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## Short Communication

# Enhancing the bioremediation by harvesting electricity from the heavily contaminated sediments



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## HIGHLIGHTS

- A 100 L SMFC was operated in heavily contaminated sediment for over 2 years.
- The sediments could provide sufficient electron donors for long-term SMFC operation.
- SMFC–PMS system is a durable and efficient tool in bioremediation and power-supply.

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## ABSTRACT

To test the long-term applicability of scaled-up sediment microbial fuel cells (SMFCs) in simultaneous bioremediation of toxic-contaminated sediments and power-supply for electronic devices, a 100 L SMFC inoculate with heavily contaminated sediments has been assembled and operated for over 2 years without external electron donor addition. The total organic chemical (TOC) degradation efficiency was 22.1% in the electricity generating SMFCs, which is significantly higher than that in the open-circuited SMFC (3.8%). The organic matters including contaminants in the contaminated sediments were sufficient for the electricity generation of SMFCs, even up to 8.5 years by the present SMFC theoretically. By using a power management system (PMS), the SMFC electricity could be harvested into batteries and used by commercial electronic devices. The results indicated that the SMFC–PMS system could be applied as a long-term and effective tool to simultaneously stimulate the bioremediation of the contaminated sediments and supply power for commercial devices.

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## 1. Introduction

Due to the hydrobiological metabolisms and geochemical processes (e.g. surface runoff, adsorption and precipitation), a significant partial of biomass and organic contaminants on the Earth accumulate in aquatic sediments which can be considered as an energy reservoir if the chemical energy stored in sediments could be extracted (Tender et al., 2002). Microbial fuel cells (MFCs) have been developed as a promising tool to harvest energy from organic matters in various environments. Compared with normal aquatic MFCs, sediment-deployed MFCs (SMFCs) face more limits due to the heterogeneous microenvironments, higher contaminant concentration and low chemical diffusion efficiency in sediments.

MFCs are developed for application in practical environments. Although most reported MFCs were operated with small volumes (<1 L) and within short operation periods (from hours to months), increasing scaled-up MFCs toward practical application have been reported (Cui et al., 2014; Feng et al., 2014; Hong et al., 2008; Zhang et al., 2011). However, the output electric power (generally  $\leq 0.8$  V) of MFCs, especially SMFCs, is too low to be used directly by commercial electronic devices, and usually do not increase with the scale (Wang et al., 2012). Serial or parallel stack of MFCs can enhance the voltage or current level, but are not applicable for SMFCs which need long-term operation in sediments due to voltage reversal or short circuit (Donovan et al., 2011; Ieropoulos et al., 2013). To harvest the electricity generated from SMFCs and elevate it to an applicable level, recently developed power management systems (PMSs) are considered to be indispensable, by which the electricity from MFCs can be continuously harvested and stored into capacitors or batteries to power electronic devices

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(Alaraj et al., 2014; Park and Ren, 2012). However, little is known about the long-term applicability of a scaled-up SMFC–PMS system to date.

Bioremediation of contaminated sediments is one of the most important aims of SMFC development (Yan et al., 2012; Zhang et al., 2010). Due to the industry and urbanization processes, the sediments always absorb a diversity of toxic contaminants which could significantly affect the performance of SMFCs. The long-term bioremediation performance as well as the organic-to-electric energy efficiency of scaled-up SMFCs in heavily contaminated sediments have not been investigated.

To test whether or not the scaled-up SMFCs–PMS system could be applied as a long-term and simultaneous bioremediation tool and power provider in heavily contaminated site, a 100 L SMFC integrated with a PMS has been operated for over 2 years. The results indicated that the SMFC–PMS system can function spontaneously and durably in heavily contaminated sediments for simultaneous bioremediation and supplying applicable electricity.

## 2. Methods

### 2.1. Characteristics of the sediment inoculation

The sediments were obtained from an heavily electronic waste-contaminated river in Shunde, China (22°45' N, 113°15' E) as previously reported (Xu et al., 2014). In addition to the visually black and a strong odor of the sediments, high concentrations of toxic contaminants such as various heavy metals (Cu, Hg, Cd, Cr, Zn, As), polycyclic aromatic hydrocarbons (PAHs, 1.2 mg/kg (wet weight)) and polybrominated diphenyl ethers (PBDEs, 178 µg/kg (wet weight)) were detected. The sediments were sieved through a 0.5 cm sieve to remove coarse debris before inoculation.

### 2.2. SMFC assembly and operation

120 L plastic barrels were used as the SMFC container (Fig. S1). 50 L of the sediment was added to the barrel and covered with 50 L of the river water. Five pieces of plain graphite plate (20 × 20 cm for each) connected in parallel with rubber-coated copper wires were embedded within the sediment and used as anode. A piece of foam-bonded plain graphite felt (40 × 40 cm) was placed on the surface of the water and used as cathode. No additional electron donor was added to the SMFC. The anode and cathode were connected with a rubber-coated copper wire including a 1000 ohm resistor. Each wire connection sites in the systems was protected by silica gel from corrosion. Voltage of the resistor was recorded using a multimeter. After the voltage reached and maintained at the maximal value, the polarization curve was plotted by changing the external resistor. Control reactors (open-circuited SMFC) was tested in parallel with the closed-circuit SMFCs in natural temperature from July 2011 to August 2013.

### 2.3. Chemical analysis

The variation of pH, oxidation reduction potential (ORP) and dissolved oxygen (DO) in the sediment were determined by a multimeter (WTW, multi 3430). Anode sediments from different sites were mixed for subsequent chemical analysis. The sediment total organic carbon (TOC) was determined by Elementar Liqui TOC II analyzer. The loss on ignition (LOI) was determined as described by Hong et al. (2008). The readily oxidizable organic matter (ROOM) in the sediment was analyzed as described by Loring and Rantala (1992).

### 2.4. Description of the PMS

The PMS used in this study consisted of an energy harvesting section and a management section (Fig. S1). SMFC as the power input was directly connected to an inductor (470 Mh, L)-based energy harvesting circuit. When the metal oxide semiconductor field effect transistor (MOSFET) switched on, the electrons would flow and generated a voltage across the inductor. When the MOSFET switched off, the energy stored in inductor would be transformed to electric current and flow through a Schottky diode into a capacitor ( $C_2$ , 220 µF). Considering that most commercially electronic devices are powered by batteries, a lithium-ion battery (3.7 V, 850 mAh) from Nokia 1000 mobile phone was used as the terminate electricity storages to test the feasibility of the SMFC–PMS system.

A low power-consuming microcontroller unit (MCU, MSP430G2553, Texas Instrument) was used to monitor and control the whole PMS circuit. Once the battery voltage reached the up-threshold of 4.2 V, the MCU disabled the electricity harvest from SMFC to battery. Once the battery voltage was consumed and decreased to the low-threshold of 3.3 V, the MCU would switch to charge mode to recharge the battery. The selected frequency and duty ratio of the PMS were 1.4 kHz and 13%, respectively. The other capacitor ( $C_1$ ) was used to collect the SMFC electricity when the PMS was occupied. A constant resistor was used to test the power out-put. The PMS energy efficiency was represented by the ratio of power output and input.

## 3. Results and discussions

### 3.1. SMFC stimulated the degradation of sediment organic matters

The sediment used in this study was heavily contaminated by organic contaminants and heavy metals as reported in previous studies (Xu et al., 2014). The initial organic contents (TOC 4.5%, LOI 13.7%, ROOM 9.5%) were nearly twofold of the sediments used in other SMFC studies (Hong et al., 2008; Tender et al., 2002) due to contamination. After two-year operation, the organic contents were significantly decreased in SMFCs. In comparison with the open-circuited reactor, the sediment TOC removal efficiency in SMFC was enhanced by 4.8-fold (22.1% vs 3.8%), the LOI removal efficiency of sediment was enhanced by 1.4-fold (18.8% vs 7.8%) and the ROOM removal efficiency was enhanced by 1.5-fold (22.6% vs 8.9%) (Fig. 1), indicating that 18.3% of TOC removal, 11% of LOI removal or 13.7% of the ROOM removal could be attributed to bacterial electricity generation.

Many previous studies have demonstrated that microbial electricity generation could enhance the degradation of contaminants in lab-scale reactors or practical environments (Hong et al., 2008; Zhang et al., 2010; Yan et al., 2012). The enhanced degradation of organic matters in this study is comparable to that of a field-deployed SMFC in a noncontaminated river (Hong et al., 2008), indicating that SMFCs could also be applied as a long-term and efficient bioremediation tool in heavily contaminated sediments. Several reasons may account for the enhanced degradation rate in SMFC: (i) SMFC stimulated the microbial growth in sediments (Fig. S2); (ii) the diversity of the microbial composition was increased in SMFC compared to that in control (Fig. S3). The SMFC-specific bacteria may play important roles in the enhanced biodegradation and electricity generation in SMFC, e.g. the obligate anaerobic, sulfur compounds respiring bacteria *Caldisericum* sp. (99% identity) and some other uncultured bacteria. Moreover, organic contaminants could be absorbed by the graphite electrode and be degraded by the biofilms which have higher toxic-resistibility and biodegradation efficiency (Zhang et al., 2010).

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