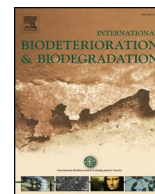




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Biodegradation of polybrominated diphenyl ethers and strategies for acceleration: A review

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs), a class of brominated flame retardants (BFRs), are widely used in various commercial products. PBDEs have attracted increasing attention due to their toxicity and bioaccumulation potential. Biodegradation associated technologies are cost-effective and environmentally friendly for possible removal of PBDEs from the environments. In this review, both aerobic and anaerobic biodegradation of PBDEs and the methods for accelerated degradation are discussed. Generally, the degradation of higher PBDEs (higher degree of bromination) is slower than that of lower PBDEs (lower degree of bromination) under both anaerobic and aerobic conditions. The aerobic degradation pathways of PBDEs include cleavage of aromatic ring, debromination, and hydroxylation, while reductive debromination dominates the initial pathway of anaerobic degradation. A number of methods to overcome the chemical inactivity and low bioavailability of PBDEs for degradation enhancement are discussed, such as the addition of external carbon sources, surfactants and vitamin B₁₂. In addition, coupling of chemical degradation and biodegradation is also reviewed.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants, which have been widely used in the production of commercial and household products, such as foams, textiles, and plastics, for over four decades (Stiborova et al., 2015). There are 209 congeners for PBDEs based on different bromine substitutions. The names and molecular structures of PBDEs appearing in this review are shown in Table 1. Three major commercial PBDEs are reported, including deca-BDEs, octa-BDEs, and penta-BDEs (De Wit, 2002). The higher toxicity and bioaccumulation has resulted in the prohibition of production and usage of penta-BDEs and octa-BDEs in Europe and USA in 2003 and 2006, respectively (Stiborova et al., 2015). However, deca-BDEs were excluded in the bans and the contribution of deca-BDEs usually occupied over 75% of the whole PBDEs (Article, 2014). Recent studies reported that deca-PBDEs could be reductively debrominated into lower PBDEs, which are one of the sources of lower PBDEs in the environment detected (Orihel et al., 2016).

Due to the increasing accumulation of PBDEs in the environment, PBDEs contamination has been found in air, water, soil, sediments, and

even biota (Wang et al., 2011; Zeng et al., 2013b). For example, high concentration of BDE-209 was detected in male birds in waste management facilities (Gentes et al., 2015), and PBDEs was even detected in the arctic biosphere (Rotander et al., 2012). In China, Guiyu Town, Guangdong Province is one of the largest e-waste recycling centers and PBDEs is detected in all of the environmental samples and even in human bodies (Jiang et al., 2014). PBDEs have attracted great attention due to its biotoxicity, such as endocrine disruption effect to mammalian tissues (Song et al., 2015). PBDEs toxicity is different from that of dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs) and pesticides. For example, the neurotoxicity and dioxin-like endocrine disruption induced by PBDEs were observed in mice (Jacobson et al., 2016; Tang et al., 2008; Zeng et al., 2013a).

Although the bioaccumulation of PBDEs in the environment is widely studied, the technology for bioremediation of PBDEs contaminated sites has not been sufficiently investigated. Biodegradation is considered as an economical and safe way for PBDEs removal (Chen et al., 2016a, 2017b; Ming et al., 2017). However, owing to high hydrophobicity and low bioavailability of the compounds the efficiency of PBDE biodegradation is relatively low, thus some methods are proposed

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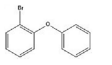
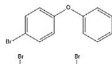
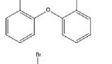
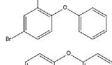



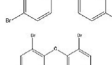
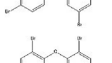
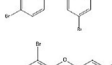

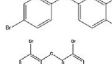
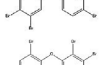
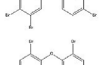
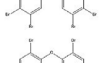
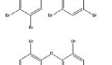
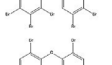
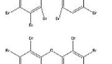
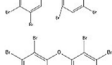
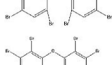

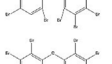

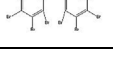
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Table 1
Names and molecular structures of the numbered PBDEs in the article.

Classification	Abbreviation	Names	Molecular Structure
Mono-PBDEs	BDE-1	2-monoPBDEs	
	BDE-3	4-monoPBDEs	
Di-PBDEs	BDE-4	2,2'-diPBDEs	
	BDE-7	2,4-diPBDEs	
	BDE-15	4,4'-diPBDEs	
Tri-PBDEs	BDE-17	2,2',4-triPBDEs	
	BDE-28	2,4,4'-triPBDEs	
Tetra-PBDEs	BDE-47	2,2',4,4'-tetraPBDEs	
	BDE-49	2,2',4,5'-tetraPBDEs	
	BDE-66	2,3',4,4'-tetraPBDEs	
Penta-PBDE	BDE-85	2,2',4,4',5-pentaPBDEs	
	BDE-99	2,2',4,4',5-pentaPBDEs	
	BDE-100	2,2',4,4',6-pentaPBDEs	
Hexa-PBDEs	BDE-138	2,2',3,4,4',5'-hexaPBDEs	
	BDE-153	2,2',4,4',5,5'-hexaPBDEs	
	BDE-154	2,2',4,4',5,6'-hexaPBDEs	
Hepta-PBDE	BDE-183	2,2',3,4,4',5',6'-heptaPBDEs	
	BDE-184	2,2',3,4,4',6,6'-heptaPBDEs	
Octa-PBDEs	BDE-196	2,2',3,3',4,4',5,6'-octaPBDEs	
	BDE-197	2,2',3,3',4,4',6,6'-octaPBDEs	
Nona-PBDEs	BDE-206	2,2',3,3',4,4',5,5',6'-nocaPBDEs	
	BDE-207	2,2',3,3',4,4',5,6,6'-nocaPBDEs	
	BDE-208	2,2',3,3',4,5,5',6,6'-nocaPBDEs	
Deca-PBDEs	BDE-209	2,2',3,3',4,4',5,5',6,6'-decaPBDEs	

to enhance the degradation, such as the use of surfactants. In addition, information regarding the detailed degradation mechanisms and limitations in complex systems is still scarce. In this paper the key factors affecting PBDEs biodegradation, such as microbes, kinetics and pathways, are discussed. Furthermore, the methods for enhancing PBDE biodegradation are summarized.

2. Anaerobic biodegradation

2.1. Microbial community

PBDEs affect the structure of microbial community depending on the degree of bromination of PBDEs (Ma et al., 2016). Bacterial community in the river sediments was altered immediately and irreversibly throughout the incubation period owing to amendment with BDE-153 and BDE-154, which were transformed less than 20% after 70 days. Similar results were observed and demonstrated that BDE-154 was one of the most influential factors on the bacterial community compared to lower PBDEs in mangrove microcosms (Wang et al., 2014). However, the bacterial community did not change significantly upon amendment of BDE-47, which was completely transformed after 63 days in the Nan-Kan River sediment. These results show that higher PBDEs have more pronounced impact on the microbial community structure than lower PBDEs.

The concentrations of PBDEs also have influence on microbial community. Low concentration (1 mg/L and 10 mg/L) of BDE-15 and BDE-209 had only small and transitory effect on the bacterial community, which was changed intensely by BDE-15 and BDE-209 at high concentration (100 mg/L) in soils (Liu et al., 2011). The result was consistent with the study of Huang et al. (2014) who also found that the bacterial community changed significantly upon exposure to a higher concentration of BDE-209. The microbial community structure in sediments from Lianjing River was highly related to the concentrations of deca-BDEs and octa-BDEs (Qiu et al., 2012). This is probably due to the increase of the toxicity of PBDEs with increasing concentrations.

2.2. Pathways of debromination

The reductive debromination, which includes *ortho*-, *meta*- and *para*-debromination, dominates the pathways of PBDEs removal under anaerobic conditions. Because BDE-209 is the key source of lower PBDEs in environment and the bioaccumulation of BDE-47 is detected frequently in biota, the debromination pathways of BDE-209 and BDE-47 are used as examples for further discussion below.

It is reported that BDE-209 can be debrominated into three types of nona-BDE and three types of octa-BDEs in sewage sludge amended with inducers (Gerecke et al., 2005), or into three nona-BDEs, five octa-BDEs, one hepta-BDE and one hexa-BDE in enrichment cultures amended with zero-valence iron (ZVI) (Chen et al., 2014). Formation of three nona-BDEs was also observed for debromination of BDE-209 in sediment and biomimetic system amended with vitamin B₁₂ (Tokarz Iii et al., 2008). nona-BDEs can be further debrominated into lower PBDEs (tri-BDEs) in sediment with addition of electron donors (Qiu et al., 2012) and in anaerobic microcosms from organic compost (Chang et al., 2016). tri-BDEs resulted from the multi-step debromination of BDE-209 were further debrominated into two di-BDEs and one mono-BDE in PBDEs-adapted sediment (Huang et al., 2014). The primary pathway of BDE-209 debromination in soils under anaerobic conditions is shown in Fig. 1.

The first step of anaerobic debromination of BDE-47 is conversion into tri-BDEs (BDE-17 or 28) under anaerobic conditions. *ortho*-debromination of BDE-47 into BDE-28 took place first, and then BDE-28 was debrominated into BDE-15 (*ortho*-debromination) in a culture consisting of *Dehalococcoides* and *Desulfovibrio* spp. (Lee et al., 2011). Ding et al. (2013) reported that BDE-47 was only debrominated into BDE-17 (*para*-debromination) and BDE-17 was debrominated into BDE-4 (*para*-debromination) in a sediment-free enrichment culture. In the mangrove sediment, more than 90% of BDE-47, however, were debrominated into both BDE-28 and BDE-17 after 7 months of incubation (Zhu et al., 2014a). Metabolism rather than complete debromination of BDE-47 was also observed in sediment microcosms. BDE-47 was decreased over 30% in several of the microcosms without a significant accumulation of by-products except for a slightly increase of BDE-17, indicating that

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