



Mediated distribution pattern of organic compounds in estuarine sediment by anthropogenic debris



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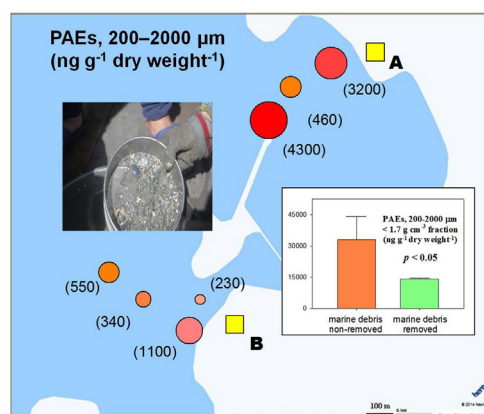
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HIGHLIGHTS

- Concentrations of OPFRs, OTs and PAEs are elevated in coarse fractions.
- Removal of debris from carbon-rich sediments can lower organics concentrations.
- Debris is responsible for the formation of “hot spots” in estuarine sediment.

GRAPHICAL ABSTRACT



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ABSTRACT

Natural organic matter and grain size are considered as important parameters dictating the transport and fate of organic compounds in sediment. However, increasing evidence suggested that manufactured debris may alter the underlying mechanisms for biogeochemical cycling of organic compounds. To examine this assumption, estuarine sediment and embedded debris were collected from a fishery base in Guangdong Province of South China and analyzed for organophosphorus flame retardants (OPFRs), phthalates (PAEs), organotin compounds (OTs) and dichlorodiphenyltrichloroethanes (DDTs). Coarse-size debris ($>200 \mu\text{m}$) were heterogeneously distributed in sediment, and most abundant near the boat maintenance facilities, aquaculture zone and shipping channel. The median concentrations of OPFRs, OTs, PAEs and DDTs in debris were 11, 0.2, 11 and $3.9 \mu\text{g g}^{-1}$ dry sample weight⁻¹, respectively, 1 to 3 orders of magnitude greater than those in bulk sediment (19, 60, 240 and 570 ng g^{-1} dry sample weight⁻¹, respectively). Furthermore, OPFRs, OTs and PAEs were mostly ($>99\%$) enriched in coarse-size (63–2000 μm) sediment, and there was no significant correlation ($p > 0.05$) between the concentrations of OPFRs, OTs and PAEs in bulk and size-fractionated sediment samples and total organic carbon or grain size, similar to the distribution pattern of DDTs reported previously. When distinct debris were removed from the light-density ($<1.7 \text{ g cm}^{-3}$) fraction of coarse-size (200–2000 μm) sediment, the concentration levels of

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OPFRs, OTs, PAEs and DDTs declined by 84%, 59%, 55% and 7%, respectively. Obviously, debris irregularly distributed in sediment can alter the sediment sorption capacity for OPFRs, OTs and PAEs, and thus may undermine the significance of organic matter and grain size to the distribution of organic chemicals in sediment. Finally, commonly used procedures for preparing sediment samples and screening of debris may disturb the grain size distribution or underestimate the abundance of heavy-density debris, resulting in flawed sediment quality assessment.

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1. Introduction

Natural organic matter and carbonaceous materials of different origins were shown to impact the distribution of organic compounds in sediment (Hung et al., 2007; Jonker and Koelmans, 2002). Particle grain size also exerts significant influences on the biogeochemical cycling of organic compounds in general (Pierard et al., 1996). Because small-sized particles have large specific surface areas, they have high adsorption capacity for organic compounds (Jonker and Koelmans, 2002; Weiler and Mills, 1965). Therefore, total organic carbon (TOC) and sediment grain size are expected to be positively and negatively, respectively, correlated with sediment organic compounds, especially if the adsorbents and adsorbates are of the same source (Ali et al., 2014; Hung et al., 2007; Koh et al., 2006). Previous studies also showed that there was no or weak significant correlation between the concentrations of organic compounds and TOC or particle grain size in heavy anthropogenically impacted regions, such as in the vicinity of industries, coasts and harbors (Casado-Martinez et al., 2009; Edgar et al., 2003; Environment Agency, 2007; Khim et al., 1999; Khim et al., 2001; Thompson et al., 1996). For instance, Edgar et al. (2003) did not find any significant correlation between the concentrations of polychlorinated biphenyls and TOC or sediment grain size in intertidal sediment of the Clyde Estuary, United Kingdom.

Anthropogenic debris are a group of manufactured or processed solid waste materials that enter into the environment either directly or indirectly after use or disposal and eventually occur in lands or oceans (Jambeck et al., 2015). The amount of debris was found to be proportional to the extent of anthropogenic impacts in regions within 200 km of the coastline (van Sebille et al., 2015). Debris encompass metals (e.g., derelict vehicles and beverage containers), glass (e.g., light bulbs and bottles), plastics and other materials (e.g., lumber, rubber and textile) (The United States Environmental Protection Agency's (US EPA) Office of Water). Additives are often used to enhance product durability (Hansen et al., 2013), which may subsequently increase the persistence of obsolete merchandises in the environment. An example is the addition of organophosphorus flame retardants (OPFRs), phthalates (PAEs) and organotin compounds (OTs) to a variety of industrial and consumer products to lessen the effects of heat and light on plastics. Debris can also act as sorbents to accumulate organic compounds from the environment, e.g., the amount of phenanthrene accumulated from seawater by plastics was 2 orders of magnitude greater than that in sediment (Teuten et al., 2007). Clearly, the distribution of organic compounds in sediment may be mediated by the occurrence of debris. For example, our previous study reported that paint flakes unevenly distributed in estuarine sediment could artificially inflate the sorption capacity of heavy-density ($>1.7 \text{ g cm}^{-3}$) sediment for DDT compounds (Wu et al., 2016).

The present study was conducted to examine the hypothesis that anthropogenically derived debris is vital to the patchy distribution of organic compounds in estuarine sediment. Target analytes include OPFRs, PAEs, OTs and dichlorodiphenyltrichloroethanes (DDTs), which are commonly used plastic additives in synthetic materials and biocides in antifouling paint (Diez et al., 2002; Yu et al., 2011). A fishing harbor in Hailing Bay, Guangdong Province of China was selected as the study region, one of the top 10 fishing harbors in China and also serves as an important aquaculture zone (Yangjiang Municipal People's Government). The objectives of the present study were to (1) determine the size and

density occurrence of the target compounds in sediment (except for DDTs) and anthropogenic debris; (2) examine how debris mediated the distribution patterns of the target compounds in estuarine sediment and (3) assess the procedures for preparing sediment samples and sorting out debris.

2. Materials and methods

2.1. Materials

Standards of the selected target analytes were supplied by Chiron AS, Trondheim, Norway (TIBP), AccuStandard, New Haven, CT, USA (TnBP, TPhP and TDCPP), Dr. Ehrenstorfer GmbH, Augsburg, Germany (TCEP, TCIPP, TBOEP, EHDPP, TCPs and TEHP) and Adamas Reagent, Shanghai, China (DCPs mixtures). The standards of OTs were purchased from J&K Scientific, Beijing, China (n-butyltin-trichloride) and ChemService, West Chester, PA, USA (Tetraphenyltin). The standards of PAEs, i.e., DMP, DEP, DIBP, DBP, DMPP, DMEP, DEEP, DPP, DHxP, BBP, DBEP, DCHP, DEHP, DNOP, DPhP and DNP, were acquired from O2si Smart Solutions, Charleston, SC, USA. The abbreviations of individual OPFRs, PAEs and OTs and related information about DDTs are presented in the Supplementary Data ("S" indicates tables and figures in the Supplementary Data afterwards) Table S2. DEHP- d_4 , PCB-67, PCB-191, *o,p'*-DDT- d_8 , TnBP- d_{27} and TPhP- d_{15} were used as surrogate standards, and *p*-terphenyl- d_{14} , PCB-82 and benzyl benzoate were used as internal standards. Among them, TnBP- d_{27} and TPhP- d_{15} were purchased from Cambridge Isotope Laboratories, *o,p'*-DDT- d_8 from C/D/N Isotopes Quebec, Canada, DEHP- d_4 from Dr. Ehrenstorfer GmbH and other standards from AccuStandard. Sodium metatungstate monohydrate powers were purchased from Alfa Aesar (Bellefonte, PA, USA).

2.2. Sample collection

The first set of surface sediment samples, used to quantify the target compounds, was collected from a fishery base in Hailing Bay, Guangdong Province of China (Fig. S1) from August 2012 to November 2014, in proximity to two boat maintenance facilities (labeled as A and B in Fig. S1). Detailed procedures for sample collection were described elsewhere (Wu et al., 2016). To further examine the role of anthropogenic debris, another set of nine surface sediment samples (labeled as MD1 to MD9; Fig. S1) were taken in the same study region in November 2015 and only used to sort out anthropogenic debris.

2.3. Separation of sediment particles and debris

The first set of sediment samples were wet-fractionated into four sizes, i.e., 200–2000 μm , 63–200 μm , 30–63 μm and $<30 \mu\text{m}$. The size-segregated sediment samples from an aquaculture zone (Site 9) and shipping channel (Site 2) were further separated into two density fractions by floating them in a sodium metatungstate solution ($\rho = 1.7 \text{ g cm}^{-3}$) (Wu et al., 2016). The procedure described in Wu et al. (2016) was slightly modified to separate anthropogenic debris from sediment. Briefly, sediment samples were wet sieved onboard with seawater, and the fractions remaining on 10- and 80-mesh sieves were collected, resulting in two size fractions, i.e., $>2000 \mu\text{m}$ and between 200 and 2000 μm . Such fractionation was based on the fact that paint flakes were mostly affiliated with coarse-size sediment (Wu et al., 2016) and

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