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Removal of methyl acrylate by ceramic-packed biotrickling filter and their response to bacterial community

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HIGHLIGHTS

- The treatment of methyl acrylate waste gas in BTF was first studied.
- Good removal performance was achieved during variation of operating conditions.
- DGGE was used to analyze the dominant strains in different layer.
- The function of each identified strains was briefly introduced.
- Bacterial community structure coupled to treatment efficiency.

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ABSTRACT

Methyl acrylate is a widely used raw chemical materials and it is toxic in humans. In order to treat the methyl acrylate waste gas, a 3-layer BTF packed with ceramic particles and immobilized with activated sludge was set up. The BTF exhibited excellent removal efficiency that no methyl acrylate could be detected when EBRT was larger than 266 s and inlet concentration was lower than 0.19 g/m^3. The 1st layer performed the best at fixed inlet concentration of 0.42 g/m^3. PCR combined with DGGE was performed to detect the differences in different layers of the BTF. Phylum *Proteobacteria* (e.g. α-, β-, γ-, δ-) was predominantly represented in the bacterial community, followed by *Actinobacteria* and *Firmicutes*. *Desulfovibrio gigas*, *Variovorax paradoxus*, *Dokdonella koreensis*, *Pseudoxanthomonas suwonensis*, *Azorhizobium caulinodans*, *Hyphomicrobium denitrificans*, *Hyphomicrobium* sp. and *Comamonas testosteroni* formed the bacteria community to treat methyl acrylate waste gas in the BTF.

1. Introduction

Acrylic ester such as methyl acrylate has many important commercial uses. Every year, more than 2.3 million metric ton of acrylic ester is consumed. Methyl acrylate is principal raw material in the manufacture of polymeric products. It is used to prepare thermoplastic coatings and synthesize higher acrylic esters. The polymers made of methyl acrylate are characterized by colorless transparency, easy adhesion, elasticity, and stability to light, moderate heat as well as weathering. And they are widely applied in surface coatings, textiles, adhesives, paper treatment, polishes, leather, fibers, detergents, and super-absorbent materials (Hunt, 1993). The application of methyl acrylate makes human life more convenient also brings us pollutions.

Methyl acrylate has immunotoxicant, neurotoxicant, respiratory toxicant, skin or sense organ toxicant, etc. Some acrylic acid and acrylic esters may enter wastewater treatment systems and surface waters. Additionally, some releases of acrylic esters to the environment may occur during their use and disposal. Moreover, as a kind of VOCs (volatile organic compounds), methyl acrylate is not only harmful to human health but also cause severe environmental problems (Patnaik, 1992). The vapor of methyl acrylate is a colorless air pollutant with a repellent odor. Odorous waste gases are a special kind of air pollutants. Humans can perceive even extremely small amounts of an odorant. It is estimated that only 10^8 or 10^9 molecules of odorant vapor in the nose is enough to trigger detection (Rappert and Muller, 2005). Some researchers found that persistence in air polluted area may lead several kinds of diseases (Brunekreef, 2002; Gryparis et al., 2004). Outdoor air pollution, mostly by PM 2.5, leads to 3.3 (95% confidence interval 1.61–4.81) million premature deaths per year worldwide, predominantly in Asia

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(Jerrett, 2015). So the treatment of VOCs waste gas like methyl acrylate is rather important.

Various technologies have been developed to reduce/eliminate odorous gases for improving the quality of air. Three sort methods, biological technologies include biotrickling filter (BTF), biofilter, bioscrubber, activated sludge, etc., chemical technologies included (chemical scrubbers, thermal oxidation, catalytic oxidation, ozonation, etc.), and physical technologies (condensation, adsorption with activated carbon or clean water scrubbers, etc.) have been often used for this purpose (Burgess et al., 2001). In most cases, some conventional physical technologies are often unsatisfactory due to organic pollutants only being transferred from gaseous to other phases, and still not being fully destroyed; while chemical technologies are always expensive (Smet et al., 2010). However, biological process has been found to be a very promising technology for the removal of odorous or toxic volatile organic compound waste gas because of low operating costs, low energy requirements as well as no by-products produced for further treatment or disposal (Delhomenie and Heitz, 2005). In addition, BTF has an advantage over other biological treatment technologies in terms of mineralized efficiency, especially for high concentration acidifying pollutants containing waste gas streams, such as, sulfur, chlorine or nitrogen containing compounds (Sercu et al., 2005). BTF, also a typical technologies of biological treatment has succeed in treating many kinds of VOCs (Balasubramanian et al., 2012; Zhang et al., 2015). However, the treatment of methyl acrylate waste gas using BTF has not been reported. Therefore, it's very necessary to set up a lab-scale BTF to determine the removal efficiency (RE) and elimination capacity (EC) during the treatment of methyl acrylate waste gas.

However, in a biological system, the gas-phase contaminant generally has to be transferred from the gas to the liquid phase and subsequently to the biofilm where it is biologically degraded to harmless compounds (Gabriel and Deshusses, 2003). The analysis of the component of biofilm is important by doing that we can understand which microbes play a major role during the procession of the degradation of methyl acrylate. In recent years, molecular techniques have been increasingly applied to investigate microbial community composition in various ecosystems, including in biofilter and BTF. PCR-based fingerprinting methods brought a remarkable advancement in microbial community analysis, since traditional laboratory cultivation approaches capture only about 1% of the population diversity (Torsvik et al., 1996). Cloning and sequencing of 16S rRNA is the most powerful technique for analyzing microbial diversity in natural samples. With genetic fingerprinting techniques such as denaturing gradient gel electrophoresis (DGGE), multiple samples can be analyzed simultaneously, as is required for studying the complex dynamics of microbial communities (Muyzer, 1999).

In this study, a three-layer lab-scale aerobic BTF packed with common ceramic particles was set up and be inoculated with activated sludge to deal with the methyl acrylate waste gas. We aimed to study the removal performance by changing the inlet concentrate or empty bad retention time (EBRT) during the procession of degradation. PCR-DGGE was used to gain an insight into the dynamic diversity of the bacterial community in different layer of the BTF. Then the roles of predominant strains in the process of methyl acrylate removal were analyzed. The predominant strains may be used as a single strain to treat methyl acrylate or some other VOCs vapors in our future research.

2. Methods

2.1. Experimental setup

The BTF was made of a clear polymethyl methacrylate column with an inside diameter of 120 mm and a height of 1100 mm (Fig. 1). The column had three layers. From the bottom to the top, each layer's packing height was 220, 220 and 150 mm, respectively. Each layer was packed with commercially available ceramic particles made by SiO_2. Below each layer, there was a perforated steel plate to prevent the column packed from dropping off, while at the same time ensuring that the gas and water could pass through easily. In the middle of each layer, there was a sample port, via these sample ports the ceramic particles were taken out and the biofilm was analyzed. To investigate the methyl acrylate removal along the depth of the packing bed, the polymethyl methacrylate column was designed with three intermediate gas-sampling ports 50 mm over each layer. The BTF was operated in an up-flow, countercurrent mode, and fed with an aerobic mixed gas containing methyl acrylate (Guangfu Fine Chemical, Tianjin, China). The mixed gas was an air stream contaminated with methyl acrylate and produced with two gas flows which were previously humidified and supplied from an air compressor. Each flow rate was controlled by a flow meter. One gas flow first flowed through a 500 mL gas scrubber bottle that contained 250 mL liquid methyl acrylate. By changing the flow rate and the temperature of the water bath instrument, different concentrations of methyl acrylate can be produced then flowed into a mixing bottle. The other flow was directly introduced into the mixing bottle to be blended with the methyl acrylate flow, thus allowing the BTF to regulate the inlet concentration (varies from 120.2 to 7505.6 mg/m^3 in this study) of methyl acrylate without changing the EBRT of the mixed gas, as well as regulating the EBRT (varies from 200 to 400 s) without changing the inlet concentration. Trickling liquid provided a convenient means to control pH, salt, or metabolite concentration and supplemented nutrients to the biomass (Xue et al., 2013). The additional nutrients, pH control, and larger gas/liquid interfacial resulted in substantially higher removal efficiencies than that of biofilter system. The inflow medium stored in an 8 L nutrient tank was intermittently recirculated by a peristaltic pump and sprayed from the top of the BTF at 1 L/min for 10 min at 6 h intervals.

2.2. Microbial culture and inoculation

The activated sludge was collected from an aeration basin of a sewage treatment plant in Yanji, Jilin province, China. The activated sludge was poured into a 10 L tank and allowed to stand for 2 h. The supernatant was drawn off and then mixed with 8 L inflow media. After 22 h of aeration then stand for 2 h, fresh inflow media was used to replace the supernatant. After 7 days of culture, the activated sludge solution was poured into the BTF. After 24 h of soaking, the packing media has already accumulated some biofilms, and then the activated sludge was exhausted. Low concentration of methyl acrylate vapor was introduced into the BTF at the same time. The inflow medium (contains 0.6 g/L glucose, 1.0 g/L K_2HPO_3, 1.0 g/L KH_2PO_3, 0.4 g/L $(NH_4)_2SO_4$, 0.6 g/L $NaHCO_3$, 0.4 g/L NaCl, 0.1 g trace elements solution, pH 7.2) was intermittently recirculated by a peristaltic pump and sprayed from the top of the BTF at 1 L/min for 10 min at 6 h intervals. Every 2 days, the content of glucose was decreased 0.1 mg/L. Every day the inflow medium was sprayed into the BTF, and then stabilizing for 2–3 h. The inlet and outlet gas was then injected into GC (gas chromatography) to detect the concentration of methyl acrylate until the RE reached 90%.

2.3. Analytical methods

Methyl acrylate concentration was measured by a GC-2010 gas chromatograph (SHIMAZDU, Tokyo, Japan) fitted with an Rtx-1701 column and equipped with a flame ionization detector. A 1 mL gas injector was used for injecting gas sample into the GC system. N_2 was used as carrier gas. The following conditions were maintained