



Short Communication

Sediment microbial fuel cell prefers to degrade organic chemicals with higher polarity

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HIGHLIGHTS

- The degradation rates of 68 organic chemicals in a 30 L SMFC were compared.
- SMFC prefers to stimulate the degradation of organic chemicals with higher polarity.
- The results are crucial for choosing a suitable method for a given contaminated site.

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ABSTRACT

By operating a SMFC in heavily contaminated sediment and analyzing its global organic chemical degradation profile, this study showed a brief trend that SMFC prefers to stimulate the degradation of organic chemicals with higher polarity. As a comparison, adding nitrate as a microbial respiration-based sediment remediation strategy preferred lower polarity chemicals. Both SMFC and nitrate reactors showed high degradation capacity in benzene homologs. These results provide crucial information for the selective and proper application of SMFC in bioremediation.

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1. Introduction

Microbial respiration is a central driving force for various biogeochemical cycles and bioremediation processes. Lacking of favorable electron acceptor is an important factor that limits microbial contaminant degradation in subsurface environments (e.g. sediments, soils) on the earth. To stimulate the intrinsic bioremediation process in contaminated subsurface environments, adding electron acceptors such as oxygen, sulfate, nitrate or ferric iron have been suggested and proved to be an efficient strategy (termed respiration-regulating remediation, RRR). The anode in a bioelectrochemical system (BES), particularly microbial fuel cell (MFC), is a recently developed artificial electron acceptor to stimulate the microbial degradation of various organic contaminants simultaneously with electricity generation.

Generally, the electron acceptors in microbial respirations can be divided into two forms: soluble (e.g. oxygen, sulfate or nitrate) and insoluble (e.g. mineral oxides, anodes). Given the different physicochemical properties of those electron acceptors, different effects on the microbial community composition and function can be assumed in RRR. In support, elevated sulfate reduction caused significantly higher polycyclic aromatic hydrocarbons (PAHs) degradation efficiency than nitrate reduction and methanogenesis in sediments (Chang et al., 2003; Lovley, 1995). Sediment-deployed MFC (SMFC) and amorphous ferric hydroxide showed different degradation efficiencies on phenanthrene and pyrene (Yan et al., 2012). However, to date, most RRR studies used sole electron acceptor or focused only on one or a group of contaminants (Lovley, 1995; Muyzer and Stams, 2008; Wang et al., 2012). The wide degradation pattern of organic chemicals in RRR has not been reported independently or comparatively, even though it could provide crucial information for choosing the suitable remediation method in a given contaminated environments.

The main aim of this study is to provide the wide degradation pattern of sediment organic chemicals in SMFC. It has been

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reported that SMFC electrode and biofilms can adsorb contaminants and co-localize the contaminants, microbial degraders and electron acceptor on the same surface (Zhang et al., 2010). Considering the low mobility of chemicals with low polarity in sediments, we hypothesized that chemicals with higher polarity would be more readily degraded as its higher mobility and electrode-accessibility in the SMFCs. The results showed that SMFC indeed preferred to stimulate the degradation of high-polar compounds while, as a comparison, addition of nitrate as an RRR preferred low-hydrophobic compounds. The differential degradation patterns shown here provide crucial information for a selective application of SMFC and highlight the necessity of such an investigation before choosing the suitable bioremediation method for a given contaminated site.

2. Methods

2.1. Construction of the SMFC and nitrate treatment reactors

Sediment and water samples heavily contaminated by electronic industry and sewage discharge were collected as described in our previous study (Xu et al., 2014). Plexiglass bioreactors with approximately 30 L volume were used to perform the biodegradation experiments (Fig. S1). The bioreactors were firstly supplemented with sediments (17 cm thick) and then covered with 9 L of the sampled water. For each SMFC, six connected pieces of graphite felt (10 × 10 cm) were vertically inserted into the sediment as an anode array. A foam-bonded graphite felt (20 × 20 cm) located right below the surface of the upper water was used as cathode (Fig. S1). The anode array and cathode were connected with rubber-sealed copper wire and all bonding-sites were covered with silicone rubber to avoid corrosion. The voltage of a resistance of 1000 Ω between the anode and cathode was recorded. Nitrate-treatment reactors containing the same volumes of sediment and waters were operated as comparisons to the SMFCs. To provide continuous nitrate-respiration, 150 mg/L of nitrate was injected into the sediments per day according the nitrate-reduction rate in the sediments. Open-circuit SMFC or no nitrate-addition reactors were operated as controls. All bioreactors were operated in nature temperature (25 ± 5 °C).

2.2. Chemical and electrochemical analyses

The sediment at different height and different distance to anode were sampled, homogenized and stored under −20 °C before analysis. For GC–MS analysis, 10 g of freeze-dried sediment was extracted with 200 mL of dichloromethane (DCM) for 48 h using a Soxhlet extractor. In order to remove the elemental sulfur, activated copper granules were added during extraction. To obtain a wide degradation pattern, the organic compounds were eluted with 30 mL of *n*-hexane (HEX), *n*-hexane/dichloromethane (4/1, HD), dichloromethane (DIC), or methanol (MET) separately. The eluates were evaporated to 1 mL, 1 mL, 1 mL, 25 mL respectively under N₂ stream. All extracts were analyzed by a Thermo Finnigan Trace DSQ GC–MS. Compounds were separated using a 30 m × 0.25 mm i.d. fused-silica capillary column coated with a 0.25 μm film of 5% diphenyl and 95% dimethyl polysiloxane.

Organic compounds were identified using the NIST mass spectral search program and NIST/EPA/NIH mass spectral library ver. 2.0. The compounds with the maximum matching degree >90% were chosen. The relative amounts of the compounds were represented by their absolute areas. The degradation rate in the SMFC was calculated as: $DR_{SMFC} = (A_0 - A_{SMFC})/A_0 \times 100\%$, wherein A_0 is the compound area of in the original sediment, A_{SMFC} is the area of the compounds in SMFC-sediment. The same method was also

used to calculate the degradation rate in nitrate ($DR_{nitrate}$) and control reactors ($DR_{SMFC-control}$, $DR_{nitrate-control}$).

3. Results and discussions

3.1. Current generation of SMFCs

Multi-anode has been suggested to be a suitable strategy to enhance the performance of scaled-up or field-applied SMFCs due to the increased treatment scope and electric charges (Hsu et al., 2013). The voltage of our SMFC increased from 0.16 V to a relative stable level of 0.6 V within 20 days (Fig. S2). The maximum power density of the SMFCs was 4.32 mW/m², while the current density was 9.29 mA/m². The internal resistance was approximately 96.68 Ω calculated from the polarization (Fig. S2). The power density was comparable to many other reported SMFCs with different scales (Hsu et al., 2013; Zhang et al., 2011; Zhou et al., 2014; Yang et al., 2015). Generally, SMFCs generate lower power density compared to the MFCs with anodes exposing to water, because (i) the sediment microbial metabolisms were several orders slower than that in aquatic environments (Hoehler and Jorgensen, 2013); (ii) the low chemical diffusion efficiencies of organic matters and protons in sediments might constraint the electron flux and cause local pH decrease, respectively; (iii) the heterogeneity of sediments may cause unavailable regions for biofilm development.

Microbial community analysis showed that the microbial community on the anode had a lower diversity and was significantly different with other sites in the reactors, as indicated by the Shannon-Wiener Index (Fig. S3). This is consistent with the assumption that SMFC current generation might have minor effects on the microbial community distant from the anode (Wang et al., 2012). Most of the detected bacteria were related to contaminated environments or anode-respiring biofilms. The most frequently detected current-generating bacteria, *Geobacter* species, in SMFCs were not detected. Alternatively, *Geothrix fermentans* (100% identity) or some other unreported current-generating bacteria on the anode may catalyzed the current generation (Bond and Lovley, 2005).

3.2. The enhanced biodegradation of organic matters in SMFC

68 putative compounds (Table S1) in the sediments were detected using GC–MS and divided into four groups: HEX, HD, DIC and MET according to the eluents used, of which the polarity order can be assumed as: HEX < HD < DIC < MET.

HEX-group consisted of 13 compounds, mainly linear or methyl-branched alkanes and alkenes from C11 to C43 with main contents of C13–C23. The degradation rates of those 13 compounds were relatively high in SMFC (20–48%) than that in control (5–26%) (Fig. 1A and Table S1). Generally, the compounds with more than 22 carbon atoms degraded slower than C11–C19 compounds in both SMFC and control reactors. For C11–C19 compounds, the degradation rates decreased from 48% to less than 30% in SMFC with the increase in carbon atoms while a contrary trend was observed in control reactors. The results suggested that the SMFC not only enhanced the degradation rates of those compounds but also altered the degradation pattern compared with the natural degradation process.

HD-group (Fig. 1B) contained 18 compounds with different molecular structures such as aromatic hydrocarbons, cyclopentane or cyclohexane hydrocarbons with nitrogen or sulfur atoms. Due to the complex molecular structures, these compounds showed relative low degradation rates in both SMFC and control reactors (<15%) compared with other groups, except for that four

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