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How wastewater with different nutrient levels influences microbial degradation of 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47) in anaerobic sediments

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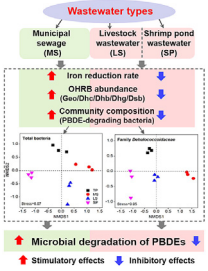
HIGHLIGHTS

- Effect of wastewater on BDE-47 biodegradation was dependent on its nutrient level.
- Wastewater type-specific effect was related to iron reduction and OHRB abundance.
- Bacterial community composition differed significantly among wastewater treatments.

GRAPHICAL ABSTRACT



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ABSTRACT

While wastewater and polybrominated diphenyl ethers (PBDEs) are commonly both discharged into aquatic ecosystems, little information is known about how wastewaters with different nutrient levels impact on microbial degradation of PBDEs. In this study, we used an anaerobic microcosm experiment to examine how the removal rates of 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47) from contaminated sediment varied when exposed to three wastewaters with different nutrient properties, namely livestock wastewater (LS), municipal sewage (MS), and shrimp pond wastewater (SP), and to determine the microbial controls on removal processes. We found that BDE-47 degraded relatively rapidly in MS, which had low carbon and nitrogen concentrations, but degraded much more slowly in LS and SP, which had relatively high nutrient concentrations. The variations in BDE-47 removal in different wastewater were related to iron reduction rates and the abundances of organohalide-respiring bacteria (OHRB). The community compositions of both total bacteria and OHRB from the family *Dehalococcoidaceae* differed significantly among the wastewater treatments. Compared with other treatments, some bacterial groups with PBDE degradation abilities were more abundant in MS where the PBDE-degradation efficiencies were higher. Our results should help support evaluations of the bioremediation potential of sites that are contaminated with both halogenated organic compounds and nutrient-rich wastewater.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are halogenated organic compounds that are widely used as brominated flame

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retardants in various commercial and household products (Wang et al., 2007). Because of their stability and persistence, these compounds are ubiquitous in the environment, where they accumulate in various environmental media, including air, water, soil, sediment, and biota (Yin et al., 2017), and pose risks to the health of animals and human beings (Bayen, 2012; Yang and Chan, 2015). In aquatic ecosystems, sediments with a high capacity to adsorb organic pollutants act as a major sink for PBDEs, and play an important role in their migration, transformation, and degradation (Wang et al., 2018; Zanaroli et al., 2015). Polybrominated diphenyl ethers have been detected at concentrations of up to mg g^{-1} dry weight (dw) in aquatic sediments worldwide (Wang et al., 2007; Yang et al., 2017), and, for example, have been found at particularly high concentrations in rivers or lakes close to e-waste recycling sites, such as the Maozhou River, China (Sun et al., 2013), and Shihwa Lake, Korea (Moon et al., 2012).

Polybrominated diphenyl ethers can be removed from contaminated environments by microbial degradation (Jugder et al., 2016; Song et al., 2015). Apart from the uppermost few centimeters, aquatic sediments are often anaerobic (Zanaroli et al., 2015). Under anaerobic conditions, various halogenated organic pollutants, e.g., polychlorinated biphenyls (PCBs) (Yu et al., 2016), tetrabromobisphenol A (Lefèvre et al., 2018), and pentachlorophenol (PCP) (Tong et al., 2014), have been attenuated naturally by microbial reductive dehalogenation, during which highly halogenated pollutants are transformed to less halogenated compounds that biodegrade readily. Also reported previously, PBDEs in sediments may be degraded and removed by anaerobic reductive dehalogenation (Pan et al., 2018; Yang et al., 2017). Organohalide-respiring bacteria (OHRB), which use halogenated organic pollutants as terminal electron acceptors and substitute chlorine/bromine atoms with hydrogen through the dehalorespiration process, are the most important functional group in reductive dehalogenation (Jugder et al., 2016; Zanaroli et al., 2015). These species generally belong to the genera *Dehalobacter*, *Dehalococcoides*, *Dehalogenimonas*, *Desulfitobacterium*, *Geobacter*, and *Sulfurospirillum*, which participate in reductive dehalogenation and also generate energy for their growth and reproduction (Ding and He, 2012; Maphosa et al., 2010). The genera *Dehalobacter*, *Dehalococcoides* and *Dehalogenimonas* strictly depend on organohalide respiration for growth, using hydrogen as the sole electron donor, thus their isolates are very difficult to culture (Maphosa et al., 2010). In contrast, the genera *Desulfitobacterium*, *Geobacter*, and *Sulfurospirillum* isolates are more easily to be identified due to the fact that they can metabolite a wide range of halogenated organic compounds (Maphosa et al., 2010). Some OHRB, especially from the family *Dehalococcoidaceae* and genus *Geobacter*, were reported to take part in reductive debromination of PBDEs that were anaerobically enriched (He et al., 2006), in biological reactors (Yang et al., 2013), or in sediment slurries (Chen et al., 2018a). In particular, hepta- and octa-BDE were detected in *Sulfurospirillum multivorans* culture exposed to BDE-209, and stepwise reductive debromination of octa-BDE to a variety of hepta-through di-BDEs were found in *Dehalococcoides*-containing culture (He et al., 2006). The genera *Dehalobacter*, *Dehalococcoides*, *Dehalogenimonas*, *Desulfitobacterium*, and *Geobacter*, have also contributed to *in situ* bioremediation of PBDE-contaminated sediments (Chen et al., 2018b; Pan et al., 2017).

While aquatic ecosystems are vulnerable to PBDE contamination, they are also threatened by discharges of various types of wastewater that are rich in carbon (C), nitrogen (N), and phosphorus (P), such as effluents from aquaculture facilities, livestock wastewater, and municipal sewage (Wang et al., 2016). There is wide variation in nutrient concentrations of different wastewaters (Chen et al., 2011; Tam et al., 2009). Previous studies have reported that nutrient inputs from wastewater can change the activity, abundance, and community composition of microorganisms in sediments (Gao et al., 2013; Saarenheimo et al., 2017). It is therefore reasonable to speculate that wastewater might affect the potential for microbial reductive dehalogenation of halogenated pollutants. Liu et al. (2010) showed that, when used as an electron acceptor, nitrate (NO_3^-) inhibited the reductive dechlorination of hexachlorobenzene (HCB) in paddy soils, but that, at appropriate concentrations, ammonium (NH_4^+) triggered HCB dechlorination by anaerobic microbes. Chen et al. (2015) also reported that NH_4^+-rich wastewater stimulated PBDE reductive debromination in the short-term by increasing the abundances of the genera *Dehalobacter* and *Dehalococcoides* in mangrove sediments. Dissolved organic carbon (DOC), amended with biochar to high concentrations, was reported to have positive effects on the growth and enrichment of OHRB and also enhanced PBDE degradation in anaerobic sediment slurries (Chen et al., 2018a). While these studies have provided useful information about how individual nutrients influence reductive dehalogenation, we still have a limited understanding of how wastewater that contain complex mixtures of nutrients influence the process.

As the third largest freshwater lake in China, Taihu Lake is one of the most polluted in the country, because of the rapid economic and social development over recent decades in its basin (Xu et al., 2016). Various types of nutrient-rich wastewater from feedlots, aquaculture facilities, and residential areas around the lake are either not, or are inadequately, treated, before being discharged into the lake, and have caused serious eutrophication and enrichment of sediment (Stone, 2011). Various organic pollutants like PCBs and organochlorine pesticides have been detected in Taihu Lake sediments (Wu et al., 2014; Yin et al., 2017). In particular, PBDEs contamination level varied from different areas of Taihu Lake (Wang et al., 2015a; Zhou et al., 2012), and the concentrations were highest in Zhushan Bay at 785 ng g^{-1} dw (Chen et al., 2018b). While nutrient-related variables such as organic carbon have been recently reported to drive the spatial distribution of OHRB in Taihu Lake sediments (Chen et al., 2018b), we have little information about the relationships between PBDE degradation and the inputs of nutrient-rich wastewater. Therefore, this study aims to compare the degradation of BDE-47 by indigenous microorganisms in the Taihu Lake sediments exposed to three types of artificial wastewater, namely livestock wastewater (LS), municipal sewage (MS), and shrimp pond wastewater (SP). We also attempts to explore possible reasons for the variations in BDE-47 degradation induced by different wastewater treatments. We spiked the sediments with BDE-47, because it is one of the most toxic and abundant PBDE congeners in aquatic sediments (Yang and Chan, 2015; Zhu et al., 2014).

2. Materials & methods

2.1. Preparation of PBDE-contaminated sediment

We collected surface sediment to a depth of 10 cm from the southern coastal area (30.9844°N, 120.1411°E) of Taihu Lake, where PBDE contamination was known to be low (Chen et al., 2018b; Wang et al., 2015a). The fresh sediment was passed through an 8-mm sieve to remove any large stones and plant debris. The samples were split into subsamples to facilitate various analyses. We determined the background physico-chemical properties and PBDE background concentrations of the sediment on one subsample as outlined below, and the results are shown in Table S1. To sterilize the sediment, another subsample of fresh sediment was autoclaved