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Long-term effect of biochar amendment on the biodegradation of petroleum hydrocarbons in soil microbial fuel cells

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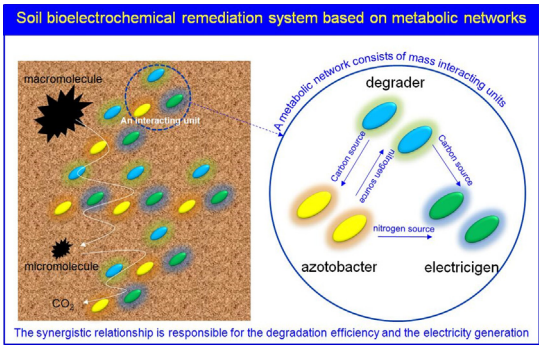
HIGHLIGHTS

- CB and SB enhanced the removal of alkane and aromatic, respectively.
- The sawdust biochar exerted the best effect on the electron transfer in soils.
- The biochar accelerated the selective enhancement of bacteria and archaea.
- The methanogenic degradation of hydrocarbons was an important process in soils.
- The degrader, azotobacter and electricigens exhibited a synergic relationship.

GRAPHICAL ABSTRACT



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ABSTRACT

Biochar is extensively applied in amendment of contaminated soils. However, the effect of biochar on the biodegradation of petroleum hydrocarbons and electricity generation in soil microbial fuel cells (MFCs) remains unclear. Here, three biochars respectively derived from poultry (chicken manure, CB), agriculture (wheat straw, SB) and forestry industries (wood sawdust, WB) were investigated after 223 days of amendment. Consequently, high removal for alkanes was in CB with the mineral nutrition and phosphorus while aromatics were in SB with the most N content and the highest molecular polarity. The lowest removal efficiency of total petroleum hydrocarbons was observed in WB with the highest surface area, whereas the most charge was obtained. The different performance of soil MFCs was due to physicochemical properties of biochar and colonized microbial communities of bacteria and archaea. The abundance of *Actinotalea* increased by 144–263% in SB and CB while that of *Desulfatitalea* distinctly increased in WB. Meanwhile, species from *Methanosarcina*, *Methanoculleus*, *Halovivax* and *Natronorubrum* exerted probably a methanogenic degrading role. This study revealed that the degrader, azotobacter and electricigens exhibited a close relationship in order to degrade hydrocarbons and generate electricity in soil bioelectrochemical remediation systems.

1. Introduction

The bioelectrochemical technology is increasingly concerned with advantages of low energy consumption and by-products, instead resource recovery (He, 2012; W. Li et al., 2015; Logan and Rabaey, 2012;

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Lovley, 2012). The bioelectrochemical removal of petroleum hydrocarbon has been approved in terms of biocurrent enhanced degradation and simultaneously bioelectricity generation (Daghio et al., 2017; Li et al., 2017; Lu et al., 2014; Venkidusamy et al., 2016; Wang et al., 2012). However, the difficult mass transfer including the substrate bioavailability and electron migration in petroleum hydrocarbon contaminated soils limits the biodegradation efficiency and charge output in soil microbial fuel cells (MFCs). The bioavailability of contaminants, especially for the aged hydrocarbons determines the degrading process and thus electricity generation in soil MFCs. Then, the solubilization effect of surfactant played an important role, and consequently the addition of ampholytic surfactant (lecithos) showed >33-fold and 328% increase of charge output and degradation of petroleum hydrocarbons in soil MFC, respectively (X. Li et al., 2018).

As for soil properties, the increase of soil moisture from 23 to 33% resulted in a 83% decrease of internal resistance and thus an improvement of soil MFC performance (Wang et al., 2012). The enlarging of soil porosity from 44.5 to 51.3% decreased by 46% of internal resistance and simultaneously enhanced the H^+ and DO diffusion, and thus increased charge output from 2.5 to 3.5 C/g soil and by 268% of petroleum hydrocarbon degradation (X. Li et al., 2015). Moreover, the internal resistance of soil MFC decreased by 58% after mixing 1% of carbon fiber into soils, leading to 15-fold increment of electricity production and 329% higher removal of petroleum hydrocarbon (Li et al., 2016c). As far as the anode is concerned, the current density of soil MFC with a biochar anode (25.6 Ω, 35.2 mA/m^2) was inferior to that with a graphite anode (11.1 Ω, 70.4 mA/m^2) for the hydrocarbon degradation in soils. Generally, the improvement of soil properties and configuration of electrode mainly reduce the ohmic polarization of soil MFCs, which is undeniably a determinant for the successful remediation of contaminated soils.

The pyrolytic biochar possesses the well electrical conductivity and uses as a sustainable electrode material in microbial fuel cells, with an advantage of the reduced cost and carbon footprint (Huggins et al., 2014). From a kinetic point of view, the interface electron transfer of pyrogenic carbon is dominated by quinone groups to carbon matrices as a result of greater graphitic structures with lower O/C and H/C ratios (Sun et al., 2017). Before microbial colonization, the access to or sensing of biochar by microbes is potentially the rate-limiting step for the electron transfer. However, after microbial colonization the electron transfer kinetics and pathways of biochar will become more important in soils, because many microorganisms favourably colonized in pores of biochar (Vanek and Lehmann, 2015). These microorganisms including degrading bacteria metabolize the organic contaminants adsorbed in/on the biochar (Denyes et al., 2016; Kong et al., 2018). Nevertheless, the enhancement of biochar for the hydrocarbon degradation and electricity generation in soil MFCs remains unknown.

The different contaminants are processed by exoelectrogenic biofilms by combining the emergence of substrate-specific microbial communities. Consequently, syntrophic interactions that exist in soil MFCs enable an exoelectrogenic biofilm to successfully convert virtually any substrate into electrical current (Logan, 2009; Lovley, 2012). This community profiles indicate a hierarchical community structure with certain microbes degrading (hydrolyzing and fermenting) complex macromolecular hydrocarbons, and others using these by-products for current production (Dombrowski et al., 2016; Li et al., 2016b; X. Li et al., 2018). Over time, the depleting of easily biodegradable fermentation end products makes the generated electricity step slower and more demotivated (X. Li et al., 2015; Li et al., 2014a; Lu et al., 2014). The differing performances in these two steps were probably due to upstream metabolic limitations associated with the degradation of the macromolecular hydrocarbons. Thus, the primary fermentation and biological interaction between the electricigens and degraders are very important in soil MFCs.

In this study, three biochar respectively derived from poultry (chicken manure), agriculture (wheat straw) and forestry industries (wood sawdust) were investigated in petroleum hydrocarbon contaminated soil MFCs after 223 days of amendment (until voltage < 20 mV, 1000 Ω). It aims to reveal: 1) the biodegradation efficiency of petroleum hydrocarbon in soil MFCs; 2) the bioelectricity generation of soil MFCs; 3) the shifts of bacterial and archaea communities after addition of biochar; 4) the preliminary biological interaction between bacteria and archaea.

2. Materials and methods

2.1. Soils preparation and biochars addition

The petroleum hydrocarbon contaminated soils were collected from the Dagang Oilfield (Tianjin, China), air-dried and passed through a 2 mm sieve. The tested soils were well-aged as the previous analysis (X. Li et al., 2018). The biochars were produced by using chicken manure, wheat straw and wood sawdust at 600 °C in a muffle (Kong et al., 2018), which were typical solid wastes in poultry, agriculture and forestry industries. The preparation and characterization of biochar were detailed in supplementary materials, and their properties were summarized in Table S1. The soil MFCs were filled with three kinds of biochars as a mass ratio of 2% and marked as CB, SB and WB respectively. The control (no biochar) was marked as CK. The dry soil and biochar of 100 g mixture were mixed homogeneously with 40 mL of distilled water and filled into each soil MFC. No exogenetic inoculation or buffer solution was added.

2.2. Soil MFCs configuration and operation

The soil MFC (6 cm × 6 cm × 6 cm) was assembled of a rolling air-cathode and carbon mesh anode. The air-cathode was consisted of an activated carbon catalyst layer, a carbon black gas diffusion layer and a steel stainless mesh current collector, and manufactured as previous methods (Li et al., 2014b; Zhang et al., 2014). The anode was one-layer carbon mesh pretreated by acetone overnight (Wang et al., 2009). Each treatment had a duplicate and placed in a 30 °C incubator. The soils in MFCs were sealed with distilled water during the experiment of 223 days. Therein, a natural place (open circuit treatment, no water seal and ambient temperature) of 5 months was set for all the soil MFCs. After remediated, a mini soil column (~1 cm of diameter) from top to bottom was collected for the biological analysis and all remaining soil samples were uniformly mixed for the physico-chemical properties and the hydrocarbon content analysis.

2.3. Electrochemical and chemical analysis

The voltage (U) of external loading of 1000 Ω (R) was recorded by a data acquisition system (PISO-813U, Shanghai, China). The accumulated charge out (Q) was calculated as $Q = \int_0^T \frac{U}{R} dt$, the cycle time ($T$) was 1800 s. The electrochemical impedance spectrum (EIS) was conducted over a 100 kHz–1 Hz of frequency range with a 10 mV of amplitude by a potentiostat (Autolab PGSTAT 302N, Metrohm, Switzerland) at the open circuit potential (stabilized for 6 h). The anode was used as the working electrode while the cathode as the counter and reference electrodes (two-electrode system). The fitting program for the Nyquist plots of EIS was ZsimpWin 3.10 as an equivalent circuit (Li et al., 2016d). The open circuit voltages of soil MFCs were gained on day of 40.

The total petroleum hydrocarbons (TPHs), alkanes (*n* alkane, *i* alkane and cycloalkanes), aromatics (monocyclic, dicyclic and polycyclic hydrocarbons), polar material and asphaltene (mixture of sulf-hydrocarbons, nitric-hydrocarbons, high-molecular hydrocarbons and nonmetallic derivatives), and 16 priority PAHs were measured according to previous procedures (Peng et al., 2009; Wang et al., 2012). The hydrocarbon contents of original soil were in Table S2. The pH and electrical conductivity (EC) of soils were determined in a mixture of soil: