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Effect of the continuous addition of ozone on biomass clogging control in a biofilter treating ethyl acetate vapors

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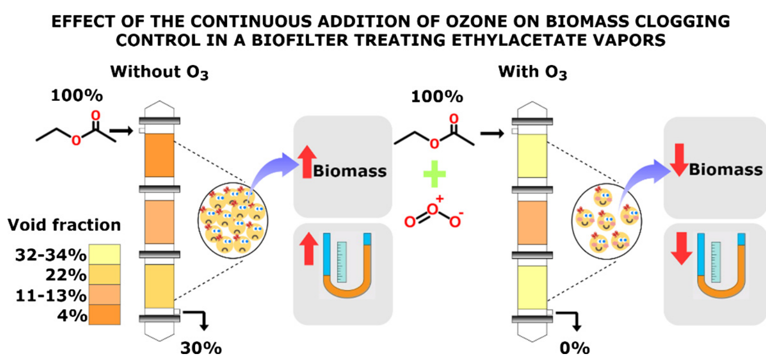
HIGHLIGHTS

- Ozone addition had a positive effect over biomass clogging control.
- Ozone addition improves the performance and extends the life time of the biofilter.
- Mineralization of ethyl acetate was higher with ozone addition than without.
- Eventually the biofilter will clog even if ozone is added.

GRAPHICAL ABSTRACT



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ABSTRACT

Biofiltration systems have been recognized as a cost-effective and environmentally friendly control technique for volatile organic compounds (VOC) removal. However, the long-term operation of biofilters causes biomass accumulation, and thus the occurrence of bed clogging, leading to a major decrease in biofilter performance. Control methods have been carried out in order to solve clogging problems, including backwashing, bed stirring, modification of flow patterns, predation, starvation and others. Ozone (O_3) has been used in biofiltration systems at low concentrations to control the excess of biomass. It is worth mentioning that all these biofiltration studies involving O_3 treated recalcitrant pollutants such as chlorobenzene, formaldehyde and toluene, which do not produce enough biomass to effectively prove clogging prevention. Thus, this study evaluated the effect of the continuous addition of O_3 as a chemical oxidant at a very low concentration (90 ppb_v) as a practical solution to overcoming clogging in a process of biofiltration of ethyl acetate (EA), a readily degradable molecule. The maximum elimination capacities achieved ranged from 200 to 120 g m^{-3} h^{-1}, with and without O_3, respectively. The biomass concentrations in these systems ranged from 23.3–180.1 to 43.31–288.46 $mg_{biomass}$ $g_{perlite}^{-1}$ with and without O_3 addition, respectively. Based on the results, it was concluded that the continuous addition of O_3 could be an attractive solution to improving biofilter performance and extending the lifetime of the filter bed.

1. Introduction

Volatile organic compounds (VOCs) are the most common air pollutants emitted from chemical, petrochemical, and allied industries. VOCs are the main precursors of photochemical reaction in the atmosphere

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leading to various environmental hazards. Two main groups of technologies have been developed in order to treat these emissions, the biological and physico-chemical processes. Even if physico-chemical processes are highly efficient, no technology can be considered fully satisfactory due to their different drawbacks. The Process economics are important to consider for example, for physico-chemical processes such as adsorption, the main issue is the cost and lifespan of the activated carbon and in the case of chemical scrubbers it is the constant chemical consumption (Estrada et al., 2012). Other drawbacks are related to high energy consumption and the use of hazardous and toxic chemical substances (Estrada et al., 2011). Also, it is worth mentioning that some of these methods are not destructive, as they only transfer the pollutant from one phase to another. The main environmental impact in biological treatments is caused by the high level of water consumption to maintain biological activity (although the use of secondary effluent water can reduce this issue and its operation cost) (Estrada et al., 2011). From a process economics viewpoint, bioscrubbing operating costs are one-half to one-fourth of the operating costs of a chemical scrubber (Hansen and Rindel, 2000) and biofiltration and chemical scrubbing presented similar operating cost: 0.21 and 0.27 € $(1000\ m^3_{treated})^{-1}$ ($0.29 and 0.38 $(1000\ m^3_{treated})^{-1}$), respectively (Estrada et al., 2011), and investment costs in a biofilter are lower compared to a chemical scrubber (225,000 and 90,000 € (Estrada et al., 2012). Biofiltration is a cost-effective and eco-friendly technology that can treat VOCs and inorganic gases with the advantages of generating only innocuous by-products. Microorganisms are used to metabolize organic compounds at ambient temperature and pressure, which entail a smaller carbon fingerprint due to less energy being required. Biofiltration presents similar energy consumption to that of biotrickling filters and chemical scrubbers, with the lowest being activated carbon (Estrada et al., 2012). The most common types of biological treatment units are, biofilters, biotrickling filters, membrane bioreactors and bioscrubbers. A major problem in biofilter systems is their instability due to rapid biomass accumulation over long-term operation or when high inlet loads are applied. If no action is taken, the biofilter will clog. "Clogging" is a typical major problem which can lead to issues such as high pressure drop, channeling and dead zones, non-homogeneous microbial growth and uneven biodegradation activity along the filter bed, which could lead to a negative effect on the contaminant removal efficiency (Alonso et al., 1997). Biomass growth in biofilters packed with inert or organic carriers needs to be controlled and optimized. Different physical, chemical and biological strategies have been evaluated for biomass control in biofiltration systems. Among them, splitting the feed and different speed ratios (Mendoza et al., 2003), bed stirring and nutrient control (Delhoménie et al., 2003), filling and draining with water and air sparging (Mendoza et al., 2004), step feed biofiltration (Estrada et al., 2013), starvation periods (Dorado et al., 2012), protozoa predation (Cox and Deshusses, 1999), mite predation (Woertz et al., 2002), backwashing with different chemicals and water (Smith et al., 1996; Sorial et al., 1997). Within the efforts of solving the clogging issue, O_3 treatment has also been proposed (Wang et al., 2009; García-Pérez et al., 2013; Maldonado-Diaz and Arriaga, 2014; Xi et al., 2014). Contrary to previous studies, the present study tested constant and continuous ozone injection during long term operation (230 days), starting from the first days of operation, in order to observe the effect of ozone on biomass formation and not only on clogging prevention. Moreover, even though O_3 seems to be a promising solution to controlling the excess of biomass in the treatment of recalcitrant VOCs, it is important to analyze its effectiveness for a readily degradable molecule in order to produce enough biomass and effectively prove clogging prevention. Ethyl Acetate (EA) is an irritant and explosive compound with fragrant odour that is used widely in printing, coating, paint manufacturing, pharmaceutical applications, artificial fruit essences and leather, and textile cleaning (Budavari, 1996). Above all, the aim of this study is to prove O_3 effectiveness as a biomass control method in a biofiltration system treating a highly biodegradable pollutant, such as EA.

2. Methodology

2.1. Experimental setup of biofilters

In brief, two biofiltration systems of 0.097 m diameter were assembled, each comprised of three identical modules 0.45 m high and with a volume of 1.1 L (total effective volume of 3.3 L) named M1, M2, and M3, in which M1 corresponds to the entrance of the saturated EA stream (Fig. 1). One biofilter was operated without O_3 addition and served as a control and the other worked under O_3 addition. The packed material used was 3.35 mm expanded Perlite from Termolita México. Reactors were inoculated with activated sludge from a waste water treatment plant in Tangamanga Park I in San Luis Potosi, Mexico. The mineral medium was prepared with 0.5 g L^{-1} $(NH_4)_2SO_4$, 0.7 g L^{-1} KH_2PO_4, 0.7 g L^{-1} K_2HPO_4, 0.3 $MgSO_4 \cdot 7H_2O$ and trace elements, and its pH was adjusted to 7 with 1N-NaOH solution. EA gas phase was generated in a stripping reactor and its flux was controlled with a needle valve. Biofilters were fed in downward mode with an empty bed retention time (EBRT) of 60 s. O_3 was produced by a A2ZS-3GLAB OZONE GENERATOR system and its concentration (90 ppb_v) was monitored with the yodimetric method (Rakness, 1996). Pressure drop was checked daily by height difference with a U-tube filled with water. EA and CO_2 concentrations were measured daily along the biofilter modules, pH and total organic carbon (TOC) contents in leachates were measured along the operation.

2.2. Stages of operation

The EA treatment experiments were carried out over a 230 days' period in two phases (Table 1). The first phase was defined with the objective of achieving the maximum inlet load that the systems were able to work with while maintaining a satisfactory removal efficiency. Accordingly, three stages were obtained with three different inlet loads: A = 60 g m^{-3} h^{-1}, B = 120 g m^{-3} h^{-1} and C = 180 g m^{-3} h^{-1}. As the removal efficiency dropped at stage C in the biofilter without O_3 addition, the same inlet load was maintained in the following stages (D, E, F y G = 180 g m^{-3} h^{-1}, Phase 2) in order to continue to operate the two

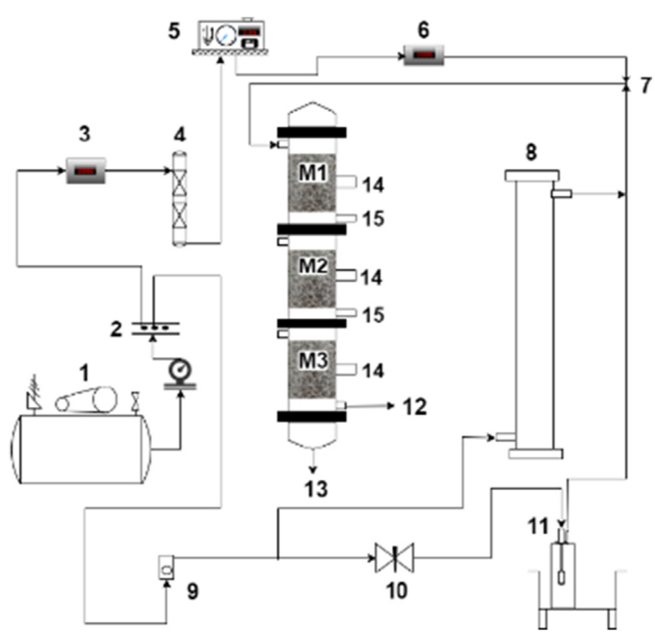

Fig. 1. Biofilter system with O_3 addition. M1, M2, M3 (Modules 1, 2 y 3). 1. - compressor; 2. - air distributor; 3. - and 6. - mass flow controller; 4. - desiccator; 5. - ozone generator; 7. - union between ozone and EA gas flow; 8. - humidifier; 9. - rotameter; 10. - needle valve; 11. - EA stripping reactor; 12. - treated air; 13. - leachate; 14. - biomass sampling ports; 15. - gas phase sampling ports.