Chemosphere 162 (2016) 139–147

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Enhanced stable long-term operation of biotrickling filters treating VOCs by low-dose ozonation and its affecting mechanism on biofilm

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HIGHLIGHTS

- Ozone was effective in preventing excess accumulation of biomass.
- Low-dose ozone contributed to the stable operation of biotrickling filters (BTFs).
- Control of ozone dose was a key strategy for achieving enhancing performance of BTFs.
- Ozone reduced the contents of extracellular polymeric substances within biofilm.
- Microbial community adapting to ozonation contributed to stable operation of BTFs.

ARTICLE INFO

Article history:
Received 1 February 2016
Received in revised form
13 July 2016
Accepted 23 July 2016

Handling Editor: Y Liu

Keywords:
Biotrickling filter
Ozone
VOCs
Biomass accumulation
EPS
Microbial community

ABSTRACT

For long-term operation of highly loaded biotrickling filters (BTFs), the prevention of excess biomass accumulation was essential for avoiding BTF failure. In this study, we proposed low-dose ozonation as a biomass control strategy to maintain high removal efficiencies of volatile organic compounds (VOCs) over extended operation of BTFs. To obtain an optimized biomass control strategy, the relative performance of five parallel BTFs receiving different ozone doses was determined, and the affecting mechanism of ozonation on biofilm was elucidated. Experimental results showed that the decline in ozone-free BTF performance began from day 150, which was correlated with excess biomass accumulation, abundant excretion of extracellular polymeric substances (EPS) and a decline in metabolic activity of biofilm over extended operation. Ozone of 5–10 mg m^{-3} was effective in preventing excessive growth and uneven distribution of biomass, and eventually maintaining long-term stable operations. Ozone of over 20 mg m^{-3} possibly inhibited microorganism growth severely, thereby deteriorating the elimination performance instead. Comparison of the biofilm EPS indicated that the presence of ozone reduce EPS contents to different extents, which was possibly beneficial for mass transfer and metabolic activity. Comparative community analysis showed that ozonation resulted in different microbial communities in the BTFs. *Dyella* was found to be the most abundant bacterial genera in all BTFs regardless of ozonation, indicating strong resistance to ozonation. *Chryseobacterium* and *Burkholderia* members were markedly enriched in the ozone-added biofilm, implying good adaptation to ozone presence. These findings provided an improved understanding of low-dose ozonation in maintaining a stable long-term operation of BTF.

1. Introduction

At present, biotrickling filter (BTF) reactors are an efficient method to treat air contaminated with VOCs (Munoz et al., 2012). Biofiltration is attractive because of its low operation and maintenance costs, high removal efficiency, and environmentally benign end-products of water and carbon dioxide (Rene et al., 2012). The growing emphasis on sustainability will even more stimulate the application of biological processes for gas treatment. Although BTFs present many advantages, there are several limitations for existing biofiltration technologies. Included among them in long-term operation are the excessive biomass growth and uneven

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http://dx.doi.org/10.1016/j.chemosphere.2016.07.072
0045-6535/

distribution. Biomass is a critical factor in gas biofiltration, and excess accumulation and uneven distribution often result in operating problems such as clogging, increase in airflow resistance, formation of gas channeling within BTF beds and subsequent declining of VOC removal efficiencies (Kan and Deshusses, 2008; Dorado et al., 2012; Garcia-Perez et al., 2013). The stability of BTF performance for long-term operation cannot be assured. Thus, coordinated biomass control is deemed necessary for achieving long-term performance of the BTFs.

A few previous studies discuss the coupling of UV photolysis and biofiltration (Moussavi and Mohseni, 2007; Wang et al., 2009b; Cheng et al., 2011), which shows that the produced ozone by 185 nm UV irradiation prevents excessive biomass growth and enhances the VOC removal. Garcia-Perez et al. (2013) utilize pulses of ozone as a complementary chemical oxidant at low concentrations to overcome excessive biomass growth and biological reaction limitations. A further study also showed that low-dose ozone could notably enhance the performance of biofilter treating toluene (Xi et al., 2014). All of the previous studies demonstrate that low-dose ozonation opens new possibilities for biomass control, thereby maintaining extended and stable operation of highly efficient BTFs. However, the previous studies mainly focus on the enhanced performance of BTFs by ozonation, while the role mechanisms of low-dose ozonation on microorganisms in BTF are still not clear to date. It is of great significance to further delve into the essence of improved biofiltration performance under a long-term operation by ozonation, which will undoubtedly provide directions and inspirations for future researches.

It is generally accepted that extracellular polymeric substances (EPS) plays a significant role in biofilm formation and maintenance of the structure integrity (Li and Yu, 2011), and is the most significant factor affecting biofilm properties (Ying et al., 2010). Aggregate of EPS forms a gel-like biofilm matrix that account for roughly 50–90% of the total organic matter in biofilms (Barry et al., 2014). In biofilm processes, EPS act as a fixation matrix, favoring the adhesion of biomass to surfaces and carrier materials (Bassin et al., 2012). This will facilitate the initial attachment of bacteria by adsorption on the surface of the carrier material and the subsequent growth of microcolonies, which will cover the carrier surface area available for biofilm deposition. Thus, the biofilm formation is accompanied with the EPS increase with the operation. The biofilm also possesses an increased hydrophobicity, which is associated with the elevated levels of EPS, especially the increased protein contents with the operation (Ying et al., 2010). A few previous studies have investigated the mechanism of membrane filterability amelioration by ozonation (Liang et al., 2010; Barry et al., 2014; Siembida-Losch et al., 2015). The appropriate ozonation will increase the sludge hydrophobicity and introduce more acidic functional groups to soluble microbial products (SMP) and EPS. Consequently, the increased sludge particle size and decreased SMP concentration jointly contribute to the amelioration of membrane filterability (Zhang et al., 2010). However, the above mechanism analyses on sludge or biofilm ozonation are mainly relevant to wastewater and sludge treatment processes. The affecting mechanisms of ozonation on the microorganisms in gas-solid phase BTF systems are still not well understood.

In this study, we focused on three aspects that had not been thoroughly considered in the previous studies. The feasibility of injecting low-concentration ozone for preventing biomass accumulation was systematically investigated in 5 parallel toluene-degrading BTFs to maintain extended and stable operation of BTFs. The impacts of ozonation on biofilm surface characteristics including EPS content and component and biofilm hydrophobicity were examined during the whole operation. The changes in microbial community composition and metabolic activities were analyzed via high-throughput pyrosequencing and BIOLOG plate, respectively. Based on the analyses, a more reliable ozonation approach for control of excess biomass was proposed and the rationale behind the BTF performance improvement and biomass control by ozonation was elucidated.

2. Materials and methods

2.1. Biotrickling filter setup

A schematic diagram of the experimental setup is shown in Fig. S1. The BTF was constructed from a Perspex pipe with a 0.95 m high and an internal diameter of 9 cm. The active packed bed height was 48 cm, and thus the bed volume was 3.1 L. A liquid sprinkler was located at the top interior of the reactor. Liquid in a peristaltic pump moved from the reservoir to the sprinkler nozzle. Then, the liquid stored in the sprinkler trickled downward via gravitational force. The gas inlet and outlet ports were located at the bottom and the top of the media bed, respectively. Five gas sampling ports were evenly distributed axially along the medium bed to allow for periodic gas sampling at intermediate locations. Additional four ports separated by a distance of 25 cm were used to recover bed particles for microbial analyses. The BTFs were packed with pelletized polyurethane foam (PUF) with a diameter of 10–15 mm. The palletized PUF had an initial porosity of 91.0%, and a specific surface area of 380 $m^2\ m^{-3}$. The bioreactors were located in the reactor room where the temperature was held at 25–30 °C.

2.2. Inoculation of the BTF and nutrient medium composition

Aerobic sludge from a local municipal wastewater treatment plant was acclimated on toluene as a sole carbon source for about 3 weeks. The toluene-degrading microbial consortium obtained above was mixed with 2 L nutrient medium to obtain a uniform suspension of the initial inoculum. The initial biomass concentration in the recycling nutrient solution was 1.2 g of suspended solids (SS) L^{-1}. The inoculation was performed by recirculating the inoculum for the next 4 days, until visible biomass remained attached to the PUF packing carrier. The nutrient solutions for BTFs consisted of the following compounds (mg L^{-1}): K_2HPO_4 (0.110), KH_2PO_4 (0.400), NH_4Cl (0.545), $MgSO_4$ (0.067), $CaCl_2$ (0.036), $FeCl_3$ (0.250), $MnSO_4$ (0.034), $ZnSO_4$ (0.043), and $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ (0.035).

2.3. BTF operation and biofilm sampling

Our experimental studies employed 5 identical BTF reactors that were operated with five different operating strategies in terms of ozone dose. Four of the BTF reactors were operated with different ozone additions, and the remaining one without ozone addition was the control BTF reactor. This ozone stream was provided by passing dry air through a GF-YG10 (Beijing Shanmeishuimei Environmental Protection Technology Co., Ltd., China) ozone generator. Ozone gas concentrations in the range of 5–30 mg m^{-3} were fed to the BTFs from the bottom of the reactors. The BTF reactors were operated in a counter-current mode with the gas flowing upward and recycled mineral medium flowing downward. As illustrated in Fig. S1, the toluene-polluted air was produced from another air stream and it was fed separately from the ozone. Compressed air was split into one main stream and one side stream. The side stream air was bubbled through liquid toluene in a flask to generate toluene-contaminated air stream. Flow rates were adjusted using mass flow controllers (Aalborg, USA). The two streams were then mixed in a mixing unit and fed from the bottom of the reactors. The total gas flow rate was varied to achieve different empty bed residence times (EBRTs) in the reactors. The liquid mineral medium in