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Contamination and distribution of heavy metals, polybrominated diphenyl ethers and alternative halogenated flame retardants in a pristine mangrove

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ABSTRACT

Owing to the expanding metal and electronics industries, pollution in the Pearl River Estuary needs special concern. Given the hydrodynamic effect, the pristine mangrove in Qi'ao Island would be contaminated by tidal flushing. Thus, we examined (1) the contamination of pollutants in this mangrove, including heavy metals, polybrominated diphenyl ethers (PBDEs) and alternative halogenated flame retardants (AHFRs), and (2) how habitat characteristics and sediment properties affect their distribution. Results showed that the sediment in Qi'ao mangrove had higher concentrations of heavy metals, PBDEs and AHFRs than that in other pristine mangroves, and similar concentrations to those mangroves impacted by point sources. Heavy metal concentrations were lower in the vegetated areas than mudflat, while the opposite was found for PBDEs and AHFRs. The findings imply that tidal flushing was an important pollution source, while mangrove plants have the capacity to minimize the impact of heavy metals, but not PBDEs and AHFRs.

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Urbanization and industrialization have been growing expeditiously in developing countries over the last few decades, where China is the leading one with respect to productivity and economic growth. In particular, Guangdong province is one of the most productive regions in China, accounting for more than 10% total gross domestic product (National Bureau of Statistics of China, <http://data.stats.gov.cn>) mainly contributed by metal and electronics industries (Chen et al., 2013; Xiao et al., 2013). However, the rapid economic growth in this region inevitably leads to ensuing environmental pollution in the coastal area of the Pearl River Estuary (PRE). As such, heavy metals and polybrominated diphenyl ethers (PBDEs) are of special concern due to their toxicity, persistence and bioavailability (Montouris et al., 2002; Darnerud, 2003). Since the production of technical PBDE products has been restricted (VECAP, 2014), alternative halogenated flame retardants (AHFRs) have drawn more global attention in recent years because they are increasingly used as the substitutes for PBDEs in electronic products (Covaci et al., 2011).

Undoubtedly, environmental pollution in coastal areas is mainly attributed to point sources (e.g. sewage discharge from smelters, electroplating plants and e-waste recycling plants) (Mai et al., 2005; Li et al., 2013; Wu et al., 2014; Zhang et al., 2015), but the influence of non-point sources, especially tidal flushing, cannot be overlooked since heavy metals, PBDEs and AHFRs are readily bound to suspended particles which are dispersed by water current (Williams et al., 1994; Rahman et al., 2001; Covaci et al., 2011). In addition, non-point sources are more difficult to control and able to affect wider areas, including pristine areas. Given the huge amount of suspended particles discharged from the distributaries into the PRE, the pollutant input from tidal flushing warrants investigation. In the coastal area, however, the impact of pollutant input may be reduced by the presence of mangrove plants because of their capacity to immobilize and even extract pollutants in water (Weis and Weis, 2004). Furthermore, mangrove plants can potentially alter the distribution of pollutants by changing habitat characteristics and/or sediment properties (Williams et al., 1994). How they affect the distribution of heavy metals has been reported (e.g. Ratheesh Kumar et al., 2010; Leung and Tam, 2013), but studies on PBDEs and AHFRs remain scant.

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In the present study, Qi'ao Island Mangrove Nature Reserve ("Qi'ao mangrove" hereafter), a pristine mangrove in South China featuring high diversity of mangrove species, was selected as the study area. Although this island is sparsely populated and dissociated from industrial activities, it is located in the western shore of the PRE where the highest turbidity and concentrations of pollutants are found due to hydrodynamic effect (Wang et al., 2013), making Qi'ao mangrove ideal for studying the impact of pollutant input from tidal flushing. Besides, how mangrove plants affect habitat characteristics, sediment properties and hence distribution of heavy metals, PBDEs and AHFRs was examined.

Three sampling areas in Qi'ao mangrove (22°26'N, 113°38'E) with different habitat characteristics were chosen for sample collection: (1) a native mangrove dominated by *Aegiceras corniculatum* and *Kandelia obovata* with random distribution (ca. 20 ind. per 100 m²; average height: 3 m); (2) an artificial mangrove dominated by mature *Sonneratia apetala* with uniform distribution (ca. 25 ind. per 100 m²; average height: 8 m); (3) a mudflat without vegetation as the reference site. A total of nine sampling points were uniformly chosen in each sampling area (Fig. 1). Five sub-samples of sediment were randomly collected by a rectangular sampler (10 cm long × 10 cm wide × 15 cm deep) within a 5 m × 5 m quadrat at each sampling point, followed by mixing to make a composite sample.

Redox potential and pH were measured *in situ* at 8 cm depth using a pH/mV/Temperature meter (IQ150, Scientific Instruments, USA). Particle size was measured using a particle size analyzer (Mastersizer 2000, Malvern Instruments, UK). Then, the sediment samples were freeze-dried, ground into powder and passed through a 2 mm sieve. Total organic carbon (TOC) was determined using a CHN elemental analyzer (Perkin-Elmer 2400, Perkin Elmer Corp., USA) after rising the sediment with 10% hydrochloric acid to remove carbonate.

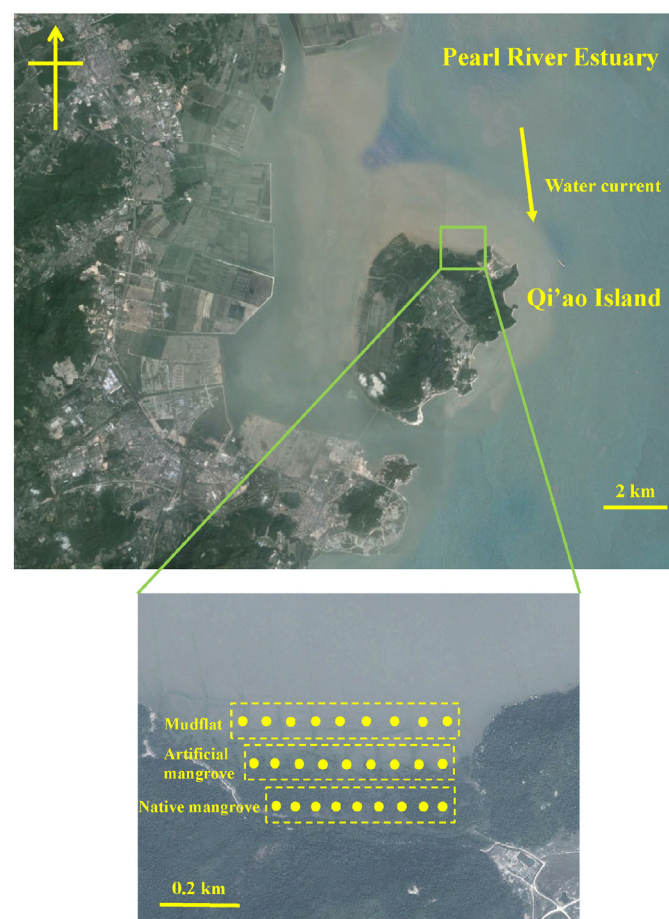


Fig. 1. The sampling locations in Qi'ao mangrove (Retrieved from Google Earth).

To extract heavy metals, ca. 0.3 g sediment sample was digested by a mixture of concentrated hydrochloric acid and nitric acid (3:1, v/v) using automatic digestion block (ST40, Beijing Polytech Instrument Ltd., China). The concentrations of chromium (Cr), copper (Cu), manganese (Mn), lead (Pb) and zinc (Zn), were determined by inductively coupled plasma optical emission spectrometry (Optima 5300DV, Perkin-Elmer Instruments, USA), while cadmium (Cd) by atomic absorption spectrometer (AAAnalyst 800, Perkin-Elmer Instruments, USA). A recovery test was conducted using a certified reference material from the State Oceanic Administration of China (GBW 07334) and the recovery for all heavy metals ranged from 86.3% to 98.2% with RSD less than 8.5%.

The methods described in Tian et al. (2012) and Zhang et al. (2015) with minor modifications were used to extract PBDEs and AHFRs. Briefly, ca. 15 g sediment plus small quantity of copper were spiked with surrogate standards (i.e. BDEs 77, 181, 205 and ¹³C-BDE 209), followed by Soxhlet extraction for 48 h using 200 mL hexane/acetone (1:1, v/v). The extract was concentrated to approximately 1 mL using a rotary evaporator, followed by purifying in an alumina/silica column. 30 μL internal standard containing BDE 118, BDE 128, 4-F-BDE 67 and 3-F-BDE 153 were added into each sample. The concentrations of BDEs 28, 47, 66, 99, 138, 153, 154, 183 (tri- to hepta-BDE congeners), hexabromobenzene (HBB), pentabromoethylbenzene (PBEB), pentabromotoluene (PBT) and 2,3,5,6-tetrabromo-*p*-xylene (pTBX) were analyzed by Agilent 7890 gas chromatograph equipped with an Agilent 5975 mass spectrometer using electron capture negative ionization (ECNI) in the selective ion monitoring (SIM) mode, separated by a DB-XLB column (30 m × 0.25 mm × 0.25 μm, J&W Scientific). The concentrations of BDEs 196, 197, 202, 203, 206, 207, 208, 209 (octa- to deca-BDE congeners), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), decabromodiphenyl ethane (DBDPE), *anti*-dechlorane plus (*a*-DP), *syn*-dechlorane plus (*s*-DP), *anti*-undecachloropentacyclooctadecadiene (*a*-Cl₁₁-DP) and *anti*-decachloropentacyclooctadecadiene (*a*-Cl₁₀-DP) were analyzed by a Shimadzu 2010 gas chromatograph coupled with a QP 2010 mass spectrometer also using ECNI in the SIM mode, but separated by a DB-5HT column (12.5 m × 0.25 mm × 0.10 μm, J&W Scientific). The GC conditions were described in Tian et al. (2012). Procedural blanks were analyzed after each set of ten samples to ensure the consistency of instrumental performance. Surrogate recoveries were 81.2% for BDE 77, 115.6% for BDE 181, 106.7% for BDE 205 and 93.6% for ¹³C-BDE 209 (RSD < 15.0% for all surrogate standards).

One-way analysis of variance (ANOVA), followed by Tukey's test, was used to test the spatial variation in the concentrations of heavy metals, total PBDEs and total AHFRs. Normality and homoscedasticity were tested by Shapiro-Wilk and Levene's tests, respectively. Logarithmic transformation was applied if either one of the assumptions was violated. Spearman correlation analysis was applied to correlate all the pollutants with sediment properties. The aforementioned analyses were performed using software SPSS 20.0 for Windows.

The habitat characteristics and sediment properties in different habitats are shown in Table 1. The sediment was more acidic in the native mangrove (pH 5.52) than the artificial mangrove and mudflat (pH 7.32–7.33) probably because of the huge amount of leaf litter in the native mangrove, which can form humic acid upon decomposition. However, the limited amount of leaf litter in the artificial mangrove did not significantly affect the pH in sediment. The decrease in redox potential from 72.7 mV in the native mangrove to −58.7 mV in the mudflat was attributed to the greater amount of belowground vegetation in the vegetated areas, which can oxygenate the sediment (Vervaeke et al., 2004). TOC was significantly higher in the native mangrove (4.98%) than the other habitats (1.46–1.68%) because of more leaf litter in the former. The sediment remained silty and clayey across habitats.

The concentrations of heavy metals were generally higher in the mudflat than the vegetated areas (Table 2). Significantly higher concentrations of Cr, Cu, Mn and Zn were found in the mudflat than the native

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