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Aerobic and anaerobic biodegradation of BDE-47 by bacteria isolated from an e-waste-contaminated site and the effect of various additives

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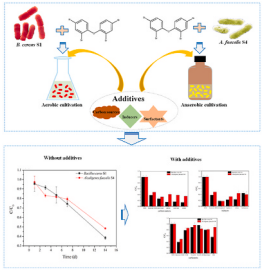
HIGHLIGHTS

- Aerobic and anaerobic BDE-47 degradation strains were isolated from real e-waste sites.
- The isolated *B. cereus* and *A. faecalis* degraded BDE-47 as the sole carbon source.
- Aerobic condition was more favorable than anaerobic for BDE-47 degradation.
- Carbon sources and inducers promoted BDE-47 degradation and surfactants inhibited.

GRAPHICAL ABSTRACT



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ABSTRACT

Degradation experiments are conducted to specifically compare the degradation of 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47) by aerobic and anaerobic strains isolated from real e-waste sites contaminated by BDE-47. The effect of carbon sources, inducers and surfactants on the degradation was examined to strengthen such a comparison. An aerobic strain, *B. cereus* S1, and an anaerobic strain, *A. faecalis* S4, were obtained. The results indicated that BDE-47 could be used as the sole carbon source by *B. cereus* S1 and *A. faecalis* S4 under aerobic and anaerobic conditions, respectively. The degradation of BDE-47 by *B. cereus* S1 and *A. faecalis* S4 was illustrated a first-order kinetics process obtaining a removal efficiency of 61.6% and 51.6% with a first-order rate constant of 0.0728 d^{-1} and 0.0514 d^{-1}, and corresponding half-life of 8.7 d and 13.5 d, respectively. The addition of carbon sources (yeast extract, glucose, acetic acid and ethanol) and inducers (2,4-dichlorophenol, bisphenol A and toluene) promoted BDE-47 degradation by both *B. cereus* S1 and *A. faecalis* S4 under aerobic and anaerobic conditions, while hydroquinone as the inducer inhibited the degradation. All of the surfactants tested (CTAB, Tween 80, Triton X-100, rhamnolipid and SDS) showed inhibitory effect. BDE-47 degradation by *B. cereus* S1 under aerobic condition was more efficient than *A. faecalis* S4 under anaerobic condition whether with or without the additives. The results of the study indicated that in the field sites contaminated by BDE-47, the aerobic condition can be more favorable for BDE-47 removal and the degradation can be further enhanced by applying suitable carbon sources and inducers.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) are known as brominated flame retardants and have strong hydrophobicity and low volatility (Liu et al., 2011; Zhao et al., 2018; Yao et al., 2021). To date, they have been widely used in various commercial and household products, including foam, textiles, circuit boards and plastics, etc. There are 209 congeners for PBDEs according to the number and position of bromine atoms (Zhao et al., 2018). Among these congeners of PBDEs, a lower-brominated PBDE, 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47), has received the most attention for its widespread use and in the meantime the high persistency (Ding et al., 2013). It can interfere with normal secretion of human thyroid hormones, produce neurotoxicity to human body, affect the fertility of human and the normal development of infants, and even increase the possibility of carcinogenesis and teratogenesis. E-waste as the major source of BDE-47, the long-term open-air stacking, illegal processing, incineration and other means of improper disposal of the e-waste are the ways for BDE-47 to enter water and soil. As the most important receptor, soil plays a key role in the migration, transformation and circulation of BDE-47 (Oros et al., 2005; Ni et al., 2012). In the study of Zhang et al. (2014), the results showed that the total concentration of PBDEs in residential areas of Guiyu, Shantou, China, was much higher than 2 mg/kg. Jiang et al. (2019) indicated that in the area near electronic-waste dismantling sites, the total amount of PBDEs was estimated in the range of 0.1–10 mg/kg.

The technologies for removal of BDE-47 from water and soil include photolysis (Pan et al., 2016; Deng et al., 2018), biodegradation (Nie et al., 2020), phytoremediation (Li et al., 2018; Farzana et al., 2019), physical and chemical degradation (Shih and Tai, 2010; Zhuang et al., 2010; Yang et al., 2016; Zhang et al., 2017; Xiang et al., 2020), and the combined use of these technologies (Yao et al., 2021). While compared to biodegradation, the other technologies were limited by secondary pollution, high cost, low remediation efficiency, and inflexibility for practical applications. Alternatively, the biodegradation, as an efficient, environmentally friendly and low-cost technology, plays a fundamental role for BDE-47 removal (Yu et al., 2020; Tang et al., 2021). Hence, isolation, screening, and mediation of strains for degradation of BDE-47 have attracted extensive attention. In recent decades, many studies have reported the degradation of BDE-47 by microbes under individually anaerobic or aerobic condition (Lee et al., 2011; Ding et al., 2013; Lv et al., 2016; Wang et al., 2016). BDE-47 is usually monitored under anaerobic condition due to their high hydrophobicity and adsorbability, and thereby easily combining with organic compounds in soil, where the anaerobic bacteria play a dominant role (Zhu et al., 2014). The anaerobic debromination of BDE-47 is an incomplete process. BDE-47 is generally first to be transformed into tri-BDEs (i.e., BDE-17 or 28) by natural subsurface strains under anaerobic condition (Lee et al., 2011; Ding et al., 2013). The different microbial community structure of natural subsurface strains, however, will lead to different rates and pathways to debrominate parent BDE-47 (Wang et al., 2021). Comparing to anaerobic debromination, some other researches are focused on the aerobic degradation of BDE-47 (Robrock et al., 2009). Aerobic degradation of BDE-47 does take place and sometimes is different from anaerobic debromination with some unique key features, e.g., cleavage of aromatic ring and hydroxylation (Gu et al., 2021; Guo et al., 2021). Effective comparison between anaerobic and aerobic degradation of BDE-47, which is implemented under comparable culture conditions, is therefore important for better understanding on the processes of BDE-47 degradation and the development of microbial resources for BDE-47 bioremediation.

The objective of this study is to systematically investigate and compare (i) the degradation of the target pollutant BDE-47 under aerobic and anaerobic conditions by two indigenous bacteria isolated from an e-waste-contaminated soil at Guiyu town, Guangdong province, China; (ii) and the effect of a variety of additives, including carbon sources, inducers and surfactants, to mediate the degradation under both aerobic and anaerobic conditions. The systematical insights into the difference between the anaerobic and aerobic degradation of BDE-47 would not only provide a solid theoretical proof for BDE-47 bioremediation but help evaluate the biodegradation fate and risk of BDE-47 in environment.

2. Materials and methods

2.1. Chemicals and media

BDE-47 (molecular structure shown in Fig. 1) was purchased from Sinopharm Chemical Reagent Co., Ltd., China (purity above 99%). Carbon sources including yeast extract (purity>99%), glucose (purity>99%), acetic acid (purity>99%), and ethanol (purity>99%) were from Hengxing Chemical Reagent Co., Ltd (Tianjing, China). The inducers including 2,4-dichlorophenol (99% purity), bisphenol A (99% purity), toluene (99% purity), hydroquinone (99% purity) were from Sinopharm Chemical Reagent Co., Ltd (Beijing, China). Cetyltrimethylammonium ammonium bromide (CTAB), rhamnolipid, sodium dodecyl sulfate (SDS), Triton X-100 and Tween 80 used as surfactants were from Merck Life Science Co., Ltd (Shanghai, China). All of the other chemicals used in this study were at least of analytical reagent grade. The agar medium for bacteria isolation consisted of (in g/L) beef extract 5, peptone 10, NaCl 5, and agar 20%. The mineral salt medium (MSM) used in all experiments consisted of (in g/L) $Na_2HPO_4{\cdot}12H_2O$ 5, KH_2PO_4 2.5, NH_4Cl 5, $MgSO_4{\cdot}7H_2O$ 0.5. The medium for enrichment was obtained by adding 0.5 g/L yeast extract into the MSM medium. The pH of the medium was adjusted to 7.0 using 10% NaOH or 10% HCl.

2.2. Screening of BDE-47 degradation strains

Soil samples that may contain aerobic and anaerobic BDE-47 biodegradation microbes were collected from the heavily polluted area by PBDEs in Guiyu Town, Guangdong Province, China (E116°20′, N23°19′). The aerobic BDE-47 biodegradation microbes were collected from the topsoil of an industrial park, shoreside of Lianjiang river, and an e-waste dump site. The anaerobic strains were isolated from subsoil (approximately 1 m below the surface) of the industrial park, a farmland site and the e-waste dump site, which were covered with water, sludge and fine soil, respectively. For strains extraction, 5 g of each of the abovementioned topsoil samples was mixed with 95 mL ultra-pure water in a 250-mL flask, while the subsoil samples were mixed with ultra-pure water in 100-mL serum bottles. After the mixing, large particles in the six suspensions were allowed to precipitate by slow centrifuge to obtain the supernatant containing the microbes. For enrichment, 5 mL of the supernatant from each of the centrifuged soil suspensions was inoculated into a conical flask or a serum bottle, both of which containing 45 mL of newly autoclaved enrichment medium. Then the flasks and serum bottles were incubated at 150 r/min, 30 °C for 24 h in dark. For screening of the BDE-47 degradation strains, certain amount of BDE-47 dissolved in n-hexane was added into flasks and serum bottles, both of which containing 45 mL hot MSM. After complete volatilization of n-hexane and cool-down of the media, 5 mL of the above-mentioned enrichment culture were added to these flasks and serum bottles and then incubated in dark under 150 r/min, 30 °C for 7 d. This process was repeated for 3 times with a gradient concentration of BDE-47 (5, 10, and 20 mg/L) to obtain the strains with high BDE-47 degradation potential.

For isolation of BDE-47 degradation bacteria, the plate streaking method was used as described by Chen et al. (2009). After isolation, three aerobic microbes, i.e., S1, S2 and S3, were obtained from the topsoil of the industrial park, the shoreside of Lianjiang river, and the e-waste dump site, respectively. Three anaerobic strains, i.e., S4, S5 and S6, were from the subsoil of the industrial park, the farmland and the e-waste dump site, respectively. For screening the aerobic strain with the highest BDE-47 degradability from S1 to S3 and the anaerobic strain from S4 to S6, 3 mL of the suspension of aerobic strains were inoculated

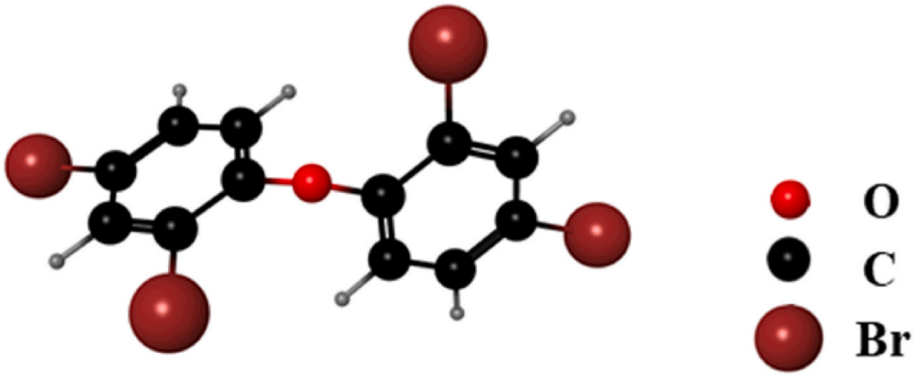

Fig. 1. The molecular structure of BDE-47.

into flasks and that of anaerobic strains into serum bottles, respectively, both of which containing 27 mL MSM with an appropriate amount of BDE-47 (0.115 mg/L) and yeast extract (50 mg/L). The incubation was implemented for 14 d in dark under aerobic or anaerobic conditions. At the end of cultivation, the flasks or serum bottles were sacrificed for BDE-47 analysis. The medium from each flask or serum bottle was poured into a separatory funnel. Then equal volume of n-hexane was added to extract the residual BDE-47. This process was repeated for 3 times. All of the n-hexane phase was collected. After vacuum distillation with the use of rotary evaporator, the solution-hexane remained was transferred into a 5-mL graduated cylinder and concentrated in nitrogen blowing apparatus to 3 mL for the analysis of BDE-47. To determine the recovery ratio of the extraction method on BDE-47, BDE-47 dissolved in 30 mL sterile MSM with a final concentration of 0.115 mg/L was immediately extracted with the used of above-mentioned method for analysis. This process was carried out in triplicate. After measurement, the average recovery ratio of BDE-47 was proved to be as high as 98.2%. No strains were added into the abiotic control groups to examine the abiotic removal of BDE-47 under aerobic (MSM1) and anaerobic (MSM2) conditions. For anaerobic cultivation, all of the operational processes mentioned above were conducted in anaerobic operating box. The culture media for long term cultivation was purged with nitrogen for 5 min to produce an oxygen-free condition.

2.3. Identification of BDE-47 degradation strains

After the aerobic strain and the anaerobic strain with the strongest BDE-47 degradability were obtained, the species identification based on the standard rRNA-PCR method was implemented and the procedures were shown in **Text SI-1** in supporting information. Some other biochemical characteristics of strains were also tested. The morphological characteristics of colony, such as shape, color and size, were observed. Gram staining experiment was conducted as described by Zhang et al. (2013). Other biochemical tests including urease reaction, gelatin liquefaction reaction, hydrogen sulfide reaction, indole reaction, methyl red reaction and Voges-Proskauer test (V–P test) were performed as proposed by Ma et al. (2019) and Zhang et al. (2013).

2.4. BDE-47 degradation experiments

The isolated aerobic *B. cereus* S1 and anaerobic *A. faecalis* S4 with highest BDE-47 degradation efficiency were used in the biodegradation experiments. MSM solution of 27 mL was added into flasks and serum bottles for aerobic and anaerobic cultivation, respectively, and the media were sterilized at 121 °C for 30 min. Then appropriate amount of BDE-47 dissolved in n-hexane was added into the hot incubation medium. The *B. cereus* S1 and *A. faecalis* S4 suspensions were obtained after the strains were activated on the agar slants and cultivated in enrichment medium at 150 r/min and 30 °C for 24 h. After complete volatilization of the n-hexane and cooling-down of the medium, 3 mL of bacterial suspension were inoculated. *B. cereus* S1 was inoculated into the flasks and cultivated aerobically at 150 r/min, 30 °C in dark. *A. faecalis* S4 was inoculated into the serum bottles and cultivated under anaerobic condition at 150 r/min, 30 °C in dark. At each predetermined cultivation time for analysis, i.e., 1, 3, 5, 7, 14 d, three flasks and serum bottles were sacrificed for BDE-47 analysis. The sampling process was exactly the same as that of the screening process of the two highest BDE-47 degradation strains in section 2.2.

To investigate the effects of additional carbon sources on biodegradation of BDE-47 by the two isolates, appropriate amount of yeast extract, ethanol, acetic acid or glucose was added into medium with a final concentration of 50 mg/L. Then the medium was autoclaved. After sterilization, BDE-47 was added using the hexane method mentioned above with a final concentration of 0.115 mg/L. After inoculating 3 mL of *B. cereus* S1 or *A. faecalis* S4 suspension, the culture medium was cultivated at 150 r/min, 30 °C for 14 d in dark. The sampling process was also exactly the same as that of the screening process of the two highest BDE-47 degradation strains in section 2.2. The experimental procedures for investigating the effect of inducers and surfactants on the biodegradation of BDE-47 by the two isolates were same as that for carbon sources. One of 2,4-dichlorophenol, bisphenol A, toluene and hydroquinone was used as the inducer and the concentration was 50 mg/L. One of CTAB, Tween 80, Triton X-100, rhamnolipid and SDS was the surfactant and the concentration was 100 mg/L. MSM with additives but without the isolates, i.e., the abiotic control groups, were used to examine the abiotic removal of BDE-47. The bacterium + MSM with isolates but without additives, i.e., the biotic control groups, were conducted to show the facilitating effect of additives on BDE-47 biodegradation. All the experiments were carried out in triplicate and the results are represented as means ± standard deviations (SD).

2.5. Analytical methods

The concentration of BDE-47 in medium was measured using Shimadzu QP-2010 gas chromatography-mass spectrometry (GC-MS). DB-5 ms (0.0.25mm × 30 m × 0.25um) was used as the GC column and helium was used as the carrier gas. One microliter of the sample was injected under a split injection mode. Temperatures of the injection port and the detector are 230 °C and 280 °C, respectively. The initial temperature of oven was 70 °C holding for 2 min, and then the temperature ramped to 280 °C at the rate of 20 °C/min and held for 10 min. The MS was equipped with electron impact ion source with a temperature of 230 °C. The single ion monitoring (SIM) model was used for quantification. The degradation efficiency of BDE-47 was calculated using the following equation:

$$\text{Degradation efficiency } (\%) = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)$$

where C_0 is the initial concentration of BDE-47 in substrate, C_t is the BDE-47 concentration at time t.

The pseudo-first-order model was employed to describe the biodegradation kinetics of BDE-47, which was described as follows:

$$C(t) = C_0 e^{-kt} \quad (2)$$

where k is the pseudo-first-order rate constant (d^{-1}), and t is the reaction time.

3. Results and discussion

3.1. Screening of BDE-47 degradation strains

After three rounds of domestication with gradient concentration of BDE-47 and the follow-up plate streaking isolation, three aerobic strains (S1, S2 and S3) and three anaerobic strains (S4, S5 and S6) were obtained. The results of BDE-47 degradation by the six strains were shown in Fig. 2. The residual BDE-47 in abiotic control groups showed no difference to the initial after inoculation under both aerobic (MSM1) and anaerobic (MSM2) conditions for 14 d, indicating that BDE-47 could not be removed through abiotic pathway under the experimental conditions. All of the six strains were able to degrade BDE-47 but to different degrees. Under the aerobic condition, strain S1 showed the highest removal efficiency, which was 73.9%. Under the anaerobic condition, the highest removal efficiency of 67.7% was observed for strain S4. Based on these results, the strains S1 and S4 were used as the strains for further experiments under aerobic and anaerobic conditions, respectively. Strains S1 and S4 were both from the site of industrial park which had a long history of BDE-47 contamination, indicating that the microbes from the aged site may have stronger ability to degrade the contaminant.

3.2. Identification of the two highest BDE-47 degradability strains

Based on the result of 16s rRNA sequences and the neighbor-joining method, the phylogenetic tree of isolated trains is constructed (Fig. 3). The 16s rRNA sequence of S1 shown in supporting information Fig. SI-1 exhibits 99% homology with that of *Bacillus cereus* ATC14579. S4 is affiliated within *Alcaligenes faecalis* cluster with the closest relation to *Alcaligenes faecalis* strain IAM12369 based on the BLASTN search as shown in Fig. SI-2. The physiological and biochemical characteristics of the two strains were summarized in supporting information Table SI-1. S1 and S4 are Gram-positive and Gram-negative, which are the same as *Bacillus cereus* and *Alcaligenes faecalis*, respectively. In addition, S1 has strong similarity with *Bacillus cereus* WJ1 and *Bacillus megaterium* YJB3 in the shape of colonies (flat and dry) and the result of indole test (all positive) (Zhang et al., 2013b; Feng et al., 2018). Strain S4, *Alcaligenes faecalis* B14 and *Alcaligenes faecalis* XF1 are biochemically and physiologically similar in oyster white and rough colonies, and positive reaction of methyl red test (Mehandia et al., 2020). Given the results above, the strains S1 and S4 were renamed *B. cereus* S1 and *A. faecalis* S4, respectively.

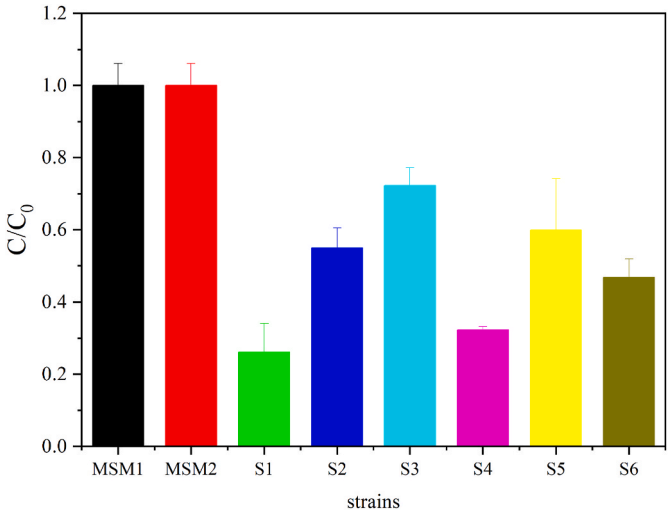

Fig. 2. BDE-47 degradation efficiency by the screened strains under aerobic (S1, S2, and S3) and anaerobic conditions (S4, S5, and S6). The error bars represent mean of triplicate experiments ± standard deviations. MSM1 and MSM2 represent the abiotic control groups under aerobic and anaerobic conditions, respectively.

3.3. Degradation of BDE-47 by B. cereus S1 and A. faecalis S4

The results of BDE-47 degradation by *B. cereus* S1 and *A. faecalis* S4 under aerobic and anaerobic conditions and corresponding first-order reaction kinetics are shown in Fig. 4. With the increase of cultivation time from 1 to 14 d, the removal efficiency of BDE-47 increased from 4.5% to 61.6% for *B. cereus* S1 under aerobic condition, and from 2.7% to 51.6% for *A. faecalis* S4 under anaerobic condition (Fig. 4a). BDE-47 degradation under both aerobic and anaerobic conditions obtained a first-order rate constant of 0.0728 d^{-1} and 0.0514 d^{-1}, and a half-life of 8.7 d and 13.5 d, respectively (Fig. 4b). Compared with the anaerobic degradation of BDE-47 by A. *faecalis* S4, the degradation by *B. cereus* S1 under the aerobic condition shows a higher efficiency, which is attributed to the stronger reproductive and metabolic ability of bacteria under aerobic condition. The growth curves of the two strains in Fig. 5 show that the aerobic *B. cereus* S1 reached the stationary phase within 6 h, while it was 21 h for the anaerobic *A. faecalis* S4.

The results indicated that aerobic degradation of BDE-47 by B. *cereus* S1 is more efficient than anaerobic degradation by *A. faecalis* S4. Similar results were also obtained in previous studies for the biodegradation of BDE-47 individually under aerobic and anaerobic conditions. For aerobic incubation, Xin et al. (2014) isolated a BDE-47 degradative strain *Pseudomonas putida* from soil of e-waste recycling plant obtaining a degradation efficiency of 50.0% within 7 d with an initial BDE-47 concentration of 0.05 mg/L. The result of kinetics reaction showed that the degradation of BDE-47 by *Pseudomonas putida* sp. had a rate constant of 0.076 d^{-1} and a half-life about 9.2 d. Zhang et al. (2013a) found that BDE-47 could be used as the sole carbon source for *Pseudomonas stutzeri* isolated from soil of the brominated flame-retardant production area. At the initial BDE-47 concentration of 0.02 mg/L, the removal efficiency of 97.9% was obtained in 14 d with a rate constant of 0.32 d^{-1} and a half-life of 2.2 d. For anaerobic cultivation, Zhu et al. (2014) reported that degradation efficiency of 82.4% and 62.0% with an initial BDE-47 concentration of 1.003 mg/kg were obtained after incubating for 90 d with anaerobic strains isolated from Mai Po mangrove sediment (MP_m) and fresh water pond sediment (MP_f), respectively. The degradation followed the first-order reaction kinetics with a rate constant of 0.009 d^{-1} and 0.013 d^{-1} and half-life of 76.2 d and 56.9 d, respectively. Tokarz et al. (2008) found that without adding other substances, the removal efficiency of BDE-47 with an initial concentration of 5 mg/kg by anaerobic sludge was 30% within 8 months obtaining a half-life longer than 8 months. These results show that anaerobic degradation of BDE-47 has a much lower first-order rate constant and a longer half-life compared to aerobic degradation. These results, however, were obtained using strains isolated from different sources and cultivated under very different culture conditions. In contrast, the isolates in this study were from exactly one contaminated site and the culture conditions were also same to each other except for the oxygen input. The results confirm that aerobic degradation of BDE-47 is faster than anaerobic degradation. The difference in degradation rate, however, is not that much as that shown in the above literature studies.

The different degradation processes between aerobic *B. cereus* S1 and anaerobic *A. faecalis* S4 would lead to the different degradation

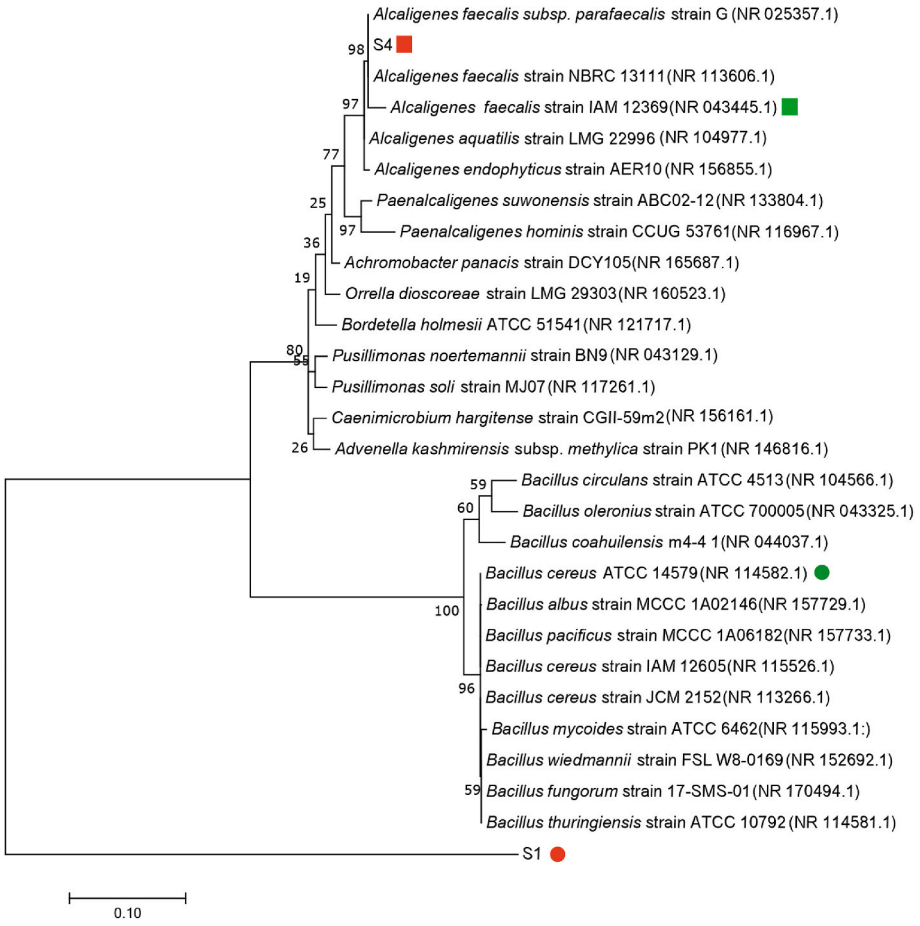

Fig. 3. Phylogenetic tree based on 16s rRNA gene sequences. Red dots show the strains in present study. Green dots indicate the closest strains. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

efficiency of BDE-47. In general, for aerobic incubation, BDE-47 firstly undergoes hydroxylation, methylation or methoxylation under the catalysis of dioxygenase to open the ring to generate various small organic molecules. Then, these molecules enter the tricarboxylic acid (TCA) cycle and eventually are mineralized, which promotes the growth and reproduction of aerobic bacteria and in turn enhances the degradation of BDE-47 (Chou et al., 2016; Wang et al., 2016). Compared to aerobic incubation, the anaerobic cultivation is generally an incomplete process. BDE-47 mainly undergoes reductive debromination reaction including ortho-, meta- and para-debromination, and generates low brominated biphenyl ether which is hard to further break under anaerobic conditions (Lee et al., 2011; Ding et al., 2013).

3.4. The influence of carbon sources on BDE-47 degradation

The addition of carbon sources (i.e., yeast extract, glucose, acetic acid and ethanol) significantly enhanced the BDE-47 degradation by *B. cereus* S1 and *A. faecalis* S4 (Fig. 6). Under aerobic condition with *B. cereus* S1 as the degrader, the addition of yeast extract, glucose, acetate acid and ethanol increased the degradation efficiency by 12.3, 23.5, 22.4, and 24.8%, respectively, compared to the biotic control in which no carbon source was added (61.6%). Under anaerobic condition with *A. faecalis* S4 as the degrader, the degradation efficiency was 51.6% for the biotic control with the absence of carbon sources. The BDE-47 degradation efficiency was increased by 16.1, 7.2, 9.6 and 13.4% with the addition of same carbon sources and yeast extract was the best. While compared to the anaerobic *A. faecalis* S4, the aerobic *B. cereus* S1 achieved relatively high biodegradation efficiency with the addition of same carbon source. It is the co-metabolism between BDE-47 and the added carbon sources that enhances the biomass of microorganisms, thereby increasing the removal efficiency of BDE-47 under both aerobic and anaerobic conditions (Lu et al., 2013; Shi et al., 2013). Lv et al. (2016) indicated that the addition of glucose made a *Pseudomonas putida* grow fast in the first two days, then become slow from the third day, and reach stability after seven days. The BDE-47 degradation efficiency was therefore enhanced by 13.7% within 10 d. While Zhang et al. (2013a) showed that the addition of biphenyl, which had a similar molecular structure to BDE-47, induced the production of enzymes and enhanced the BDE-47 removal efficiency from 87.3% to 94.0%. In addition, ethanol in this study can be used as not only carbon source but also cosolvent to improve the solubility of BDE-47 in solution, and thus increase the bioavailability of BDE-47 and eventually enhance the removal efficiency of BDE-47 under both aerobic and anaerobic conditions. The addition of excessive carbon source, however, could inhibit the degradation efficiency of BDE-47. This is because the microorganisms will first use the excessively added and readily degradable carbon sources without using BDE-47, resulting in a decrease of BDE-47 degradation efficiency (Shi et al., 2013).

3.5. The influence of inducers on BDE-47 degradation

The effect of inducers (2,4-dichlorophenol, bisphenol A, toluene and hydroquinone) on BDE-47 biodegradation were shown in Fig. 7. For aerobic *B. cereus* S1, the degradation efficiency of BDE-47 was increased from 61.6% to 82.1, 77.8 and 76.9% with the addition of 2,4-dichlorophenol, bisphenol A and toluene, respectively. The addition of hydroquinone decreased the removal ratio to 55.3%, indicating inhibitory effect of hydroquinone on BDE-47 degradation by *B. cereus* S1. While for

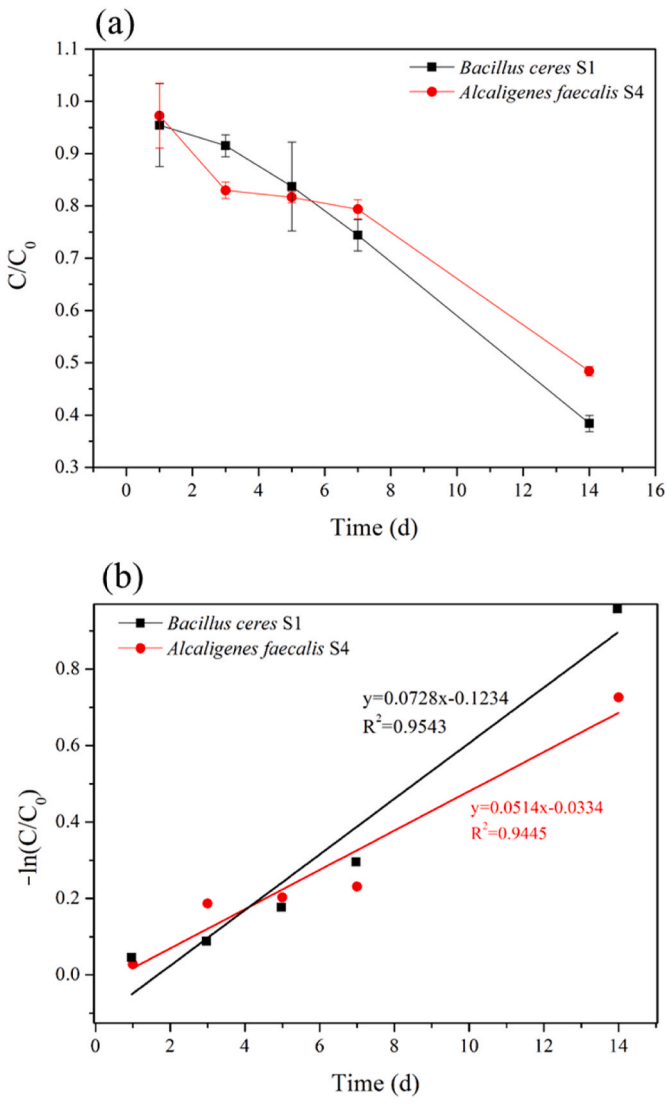

Fig. 4. The degradation of BDE-47 by B. *cereus* S1 and *A. faecalis* S4 under aerobic and anaerobic conditions (a) and corresponding first-order degradation kinetics (b). The error bars represent mean of triplicate experiments ± standard deviations.

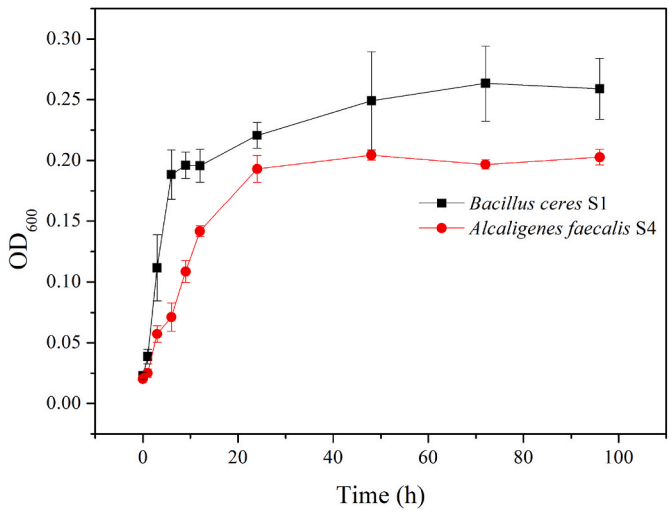

Fig. 5. Growth curves of aerobic *B. cereus* S1 and anaerobic *A. faecalis* S4 under aerobic and anaerobic conditions, respectively. The error bars represent mean of triplicate experiments ± standard deviations.

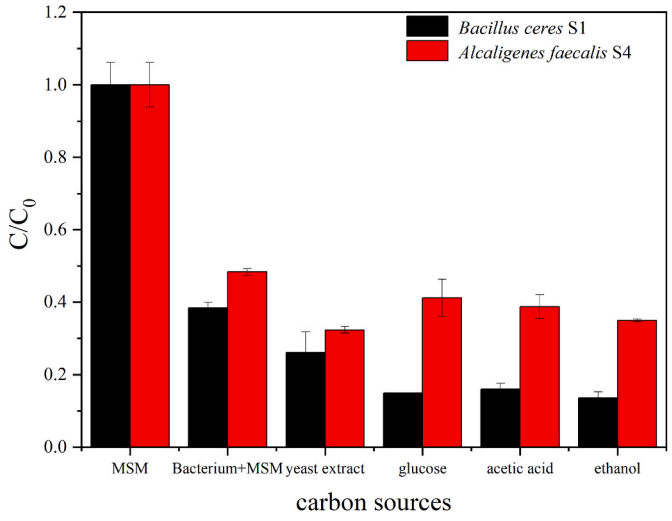

Fig. 6. Effect of carbon sources on BDE-47 degradation by *B. cereus* S1 under aerobic condition and *A. faecalis* S4 under anaerobic condition. The error bars represent mean of triplicate experiments ± standard deviations. The MSM group represents abiotic control. The bacterium + MSM group represents the biotic control under experimental conditions with no carbon sources added.

A. faecalis S4 cultivation under anaerobic condition, the BDE-47 degradation efficiency was increased from 51.6% to 82.6, 67.4, 55.6, and 60.2% with the addition of 2,4-dichlorophenol, bisphenol A, toluene, and hydroquinone, respectively.

BDE-47 with a low bioavailability corresponds to a low level of enzyme production during its biodegradation, which eventually affects the degradation process. Therefore, the inducers which have a molecular structure similar to BDE-47 but more easily utilized by bacteria will induce a more efficient production of enzymes and promote the degradation of BDE-47 (Zhao et al., 2018). Kim et al. (2007) indicated that the addition of diphenyl ether could induce the *Sphingomonas* sp. PH-07 to produce dioxygenases, which cleaved the aromatic ring of PBDEs and eventually increased the capability of degrading PBDEs under aerobic condition. Stiborova et al. (2015) also illustrated that the addition of 0.6 mg/L 4-bromobiphenyl stimulated the activity of dioxygenases, and hence remarkably improved the degradation efficiency of PBDEs in Hradec Kralove (a wastewater treatment plant). During investigating the effect of inducers on BDE-209 degradation in anaerobic sludge, Gerecke et al. (2005) found that the addition of inducers resulted in increase of the removal rate of BDE-209 by 2 fold. In the present study, the degradation efficiency of BDE-47 by bacterium with the addition of 2, 4-dichlorophenol was higher than other inducers both under aerobic and anaerobic conditions. A possible reason is that the chlorine atoms of 2,4-dichlorophenol are in the same position as bromine atoms on the benzene ring of BDE-47, and therefore induces a greater production of enzyme for BDE-47 biodegradation. In addition, the inducer can also be used as a carbon source to increase the biomass of bacteria and promote the degradation of BDE-47 (Wu et al., 2018).

3.6. The influence of surfactants on BDE-47 degradation

The addition of surfactants at the concentration of 100 mg/L to the medium significantly inhibited the degradation efficiency of BDE-47 under both aerobic and anaerobic conditions (Fig. 8). For aerobic incubation, BDE-47 removal efficiency was reduced from 61.6% to 21.7, 11.1, 3.9, 20.1 and 44.5% with the addition of CTAB, Tween 80, Triton X-100, rhamnolipid and SDS, respectively. While for anaerobic degradation, the removal efficiency was decreased from 51.6% to 14.0, 3.7,

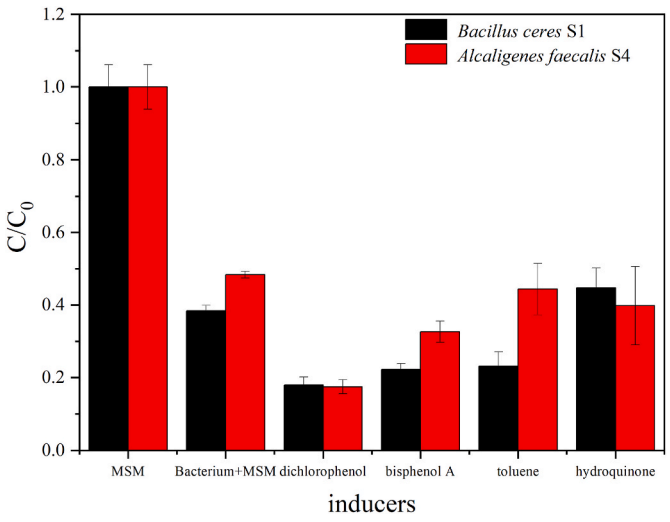

Fig. 7. Effect of inducers on BDE-47 degradation by *B. cereus* S1 under aerobic condition and *A. faecalis* S4 under anaerobic condition. The error bars represent mean of triplicate experiments ± standard deviations. The MSM group represents abiotic control. The bacterium + MSM group represents the biotic control under experimental conditions with no inducers added.

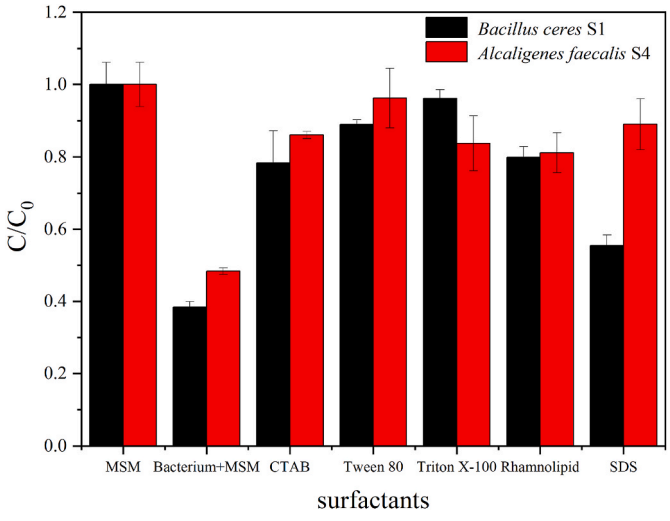

Fig. 8. Effect of surfactants on BDE-47 degradation by *B. cereus* S1 under aerobic condition and *A. faecalis* S4 under anaerobic condition. The error bars represent mean of triplicate experiments ± standard deviations. The MSM group represents abiotic control. The bacterium + MSM group represents the biotic control under experimental conditions with no surfactants added.

16.2, 18.8, and 11.0%, respectively. However, there are many studies in which the enhancing effect of surfactants on biodegradation of PBDEs is reported. For example, the degradation efficiency of BDE-209 was enhanced by 14% with the addition of biosurfactant tea saponin after incubating with *Brevibacillus brevis* for 5 d (Tang et al., 2014). Similarly, Zhou et al. (2007) indicated that Tween 80 exhibited a positive effect on BDE-209 degradation by *white rot fungi* at relatively low concentration (500 mg/L). A widely accepted reason for the enhancing effect of surfactants is that surfactants at concentrations higher than critical micelle concentration (CMC) can increase aqueous solubility of PBDEs and thus the bioavailability of PBDEs to the bacteria. This, however, is not true for the present study, showing that such an effect depends on the combination of contaminants, surfactants and bacteria (Zhao et al., 2018). There is the possibility that when the surfactant concentration is higher than CMC, the BDE-47 will be incorporated into the central zone of surfactant micelles (Huo et al., 2020). Therefore, in order to degrade BDE-47, bacteria must destroy the surfactant layer of micelles, which further increases the difficulty of degradation of BDE-47. There may be some other reasons accounting for the negative effect of surfactants. Surfactant would adsorb on the surface of bacterium, which would occupy the adsorption sites of BDE-47 indicated by bacteria and reduced the degradation of BDE-47 (Sun et al., 2016). In addition, the toxicity of surfactant also poses inhibitory effect on BDE-47 biodegradation.

4. Conclusion

This study investigated aerobic and anaerobic degradation of BDE-47 by the bacterial strains isolated from the real sites contaminated by e-wastes. BDE-47 can be used as the sole carbon source by both aerobic and anaerobic strains isolated from an e-wastes site and the ones with highest degradation efficiency were Gram-positive *B. cereus* S1 and Gram-negative *A. faecalis* S4 for aerobic and anaerobic conditions, respectively. Carbon sources can enhance BDE-47 degradation by both *B. cereus* S1 and *A. faecalis* S4. Inducers basically can enhance the degradation but with hydroquinone as an exception. Surfactants, however, exhibit inhibitory effect. The aerobic process can be more effective than the anaerobic process for degradation of BDE-47, which was demonstrated by the comparison between the two strains for BDE-47 degradation. These results indicate that for on-site remediation of soils contaminated by BDE-47, aerobic condition may be more favorable for the degradation and should be seriously considered. The additives of carbon sources, inducers and surfactants may have varied effect and great care should be taken for their application, especially for surfactants. A pre-test on their effect before applying to the sites is strongly recommended.

Author contributions statement

Lili Huo wrote the manuscript and polished it; **Chenghao Zhao** did the experiments-getting the experimental data; **Tianyuan Gu** processed the experimental data-making figures; **Ming Yan** guided the design and operation of experiments; **Hua Zhong** directed the preparation and revision of the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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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.2022.133739.

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