Methane removal using materials from biofilters at composting plants

Loại bỏ khí Mê-tan bằng cách sử dụng vật liệu trong các bộ lọc khí sinh học ở các cơ sở sản xuất phân vi sinh

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Methane (CH4) source of Greenhouse Gases should be considered; CH4 is formed by composting under anaerobic conditions. Using microbial Methane Oxidation is a solution with low cost and effective. In this study, 27 bio-filters and 18 laboratory-scale bioreactors were used to investigate the potential for CH4 removal in biogas. The CH4 Dinitrogen monoxide (N2O) and Carbon dioxide (CO2) concentrations at the inlet and outlet of the air purifier were measured by gas chromatography. The results showed that the CH4 concentration decreased in experiments while the CO2 and N2O content increased in all experiments. An experiment was conducted with 1 kg of biofilter material with the input of 800 ppm CH4 contained in a 5-liter flask for 49 hours containing. The results also showed that the CH4 concentration decreased by 71% after 20 hours and N2O was formed in the reactor.

Mê-tan (CH4) là nguồn khí gây nên hiệu ứng nhà kính cần được quan tâm, khi CH4 được sinh ra trong quá trình xử lý sinh trong điều kiện kí khí. Một giải pháp với chi phí thấp là sử dụng vi sinh vật oxy hóa khi CH4 cố định trên giá thể là vật liệu sử dụng trong thiết bị lọc sinh học. Trong nghiên cứu này, 27 thiết bị lọc sinh học trên thực tế và 19 bình lọc tại phòng thí nghiệm đã được sử dụng nhằm mục đích khảo sát khả năng loại bỏ CH4 cố định trong khí sinh học. Nồng độ khí CH4, N2O và CO2 đều tăng và điều điều lỏng khí CH4 cố định trong khí sinh học. Kết quả cho thấy nồng độ khí CH4 giảm sau khi quá hệ thống lọc sinh học ở một số bình, trong khi nồng độ khí CO2 và N2O lại tăng lên ở tất cả các bình. Khoảng số khí nồng độ CH4 đã được giảm 71% sau 20 giờ. Tuy nhiên, N2O đã được phát hiện có hình thành trong bình phản ứng.

Keywords: greenhouse gas, emissions, composting, windrows, organic waste, methane, biofilter

1. Introduction

Methane (CH4) and Dinitrogen monoxide (N2O) are considered to be strong Greenhouse Gases (GHGs), whereas NH3 is identified as an odour component and an indirect GHG. According to the Intergovernmental Panel on Climate Change (henceforth IPCC) Report in 2007, the Global Warming Potential (GWP) of CH4 and N2O in 100 years are respectively 25 and 298 times higher than that of CO2. CH4 emissions from livestock account for 37% of the total emissions of CH4 in global, mainly for ruminants in digestion. In addition, landfill gas collection has been established to generate electricity and heat in many countries. The CH4 emitted from landfills is one of the major sources accounting for approximately 30% of total CH4 emissions (Kára et al., 2010). In other cases, flares are operated to burn CH4 and other organic trace compounds contained in the landfill gas before releasing to the atmosphere. However, these processes cannot be applied in cases of low CH4 concentrations. Up to now, the mitigation of emissions with low CH4 concentrations often is neglected.

CH4 removal was observed with low gas flows and high concentrations in the soil or in biofilters (Bender & Conrad, 1992). In a biofilter, an autochthonous methanotrophic biofilm can develop and use CH4 as a carbon source. However, the CH4 removal in biofilters varied from case to case due to different working conditions e.g. ambient temperature, humidity and nutrient content.

Most of the previous reports (Scheutz et al., 2009; Melse & Van De Werf, 2005 and Bender & Conrad, 1992) showed that a good CH4 oxidation rates of 20% CH4 in the supply air and 2.5% CH4 in the exhaust air. Biofilter has a high flow rate in a combination of low concentrations of CH4. Low CH4 concentrations range of below 0.07% CH4 (700 ppm ~ 460 mg CH4/m³) in combination with a high exhaust air volume flow in the range of 20,000 - 50,000 m³/h. This results in...
in a biofilter had a load of about 100 m³/m³·h. The efficiencies of CH₄ reduction range from 0% - 20% (Scheutz et al., 2009). In addition, Melse and Van De Werf (2005) reported a CH₄ reduction of 80% in the biofilter with a load of 0.75 m³/m³·h.

CH₄ oxidation is therefore considered as an important method to CH₄ emissions reduction. At present, only a few scientific publications on biological waste air treatment are available. Studies related to test the CH₄ removal capacity of a new material have not yet been done. Looking into the real conditions of the biofilter, among different microorganisms, CH₄ oxidation bacteria needs to be found. Then, they will be enriched populations, which are added to the biofilter or bioscrubber on demand.

The aim of this work was to investigate CH₄ removal in different biofilters and to develop a new method to find materials for CH₄ oxidation.

2. Materials and methods

2.1 Screening of biofilters

Twenty-seven biofilters in composting plants in Germany were investigated in the study. The study was carried out from 2012 to 2014. An overview of a capsulated biofilter is shown in Figure 1. The gas before and after biofilter was analysed at each plant. At capsuled biofilters, the treated air left the biofilter in a chimney. Here the gases were measured. At the open biofilter of 16m² (4x4m) was covered by a thin foil. Concentrations of the treated gases were measured under the foil.

Exhaust gases were sampled manually by evacuated headspace vials and subsequently analysed for CH₄, CO₂ and N₂O by gas chromatography (ECD/FID, SRI 8610C, USA) in the laboratory in Bonn University, Germany. It was assumed that the volumes of treated and untreated air were the same.

![Figure 2. Test kit for determination of CH₄ removal capacity](image)

![Figure 3. Material of biofilter for the testing of CH₄ removal capacity](image)

The removal performance was determined by mass balance according to the following equation:

\[
\text{CH}_4\text{ox}_{-i} = \left(\frac{\text{flux}_\text{in} - \text{flux}_\text{out}}{\text{flux}_\text{in}}\right) \times 100
\]

With

\[
\text{CH}_4\text{ox}_{-i}: \text{At the time } i \text{ oxidized portion of inlet flow } [\%]
\]

\[
\text{Flux}_{\text{ox}}: \text{CH}_4 \text{ concentration flow into the reactor at time } i \text{ (i=} 1, 2, 19, 22, 24, 26 \text{ and } 49 \text{ hours})
\]
Flux<sub>out</sub>: CH<sub>2</sub> concentration flow out the reactor at time i (i = 1, 2, 19, 22, 24, 26 and 49 hours)

3. Results and discussion

3.1 CH<sub>4</sub> removal from biofilters at composting plants

The inlet concentration of CH<sub>4</sub> and CH<sub>4</sub> reduction of biofilters were shown in Figure 4. CH<sub>4</sub> was reduced and produced in the biofilters. The removal efficiency ranged from production of ~260% to reduction of 80% for CH<sub>4</sub>. The mean concentration of CH<sub>4</sub> before biofilter was (106 ± 73) ppm. The average CH<sub>4</sub> removal efficiency was 25% at 13 biofilters. With regard to other literature, CH<sub>4</sub> was reduced by 15% (Amlinger et al., 2008). As waste gas permeates the biofilter layer, CH<sub>4</sub> is oxidized to CO<sub>2</sub> and H<sub>2</sub>O by methanotrophic bacteria present in the biofilter’s material. The effect on the production of CH<sub>4</sub> emissions in the biofilter may be explained by anaerobic degradation of organic substances in the biofilter (Nguyen & Cuhls, 2016). However, it is not yet well understood.

The reductions of CH<sub>4</sub> were lower than in previous studies (Table 1). Melse and Van De Werf (2005) reported the CH<sub>4</sub> reduction of 80% in a biofilter with the load of 0.75 m<sup>3</sup>/m<sup>3</sup>.h. However, this loading factor is 130 times lower than in practice. Previous studies illustrated that the process-controlling factors of the CH<sub>4</sub> oxidation in low-concentrations are the O<sub>2</sub> availability, storage density and gas permeability as well as the volume flow of the gas to be treated (Bender & Conrad, 1992: Benstead & King 1997 and Dunfield et al., 1999). In fact, the low CH<sub>4</sub> removal efficiency in practice could be explained by exposure time due to a short duration of the waste air treatment in biofilters.

3.2 Methane oxidation reaction

3.2.1 The value of pH

The pH of biofilter’s material was from neutral to a light alkaline (ranged from 7.16 to 7.82). The pH values were suitable for methanotrophic activities. The CH<sub>4</sub> removal process was observed in a broad pH range, from pH< 4 in a sand soil to pH> 9 in a bog soil. The optimal pH for methanotrophs growth is between 6 and 8 (Cao & Staszewska, 2011).

3.2.2 Methane removal and carbon dioxide generation

CH<sub>4</sub> removal and CO<sub>2</sub> generation were investigated for the biofilter’s materials. The observed decrease in CH<sub>4</sub> concentration in Figure 5 and 6 can be explained by oxidation since O<sub>2</sub> was available and CO<sub>2</sub> was increased. After an incubation for 49 h at 25°C, 99% of CH<sub>4</sub> was consumed. The CH<sub>4</sub> removal rate increased when the O<sub>2</sub> concentrations increased from 2.5% to 15% (Cao & Staszewska, 2011).

By comparison, the CH<sub>4</sub> concentration in the reactor was 200 ppm after 20 hours (500 ppm reduction); whereas, the CO<sub>2</sub> concentration was 3000 ppm (2500 ppm generation). This could be explained by aerobic decomposition of material in the reactor due to the availability of O<sub>2</sub>. Maximum CH<sub>4</sub> consumption and maximum CO<sub>2</sub> generation were observed at the first phase of incubation. Previous studies have shown that the rate of CH<sub>4</sub> removal grows with the increase of CH<sub>4</sub> concentration. The increase of CH<sub>4</sub> concentrations from 2 to 16 % led to 1.1 - 2.5 folds increase of the methanotrophic activity (Pawlowska & Stepniewski, 2004). Another author found that a value (2.3-folds) in the methanotrophic activity increase was observed, where the measured CH<sub>4</sub> concentration varied from 25 to 200 ppm (Whalen & Reeburgh, 1996).

![Figure 4. Inlet CH4 concentrations and removal efficiencies of CH4 at the studies biofilters](image)

![Figure 5. Inlet CH4 concentrations and removal efficiencies of CH4 at the studies biofilters](image)

Table 1. Comparison of removal efficiency of biofilters: this study and data from literature (Scheutz et al., 2009)

<table>
<thead>
<tr>
<th>Reference</th>
<th>CH4 removal (%)</th>
<th>CH4 removal rate max (g/m&lt;sup&gt;3&lt;/sup&gt;.day)</th>
</tr>
</thead>
<tbody>
<tr>
<td>This study</td>
<td>7 to 27</td>
<td>5-56</td>
</tr>
<tr>
<td>Amlinger et al., 2008</td>
<td>15</td>
<td>-</td>
</tr>
<tr>
<td>Trimborn, et al., 2003</td>
<td>up to 26</td>
<td>-</td>
</tr>
<tr>
<td>Du Plessis et al., 2003</td>
<td>-</td>
<td>87</td>
</tr>
<tr>
<td>Wilhusen et al., 2004</td>
<td>-</td>
<td>96-276</td>
</tr>
<tr>
<td>Streese &amp; Stegmann, 2003</td>
<td>-</td>
<td>341</td>
</tr>
<tr>
<td>Melse &amp; Van De Werf, 2005</td>
<td>-</td>
<td>377</td>
</tr>
<tr>
<td>Park et al., 2005</td>
<td>-</td>
<td>435</td>
</tr>
<tr>
<td>Sly et al., 1993</td>
<td>-</td>
<td>586</td>
</tr>
<tr>
<td>Haubrichs &amp; Widmann, 2006</td>
<td>-</td>
<td>592</td>
</tr>
</tbody>
</table>

Note: "-" means data was not available
was transformed into N₂O. Clemens and Cuhls (2003) reported that 26% of NH₃-N was transformed and released as N₂O and NH₄⁺.

The increase of N₂O concentrations in the reactors during incubation time is significant. The total cumulative N₂O generation after 50h incubation was 1.4 ppm was much lower than the reduction of CH₄ (823 ppm). After the end of the experiment, the contribution of N₂O to the total GHG concentration in the outlet air is insignificant.

CO₂ concentration was increased steadily (Figure 6). CO₂ generation was increased 15 times after 49 hours of incubation (from 3,400 ppm to 57,000 ppm). Although the amounts of CH₄ removal by 3 replications were similar and decreased slightly after 20 hours’ incubation, CO₂ generation increased sharply. O₂ is a significant factor influencing the CH₄ oxidation process (Cao & Staszewska, 2011).

Results from experiments using biofilter’s material showed that the material was an excellent property as a carrier for CH₄ oxidation processes. Application of the microbial oxidation is a promising way to control the CH₄ emission from organic waste treatment.

CH₄ reduction rates in some biofilters were significantly lower than other. In biofilters, CH₄ is either oxidized or produced due to the O₂ availability. The CH₄ oxidation rate was lower in some part of biofilters because of the anaerobic condition. Therefore, the designing the biofilters correspondingly should effectively promote the microbial CH₄ removal of exhausted gas.

Further study is needed to understand and explain the CH₄ formation and removal in the biofilter in order to optimize the CH₄ and N₂O reduction processes. More studies with the continues reactor should be carried out because GHG emission of N₂O in the outlet air of the batch reactor was 3 times higher than the inlet gases. If it is the same tendency in the continuous reactor, it will be concerned more when the concentration of CH₄ (GPW = 298) increases three times in biofilter.

4. Conclusions

Biofilter has the capacity of CH₄ removal and reduction. This is an attractive option for CH₄ removal at low concentration in the organic waste treatment plants.

3.2.3 Nitrous oxide formation

N₂O generations were low at the start of the experiment and increased steadily to a maximum rate (Figure 7). The increase in N₂O concentrations during the experiment is attributed to the nitrification process.

The formation of N₂O during the further duration of the experiment indicates that some N₂O production occurred in the biofilter material. The increase of N₂O may be explained by the fact that N is converted to N₂O by nitrification due to continuous aerobic conditions in the biofilters (Melse & Van der Werf, 2005). According to previous studies, around one third NH₃ that enters biofilters can be transformed and released as N₂O (Trimborn, 2003). Similarly, Clemens and Cuhls (2003) reported that 26% of NH₃ was transformed into N₂O in biofilters.

Figure 5. CH₄ concentrations in the reactors during incubation time

Figure 6. CO₂ concentrations in the reactors during incubation time

Figure 7. N₂O concentrations in the reactors during incubation time

3. References


