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Effect of mangrove species on removal of tetrabromobisphenol A from contaminated sediments

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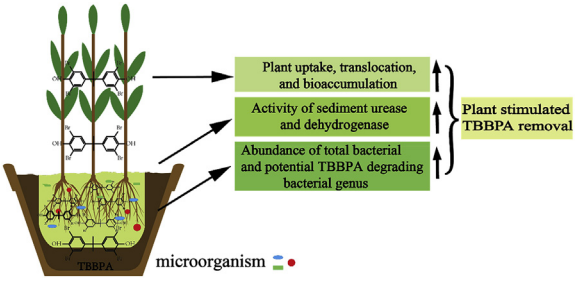
HIGHLIGHTS

- Higher TBBPA degradation in mangrove-planted sediments compared to unplanted ones.
- Mangrove plants can uptake, translocate, and bioaccumulate TBBPA.
- Mangrove plants can enhance microbial activities and abundance in rhizosphere.
- Plant uptake contributes less to TBBPA removal than microbial degradation.
- *K. obovata* is more suitable for remediation of TBBPA-contaminated sediments.

GRAPHICAL ABSTRACT



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ABSTRACT

The increase levels of tetrabromobisphenol A (TBBPA) in mangrove wetlands is of concern due to its potential toxic impacts on ecosystem. A 93-day greenhouse pot experiment was conducted to investigate the effects of mangrove plants, *A. marina* and *K. obovata*, on TBBPA degradation in sediment and to reveal the associated contributing factor(s) for its degradation. Results show that both mangrove species could uptake, translocate, and accumulate TBBPA from mangrove sediments. Compared to the unplanted sediment, urease and dehydrogenase activity as well as total bacterial abundance increased significantly ($p < 0.05$) in the sediment planted with mangrove plants, especially for *K. obovata*. In the mangrove-planted sediment, the *Anaerolineae* genus was the dominant bacteria, which has been reported to enhance TBBPA dissipation, and its abundance increased significantly in the sediment at early stage (0 −35 day) of the greenhouse experiment. Compared to *A. marina*-planted sediment, higher enrichment of *Geobater, Pseudomonas, Flavobacterium, Azoarcus*, all of which could stimulate TBBPA degradation, was observed for the *K. obovata*-planted sediment during the 93-day growth period. Our mass balance result has suggested that plant-induced TBBPA degradation in the mangrove sediment is largely due to elevated microbial activities and total bacterial abundance in the rhizosphere, rather than plant uptake. In addition, different TBBPA removal efficiencies were observed in the sediments planted with different

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mangrove species. This study has demonstrated that *K. obovata* is a more suitable mangrove species than *A. marina* when used for remediation of TBBPA-contaminated sediment.

1. Introduction

Tetrabromobisphenol A (TBBPA), a most widely used brominated flame retardant, is extensively used in manufactured electronic devices, plastics and textiles to prevent materials against ignition (Covaci et al., 2009). There were various studies on the occurrence of TBBPA in natural environment such as water, biota samples, soil and sediments, and the mainly emission sources were e-waste recycling sites and wastewater treatment plants (Kim et al., 2016; Malkoske et al., 2016). Due to the high hydrophobility of TBBPA (K_{ow} = 4.5–6.5), it is frequently adsorbed to soil and sediment particles which has been known as a final sink for TBBPA (Malkoske et al., 2016). The highest TBBPA concentration was reported up to 732 mg kg^{-1} (dw) in wastewater sewage sludge (Liu et al., 2016b), and recently reports showed that TBBPA concentration ranged from 0.02 to 304 ng g^{-1} (dw) in sediment from Pearl River Estuary, South China (Feng et al., 2012; Hu et al., 2019). Previous studies have showed that TBBPA could disrupt thyroid and estrogen hormone function, induce liver toxicity, cytotoxicity, and immunotoxicity, and increase uterine cancer risk in mammals (Lai et al., 2015). At present, many efforts have been made to remove TBBPA from water-body, soil and sediment including advanced oxidation degradation, thermal decomposition and biodegradation (Liu et al., 2018).

TBBPA could be degraded in soil, sediment, and sewage sludge via microbial-mediated reductive debromination, yielding lower brominated intermediates and bisphenol A (Arbeli and Ronen, 2003; Arbeli et al., 2006; Lefèvre et al., 2018). In aerobic sediment, methylation of TBBPA by aerobic bacteria yields mono- and dimethyl ether derivatives which are highly lipophilic than the parent, therefore, exhibited higher bioaccumulation in the food chain (George and Haeggblom, 2008; Li et al., 2015). An et al. (2011) reported that one strain of *Ochrobactrum* sp. *T* was capable of, within 72 h, simultaneously debrominating and mineralizating TBBPA in aerobic conditions. Several other studies have demonstrated that amendment of TBBPA-degrading bacteria into soil or sediment stimulated the remove efficiency of TBBPA (Li et al., 2014, 2016). Furthermore, research has shown that adding yeast extract, surfactant, humic acid, or sodium chloride, etc, could also enhance TBBPA co-biodegradation in bioreactors (Peng and Jia, 2013; Yang et al., 2016). However, there is limited practical field studies on TBBPA bioremediation.

Phytoremediation, recognized as a reliable approach for removing organic contaminates from in soil or sediment, is defined as *in situ* process of plants uptake and rhizodegradation (Wenzel, 2009; Feng et al., 2016). TBBPA uptake by plant roots from soil and further translocation to aboveground through transpiration stream have been widely reported in previous studies, but most of these studies have focused on herbaceous plant (Dogan et al., 2010; Li et al., 2011; Hou et al., 2019). Mangrove swamps, located in the tropical and subtropical estuary and coastline zones provide a vital eco-service for human society and are important habitats for diverse living creature (Bayen, 2012). The phytoremediation of PBDEs and PAHs by mangrove plants have been substantially investigated (Zhu et al., 2014; Chen et al., 2017), all of which has suggested that mangrove plants have great potential for remediation of organic pollutants in sediments. However, knowledge on the uptake and biodegradation of TBBPA by mangrove plants is still limited.

It is well known that both plant uptake and plant induced microbial degradation are mainly factors in controlling the dissipation of organic pollutants in soil and sediment (Santos et al., 2011; Qin et al., 2014; Storey et al., 2014). In addition, previous research has suggested that rhizosphere microbial activity indicated by activities of enzymes such as urease and dehydrogenase are closely related removal of persistent organic compounds from sediment (Lu et al., 2011; Wang et al., 2014; Chen et al., 2015). However, it is still unclear that whether mangrove species can stimulate TBBPA degradation by plant uptake and alteration of microbial community in sediment.

Therefore, this work aims to (i) investigate the effect of two mangrove species, *Kandelia obovata* (*K. obovata*) and *Avicennia marina* (*A. marina*), on TBBPA degradation in mangrove sediments; (ii) determine the ability of both species to take up and translocate TBBPA; and (iii) explore the rhizosphere microbial community structure and the microorgansisms that are responsible for the planting-induced dissipation of TBBPA.

2. Materials and methods

2.1. Sediment and plants collection and treatment

Surface sediment (0–20 cm) and mangrove propagules used in the experiment were obtained from a mangrove forest in the Jiulongjiang River Estuary (24°20′N, 117°45′E), Fujian Province, China. Before experiment, all the sediments were sieved through a 0.5 cm sieve to remove coarse debris and manually homogenized (Ghoveisi et al., 2014), and after that was aged for one month to make the sediment properties thoroughly stabilize (Ghoveisi et al., 2018). Selected physicochemical properties of the sediment used as shown in Table S1.

The freshly collected sediment was divided into two portions, with one sterilized by autoclaving three times, each time for 2 h at 121 °C and 147 kP (STIK MJ-54A, USA), on three consecutive days and the other portion remained as non-sterilized. Each portion of sterilized and non-sterilized sediments was spiked with TBBPA (TCI, Tokyo, Japan), respectively, and the procedure according to the previous study by Lu et al. (2011). The average TBBPA concentration of 6 replicates collected from different locations in sterilized and non-sterilized sediment were 10.83 ± 0.46 and 10.78 ± 0.18 mg kg^{-1} dw, respectively. The spiked sediment was thoroughly stirred and aging for 7 days before the greenhouse pot experiment (Ghoveisi et al., 2018). Same size mangrove propagules were selected and sand cultivated with Hoagland nutrigen solution for three months until each seedling grew 2 to 3 pairs of leaves and 15–18 cm height. Healthy and vitality *A. marina* or *K. obovata* seedlings with similar size were then selected for planting in the pots for the greenhouse experiment.

2.2. Greenhouse experimental design and sampling

Polyvinyl chloride pots (200 mm in diameter for the top of the opening, 160 mm in height, 180 mm in diameter for the bottom) were used. After adding the TBBPA-spiked sterilized or non-sterilized sediment to each pot, three healthy *A.* marina or *K. obovata* seedlings with similar sizes were planted into each pot. For the *A. marina* and *K. obovata*, 1.5 and 2.0 kg sediment/pot was used, respectively. No nutrients were added to each pot during the entire

greenhouse experiment. The pots were randomly arranged within the greenhouse to ensure uniform conditions. To simulate the tidal characteristic of mangrove swamp and minimize sediment alternation, a 24-h high tide and 24-h low tide was applied according to Chen et al. (2017). The daily temperature and relative humidity in the greenhouse were maintained at 25 ± 5 °C and 60–80%, respectively. There was 12 h daily light exposure at intensity of 800–1200 μmol m^{-2} s^{-1}. There were four treatments with triplicates/treatment: a) sterilized sediment without mangrove plants; b) non-sterilized sediment without mangrove plants; c) non-sterilized sediment planted with *A. marine*; and d) non-sterilized sediment planted with *K. obovata*. For the treatments with plants, plant rhizosphere and non-rhizosphere were divided by using 500 mesh nylon rhizobox according to method by Chen et al. (2015).

In the greenhouse, the plants were grown for up to 93 day (5–6 pairs of leaves and 25–30 cm height of plant), during which triplicate pots from each treatment group were retrieved at 0, 7, 14, 21, 35, 49, 63, 93 day to sacrificially collect the plants and sediment sample. All plant samples were first carefully washed with tap water and sequence washed with DI water carefully, separated into roots, stems, and leaves, immediately freeze dried, weighted, ground into powder, homogenized by sieving through a stainless steel 80-mesh (0.2 mm) sieve, and then analyzed for TBBPA. From each pot, the sterilized sediment, non-sterilized unplanted sediment, planted mangroves rhizosphere sediment were collected and a portion of the fresh sediment stored at 4 °C for determination of enzyme activity within one week. A second portion of the collected sediment was immediately freeze-dried, homogenized by grinding and sieving through a stainless steel 80-mesh (0.2 mm) sieve, and then analyed for TBBPA. A third portion of the collected sediment sample was kept at −80 °C until later analysis for microbial community using 16S rRNA sequencing.

2.3. Chemical analysis of samples

2.3.1. Extraction and purification of TBBPA

A precise weight close to 2.5 g freeze-dried sediment or 0.5–1.0 g freeze-dried plant sample was spiked with 50 ng of $^{13}C_{12}$-TBBPA (50 μg mL^{-1} in methanol, 99%, CIL, USA) as surrogate standard and placed at room temperature for 24 h in darkness in order for carrier solvent methanol to completely evaporate before extraction and purification of TBBPA. The sample was extracted with dichloromethane (DCM)/acetone (2/1, v/v, TEDIA, USA) on an accelerated solvent extractor (Dionex ASE 350, Thermo Fisher Scientific, USA). The detailed extraction conditions are listed in the Supporting Information. Each extract was cleaned up by eluting through an ENVI-Carb SPE (Supelco, Sigma-Aldrich, USA) cartridge using method modified based on that by Liu et al. (2016a). The cleaned up eluent was further concentrated to dryness under a gentle stream of high purity nitrogen gas, resolved in 500 μL methanol (Merck, GER), and filtered through a 0.22 μm PTFE syringe filter before analysis on ultra-performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS).

2.3.2. Measurement of sediment enzyme activity

Colorimetric method was used to determine urease and dehydrogenase activity (Lu et al., 2011). Sediment urease activity was determined by the enzymatic reaction of urea in citric acid buffer solution (pH 6.7) at 37 °C and was expressed as mg NH_4^+-N released kg^{-1} dry sediment. Sediment dehydrogenase activity was measured by the reduction of 2, 3, 5-triphenyl tetrazolium chloride (TTC) to 1, 3, 5-triphenyl formazan (TPF) according to Cheema et al. (2009), and it was calculated as 1.00 μg TPF g^{-1} dry sediment.

2.4. Instrumental analysis

UPLC-MS/MS (1290 Infinity UPLC, 6490 Triple Quadrupole Mass Spectrometer, Agilent, USA) coupled with electron spray ionization negative mode was used to quantify TBBPA and its debromination products by select ion mode (Zhang et al., 2015; Lefèvre et al., 2018). Liquid chromatography separation used a ZORBAX Eclipse Plus C18 column (Rapid Resolution High Definition, 2.1 × 100 mm^2, 1.8 micros, Agilent Technologies, location of the company) front a guard column (ZORBAX Eclipse Plus C18, 2.1 × 5 mm 1.8 μm, Agilent, USA). The mobile phase consisted of Milli-Q water (Mobile Phase A) and acetonitrile:methanol (30:70, v/v, Merck, GER) (Mobile Phase B) and its gradient steps are listed in Table S2 of the Supporting Information. The mobile phase flow rate was 0.3 mL min^{-1}. Sample injection volume was 1.0 μL. The mass spectrometry operation parameters are listed in Table S3 of the Supporting Information.

2.5. DNA extraction and PCR amplification

The sediment samples collected at 14, 35, 93 day were extracted for total DNA using the E.Z.N.A.® soil DNA Kit (Omega Bio-tek, Norcross, GA, U.S.) following the protocol provided by the manufacturer. The concentration of the extracted and purified DNA from each sample was determined by a NanoDrop 2000 UV–vis spectrophotometer (Thermo Scientific, Wilmington, USA), and the DNA quality was checked by 1% agarose gel electrophoresis. The V3 − V4 hypervariable regions of the bacteria 16S rRNA gene were amplified using universal primers 338F (5′-ACTCCTACGGGAGGCAGCAG-3′) and 806R (5′-GGACTACHVGGGTWTCTAAT-3′) on a thermocycler PCR system (GeneAmp 9700, ABI, USA). The detailed PCR amplification procedure is provided in the Supporting Information. Purified amplicons from each sample were normalized to equimolar and paired-end sequenced (2 × 300 bp) on an Illumina MiSeq platform (Illumina, San Diego, USA) according to the standard protocol provided by the Majorbio Bio-Pharm Technology Co. Ltd. (Shanghai, China). QIIME was used to analyze the Illimina MiSeq generated data. The taxonomy of each 16S rRNA gene sequence was analyzed by the RDP Classifier algorithm against the Silva (SSU123), 16S rRNA database using confidence threshold of 70%.

2.6. Quality assurance and control

The recovery rate of TBBPA analytical method was tested by the surrogate standard ($^{13}C_{12}$-labeled TBBPA) that was spiked to each sample before the extraction and purification steps. Neither TBBPA nor its metabolites were detectable in the procedural blanks sample. The surrogate standard recoveries of $^{13}C_{12}$-TBBPA from sediment, roots, stems, and leaves ranged from 86 ± 9.7% to 110 ± 6.9%, from 52.1 ± 6.8% to 91.7 ± 15.1%, from 62.0 ± 10.7% to 97.1 ± 13.9%, and from 45.0 ± 2.3% to 120.8 ± 6.5%, respectively. The method detection limits of TBBPA based on signal-to-noise ratio of 3:1 were 0.08, 0.12, 0.33, and 0.25 ng g^{-1} for sediment, roots, stems, and leaves, respectively. The final TBBPA concentration in a sample was corrected relative to the surrogate recovery of the sample.

2.7. Statistical analysis

The OriginPro 2016 (Origin Lab, USA) and SPSS Statistics 20 (IBM SPSS, USA) were used to analysis the data. Degradation of TBBPA was fitted using the first-order kinetics equation $C = C_0 e^{-k t}$, where C_0 and C_t are the concentrations at time 0 and t, respectively, and k is the degradation rate constant (Ghoveisi et al., 2014). The half-life ($T_{1/2}$) was calculated using $T_{1/2} = \ln 2/k$. A one-way analysis of variance (ANOVA) was performed to test statistically significant difference among treatments at $p < 0.05$.

3. Results

3.1. TBBPA degradation in sediments

The concentrations of TBBPA in the sediments of different treatments decreased with time (Fig. 1). However, the reduction of TBBPA in the sterilized sediment was the slowest with time comparing to other treatments. At day 93, 78.42% of the TBBPA still remained in the sediment. The level of TBBPA in the unplanted non-sterilized sediment declined rapidly with time during the first 21 day, from 10.83 to 3.12 mg kg^{-1} (dw), and then slowly decreased to 0.35 mg kg^{-1} (dw) at day 93. Compared to the unplanted non-sterilized sediment, the TBBPA levels in the sediments planted with mangroves were significantly lower at any given time except at day 93 ($p < 0.05$), suggesting mangrove plants further enhanced TBBPA degradation. Comparing the two planted sediments, TBBPA concentrations in the *K. obovata*-planted sediment was significantly lower than that planted with *A. marina*. The degradation of TBBPA followed the first order kinetics with TBBPA half-lives of 21.66 day, 20.38 day and 16.50 day for the unplanted non-sterilized sediment, *A. marina*-planted, and *K. obovata*-planted non-sterilized sediments, respectively (Table S4). After 93-day incubation, the debromination products of TBBPA, namely tribromobisphenol A (TriBBPA), dibromobisphenol A (DiBBPA), monobromobisphenol A (MonoBBPA), and bisphenol A (BPA), were all detected in non-sterilized sediment, but not detected in the sterilized sediment (Fig. S1, Table S3). The result indicated that the debromination of TBBPA constantly took place in contaminated mangrove sediment.

3.2. Uptake and translocation of TBBPA in mangrove plants

In the present study, TBBPA was detectable in all plant tissue of both *A. marina* and *K. obovata* on the 14th, 35th, and 93rd day of the greenhouse experiment (Fig. 2). Within the first 14 day of planting, substantial amount of TBBPA was taken up by the roots of both mangrove species and also translocated to aboveground plant tissues. On the 14th day, the concentrations of TBBPA in roots tissue of *A. marina* and *K. obovata* were similar and the highest compared to those tested at later times (Fig. 2). The TBBPA concentration in the roots continued to reduce with time, however, this reduction with time is more rapid for the *A. marina* compared to *K. obovata*. The concentration of TBBPA in the *K. obovata* roots was 3.52 and 2.18-times higher than that in the *A. marina* roots on the 35th and 93rd day, respectively. The levels of TBBPA in the stems of both plants reduced with time. However, on the 14th and 35th day, 4.83 and 3.25 times, respectively, higher levels of TBBPA were found in the *K. obovata* stems than the *A. marina* stems and on the 93rd day its levels became similar in the stems of both plants at 119.44 and 94.64 ng g^{-1} dw, respectively. This result suggests higher bioaccumulation of TBBPA in the *K. obovata* stems compared to *A. marina*. Although on the 35th day the TBBPA level was slightly higher in the *K. obovata* leaves compared to that of *A. marina* leaves, its levels were fairly consistent with time and between the two plant species.

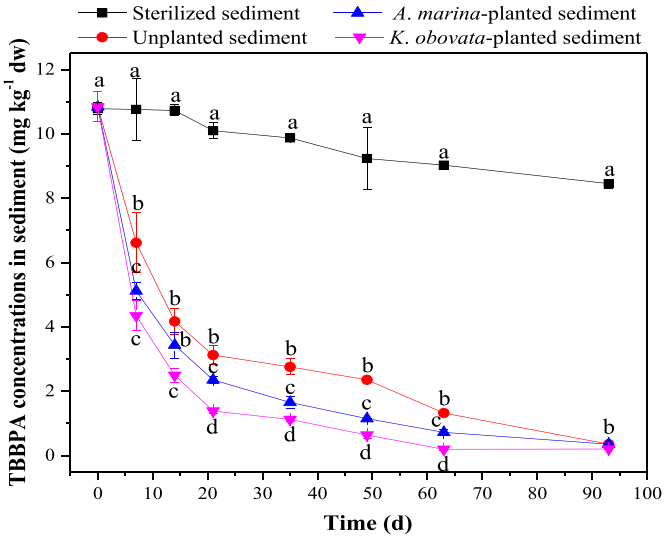

Fig. 1. Concentrations of TBBPA in sterilized sediment and non-sterilized sediment without and with *A. marina* or *K. obovata* at different time. Different letters at the same time point indicate statistically significant difference at $p < 0.05$.

Bioconcentration factor (BCF) and translocation factor (TF) of TBBPA for *A. marina* and *K. obovata* are shown in Table 1. The root and whole plant average BCF values for *A. marina* decreased from 1.13 to 1.18 on the 14th day to 0.49 and 0.56 on the 35th day, and then increased to 0.85 and 1.13 on the 93rd day. On the contrary, the average BCF values for *K. obovata* root and whole plant increased with time, reaching to the highest average values of 3.72 and 4.50 at 93 day for the root and whole plant, respectively. Comparing to *A. marina,* the BCF values for *K. obovata* were significantly higher for both the root and the whole plant during the 93-day incubation. The average root-stem translocation factor ($TF_{r\text{-}s}$) values for *K. obovata* decreased from 0.16 on the 14th day to 0.092 on the 35th day and then increased back up to 0.16 on the 93rd day, while the average $TF_{r\text{-}s}$ values for *A. marina* continued to increase during the entire 93-day experimental period. Comparing to *A. marina,* the $TF_{r\text{-}s}$ value for *K. obovata* was significantly higher on the 14th day, but the opposite was observed on the 93rd day. The average stem-leaf translocation factor ($TF_{s\text{-}l}$) values for both *A. marina* and *K. obovata* increased with time. The average $TF_{s\text{-}l}$ values for *A. marina* and *K. obovata* were similar on the 14th and 35th day, but it was significantly lower for *K. obovata* comparing to that for *A. marina* on the 93rd day.

3.3. Enzyme activities in sediment

Urease activity, as an indicator of soil nutrition properties, is shown in Fig. 3A. Within the first 35 day, the urease activity decreased with increasing time for all treatments. However this reduction was faster for the unplanted sediment than the planted sediments. From 35th to 93rd day, the urease activity went back up with time for the unplanted sediment but for the planted sediments this increase with time was not as significant. The urease activity was similar between the *K. obovata*-planted and the *A. marina*-planted sediments except on the 49th and the 63rd day. However, both planted sediments had significantly higher levels of urease activity than the unplanted sediment during the 93-day growth period. As shown in Fig. 4B, the dehydrogenase activity, an indicator for soil microbial metabolism activity, increased with time in all three types of sediments within the first 49 day and then slowly decreased with time thereafter until the 93rd day. However, this initial increase of dehydrogenase activity was more rapid in the *K. obovata*-planted sediment than the *A. marina*-planted and the unplanted sediments, resulting in overall higher levels of dehydrogenase activity in the *K. obovata*-planted sediment comparing to the other two sediments during the entire 93-day greenhouse plant growth period.

3.4. Microbial community α-diversity and β-diversity

High-throughput sequencing of DNA extracted from a total of 27 sediment samples that were collected on 14th, 35th, and 93rd day

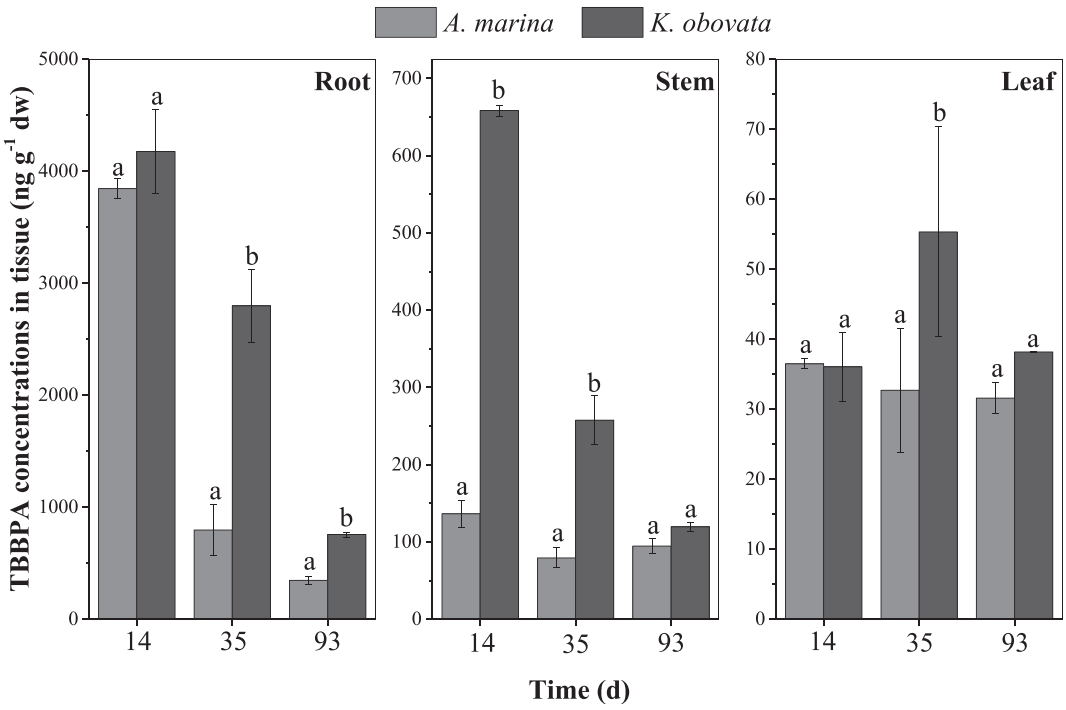

Fig. 2. Concentrations of TBBPA in roots, stems, and leaves of *A. marina* and *K. obovata* on the 14th, 35th, and 93rd day of planting. Different letters at the same time point indicate statistically significant difference at $p < 0.05$.

Table 1
Bioconcentration factor (BCF) and translocation factor (TF) of *A. marina* and *K. obovata* in TBBPA contaminated sediment on the 14th, 35th, and 93rd day.

		BCF		TF	
		Root	Whole plant	Stem ($TF_{r\text{-}s}$)	Leaf ($TF_{s\text{-}l}$)
14 day	*A. marina*	1.13 ± 0.12 a	1.18 ± 0.13 a	0.035 ± 0.002 a	0.01 ± 0.00 a
	K. obovata	1.69 ± 0.31 b	1.97 ± 0.33 b	0.16 ± 0.017 b	0.009 ± 0.00 a
35 day	*A. marina*	0.49 ± 0.20 a	0.56 ± 0.21 a	0.11 ± 0.052 ab	0.047 ± 0.024 ab
	K. obovata	2.69 ± 0.96 bc	2.99 ± 1.05 bc	0.092 ± 0.009 b	0.020 ± 0.008 ab
93 day	*A. marina*	0.83 ± 0.13 a	1.13 ± 0.15 a	0.28 ± 0.04 c	0.092 ± 0.012 c
	K. obovata	3.72 ± 0.29 c	4.50 ± 0.35 c	0.16 ± 0.017 b	0.051 ± 0.004 b

Note: BCF_{root} = TBBPA concentration in plant tissue/TBBPA concentration in sediment; $BCF_{whole\ plant}$ = TBBPA concentration in whole plant/TBBPA concentration in sediment; TF = TBBPA concentration in aboveground plant tissue/TBBPA concentration in root; $TF_{r\text{-}s}$: root-stem translocation factor; $TF_{s\text{-}l}$: stem-leaf translocation factor. Different letters within the same column indicate statistically significant difference ($p < 0.05$).

generated a total of 1147311 high quality sequences with 42493 sequences in each sample. These sequences were clustered into a total of 10487 OTUs with a similarity level of 97%, which contained 61 phylum, 164 class, 318 order, 567 family, 1105 genus and 2419 species (Table S5). Chao1 index-based rarefaction curves increased with increasing sequence depth and plateaued when the number of reads reached to 40,000 and the Good's coverage were all above 95% (Fig. S2, Table S6). This result suggests that the data was representative for the microbial diversity analysis in each sample. On the 14th day, the α-diversity index analysis (Table S6) showed that the Shannon index in the planted sediments was significantly higher than the unplanted ones, and the chao 1 index was higher in the *K. obovata*-planted sediment than that in the unplanted sediment. In the *K. obovata*-planted sediment, the Shannon and chao 1 index was significantly higher than the other two treatments on the 35th day, however, the difference became insignificant among the three treatments on the 93rd day. Gene copies of total bacterial in the planted sediments were significantly higher than that in the unplanted sediment and it was higher in the *K. obovata*-planted sediment than that in the *A. marina*-planted sediment during the incubation (Table S6).

The OTU-based hierarchical cluster and UniFrac-based principal co-ordinate analysis showed that the sediment samples from different treatments were distinctly separated at each sampling time (Fig. S3). In addition, the analysis of similarity (ANOSIM) tested (Table S7) shows that treatment ($R = 0.5067$, $p = 0.001$) was the major factor but the length of plant growth in the sediments ($R = 0.1107$, $p = 0.029$) was also a factor in controlling microbial community evolution in the sediments. The R-values were 0.9424 ($p = 0.002$), 0.6214 ($p = 0.006$), and 0.7119 ($p = 0.019$) in the sediments collected on the 14th, 35th, and 93rd day, respectively, indicating the most significant differences in microbial community composition among three treatments occurred on the 14th day.

3.5. Microbial community composition and evolution

The sequencing results showed that the relative abundance of 16 dominant microorganisms accounted for above 93.34% of the entire bacterial community at the phylum level across all sediment samples (Fig. 4, Fig. S4). The mainly phylum were *Proteobacteria*, *Chloroflexi*, *Firmcutes*, *Actinobacteria*, *Bacteroidetes*, *Acidobateria*, *Nitrospira*, and *Cyanobacteria* in the sediments. Together, they

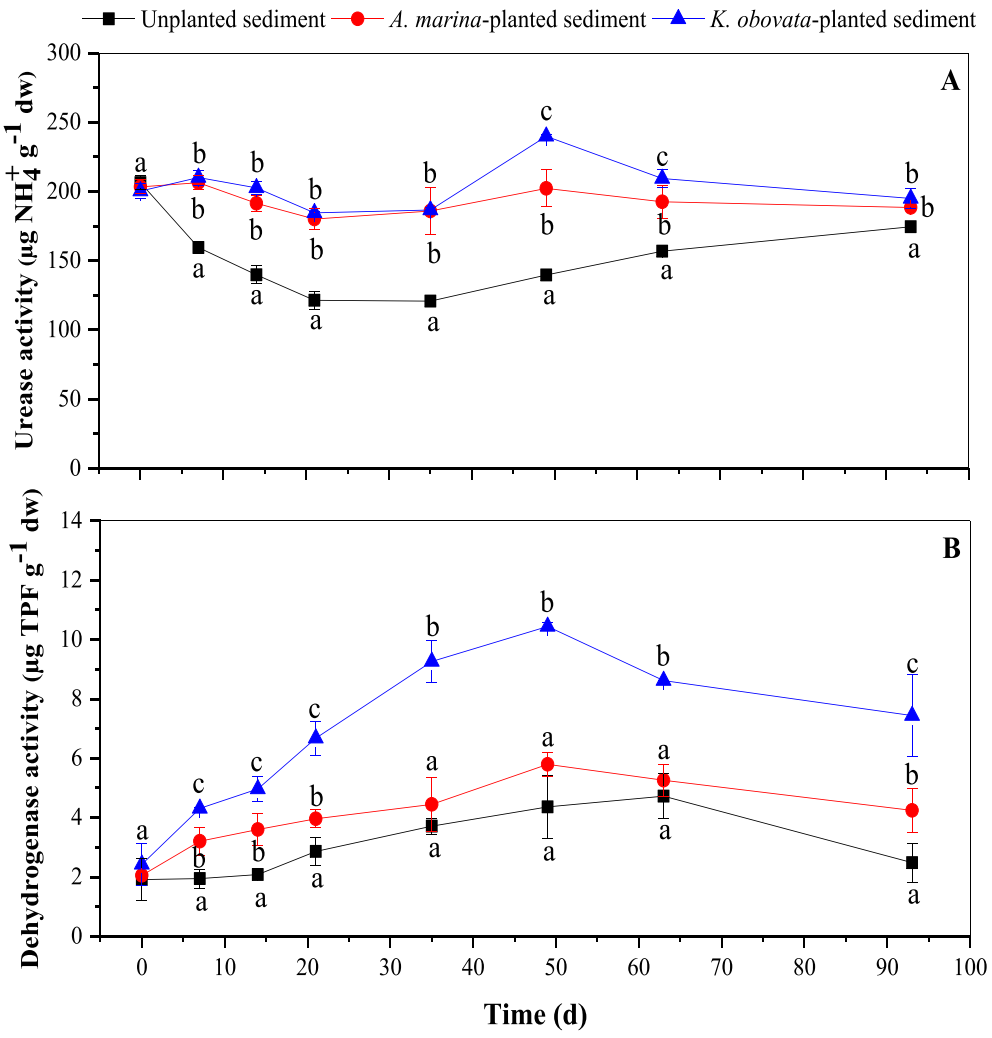

Fig. 3. (A) Urease activity and (B) dehydrogenase activity in non-sterilized sediment without and with *A. marina* or *K. obovata* at different time. Different letters at the same time point indicate statistically significant difference at $p < 0.05$.

accounted for above 80% of total microbial community in a sediment (Fig. S4). On the 14th day, the relative abundance of *Proteobacteria* and *Nitrospira* was significantly higher in the *K. obovata*-planted sediment than the other two treatments, the relative level of *Chloroflexi* in the *A. marina*-planted sediment was higher than the other two treatments, and the relative abundance of *Bacteroidetes* phylum was significantly higher in the unplanted sediment than the two planted sediments (Fig. 4A). However, the above described differences between treatments became insignificant at the two later dates. The relative abundance of *unclassified-k-norank* was the higher in the *K. obovata*-planted sediment than the other two treatments at all 3 sampling times. The relative abundance of *Cyanobacteria* phylum was significantly higher in the unplanted sediment than the other two treatments at the 35 day (Fig. 4B). On the 93rd day, the relative abundance of *unclassified-k-norank* and *Gracilibacteria* phylum were significantly higher in the *K. obovata*-planted sediment than the other two treatments (Fig. 4C). These results indicates that different treatments affect the microbial community structure evolution at phylum level during TBBPA degradation.

The top 30 genus composition (relative abundance > 0.01) and the relative abundance of 4 known genus involved with TBBPA-degrader (relative abundance < 0.01) were analyzed (Figs. 5 and 6). The major genera in three treatments included *Anaerolineae*, *Acidobacteria*, *Sulfurovum*, *Thiobacillus*, *Desulfobulbus*, *Nitrospira*, *unclassified-p-Chloroflexi*, *Aminicenantes*, *Cyanobacteria* (Fig. 5, Fig. S5). During the first 35 day, the relative abundance of 25 identified genera were higher in the planted sediments than the unplanted sediment (Fig. 5 A and B). In the *K. obovata*-planted treatment, the mainly genus *Sulfurovum* and *Nitrospira* and the *unclassified-k-norank*, *Gemmatimonadaceae*, *Gammaproteobacteria* genus abundance were significantly higher than the other two treatments, and the abundance of *Sva0485*, *Gemmatimonadetes* genus were significantly higher than *A. marina*-planted sediment, on the 14th day (Fig. S6). The *Anaerolineae* abundance significantly increased in the *A. marina*-planted sediment at 14 day, and its relative abundance significantly increased from 7.67% at 14 day to 14.35% at 35 day in the *K. obovata*-planted sediment (Figs. S6 and S7). At 35 day, the abundance of *Sulfurovum* and *unclassified-p-Chloroflexi* significantly increased in the planted sediment, especially in the *K. obovata*-planted sediment, however, the *Cyanobacteria* genus exhibited significantly higher abundance in the unplanted sediment than the planted sediments (Figs. S6 and S7). Moreover, planting also increased the abundance of genus *Acidobacteria*, *Thiobacteria*, *Aminicenantes*, *Desulfobulbus*, *Desulfobacca*, *Desulfatiglans*, *Bacillus* during the 35 day incubation. At 93 day, the

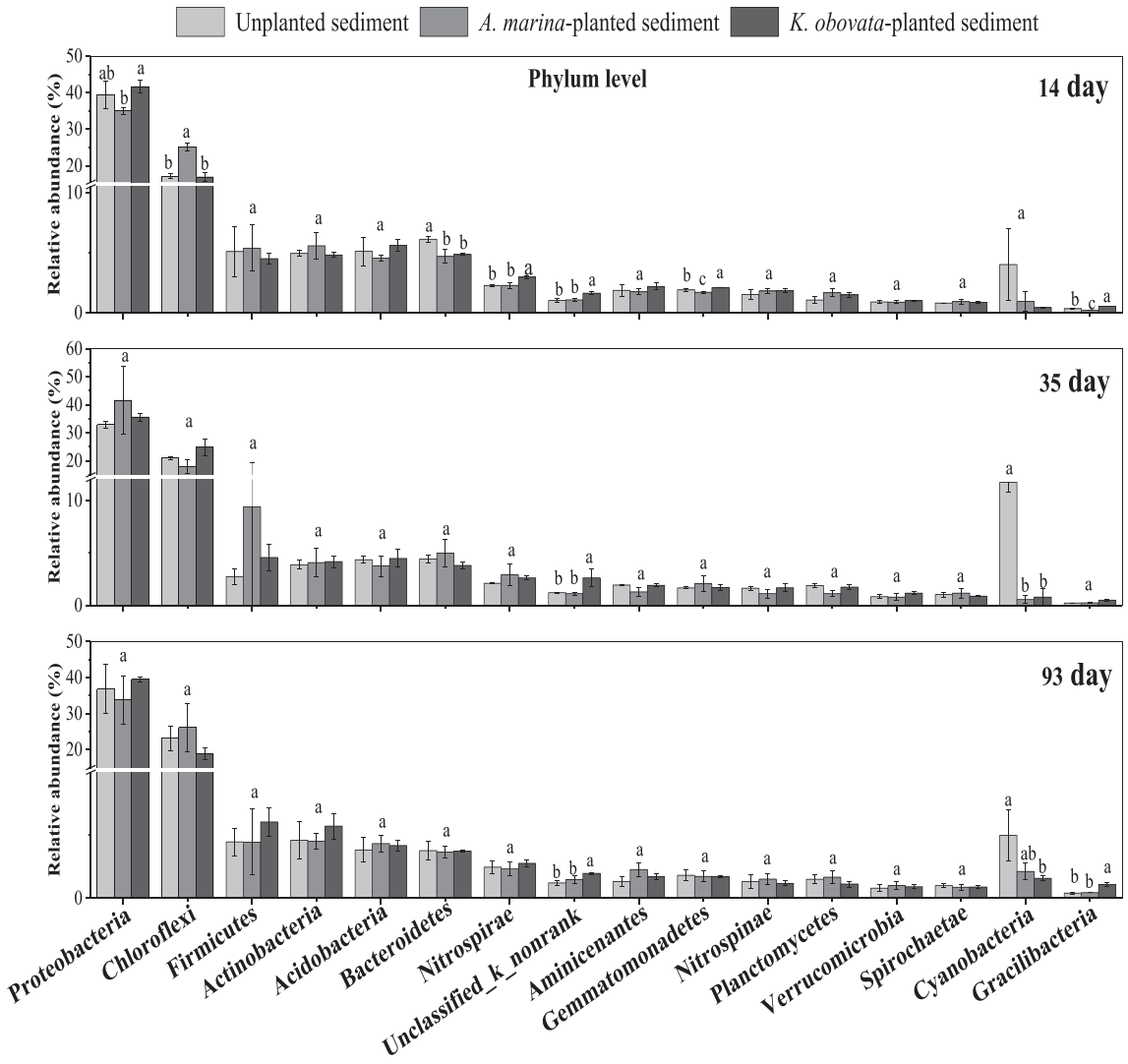

Fig. 4. Relative abundance of dominant microorganisms at phylum level (relative abundance > 0.01) in the non-sterilized sediments without and with *A. marina* or *K. obovata* on the 14th, 35th, and 93rd day. Different letters at the same time point indicate statistically significant difference at $p < 0.05$.

relative abundance of 22 genus was higher in the planted sediments than the unplanted sediment, but the mainly genus abundance was not significantly different among three treatments, while the relative abundance of *SJA-68* and *unclassified-k-norank* were significantly higher in the *K. obovata*-planted sediment than the other treatments (Fig. S8). The 4 known genus associated with TBBPA degradation have been reported by others (Peng et al., 2012; Yang et al., 2016) at relative abundance < 0.01. Our result has shown that *K. obovata* could significantly stimulate the abundance of *Geobacter*, *Flavobacterium* at 14 day (Fig. 6A and B). Even on the 93rd day of incubation, *Geobacter*, *Flavobacterium*, *Pseudomonas*, *Azoarcus* genus still exhibited significantly higher abundances in the *K. obovata*-planted sediments than the other treatments (Fig. 6).

4. Discussion

4.1. Effect of mangrove plants on TBBPA dissipation

The results from this study, for the first time, suggest that the rhizosphere process of both mangrove species is likely to significantly enhance TBBPA removal from contaminated sediments within 93 day of plant growth (Fig. 1). Similarly, Ravit and coworkers have observed positive effect of *Spartina alterniflora* and *Phragmites australis* in the rhizosphere sediment on TBBPA degradation after the 130 day incubation and suggested microbial reductive dehalogenation process for TBBPA (Ravit et al. (2005). Recently, Sun et al. (2014) investigated the effect of rice (*Oryza sativa*) and reed (*Phragmites australis*) on TBBPA removal in a paddy soil and also demonstrated that TBBPA dissipation was slightly accelerated in the rice soil and strongly enhanced in the reed soil after 66 day incubation. In our study, TBBPA in the contaminated sediment could be taken up by roots of *A. marina* and *K. obovata* and them translocated to aboveground tissues, resulting in significantly higher concentration of TBBPA in roots than stems and leaves (Fig. 2). This observation is in consistent with former reports by Li et al. (2011) and Sun et al. (2014). In the mangrove sediment, the microbial reductive dehalogenation has been recognized as an important mechanism for the removal of halogenated organic pollutants (Zhu et al., 2014; Chen et al., 2017). In the present study, the reductive debromination of TBBPA in non-sterilized mangrove sediment also contributed to the microbial degradation.

Based on the BCF and TF of TBBPA in both species, *K. obovata* exhibited stronger TBBPA accumulation ability in root and whole plant, but *A. marina* has stronger capability of translocation TBBPA

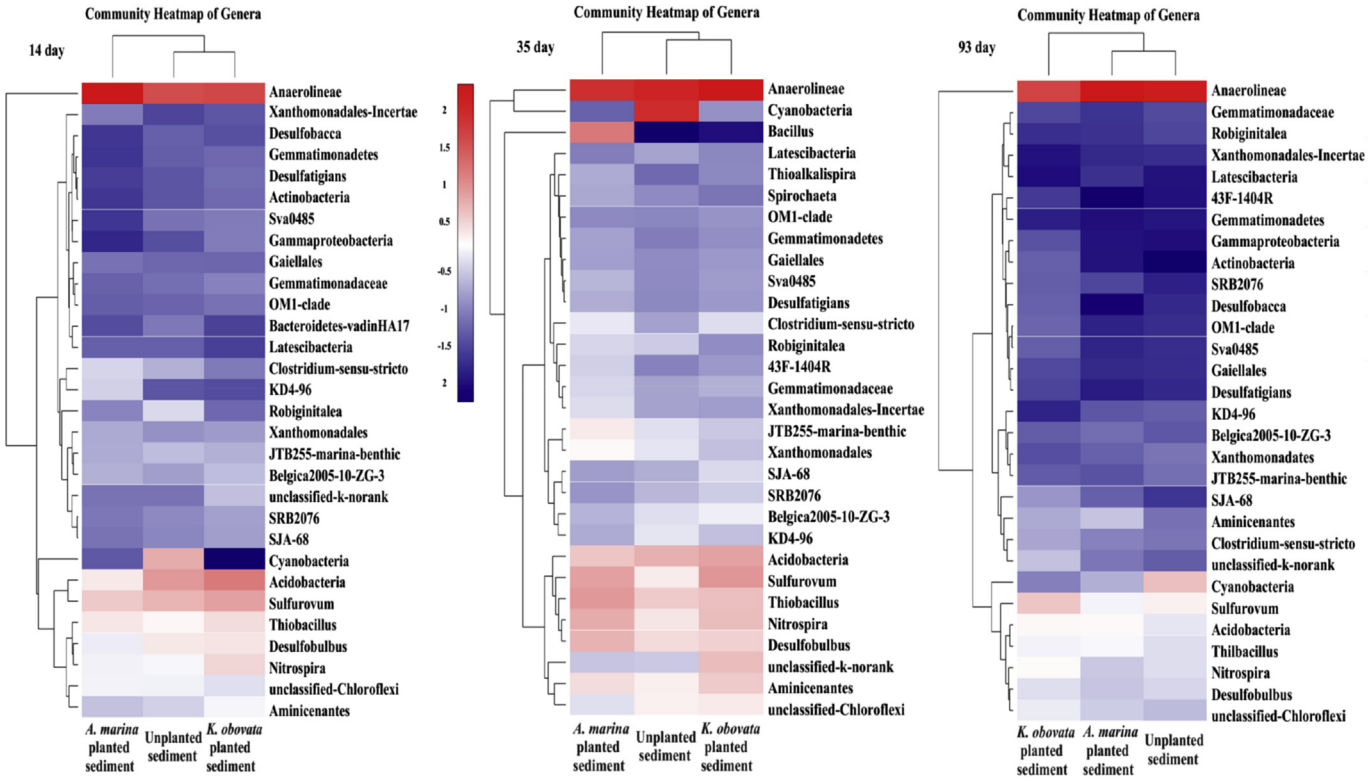

Fig. 5. Hierarchical cluster analysis showing the relative abundances of top 30 genera (relative abundance > 0.01) in the non-sterilized sediments without and with *A. marina* or *K. obovata* on the 14th, 35th, and 93rd day. The color-coded panel indicates the changes of relative abundance. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

to stem and leaves (Table 1). The result was in line with observation by Li et al. (2019), who found that *K. obovata* exhibited the highest RCF of TBBPA compared to *A. marina*, but *A. marina* exhibited higher TF_{s-l} than *K. obovata*. This observation indicated different enrichment capacity of TBBPA in different mangrove plants. In the current study, we also found that the concentration of TBBPA in root and stem declined with time which is similar to what was observed by Hou et al. (2018). In the study by Hou and coworkers, it was found that TBBPA biotransformated to TBBPA-methyl ether derivatives, 2,6-dibromo-4-(2-(2-hydroxyl)-propyl)-phenetole in pumpkin tissue during 15 day growth period. It is clear that the transformation of TBBPA seems to be taken place within mangrove tissue, however, knowledge on TBBPA transformation pathway is still unknown, partly due to the low concentration of TBBPA detected in mangroves (Li et al., 2019).

Mass balance of TBBPA in microcosm showed that the contribution of plants uptake to the removal of TBBPA from sediment was less than 0.26% during the incubation, which might be negligible (Table S8). Similar observations were also made for the removal of other organic pollutants such as PBDEs by mangroves (Chen et al., 2015, 2017). At the end of 93 day mangrove growth period, over 95% TBBPA was removed from three treatment sediments. This removal should be largely due to microbial-driven biotransformation, bioaccumulation, and biodegradation.

4.2. Effect of mangrove rhizosphere process on sediment microbial community and TBBPA removal

Significantly higher urease and dehydrogenase activities were observed in the rhizosphere sediment of *A. marina* and *K. obovata*, especially in *K. obovata* (Fig. 3A and B). This might contributed to greater TBBPA dissipation in the planted sediments than the unplanted sediment as observed in the present study (Fig. 1). Previous reports have demonstrated that both urease and dehydrogenase activities were positively correlated with the removal of organic contaminants in sediment (Lu et al., 2011; Chen et al., 2015). In addition, larger Shannon, Chao 1 index, and total gene copies were observed in the planted sediment, especially in the *K. obovata*-planted sediment (Table S6), indicating higher microbial diversity, activity and abundance in planted sediment. This might also be responsible for the TBBPA removal. Similar reports on enhanced organic contaminates dissipation in rhizosphere due to higher microbial activity, diversity and abundance have been reported (Tu et al., 2011). It has been reported that labile substrates released by plants into rhizosphere could enhance carbon or nitrogen levels and in turn increase microbial activity and abundance. As a result organic compounds degradation in the rhizosphere is stimulated (Martin et al., 2014). However, to further explore the in depth mechanisms of microbial-driven TBBPA degradation in plant rhizosphere, the response of plant-induced TBBPA-degrading relative microbial community must be investigated.

On the 14th day, *K. obovata* enriched the mainly phylum of *Proteobacteria* and *Nitrospira* and *A. marina* enriched *Chloroflexi*, indicating that bacterial enrichment by mangrove plant seems to be species-specific. Similar reports were made previously (Phillips et al., 2012; Chen et al., 2017). The top 30 microbial community of sediments at genus indicated that the *Anaerolineae* might play an important role in the period of TBBPA degradation (Chen et al., 2019). *A. marina* significantly stimulated *Anaerolineae* genus in the sediment at 14 day and *K. obovata* obviously increased the relative abundance of *Anaerolineae* at 35 day (Fig. 5A and B). The *Anaerolineae* might enhanced the degradation of TBBPA at early planting stage. Both *Sulfurovum* and *Nitrospira* were present in the mangrove sediment stimulating TBBPA anaerobic degradation

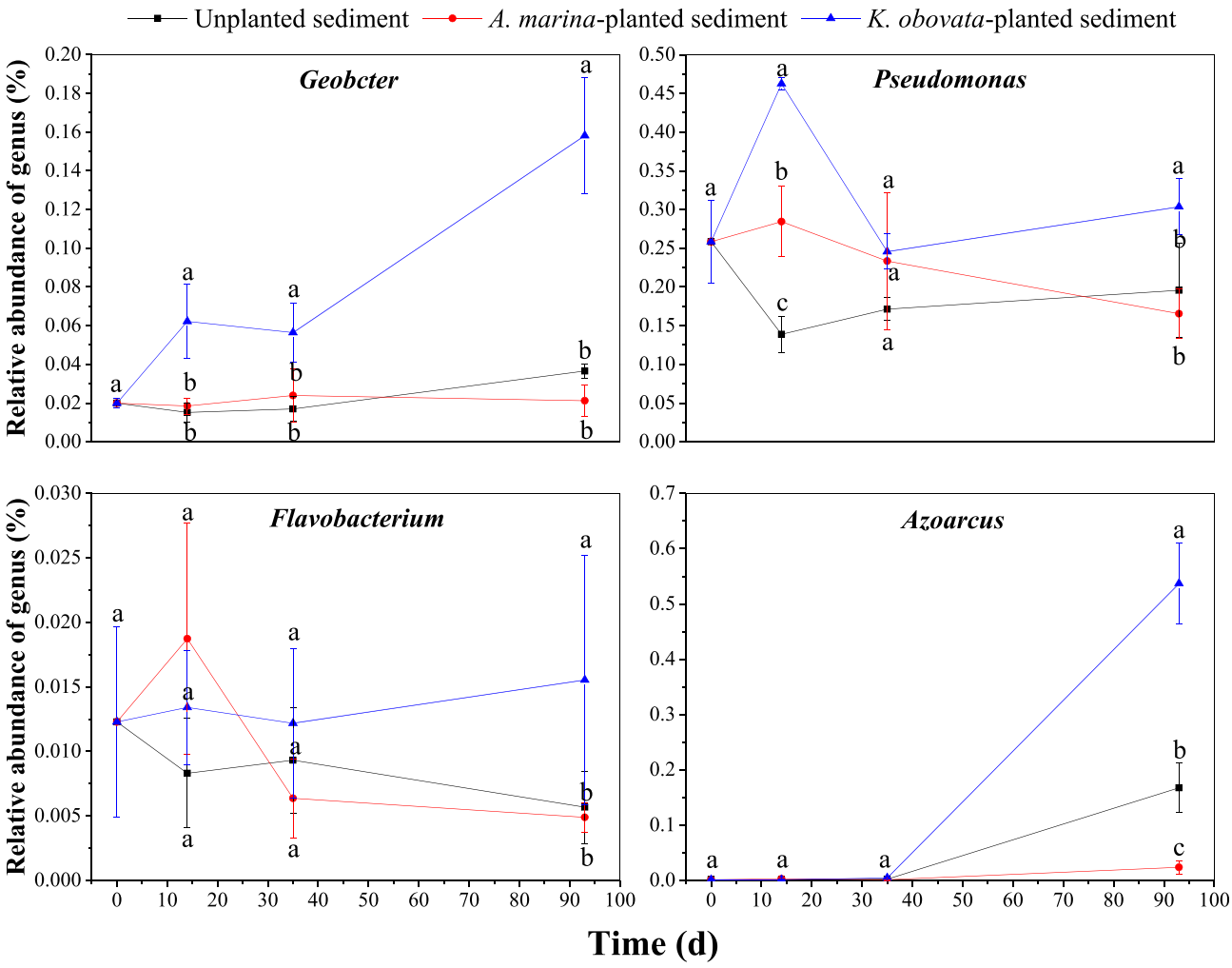

Fig. 6. Relative abundances of the genus (relative abundance < 0.01) associated with TBBPA degradation in non-sterilized sediments without and with *A. marina* or *K. obovata* at different time. Different letters at the same time point indicate statistically significant difference at $p < 0.05$.

(Yang et al., 2019). Li et al. (2015) also found Nitro-debromination of TBBPA by nitrifiers in nitrifying activated sludge. We observed that *K. obovata* significantly stimulated the genus of *Sulfurovum* and *Nitrospira* at 14 day, and *Sulfurovum* at 35 day (Figs. S6 and S7). The genus of *Gammaproteobacteria*, known as TBBPA-degrading bacteria (Peng et al., 2012) was significantly enhanced in the *K. obovata*-planted sediment at 14 day (Fig. S6). Furthermore, a substantial of bacterial communities associated with organic pollutants degradation in mangrove sediment (Zanaroli et al., 2015) including the genus of *unclassified-k-norank*, *Gemmatimonadaceae*, *Sva0485*, *Gemmatimonadetes* were significantly enriched in the *K. obovata*-planted sediment at 14 day (Fig. S6), which are likely to be the potential bacteria for TBBPA degradation in the *K. obovata*-planted sediment. The selective bacterial enrichment by mangrove plants depends on the plant species, the characteristic of exudates, and the concentration and composition of organic pollutants (Bourceret et al., 2015; Guo et al., 2017). *Desulfobulbus*, *Desulfobacca*, *Desulfatigians* are strictly anaerobic bacteria, which are regarded as sulfur-reducing bacteria and the reductive debromination of TBBPA might act as a terminal electron acceptor (Zanaroli et al., 2015). The enrichment of these sulfur-reducing bacterial in planted sediments compared to the unplanted sediment might be due to higher dissolve organic carbon in rhizosphere. It has been reported that dissolve organic carbon is the main factor that influences the growth of sulfur reducing bacterial (Muhammad et al., 2014). In the early stage, plants also stimulated the abundance of *Bacillus*, *Thiobacteria*, *Clostridium*, *Acidobacteria*, *Aminicenantes*, all of which are believed to be associated with the biodegradation of TBBPA (Lefevre et al., 2016; Xiong et al., 2017; Xie et al., 2018). The abundance of *Cyanobacteria* in the unplanted sediment increased significantly at 35 day compared to other two planted sediment. Peng et al. (2014) reported that microalgae could use TBBPA as carbon source for growth and might also be involved in TBBPA biodegradation in mangrove sediment.

Nevertheless, previous research has suggested that the functional microbial genus associated with organic pollutants degradation deserve more attention (Tejeda-Agredano et al., 2013). In the present study, *K. obovata* significantly enhanced the abundance of *Geobater* and *Pseudomonas* abundance at 14 day and the abundance of *Geobater*, *Pseudomonas*, *Flavobacterium*, *Azoarcus* at 93 day (Fig. 6), all of which might accelerate the removal of TBBPA in *K. obovata*-planted sediment. *Geobater* genus is one of orgnohalide-respiring bacteria, which involves in reductive debromination process of halogenated organic compound (Zhang et al., 2013; Zanaroli et al., 2015). *Pseudomonas*, *Flavobacterium*, and *Azoarcus* genus have been considered as TBBPA-degrading bacteria (Peng et al., 2012; Fan et al., 2017). Different impact of *A. marina* and *K. obovata* on enrichment of bacterial genus in the sediments at

early stage (0–35 day) of plant growth, especially at 14 day, might explain the difference in the TBBPA removal from the *A. marina*- and *K. obovata*-planted sediments. In addition, the stimulatory effect of *K. obovata* on *Geobater*, *Pseudomonas*, *Flavobacterium*, *Azoarcus* in the sediments were significantly higher than *A. marina* (Fig. 6), likely leading to the higher TBBPA degradation efficiency in the *K. obovata*-planted sediment.

5. Conclusion

This study demonstrates that both *A. marina* and *K. obovata*, especially *K. obovata,* were able to promote TBBPA degradation in mangrove sediment. It was observed that *K. obovata* exhibited higher ability of TBBPA bioaccumulation and lower capability of TBBPA translocation from roots to above ground tissues compared to *A. marina*. In the rhizosphere of *A. marina* and *K. obovata,* the urease and dehydrogenase activities and total microbial abundance were significantly higher compared to non-rhizosphere sediment, likely resulting in enhanced TBBPA biodegradation in the mangrove-planted sediments. Our mass balance results strongly suggests that the degradation of TBBPA in the mangrove-planted sediment is largely due to enhanced microbial activity and abundance in the rhizosphere, rather than plant uptake. Different mangrove species demonstrated different capacity for enrichment of potential TBBPA-degrading bacterial genus and, therefore, different impact on TBBPA degradation in the sediment. Our study has shown that *K. obovata* would be a more preferred mangrove species than *A. marina* when used for phytoremediation of TBBPA-contaminated mangrove sediment.

Declaration of competing interest

There is no conflict of interest in the submission of this manuscript. The manuscript has been approved by all authors for submission. I would like to declare on behalf of my co-authors that the work described in this manuscript is original research that has not been published previously, and has not been under consideration for publication elsewhere, in whole or in part.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2019.125385.

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