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## Q4 Sulfur dioxide and o-xylene co-treatment in biofilter: 2 Performance, bacterial populations and bioaerosols emissions

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### A B S T R A C T

Sulfur dioxide (SO<sub>2</sub>) and benzene homologs are frequently present in the off-gas during the process of sewage sludge drying. A laboratory scale biofilter was set up to co-treat SO<sub>2</sub> and o-xylene in the present study. SO<sub>2</sub> and o-xylene could be removed simultaneously in a single biofilter. Their concentration ratio in the inlet stream influenced the removal efficiencies. It is worth noting that the removal of SO<sub>2</sub> could be enhanced when low concentrations of o-xylene were introduced into the biofilter. *Pseudomonas* sp., *Paenibacillus* sp., and *Bacillus* sp. were the main functional bacteria groups in the biofilter. Sulfur-oxidizing bacteria (SOB) and o-xylene-degrading bacteria (XB) thrived in the biofilter and their counts as well as their growth rate increased with the increase in amount of SO<sub>2</sub> and o-xylene supplied. The microbial populations differed in counts and species due to the properties and components of the compounds being treated in the biofilter. The presence of mixed substrates enhanced the diversity of the microbial population. During the treatment process, bioaerosols including potentially pathogenic bacteria, e.g., *Acinetobacter lwoffii* and *Aeromonas* sp., were emitted from the biofilter. Further investigation is needed to focus on the potential hazards caused by the bioaerosols emitted from waste gas treatment bioreactors.

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### 46 Introduction

47 With increased treatment rates and treatment capacity of  
 48 municipal sewage treatment plants, the production of sewage  
 49 sludge with 80% moisture content from wastewater treatment  
 50 plants reached up to 27.86 million tons in 2016. Besides  
 51 moisture, organic matter, and inorganic salts, the sludge  
 52 contains hazardous substances such as pathogens and heavy  
 53 metals, which pose threats to human health and the environ-  
 54 ment. Sanitary landfills, composting, and incineration are three  
 55 main technologies that are frequently used in sludge treatment.  
 56 Among these methods, sludge incineration is the most effective

and reliable technology because it minimizes the sludge volume  
 and recovers dry sludge. The pathogens and other toxic  
 compounds can be destroyed during the incineration process.  
 Co-processing of sewage sludge in cement kilns is an emerging  
 technology, characterized by feeding the sewage sludge into the  
 cement kiln for incineration (Lv et al., 2016). The moisture  
 content of the sludge should be reduced to below 30% before  
 incineration in the cement kiln. Thermal drying by waste heat  
 of the cement kiln exhaust gases is an economic way of sludge  
 desiccation. In Europe, particularly in Germany, the sludge  
 drying method has been frequently applied in cement kilns and  
 coal-fired power stations (Kelessidis and Stasinakis, 2012). This

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environmentally friendly disposal method has attracted increasing attention in China and has been employed in Beijing, Shanghai, and Guangzhou in recent years.

In the sludge drying process, the temperature reaches up to 290°C to sterilize the sludge and destroy its gel structure. Large amounts of off-gases containing odors and volatile organic compounds (VOC) are released into the atmosphere during the process, which cause odorous air pollution that is harmful to human health and the environment. SO<sub>2</sub>, NH<sub>3</sub>, and VOC are frequently present in the emissions from the sewage sludge desiccation process. Effective treatment technologies are needed to reduce these emissions. Biofiltration is a method with low operating costs and small amounts of hazardous residual waste, and is efficient in the purification of off-gases with large flow rates and low concentrations. Biological technologies were used to remove hydrogen sulfide, mercaptans, ammonia, and amines from air. Approximately 10 years later, this technology has been extended to the removal of VOC, volatile organic sulfur, and SO<sub>2</sub>. Several studies have reported effective elimination of aromatic VOC, including BTEX by these technologies (Jeong et al., 2009; Rahul et al., 2013; Wang et al., 2013). Philip and Deshusses (2003) developed a combined bioreactor system containing a biotrickling filter followed by a biological post-treatment unit to treat SO<sub>2</sub> from flue gases. Their results showed that nearly 100% SO<sub>2</sub> removal efficiency was achieved by using this treatment system. Although biofilters have many advantages, there are still some problems with those methods and the performance of biofilters in the treatment of gases containing complex pollutants needs further improvements. In addition, biofilters inhabited by bacteria and fungi are sources of microbial bioaerosol emissions (Martens et al., 2001). Because of arising public concern about such emissions and possible health risks to the exposed people, further research should be conducted to understand the degree of emissions from airborne microorganisms in biofilters during the odor or VOC treatment processes.

In the present study, a laboratory-scale biofilter was established to treat the waste gas containing SO<sub>2</sub> and *o*-xylene. SO<sub>2</sub> and *o*-xylene were selected as representatives of sulfur-containing compounds and monoaromatic hydrocarbon compounds that are usually present in the sludge-drying tail gases. The performance of the biofilter was evaluated for over 6 months by varying the inlet load of *o*-xylene and SO<sub>2</sub>. The microbial population in the biofilter was analyzed using PCR-DGGE (denaturing gradient gel electrophoresis) methods to investigate the relationship between microbial characteristics and the removal of waste gas mixture with *o*-xylene and SO<sub>2</sub>. Bioaerosol emissions from the biofilter were also studied.

## 1. Materials and methods

### 1.1. Experiment and procedure

The co-treatment of SO<sub>2</sub> and *o*-xylene was carried out in a biofilter with a height of 2.5 m and a diameter of 0.2 m (Fig. 1). The effective volume of the biofilter was 0.056 m<sup>3</sup>. Polyurethane foam cubes with 97% of porosity were packed in the biofilter column. The volume of each cube was approximately 1 cm<sup>3</sup>. The packing density was 12–15 kg/m<sup>3</sup>. Bacteria or other

microorganisms were grown on polyurethane foam cubes. SO<sub>2</sub> and *o*-xylene were selected as representative compounds to be treated by the biofilter. Three streams of pure SO<sub>2</sub> (Beijing Hua Yuan Gas Chemical Industry Co., Ltd.), pure *o*-xylene (Beijing Hua Yuan Gas Chemical Industry Co., Ltd.), and air were mixed to generate the synthetic gaseous waste stream. The desired concentrations of SO<sub>2</sub> and xylene in the inlet stream were maintained by adjusting the rates of the three airflows. The gaseous stream with SO<sub>2</sub> and *o*-xylene was passed through the biofilter column with a flow rate of 3.5 m<sup>3</sup>/hr. The corresponding empty bed residence time was 51 sec.

Initially, the polyurethane foam cubes were inoculated with microbial cultures containing sulfur-oxidizing bacteria (SOB) obtained from a bioreactor for sludge-drying tail gas purification. Approximately 50 g of packing material was soaked in 10.0 L nutrient solution containing 2.0 g/L of Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>·5H<sub>2</sub>O, 0.5 g/L of beef extract, 2.5 g/L of NH<sub>4</sub>Cl, 4.5 g/L of KH<sub>2</sub>PO<sub>4</sub>, 0.1 g/L of FeSO<sub>4</sub>·7H<sub>2</sub>O, and 0.1 g/L of MgSO<sub>4</sub>·7H<sub>2</sub>O (pH 4.5–5.0). After shaking the solution for 30 min in an ultrasonic oscillator (KQ-250B, Kunshan, China), the liquid with suspended cells was cultured in 5.0 L nutrient solution at 36°C and 120 r/min for 3 days. Then the liquid was transferred into the same nutrient solution to obtain the enrichment capable of degrading SO<sub>2</sub>. The inoculums containing *o*-xylene-degrading microbial population were obtained from an acclimatized culture seeded with soil from a place close to the petroleum factory, where one can expect to find xylene-degrading microorganisms. The soil was sieved to separate particles with diameter larger than 1 mm. Next, 100 g soil was soaked into 1.0 L of nutrient solution containing 2.0 g/L of NH<sub>4</sub>Cl, 3.5 g/L of KH<sub>2</sub>PO<sub>4</sub>, 0.1 g/L of FeSO<sub>4</sub>·7H<sub>2</sub>O, and 0.1 g/L of MgSO<sub>4</sub>·7H<sub>2</sub>O (pH 5.0–5.5) and was shaken for 30 min in an ultrasonic oscillator (KQ-250B, Kunshan, China). Then, 200 mL liquid with suspended cells diluted with 1.0 L of the same nutrient solution was placed in an aerated batch reactor. The reactor was continuously fed with *o*-xylene at a rate of 0.6 g/hr for 35 day. The microbial cultures were harvested by refrigerated centrifugation at 3000 r/min for 20 min (Biofuge Stratos, Heraeus, Germany). The concentrated cells were mixed with 3.0 L nutrient solution and seeded onto the biofilter packing material.

The biofilter was irrigated periodically by the mineral nutrient solution, which contained 5.0 g/L of KH<sub>2</sub>PO<sub>4</sub>, 1.0 g/L of NH<sub>4</sub>Cl, 0.1 g/L of FeCl<sub>2</sub>·7H<sub>2</sub>O, and 0.2 g/L of MgCl<sub>2</sub> to sustain moisture and provide nutrients for microorganism growth. The sampling ports were located at the bottom and the top of the biofilter column. Samples were regularly collected from the sampling ports to determine the performance of the biofilter.

### 1.2. Airborne microorganisms capture

A Single Stage Andersen Cascade Impactor (N6, Thermo Scientific™, USA) with 400 precision machined jet orifices was used to capture airborne microorganisms emitted from the biofilter. Air stream was drawn through the sampler using a pump at a constant flow rate of 28.3 L/min to achieve the sharp cut point diameter of 0.65 μm. Airborne microorganisms were enriched from the collected air samples in Petri dishes containing different agar media and were cultivated. All inside surfaces were maintained in a sterile condition until sampling. The geometric mean of the replicates was calculated and the

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