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Performance evaluation and optimization of field-scale bioscrubbers for intensive pig house exhaust air treatment in northern Germany

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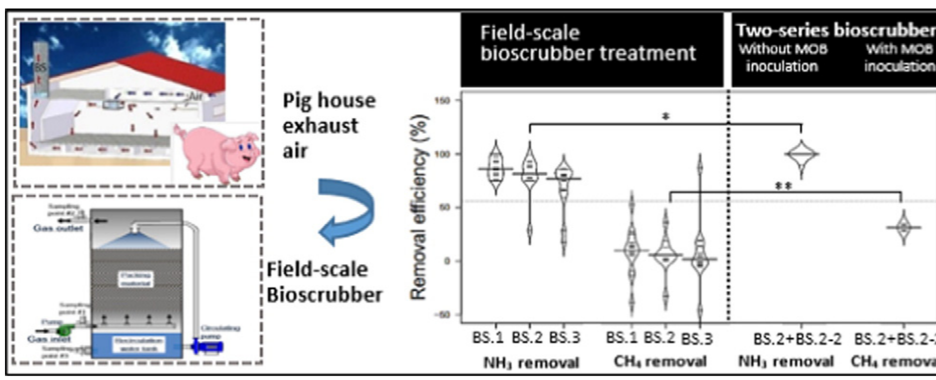
HIGHLIGHTS

- Bioscrubbers perform stable and high NH_3 removal under annual load variations.
- Two-series connected bioscrubber can promote NH_3 removal but not CH_4 removal.
- Isolated methanotrophic bacterial inoculation can improve CH_4 removal significantly.
- NH_3- and CH_4-oxidizers were *Nitrosomonas* sp. and Type I methanotrophs, respectively.

GRAPHICAL ABSTRACT



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ABSTRACT

The treatment of exhaust air from three intensive pig houses in northern Germany by field-scale bioscrubbers (BS.1, BS.2, and BS.3) were evaluated monthly in 2015. The simultaneous removal of NH_3 and CH_4 was investigated by connecting a second bioscrubber (BS.2-2) to one of the three bioscrubbers (BS.2) to create a two-series connected bioscrubber (BS.2 + BS.2-2). Additionally, whether isolated methanotrophic bacterial inoculation in BS.2-2 intensified CH_4 removal was examined.

Average NH_3 removal efficiencies of 86%, 80%, and 77% were observed for BS.1, BS.2, and BS.3, respectively, under fluctuate NH_3 inlet concentrations (variation of 22%–54%) throughout the study year. However, average CH_4 removal efficiencies were lower than 10% in the three bioscrubbers. The pH of the recirculation water, which ranged from 5.7 to 8.1, was demonstrated to be an important factor for NH_3 removal and negatively correlated with NH_3 removal and NH_4^+-N concentration in the recirculation water. The dominant NH_3-oxidizing and methanotrophic bacteria in the bioscrubbers, analysed by transmission electron microscopy, were *Nitrosomonas* sp. and Type I methanotrophs, respectively. NH_3 removal efficiency reached 100% in the two-series connected bioscrubber, however, CH_4 removal was still low (average of 2%). After inoculating isolated methanotrophic bacteria into BS.2-2, the average CH_4 removal was enhanced to 35%, offering a great option for bioscrubbers application to intensify CH_4 removal. Therefore, a two-series connected bioscrubber inoculated with methanotrophic bacteria would be an option for simultaneous removal of NH_3 and CH_4 from the exhaust air of animal houses.

1. Introduction

Intensive pig production is of great importance to the economies of many western European countries, however, it is also connected to a

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number of environmental effects in terms of emissions of NH_3 and the greenhouse gases, CH_4, N_2O, and CO_2 (Hamon et al., 2012; Melse et al., 2009b). Niedersachsen in northern Germany is one of the biggest pig production areas in Europe, contributing approximately 19% of the pig production on the continent (European Commison, 2003). Recently, to treat the exhaust air of these pig houses, bioscrubbers have become a state-of-the-art technique applied on a large scale due to their robust, cost-efficient, and ecological characteristics (Estellés et al., 2011; Hahne, 2011; Melse et al., 2009a).

To our knowledge, research on the annual performance of field-scale bioscrubbers for intensive treatment of pig house exhaust air is scarce. One previous study by Melse et al. (2012b) lasted 7 months (including winter and summer) for a field-scale bioscrubber equipped at a pig farm in the Netherlands; however, it focused merely on NH_3 removal, which varied between 42 and 92% but was mostly above 70%. Another study by Lafita et al. (2012) reported that a field-scale biotrickling filter (bioscrubber) performed stably and high biological removal efficiencies of hydrogen sulphide and odour were observed during nearly one year of monitoring when treating the waste gas from wastewater treatment plants. However, annual fluctuation in exhaust air inlet loading and seasonal temperature variation are expected to influence NH_3, CH_4, N_2O, and CO_2 treatment through biodegradation in the bioscrubber. For pig house exhaust air treatment, most previous studies have merely focused on short periods (≤two seasons) of monitoring NH_3 and N_2O removal (Melse and Mol, 2004; Melse and Mosquera, 2014), treating CH_4 and CO_2 (Aguilar et al., 2010), mitigating odour (Melse and Mol, 2004) and reducing particulate matter (Melse et al., 2012a). Therefore, an annual performance evaluation of field-scale bioscrubbers is needed.

In previous studies, it was demonstrated that field-scale bioscrubbers can achieve 45–99% NH_3 removal efficiencies (Belzile et al., 2010; Liu et al., 2016; Melse and van der Werf, 2005; Nisola et al., 2009; Sharvelle et al., 2008), whereas, their CH_4 removal rate is extremely low, ranging from 0.9–6% (Aguilar et al., 2010; Belzile et al., 2010). This is mainly because of the low transfer rate of CH_4 from the gas to the aqueous phase for further degradation and the possible inhibition of the methanotrophic bacteria from the high ammonia concentration in the aqueous phase. Thus, optimization of field-scale bioscrubbers to intensify CH_4 removal is needed urgently but studies on such are lacking. An easy and simple option is to connect another bioscrubber onto an existing one forming a two-series connected bioscrubber treatment system, which would allow the exhaust air to travel through two bioscrubbers to increase the air-water-biofilm contact time and biodegradation possibility during the second bioscrubber stage. Based on this concept, a multistage bioscrubber with a higher average air residence time was reported to achieve higher particulate matter (PM_{10}) removal than a single stage bioscrubber (Melse et al., 2012a). Nevertheless, whether this optimization can benefit CH_4 removal in a bioscrubber has never been addressed. Moreover, using various biological and chemical additives is also a common optimization method to improve pollutants removal by stimulating microbial biodegradation (Schmidt and Ahring, 1993; Sreekrishnan et al., 2004). Avalos Ramirez et al. (2012) reported that the performance of lab-scale bioscrubbers treating CH_4 was improved from 5 to 12% to 13–33% after adding nonionic surfactants (Brij 35) to the recirculation water. However, the packing materials were clogged quickly due to the detached biomass when Brij 35 was added. Methanotrophs were found as the main bacteria for methane removal in bioscrubber (biotrickling filter) and biofilter systems (Estrada et al., 2014; Yoon et al., 2009). The methane oxidation activities showed positively correlated with the amount (cell number) of methanotrophic bacteria (Gebert et al., 2003). Therefore, a further perspective to increase the removal efficiency of methane would be the inoculation with methanotrophs. Nevertheless, the direct effect of extra methanotrophs addition, as biological additive, on the methane removal in bioscrubbers need to be studied.

In this study, three field-scale bioscrubbers in the Niedersachsen area of northern Germany were selected for our case study to evaluate the treatment of intensive pig house exhaust air during the whole year of 2015. The inlet and outlet concentrations of NH_3 and the greenhouse gases CH_4, N_2O, and CO_2 were measured monthly. Transmission electron microscopy (TEM) was used to detect the dominant NH_3-oxidizing and methanotrophic bacteria in the three selected bioscrubbers. To achieve simultaneous removal of NH_3 and CH_4, the performance of a two-series connected bioscrubber was evaluated and the effect of methanotrophic bacterial inoculation on CH_4 removal was examined as well.

2. Materials and methods

2.1. Field-scale bioscrubbers

This research was conducted at three field-scale bioscrubbers (BS.1, BS.2, and BS.3) equipped at three different intensive pig production houses in Niedersachsen (Cloppenburg area), northern Germany. The pig in the selected pig production houses were 70–150 day old, and the pig numbers were similar in the range of 1160–1700 (Table 1). Each pig house was equipped with the same mechanical ventilation channel setup. The exhaust air passed through the channel and flowed into each field-scale bioscrubber with an average inlet air ventilation rate (IAVR) of 27.8–36.4 $m^3\ s^{-1}$. Fig. 1 presents a schematic of a single bioscrubber system. Each bioscrubber included an air pump at the pig house exhaust air inlet, a packed bed for biological treatment, a recirculation water tank and water pump for water recirculation. The packing was formed by stacks of coarse plastic square plate (polyethylene, 0.3 m in length and 1.0 cm in thickness). The distance of each plate was around 5 cm. The packing stacks were placed with 45° tilt to the airflow direction (Fig. 1). The exhaust air flowed upwards through the packed bed while water was sprayed simultaneously from the top of the bioscrubber. These opposite flow directions of the water and exhaust air can provide intensive contact between gas and liquid. The moisture condition caused by the water spray can also promote bacterial growth and extend the contact time between bacteria and pollutants. The characteristics of the selected field-scale bioscubbers are summarized in Table 1. BS.1, BS.2, and BS.3 were constructed in 2013, 2014, and 2011, respectively. They differ slightly in their effective packing materials volume (44–57 m^3) and recirculation water tank volume (16–21 m^3), but have the same empty bed air residence time (EBRT) of 1.6 s and recirculation water flow rate of 10 $m^3\ h^{-1}$. Once the electrical conductivity (EC) of the recirculation water exceed 22 mS cm^{-1} (around every 1–2 month), half volume of the water was discharged and fresh water was filled up in each recirculation tank.

2.2. Experimental and sampling strategy

2.2.1. Sampling from the three field-scale bioscrubbers

The sampling activities were conducted from January to December of 2015. Samples were taken every month, except February and October, from the inlet (gas; sampling point #1 in Fig. 1), outlet (gas; sampling point #2 in Fig. 1), and recirculation water tank (water; sampling point #3 in Fig. 1) of all three selected field-scale bioscrubbers (BS.1, BS.2, and BS.3) for performance evaluation (Fig. 2). The gas samples were taken using an electric air pump (SP 750 EC/50 Hz, Schwarzer Precision, Germany) at an air flow rate of 6 L min^{-1}. During gas sampling, one side of the pump was connected to the sampling site (Fig. 1), while the other side was connected to a vacuum glass tube (10 mL) equipped with a single polypropylene fitting. Additionally, each gas sample was only taken after the first 10 min discarded when the gas flow statically and the sample was assumed to be representative. Water samples (1 L) were collected from the recirculation water tank into an amber flask (full volume). Moreover, biofilm samples (10 mL) from the packing material of BS.1, BS.2, and BS.3 were randomly collected from 3 places to evaluate the morphological properties and quantification of the NH_3- and CH_4-degrading functional bacterial communities