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Spatiotemporal variation and source apportionment of organotin compounds in sediments in the Yangtze Estuary

Chunzhao Chen[1,2], Ling Chen[2,3], Rui Xue[3], Qinghui Huang[1,3], Lingling Wu[1,2*], Shufeng Ye[4] and Wen Zhang[5]

Abstract

Background: The Yangtze Estuary is a vital habitat and breeding ground for many rare species (e.g., Chinese sturgeon, Chinese paddlefish). From the view of marine ecosystem health, organotin compounds (OTCs) should be of great concern in this area based on their broad applications and potential threats. At present study, five OTC species were determined in 28 and 26 surface sediments from the Yangtze Estuary (including the Nearshore area, Estuarine Turbidity Maxima (ETM) zone and Plume zone) in 2015 and 2016, respectively. Meanwhile, four sludge samples were collected from the adjacent Shidongkou Wastewater Treatment Plant to perform a source apportionment.

Results: The sum of OTCs presented a decreasing trend towards the open sea, averaging 4.8 ± 6.0 ng Sn g^{-1} dw in the Plume zone. While OTC levels in the Nearshore area (2.9–34.6 ng Sn g^{-1} dw) were similar with those in the ETM zone (5.6–36.0 ng Sn g^{-1} dw), and the sites belonging to the Deepwater Navigation Channel showed heavier contamination in the ETM zone than the Nearshore area. There are abundant suspended particles and organic matters in the ETM zone that can effectively capture the hydrophobic compounds. Besides, OTC contamination in shoreside zone was more serious than those in channel center. High OTC loads (average 633 ± 124 ng Sn g^{-1} dw) were also found in sludge samples from adjacent wastewater treatment plant.

Conclusion: Land-based sources (e.g. sewage discharge, runoff) are probably one of the predominant pathways of OTCs entering the ocean. Combining with previous observation in 2014, OTC contamination in the same ten sites varied slightly with years. Risk assessment indicated that the concentrations of tributyltin (TBT) are sufficient to pose ecosystem threats especially in the ETM zone. Hence, OTC contamination issues in the Yangtze Estuary still can not be neglected.

Keywords: Organotins, Estuary contamination, Estuarine Turbidity Maxima zone, Trapping ability, Sources

Introduction

Organotin compounds (OTCs) are typical persistent organic pollutants and endocrine disruptors. Since 1960s, butyltins and phenyltins have been widely used as antifouling agents, pesticides, plastic stabilizers and wood preservatives, accounting for their ubiquitous occurrence in water environments [1, 2]. Exposure to OTCs could cause many harmful effects on marine organisms, and imposex is of particular concern since it can result in female sterility and species extinction. It was reported that TBT has caused imposex in more than 260 gastropods worldwide [3]. Thus TBT-based products (mainly antifouling paints) have been banned in many countries and the global prohibition proposed by the International Maritime Organization (IMO) came into effect in 2008 [4–7].

Estuaries are the transition zone between rivers and seawaters with obvious land-ocean interactions (e.g., the influxes of saline water, flows of fresh water and high nutrients), making them become the most productive natural habitats [8, 9]. The Estuarine Turbidity Maxima

*Correspondence: wulingling@tongji.edu.cn; wulleco@hotmail.com
[1] Key Laboratory of Yangtze River Water Environment of the Ministry of Education, College of Environmental Science and Engineering, Tongji University, Shanghai, China
Full list of author information is available at the end of the article

(ETM) zone has much more amounts of suspended particles than its upstream and downstream [10]. They can trap land-based debris (e.g., fine sediments, organic matters and pollutants) and transferred them into bottom sediments [11, 12]. Sediments are widely known as long-term reservoir and continuous sources for OTC residues based on their more than 10 years of half-life [13, 14]. The Yangtze Estuary, as a typical tidal estuary, also includes the ETM zone where many OTC-sensitive species inhibit (e.g., snails, clams) [15, 16]. In recent years, this estuary has been experiencing significant industrialization and urbanization and became one of the most important shipbuilding bases in China. Local human activities greatly disturb the normal marine ecosystem. Chen et al. [15] found a widespread distribution of OTCs in the Yangtze Estuary in 2014, but no data is available about their temporal variation, potential sources and environmental behaviors based on the specific geographic conditions (e.g., hydrophobic pollutants "accumulation" effects in the ETM zone) locally.

In this study, five OTCs (TBT, dibutytin (DBT), monobutyltin (MBT), triphenyltin (TPhT) and diphenyltin (DPhT)) were determined in surface sediments from the Yangtze Estuary including the Nearshore area, the ETM zone and the Plume zone in 2015–2016, as well as sludges from adjacent Shidongkou Wastewater Treatment Plant during 4 different months. This study is aimed to a comprehensive overview of OTCs' distribution, sources and environmental fates in marine environment especially the high turbid zones.

Materials and method

Study area

The Yangtze Estuary is a typical middle tidal zone with abundant runoff and sediment fluxes. After more than 200 years' evolution, a stable pattern of three order bifurcations and four outlets into the sea has been formed (Fig. 1). The suitable environment and climate facilitate many rare species (e.g., Chinese sturgeon, Chinese paddlefish and finless porpoise) for inhabitation and reproduction. However, the Yangtze Estuary has been subjected to heavy shipping activities recently. The Deepwater Navigation Channel (12.5 m in depth) in its north passage was completed in March 2010 (Fig. 1), greatly increasing the large vessels transport through-out the estuary. Meanwhile, the Yangtze Estuary was estimated

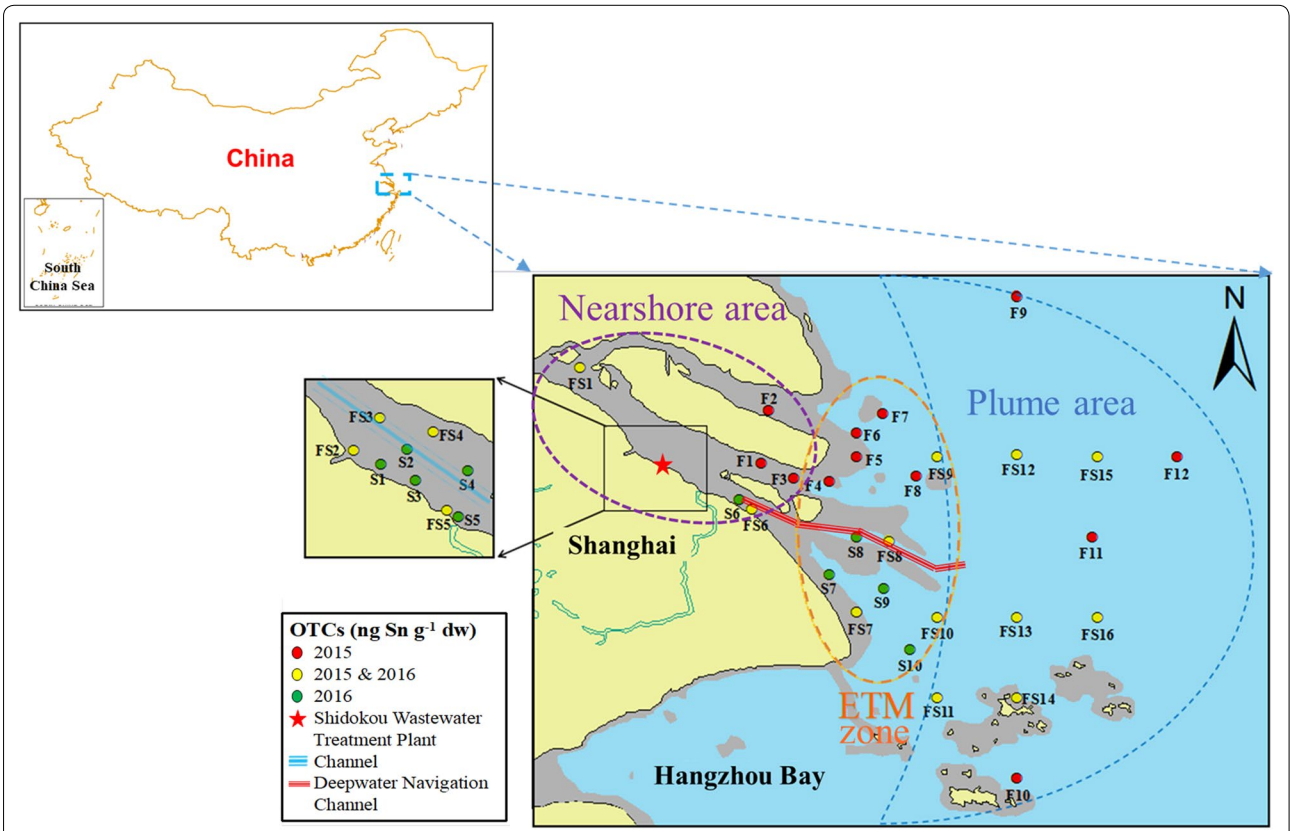

Fig. 1 Sampling sites in the Yangtze Estuary and nearby coastal areas. The pink dotted circle represented the Nearshore area, the orange dotted circle represented the Estuarine Turbidity Maxima (ETM) zone and the blue dotted sector represented the Plume zone

to receive about 5×10^6 tons of municipal and industrial wastewater per day [17]. These facts demonstrated that the ecosystems especially long-lived species in the Yangtze Estuary have suffered strong anthropogenic pressures.

Sample collection

Surface sediments (0–20 cm) were collected from the Yangtze Estuary and adjacent coastal zones in August 2015 (26 sampling sites) and August 2016 (28 sampling sites). The sites encoded by "F" or "S" represented the sampling campaign in 2015 or in 2016, while "FS" represented the sites visited in both years (Fig. 1). The concentrations of suspended particles were determined in the corresponding waters in 2015 (Additional file 1: Table S1). Sludge samples were obtained from the nearby Shidongkou Wastewater Treatment Plant in November and December 2016 as well as in February and May 2017. Each sample was kept in an OTC-free polyethylene bag and transferred in ice boxes immediately. After arrival to the laboratory, they were freeze–dried, grounded, sieved (200-mesh), and finally stored at − 20 °C until analysis.

Organotin determination

The whole pretreatment, analysis and quantification procedure was performed using our established method [18]. Briefly, 2 g of sediment powder and 10 mL of methanol-acetic acid mixed solutions ($v/v = 9{:}1$, containing 0.03% tropolone) were added in a 70-mL glass vial and ultrasonic extracted for 30 min at room temperature. Then 5 mL of supernatant was derivated by $NaBEt_4$ (w/w = 1%, 600 μL) in another glass vial containing 40 mL of acetic acid-sodium acetate buffer (pH 4.5, 1 mol L^{-1}), 1 g of NaCl, 4 mL of hexane and 100 μL of TPrT (internal standard). The mixture was horizontally shaken for 1 h to achieve complete derivatization and extraction. Then 2 mL of hexane phase was cleaned up by passing through a Florisil column (500 mg/6 mL, CNW) and concentrated to 0.5 mL.

OTCs were determined by gas chromatography coupled with mass spectrometer (Shimadzu QP 2010 Plus, Japan). A HP-5 capillary column (30 m × 0.25 mm × 0.25 μm) was used to separate the OTC targets. The initial column temperature was 60 °C (1 min), increased to 150 °C at a rate of 10 °C min^{-1} and then to a final temperature of 290 °C at 8 °C min^{-1} (2 min). The injector was carried out on a split-less mode. Mass spectrometer was operated in the electron impact mode with 70 eV of ionization energy. The determination was in the selected ion monitoring mode (SIM) using two qualifier ions and one quantifier ion.

Method variation and statistical analysis

The methodology was verified by analyzing the certified reference material (BCR-646, IRMM, Geel, Belgium). The determined levels of butyltins and phenyltins were in good agreement with the certified values (Additional file 1: Table S2). In each batch (12 samples), a blank sediment sample spiked with 100 ng Sn g^{-1} dw of OTC standards was analyzed and obtained 80%–110% of recoveries. The relative standard deviations (RSD) for seven replicate measurements were less than 7%. Limits of detection and quantification estimated as the concentrations corresponding to three and ten times of signal to noise (N/S) in blank sediments were 0.43–0.98 ng Sn g^{-1} dw and 0.82–3.1 ng Sn g^{-1} dw, respectively. The reagent blank showed no contamination of the analytical system and glassware.

Statistical analysis was performed by SPSS Statics 19.0 (SPSS Inc., USA). The relationships of five targeted OTCs were studied using the non-parametric Spearman correlation, whilst their spatial and temporal variations were analyzed by the non-parametric Kruskal–Wallis test. Significance was set at $p < 0.05$.

Results and discussion

Organotin contamination in sludge samples of the Shidongkou Wastewater Treatment Plant

The sum of OTC concentrations ranged from 542 to 807 ng Sn g^{-1} dw among four sampling months (Table 1). Phenyltin contamination was relatively much lower with a mean level of 10.0 ± 8.5 ng Sn g^{-1} dw. DBT was the major component, accounting for more than 68% of total OTCs. On the contrary, Lu et al. [19] observed the dominance of TBT in sewage sludges from 18 Chinese cities (the concentration ratios of TBT and DBT varied from 1.8 to 10.5). This is probably due to the different sampling locations and sampling times. Agricultural use as farmland fertilizers is the main application for sludges, which can transfer the nutrients but also to some extent the organic and inorganic pollutants (i.e., OTCs) to farm soils [20]. These sludge-mixed soils may probably enter the ocean under the influence of local topography, runoff and bad weather conditions. On the other hand, although most of the terrestrial pollutants (i.e., pesticides, plasticizers, heavy metals) are absorbed by sludge deposits via multi-stage wastewater treatment arrangements, some fractions could still be preserved in water phases and directly discharged to the marine environment [21].

Organotin contamination in surface sediments of the Yangtze Estuary

OTCs were found in the majority of sampling sites in 2015 and 2016, with the peak levels of 36.0 ng Sn g^{-1} dw

Table 1 Organotin concentrations (average ± SD) in sludge samples from Shidongkou Wastewater Treatment Plant. Three replicates of each sample were analyzed

Sampling date[a]	MBT	DBT	TBT	DPhT	TPhT	Total
	ng Sn g^{-1} dw					
11/2016	78.4 ± 10.5	470 ± 11.6	ND	ND	ND	548
12/2016	148 ± 12.3	436 ± 20.5	41.7 ± 5.2	5.7 ± 2.4	4.3 ± 1.7	636
02/2017	ND	712 ± 15.8	86.4 ± 11.4	ND	9.0 ± 1.0	807
03/2017	ND	456 ± 6.2	65.6 ± 10.5	13.6 ± 3.6	7.2 ± 2.3	542

ND, below the limits of detections

[a] Sampling date are in MM/YY format

and 34.6 ng Sn g^{-1} dw, respectively. The contamination pattern of OTCs was similar with those observed in other domestic coastal zones such as Kaohsiung harbor and Three Gorges Reservoir, but much lower than some developing countries (e.g. Chile, Brazil, Argentina) (Table 2). In the first sampling campaign (2015), butyltin concentrations in the Nearshore area and the ETM zone (average 14.6 ± 11.0 ng Sn g^{-1} dw, 14.9 ± 5.4 ng Sn g^{-1} dw) were generally higher in comparison with phenyltins (average 2.4 ± 2.5 ng Sn g^{-1} dw, 3.4 ± 2.8 ng Sn g^{-1} dw). Whereas in the Plume zone only 2 sites were slightly contaminated by butyltins and 4 sites by phenyltins (Fig. 2a). A similar distribution was also found in 2016 with the dominance of DBT and MBT (the sum over 56% of total OTCs) in the sampling area except the Plume zone, where the five targeted OTCs were at the same low concentration levels (average 4.4 ± 3.9 ng Sn g^{-1} dw) (Fig. 2b).

Spatial variation of organotins-trapping by the ETM zone

In the two sampling campaigns, total OTC concentrations decreased rapidly with the distance to the Plume zone. Water exchange between the Plume zone and open sea is strong based on the Taiwan warm current, Yangtze diluted water, coastal current, etc., leading to a larger-scale transport and more intense dilution of OTC pollutants in this area than the semi-closed regions. No significant variation of OTC levels was found between the Nearshore area and the ETM zone (Fig. 3). Meanwhile, sites S8 and FS8 belonging to the ETM zone and S6 belonging to the Nearshore area were located in the Deepwater Navigation Channel. OTC contamination theoretically should be similar in these three sites due to the same sources (mainly large ships) and small scaled sampling. While their concentrations especially TBT was much higher in S8 and FS8 than in S6 (Additional file 1: Fig. S1). In comparison with the upriver and downriver areas, the ETM zone has much more abundant suspended particles (Additional file 1: Table S1), as well as

Table 2 Organotin distribution in surface sediments from China and some other foreign countries after the TBT global ban

Country	Sampling site	Sampling year	MBT	DBT	TBT	DPhT	TPhT	References
			ng Sn g^{-1} dw					
China	Kaohsiung Harbor	2009	2.6–30.3	2.9–8.3	12.7 ± 18.8[a]	NG	NG	[5]
	Three Gorges Reservoir	2012/2013	2.0–23.3	17.8–186	1.2–6.2	ND–9.1	ND–4.9	[22]
	Yangtze Estuary	2014	10.0–17.0	ND–17.4	ND–15.7	3.9[b]	3.7[b]	[15]
	Yangtze Estuary	2015/2016	ND–11.3	ND–11.8	ND–10.5	ND–5.6	ND–10.4	This study
Brazil	Todos os Santos Bay	2010/2011	< 3–4.5	4–21	< 2–262	NG	NG	[23]
		2012	< 3–108	< 2–72	< 2–77	NG	NG	
	Santa Catarina harbor	2008	ND–312	ND–394	ND–1137	NG	NG	[24]
	Brazilian coast	2009	< 25–289	< 11–338	< 3.2–279	NG	NG	[25]
Argentina	Bahía Blanca Estuary	2013	31.7–293	14.3–156	7.7–32.0	NG	NG	[26]
France	Toulon Bay	2008/2009	0.5–1000	< 0.5–1800	< 0.5–2700	NG	NG	[27]
Chile	Caldera coast	2015	1.3–204	< 0.2–369	90.4–662	NG	NG	[28]

NG, not given; ND, below the limits of detection

[a] mean level

[b] maximum level

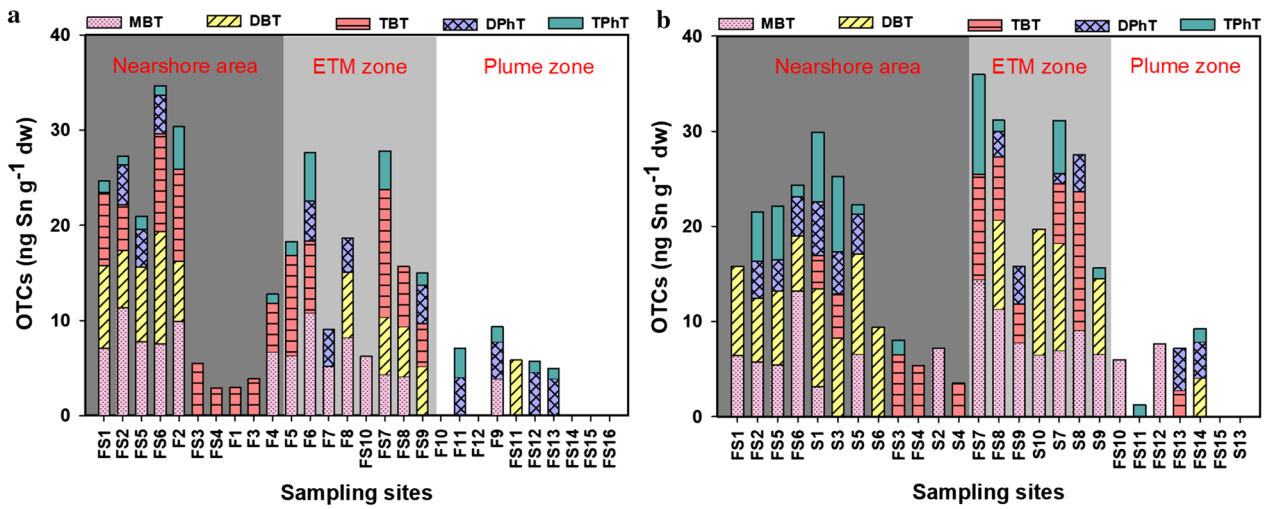

Fig. 2 Organotin concentrations in surface sediments from the Nearshore area, the ETM zone and the Plume zone in 2015 (**a**) and 2016 (**b**)

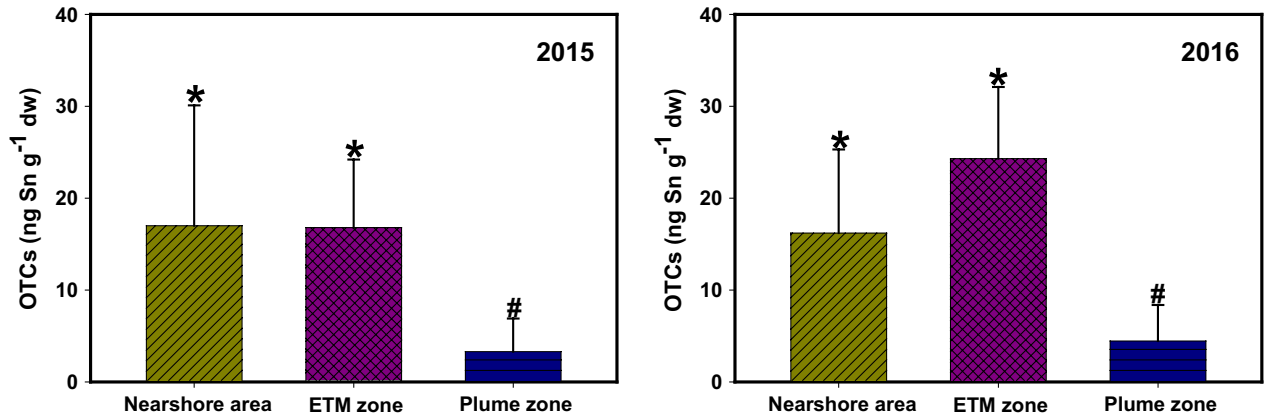

Fig. 3 Spatial variation of total organotins in the Nearshore area, the ETM zone and the Plume zone in 2015 and 2016. Different letters (# and *) above the error bar mean significant difference based on non-parametric Kruskal–Wallis test ($p < 0.05$)

organic matters reported in our previous work and other literature [29–31]. These materials can easily sorb hydrophobic OTCs and then transfer OTCs into sediments via flocculation and sedimentation of suspended particles [32]. Thus most of the OTC residues may probably be trapped and conserved in the ETM zone during the past few years.

Temporal variation of organotins

In 2014, OTC determination was also carried out in surface sediments from the same 10 sampling sites [15]. The temporal variation of OTC levels was evaluated from 2014 to 2016 based on the reported and current findings (Fig. 4). The median levels of total OTCs degreased slightly among three sampling campaigns ($p > 0.05$). While phenyltins exhibited an increasing temporal trend and the highest median value for butyltins was observed in 2015. High imposex frequency and OTC residues were also observed in recent studies in the Mediterranean, the Atlantic and the Polish coast of the Baltic sea [33, 34]. These findings were not surprised since the long-term preservation of OTCs in sediments and the possibility of their illegal usage [14]. Hence, marine OTC contamination are not effectively alleviated even 10 years after the TBT global ban.

Possible source apportionments of organotins in the Yangtze Estuary

For the Nearshore area, OTC composition and distribution in the shoreside zone were different from the

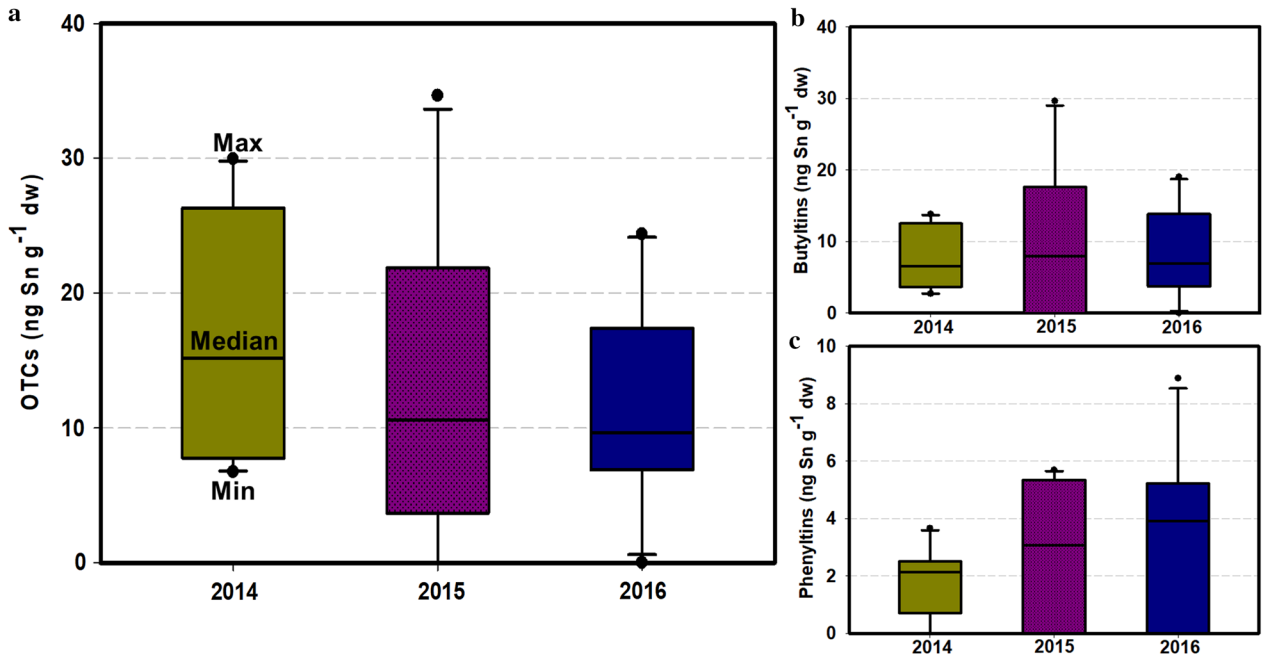

Fig. 4 Temporal variation of total organotins (**a**), butyltins (**b**) and phenyltins (**c**) in the same 10 sites during 2014–2016 (Min and Max range represented the organotin concentrations in the 10th to 90th percentile range)

Table 3 Spatial distribution of organotin compounds (average of three replicates ± SD) in the Nearshore area including the shoreside zones and the center area of channel

Sampling year	Sampling area	Number	MBT	DBT	TBT	DPhT	TPhT	Total
			ng Sn g^{-1} dw					
2015	Shoreside zone	FS2	11.3 ± 2.4	6 ± 2.5	4.8 ± 0.7	4.2 ± 2.1	ND	26.3
		FS5	7.7 ± 1.5	7.9 ± 1.3	ND	4.0 ± 2.1	3.4 ± 1.0	23
		FS6	7.5 ± 3.8	11.8 ± 2.1	5.3 ± 2.6	4.0 ± 1.5	2 ± 1.4	30.6
		F2	9.9 ± 4.0	6.3 ± 1.1	9.6 ± 1.1	ND	4.5 ± 1.1	30.3
	Channel center	FS3	ND	ND	5.5 ± 1.3	ND	ND	5.5
		FS4	ND	ND	2.9 ± 1.5	ND	ND	2.9
		F1	ND	ND	2.9 ± 2.0	ND	ND	2.9
		F3	ND	ND	3.9 ± 1.8	ND	ND	3.9
2016	Shoreside zone	FS1	6.4 ± 2.3	9.4 ± 0.6	ND	ND	ND	15.8
		FS2	5.8 ± 1.4	6.7 ± 2.2	ND	3.9 ± 1.0	5.2 ± 4.1	21.6
		FS5	5.4 ± 3.2	7.8 ± 2.5	ND	3.3 ± 2.6	5.6 ± 1.3	22.1
		FS6	13.2 ± 2.9	5.8 ± 1.6	ND	4.1 ± 3.3	3.2 ± 2.0	27.3
		S1	3.1 ± 2.1	10.3 ± 2.6	3.5 ± 2.4	5.6 ± 1.2	7.3 ± 5.3	29.8
		S3	ND	8.2 ± 3.5	4.7 ± 1.8	4.4 ± 3.2	8.0 ± 2.5	25.3
		S5	6.6 ± 1.7	10.5 ± 1.7	ND	4.2 ± 4.0	6.5 ± 1.6	27.8
		S6	11.3 ± 2.1	9.4 ± 4.3	ND	ND	ND	20.7
	Channel center	FS3	ND	ND	6.5 ± 2.7	ND	1.5 ± 1.0	8
		FS4	ND	ND	5.4 ± 1.9	ND	ND	5.4
		S2	7.1 ± 1.1	ND	ND	ND	ND	7.1
		S4	ND	ND	3.5 ± 2.8	ND	ND	3.5

ND, below the limits of detection

channel center (Table 3). The sum of OTCs in the shoreside zone ranged from 15.8 to 30.6 ng Sn g^{-1} dw, among which DBT and MBT were the predominant species with a proportion of 32%–100%. Low levels of TPhT and DPhT were also prevalent except for sites FS1 and S6 in 2016 with no phenyltin contamination. While in the central area of channel OTC mean concentration was just 4.9 ± 1.9 ng Sn g^{-1} dw. Butyltins were dominated by TBT and only FS3 in 2016 had a detectable phenyltin level (1.5 ng Sn g^{-1} dw of TPhT). A similar trend was found in the Three Gorges Reservoir Region, China where OTC concentrations decreased toward the harbor's central area [35]. DBT, MBT and TPhT have both marine and land-based origins. Except being from TBT's degradation, DBT and MBT have been used as stabilizers for polyvinyl chloride (PVC) plastics; TPhT is applied as fungicides in aquacultural and agricultural fields, but also was as cotoxicant with TBT in some long performance antifouling paints, which is the main usage of TBT compound before its prohibition in 2008 [5, 14]. In the shoreside area, the concentrations of TBT were not correlated with DBT, MBT and TPhT ($p > 0.05$). Meanwhile, sludge samples from adjacent Shidongkou Wastewater Treatment Plant showed much heavier DBT and MBT loads and a similar low TPhT load with the shoreside sediments (mentioned above). This could be an indication that OTC pollution (represented by DBT, MBT and TPhT) in the shoreside area were mainly derived from land-based sources such as the municipal and industrial wastewater discharge, being less associated with TBT. While the slight contamination of OTCs (represented by TBT) in channel center may originate from the historic residues as well as the illegal usage of TBT-based products.

Risk assessment for TBT compound

As a well-known endocrine disruptor, TBT has been of particular concern especially considering its threats to marine organisms. The risk assessment of TBT compound was performed referring to the low trigger values of Australian and Spain sediment quality guidelines (SQG) (Fig. 5) [36–38]. During the two sampling campaigns, at least half of the samples exceeded the Australian SQG low threshold (5 ng Sn g^{-1} dw). Meanwhile, two sites in 2015 and three sites in 2016 in the EMT zone had the levels above 10 ng Sn g^{-1} dw of Spanish SQG low trigger value. It should be noted that these five sites were all closed to the Nature Reserve for Chinese Sturgeon. As benthic feeders, Chinese Sturgeon in particular their juveniles are highly sensitive to chemicals and have been listed in the IUCN Red List (critically endangered) in 2012. TBT compound, even in ppb concentrations, can induce harmful effects to benthic organisms [38, 39]. All of these indicated that potential ecological risks related to TBT still be a serious problem in the Yangtze Estuary.

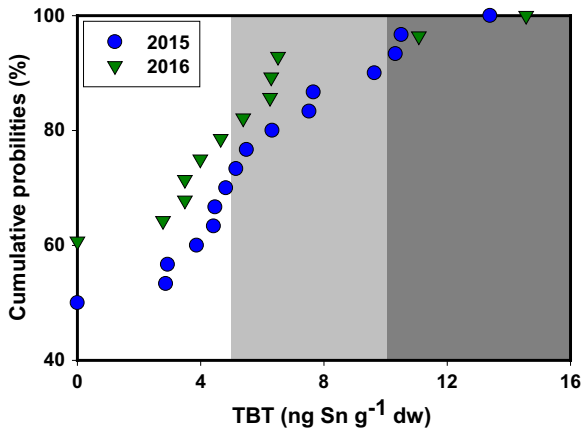

Fig. 5 TBT risk assessment based on the low trigger values of Australian sediment quality guidelines (5 ng Sn g^{-1}) and Spanish sediment quality guidelines (10 ng Sn g^{-1})

Conclusion

The spatiotemporal distribution and potential sources of five OTCs were investigated in the Yangtze Estuary between 2015 and 2016. Although OTC loads decreased greatly towards the open sea, their contamination levels were similar between the ETM zone and the Nearshore area. The possible reasons are the poor water circulation in the semi-closed ETM zone and its strong trapping capacity based on the abundant suspended particles and organic matters. In addition, both butyltins and phenyltins showed obvious spatial variation in the Nearshore area, where the levels found in the shoreside area were much higher than the central area of the channel. Combining with the heavy OTC residues in adjacent sludge samples, these pollutants may mainly originate from the land-based sources. Besides, the annual mean levels of OTCs varied less during 2014–2016, and risk assessment indicated the potential harmful effects of TBT in the ETM zone.

Additional file

Additional file 1: Figure S1. The concentrations of total organotins (**a**) and TBT compound (**b**) in the Deepwater Navigation Channel in 2016. Site S6 is located in the Nearshore area. Sites S8 and FS8 are located in the ETM zone. **Table S1.** Suspended particle concentrations in the Nearshore area, the ETM zone and the Plume zone. **Table S2.** Determination of organotin compounds in the certified reference material BCR-646 (n = 5).

Abbreviations
OTC: organotin compound; TBT: tributyltin; DBT: dibutyltin; MBT: monobutyltin; TPhT: triphenyltin; DPhT: diphenyltin; TPrT: tripropyltin; NaBEt4: sodium tetraethylborate; RSD: relative standard deviations; IMO: International Maritime Organization; SIM: selected ion monitoring; ETM: estuary turbidity maxima; PVC: polyvinyl chloride; SQG: sediment quality guideline; IUCN: International Union for Conservation of Nature.

Authors' contributions
LC designed the experiments. CC performed chemical treatments of sediment samples, data analysis and manuscript writing. RX and SY made contributions to sampling design and sample collection. QH gave good suggestions on the interpretation of experimental data. LW and WZ paid much attention on manuscript revision. All authors read and approved the final manuscript.

Author details
[1] Key Laboratory of Yangtze River Water Environment of the Ministry of Education, College of Environmental Science and Engineering, Tongji University, Shanghai, China. [2] Shanghai Institute of Pollution Control and Ecological Security, Shanghai, China. [3] State Key Laboratory of Pollution Control and Resource Reuse, College of Environmental Science and Engineering, Tongji University, Shanghai, China. [4] East China Sea Environmental Monitoring Center, East China Sea Branch of the State Oceanic Administration, Shanghai, China. [5] John A. Reif, Jr. Department of Civil and Environmental Engineering, New Jersey Institute of Technology, Newark, NJ, USA.

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Competing interests
The authors declare that they have no competing interests.

Availability of data and materials
The datasets supporting the conclusions of this article are included within the article and its additional file.

Consent for publication
Not applicable.

Ethics approval and consent to participate
Not applicable.

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