



Effect of static magnetic field on trichloroethylene removal in a biotrickling filter



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HIGHLIGHTS

- The MF-BTF was first applied in the treatment of waste gas.
- Higher ECs and REs were achieved in the MF-BTF under the low MF intensity.
- The static magnetic field improved the performance of the BTF.
- The relative abundance of predominant genus *Acinetobacter* was higher in MF-BTF.

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ABSTRACT

A laboratory-scale biotrickling filter combined with a magnetic field (MF-BTF) and a single BTF (S-BTF) were set up to treat trichloroethylene (TCE) gas. The influences of phenol alone and NaAc-phenol as co-substrates and different MF intensities were investigated. At low MF intensity, MF-BTF displayed better performance with 0.20 g/L of phenol, 53.6–337.1 mg/m³ of TCE, and empty bed residence times of 202.5 s. The performances followed the order MF-BTF (60.0 mT) > MF-BTF (30.0 mT) > S-BTF (0 mT) > MF-BTF (130.0 mT), and the removal efficiencies (REs) and maximum elimination capacities (ECs) corresponded to: 92.2%–45.5%, 2656.8 mg/m³h; 89.8%–37.2%, 2169.1 mg/m³h; 89.8%–29.8%, 1967.7 mg/m³h; 76.0%–20.8%, 1697.1 mg/m³h, respectively. High-throughput sequencing indicated that the bacterial diversity was lower, whereas the relative abundances of *Acinetobacter*, *Chryseobacterium*, and *Acidovorax* were higher in MF-BTF. Results confirmed that a proper MF could improve TCE removal performance in BTF.

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1. Introduction

Recently, several severe haze events occurred concurrently worldwide (Li and Han, 2016), which were attributed to volatile organic compounds (VOCs). VOCs play a significant role in the formation of secondary organic aerosols and ground-level ozone (Ran et al., 2011; Zhang et al., 2014) and significantly affect the chemistry of the atmosphere and human health (Dumanoglu et al., 2014). Trichloroethylene (TCE) is a persistent VOC contaminant that infiltrates soil and ground water and can contribute to long-term contamination (Popat et al., 2012). It has been extensively used as a solvent such as degreasing of metals, applications in the textile industry and electronics industries, etc. (Ohlen et al., 2005). TCE is suspected of being carcinogenic and mutagenic as a

chlorinated aliphatic hydrocarbon (Yu and Semprini, 2004). The International Agency for Research on Cancer has reassessed the carcinogenicities of TCE and its metabolites in humans and mammals, and classified them as level I carcinogens (Neela et al., 2012). Thus, TCE was listed as one of 129 kinds of priority control pollutants by the U.S. Environmental Protection Agency, and the specified concentration limit in drinking water is no more than 5 µg/L according to the World Health Organization (Chiu et al., 2006), thereby making the efficient removal of TCE highly essential.

Several conventional treatments of waste gas, including physical and chemical methods, have been developed and used by researchers to remediate TCE in the environment, but these techniques are usually expensive and generate secondary pollutants that require further treatment (Tabernacka et al., 2014). Recently, biological techniques for VOCs removal have been shown to be cost effective and environmentally friendly, among which bioscrubbers,

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biofilters, and biotrickling filters (BTF) have been used or developed (Bhatt et al., 2007; Balasubramanian et al., 2012). Compared with other biological techniques, the BTF has a distinct advantage in terms of lower operating and capital costs, low pressure drop, and the capability to treat acid degradation products of VOCs (Lebrero et al., 2012). But BTF could not achieve satisfactory performance for some recalcitrant VOCs because of slow microbial adaptation and growth (Hu et al., 2016). So it is desirable to develop an efficient BTF to treat TCE waste gas.

A recent advance is the combination of single technologies that overcome the limitations of individual approaches and remove more VOCs (Yu et al., 2014). The use of a magnetic field (MF) has become a focus for environmental microbe enhancement, for the advantages of lower operational cost as well as ecofriendliness and convenience. Especially in the treatment of wastewater, MF strengthening can result in positive effects (Niu et al., 2014; Ji et al., 2010). Through periodical exposure to an MF induction of 40 mT using permanent magnets, the transformation of nitrogen compounds was more effective and the oxygen uptake rate of second-phase nitrifiers was intensified (Tomska and Wolny, 2008). A 7 mT MF increased formaldehyde removal by 20% with a formaldehyde concentration of 1600 mg/L and also had a positive effect on the efficiency of chemical oxygen demand removal, as well as bacterial and activated sludge biomass growth (Lebkowska et al., 2013). The MF had an impact on phenol biodegradation, *Rhodococcus erythropolis* growth, and respiration activity in a fed-batch reactor (Křiklavová et al., 2014). Enhancement of the MF intensity from 20 mT to 40 mT caused microorganisms to produce more unsaturated fatty acids to stimulate TTC dehydrogenase activity and adaptation to low temperature (Niu et al., 2014).

Although the application of an MF can improve the treatment efficiency of wastewater by activated sludge, a performance evaluation of a BTF treating VOCs under a static MF has not been reported. Besides, selection of the carbon source is a key factor for a BTF treating TCE, as TCE cannot be used as the carbon source or energy source for microbial growth under aerobic conditions, and can only be utilized based on co-metabolism. Methane, phenol, toluene, propane, ortho-xylene, and ammonia are generally used as growth substrates for co-metabolic TCE degradation. Of those, toluene and phenol had the better performance. In this study, phenol was chosen as the co-substrate because of its ability to cultivate a robust biomass with considerable TCE catalytic activity (Vannelli et al., 1990), and also because of its greater water solubility and lower toxicity compared to toluene. What is more, 50% of gram-negative bacteria were reported to use phenol as a co-substrate for TCE co-metabolic degradation, and the results were better than for toluene (Suttinun et al., 2013). Thus, the objective of this study was to provide the proof of concept for the effect of static MF on TCE removal in a BTF. A laboratory-scale BTF combined with an MF (MF-BTF) and a single BTF (S-BTF) system were set up in parallel. The aim was to evaluate their performances in terms of removal efficiency (RE) and elimination capacity (EC) in treating TCE. In addition, this study aimed to assess the changes in the bacterial community through a high-throughput sequencing of the contents of the S-BTF and MF-BTF. This study is expected to provide a useful reference for the treatment of hydrophobic and recalcitrant VOCs industrially with MF.

2. Materials and methods

2.1. Biotrickling filter setup

The study was carried out in two lab-scale BTFs (S-BTF and MF-BTF), each consisting of a cylindrical jacketed PMMA column with a height of 1000 mm, inner diameter of 110 mm, and a working vol-

ume of 2.7 L (Fig. 1). The S-BTF was the control experiment without an MF and the MF-BTF experiment was set up in the heteropolar MF; the MF consisted of two magnet blocks connected by a keeper placed on the outer sides of the poles and joined by the expanders of the MF; the MF intensity was regulated by moving the magnetic keeper, and measured by using a Tesla-Meter HT20 (Xiangtong Magnetolectricity Technology Co., Shanghai, China). Magnet blocks were composed of NdFeB and were of 600 mm length and 100 mm width. The two BTFs were packed with lava rocks of 8–10 mm particle diameter, which were obtained from the Changbai Mountains located in Jilin province, China.

Acclimated activated sludge was poured into the S-BTF and MF-BTF and discharged after 24 h. TCE-containing waste gas was generated from two air streams using an aquarium air pump (Sebo, Zhongshan, China). One air stream was bubbled through liquid TCE in a 500 mL gas scrubber bottle. The other air stream was humidified to the desired relative humidity, and then the two streams were mixed in a buffer bottle of 4 L. Finally, the two BTFs were continuously fed from the bottom of the two BTFs. The TCE inlet concentration and the flow rate of the mixed air stream were controlled by mass flow meters; the empty bed residence time (EBRT) was set at 202.5 s, corresponding to a gas flow of 0.8 L/min. Mineral salt medium stored in an 8 L nutrient tank was periodically trickled from the top of each BTF for 15 min at 8 h intervals by pumping at a rate of 3 L/h. The details of operational parameters are listed in Table 1; the S-BTF and MF-BTF were operated identically except for the presence of the magnet blocks.

2.2. Microorganisms

The aerobic activated sludge was obtained from an aeration basin of a wastewater treatment plant (Yanji, China). The activated sludge was acclimated with TCE and phenol for 60 days in a tank. The composition of the mineral salt medium was 0.60 g/L NaAc, 0.46 g/L K_2HPO_3 , 0.38 g/L KH_2PO_3 , 0.48 g/L $(NH_4)_2SO_4$, 0.60 g/L $NaHCO_3$, 0.40 g/L NaCl, 0.1 g trace elements solution, pH 7.2. The composition of the trace elements solution was (g/L): $CuSO_4 \cdot 5H_2O$, 0.01; KI, 0.01; $CaCl_2 \cdot 2H_2O$, 0.132; $Na_2MoO_4 \cdot 2H_2O$, 0.02; $KAl(SO_4)_2 \cdot 12H_2O$, 0.02; $MnSO_4 \cdot H_2O$, 0.045; $CoCl_2 \cdot 6H_2O$, 0.05; $FeCl_3$, 0.12; H_3BO_3 , 0.05; and $ZnSO_4 \cdot 7H_2O$, 0.075.

2.3. Analytical methods

TCE concentration was measured using a GC-2010 gas chromatograph (SHIMAZDU, Tokyo, Japan) equipped with an Rtx-1701 capillary column (30 m × 0.25 mm × 0.25 μm) and a flame ionization detector. The following conditions were maintained throughout the analysis: the temperatures of the injector and detector were 220 °C, the oven temperature was 80 °C, the flow rate of N_2 as the carrier gas was 136.3 mL/min, and the column flow rate was 1.32 mL/min. Injections were made in the split mode with a split of 100; a 1000 μL gas injector was used for injecting the gas sample into the gas chromatography.

2.4. Bacterial community analysis

2.4.1. Genomic DNA extraction

To analyze the bacterial community of the BTF biofilms, the mixed biofilms were collected from the bottom port at four different positions of the same height on 121 days. For comparison, the initial activated sludge (IAS) that was taken from an aeration basin of a wastewater treatment plant (Yanji, China) was also examined. Total genomic DNA was extracted using a PowerSoil® DNA isolation Kit (MoBio Laboratories, Carlsbad, CA, USA) according to the manufacturer's instructions. DNA quality was assessed using 1%-TAE (tris-acetate-EDTA) agarose gel electrophoresis stained with

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