



Greenhouse Gas Emissions From Biofilters for Composting Exhaust Ammonia Removal

Bin Shang*, Tanlong Zhou, Xiuping Tao and Yongxing Chen

Key Laboratory of Energy Conservation and Waste Management of Agricultural Structures, MARA, Institute of Environment and Sustainable Development in Agriculture, Chinese Academy of Agricultural Sciences, Beijing, China

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*Correspondence:

Bin Shang
shangbin@caas.cn

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Emissions of odorous compounds, such as ammonia (NH_3), from composting have negative agronomic and environmental impacts. A biofilter is widely used for NH_3 removal, with one of its potential detrimental by-products being nitrous oxide (N_2O), which is a higher warming potential greenhouse gas (GHG). The aim of the study was to evaluate the effect of empty bed retention time (EBRT) on GHG emissions from biofilters for removing NH_3 from composting. Composting experimental trials lasted 6 weeks, and composting materials were mixtures of dead pigs and manure. Three groups of biofilters with 1.2 m-height, 0.3 m-inner diameter, and 1.0 m media depth were conducted with EBRT of 30, 60, and 100s, respectively. Each treatment was performed in triplicate, and the gas was monitored using the dynamic emission vessel method. The Spearman's correlation analysis showed a significantly positive correlation between inlet concentrations (ICs) of NH_3 and increased N_2O concentrations: $\rho = 0.707, 0.762, \text{ and } 0.607$ with $p \leq 0.0001$ for biofilters with EBRT of 30, 60, and 100s, respectively. The fraction of $\text{NH}_3\text{-N}$ denitrified into $\text{N}_2\text{O-N}$ in biofilters with EBRT of 60 and 100s was higher than that with EBRT of 30s. The total global warming potential (GWP) increased by 126%, 162%, and 144% for biofilters with EBRT of 30, 60, and 100s, respectively. These results indicated that biofilters with longer EBRT will lead to higher GWP production. Future research on odorous mitigation for composting with biofilters should focus more on greenhouse gas emissions.

Keywords: empty bed retention time, ammonia biofilter, greenhouse gases, nitrous oxide, waste management

INTRODUCTION

Composting has been applied worldwide as an environmentally friendly and cost-effective method for sanitation and recycling animal waste (Loyon, 2017; Zheng et al., 2020). However, one major complication during composting is the odor emission (Cheng and Hu, 2010; Huang et al., 2012; Paulot, et al., 2014), which not only poses a problem to the general public and environmental health but also causes adverse effects on vegetation surrounding the composting plants (Han et al., 2019). Ammonia (NH_3) is considered the major contributor to odor from composting (Zhang et al., 2016; Zhu et al., 2016), and the precursors of particulate matter can be produced by the reactions of NH_3 with sulfuric and nitric acid aerosols (Pinder, et al., 2007; Renner and Wolke, 2010; Kong et al., 2020).

Biofilters are widely used to reduce NH_3 emissions from composting (Turan et al., 2009; Mudliar et al., 2010; Janni et al., 2014). Many studies have focused on the physical, chemical, and biological

TABLE 1 | Experimental performance of biofilters.

Treatment	Biofilter	Volume per biofilter (m ³)	Empty bed retention time (EBRT, s)
1	1,2, and 3	0.071	30
2	4,5, and 6	0.071	60
3	7,8, and 9	0.071	100

parameters influencing the biofiltration process (Park, et al., 2002; Yuan et al., 2019). A number of studies have shown that nitrous oxide (N₂O) generation in biofilters is often accompanied by NH₃ removal (Maia et al., 2012b; Yang et al., 2014a; Kong et al., 2020). Akdeniz and Janni (2012) observed that N₂O generation ranged from -29.2% to 4.0% for a flat-bed biofilter with an empty bed retention time (EBRT) of 5s. Clemens and Cuhls (2003) observed that approximately 26% of NH₃-N entered in the biofilters is converted into N₂O-N. N₂O emissions from biofilters can be affected by several factors, such as inlet NH₃ concentration, temperature, moisture content, and pH value (Maia et al., 2012b; Yang et al., 2014a; Yang et al., 2014b; Dumont et al., 2014). High moisture content can cause regional anaerobic zones and increase the microbial activity, which favors N₂O emission (Yang et al., 2014a). Compared to a high pH value (8.0–9.5), a low pH value (4.5–6.0) of the media can inhibit the generation of N₂O reductase and reduce the emission of N₂O (Yang et al., 2014b). N₂O from biofilters was correlated significantly with the NH₃ in the biofilters (Clemens and Cuhls, 2003). EBRT can affect microorganism absorption and the conversion process of NH₃ in the biofilters (Shang et al., 2020). Shorter EBRT means faster gas flow velocity, which can change the oxygen (O₂) gradients in the biofilter media and can lead to changes in denitrification (Maia, et al., 2012b). In addition, higher air flow can increase the emission rate of gas from composting. EBRT is one of the key parameters of biofilters for NH₃ removal (Liu et al., 2017); however, few studies have investigated the effects of EBRT on emissions of N₂O. A better understanding of the effects of EBRT on the generation and emission of N₂O needs to be elucidated. In this study, the effects of EBRT on greenhouse gas (N₂O and methane, CH₄) emissions from pilot-scale biofilter systems were studied. The results can provide a promising tool for greenhouse gas reduction from full-scale biofilters.

CH₄ is another important greenhouse gas emitted during composting (Zhu-Barker, et al., 2017). Biofilters also can be used to reduce CH₄ emissions (Haubrichs and Widmann, 2006). Many studies performed on CH₄ biofiltration utilized an EBRT of at least 4 min (La et al., 2018), which posed obstacles to the biofilter application. Little information is available in the literature about CH₄ reduction capabilities during NH₃ biofiltration.

Global warming potential for N₂O and CH₄ is 296 and 23 times higher than that of carbon dioxide (CO₂), respectively (Pachauri et al., 2014). It is very important to examine the generation of greenhouse gas due to odor treatment by biofilters. Thus, the objectives of this study were 1) to investigate the emissions of N₂O from biofilters with different EBRT and 2) to assess the greenhouse gas (N₂O and CH₄) emissions from biofilters for composting NH₃ removal.

MATERIALS AND METHODS

Experiment Materials

This study was conducted in the Beijing Anding pig farm, located in Daxing District, Beijing, China (39°62'N, 116°50'E). The composting materials and composting process have been described by Shang et al. (2020).

The mature compost, composted for about 4 months, was used as the medium material. Before the experiment, the mature compost was inoculated with activated sludge from the aerobic fermentation plant for wastewater treatment in the pig farm, and the water content of packing materials at the beginning of composting was adjusted to 57.4 ± 2.5%, according to Akdeniz and Janni (2012). The mature compost used for biofilter media had total carbon (C) and nitrogen (N) contents of 30.8 ± 2.8% and 2.9 ± 0.7%, respectively, with a pH of 7.0.

Biofilter Design and Operations, Gas Sampling, and Analytical Method

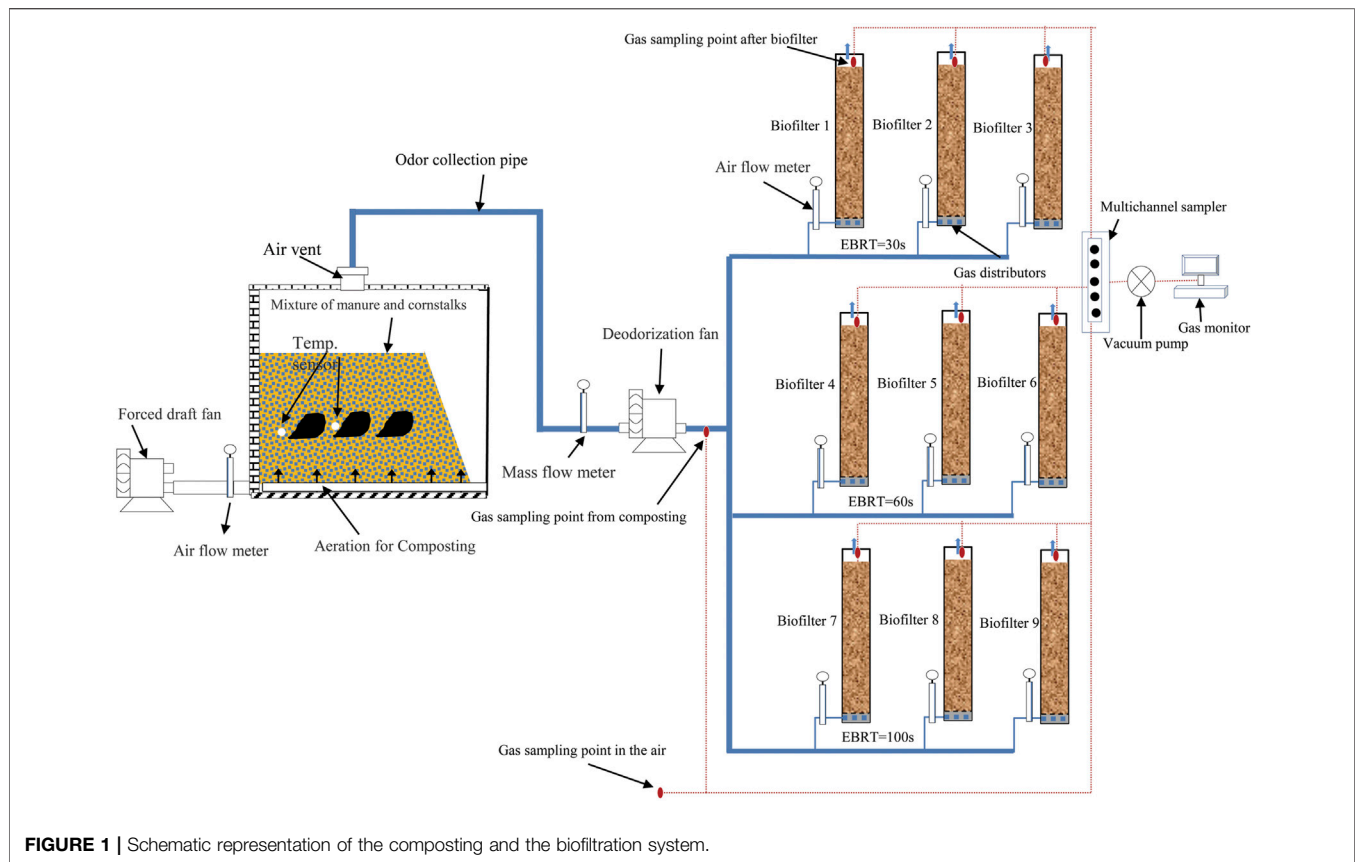
The biofilters were constructed with circular unplasticized polyvinyl chloride (UPVC) pipes. The dimensions were 1.2 m (height) and 0.3 m (inner diameter), with a 1.0 m depth of media (corresponding to a bed material volume $V = 0.07 \text{ m}^3$). Nine biofilters were divided into three treatments, and different EBRT (30s, 60s, and 100s) were set according to previous studies (Pagans et al., 2005). Each treatment was conducted with three replicates. $\text{EBRT} = V/Q$, where Q is the air flow rate in $\text{m}^3 \cdot \text{s}^{-1}$. The experimental arrangements are shown in **Table 1**. NH₃, CH₄, and N₂O concentrations of air outside (1 sampling point), a gas inlet of biofilters (3 sampling points), and a gas outlet of biofilters (9 sampling points) were measured continuously and simultaneously using a photoacoustic multigas analyzer (model Innova 1412i, LumaSense Technologies, Ballerup, Denmark) every day. The deodorization system was constructed, as previously described (Shang et al., 2020), and the schematic is shown in **Figure 1**. The experiment corresponding to the composting period was carried out for 42 days. During the experiment, no water was supplied to the media of biofilters.

Data Analyses

All data were analyzed by Microsoft Excel 2016 and SPSS 22. The variables were tested for significance using Spearman's correlation coefficient. The ratios of gas concentrations in outlet and inlet biofilters were calculated (Dumont, 2018). A ration >1 indicates gas formation in the biofilter, and a ration <1 indicates gas depletion in the biofilters. The t -test (one side $p < 0.05$) was used to determine if there was a significant deviation from 1.

Global Warming Potential Calculations

Global warming potential (GWP) was quantified as CO₂ equivalent with a 100-year timescale: 1 kg CH₄ and N₂O emitted are equivalent to 23 and 298 kg CO₂, respectively (Pachauri et al., 2014). The CH₄, N₂O, and GHG emissions (in kg eqCO₂) were calculated by multiplying the aerobic rates, the concentration, and the GWP factor during the whole experiment which lasted for 42 days. In this study, the GWP of



CO₂ was not included because it is not considered GHG of agriculture (Buendia et al., 2019).

RESULTS AND DISCUSSION

Emission of Nitrous Oxide From Biofilters

NH₃, N₂O, and CH₄ concentrations in the air during the composting period were 5.6 ± 2.5 , 1.0 ± 1.6 , and 3.2 ± 1.6 mg m⁻³, respectively. The NH₃ removal efficiencies have been described by Shang et al. (2020). The daily mean concentration of N₂O from composting ranged between 1.5 and 11.5 mg m⁻³, while the daily mean N₂O concentrations from biofilters were about around 1.0 mg m⁻³ (Figure 2). The large variation in N₂O production was due to the different concentrations of NH₃ and N₂O from composting. The N₂O concentrations at the biofilters' outlet were in the range of 20%–1250%, 23%–1652%, and 3–1352% higher than those at the biofilters' inlet for EBRT of 30, 60, and 100s. The outlet concentrations of N₂O of biofilters with EBRT of 100 and 60s were higher than those with EBRT of 30s (Figure 3). But there are no significant differences between the biofilters with different EBRT.

With biofiltration systems, about half of the inlet NH₃ is converted to nitrites or nitrates, and the other half is absorbed into the water as ammonium (Ottosen et al., 2011; Yasuda et al., 2017). Both ammonia oxidizer and denitrifier can produce N₂O

in a biofilter, but the majority of N₂O was generated from denitrification (Kong et al., 2020). Maia et al. (2012a) pointed that higher EBRT may result in non-uniform oxygen distribution, which favors conditions for denitrification to generate N₂O. In this study, the higher EBRT means a slower air flow rate, and the increasing N₂O concentration from biofilters with increasing EBRT may result from more denitrification. In addition, when the air continuously goes through media in the biofilter, the higher EBRT implies that there is more content time of air and media, which can lead to high NH₃-sorption, which favors the occurrence of nitrification–denitrification (Maia et al., 2012a). In this study, N₂O concentrations were found to be higher, up to 16 times, than inlet concentrations, which may be due to the longer EBRT and higher inlet concentrations of NH₃ (Dumont et al., 2014; Liu et al., 2017).

The inlet concentrations, inlet loads, and elimination capacities of NH₃ may affect N₂O generation. The Spearman's correlation analysis showed a strong positive correlation between inlet NH₃ concentrations and increased N₂O concentrations: $\rho = 0.707, 0.762, \text{ and } 0.607$ with ($p \leq 0.0001$) for EBRT of 30, 60s and 100s, respectively. Inlet loads of NH₃ and increased N₂O concentrations were positively correlated: $\rho = 0.685, 0.750, \text{ and } 0.579$ ($p \leq 0.0001$) for EBRT of 30, 60s, and 100s, respectively. Elimination capacities of NH₃ and increased N₂O concentrations were also positively correlated: $\rho = 0.706, 0.761, \text{ and } 0.602$ with $p \leq 0.0001$ for EBRT 30, 60, and 100s, respectively. In this study, the inlet NH₃ concentrations were between 12 and 447 mg m⁻³

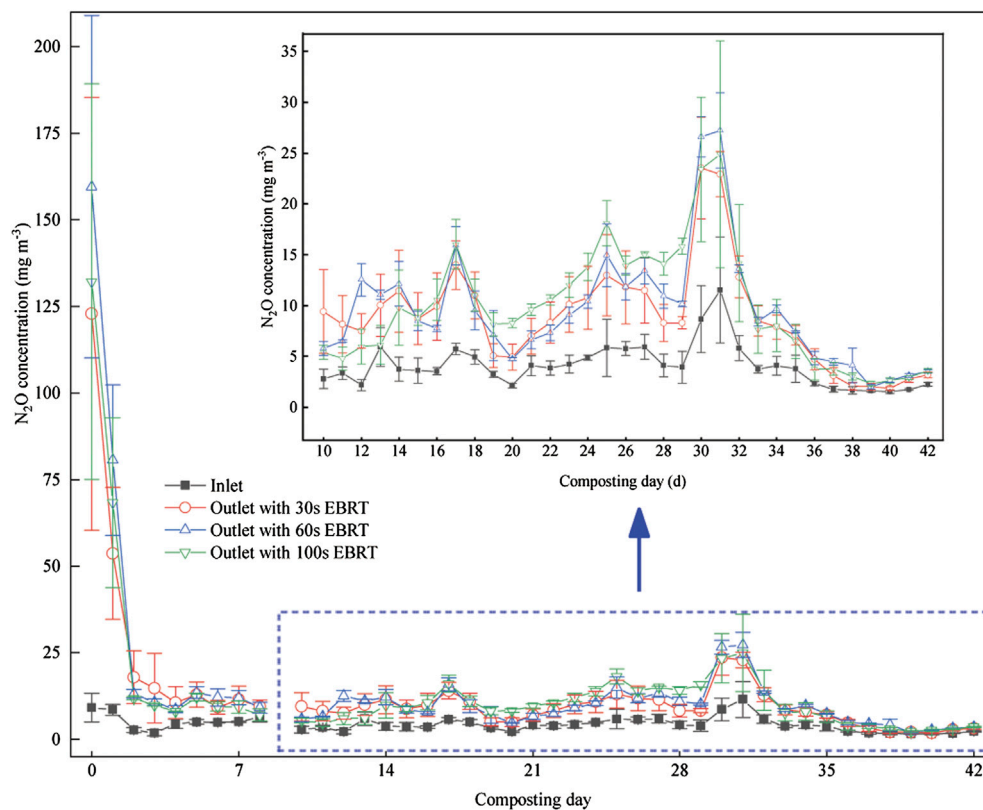


FIGURE 2 | Daily N_2O concentrations (mean \pm SE) at the biofilter inlet (IC) and outlet (OC).

due to different composting processes, while the overall removal efficiencies were 85.4%, 88.7%, and 89.0% for EBRTs of 30, 60, and 100s, respectively. The elimination capacities of NH_3 have been discussed by Shang et al. (2020). NH_3 removal was considered the net source of N_2O in biofilters (Maia et al., 2012a). N_2O is considered normal at biofilters treating NH_3 -containing air, being a byproduct of nitrification and denitrification, and the pathways of N_2O formation reported in the literature, however, are complex. Kong et al. (2020) found that the inlet NH_3 concentration can affect the nitrification process and the substrate availability for denitrification. In the present study, high inlet loads of NH_3 promoted N_2O generation, which were consistent with the other study (Kong, et al., 2020); the denitrification is the main pathway for N_2O formation.

Fraction of NH_3 -N Denitrified Into N_2O -N

In the present study, there were significant differences in N_2O emissions between biofilters with different EBRT due to different inlet loads of NH_3 resulting from different EBRT. The fractions of NH_3 -N converted into N_2O -N were 4.6%, 5.6%, and 5.1% for biofilters with EBRT of 30, 60, and 100s, respectively. The results were consistent with other studies (Table 2). Denitrification is considered the main source of N_2O emission while nitrification is a trigger (Yang et al., 2014a; Dumont et al., 2014; Kong et al., 2020), and the presence of oxygen can inhibit the denitrification process. In this study, low EBRT means high gas flow rate, which

results in more oxygen penetration into the media of the biofilter, further reducing the denitrification rates. Except for the EBRT, some other factors, such as the kind of media and moisture content may also affect N_2O generation in biofilters. In Maia et al. (2012a), the compost of horse manure, cattle manure, chicken waste, woodchips, sawdust, and other materials were used as media for NH_3 removal, fraction of NH_3 -N denitrified into N_2O -N was 14% and 19% for one biofilter and two other biofilters, respectively. The results of Yasuda et al. (2009) confirmed that NH_3 could be treated by biofilter with rockwool mixture without an extra increase of N_2O . The percentage of N_2O reduction efficiency ranging from 0.13 to 0.73% was found by Akdeniz (et al., 2011), who studied the removal of NH_3 and N_2O using biofilters with lava rock as media at 5s EBRT. The part of NH_3 -N converted into N_2O was estimated to range from 10% to 40% (Dumont et al., 2014). Yang et al. (2014a) found that there was a slight increase in N_2O when the media moisture content increased from 35 to 55%, but further increasing the moisture content to 63% triggered N_2O generation rapidly. Yasuda et al. (2009) observed a higher N_2O generation when the moisture content ranged from 65% to 52%; when the moisture content decreased to 48%, the N_2O net generation decreased to nearly zero; when the moisture content decreased from 44% to 13%, N_2O generation decreased. A moisture content of about 50% is recommended for efficient NH_3 removal and less N_2O generation. Yang et al.

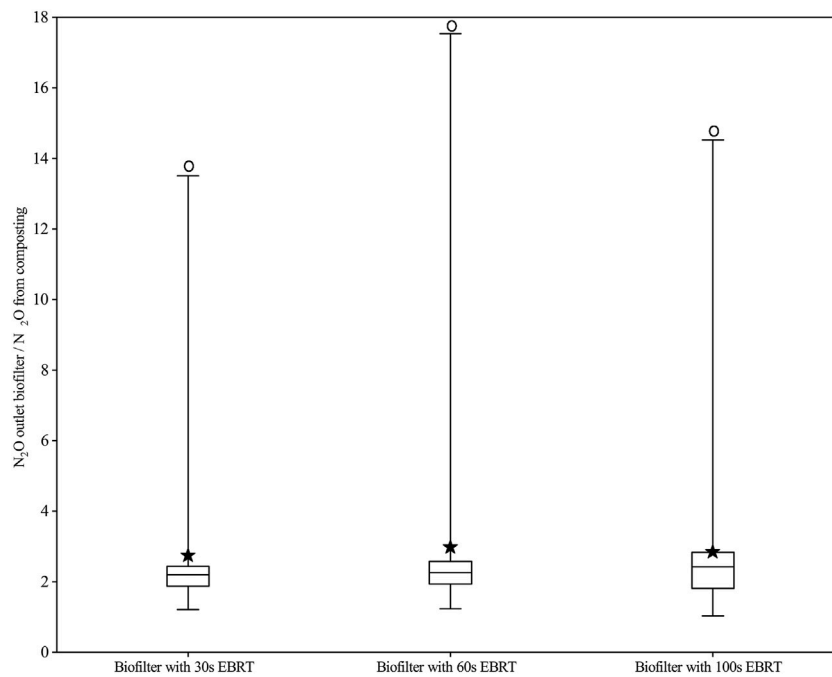


FIGURE 3 | Boxplot: Ratio of N_2O concentration of the outlet and inlet at different biofilters (Box border means 25 and 75% percentile, the solid line in the box means the median value, the solid pentacle means the mean value, whisker means the maximum and minimum values, and the white circle means significant differences between the inlet and outlet).

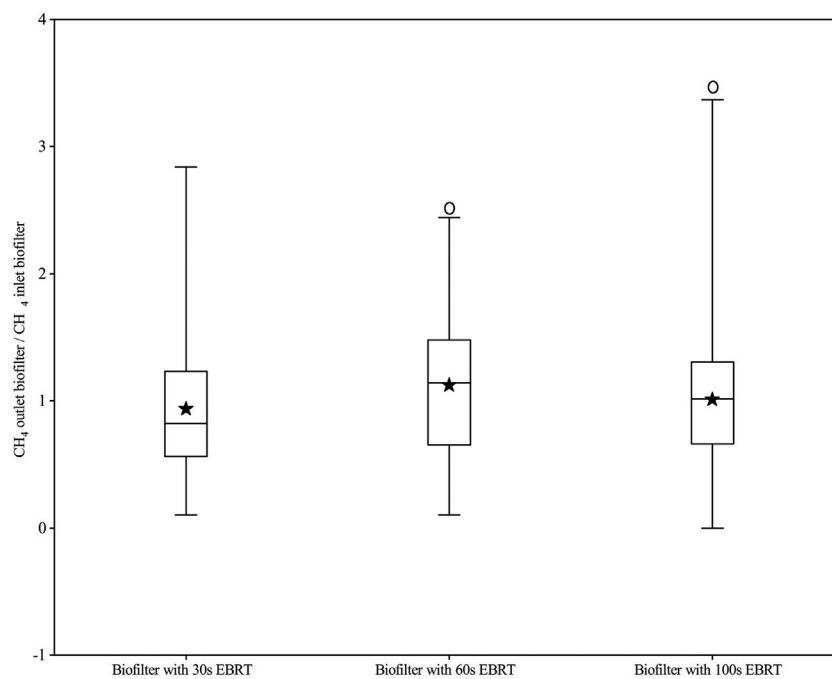


FIGURE 4 | Boxplot: Ratio of CH_4 concentrations of the outlet and inlet at different biofilters (Box border means 25 and 75% percentile, the solid line in the box means the median value, a solid pentacle means the mean value, a whisker means the maximum and the minimum values, and the white circle means significant differences between the inlet and outlet).

TABLE 2 | Literature overview of N₂O generations in biofilters.

Biofilter media material	Empty bed retention time (s)	Experiment periods (d)	Inlet NH ₃ load (g.m ⁻³ .h ⁻¹)	Inlet NH ₃ concentration (mg.m ⁻³)	N ₂ O-N emission (g.m ⁻³ .h ⁻¹)	Inlet NH ₃ -N to N ₂ O-N (%)	Reference
Woodchips inoculated with activated sludge	12	124	2.4–3.0	8–12	Maximum was around 1	10–40	Dumont, et al. (2014)
Compost (mixture of horse manure, cattle manure, chicken waste, woodchips, sawdust, and others)	25	100	0.99	17.5	0.2	/ ^a	Maia, et al. (2012b)
Compost (mixture of horse manure, cattle manure, chicken waste, woodchips, sawdust, and others)	20	21	0.47	11.2	/	14–19	Maia, et al. (2012a)
Mixture of wood chip and compost	34	22–35	5.24	31	/	1.9–2.3	Yang, et al. (2014a)
Mixture of pine wood chips and peat soil	42	/	1.5–3	13.7–26.6	/	5.2–14.8	Kong, et al. (2020)
Woodchips	1.4–3.3	/	8.7–67	19–86	0.2–0.5	1.3–21	Melse and Hol, (2017)
Mature compost	30–100	42	0.5–53.6	13–447	0.3–1.6	4.6–5.6	The present study

^a/ⁿ means no data.

TABLE 3 | Cumulative nitrous oxide (N₂O) and methane (CH₄) emissions from biofilters used for composting exhaust ammonia (NH₃) removal (values are means ± SE, n = 3)^a.

	Biofilter inlet			Biofilter outlet		
	CH ₄ (kg.m ⁻³ biofilter)	N ₂ O (kg.m ⁻³ biofilter)	Total GWP [kg (CO ₂ eq.) m ⁻³ biofilter] ^b	CH ₄ (kg.m ⁻³ biofilter)	N ₂ O (kg.m ⁻³ biofilter)	Total GWP [kg (CO ₂ eq.) m ⁻³ biofilter]
Biofilter with EBRT 30s	3.8 (0.5)	0.52 (0.09)	240 (40)	3.2 (0.6)	1.6 (0.4)	544 (131)
Biofilter with EBRT 60s	1.9 (0.5)	0.26 (0.05)	120 (20)	2.0 (0.1)	0.9 (0.1)	314 (33)
Biofilter with EBRT 100s	1.1 (0.2)	0.16 (0.03)	72 (12)	1.1 (0.3)	0.5 (0.1)	176 (39)

^aThe nitrous oxide (N₂O) and methane (CH₄) emissions were the cumulative amount emitted from biofilters during the whole experiment which lasted for 42 days.

^bGWP_{CH4} = 23 and GWP_{N2O} = 296 (Pachauri et al., 2014).

(2014b) showed that N₂O concentrations ranged from 0.1 to 0.4 ppm with a pH of 8.0, and the acidified biofilters showed higher N₂O concentrations than alkalized biofilters. In the present study, the values of the moisture content and pH of media change very little through the experiment. During the experiment, the pH of media was maintained at around seven for all biofilters, while the average moisture contents of media (mean ± SE, n = 12) were 48.3 ± 0.4, 48.5 ± 0.5, and 49.1 ± 0.4 for the biofilters with EBRT of 30, 60, and 100s, respectively.

Emissions of CH₄ From Biofilters

The daily mean concentrations of CH₄ from composting ranged between 5 and 149 mg m⁻³, while the concentrations from biofilters ranged between 2 and 241 mg m⁻³. The average daily REs of CH₄ were 6.5%, -10.5%, and -6.0% for EBRT 30, 60s, and 100s, respectively. Lim et al. (2012) reported the CH₄ reductions ranged from 0.1% to 1.9% in biofilters with 0.3s–0.6s EBRT. Fedrizzi et al. (2018) attained nearly 100% CH₄ 100% removal efficiencies with 756s EBRT, so enough EBRT is needed for CH₄ removal. Akdeniz et al. (2011) achieved the removal efficiencies of 6.9%–25% for CH₄ by using the pilot-

scale biofilters with lava rock media at the high moisture levels and low inlet CH₄ concentrations (90% moisture content and average of 31.0 ppm of inlet CH₄ concentration), while the EBRT was just 5s. Melse and van der Werf (2005) reported that the biological conversion of CH₄ in a biofilter is a slow process due to the low water solubility of methane (Henry's law constant = 1.5 × 10⁻³ M atm⁻¹). Biofilters with EBRT of 83–199s cannot reduce concentrations of CH₄ emitted from municipal solid waste composting, which has been reported by Clemens and Chuls (2003). Yasuda et al. (2009) also pointed out that several microbial CH₄ production and consumption reactions occurred, but a large portion of the CH₄ seemed to pass straightly through the biofilter with the EBRT of 100–200s (2014; Devanny, et al., 1999). In the present study, negligible CH₄ removal and no significant difference were found among three EBRTs, (**Figure 4**) and the result was similar to that of Akdeniz et al. (2011), Yasuda et al. (2009), and Lim et al. (2012). The possible reason is that the EBRT in this study is not enough for the transfer of CH₄ from the gas phase to the biofilm phase (Melse and Van der Werf, 2005). On the other hand, although the anaerobic methanogens were not tested in this study, the retention time for anaerobic suspended growth in

biofilters was about 50 days (Maia et al., 2012b); the slow acclimation of methanogens in the biofilter will lead to low CH₄ removal efficiency.

Global Warming Potential Emissions

Based on the measurements, the total global warming potential (GWP) emissions were calculated. **Table 3** shows the GWP loading rate before and after passing through the biofilter for all the biofilters. The total GWP emission increased by 126%, 162%, and 144% for biofilters with EBRT values of 30, 60, and 100s, respectively. Significant differences were found in the total CO₂-eq emissions from the biofilters. A lower percentage of GWP was emitted from the biofilter with 30s EBRT than in biofilters with 60 and 100s EBRT because the fraction of NH₃-N denitrified into N₂O-N is lower for the biofilter with 30s EBRT than that of 60 and 100s EBRT. These results showed that prolonged EBRT can improve RE of NH₃ but increase the GWP emissions accordingly. Melse and Hol (2017) evaluated three kinds of biofiltration of exhaust air from animal houses in which the total GWP emissions increased by 60%, 45%, and 0 in biofilters with EBRT of 1.4, 2.6, and 3.3s, respectively. In their experiment, the average inlet concentrations of NH₃ were 66, 10, and 15 ppm, respectively, and the removal efficiencies were 74%, 42%, and 38%, respectively. In the present study, the average inlet NH₃ concentrations were 124–163 mg m⁻³, and the removal efficiencies were 82%–89%; both average inlet NH₃ concentrations and removal efficiencies of biofilters were higher than Melse and Hol (2017).

With regard to GHG emissions, it can be concluded that the lower EBRT is more suitable for biofilter systems of composting, and the parameters of the control process, such as pH, temperature, dissolved oxygen, and water content, will be useful to prevent N₂O formation and guarantee a good NH₃ removal efficiency.

CONCLUSION

The increased N₂O concentrations from the biofilter were strongly and positively correlated with the elimination

capacities of NH₃. The total GWP emission increased by 54%, 62%, and 61% for biofilters with 30, 60, and 100s EBRT, respectively. The total GWP emission from biofilters increases by over 50% compared to a composting system without biofilters. More NH₃ converted into N₂O due to higher EBRT suggested that lower EBRT is useful to prevent GHG from biofilters.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**; further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

BS: conceptualization, data curation, formal analysis, funding acquisition, writing—original draft, and writing—review and editing. TZ: conceptualization, data curation, formal analysis, and writing—review and editing. XT: formal analysis, investigation, methodology, and writing—review and editing. YC: investigation, validation, and writing—review and editing.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fbioe.2022.918365/full#supplementary-material>

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