of 4 ng Sn g^{-1} and 5 ng Sn g^{-1} respectively **(Haynes and Loong 2002)** are among the least contaminated reported sediments by tri-organotin for coastal zones worldwide.

Similar trend was found in other countries where tri-organotins antifouling paints were restricted, contamination diminished gradually, these countries were among the following: the Gulf of Oman (<0.01–60 ng Sn g^{-1}) **(De Mora *et al.*, 2003)**, Atlantic Ocean coast of Portugal (<3.8–12.4 ng Sn g^{-1}) (Diez and Bayona, 2009) and Øresund strait between Sweden and Denmark (1–19 ng Sn g^{-1}) **(Strand *et al.*, 2003)** which appears to be lower than the compared results of the present study.

On the other hand, some Asian countries have no specific legislation regulating the application of tri-organotins. In these countries, high levels of tri-organotins contamination are still found in sediments. Taiwan coastal areas were reported to be extremely contaminated by organotins that are highly concentrated with tri-organotins up to 25,300 ng Sn g^{-1} ww. in marine sediment collected from the Kaohsiung's harbor area **(Jang 2004)**, as well as in the estuary area of Love River's estuary area in Kaohsiung, where TBT's concentration amounts to 30,222 ng g^{-1} ww. **(Chi 2004)**.

By comparing the results of TBT of the present study with the work carried out by Harino *et al.*, (2012) in Southeast Asia, we deduced that higher of TBT concentrations were evident in sediments found in Malaysia and Thailand, especially in the coastal areas. The utmost concentration value of TBT amounted to 1246 ng g^{-1} dw which was found in industrial area sediments; this area is in close proximity to shipyards located in Thailand. This finding resonated with Kan-Atireklap's study in 1997 in Thailand where high concentrations of TBT in

the marine sediment, sometimes amounting to 4500 ng g^{-1} dw. Unlike organotins concentrations in sediments in Hong Kong which fluctuated over time but maintained a diminishing temporal trend. The concentration of TBT in sediment was as high as 1,690 ng g^{-1} dw in the early 1990s **(Lau 1991)**. However, Cheung et al. (2003) found concentrations of TBT amounting to 560 ng g^{-1} dw in sediments from Yam O. but still exceeds those of our results. In Jinhae Bay in South Korea, it was found that the concentration of TBT in biota and the water column diminished after actions to deter antifouling uses of TBT; however, sediment concentrations remain the same, statistically, ten years after the ban **(Kim *et al.*, 2014)**.

Conditions similar considering the limited data on contamination of organotins in sediments and results when comparing the current study with prior studies in China. Zhang *et al.* (2003) that gauged concentrations of tri-organotins in the sediments acquired from Pearl River Delta, where concentrations ranged from 1.7 to 379.7 ng g^{-1} dw, lower than amounts found in other parts of the world while similar with the results of the current work. The peak of concentration values of tri-organotins was found in locations close to harbors and shipyards. From Pearl River to the estuary, there was a downward-sloping gradient of tri-organotins concentrations; this implies that tributyltin is sourced from sewage discharges and extensive riverine shipping activities; there was a dilution effect induced by seawater to the contaminants in the estuary. Huang *et al.* (2005) assessed organotins' concentrations in the sediments collected from three harbors along the coast of Southeastern China where tri-organotins were found to be within 0.3 and 174.7 ng g^{-1} dw.

In Japan, high levels of organotins in marine systems have not only been correlated to activities related to recreational and commercial boating or dry docks and facilities conducting vessel repair but also with antifouling paints used in cages and fishnets in aquaculture (**Takeuchi et al., 2004**). Even after 7–9 years after the partial ban on TBT (initiated in 1990), butyltins are still widely traceable along the Japanese coast, including international ports and unpopulated areas. High levels of organotins, compared with the current study's, were reported by Harino *et al.,* (2007) in sediments collected from Otsuchi Bay, Japan with butyltins levels ranging from 1 to 14,000 ng g^{-1} for TBT, 2 - 3.400 ng g^{-1} for DBT and 1 - 3.300 ng g^{-1} for MBT.

Table 3. Comparison of the present study results with previous works

Location	TBT	DBT	MBT	References
	Concentration range (ng Sn g^{-1})			
Red Sea coast, Egypt	7-220	3.5-48.8	4.2-78	Present study
Marinas and harbors of Stockholm, Sweden	10–1300	25–1400	10–990	Eklund et al., 2010
Barcelona harbor, Spain	9 8–4702	67–2607	35–440	Diez et al., 2006
Santos-Sa˜o Vicente Estuarine, Brazil	13.72–159.2	14.26–5.66	9.98–22.48	Buruaem et al., 2013
Bahı´a Blanca Estuary, Argentina	7.68–36.75	14.32–56.9	31.05–292.62	Pamela et al., 2016
Marinas and Ports of Mediterranean Sea coast of France	37–4402	34–3025	65–3682	Cassi et al., 2008
lake Mariut, Egypt	35-975	10-305	ND-330	Aboul Dahab et al., 1990
Egyptian Mediterranean coast of Alexandria	3-1600	1-330	-	Shreadah et al., 2006
St. Lawrence Estuary's sediments, Canada	3 - 6			(Michaud and Pelletier, 2006)
San Francisco Bay 's sediments, USA	0.3 - 3.6	0.6 – 2.5	0.4 – 2.0	Pereira et al., 1999
Townsville and Cairns marinas, Great Barrier Reef, Australia	4.2-5.5	<1	<1 - 33	Haynes and Loong 2002
AGulf of Oman	<0.1–60	<0.1 – 2.0	<0.2 – 9.7	De Mora et al., 2003
Atlantic Ocean coast of Portugal	<3.8–12.4	<5.3–65	<5.2–78	Diez and Bayona, 2009
Øresund strait between Sweden and Denmark	1–18.8	-	-	(Strand et al., 2003)
Kaohsiung's harbor area, Taiwan	25,300	-	-	Jang, 2004
Love River, Kaohsiung, Taiwan	30,222	-	-	Chi, 2004
Shipyards, Thailand	1.6 -1246	0.7 -368	1 -293	Harino et al., 2012
Mariculture sites, marinas and typhoon shelters, Hong Kong.	14 - 1,690	-	-	Lau, 1991
Yam O., Hong Kong	<0.1 - 560	-	-	Cheung et al., 2003
Coastal areas of Thailand	7 - 410	2 - 1900	4 - 4500	Kan-Atireklap, 1997
Pearl River Delta, China	1.7 -379.7	8.5 -143.3	ND-62.6	Zhang et al., 2003
Southeast coast of China	0.3 -174.7	-	-	Huang et al., 2005
Otsuchi Bay, Japan	1 - 14000	2 - 3400	1 - 3300	Harino et al., 2007
Spanish North-western coast	124-18722	92-6860	7-1674	Diez et al., 2002
Spanish South-western coast	200- 3868	59- 813	86- 785	Diez et al., 2002
Argentinian Atlantic Ocean coast	3288	1645	-	Delucchi et al., 2007
Gipuzkoa, Spain	50-5480	150-710	860-2870	Arambarri et al., 2003
Southern Venice lagoon, Italy	21 – 39300	2 – 12623	-	Berto et al., 2007

German Baltic Sea and North Sea marinas	80–17,000	30–14,000	10–1300	Biselli et al., 2000
Gdańsk Port, Baltic Sea, Poland	1066 – 16400	1 020 – 21 420	816 – 31 280	Senthilkumar et al., 1999
Gdynia Port, Poland	13 – 15775	9 – 2060	7 – 68	Filipkowska et al., 2011
Gdynia Port, Poland	1 143 – 6 408	250 – 2716	134 – 968	Radke et al., 2012
Commercial harbours, Great Barrier Reef, Australia.	7 500 – 340 000	660 – 32 000	660 – 61 000	Haynes and Loong 2002

Moreover, Arp *et al.* (2014) conducted a study assessing whether levels of tributyltin in water and sediments in the Drammensfjord (Norway) diminished by time (2005–2013) due to natural recovery showed a substantial drop in the average values of TBT concentrations from 2005 to 2008 and moderate diminishes in the port area since 2008 **(Arp *et al.*, 2014)**. Levels of tributyltin in water and particularly sediments located away from the major site of legacy tributyltin emission seemed to diminish at a much slower rate **(Arp *et al.*, 2014)**. This indicates that even though the efforts to limit the tributyltin usage on and in ships may effectively decrease water and biota concentrations, sediments are still a reservoir for future tri-organotins contamination. Therefore, TBT concentration in sediments from the Spanish south eastern and north eastern coasts were 200- 3868 ng g^{-1} d.w. and 124-18 722 ng g^{-1} d.w. respectively **(Diez *et al.*, 2002)**, from Port of Belgrano, Argentinian Atlantic Ocean coast were 3288 ng g^{-1} d.w. **(Delucchi *et al.*, 2007)**, from river estuaries of Gipuzkoa, North Spain 50–5480 ng g^{-1} d.w. **(Arambarri *et al.*, 2003)**, from Southern Venice lagoon in Italy 21–39300 ng g^{-1} d.w. **(Berto *et al.*, 2007)**, from the German Baltic Sea and North Sea marinas 80 – 17000 ng g^{-1} d.w. **(Biselli *et al.*, 2002)**, from sediments of the Baltic Sea in Poland, Port of Gdańsk in 1999 (1066–16400 ng g^{-1} d.w.) by Senthilkumar *et al.*, (1999), and after 12 years, TBT concentration did not decrease considering the results obtained by Filipkowska et al., 2011 where TBT levels were within 13 and 15,7759 ng g^{-1} d.w.. A similar trend was deduced in Port of Gdynia sediments (1143–6408 ng g^{-1} d.w.) by Radke et al., (2012) which are still surpassing those of the current study.

It is still possible to detect butyltins in surface sediments and hermit crabs even after the international and Brazilian bans, from 7 to 1304 ng Sn g^{-1} in sediments **(Sant'Anna *et al.*, 2014)**. Alternatively, the TBT levels recorded in the gastropod Hexaplex trunculus reduced over time in Bizerta channel, northern

Tunisia, rendering the compound less frequent among butyltins **(Lahbib *et al.*, 2011)**. An earlier study demonstrated higher concentrations of butyltins in the United Kingdom where concentrations ranged from 46.2 to 3935 ng Sn g^{-1} **(Dowson *et al.*, 1992)**, 34 to 4402 ng Sn g^{-1} in marinas and ports of Mediterranean Sea coast of France **(Cassi *et al.*, 2008)**, 25 to 1400 ng Sn g^{-1} in Stockholm harbors and Marinas, Sweden **(Eklund *et al.*, 2010)**, 35 to 4702 ng Sn g^{-1} in Barcelona harbor, Spain (Diez et al., 2006), 100 to 780 ng Sn g^{-1} in Slovenia **(Scancar *et al.*, 2007)**, and 7.68 to 292.62 ng Sn g^{-1} in Bahı´a Blanca Estuary, Argentina **(Quintas *et al.*, 2016).**

The present study's were almost similar with the results of butyltins recorded in Santos-Sa˜o Vicente Estuarine, Brazil **(Buruaem *et al.*, 2013)** and Mzoughi et al. (2005) for sediments collected from North Africa, specifically from sediments of Bizerte Lagoon, Tunisian the Mediterranean Sea, while the results of the present work were lower than sediments of commercial harbours of the Great Barrier Reef World Heritage Area, Australia **(Haynes and Loong, 2002).**

We can conclude that the decreasing of organotin levels is not significantly observable in costal sediments of some areas around the world after the regulations to restrict the usage of TBT as antifouling paints. Organotin are very persistent in sediments and are stored for years up to a decade or longer and still need continuous monitoring especially in the critical areas of maritime activities.

International regulations implementation might be effective enough to curb organotin contamination widespread in certain areas. However, coastal areas managements need to contend with these unsolved problems in the future.

According to Dowson *et al.,* (1993), investigated sediments from all stations were ranked, according to the classification proposed, as highly contaminated with TBT, except sites IX and X, established to be of low TBT contamination (lightly contamination) (Figure 4). As per the HELCOM recommendation, the threshold level for Good Environmental Status (GES) of sediments' TBT is 1.6 μg kg−1 d.w., i.e., 0.66 ng Sn g−1 d.w. Yet, this is merely a value for the test's threshold according to the sediment of 5% of total organic carbon **(HELCOM, 2018)**. The priorly recommended GES TBT boundary in sediments was 0.02 μg kg−1 d.w., i.e., 0.008 ng Sn g−1 d.w. **(HELCOM, 2012)**. These values are very low for thresholds concerning the concentration of TBT as exhibited in this study.

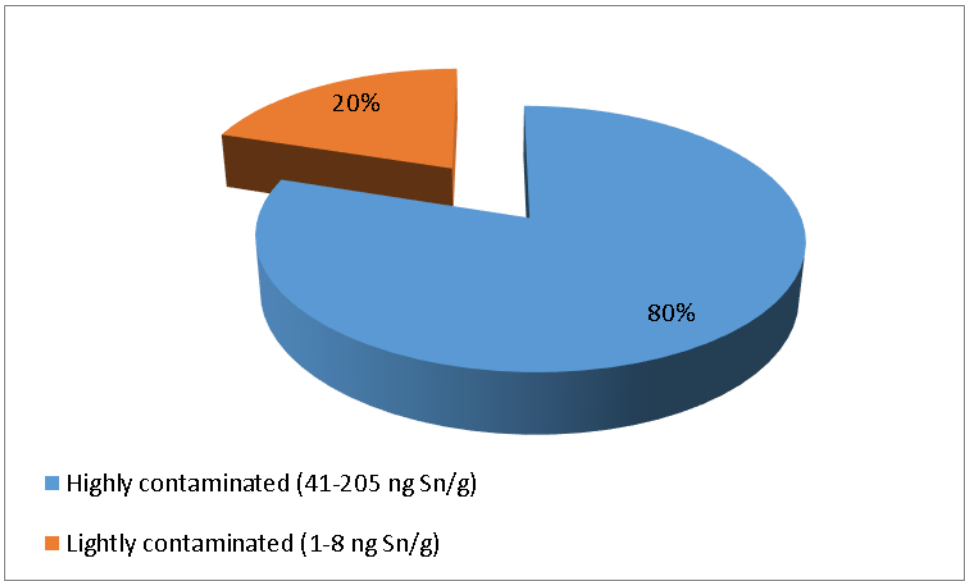

Fig. 4. Investigated sediments classification based on tributyltin levels according to Dowson et al. (1993).

Ten years following the total ban on TBT in antifouling paints, concerns in the area of study remain because of the high levels of TBT sediments. Moreover, the TBT percentage in the total sum of BTs is high, indicating that the process of degradation of TBT is slow and continuous. Additionally, butyltins might have been used illegally in recent years. In the need, the results above necessitate more accurate data on compounds if butyltins in the area of study in the next years, since the samples presented were beyond the GES threshold for TBT in sediments.

CONCLUSION

In conclusion, the highest concentrations of butyltin contamination in sediments of the Egyptian Red Sea Coast were found at station VI (El Hamarawein, North Port), implying the occurrence of alternative causatives of pollution by butyltin in the El Hamarawein, North Port, besides the leaching of antifouling paints from boats and ships. TBT various levels ranked the sampling sites classification to rank from lightly to highly contaminated, and a higher income of these pollutants was recorded during summer.

Contamination patterns of derivatives of butyltin in the sediments of the area of study were TBT > MBT > DBT and most of the investigated samples had BDI values around one which proves no new addition of TBT and a high extent of TBT degradation. Butyltin compounds concentration differed broadly alongside the coast, but the major composition of the butyltins was TBT. As per the HELCOM recommendation, investigated sediments of all sites surpassed the

Good Environmental Status boundary for tributyltin in sediments. However, general organotin levels were lower than those previously reported in different areas.

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