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Removal of greenhouse gas in biofilter using organic and inorganic media

Nguyen Thanh Tung[1], Wilai Chiemchaisri[1], Chart Chiemchaisri[1*], Samunya Sanguanpak[2]

[1]Department of Environmental Engineering, Faculty of Engineering, Kasetsart University, Thailand
[2]National Metal and Materials Technology Center, National Science and Technology Development Agency, Thailand

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Abstract:

A biofilter using organic (compost) and inorganic (pumice, porous silica pellet or poremat) media was applied to the removal of methane (CH_4) and nitrous oxide (N_2O) to minimize the impact of off-gasses from municipal solid waste disposal global. The objective was to determine the appropriate biofilter media for CH_4 oxidation and N_2O conversion. The off gas (59.6% CH_4, 1.0% N_2O) was fed simultaneously with air (1:3 ratio) into the biofilter. CH_4 oxidation and N_2O conversion rates were observed over a 101-day period through the analysis of gas concentration along the biofilter's depth using gas chromatography. Higher CH_4 oxidation in the biofilters containing organic and inorganic media was achieved, especially for the compost-poremat biofilter with 70.1 g $CH_4/m^3/d$ realised. The most active methane oxidation zone was found near the gas inlet at the bottom of the biofilter. The presence of inorganic material helped promote aerobic conditions for CH_4 oxidation, especially during the initial period. N_2O was also more completely removed with the biofilter containing inorganic media. Higher methanotrophic activities in matured biofilter media and the presence of methanotrophs type I, which prefer oxygen-rich conditions, were confirmed.

Keywords: **biofilter, compost, greenhouse gas, inorganic media, methane oxidation.**

Classification number: **5.1**

Introduction

Municipal solid waste (MSW) disposal on land either by sanitary landfilling or open dumping are the most common methods used in Asian countries [1]. Their major environmental impacts include pollution from leachates and greenhouse gas emissions. Highly potent greenhouse gases, including methane (CH_4) and nitrous oxide (N_2O), could be produced during solid waste decomposition in waste landfills as well as leachate treatments [2-5]. Therefore, the mitigation of CH_4 and N_2O emissions from emitted gas is required to minimize the overall environmental impact from MSW management.

Biofiltration is considered to be a cost-effective and efficient means to mitigate diffuse CH_4 emissions [6]. In a biofilter, CH_4 is oxidized to CO_2 by methanotrophic bacteria under the presence of oxygen [7]. Previous studies have reported the application of biofilters to alleviate CH_4 in off-gases from landfills [8] and wastewaters [9, 10]. Different organic materials have been applied successfully as biofilter media including compost [11] and stabilized wastes [12]. It was reported that the loose texture of the organic media provided sufficient oxygen while supplementing nutrients for methanotrophic activities [11]. Nevertheless, high CH_4 oxidation could not be sustained over long-term operation due to the shortage of oxygen supply from media clogging as a result of excessive extracellular polysaccharide (EPS) production from methanotrophic activities [13]. On the other hand, inorganic media such as gravels and porous clay particles have been used as biofilter media [14], but the optimum nutrient solution needs to be continuously supplemented to maintain its methanotrophic activities [15]. To deal with these difficulties, a combination of organic and inorganic packing materials for CH_4 biofiltration systems were studied in this research. In our hypothesis, the organic materials played a role in supplying nutrients for methanotrophs while the clogging problems of filter media were alleviated by the presence of granular inorganic materials that preserve the stability of the filter media structure and allow uniform gas distribution. Moreover, we also investigated N_2O conversion, which occurred simultaneously with CH_4 oxidation, during gas treatment in the biofilter.

**Corresponding author: Email: fengccc@ku.ac.th*

Materials and methods

In this study, three different materials, including one organic material (compost) and two inorganic materials (pumice, porous silica pellet), were used to investigate their methane removal capacities. Specifically, the organic material was commercial grade leaf compost for garden applications and the inorganic materials were pumice of 3-5 mm diameter (commonly used as filter material in fishponds or aquariums and for garnishing soil for flowering and ornamental plants) and porous silica pellets called "poremat", which is commonly used as media in biological water treatment and controlled planting systems.

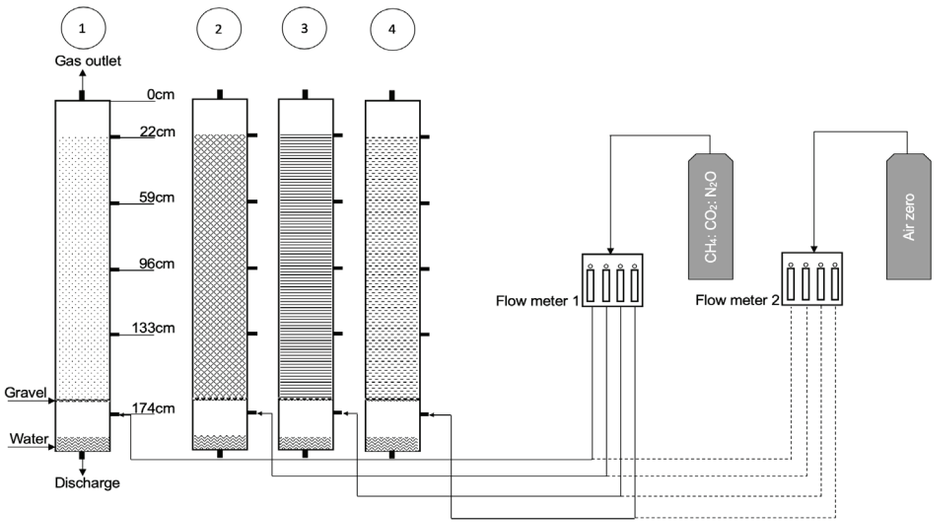

Fig. 1. Schematic of experimental column with different media: (1) compost, (2) compost-pumice, (3) compost-poremat, and (4) compost-pumice-poremat.

Four acrylic closed-top columns of 5 cm diameter and 180 cm length (Fig. 1) were used. The columns were equipped with gas sampling ports sealed with butyl rubber stoppers. Each column contained 3 l of filter material placed over 2 cm of gravel and a perforated plate located 15 cm from the bottom of the column. The 1st column was filled with 100% compost whereas the 2nd, 3rd, and 4th columns were filled with a mixture of compost and pumice at 70:30% (w/w), a mixture of compost and poremat at 70:30% (w/w), and a mixture of compost, pumice, and poremat at 70:15:15% (w/w), respectively. The placement of the biofilter material was performed by homogeneously mixing the indicated material proportions and placing it into the biofilter as a 10 cm layer until the total media depth (150 cm) was reached. The amount of media placed into each biofilter was as follows: column 1 had 1,708 g of compost; column 2 had 1,340 g of compost and 574 g of pumice, column 3 had 1,381 g of compost and 592 g of poremat, and column 4 had 1,361 g of compost, 292 g of pumice, and 292 g of poremat. The mixed media samples were also kept for characterization. The physicochemical characteristics of the biofilter materials in each column were analysed following standard methods [16] and the results are shown in Table 1. Artificial gas (CH_4:CO_2:N_2O=59.94:39.96:0.10%) and laboratory grade purified air (or air zero) were continuously fed to the bottom of the columns at 0.5 and 1.5 ml/min flow rates. These gas concentrations were set to simulate the emission of CH_4 (20-60%) and N_2O concentrations (up to 300 ppm) from open dumpsites and aerated closed landfill sites reported in previous studies [17, 18]. The experiment was conducted at room temperature (28-30°C) and the moisture content of each media were adjusted from their initial values of 10-15% (Table 1) to 25% after being placed in the biofilter to optimize methanotrophic activities [19]. At the end of the column experiment, the biofilter materials in the columns were sampled and analysed for methanotrophic activity and responsible microorganisms.

Table 1. Characteristics of biofilter media.

Parameters (unit)	Column No.			
	1	*2*	*3*	*4*
pH	7.5	7.5	7.7	7.6
Moisture content (%)	15.2	10.4	10.6	10.5
Porosity (%)	39.7	61.5	54.6	58.3
Bulk density (g/cm^3)	0.58	0.65	0.67	0.66
Total Nitrogen (µg/g)	10,008	7,718	7,823	5,465
NH_4^+ (µg/g)	69.9	39.7	19.3	27.0
NO_3^- (µg/g)	1,059	942	833	726
PO_4^{3-} (µg/g)	5,268	4,010	3,941	3,803

The column experiment was initialized one month prior to the methane oxidation rate (MOR) measurements. There was no initial inoculation of methanotrophs in the biofilter media, thus an initial start-up operation was required to allow indigenous methanotrophic bacteria to develop naturally in the biofilters. During the experiment, gas samples were collected from the sampling ports and analysed for their composition (CH_4, CO_2, O_2 and N_2O) using gas chromatography (GC). For CH_4, CO_2, and O_2 gas analysis, an Agilent GC6890 equipped with a CTR I (Alltech®) column and thermal conductivity detector (TCD) was used. The operating conditions were an inlet temperature of 105°C, column temperature of 35°C, and a detector temperature of 150°C with helium (He) carrier gas

flow rate of 65 ml/min. Meanwhile, a Perkin Elmer Clarus 580 GC equipped with a Heyesep D column and electron capture detector (ECD) was used for N_2O analysis. The operating temperatures of the injector, column, and detector temperatures of 400, 60, and 300°C, respectively, with a He carrier gas flow rate of 20 ml/min.

For CH_4 and N_2O, the MOR and nitrous oxide conversion rate (NCR) were calculated from the inflow and outflow gas concentrations from each section of the biofilter media depth as shown in the following equation:

$$R\ or\ NCR = \frac{Q[(C)_{in}-(C)_{out}]}{V_{media}} \quad (1)$$

where MOR = methane oxidation rate (g $CH_4/m^3/d$); NCR = nitrous oxide conversion rate (g $N_2O/m^3/d$); Q = gas flow rate (m^3/d); $(C)_{in}$ = inflow CH_4 or N_2O gas concentration (g/m^3); $(C)_{out}$ = outflow CH_4 or N_2O gas concentration (g/m^3); V_{media} = volume of filter media at corresponding depth (m^3). The t-test analysis was used to verify any significant difference between the experimental columns at 95% confidence interval ($p<0.05$). Microsoft Excel version 2019 was used to perform the statistical analysis.

Batch experiments were conducted to evaluate the methanotrophic activities and carbon dioxide production rates of biofilter media. Ten grams of media were taken from the columns at the end of experiment (day 101) and transferred into 108-ml serum bottles capped with rubber septa and aluminium rings. Each experiment was conducted in duplicate. Subsequently, 10 ml of artificial gas (CH_4:CO_2:N_2O) was added to the serum bottle and incubated at room temperature (28-30°C), then the gas composition in the headspace was analysed by GC daily over a 3-day period. During the batch experiments, the methanotrophic activity (MOA) and carbon dioxide production rate (CPR) of the biofilter media and methane or carbon dioxide concentration gradients were determined and input into the following equation:

$$MOA\ or\ CPR = \frac{pV_{gas}}{M}\frac{dC}{dt} \quad (2)$$

where MOA = Methanotrophic activities (mg CH_4/g media/h); CPR = Carbon dioxide production rates (mg CO_2/g media/h); ρ = CH_4 or CO_2 gas density at room temperature (mg/cm^3); V_{gas} = head space gas volume (cm^3); M = dried mass of media (g); dC/dt = CH_4 or CO_2 concentration gradient (vol. fraction/h).

The microbial population attached on the organic (compost) and inorganic (poremat) media were analysed by next generation sequencing. The isolation of the microbial genomic DNA from the test materials were performed by the DNeasy PowerSoil Pro Kit. The 16S rRNA gene was amplified using 341F and 805R primers (Quanta bio, USA). Cluster generation and 250-bp paired-end read sequencing were performed on an Illumina MiSeq and examined using FASTQC software. The methanotrophic, nitrifying, and denitrifying microorganisms responsible for CH_4 oxidation and N_2O production were targeted in this study.

Results and discussion

Methane oxidation rate (MOR) in biofilter

Figure 2 shows the MOR observed among the different biofilters after the start-up period. During the initial period (day 39-60), the biofilters containing organic and inorganic media provided a higher MOR compared to the compost material. Among them, the highest average MOR of 79.7 g $CH_4/m^3/d$ was observed in the compost-poremat media. Afterwards, the MOR of the compost media gradually increased from 61.1 g $CH_4/m^3/d$ during day 64-88 to 70.6 g $CH_4/m^3/d$ during day 91-101. During the biofiltration progression, the MOR in the biofilters with compost-pumice, compost-poremat biofilter, and compost-pumice-poremat biofilter were found to be slightly reduced over time. Nevertheless, the average MOR over the entire operation period of 101 days was found to be higher in the biofilters containing both organic and inorganic media. Among them, the highest MOR was achieved in compost-poremat biofilter at 70.1 g $CH_4/m^3/d$ (Table 2). Compared to other low CH_4 loading biofilters operated under passive gas flow conditions, the MOR achieved in the biofilter containing both organic and inorganic materials in this study was found in between those of sandy loam (96-176 g $CH_4/m^3/d$) [11], compost (176-224 g $CH_4/m^3/d$) [11], and stabilized wastes (7.6-34 g $CH_4/m^3/d$) [12]. Nevertheless, a higher MOR was also reported in a biofilter containing a compost and pumice mixture than that of compost media alone, even when the biofilter was operated under active gas flow conditions [20].

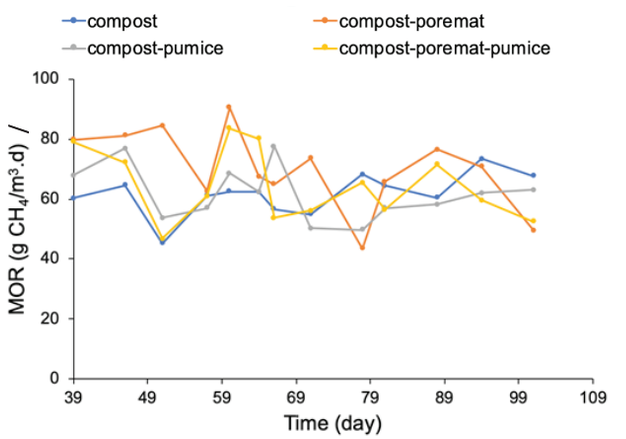

Fig. 2. Methane oxidation rate observed in biofilters containing

different media.

Table 2. MOR (g CH_4/m^3/d) in biofilters at different operation period.

Biofilter media	day 39-60	day 64-88	day 94-101	overall
Compost	58.7 (7.7)[a]	61.1(4.9)	70.6 (4.0)	61.7 (6.9)[a]
Compost-poremat	79.7 (10.4)[a,b]	65.3(11.6)	60.2 (14.9)	70.1(13.3)[a,b]
Compost-pumice	64.8 (9.4)[b]	59.1(10.2)	62.5 (0.8)	61.8 (9.0)[b]
Compost-pumice-poremat	68.5 (14.9)	63.9 (10.5)	56.0 (5.1)	64.4 (11.9)

Note: the numbers show average (SD) values; a and b show significant difference ($p<0.05$) between the biofilters, i.e. compost and compost-poremat biofilter, compost-poremat and compost-pumice, respectively.

The results suggest that the incorporation of inorganic media helped to achieve higher CH_4 oxidation rates in the biofilter, especially during the initial period. The CH_4 oxidation reaction produces water vapor as a product, so the increase and condensation of water within the biofilter media could reduce the mass transfer of CH_4 into the biofilm by blocking gas diffusion into available material pore space [21], thus decreasing CH_4 oxidation. Owing to the high porosity characteristics of inorganic materials, the mixture of inorganic materials and compost helped to increase the porosity of the biofilter media from 39.7 to 61.5, 54.6, and 58.3% in the biofilters with compost-pumice, compost-poremat, and compost-poremat-pumice media, respectively (Table 1). Under high porosity conditions, an enlarged pore space within the biofilter media could facilitate the diffusion of CH_4 and O_2 into methanotrophic biofilm CH_4 oxidation. This could explain the reason for achieving higher CH_4 oxidation in the biofilters containing both organic and inorganic media. In previous studies, it was also reported that the high porosity of biofilter media was favourable for CH_4 oxidation [8, 22]. Oppositely, a higher porosity media in compost-pumice biofilter and compost-pumice-poremat biofilter yielded a lower MOR as compared to the compost-poremat biofilter. This might be due to their shorter CH_4 retention times in the filter media. With the low solubility of CH_4 in the biofilm layer and the fact that only dissolved CH_4 can be consumed by methanotrophs, the retention time should be sufficient to ensure that CH_4 is converted to a form in which it becomes available to methanotrophic bacteria [18]. Therefore, these results reveal that the porosity of the biofilter media should be properly designed to achieve high CH_4 removal in the biofilter.

Figure 3 presents the gas concentration profiles within the biofilter, which confirmed the presence of CH_4 oxidation. An increase in CO_2 concentration and a decrease of CH_4 and O_2 clearly demonstrate ongoing biological methane oxidation. The most active zone of methane oxidation in all four biofilters was observed at the layer of 1.33-1.74 m depth from the top of biofilter. Meanwhile, there were smaller changes to the CH_4, CO_2, and O_2 concentrations in the upper part of the biofilter. Fig. 3A demonstrates that the CH_4 concentration in the biofilter with organic-inorganic media decreased to a larger extent than that within the compost biofilter in the active zone. Among the tested biofilters, the compost-poremat biofilter had the largest decrease in CH_4 concentration from 17% at the inlet to 6.2% at 1.33 m depth and the CH_4 concentration was mostly unchanged at the top of biofilter. Meanwhile, the CH_4 concentration in the compost biofilter was reduced to 8% at 1.33 m depth and slightly decreased to 7% at the top of the biofilter. The decrease in O_2 concentration corresponded to CH_4 oxidation by methanotrophs. However, it was noticed

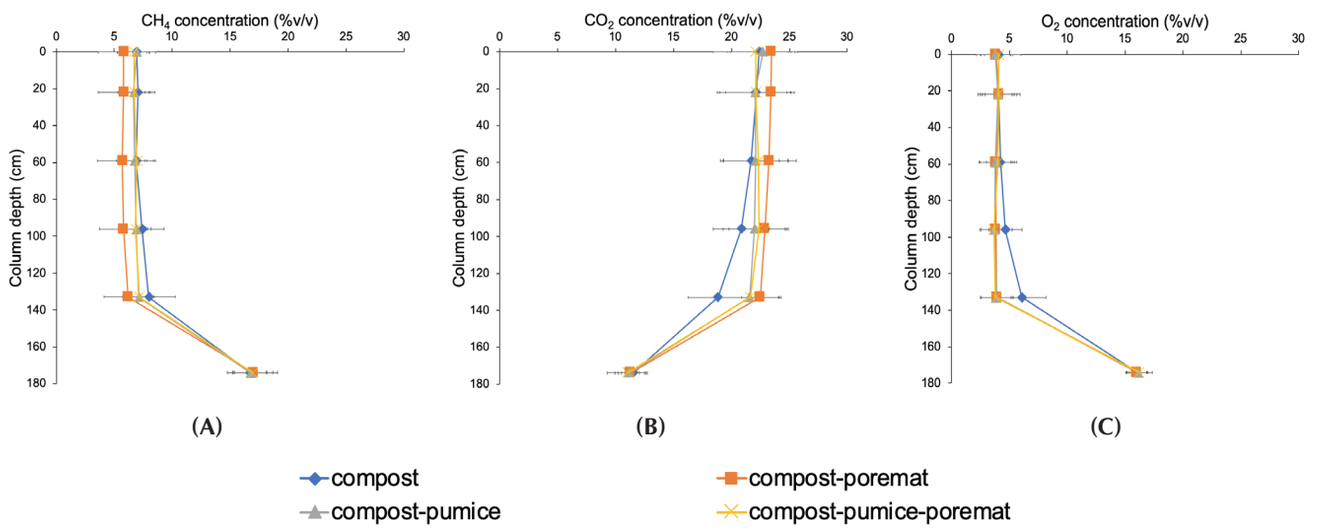

Fig. 3. Profiles of (A) CH_4, (B) CO_2 and (C) O_2 concentrations along the biofilter depths. The error bar indicates standard deviation of gas concentrations.

that O_2 concentrations became limited (<5%) in the biofilter above 1.33 m depth and this might indicate that microbial CH_4 oxidation might be inhibited by the shortage of O_2 above this height. Therefore, a limited CH_4 oxidation was observed in the biofilter above 1.33 m depth.

Methanotrophic activities in biofilter media

At the end of the experiment, 10 g of filter media from the most active CH_4 oxidation zone (1.33-1.74 m depth) in each biofilter was collected to investigate the methanotrophic activities via batch experiments. As shown in Table 3, the filter media with different physiochemical properties exhibited different methanotrophic activities. The highest methanotrophic activities were observed for compost-poremat media at 35.4 µg CH_4/g dried wt/h. Meanwhile, the lowest methanotrophic activities were observed in compost at 22.3 µg CH_4/g dried wt/h. These results imply that the presence of the inorganic biofilter (with high porosity) helped facilitate oxygen transfer into the media while the compost material supplied nutrients for methanotrophic activities. Therefore, the co-existence of organic and inorganic media could benefit CH_4 oxidation. The results obtained from the batch experiment were in agreement with those observed during column operation. During the batch experiments, the CPR trends followed that of the MOR. The CH_4:CO_2 mol ratio observed during batch experiments was found to be 1:1.25-1.43, which suggests that the presence of organic oxidation comes from other organic carbon contained within the biofilter media.

Table 3. Methanotrophic activities of different biofilter materials.

Biofilter media	MOA ($x10^{-3}$ mg/g media/h)	CPR ($x10^{-3}$ mg/g media/h)	CH_4:CO_2 (mol:mol)
Compost	22.3	81.63	1:1.33
Compost-poremat	35.4	121.9	1:1.25
Compost-pumice	25.1	99.1	1:1.43
Compost-pumice-poremat	30.9	109.4	1:1.28

Note: MOR: methane oxidation rate; CPR: carbon dioxide production rate.

Methanotrophic bacterial population

The methanotrophic bacteria population in compost-poremat media, which exhibited the highest MOR in the column experiment and largest methanotroph activities, was investigated. The sequencing of the 16S rRNA genes amplified from the active bacterial communities of the compost-poremat biofilter revealed the presence of microorganisms classified in 21 phyla, 47 classes, 114 orders, 188 families, and 332 genera in total. Among them, they predominantly belonged to Actinobacteria (46.6±1.9%) followed by Proteobacteria (20.5±1.7%) and Firmicutes (18.0±2.3%). The presence of methanotrophs (Fig. 4) included *Methylobacter* and *Methylocaldum*, both classified as Methanotroph type I. Previous research has reported that a high oxygen environment favours the growth for methanotroph type I whereas type II was found to favour methane-rich conditions [19]. There was a similar microbial profile detected in organic and inorganic media. In the compost, *Methylobacter* and *Methylocaldum* accounted for 9.2 and 2.1% of the total bacteria, respectively, and were found to be 8.5 and 2.6% in the poremat media, respectively. These results suggest that the biofilter media provided favourable conditions for Methanotroph type I growth in both organic and inorganic media. The predominance of similar microbial species was also reported in biofilters using a mixture of compost and lava rock and compost and a wood-based biochar mixture [22].

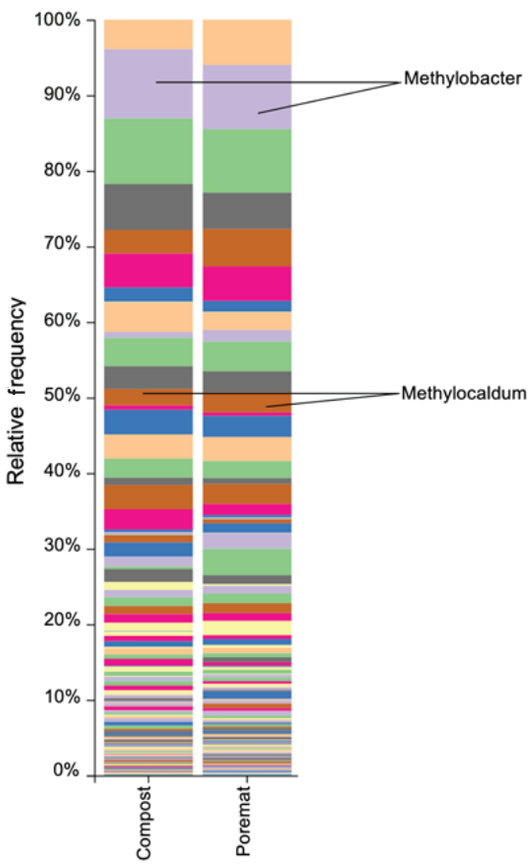

Fig. 4. Bacterial population in compost-poremat media at 1.50 m depth.

N_2O removals in the biofilter

Figure 5 shows the NCR in the biofilters with different media. In the compost biofilter, negative conversion rates were determined that indicated N_2O production during day

39 to day 81, but they became positive or were removed afterwards until the end of experiment. The production of N_2O in the compost biofilter could possibly be provided from a high nitrogen content maintained in the compost biofilter together with the formation of preferential air flow and stagnant zones. These conditions can create zones with low oxygen penetration leading to the development of potentially anoxic/anaerobic microbial metabolic conditions that yield incomplete nitrification. Diffusion limitations in thick biofilms could also create the formation of oxygen-depleted anoxic zones. When nitrification proceeds under oxygen-limiting conditions, ammonia-oxidizing bacteria use NO_2^- as the terminal electron acceptor instead of O_2, which leads to higher N_2O production as a by-product during incomplete nitrification [23]. Similar observations of N_2O production in organic material-based biofilters were also reported [24].

A shift from N_2O production to removal in the compost biofilter occurred simultaneously with an increase in CH_4 oxidation in the same biofilter (Fig. 2). This observation revealed the improvement of aerobic conditions in the compost biofilter over time. Meanwhile, the compost-pumice, compost-poremat, and compost-pumice-poremat biofilters yielded N_2O removal during the entire experimental period (Table 4). The removal of N_2O concentrations in these biofilters suggest that the use of organic and inorganic materials could promote oxygen availability to achieve complete nitrification in the biofilter media. In previous research, it was reported that N_2O emissions from biofilters containing inorganic (pumice) media was negligible due to a much lower microbial N content in compost compared to that in compost-based biofilters [20].

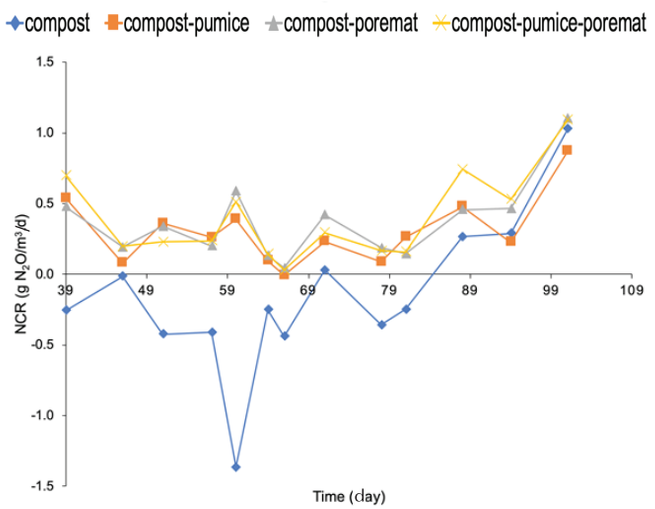

Fig. 5. N_2O emission rates in different filter materials.

Table 4. NCR (g $N_2O/m^3/d$) in biofilters at different operation period.

Biofilter media	day 39-60	day 64-88	day 94-101	overall
Compost	-0.5 (0.5)^{a,b,c}	-0.2 (0.3)	0.7 (0.5)	-0.2 (0.5)^{a,b,c}
Compost-poremat	0.4 (0.2)^{a}	0.2 (0.2)	0.8 (0.5)	0.4 (0.3)^{a}
Compost-pumice	0.3 (0.2)^{b}	0.2 (0.2)	0.5 (0.5)	0.3 (0.2)^{b}
Compost-pumice-poremat	0.4 (0.2)^{c}	0.3 (0.3)	0.8 (0.4)	0.4 (0.3)^{c}

Note: the numbers show average (SD) values; a, b, and c significant difference ($p<0.05$) between the compost biofilter and other biofilters respectively.

Conclusions

The compost-poremat biofilter was found to be the most effective filter material providing an average methane oxidation rate of 70.1 g $CH_4/m^3/d$ over a 101-day period. The high porosity of the biofilter media containing a mixture of organic and inorganic media allowed sufficient oxygen transfer for methane oxidation, even at the bottom of biofilter media bed of 1.5 m depth. High methanotrophic activities and the presence of type I methanotrophs confirmed active methane oxidation under oxygen-rich conditions in the biofilter media. N_2O was also found to be removed from the biofilter containing inorganic media, while it was mainly produced in the compost biofilter.

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COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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