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Removal of Wastewater-borne Polybrominated Diphenyl Ethers by Mangrove Wetland Microcosms

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Abstract. Polybrominated diphenyl ethers (PBDEs) are persistent and toxic organic pollutants in environments. They are difficult to remove by conventional wastewater treatment processes, and alternatives are needed. A greenhouse experiment was conducted to investigate the removal of wastewater-borne PBDEs by constructed mangrove wetland planted with one-year old seedlings of Kandelia obovata (Ko) under different tidal regimes, that is, everyday (Te), every-two-day (To), every-two-week (Tt) and no tidal flushing (Tn). During the 8-month experiment, all six congeners of PBDEs in wastewater, including BDE-47, -100, -99, -154, -153 and -209, were not detected in effluents, suggesting the constructed mangrove wetland was effective in removing PBDEs under all tidal regimes. Around 10 to 42% of the wastewater-borne PBDEs were retained in sediments, depending on types of congeners and duration of tidal regimes. Significantly higher concentrations of PBDEs were found in upper than bulk sediments, and rhizosphere sediment had the lowest retention. Tidal regimes also affected the accumulation of PBDEs in sediment. The lowest concentration was found in Te regime with frequent tidal flushing creating the most anaerobic sediment, suggesting that PBDEs retained in sediment were then removed by reductive debromination. This study revealed that mangrove wetland could be constructed to remove PBDEs, and the immobilization of PBDEs congeners in sediment depended on sediment locations, types of congeners and tidal regimes.

Keywords: tidal regime, persistent organic pollutants, mangrove plants, Kandelia

1. Introduction

Polybrominated diphenyl ethers (PBDEs), common brominated flame retardants, are widely used in industries as additives to products like plastics, textiles and foams, etc. They are persistent organic pollutants (POPs) with high toxicity. The potential sources of contamination are discharges from industries and households, and PBDEs are ubiquitous in environments. Theoretically, there are 209 congeners of PBDEs according to the numbers of bromines and their substitution positions. The three major commercial PBDEs mixtures are penta-BDE (BDE-47, -99, -100, -153 and -154), octa-BDE (BDE-183) and deca-BDE (BDE-209), having 5, 8 and 10 bromines, respectively. Due to their toxicity, the use of penta- and octa-BDEs have been phased out in Europe and North America since 2006 [1]. However, the potential environmental hazards posed by these BDE compounds will not disappear immediately because of new productions with recycled BDE-containing materials and disposal of e-waste [2]. Deca-BDE is still produced and applied around the world. Conventional wastewater treatment plants are not designed to effectively remove PBDEs in wastewater, and high concentrations of PBDEs are often found in influent, effluent and sewage sludge [3], [4]. Research on the removal of PBDEs is limited as previous studies mainly focused on the contamination levels of PBDEs in the environment. In recent decades, constructed wetland (CW) has been used as a cost-effective and environmentally friendly technology for treating industrial, municipal and metal-contaminated wastewater [5]. However, there is currently a dearth of information on the application of CWs to treat wastewater containing PBDEs.

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Natural mangrove wetlands are one of the most important ecosystems found along tropical and subtropical coastlines, and are often sinks of many POPs, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), etc. [6], [7]. Mangrove plants were shown to have high tolerance to toxic organic pollutants like oil and PBDEs [8], [9]. Constructed mangrove wetlands could effectively remove nutrients and toxic pollutants such as heavy metals in municipal and industrial wastewater [10]-[12]. Mangrove sediment also harbored a high diversity of microorganisms capable of degrading POPs like PAHs [13], [14]. However, its ability to remove and degrade PBDEs is still unclear.

Mangrove wetland in inter-tidal location is regularly flooded by incoming tides. This leads to alternating wetting and drying periods and creates anaerobic and aerobic conditions in mangrove sediment, respectively. The degree and duration of aerobic and anaerobic conditions depend on tidal flushing regimes, the longer the flooding time, the more the anaerobic condition. Anaerobic condition is important for the debromination of PBDEs. The highly brominated PBDEs such as deca- or octa-BDEs could be debrominated to the less brominated congeners [15], [16]. So far, effects of tidal regimes on the removal and the behaviour of wastewater-borne PBDEs in mangrove wetland systems have never been reported. The present study, based on greenhouse microcosms, aims to evaluate the removal of a mixture of PBDE congeners in wastewater by constructed mangrove wetlands under different tidal regimes and their immobilization in sediments.

2. Materials and Methods

A total of 24 small pots, each with a dimension of 14 cm in diameter, 13.5 cm in height and filled with approximately 2 kg fresh sediment to a depth of 10 cm, were used as the microcosms. The sediment was collected from a relatively clean mangrove swamp in Hong Kong, and its properties were reported by Wang et al. [17]. Each pot was planted with one seedling of K. obovata (1-year old) and acclimated for one month. After acclimation, the pots were divided into four tidal flushing regimes: (i) everyday (Te): pots received tidal flushing daily, with seawater flooded onto the sediment surface to a water depth of 4 cm to simulate high tide for 18 hours and seawater was retreated to let sediment exposed for 6 hours to simulate low tide; (ii) every-two-day (To): pots received tidal flushing for one-day but without for the following day, and this cycle repeated; (iii) every-two-week (Tt): pots received two tidal flushing per month (on the 1st and 15th day); and (iv) no tidal flushing (Tn): pots did not receive any tidal flushing and were the control. Each tidal regime had six replicated microcosms. To simulate tidal flushing, pots were placed into a plastic basket filled with roughly 2 L artificial seawater at a salinity of 15 parts per thousands (‰) during high tide flushing, and were taken out of the baskets to dry at low tides. Deionized water was added to the control and pots when they did not receive tidal flushing to keep the sediment moist.

During low tide period (at about 15:00 of the day), each pot was irrigated daily with 80 mL artificial wastewater containing six congeners of PBDEs, including BDE-47, -100, -99, -154, -153 and -209, purchased from AccuStandard (New Haven, CT, USA) and other pollutants such as PAHs. The measured concentrations of BDE-47, -100, -99, -154, -153 and -209 in the artificially prepared wastewater were 10.94, 3.34, 16.86, 1.21, 1.56 and 18.77 μg L-1, respectively. The experiment lasted eight months. At every two months, 150 mL effluent, consisting of wastewater collected in three consecutive days (around 50 mL per day), was collected from each pot, and concentrations of BDE-47, -99, -100, -153, -154 and -209 were determined by liquid-liquid extraction. In brief, 100 mL of each water sample was extracted with 20 mL n-hexane, with PCB-209 as the internal standard. The extraction was repeated three times and extracts were combined. Gas chromatograph (6890N, Agilent Technologies Inc.) coupled with mass selective detector (5875, Agilent Technologies Inc.) was used to quantify PBDEs. The mass spectrometer was equipped with a chemical ionization source. A 15-m DB-5HT column (0.25 mm i.d. × 0.10 μm) was used. 2μL sample was injected to the column in splitless mode at 240 ℃. The column oven temperature was programmed from 150 ℃ (2 min) to 300 ℃ (3 min) at a rate of 6 ℃/min, with a post-run time of 2 min.

For each tidal regime, microcosms in triplicates were retrieved at the end of 4- and 8-month of the experiment. Sediment in each pot was divided into three portions: (i) upper sediment: about 4 cm depth from the top of sediment and did not contain any root, (ii) bulk sediment: all below the upper sediment but distant from roots and (iii) rhizosphere sediment: attached onto root surface [17]. To extract PBDEs, 2-10g freeze-dried sediment samples were firstly mixed with diatomaceous earth and extracted with n-hexane by

Accelerated Solvent Extractor (ASE200, Dionex, Sunnyvale, USA) according to Application Note 3050. PCB-209 was added as an internal standard prior to extraction. After ASE extraction, extracts were evaporated with a rotary evaporator to about 1 mL, further dried by nitrogen gas, and n-hexane was added to bring it up to 200 μL. The identification and quantification of PBDEs in sediment were performed by GC-MS analyses, same as water samples. Recoveries of the six PBDE congeners in sediment and water samples were checked by spiking a mixed standard of PBDE congeners into originally freeze-dried sediment samples from Sai Keng and deionized water, respectively. The mean recoveries of three replicates ranged from 76.4 to 82.9% for all PBDE congeners, with relative standard deviations <7%.

3. Results and Discussion

PBDEs were not detected in all effluent samples, indicating the simulated mangrove microcosms, irrespective to tidal regimes, effectively removed PBDEs in wastewater. It is possible that PBDEs were immobilized in sediment because of their hydrophobic properties, with log K_{ow} values varying from 4 to 10. Previous studies found that PBDEs in water tended to retain and accumulate in soil, which became a primary reservoir of PBDEs [18], [19]. Mueller et al. [20] also reported that the abiotic sorption of penta-BDEs onto soil constituents as the most important factor in determining the short-term fate of PBDEs in soil.

The concentrations of BDE-47, -99 and -209 in the sediment before the experiment were 0.14 ± 0.03, 0.07 ± 0.02, 1.24 ± 0.32 μg kg^{-1}, respectively (mean and standard deviation of three replicates), and the other PBDE congeners were below detection limits. These background levels were very low compared to the concentrations found in the sediment at the end of 4- and 8-month of wastewater discharge (Table 1). The sediment concentrations between the two sampling times clearly showed that the longer the discharge of wastewater, the higher the concentration. This indicated that PBDEs in wastewater were immobilized in sediments. For all congeners and under all tidal regimes, sediments collected from the upper layer where artificial wastewater firstly passed through during irrigation retained most of the PBDEs in wastewater, followed by bulk sediment, and rhizosphere sediment had the lowest concentration. This declining trend of upper > bulk > rhizosphere sediments was the same under all tidal regimes. Similarly, Yang et al. [21] also found that highest concentrations of 37 PBDE congeners appeared in the surface soil, which were 18-29 times higher than the second-lower-layer soil in a typical electronic wastewater recycling area in South China. The residual BDE-209 concentrations was the lowest in the root compartment, and obviously rose with increasing distance from the roots [22]. These previous results were in good agreement with the present study that upper layer sediment had highest concentrations of all PBDE congeners.

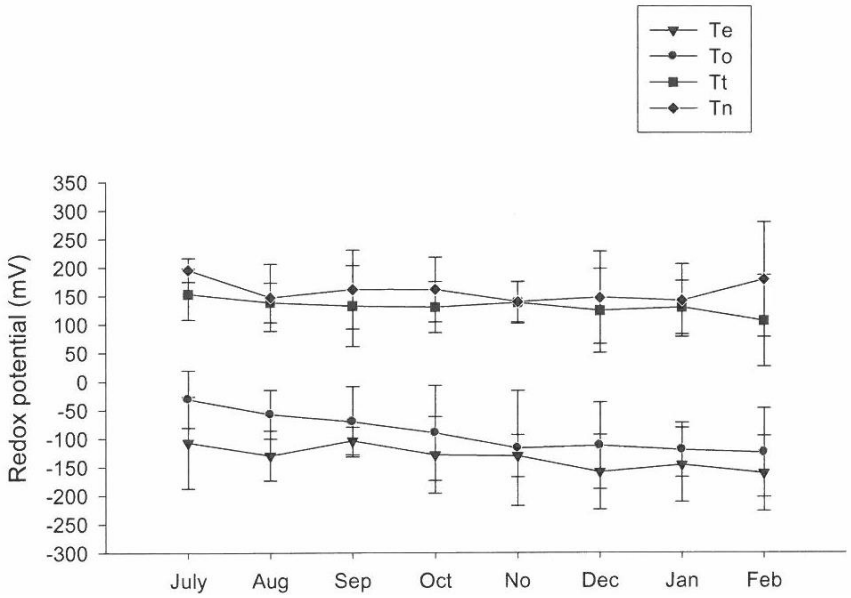

Fig. 1: Redox potential in sediment at a depth of 3 cm during 8-month wastewater discharge experiment (Mean and standard deviations of 3 replicates were shown; Te: everyday tidal flushing; To: one day with tidal flushing and one day without; Tt: every-two-week tidal flushing; Tn: control and without tidal flushing)

Table 1: Concentrations of six PBDE congeners (μg kg-1) in upper, bulk and rhizosphere sediments at the end of 4- and 8-month experiment (Means and standard deviations of triplicates were shown; Te: everyday tidal flushing; To: one day with tidal flushing and one day without; Tt: every-two-week tidal flushing; Tn: control and without tidal flushing; different letters in the superscript position of the same row at the same month indicate significant differences among four tidal regimes according to one-way analysis of variance at $p \leq 0.05$)

Congeners	Sediment	4-month				8-month			
		Te	To	Tt	Tn	Te	To	Tt	Tn
BDE-47	Upper	12.50 ± 3.57^{a}	14.07 ± 3.24^{a}	32.49 ± 2.41^{b}	22.69 ± 7.90^{c}	16.57 ± 1.8^{A}	28.70 ± 2.45^{B}	42.19 ± 5.46^{C}	28.95 ± 2.80^{B}
	Bulk	0.77 ± 0.16^{a}	1.38 ± 0.27^{b}	1.59 ± 0.39^{b}	1.11 ± 0.05^{b}	1.24 ± 0.47^{A}	2.21 ± 0.32^{B}	2.94 ± 0.85^{C}	1.89 ± 0.29^{B}
	Rhizosphere	0.38 ± 0.02^{a}	0.62 ± 0.11^{b}	0.52 ± 0.06^{b}	0.52 ± 0.07^{b}	0.78 ± 0.27^{A}	1.12 ± 0.14^{B}	1.28 ± 0.43^{B}	0.97 ± 0.34^{AB}
BDE-99	Upper	20.44 ± 1.06^{a}	23.84 ± 5.34^{a}	91.87 ± 7.63^{b}	83.88 ± 7.46^{b}	24.70 ± 2.43^{A}	43.20 ± 2.87^{B}	112.8 ± 5.7^{C}	105.6 ± 11.5^{C}
	Bulk	1.04 ± 0.44^{a}	2.17 ± 0.32^{b}	4.68 ± 0.84^{c}	4.51 ± 0.34^{c}	1.75 ± 0.76^{A}	2.96 ± 0.18^{B}	6.53 ± 0.86^{C}	5.65 ± 1.09^{C}
	Rhizosphere	0.45 ± 0.08^{a}	0.58 ± 0.03^{a}	0.64 ± 0.13^{a}	1.13 ± 0.28^{b}	0.55 ± 0.08^{A}	1.29 ± 0.06^{B}	1.82 ± 0.33^{BC}	2.09 ± 0.66^{C}
BDE-100	Upper	4.64 ± 0.96^{a}	5.42 ± 1.51^{a}	11.88 ± 0.74^{b}	9.89 ± 0.68^{b}	5.54 ± 1.12^{A}	8.52 ± 0.42^{B}	12.23 ± 0.49^{C}	9.85 ± 0.16^{B}
	Bulk	0.23 ± 0.06^{a}	0.47 ± 0.09^{b}	0.86 ± 0.19^{c}	0.82 ± 0.08^{c}	0.27 ± 0.08^{A}	0.61 ± 0.04^{B}	0.83 ± 0.28^{C}	0.43 ± 0.09^{D}
	Rhizosphere	0.04 ± 0.01^{a}	0.04 ± 0.00^{a}	0.08 ± 0.02^{b}	0.17 ± 0.05^{c}	0.32 ± 0.05^{A}	0.26 ± 0.07^{A}	0.32 ± 0.02^{A}	0.27 ± 0.16^{A}
BDE-153	Upper	2.56 ± 0.56^{a}	3.64 ± 0.16^{b}	7.34 ± 1.46^{c}	4.85 ± 1.68^{b}	3.49 ± 0.22^{A}	4.55 ± 0.06^{B}	5.50 ± 0.50^{C}	4.49 ± 0.43^{B}
	Bulk	0.23 ± 0.05^{a}	0.37 ± 0.01^{b}	0.58 ± 0.12^{c}	0.64 ± 0.12^{d}	0.18 ± 0.04^{A}	0.35 ± 0.25^{B}	0.44 ± 0.04^{C}	0.25 ± 0.06^{AB}
	Rhizosphere	0.05 ± 0.01^{a}	0.12 ± 0.01^{b}	0.09 ± 0.02^{b}	0.13 ± 0.02^{b}	0.11 ± 0.04^{A}	0.16 ± 0.05^{AB}	0.23 ± 0.10^{B}	0.16 ± 0.06^{AB}
BDE-154	Upper	3.26 ± 0.26^{a}	3.16 ± 0.30^{a}	6.78 ± 0.49^{b}	5.18 ± 0.44^{c}	3.57 ± 0.38^{A}	4.29 ± 0.14^{AB}	5.14 ± 0.50^{B}	4.10 ± 0.46^{AB}
	Bulk	0.18 ± 0.04^{a}	0.30 ± 0.10^{b}	0.46 ± 0.09^{c}	0.44 ± 0.05^{c}	0.14 ± 0.05^{A}	0.31 ± 0.22^{B}	0.42 ± 0.09^{B}	0.21 ± 0.05^{A}
	Rhizosphere	0.02 ± 0.00^{a}	0.04 ± 0.01^{a}	0.10 ± 0.01^{b}	0.07 ± 0.01^{c}	0.12 ± 0.06^{A}	0.14 ± 0.04^{A}	0.18 ± 0.06^{B}	0.15 ± 0.06^{AB}
BDE-209	Upper	44.01 ± 3.18^{a}	47.89 ± 0.03^{a}	105.4 ± 24.3^{b}	67.59 ± 3.42^{c}	50.24 ± 3.24^{A}	66.35 ± 7.40^{A}	126.9 ± 6.5^{B}	88.36 ± 14.33^{AB}
	Bulk	4.83 ± 0.30^{a}	5.48 ± 0.24^{a}	9.18 ± 1.03^{b}	6.45 ± 0.48^{a}	18.72 ± 2.89^{A}	22.24 ± 6.65^{A}	29.99 ± 6.59^{AB}	29.90 ± 4.82^{AB}
	Rhizosphere	3.65 ± 0.09^{a}	4.22 ± 0.38^{b}	5.21 ± 0.50^{b}	4.95 ± 0.71^{b}	9.58 ± 1.55^{A}	12.46 ± 0.08^{AB}	13.91 ± 1.86^{BC}	18.41 ± 4.46^{C}

Table 2: Percerntages of total amounts of wastewater-borne PBDEs retained in sediments at the end of 8-motnh experiment (Means of triplicates were shown; Te: everyday tidal flushing; To: one day with tidal flushing and one day without; Tt: every-two-week tidal flushing; Tn: control and without tidal flooding; total amounts of BDE-47, -99, -209 and sum of six PBDEs added at the end of the experiment was 140.10, 215.89, 240.24, 674.44 μg, respectively)

Congeners	Sediment	Te	To	Tt	Tn
BDE-47	Upper	9.46	16.39	24.09	16.53
	Bulk	0.97	1.74	2.31	1.49
	Rhizosphere	0.06	0.08	0.09	0.07
BDE-99	Upper	9.15	16.01	41.80	39.12
	Bulk	0.89	1.51	3.33	2.88
	Rhizosphere	0.03	0.06	0.08	0.10
BDE-209	Upper	16.73	22.10	42.26	29.42
	Bulk	8.57	10.18	13.73	13.69
	Rhizosphere	0.40	0.52	0.58	0.77
Sum of six PBDEs	Upper	12.35	18.46	36.15	28.62
	Bulk	3.64	4.68	6.71	6.25
	Rhizosphere	0.17	0.23	0.26	0.33

The total percentages retained in sediments were less than 50% of the wastewater-borne PBDEs (Table 2), indicating that not all PBDEs in wastewater were retained in sediments. The percentages of BDE-47, -99, -209 and total PBDEs (sum of six congeners) retained in upper sediment under all tidal regimes were 9.5-24.1%, 9.2-41.8%, 16.7-42.3% and 12.4-36.2%, respectively. These values were much higher than that in bulk and rhizosphere sediments. The higher brominated congeners such as BDE-209 had higher accumulation in sediment than BDE-47, the lower brominated congener. These results suggested that other processes such as plant uptake and transformation by microorganisms in root and sediment might also involve in the removal of PBDEs in wastewater. However, the direct contribution of plants to the removal of PBDEs through uptake and tissue accumulation was not so significant [22]. Huang et al. [23] found that the reduction of PBDEs in the planted treatment was mainly dependent on microbial metabolism and the bio-

stimulation of microbial biomass by planting. The direct and indirect roles of plants on the removal of PBDEs in wastewater, as well as the microbial transformation and degradation, deserves more in-depth research.

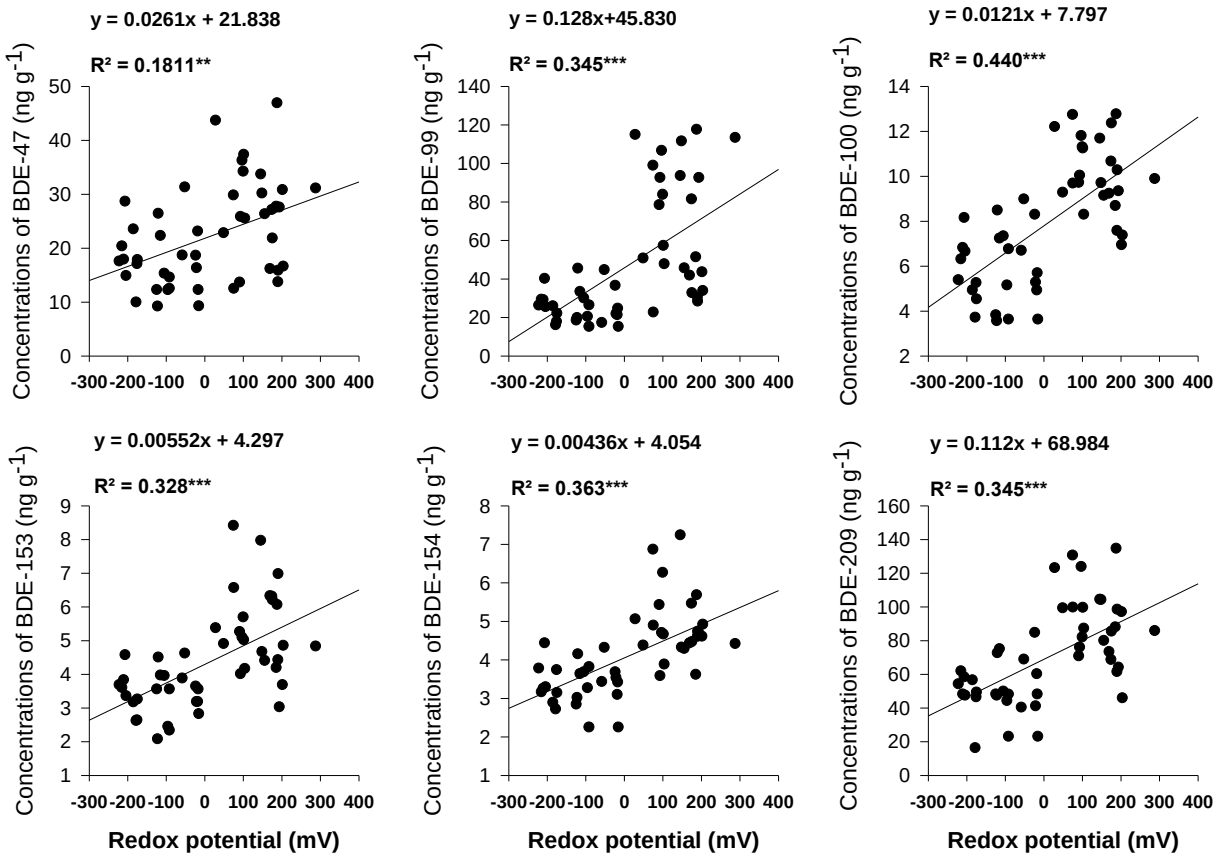

Fig. 2: Cause–effect relationships between redox potential and concentrations of six PBDE congeners in upper sediment; curves were fitted with linear regression models (y=y0+ax); R2: regression coefficients; *, ** and *** indicate the R2 are significant at p≤0.05, 0.01 and 0.001 levels, respectively).

Not only sediment position, the residual concentrations of six PBDEs congeners in sediment were also affected by tidal regimes, with significantly lower concentrations of both individual PBDE congeners and total PBDEs (sum of six congeners) in microcosms with every day tidal flushing (Te) than those with less frequent tidal flushing (Table 1). It is interesting to found that microcosms under Tt had higher residual concentrations of BDE-47, -100 and -209 in upper sediment than those without any tidal flushing (Tn). Such interesting results were not observed in bulk and rhizosphere sediments as the PBDE concentrations in these two sediments were 10-40 times less than that in upper sediment. For example, the residual concentrations of BDE-100 in upper sediments varied from 3.68 to 12.62 μg kg^{-1} after 8-month wastewater discharge, while the range in rhizosphere sediment was 0.03-0.22 μg kg^{-1}. The percentages of PBDEs retained in bulk sediment were <14% (most were only a few percent) while those in rhizosphere sediment were <0.5% (Table 2). These results might explain why the tidal regime effect on PBDE retention in bulk and rhizosphere sediments was less obvious than that in upper sediment. Tidal regime could alter the duration of waterlogging and the degree of anaerobic conditions which are important for the reductive debromination of PBDEs in sediments. Wang et al. [17], [24] revealed that the structure and function of the microbial community in sediment were affected by tidal regimes, and the key influential factor was redox potential. In this study, the redox potential in upper sediment (at 3 cm) determined by the ORP meter (TPS WP-81, Brisbane, Australia) varied from -250 to 270 mV, dependent on tidal regimes (Fig. 1). The microcosms receiving Te and To regimes had significantly more negative redox potential than those with Tt and Tn tidal flushing, according to one-way analysis of variance ($F=432.66$, $p\leq0.001$). The redox potential also gradually decreased with sampling time ($F=3.352$, $p=0.002$), especially in Te and To. Cause-effect relationships between redox potential in upper sediment and residual concentrations of each PBDE congeners were all positive (Fig. 2). The slope and R^2 values of these relationships were congener-specific, and the best fit

correlation was found in BDE-100 among six PBDE congeners. These results indicated that more negative redox potential in sediment with more frequent tidal flushing (Te and To) created more anaerobic conditions for the debromination of PBDEs, leading to more dissipation and less retention in sediment. Chen et al. [25] concluded that anaerobic reductive debromination was the main process to remove PBDEs in mangrove soil under anaerobic conditions with redox potential ranging from -93 to -156 mV, and different lower brominated congeners were detected. Similarly, Zhu et al. [26] also found that more than 90% of the spiked BDE-47 was removed within seven months, and many lower brominated congeners were detected in mangrove sediment, indicating its intrinsic debromination potential. The roles of microorganisms in debromination and degradation of PBDEs in constructed mangrove wetland system and the fate of individual congener in wastewater still need further investigations.

4. Conclusion

The present study is the first time demonstrating that constructed mangrove wetlands were effective in removing different congeners of PBDEs in wastewater under all tidal flushing regimes. However, the amounts of PBDEs retained in sediments were significantly affected by tidal regimes, with more accumulation in the sediments with more positive redox potentials, such as in the treatments receiving tidal flushing every two weeks and without tidal flushing. Most of the wastewater-borne PBDEs were immobilized in surface sediments, some were taken up by plants and transformed / degraded by microorganisms.

5. Acknowledgements

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