



## Baseline

## Occurrence and distribution of organophosphorus flame retardants/ plasticizers in coastal sediments from the Taiwan Strait in China



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## ABSTRACT

Concentrations and spatial distributions of organophosphate esters (OPEs) in marine sediment samples in the western Taiwan Strait were investigated and analyzed for the first time. The total OPE concentration ( $\Sigma$ OPE) in sediments was in the range of 5263–34,232 pg/g (dry weight), with an average value of 12,796 pg/g. Generally, there was a higher abundance of non-halogenated OPEs than halogenated OPEs and aryl OPEs, with tributyl phosphate, tris (2-ethylhexyl) phosphate, and tri-(1-chloro-2-propyl) phosphate being the main components. The  $\Sigma$ OPE concentrations in the western coastal and northern sampling sites were relatively high. The results indicated that OPEs in the study region originated mainly from terrestrial river inputs and local emission sources, as well as exogenous influxes by ocean currents.

### 1. Introduction

Organophosphate esters (OPEs) are a group of chemicals that are widely used as flame retardants and/or plasticizers due to their excellent fire resistance and flexibility. They are added to various consumer and industrial products, such as plastics, furniture and decorative materials, coatings, building materials, textiles, and electronics/electrical appliances (van der Veen and de Boer, 2012). With the banning/restriction of polybrominated diphenyl ethers (PBDEs), the production and usage of alternative flame retardants, especially OPEs, has increased significantly and is estimated to reach about 3 million tons in 2018 (Wei et al., 2015; Lai et al., 2019; de la Torre et al., 2020). During their production, application, and recycling, OPEs can be released into the ambient environment through volatilization, abrasion, and leaching (van der Veen and de Boer, 2012; Xu et al., 2019). After emission, these chemicals can be transported over long distances through the atmosphere or along rivers (Sührling et al., 2016; Ma et al., 2017). Individual OPEs from different subgroups (namely, chlorinated OPEs, alkyl OPEs, and aryl OPEs) exhibit different physicochemical properties (such as water solubility, log octanol–water partition coefficient, and vapor pressure), which could significantly affect their occurrence, distribution, and ultimate fate in the environment (van der Veen and de Boer, 2012; Wei et al., 2015).

However, it has been demonstrated that OPEs, especially chlorinated OPEs, are similar to/or more persistent than those that have

replaced PBDEs (Wei et al., 2015; Hou et al., 2016). These compounds have reached remote regions via long-distance transportation through the atmosphere or water (McDonough et al., 2018; Rauert et al., 2018). In addition, many studies have demonstrated that OPEs also have a range of adverse effects on aquatic organisms and the human body, such as neurotoxic, carcinogenic, and endocrine-disrupting activities (van der Veen and de Boer, 2012; Wei et al., 2015). Consequently, there is great concern about the risk they pose to ecosystems and human health, and increasing attention has been paid to their toxicity as well as their occurrence, distribution, and ultimate fate in the environment.

The Taiwan Strait (TWS), located between Taiwan Island and the southern Chinese mainland, is the key pathway connecting the South China Sea (SCS) and the East China Sea (ECS) and transports materials between both seas. Given its geographical location, the TWS is influenced by multiple factors, including different ocean currents, eddies, submarine groundwater discharge, and coastal upwelling. Besides receiving a large terrestrial input from the mainland and Taiwan Island, different sources of water are transported to and mixed in the TWS (please see Fig. S1), including the Kuroshio Branch (KCB), which originates from West Philippines Sea water, the Southward China Coastal Current, Zhejiang-Fujian Coastal Current (ZFCC), and the Taiwan Warm Current (TWC). In summer, the KCB and TWC are the strongest currents, while the ZFCC, which is driven by the East Asia Monsoon, is the strongest current in winter. The main water masses originate from SCS

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water. Under this unique environment, nutrients and organic carbon (OC) from different sources have been deposited and buried in the region (Liao et al., 2018; Ya et al., 2018; Huang et al., 2019). Similarly, PAHs in Jiulong River, SCS, and ZFC were transported and mixed resulting in high levels of PAHs in seawater in TWS (Ya et al., 2018; Wu et al., 2011). There has been reported that OPEs were detected in seawater in Yellow River Estuary, the Pearl River Delta and SCS. These compounds could be transported and deposited in the TWS, however, little is known about OPEs pollution in the region (Lai et al., 2019).

Given the extensive production and usage of OPEs in China (Wei et al., 2015) and the geographic and hydrologic features of the TWS (Liao et al., 2018), we investigated the levels and distribution of widely used OPEs in marine sediments in this region. A total of 32 surface sediment samples from the TWS were collected and analyzed to: i) investigate the concentrations and composition profile of OPEs in the marine sediments; ii) explore the different spatial distributions of individual OPEs; and iii) determine their possible emission sources based on the measured concentrations and spatial distribution.

Ten OPEs were selected as target analytes and standards were purchased from Sigma-Aldrich (St. Louis, MO, USA). The details of the target compounds are listed in Table S1. Four deuterated compounds,  $d_{15}$ -triethyl phosphate ( $d_{15}$ -TEP, 99.1%),  $d_{27}$ -tributyl phosphate ( $d_{27}$ -TNBP, 98%),  $d_{15}$ -triphenyl phosphate ( $d_{15}$ -TPHP, 98%), and  $d_{12}$ -tris-(2-chloroethyl) phosphate ( $d_{12}$ -TCEP, 98%) were purchased from C/D/N Isotopes (Pointe-Claire, QC, Canada) and used as surrogate standards. Hexamethylbenzene (HMB, 99.5%) was obtained from Ehrenstofer-Schäfer Bgm-Schlösser (Karlsruhe, Germany) and used as an internal standard.

All solvents were of chromatographic grade. Methanol, acetone, *n*-hexane, ethyl acetate, and dichloromethane (DCM) were purchased from Merck Co. (Darmstadt, Germany). Milli-Q water was obtained from a Milli-Q Unique-R10 water purification system (Merck Millipore Inc., Billerica, MA, USA). Neutral alumina (100–200 mesh) and silica gel 60 (70–230 mesh) were purchased from Merck Co. (Darmstadt, Germany). Anhydrous  $\text{Na}_2\text{SO}_4$  was purchased from Guangzhou chemical reagent factory (Guangzhou, China).

The silica gel and neutral alumina was extracted continuously with methanol and DCM for 48 h, activated at 180 and 250 °C, and then deactivated using 3% (w/w) ultrapure water. Finally these materials were kept in *n*-hexane prior to use. Anhydrous  $\text{Na}_2\text{SO}_4$  was baked for 4 h at 450 °C and kept in a desiccator prior to use.

The study region was located in the northern and western TWS in the range of 23.5–27°N latitude and 118.5–121°E longitude (as indicated in Fig. 1). The geographic location of each sampling site is listed in Table S2 and a brief description is given below. A total of 32 surface sediments (0–5 cm) were collected in July 2016, with a stainless-steel grab sampler onboard the R/V YANPING II. Samples were placed in an aluminum container and then transported on ice to the laboratory. In the laboratory, all sediment samples were freeze-dried, finely ground, and homogenized after the removal of organic debris and pebbles. After passing through a 60 mesh sieve, the sediment samples were stored in pre-cleaned amber bottles at –20 °C until extraction. Detailed information about the study region and its ocean currents was published previously (Liao et al., 2018).

Approximately 10 g of sediment sample was spiked with surrogates (100 ng of  $d_{15}$ -TEP,  $d_{27}$ -TNBP,  $d_{15}$ -TPHP, and  $d_{12}$ -TCEP) and Soxhlet-extracted with 120 mL DCM for 72 h. Activated copper was used to remove elemental sulfur in the sediment samples. The extracts were concentrated and solvent-exchanged into *n*-hexane, and concentrated again to about 1 mL with a rotary evaporator. The extracts were subjected to further separation and clean-up via a combined column of neutral alumina/silica gel (1:2). The fraction containing the target OPEs was concentrated almost to dryness under a gentle  $\text{N}_2$  stream, and redissolved into 200  $\mu\text{L}$  *n*-hexane. An aliquot of 100 ng internal standard (HMB) was added before the instrumental analysis.

Determination and quantification of OPEs was conducted using a gas chromatography unit equipped with a mass spectrometer (Model 2010, Shimadzu, Kyoto, Japan) coupled with a TG-5 ms column

(30 m  $\times$  0.25 mm i.d.  $\times$  0.25  $\mu\text{m}$ , Thermo Fisher Scientific, Waltham, MA, USA). The details of the extraction, clean-up, and determination of the analytes has been published previously (Zeng et al., 2018a), with further details provided in the Supporting Information.

The limit of detection (LOD) is defined as the smallest amount of a target compound that could be reliably measured for each individual OPE. In the present study, LODs were calculated by  $3 \times (\text{SD}/S)$ , where SD is the standard deviation of the response of seven repeat injections of low levels of OPEs approximate to blank samples (Zeng et al., 2018a, 2018b), with values being in the range of 1.15–6.09 ng/mL. In addition, those compounds without signal response during instrumental analyses procedure were defined as Not Detected (ND), those showing lower signal response and the calculated concentration being values lower than twice LODs were defined as LODs.

During the analytical procedure, strict quality control and quality assurance procedures were adopted for the analyses of OPEs in sediment samples. Procedural blanks ( $n = 3$ ), spiked blanks ( $n = 3$ , analytes spiked into solvents), and spiked matrix ( $n = 3$ , analytes spiked into pre-treated sediment) were analyzed in every batch of real sediment samples. Surrogate standards were added to all samples. The results indicated that low recoveries were obtained for  $d_{15}$ -TEP (35.2%  $\pm$  14.0%) due to its high volatility (Please see Table S1). The recovery rates of the other three surrogates were 91.8%  $\pm$  13.0%, 125%  $\pm$  12.3%, and 86.7%  $\pm$  12.8%, for  $d_{27}$ -TNBP,  $d_{12}$ -TCEP, and  $d_{15}$ -TPHP, respectively. The recovery rates of targets compounds from the spiked matrix varied in the range of 39.6  $\pm$  14.0% (TEP)-121  $\pm$  10.5% (TCIPP). Three congeners, TCEP, TCIPP, and TPHP were found to be below the LOD in all blank samples, and the other OPEs were not detected in any blank samples. The reported OPE concentrations in sediments were not corrected with recovery rates.

In this study, TEP and TPrP were not detected in any of the sediment samples. The other eight target OPEs were found with different detected frequencies (DFs) in the study region. The concentrations and distributions of the eight detected OPEs in the sediments are summarized in Table 1. Further detailed information is listed in Table S3. From Tables 1 and S3, it can be seen that the total concentration of eight OPEs ( $\Sigma 8\text{OPEs}$ ) in the sediment ranged from 5263 to 34,232 pg/g (dry weight), with an average value of 12,796 pg/g.

Three alkyl OPEs were detected, and TNBP and TEHP were found in all sediment samples in the range of 2994–15,332 pg/g (average 7916 pg/g) and 91.4–16,803 pg/g (average 1821 pg/g), respectively. In contrast, TBOEP was found in 20 samples, with a DF of 62.5% and concentrations varying from the LOD to 634 pg/g. For the two aryl OPEs (TPHP and TMPP), TPHP was detected at 45.6–4364 pg/g, with an average value of 549 pg/g and DF of 100%. A low level of sedimentary TMPP was measured in the range of ND–671 pg/g (average 93.8 pg/g), with a DF of about 93.8%. It was notable that the three chlorinated OPEs were found in all samples. The TCEP concentration ranged from the LOD–4232 pg/g, with an average value of 1096 ng/g, and the TCIPP concentration ranged from 442 to 3448 pg/g, with an average value of 1314 pg/g. TDCIPP was detected at the LOD in 25 samples and at concentrations up to 204 pg/g in the other seven sediment samples. Based on the average concentrations and DFs, TNBP, TEHP, and TCIPP were the dominant OPEs in these marine sediments.

As mentioned in the introduction, there is an ongoing concern from the public and scientists about the potential risk from OPEs to humans and ecosystems. Many studies have demonstrated their ubiquitous presence in the environment, as well as their toxicity to ecosystems and human beings, most of which have focused on their presence in the terrestrial aquatic and indoor environments (Wei et al., 2015; Pantelaki and Voutsas, 2019). Only a few published studies have reported OPEs in the marine environment and these are summarized in Table 2 (Tan et al., 2016; Ma et al., 2017; Zhong et al., 2018; Lai et al., 2019).

In a previous study, ten marine sediment samples were collected from the Pearl River Estuary (PRE) and analyzed (Tan et al., 2016), and the total concentrations of 11 OPEs (TEP, TiPP, TNBP, TCEP, TCIPP,

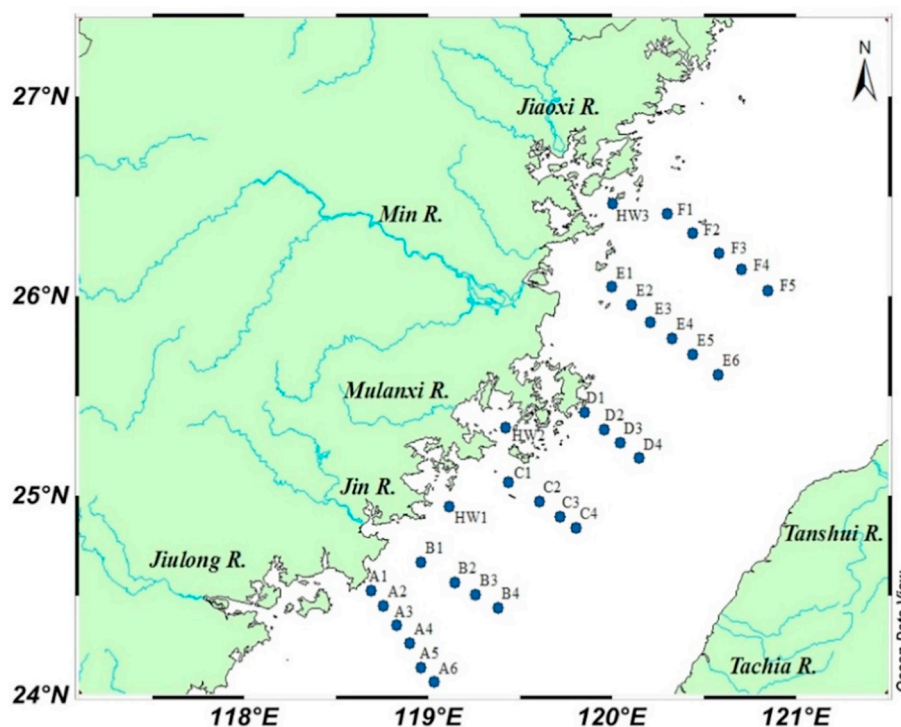


Fig. 1. Illustration of sampling location in Taiwan Strait.

TDCIPP, TBOEP, TEHP, 2-ethylhexyldiphenyl phosphate, TPHP and TMPP) varied in the range of 12–66 ng/g. Due to the significant discharges from *E*-waste recycling activity, as well as domestic and industrial wastewater discharges from the densely populated, highly urbanized, and industrialized region, the concentrations of chlorinated OPEs (TCEP 1.5–33 ng/g, TCIPP 1.5–11 ng/g, and TDCIPP ND–1.8 ng/g) in the PRE were one order of magnitude higher than the levels found in our study. Due to industrial discharges from a TCIPP manufacturer located in Jiangsu Province, the TCIPP concentration was found to be high (1521 pg/g) in the sediment of a river mouth to the Yellow Sea, which was similar to the level reported here for the TWS sediment, while levels of TCEP, TDCIPP, TPHP in the Yellow Sea were one order of magnitude lower than those in the present study (Zhong et al., 2018). According to Ma et al., (2017), seven OPEs were detected in marine sediment from the North Pacific to the Arctic Ocean at concentrations in the range of 159–4658 pg/g. Unlike the results in the present study, chlorinated OPEs and TiBP contributed most to the total OPE concentration in the region, which might be due to the deposition of chlorinated OPEs from long-distance transportation as well as river discharges from the Canadian Archipelago. In sediment from the Great Lakes (Cao et al., 2017), comparable concentrations of 14 OPEs were measured in the range of 2.2–16.6 ng/g, with different concentrations and compositions depending on the diverse range of emission sources and specific physicochemical properties for each OPE. Among their target analytes, TiBP and TNBP are the most volatile OPEs and could be partitioned into the gaseous phase and undergo long-distance

transportation to remote regions, whereas hydrophobic chemicals (e.g., TPHP, TMPP, and TEHP) were likely to be absorbed onto particles and sediment after local discharge.

The composition profile of OPEs at all sampling stations is shown in Fig. 2. Among the non-halogenated OPEs, TNBP was the predominant OPE detected in all sampling locations (with the exception of F1), making a contribution of 34.0%–81.8% to  $\Sigma$ OPEs. In sampling location F1, a significantly high level of TEHP was detected (16,803 pg/g), accounting for about 49.1% of  $\Sigma$ OPEs. The results suggest that there were local TEHP emissions adjacent to this sampling location. Among the chlorinated OPEs, TCIPP was the dominant OPE, with concentrations in the range of 442–3448 pg/g (average 1314 pg/g), followed by TCEP, with concentrations ranging from ND to 4234 pg/g (average 1096 pg/g). TDCIPP was detected in all sampling sites, with concentrations varying from the LOD to 204 pg/g. The total concentrations of chlorinated OPEs contributed approximately 4.13%–53.5% to  $\Sigma$ OPEs.

The OPE composition profile in the present study was unlike those reported by Chung and Ding (2009), who reported that chlorinated OPEs were the predominant compounds among six OPEs in two sediments adjacent to Taiwan Island. This maybe indicate the different usage pattern of OPEs in industry between mainland and Taiwan, and the different target compounds selected in these two studies. In our study, all sampling sites were situated in the region close to the mainland, whereas in their report, two marine sediment samples were collected from the southwestern TWS adjacent to Taiwan Island. The different sampling sites might be impacted by different discharges from

**Table 1**  
Statistics of OPEs in marine sediments from the Taiwan Strait (pg/g).

	Chlorinated OPEs			Alkyl OPEs			Aryl OPEs	
	TCEP	TCIPP	TDCIPP	TNBP	TEHP	TBOEP	TPHP	TMPP
Range	LOD-4232	442–3448	LOD-206	2994–15,332	91–16,803	ND-634	46–4364	ND-671
Average	1096	1314	124	7916	1821	343	549	42
DFs (%)	100	100	100	100	100	62.5	100	93.8

ND: not detected; LOD: below the limit of detection.

**Table 2**  
Comparison of OPEs concentration in marine sediments reported (pg/g).

Location	TEP	TPrP	TNBP	TBOEP	TEHP	TCEP	TCIPP	TDCIPP	TPHP	TMPP	ref
Taiwan Strait	-	-	-	-	-	1000-1400	ND-4700	ND-600	ND-2300	-	Chung and Ding, 2009
Pearl River Estuary	ND-910	ND	ND-13000	ND-10000	860-12,000	1500-33,000	1500-11,000	ND-1800	ND-16000	20-1200	Tan et al. 2016
Bohai / Yellow seas	-	-	4-54	-	8-3445	7-671	29-1521	2-54	7-209	-	Zhong et al., 2018
North Pacific / Arctic cean	-	-	19-209	-	-	81-3903	0.94-460	ND-165	ND-105	-	Ma et al., 2017
Taiwan Strait	ND	ND	2994-15,332	ND-634	91-16,803	ND-4234	442-2692	LOD-206	46-4364	ND-671	This study

the mainland and Taiwan Island, respectively. In addition, in their report, only five OPEs (TCEP, TCIPP, TDCIPP, TPHP, and methyl diphenyl phosphate) were investigated, without TNBP and TEHP, which were the dominant components in our study.

Fig. 3 shows the spatial distribution of ΣOPEs. Generally, higher levels of OPEs were found in the northern and western parts of the TWS, which was similar to the reported distribution characteristic of particle-size of sediment (Liao et al., 2018). Under the combined effects of ocean currents, such as the ZFCC, SWC, KCB, and TWC, fine particles are transported farther than coarse particles and are then deposited in the northern and western parts of the TWS (Liao et al., 2018). It was not surprising that higher concentrations of ΣOPEs were found in the area due to the sedimentation of fine particulate containing OPEs carried to the study area by ocean currents from other waters, such as from the SCS and Yellow Sea (Lai et al., 2019; Zhong et al. 2018).

According to a report by Lai et al. (2019), OPEs are ubiquitously present in water from the PRE that receives massive river discharges from the Pearl River Delta and the SCS offshore the PRE. We speculate that a portion of the OPEs could be transported to the TWS along the SWC, KCB, and TWC, after being adsorbed on particles, and then deposited in sediment in the western district under the unique hydrodynamic conditions (Liao et al., 2018). Simultaneously, high concentrations of OPEs have been reported in the Yellow River Estuary, Bohai Sea, and Laizhou Bay (Lai et al., 2019; Zhong et al. 2018). Under the action of the ZFCC, which is driven by the northeast monsoon in winter, the OPEs could be transported to the TWS, similarly to the transportation of PAHs from the Yangzi River to the western TWS (Ya et al., 2018).

The spatial distribution of several main OPE individual congeners (TNBP, TEHP, TPHP, TCEP, and TCIPP) in the TWS is shown in Fig. 3. The other individual OPEs are not shown because of their low concentrations (TMPP and TDCIPP) and/or lower DFs (TBOEP). However, it was possible to determine their main sources based on the measured concentrations of OPEs as listed in Table S3. TDCIPP was found at a quantifiable concentration higher than the LOD in only seven samples, most of which were adjacent to the mainland (e.g., HW1, HW3, and C1) or close to the Taiwan Island (e.g., D4). The results suggest river discharges from the mainland and Taiwan Island (Liao et al., 2018; Wang et al., 2018). The TMPP concentration was 671 pg/g in A6, a sampling site near Taiwan Island and the Penghu Islands, which was one order of magnitude higher than in the other samples. The results indicated a local emission source (Wang et al., 2018).

As indicated in Fig. 3, the five OPE individual congeners had heterogeneous spatial distribution characteristics. For TCEP, the highest concentration was 4234 pg/g in D1 near Pingtan County in the mainland, followed by HW3, with a concentration of 2677 pg/g. The highest TCIPP concentration was 3448 pg/g in B1, followed by 2692 pg/g in E1 and 2171 pg/g in D1. The results suggested that TCEP and TCIPP were possibly derived from river discharges from the mainland (e.g., the Jin, Mulanxi, and Min rivers) and then diffused and transported in the study region under multiple influences, including the Xiamen-Quanzhou upwelling and ZFCC (Liao et al., 2018). In general, chlorinated OPEs were more persistent than non-chlorinated OPEs and could undergo longer distance transportation, with high concentrations gradually dominant over the non-chlorinated OPEs in fresh water influxes into seas (Wang

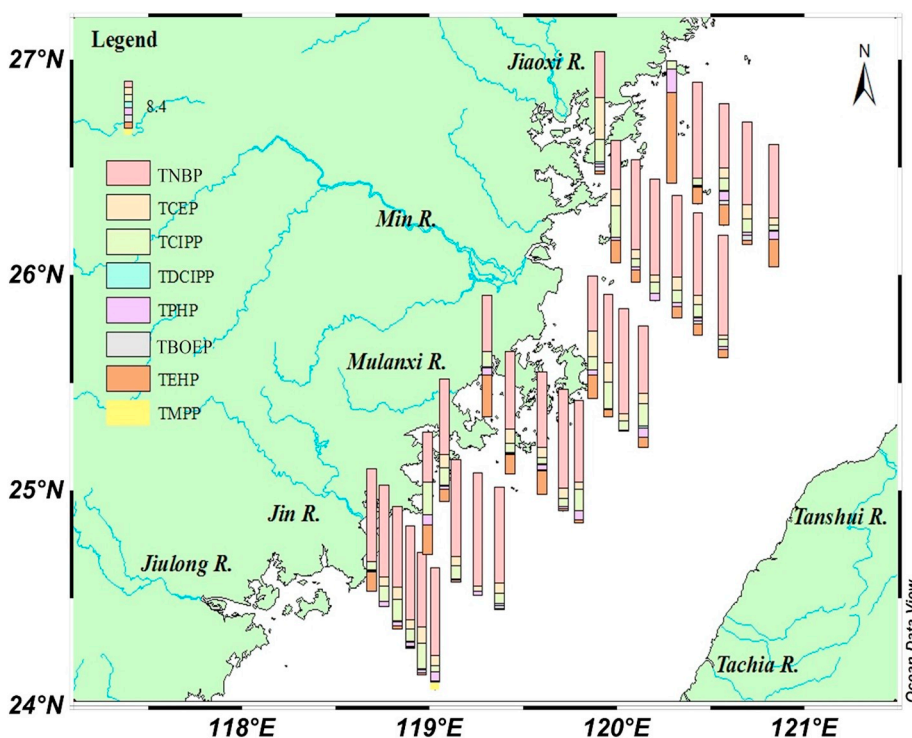


Fig. 2. Composition profile of OPEs in sediment in Taiwan Strait.



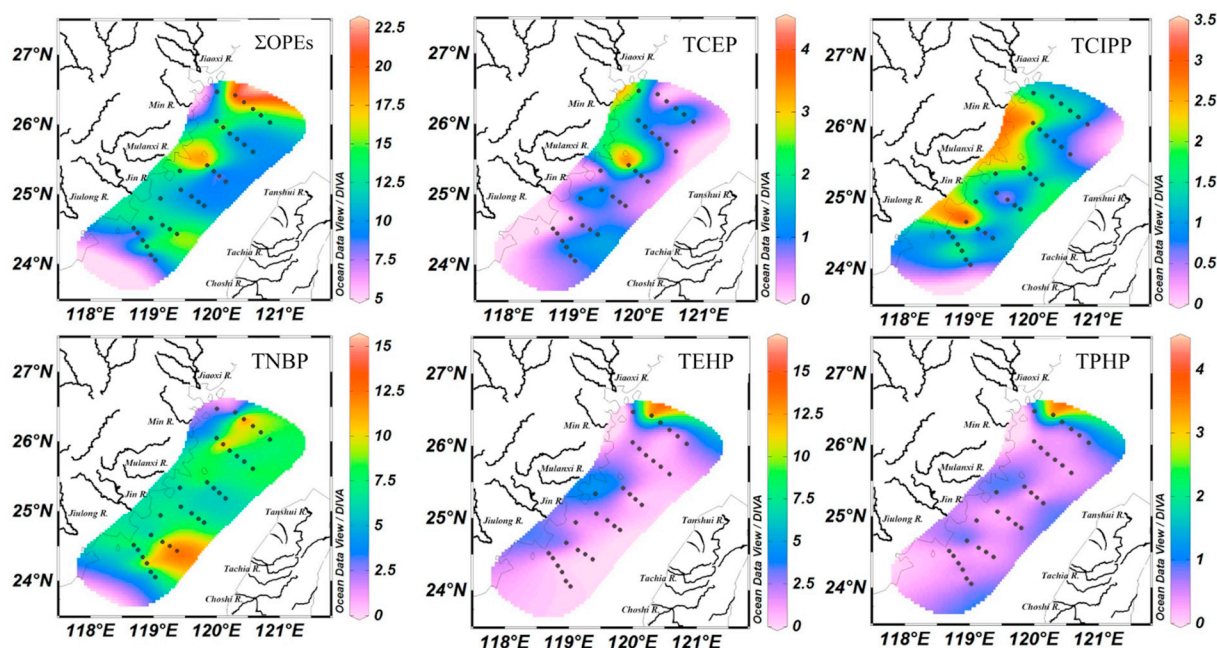


Fig. 3. Spatial distribution of target OPEs.

et al., 2011). It was not surprising that the highest concentrations of chlorinated OPEs were found in sampling sites near river mouths.

It was surprising that TEHP and TPHP shared a similar spatial distribution. In the northwestern TWS, the F1 sampling site was not located at the mouth of any river and was not near the mainland or Taiwan Island (see Fig. 1); however, it had high levels of TEHP (16,803 pg/g) and TPHP (4364 pg/g). The concentration of each OPE was at least four times higher than in the other sampling sites. The results suggested an unidentified local emission source near the sampling location.

High TNBP concentrations were measured in the study region, with considerably high TNBP concentrations found at F2, E2, B4, B3, and A4. TNBP is an important additive in hydraulic oils and lubricants (van der Veen and de Boer, 2012), consequently we speculated that high levels of TNBP might be due to extensive shipping activity (Ya et al., 2018) in the TWS connecting SCS and ECS.

Based on the above discussion, it was concluded that the OPEs in the sediments of the TWS were mainly derived from terrestrial inputs, local emission sources, and exogenous inputs from the SCS and Yellow Sea.

Mass inventory of 8 OPEs in the surface sediments in the studied area was estimated in order to assess the contamination status and the potential of the sediment as a second pollution source to the surrounding environment. The studied area was about 189,925 km<sup>2</sup>. The total inventory ( $I$ ) was calculated by the following equation:

$$I = \sum kC_iAid\rho$$

where,  $C_i$  was the average concentrations of individual OPE (pg/g),  $A_i$  was the total area of the studied region,  $d$  is the thickness of sediment sampled, which was 5 cm in the present study,  $\rho$  means the average density of the dry sediment at 1.2 g/cm<sup>3</sup> according to literature (Zhou et al., 2014). Based on the measured OPEs concentrations, the mass inventory of TNBP, TBOEP, TCEP, TCIPP, TDCIPP, TPHP and TMPP were 9.02, 0.391, 1.25, 1.50, 0.141, 0.625 and 0.0985 tones, respectively. It is noting that chlorinated OPE were much persistent than alkyl/aryl OPE, much attention should be paid to their adverse effect on aquatic organisms and potential impact to adjacent aquatic environment.

In the present study, we investigated the levels and composition profile of widely used OPEs as well as their spatial distribution in coastal sediments of the TWS. It was found that OPEs were ubiquitous in sediment in the study region, and ΣOPEs concentrations were in the

range of 5263–34,232 pg/g, with an average of 12,796 pg/g and TNBP, TEHP, and TCIPP being the dominant compounds. Riverine inputs, transportation, and ocean currents were the main factors that influenced the spatial distribution of OPEs in marine sediment. Further studies should be conducted to evaluate OPE degradation and transformation, especially for non-chlorinated OPEs, as well as their possible vertical distribution due to long-term sedimentation in the region.

#### CRedit authorship contribution statement

**Xiangying Zeng:** Conceptualization, Writing - review & editing, Funding acquisition. **Liang Xu:** Investigation, Writing - original draft. **Qiongpu Hu:** Investigation, Data curation. **Yi Liu:** Investigation. **Jianfang Hu:** Resources. **Weisen Liao:** Investigation. **Zhiqiang Yu:** Writing - review & editing, Funding acquisition.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2019.110843>.

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