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Tea saponin enhanced biodegradation of decabromodiphenyl ether by *Brevibacillus brevis*

Shaoyu Tang [a], Jieqiong Bai [b], Hua Yin [a,*], Jinshao Ye [b,*], Hui Peng [c], Zehua Liu [a], Zhi Dang [a]

[a] Key Laboratory of Ministry of Education on Pollution Control and Ecosystem Restoration in Industry Clusters, College of Environment and Energy, South China University of Technology, Guangzhou 510006, Guangdong, China
[b] Department of Environmental Engineering, Jinan University, Guangzhou 510632, Guangdong, China
[c] Department of Chemistry, Jinan University, Guangzhou 510632, Guangdong, China

HIGHLIGHTS

- Main BDE209 metabolites were quantitatively analyzed.
- BDE209 degraded to BDE-208, -207, -206, BDE-203, -197, -196 and BDE-183.
- Level of nona-PBDEs was significantly higher than that of octa-PBDEs and hepta-PBDEs.
- *Brevibacillus brevis* released and utilized nutrient ions during BDE209 degradation.
- Tea saponin efficiently increased the bioavailability and biodegradation of BDE209.

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ABSTRACT

Decabromodiphenyl ether (BDE209) is a ubiquitous persistent pollutant and has contaminated the environment worldwide. To accelerate BDE209 elimination and reveal the mechanism concerned, the biosurfactant tea saponin enhanced degradation of BDE209 by *Brevibacillus brevis* was conducted. The results revealed that tea saponin could efficiently increase the solubility of BDE209 in mineral salts medium and improve its biodegradation. The degradation efficiency of 0.5 mg L^{-1} BDE209 by 1 g L^{-1} biomass with surfactant was up to 55% within 5 d. Contact time was a significant factor for BDE209 biodegradation. BDE209 biodegradation was coupled with bioaccumulation, ion release and utilization, and debromination to lower brominated PBDE metabolites. During the biodegradation process, *B. brevis* metabolically released Na^+, NH_4^+, NO_2^- and Cl^-, and utilized the nutrient ions Mg^{2+}, PO_4^{3-} and SO_4^{2-}. GC–MS analysis revealed that the structure of BDE209 changed under the action of strain and nonabromodiphenyl ethers (BDE-208, -207 and -206), octabromodiphenyl ethers (BDE-203, -197 and -196) and heptabromodiphenyl ether (BDE-183) were generated by debromination.

1. Introduction

As common flame retardant additives, polybrominated diphenyl ethers (PBDEs) have been widely used in electronic circuit boards and manufacture materials like plastics, textiles and building materials (Hall and Thomas, 2007; Chen and Hale, 2010). Along with the wide application of PBDEs, they have become global environmental problems and caused worldwide attention (Alcock et al., 2011). PBDEs have the characteristics of high hydrophobicity, environmental persistence, bioaccumulation in the food chain and threat to the health of human and animals (Johansson et al., 2011). Therefore, most of PBDEs products have been banned or voluntarily withdrawn from use in many regions of the world (Sun et al., 2012). However, due to shorter half life and higher bioavailability than other kinds of PBDEs, BDE209 remains legal in most countries, inducing a great environmental concern. For example, the concentration of BDE209 in carp collected in the Dongjiang River of Guangdong, South China, was up to 220 ng g^{-1} (He et al., 2012). High concentration of BDE209 in water and sediment has a series of negative biological effects on aquatic organisms (She et al., 2013). Moreover, BDE209 is also subchronic/chronic toxic to the human liver and the thyroid gland (Zhang et al., 2011).

To eliminate PBDEs from terrestrial and aquatic environments, some physico-chemical activities including burying, incineration,

* Corresponding authors. Tel.: +86 20 39380569 (H. Yin). Tel.: +86 20 85226615 (J. Ye).
E-mail addresses: huayin@scut.edu.cn (H. Yin), jsye@jnu.edu.cn (J. Ye).

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chemical hydrolysis and photo-catalytic oxidation have been investigated for the potential treatment (Chow et al., 2012). Nevertheless, because of some disadvantages of these methods, such as energy consumption, high cost, solar irradiation limitation or secondary pollution, concerns have recently shifted to developing more suitable techniques, especially biodegradation for PBDEs control.

Some published studies on the degradation of PBDEs in sewage sludge and sediments had exhibited the bioremoval abilities of certain indigenous microorganisms (Yen et al., 2009). Among these reports, the effects of some environmental factors and supplemental substrates on PBDEs biodegradation were revealed to elucidate the best conditions (Zhang et al., 2013). As one of the nutrients for PBDEs co-metabolism, glucose was found to be able to enhance biodegradation of 4-monobrominated diphenyl ether in the Jhongsing sludge (Shih et al., 2012). Robrock et al. reported the aerobic transformation of tetra-, penta- and hexa-BDE, and stoichiometric release of bromide during PBDEs transformation (Robrock et al., 2009). Although the biodegradation of PBDEs has been illustrated by these studies, the quantitative analysis of metabolites and relation between PBDEs biodegradation and cellular metabolism during biodegradation process are still required further study. Moreover, since BDE209 has a relatively low degradation rate due to its high hydrophobicity, adding surfactant proved to be a comparatively straightforward and well established method to overcome these problems (Li and Zhu, 2012).

Therefore, the present study aimed to investigate BDE209 biodegradation by *Brevibacillus brevis* and cellular ion metabolism in the presence of tea saponin, a biosurfactant widely utilized for its excellent properties and biodegradability. Effects of surfactant concentration and contact time on the degradation were investigated, and the mechanisms of biodegradation were revealed through quantitative analysis of main BDE209 metabolites and examination of ion utilization and release during the degradation process.

2. Materials and methods

2.1. Strain and chemicals

B. brevis was isolated from the contaminated sediment samples of Guiyu in Guangdong Province, China, and was preserved in our laboratory.

BDE209 (98% purity) was purchased from Sigma Aldrich (St. Louis, MO, USA).

Other reagents were of analytical grade and purchased from Guangzhou Chemical Reagent Factory, China.

2.2. Microbial culture

B. brevis was inoculated into the nutrient medium containing 5 g L^{-1} beef extract, 10 g L^{-1} peptone and 5 g L^{-1} NaCl, and incubated at 30 °C on a rotary shaker at 130 r min^{-1} for 24 h. Subsequently, the strains were harvested by centrifugation at 3500*g* for 10 min, washed three times with sterile distilled water and used for further studies.

2.3. Solubilization effect of tea saponin on BDE209

Enhanced solubilization of 0.5 mg L^{-1} BDE209 by tea saponin was performed at 30 °C in 20 mL mineral salts medium (MSM), which was composed of (in mg L^{-1}) 90 $Na_2HPO_4{\cdot}12H_2O$, 30 KH_2PO_4, 5 NaCl, 10 NH_4NO_3, 4 $MgSO_4{\cdot}7H_2O$, 1 $CuSO_4{\cdot}5H_2O$, 1 $MnSO_4{\cdot}H_2O$, 1 $FeSO_4{\cdot}7H_2O$ and 1 $CaCl_2{\cdot}2H_2O$ by shaking on a rotary shaker at 130 r min^{-1}. To elucidate the extent of the solubility enhancement of BDE209 by tea saponin, the 50 mL Erlenmeyer flasks containing 20 mL MSM with 0.5 mg L^{-1} BDE209 and surfactant at 0, 1, 5, 10, 20, 40 and 60 mg L^{-1} individually were shaken at 130 r min^{-1} for 24 h, equilibrated for 12 h in the dark, and centrifugated at 3500*g* for 15 min. Then 10 mL supernatant was transferred and extracted twice by ultrasound method using isovolumetric extractant ($V_{\text{normal hexane}}$:$V_{\text{methylene chloride}}$ = 1:1). Three samples for each experiment were taken and the mean values were used in calculations.

2.4. Effect of tea saponin concentration and contact time on BDE209 biodegradation

MSM containing 0.5 mg L^{-1} BDE209, 1.0 g L^{-1} *B. brevis* and 0–60 mg L^{-1} tea saponin was incubated in the dark at 30 °C in a rotary shaker at 130 r min^{-1} for 5 d, to determine the effect of surfactant concentration on BDE209 biodegradation. Meanwhile, the control without biomass was conducted under the same condition. The influence of degradation time, which was set at 1–7 d, was investigated when surfactant concentration was 20 mg L^{-1}. After degradation, the samples were extracted twice by ultrasound method with 20 mL organic solvent ($V_{\text{normal hexane}}$:$V_{\text{methylene chloride}}$ = 1:1). The organic part was collected, followed by concentrating using a rotary evaporator at 35 °C. Subsequently, the residues were analyzed by capillary gas chromatograph – mass spectrometer (GC–MS) (QP2010, Shimadzu). The controls were run in parallel in flasks with solutions that were uninoculated.

2.5. Ion metabolism during biodegradation

After biodegradation of BDE209 under different surfactant concentrations and degradation time, the samples were centrifuged at 3500*g* for 10 min. Then the resultant supernatant was filtered using a 0.22-μm polyether sulfone filter. The concentration change of nutrient ions in solutions during BDE209 biodegradation by *B. brevis* was determined by an ICS-900 ion chromatography system (Dionex, Sunnyvale, USA).

2.6. Quantitative analysis of metabolites during BDE209 biodegradation

The same samples collected in Section 2.4 were freeze-dried, and then extracted twice with 20 mL mixture of n-hexane and acetone (1:1, V/V) by ultrasonic method for 30 min. Then 20 mL n-hexane and methylene chloride (1:1, V/V) was added into the sample to repeat again the operation. The organic part was collected and concentrated by 10 mL chromatogram class n-hexane for instrumental analysis. Metabolites of BDE209 degradation were analyzed by GC–MS, using electron capture negative ionization

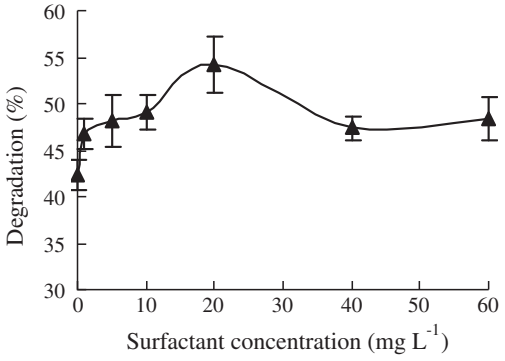

Fig. 1. Effect of surfactant concentration on degradation of 0.5 mg L^{-1} BDE209 by 1.0 g L^{-1} *B. brevis*.

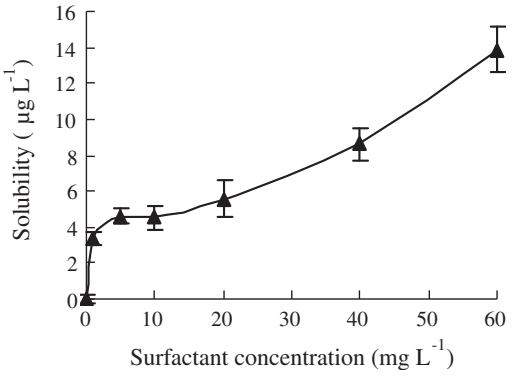

Fig. 2. Effect of surfactant concentration on BDE209 solubility.

(ECNI) in the selective ion monitoring (SIM) mode. A capillary column (15 m × 0.25 mm × 0.1 μm, J&W Scientific) was used for separation. Helium was used as the carrier gas with a constant flow at 1.5 mL min^{-1}. The column temperature program started at 110 °C, held for 5 min. Subsequently, the oven was heated to 310 °C at a rate of 10 °C min^{-1} and held for 10 min. The GC–MS interface temperature was maintained at 280 °C. Mass spectra were recorded at 1 scan s^{-1} under electronic impact with 70 eV electron energy, mass ranged 50–950 atoms to mass unit. The temperature of ion source was set at 260 °C.

3. Results and discussions

3.1. Influence of surfactant on BDE209 biodegradation

Tea saponin promoted the biodegradation of BDE209 significantly and the degradation rate increased by nearly 14% (Fig. 1). The positive impacts of surfactant were owing to certain attractive advantages tea saponin had: (i) as one of the hydrophobic organic pollutants, BDE209 could be emulsified and become more available for the further biodegradation under the influence of surfactant. The growing solubilization of BDE209 by tea saponin shown in Fig. 2 further confirmed this inference; (ii) surfactant micelles could act as hydrogen donors and thus enhance the biodegradation; (iii) surfactant could regulate the hydrophobicity of *B. brevis* (Ye et al., 2013a, b), through promoting the affinity of this strain

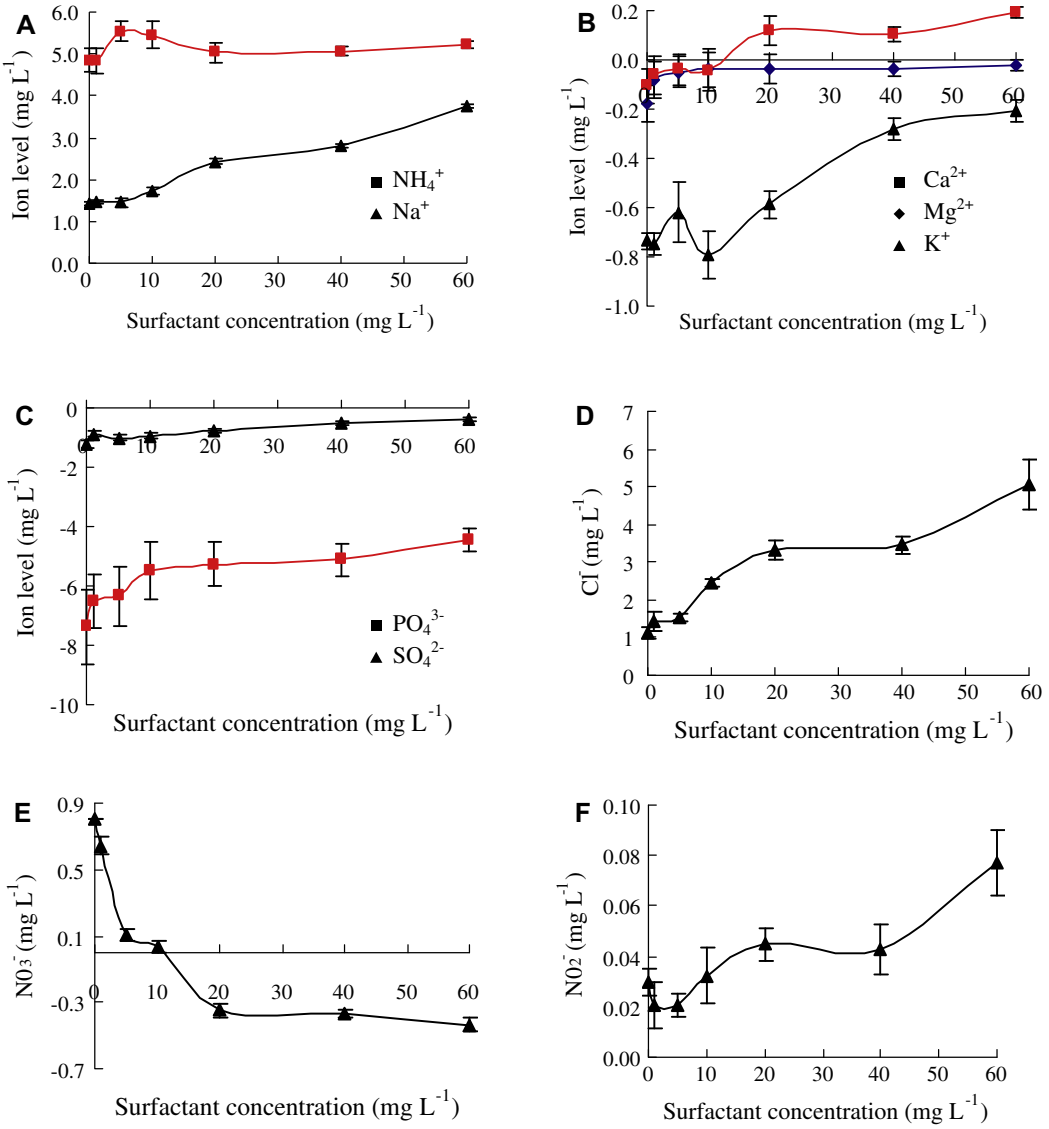

Fig. 3. Ion release and utilization during biodegradation of 0.5 mg L^{-1} BDE209 by 1.0 g L^{-1} *B. brevis* at 30 °C for 5 d.

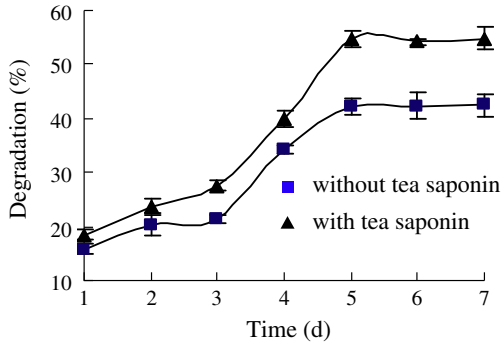

Fig. 4. Influence of contact time on biodegradation of 0.5 mg L^{-1} BDE209 by 1.0 g L^{-1} *B. brevis* at 30 °C.

to BDE209, consequently leading to a higher bioavailability of BDE209; and (iv) surfactant could also serve as nutrient for cell metabolism due to its bioavailability, which partially contributed to the promotion of BDE209 biodegradation.

However, the positive enhancement was depressed to some extent when surfactant content exceeded 20 mg L^{-1}. This was probably ascribed to the unique hydrophobic and hydrophilic domains of surfactant, which made tea saponin at high concentration excessively increase the permeability of cell membrane of *B. brevis* and induce the apoptosis of some cells, reducing the degradation kinetics (Song et al., 2010). Moreover, the exposure of *B. brevis* to higher concentration of dissolved BDE209 caused by higher surfactant level (Fig. 2) would exhibit greater negative influence on cellular activities. Thereby, surfactant at high concentration depressed BDE209 removal when compared to that at low level, but still enhanced the degradation in comparison with the control in the current study.

3.2. Nutrient utilization by *B. brevis* during BDE209 biodegradation

Some studies have showed that the extent of PBDEs bioaccumulation and biodegradation by microorganisms is partially related to the utilization of nutrient substances (Chai et al., 2013), since this process depends on metabolism. To confirm this and reveal the correlation between BDE209 biodegradation and nutrient utilization, the concentrations of some metabolically essential ions in the solutions were detected after BDE209 biodegradation.

Except for NO_3^-, the concentrations of all detected ions in solutions were obviously rising with the level of surfactant (Fig. 3). This finding revealed that surfactant at high levels used in the current experiments enhanced the release of some intracellular ions and depressed the utilization of certain extracellular nutrient ions. Surfactants are a group of compounds which are capable of altering the permeability characteristics of cell membrane where they can intercalate with lipid bilayers and increase penetration (Shaw et al., 2012). The rising penetration induced by tea saponin at high levels triggered intracellular ions leakage. Apart from penetration, similar to the effects of other nutrient on pollutants bioelimination, tea saponin could also serve as nutrient for *B. brevis* metabolism, which was partially responsible for the decreasing uptake of the nutrients in solution.

Na^+ is involved in the energy metabolism of bacteria (Buckel and Thauer, 2013). Its gradients play an essential role in conservation of energy, as well as in ionic homeostasis (Juárez and Barquera, 2012). Moreover, *B. brevis* used in the current study contained abundant intracellular Na^+, since it was inoculated into the nutrient medium containing 5 g L^{-1} NaCl. As BDE209 biodegradation was a process dependent on metabolism, 1.4–3.7 mg L^{-1} of Na^+ was released to solution when tea saponin concentration varied from 0 to 60 mg L^{-1}, respectively.

Previous researches on the relation between the role of metal cations and phosphorus removal confirmed that some metal cations, especially K^+, Ca^{2+} and Mg^{2+}, which were cotransported with

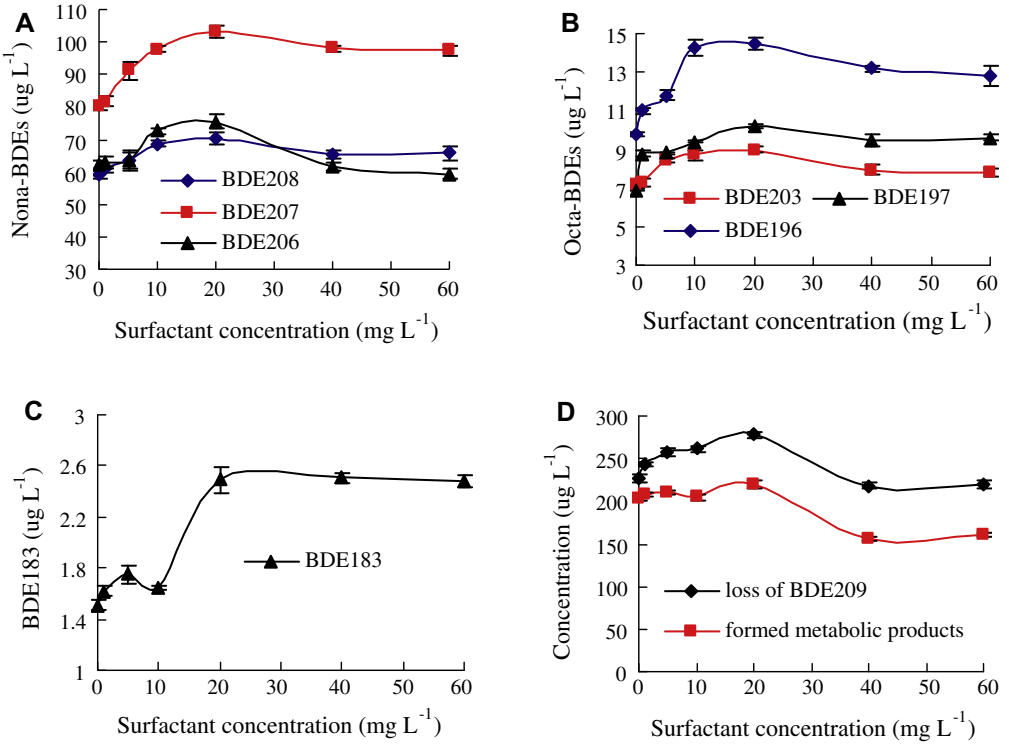

Fig. 5. Metabolites of biodegradation of 0.5 mg L^{-1} BDE209 by 1.0 g L^{-1} *B. brevis* with 0–60 mg L^{-1} surfactant at 30 °C for 5 d.

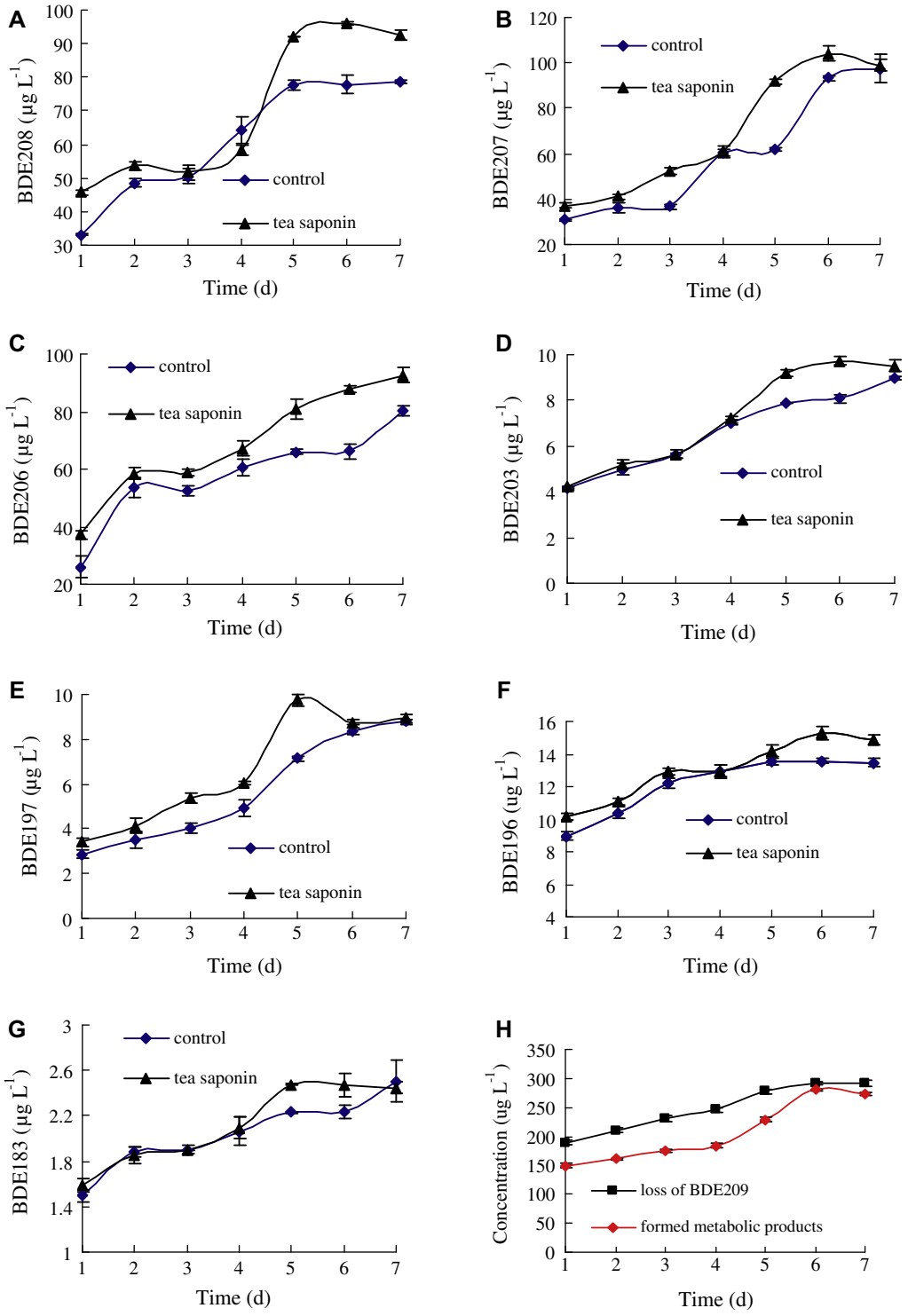

Fig. 6. Metabolites of biodegradation of 0.5 mg L^{-1} BDE209 by 1.0 g L^{-1} *B. brevis* with 20 mg L^{-1} surfactant at 30 °C for 1–7 d.

phosphate into and out of bacterial cells, were important for cells in maintaining electroneutrality inside the bacterial cell (Barat et al., 2008). This is consistent with the utilization trends of K^+, Mg^{2+} and PO_4^{3-} in the current experiment. In many ecosystems, nutrient PO_4^{3-} is always a growth limiting factor. Moreover, the uptake of organic nutrients and their further degradation are performed by some active pathways followed by catabolism through an intracellular phosphorylative pathway, which triggered the utilization of extracellular PO_4^{3-}. The degradation of BDE209 and the maintenance of cellular activities during this process were depended on the intracellular phosphorylation and dephosphorylation pathways. Fig. 3C exhibited the utilization trends of PO_4^{3-}, although high level of surfactant depressed the uptake of PO_4^{3-} to some extent.

Although MSM used in the current study was composed of 10 mg L^{-1} NH_4NO_3, *B. brevis* still exported substantial levels of NH_4^+ into MSM solution (Fig. 3A), which was mainly due to the following reasons: (i) *B. brevis* was inoculated into the nutrient

medium containing 5 g L^{-1} beef extract and 10 g L^{-1} peptone, which ensured that the intracellular concentration of NH_4^+ was higher than that in solution. (ii) NH_4^+ efflux was partially owing to the apoptosis occurred in some of the cells or the rising penetration of membrane induced by BDE209. (iii) NH_4^+ is one of the signaling molecules tightly linked to amino acid availability and transport (Čáp et al., 2012). The export of NH_4^+ was coupled with the release of amino acids which was secreted from some proteins participating in BDE209 biodegradation.

The concentration of NO_3^- and NO_2^- exhibited opposite trends with the increase of surfactant dosage (Fig. 3E and F), partially owing to the bioreduction of NO_3^-. After NO_3^- uptake by cell, the reduction to NO_2^- by nitrate reductase may be the next step in the nitrogen assimilation process (Cao et al., 2008). The substantial increase of NO_2^- in solutions during the biosorption of $Cu(NO_3)_2$ by *Stenotrophomonas maltophilia* further confirmed this inference (Ye et al., 2013a, b). Besides, part of the increased NO_2^- in solution could also come from the export from inside cells to the extracellular environment.

As common available electron acceptors, SO_4^{2-} usually participates in electron accepting processes (Schreiber et al., 2004), which are associated with pollutants biodegradation and nutrients utilization.

3.3. Influence of contact time on BDE209 biodegradation

As shown in Fig. 4, the biodegradation capacity increased notably as time increased and biodegradation equilibrium nearly arrived within 5 d. The result verified that *B. brevis* could degrade BDE209 fairly rapidly in the first 2 d due to vigorous metabolic activity the cells possessed. Thereafter, the residual BDE209, metabolites of BDE209 degradation and nutrient deficiency all had certain negative influences on strain activities, which made the increasing of degradation efficiency not obvious during the 2nd–3rd d. And then, the efficiency of BDE209 uptake increased quickly again with elongated contact time and reached a state of equilibrium on the 5th d. These results were possibly ascribed to some enzymes produced by the strains after adaptive phase, which helped to further degrade BDE209 and its metabolites.

3.4. Effect of surfactant concentration on metabolites of BDE209 biodegradation

Although BDE209 debromination has been revealed in some reports, quantitative analysis of BDE209 metabolites is still limited. It was demonstrated from samples of BDE209 biodegradation by GC–MS analysis that BDE209 was catalytically debrominated to lower brominated PBDEs, including nonabromodiphenyl ethers (BDE-208, -207 and -206), octabromodiphenyl ethers (BDE-203, -197 and -196) and heptabromodiphenyl ether (BDE-183), within a 5 d period. However, no other lower brominated metabolites were detected under current experimental condition, implying that the further debromination and degradation of the detected metabolites would be a rapid step. Among these detected metabolites, the concentrations of the higher brominated PBDEs, such as BDE208, BDE207 and BDE206 (Fig. 5A), were notably higher than those of the lower brominated ones (Fig. 5B and C), proving the above inference that the further debromination of lower metabolites was relatively quick so that an appropriate accumulation for the detection was not available. This type of debromination was also found in the photolytic degradation of decabromodiphenyl ether and decabromodiphenyl ethane (An et al., 2008; Wang et al., 2012), and was also consistent with the finding in biotransformation of PBDEs by other species, in which the metabolic rates of PBDEs in both beluga whale and rat were inversely associated with the degree of halogenation (McKinney et al., 2006).

Fig. 5D exhibited that the amount of BDE209 degraded was significantly more than that of the total detected metabolites. There may be two reasons for this discrepancy. First, some unidentified and transformation products, such as lower bromo-products through ring opening, were produced. Second, since BDE209 has 209 congeners and not all standards were available, other lower bromodiphenyl ethers that had no standards to compare formed with the decrease of BDE-209. Since higher toxic metabolites were not accumulated, the degradation of BDE209 by *B. brevis* was an effective process.

Concentrations of the metabolites elevated to the maximum when tea saponin level was 20 mg L^{-1}. Subsequently, these decreased to some extent with the increase of surfactant content, which was in accordance with the results shown in Fig. 1. This result meant that the joint influence of cells and surfactant on BDE209 debromination was mainly dependent on the concentration of surfactant. Owing to the similar trends of BDE209 debromination and biodegradation with the increase of surfactant level, it revealed that BDE209 biodegradation primarily lay on its debromination.

3.5. Metabolites during BDE209 biodegradation process

It was displayed in Fig. 6 that both BDE209 biodegradation and metabolite concentration ascended with contact time. The level of each metabolite increased quickly on the 4th–5th d, and then slowed down, implying that the further biodegradation of BDE209 by *B. brevis* was relatively depressed with elongated degradation time. Because of nutrient deficiency in MSM under this condition, the metabolic activity of *B. brevis* was negatively affected, consequently decreasing the debromination and degradation of BDE209 and its metabolites (Shi et al., 2013). Furthermore, the degradation activity was further depressed by the accumulated metabolites. Fortunately, there was no further accumulation of the detected metabolites by the end of the 6th d, except for BDE206. Compared with the control, tea saponin presented an enhanced debromination during the process.

4. Conclusions

BDE209 could be degraded effectively by *B. brevis*. Tea saponin significantly increased the solubility of BDE209 in water and enhanced its biodegradation and debromination. The degradation efficiency of 0.5 mg L^{-1} BDE209 by 1.0 g L^{-1} biomass was up to 55% within 5 d. Contact time was a major factor influencing BDE209 degradation and removal rate could reach a state of equilibrium on the 5th d. BDE209 biodegradation included bioaccumulation, ion release and utilization, and debromination to other lower brominated PBDEs. During biodegradation process, *B. brevis* metabolically released Na^+, NH_4^+, NO_2^- and Cl^-, and utilized the nutrient ions Mg^{2+}, PO_4^{3-} and SO_4^{2-}. The structure of BDE209 was transformed by the strain, nonabromodiphenyl ethers (BDE-208, -207 and -206), octabromodiphenyl ethers (BDE-203, -197 and -196) and heptabromodiphenyl ether (BDE-183) were the main metabolites generated by debromination.

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