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Ammonia and Greenhouse Gas Emissions from Organic Manure Composting: The Effect of Membrane Cover

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Abstract: The current scientific literature predominantly focuses on pilot-scale studies concerning the effectiveness of membrane covers in reducing gas emissions during cattle manure composting. Our study centers on the application of a leading market commercial membrane cover (ePTE-TEX$_{comm}$) and a locally manufactured one (ProfiCover®) at industrial processing levels, evaluating their efficacy in mitigating gas emission during the fifth day of the thermophilic phase. Taking into account material inhomogeneities, work environment impact, and efficiency, the results are characteristic of industrial-scale processes rarely discussed in the scientific literature. Our results, obtained with a portable gas sampler and FTIR spectroscopy measurements using corresponding standards, indicate that ePTE-TEX$_{comm}$ manifested a reduction of 90.8% for NH_3 and 59.6% for CO_2. CH_4 emissions increased, suggesting their potential entrapment. N_2O and propane equivalent experienced reductions of 23.1% and 44.8%, respectively. On the other hand, ProfiCover® presented emission reductions for NH_3 and CO_2 of 93.3% and 85.9%, respectively. CH_4, contrasting with ePTE-TEXc$_{omm}$, showed a significant reduction of 55.6%. N_2O and propane equivalent followed with reductions of 56.7% and 84.5%, respectively. All of this divergence in performance implies a potential trade-off in emission reduction efficacy between the covers. Knowledge sharing between researchers and industry partners is key to translating these technologies into widespread adoption.

Keywords: ammonia; manure compost; ePTFE membrane cover; gaseous emission; ammonia; carbon dioxide; methane; propane equivalent

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1. Introduction

Organic manure composting is a common practice in agriculture for managing manure and producing nutrient-rich soil amendments. However, this process can lead to the emission of greenhouse gases (GHGs) such as carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O), which contribute to climate change [1]. Additionally, the release of ammonia (NH_3) during composting can have negative impacts on air quality and human health [2]. The composting process consists of different stages: the so-called thermophilic phase, occurring after a short initial period, when the microbes multiply rapidly, heating up the substrate until the point where the easily degradable substances are depleted and the process moves to the mesophilic phase, followed by curing, when mineralization and humification occurs. One of the critical phases of composting is the thermophilic phase, with a temperature range over 45 °C. In this phase, the highest amounts of emissions are observed, starting around the fifth day from the start of composting. An assessment of three main methods of open manure composting, i.e., aerobic storage, extensive composting, and intensive composting, indicated that intensive composting had the highest NH_3 emissions, while the nitrogen content in extensive composting was lost in the form of, more favorably, N_2, and, less favorably, N_2O, due to nitrification–denitrification and the lack of oxygen control in all three cases, which led to largely anaerobic beds, resulting in uncontrolled NH_3, N_2O, and CH_4 emissions [3–5].

Mitigating NH_3 and GHG emissions from composting is crucial for environmental sustainability. To address these environmental concerns, researchers have explored various strategies to reduce GHG emissions and NH_3 volatilization during composting. One approach that has shown promise in reducing these emissions is the use of semi-permeable membrane covers. Membrane covers placed over composting piles act as impermeable barriers for certain components, minimizing gas exchange between the composting material and the atmosphere [1,6–8]. Studies have shown that semi-permeable membrane covers can effectively reduce GHG emissions [1,9–12] and the volatilization of NH_3 during composting [13]. Membrane covering has been demonstrated to be more effective than biomass covering, while also providing a physical barrier of pathogens during bacterial community succession [11,12,14]. However, these approaches need to address the reduced O_2 concentrations in composting piles, usually solved by forced aeration, thus creating aerated static pile (ASP) systems.

Reducing NH_3 emissions is important for capturing the nitrogen content in compost and improving air quality and human well-being, as airborne NH_3 can cause severe adverse environmental and human health effects. NH_3 at 30 parts per million (pm) (21 mg/m^3) can cause discomfort, irritation, or certain asymptomatic non-sensory effects in humans; human exposure to NH_3 at 110–220 ppm (77–154 mg/m^3) can result in irreversible and lasting adverse health effects; and at 390–2700 ppm (272–1880 mg/m^3), it can cause life-threatening health effects or death [15]. Moreover, high levels of atmospheric NH_3 can contribute to the formation of particulate matter and smog [2].

Wang et al. [16] conducted a laboratory study on pig manure composting and demonstrated that the use of membrane covers significantly reduced NH_3 volatilization compared to uncovered compost piles. Additionally, semi-permeable membrane covers may help to retain moisture in composting piles, promoting optimal microbial activity and nutrient retention [17]. Nonetheless, while the use of membrane covers in composting shows promise, further research is needed to fully understand their effectiveness and optimize their implementation [18]. Factors such as the type of membrane material, design, and management practices need to be considered to maximize their benefits [18]. Additionally, the long-term effects of membrane covers on compost quality, nutrient cycling, and overall sustainability need to be evaluated [19]. The existing literature highlights a significant knowledge gap concerning the effectiveness of membrane covers in mitigating NH_3 and GHG emissions during the composting process of organic manure, particularly at industrial-scale facilities. The predominant focus has been on pilot-scale studies, with a lack of comprehensive research at an industrial scale exploring different types of composting materials and varied composting conditions, such as the composting technology itself, the treated feedstock mass, and operational patterns. This gap potentially hinders the optimization of membrane cover technologies for larger-scale applications, which is necessary for significantly reducing emissions and improving air quality and environmental sustainability in agricultural waste management, especially manure composting.

This study aims to contribute to the existing knowledge by investigating the effect of membrane covers on NH_3 and GHG emissions during organic manure composting and providing recommendations for their practical implementation in agricultural systems. The primary aim of this investigation is to evaluate the emissions of NH_3, GHGs, and volatile organic compounds (VOCs) during the composting process of organic manure. Specifically, the study centers on the application of a leading, market-expanded polytetrafluoroethylene (ePTFE) membrane cover (ePTE-TEX$_{comm}$) and a membrane cover manufactured locally, also from ePTFE (Profikomp Environmental Technologies Inc., Gödöllő, Hungary), ProfiCover®, evaluating the efficiency of the two semi-permeable membrane covers in mitigating gas emissions during the thermophilic phase of composting. This phase is characterized by a temperature range over 45 °C, typically occurring from the fifth day of the composting process, and has crucial importance for biomass degradation, the means of hygienization with the highest amount of gas emissions.

This study aims to (a) understand the effectiveness of membrane cover in reducing gas emissions, (b) evaluate the difference in emissions between covered and uncovered compost piles, and (c) explore the influence of membrane cover on gas emissions inside compost piles and nitrogen retention. The investigation is confined to the evaluation of a semi-permeable membrane cover and its impact on gas emissions compared to uncovered compost piles. A unique feature of this study is that it describes industrial-scale processes carried out on a dairy farm in Hungary. The composting plant processes cattle manure and produces compost to be used in the local fields as a soil conditioner and also produces certified bagged fertilizer products on demand. This study endeavors to fill the identified research gaps by providing a thorough understanding of the impact of membrane covers on gas emissions during composting at an industrial scale. The insights gleaned from this research could contribute to the development of effective strategies for emissions reduction, thereby aiding in the mitigation of the environmental impact of organic manure composting. Moreover, the findings could provide valuable recommendations for the practical implementation of membrane cover technology in agricultural waste management systems, promoting environmental sustainability. The measurements were carried out on the day of peak NH_3 production, established based on extensive operational experience gained in the given treatment plant. Limitations of this include the specificity of the composting unit design and the singular focus on the thermophilic phase of composting.

2. Materials and Methods

2.1. Composting Units under Study

The composting units under study, located near Orosháza (Békés County, Hungary), were membrane-covered side-walled ASP systems covered with either ePTE-TEX$_{comm}$ or ProfiCover® membranes and an additional, uncovered ASP unit (Figure 1). The composting plant has a total of 6 treatment units (Figure 1a) and a maximum yearly capacity of 15 thousand tons, from which 3 treatment units were set up in the experiment in the same way, with freshly sourced manure from the monthly manure removal from the barns. All the composting units were of industrial scale with dimensions of 25 m in length and 8 m in width (Figure 1). The experiments were performed with approximately 300 m^3, i.e., 500 tons of manure biomass in each ASP. The widely used 18 min off and 2 min on aeration cycle was applied throughout the entire treatment process, resulting in 6 min/hr (2.4 hr/day) air supplied to the pile, surpassing the average theoretical stoichiometric air demand of feedstock by not more than 50% for cost-efficient operation. This operational approach was completely appropriate and in line with the operation manual of the technology supplier. During sampling, however, a manual aeration schedule was used. Consequently, the results are characteristic of industrial-scale processes (rarely occurring in the scientific literature); at the same time, experiments were often burdened with exceedingly high standard deviations, partly due to unavoidable sample inhomogeneities resulting from the aeration, feedstock, and operational patterns and partly due to the inevitably high time span of the sampling process through the ASPs. The ASP system was fully operational, and the aeration was turned on during the whole sampling period; aeration can introduce substantial variability in microbial activity and determine the decomposition rate of organic matter and, subsequently, the profile of emissions.

(**a**)

(**b**)

(**c**) (**d**)

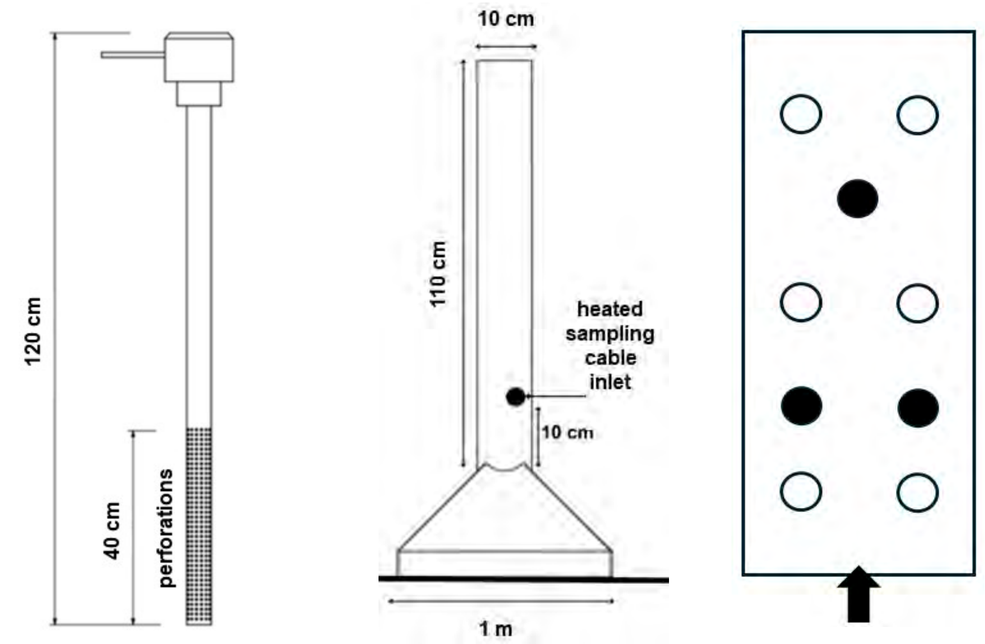
Figure 1. On-site sample measurement on the membrane-covered side-walled aerated static pile (ASP) systems covered with expanded polytetrafluoroethylene (ePTFE) membrane cover (ePTE-TEXcomm or ProfiCover®, Profikomp Environmental Technologies Inc., Gödöllő, Hungary), located near Orosháza,

Békés County, Hungary. (**a**) Aerial view of the industrial composting site (indicated by the red arrow) with the six ASPs (indicated by dashed rectangles) 25 m × 8 m in size each. (**b**) A covered ASP equipped with gas-collecting cones and a gas-analyzing unit. (**c**) A close-up view of a gas-collecting cone installed on the top of the pile. (**d**) Computerized extractive direct interface Fourier Transform infrared (FTIR) spectrometer connected by a heated sampling cable to avoid condensation. An uncovered ASP unit (shown on the right) was also simultaneously constructed.

2.2. Sampling Procedures and Techniques

2.2.1. Gas Collection

For collecting samples of gas content in the compost biomass, perforated lance probes inserted into the biomass through the cover membranes were used. For collecting samples of emitted gases, gas-collecting cones (hoods) installed on the top of the compost piles were utilized (Figure 2). This technique is suitable for the comprehensive capture of gases originating from compost piles. This study identified 6 sampling points on top of the cover laminate. This protocol was adopted to ensure uniform and consistent sampling across all three piles. For measuring the composting material's internal gaseous emissions, penetration probe sampling was used at 3 different sampling points inside each pile, where sampling in multiplicates was made possible without damaging or opening the covers via the openings on the covers for the temperature sensors, and without allowing the escape of the positive pressure and built up emissions from underneath. This involved the insertion of a probe roughly 1 m into the compost pile, providing emission measurements of its internal gaseous state in sufficient repetitions at the sampling points, with new insertions of the probe at different angles. Compared to laboratory-scale experiments, where high standard deviations would be unacceptable, in the case of industrial-scale tests in reality, this is not unusual at all, considering the inevitable inhomogeneity resulting from the nature of the materials examined and the industrial-scale, robust operation of machinery and aeration actively working. This issue is covered in more detail in Section 4. Working with an industrial process on this scale, we expected exceedingly high standard deviations, originating from a wide range of factors, primarily the inhomogeneity of the composting biomass. The 5th day of the thermophilic phase was chosen for the measurements based on extensive operational experience gained in the given treatment plant, combined with the limitations of time and cost parameters of previous trial runs. Our aim was to ensure measurable emissions during the entire sampling process before temporarily depleting the pore space of the piles by the constant flush of supplied fresh air that would, in time, prevent further measurements. The approximate location of the sampling points is illustrated in Figure 2. The ambient air temperature during the sampling fluctuated between 38.5 °C and 42.1 °C. The relative humidity levels were between 32.0% and 34.5%. The wind speeds were between 0.4 m/s and 1.5 m/s and primarily from the west/southwest direction. Sampling took approximately 15 min per sample point; therefore, the experimental design was established, taking into consideration that the aeration had to be turned off after sampling at each point in order not to flush out and deplete the piles. The sampling equipment had to be moved after each point to the corresponding point of the next pile. For internal sampling, the temperature probe openings on the membrane covers were used for reaching into the biomass, as uncovering the sides would have caused disturbances in the airflow for the entire pile, resulting in the uncontrolled loss of the positive pressure and emissions, thus preventing sampling altogether.



(a) (b) (c)

Figure 2. The devices used for sampling gas in the compost biomass and emissions. (**a**) Perforated lance probes for sampling gas content in the compost biomass. (**b**) Gas-collecting cones (hoods) installed on the top of the compost piles for sampling gas emissions through the membrane cover. (**c**) Locations of the sampling points on the aerated static piles. Locations of the perforated sampling lance probes (solid dots) and sampling hoods (hollow dots). The entrance of the composting pile, i.e., the farthest point from the aeration fan is indicated (black arrow).

2.2.2. Sampling Equipment

The sampling apparatus utilized was a portable gas sampler (product ID 09204; Gasmet Technologies Oy, Vantaa, Finland/Ansyco GmbH, Karlsruhe, Germany). It featured a Swagelok fitting (6 mm/8 mm; Swagelok Co., Solon, OH, USA). The equipment was consistently operated at a temperature of 180 °C to maintain the integrity of the gas samples. This consistent temperature ensured the preservation of the gas samples' integrity throughout the collection process.

2.3. Measurement Protocols and Techniques

2.3.1. Ammonia Measurement

The measurement of NH_3 emissions was based on the ASTM D6348-3 Standard test method due to its efficacy in determining gaseous compounds via extractive direct interface Fourier Transform infrared (FTIR) spectroscopy. The corresponding standard VDI 3862 Blatt 8 [20] was incorporated alongside the FTIR for formaldehyde emissions in combustion engine exhausts using the FTIR methodology. This study was performed according to the technical guidelines outlined in the UK Technological Guidance Note TGN M22 v.3 [21]. Airborne NH_3 concentration is expressed as ppm (1 ppm corresponding to 0.697 mg/m^3) [15].

2.3.2. Greenhouse Gas Emissions Measurements

ASTM D6348-3 Standard's FTIR spectroscopy technique [22] was used for GHG emissions measurements. This method is suitable for identifying up to 50 distinct gases based on their individual IR spectra. Some of the spectra were developed using certified material samples, facilitating device-specific calibrations; others were procured from the Gasmet spectrum library (Gasmet Technologies Oy, Vantaa, Finland/Ansyco GmbH, Karlsruhe, Germany) [23]. Airborne CO_2, CH_4, and N_2O concentrations are expressed as ppm (1 ppm corresponding to 1.800, 0.656, and 1.803 mg/m^3, respectively).

2.3.3. Volatile Organic Compound Measurements

VOC content was represented by the propane equivalent value calculated from FTIR spectroscopy determinations using the DX-4000 FTIR multigas analyzer #091610 (Gasmet Technologies Oy, Vantaa, Finland/Ansyco GmbH, Karlsruhe, Germany) with a silicon carbide ceramic infrared light source in the wave number range of 900–4200 1/cm operating at 1550 K; a gold/rhodium-coated aluminum measuring cell (0.45 dm^3) with a multi-lane constant road length (5.0 m) structure; and a thermoelectronically cooled mercury–cadmium telluride detector at atmospheric pressure and a 180 °C operating temperature. The experiment was controlled by the measuring software Calcmet Standard for Windows v.12 (Gasmet Technologies Oy, Vantaa, Finland/Ansyco GmbH, Karlsruhe, Germany). The instrument configuration allowed a wave number resolution of 8 1/cm and a measurement frequency of 10 spectra/s.

2.4. Data and Statistical Analysis

To find out the impact of the covers on the gaseous emissions from the different compost piles, a series of statistical analyses were performed. The emissions under investigation were NH_3, GHGs (including CO_2, CH_4, and N_2O), and VOCs represented by the propane equivalent (FTIR). The repetitions of each treatment for statistical comparison were generated through multiple gas sampling points within each compost pile (ePTE-TEXcomm, ProfiCover, uncovered). Gas samples were taken from various points both inside and outside each pile for comprehensive sampling, and accounting for variability within each pile. This method provided the necessary repetitions within each treatment group. The normality of the data was evaluated by the Shapiro–Wilk test [24]. Given that not all data subsets conformed to a normal distribution, the median values were used as primary metric descriptors for comparing emissions across the compost piles. Such use of median values (in contrast to averages) is appropriate for datasets with non-normal distribution as it offers a robust measure of central tendency and is unaffected by outliers or extreme values that occur from expected high variations within measured systems. For the comparison of gaseous emissions from the inside versus the outside of each compost pile, the Mann–Whitney U test [25] was used as it is a nonparametric test suitable for datasets that follow or do not necessarily follow a normal distribution and allows for a comparison of independent groups with unequal sample sizes [26]. The homogeneity of variances was assessed by Levene's test for homoscedasticity [27]. This test was applied to the inside emissions data of the covered piles in comparison to the uncovered pile. To examine the efficacy of the covered piles in comparison to the uncovered pile, the independent sample *t*-test was applied for the inside emissions. This parametric test was selected due to its increased sensitivity, allowing it to identify even subtle differences, which was a key point, given our interest in the potential advantages of the covers. Moreover, due to the industrial size of the experiments, relative standard deviations often substantially exceeded the levels routinely seen in laboratory studies. Wherever possible, standard deviations are shown for the experimental data. In given cases burdened by unusually high (>100%) relative standard deviations, those variabilities are not reported numerically; only median values (typed in italics) are shown in the text for indicational purposes.

It is important to highlight that while these statistical tests provide insights into significant differences, they do not give the magnitude or practical significance of these variances. Therefore, emission reduction percentages were calculated for both the cover types to see the efficiency of these covers in mitigating the emissions. The reduction percentage from inside to outside was calculated using Equation (1) and the reduction of emissions inside the covered piles vs. the uncovered pile was calculated using Equation (2).

$$\text{ReductionPercentage} = \frac{E_{\text{outside}} - E_{\text{inside}}}{E_{\text{outside}}} \times 100\% \quad (1)$$

where E_{inside} is the median emission inside the cover and E_{outside} is the median emission outside the cover.

$$\text{ReductionPercentageInsideCoveredPiles} = \frac{E_{\text{uncovered\_in}} - E_{\text{covered\_in}}}{E_{\text{uncovered\_in}}} \times 100\% \quad (2)$$

where $E_{\text{uncovered\_in}}$ is the median emission inside the uncovered pile and $E_{\text{covered\_in}}$ is the median emission inside the covered pile.

Percentage emission reduction values were calculated for CO_2 from volume percentage ($v/v\%$) concentrations; all other gas emissions were calculated from mass concentrations (ppm). All computational and statistical analyses were performed using Python (version 3.8.5; Python Software Foundation, 2020). The following Python libraries were employed: Pandas (version 1.1.0; Pandas Development Team, 2020) and SciPy (version 1.5.2; SciPy Developers, 2020). MS Excel 16 (Microsoft, Redmond, WA, USA) was used for data processing and calculating reduction percentages.

3. Results

The internal (gas content in the compost biomass) and external (gas emission) gas concentrations detected in the compost piles covered by the ePTE-TEX$_{comm}$ or ProfiCover® membrane are shown in Table 1. The differences between the internal and external gas concentrations were calculated and statistically analyzed. Table 1 showcases median values for each emission type, differentiated by pile type and measurement location (inside vs. outside).

Table 1. The median values for each emission type, differentiated by pile type and measurement location (inside vs. outside) for ePTE-TEX$_{comm}$ and ProfiCover® (Profikomp Environmental Technologies Inc., Gödöllő, Hungary) piles.

Measurement	ePTE-TEXcomm			Mann–Whitney U Test *p*-Value	ProfiCover®			Mann–Whitney U Test *p*-Value
	In	Out	Difference		In	Out	Difference	
Ammonia (NH_3) [ppm]	4259.8 ± 212.2	390.1 ± 77.2	3869.7 ± 289.4	0.0357	3343.8 ± 1400.7	225.8 ± 99.7	3118.0 ± 1500.4	0.0238
Carbon dioxide (CO_2) [$v/v\%$]	1.09 ± 0.25	0.44 ± 0.18	0.65 ± 0.43	0.0357	*1.42* [a]	0.20 ± 0.11	1.22 ± 0.11	0.0238
Methane (CH_4) [ppm]	39.9 ± 23.8	*81.2*	-	>0.05	*69.3*	30.8 ± 18.6	*38.5*	>0.05
Nitrous oxide (N_2O) [ppm]	1.30 ± 0.67	1.00 ± 0.20	0.30 ± 0.87	0.0357	*1.50*	0.65 ± 0.13	*0.85*	0.0256
Propane equivalent [ppm]	56.7 ± 13.9	*31.3*	*25.4*	>0.05	*79.2*	12.3 ± 6.6	*66.9*	0.0238

[a] Median value for indicational purposes (typed in italics)—the relative standard deviation exceeded 100%.

3.1. Analysis of Emissions: Inside vs. Outside of Compost Piles

3.1.1. Shapiro–Wilk Test Results on Data Distribution

Interpretation of *p*-values: $p < 0.05$ indicated a statistically significant difference or that the data did not adhere to a normal distribution; $p \geq 0.05$ indicated no significant difference or that the data were normally distributed.

The Shapiro–Wilk test results on data distribution indicated that many data subsets for specific emissions and pile types were normally distributed (*p*-values greater than 0.05). However, some data subsets did not follow a normal distribution (*p*-values less than 0.05). This was evident from the examination for emissions of CO_2, CH_4, and propane equivalent for ePTE-TEX$_{comm}$ (outside) and CH_4 and propane equivalent for uncovered piles (outside). The variation in the data distribution led to the decision to proceed with median comparison and employ the nonparametric Mann–Whitney U Test, as mentioned in the methodology. This approach is appropriate when dealing with non-normally distributed independent data.

3.1.2. Mann–Whitney U Test Results on Inside vs. Outside Emissions

The results of the Mann–Whitney U test (Table 1) indicated that there were significant differences between the emissions inside and outside of the compost piles for both compost piles covered with either ePTE-TEX$_{comm}$ or ProfiCover®. Specifically, significant differences were found for NH_3, CO_2, and N_2O emissions in the ePTE-TEX$_{comm}$ pile and for NH_3, CO_2, N_2O, and propane equivalent (FTIR) emissions in the ProfiCover® pile.

These findings corroborate the observations made by Song et al. [28], who found that covering compost piles with a semi-permeable membrane not only improved nitrogen preservation rates but also notably reduced emissions of NH_3 and H_2S. These findings are further strengthened by the study by Soto-Herranz et al. [29,30], who achieved a substantial 65% reduction in NH_3 emissions using ePTFE and other semi-permeable membrane cover systems in farm-scale composting processes. This evident reduction in NH_3 emissions mirrors the results observed, signifying the consistent performance of membrane covers across different scales and conditions. Additionally, Cao et al. [31] demonstrated a similar trend where a membrane-covered approach diminished NH_3 emissions during anaerobic composting. Such consistent findings across multiple studies indicate a reliable effect of membrane covers on NH_3 emissions.

3.2. Levene's Test for Homoscedasticity

Levene's test for homoscedasticity showed that the variances were equal across the groups for all emissions, satisfying the assumption of homogeneity of variances for the independent sample *t*-test. This indicated that the groups being compared had similar variability in emissions. For all emissions (NH_3, CO_2, CH_4, N_2O, propane equivalent), the variances were equal across the groups, satisfying the homogeneity of variances assumption for the independent sample *t*-test.

3.3. Comparative Analysis of Emissions from Covered and Uncovered Piles Using the Independent Sample T-Test

Both ePTE-TEX$_{comm}$ and ProfiCover® demonstrated a significant reduction in emissions when compared to the uncovered pile. Table 2 showcases the median values for each emission type, based on measurements of gas content inside the covered and uncovered piles, differentiated by membrane-covered and uncovered pile types for each membrane type. The comparative analysis of covered (ePTE-TEX$_{comm}$ and ProfiCover®) and uncovered piles using the independent sample *t*-test revealed interesting findings regarding emissions. For NH_3 emissions, both ePTE-TEX$_{comm}$ and ProfiCover® demonstrated a significant reduction compared to the uncovered pile. This aligns with previous research that has shown the potential of covering compost piles to mitigate NH_3 emissions [31–34].

Table 2. The median values for each emission type based on internal gas content by comparing covered and uncovered piles and ePTE-TEX$_{comm}$ and ProfiCover® (Profikomp Environmental Technologies Inc., Gödöllő, Hungary) piles.

Measurement	Uncovered	ePTE-TEXcomm	ePTE-TEXcomm—Uncovered	Mann–Whitney U Test *p*-Value	ProfiCover®	ProfiCover®—Uncovered	Mann–Whitney U Test *p*-Value
	In	In	Difference		In	Difference	
Ammonia (NH_3) [ppm]	1584.8 ± 367.2	4259.8 ± 212.2	2675.0 ± 684.0	0.0357	3343.8 ± 1400.7	*1759.0*	0.0714
Carbon dioxide (CO_2) [*v*/*v*%]	0.43 ± 0.16	1.09 ± 0.25	0.66 ± 0.50	0.0357	*1.42* [a]	*1.0*	-
Methane (CH_4) [ppm]	3.45 ± 1.50	39.9 ± 23.8	36.5 ± 25.4	0.0357	*69.3*	*65.9*	-
Nitrous oxide (N_2O) [ppm]	0.54 ± 0.39	1.30 ± 0.67	*0.76*	>0.05	*1.50*	*1.0*	-
Propane equivalent [ppm]	13.9 ± 3.51	56.7 ± 13.9	42.8 ± 17.9	0.0357	*79.2*	*65.3*	-

[a] Median value for indicational purposes (typed in italics)—the relative standard deviation exceeded 100%.

The median values for each emission type, differentiated by pile type and measurement location (inside vs. outside), highlighted variability in emissions between the inside and outside of compost piles and across different pile types. For example, the median values for NH_3 emissions were found to be substantially higher inside the covered piles (ePTE-TEX$_{comm}$ and ProfiCover®) compared to the uncovered pile. However, for CO_2, CH_4, N_2O, and propane equivalent emissions, no significant difference was found between the emissions from inside the covered piles and the uncovered pile. This suggests that the impact of membrane covers on other emissions such as CO2, CH_4, N_2O, and propane equivalent may be limited.

3.4. Emissions Reduction Analysis

To evaluate the efficacy of the compost covers in modulating gaseous emissions, an in-depth analysis focusing on emission reduction was conducted. The main emissions investigated were NH_3, GHGs (including CO_2, CH_4, and N_2O), and VOCs, denoted by the propane equivalent (FTIR). The composting piles under examination were covered by ePTE-TEX$_{comm}$ and ProfiCover®.

3.5. Reduction from Inside to Outside

The emission reduction percentages from the inside of the piles to the outside of the membrane cover were calculated using Equation (1), previously described in the methodology section, to evaluate the efficacy of the compost covers in modulating emissions. The reduction percentages for both the covered piles are presented in Table 3.

Table 3. Emission reduction percentages between inside vs. outside of membrane-covered compost piles and between inside membrane-covered compost piles relative to the uncovered pile for ePTE-TEX$_{comm}$ and ProfiCover® (Profikomp Environmental Technologies Inc., Gödöllő, Hungary) piles.

Measurement	Emission Reduction (%)			
	Between Inside and Outside of Membrane-Covered Compost Piles		Between Inside Membrane-Covered and Inside Uncovered Piles	
	ePTE-TEX$_{comm}$	ProfiCover®	ePTE-TEX$_{comm}$	ProfiCover®
Ammonia (NH_3)	90.8 ± 1.9	93.3 ± 5.7	*62.8*[a]	*52.6*
Carbon dioxide (CO_2)	59.6 ± 18.8	85.9 ± 23.5	*60.5*	*69.6*
Methane (CH_4)	n.s.d. [b]	n.s.d.	*91.4*	*95.0*
Nitrous oxide (N_2O)	*23.1*	56.7 ± 49.3	*58.3*	*63.9*
Propane equivalent	*44.8*	84.5 ± 25.4	*75.5*	*82.4*

[a] Median value for indicational purposes (typed in italics)—the relative standard deviation exceeded 100%.[b] n.s.d.: no significant difference detected.

For ePTE-TEX$_{comm}$, the reduction percentages indicated a significant reduction in NH_3 emissions (90.8%), N_2O emissions (23.1%), and CO_2 emissions (59.6%) when transitioning from inside to outside the compost pile. The reduction in propane equivalent emissions (44.8%) suggested a positive impact of the cover in reducing VOCs. Similarly, for the ProfiCover®, there was a substantial reduction in NH_3 emissions (93.3%), CO_2 emissions (85.9%), CH_4 emissions (55.6%), N_2O emissions (56.7%), and VOC (propane equivalent) emissions (84.5%) while transitioning from inside to outside the compost pile.

The observed reductions align with existing studies on the potential of cover technologies in minimizing composting emissions. For instance, the notable reduction in NH_3 emissions under both covers is supported by findings from Sun et al. [35] and Soto-Herranz et al. [29], affirming the effectiveness of membrane technologies in reducing NH_3 volatilization. However, the ProfiCover® exhibited a slightly higher reduction in NH_3 emissions (93.3%) compared to the ePTE-TEX$_{comm}$ (90.8%), suggesting a marginal superiority in performance, albeit within a close range. The primary reason for the reductions in NH_3 emissions was attributed to the condensation droplets forming under the membrane partially evaporating and NH_3 being discharged to the environment, thus decreasing the emission rate inside the membrane [36]. Similarly, the substantial reduction in CO_2 emissions, especially under the ProfiCover® (85.9%), resonates with the findings of Cao et al. [31], although at a different magnitude. The variance in the reduction percentages could be attributed to the distinct characteristics of the membrane technologies employed or the feedstock used in the composting process, as suggested by Cao et al. [31]. The reduction of N_2O emissions, although significant, was lower compared to the reductions of other gases. This is consistent with the relatively lower reduction of N_2O emissions reported by

Cao et al. [31]. The dynamics of N_2O production and emission during composting might be more resilient to modulation by cover technologies, thus necessitating further research to explain the mechanisms and improve mitigation strategies.

Starting with our primary observation, both the ePTE-TEX$_{comm}$ and ProfiCover® demonstrated considerable efficacy in reducing NH_3 and CO_2 emissions during composting. This aligns well with the study by Cao et al. [31], which reported a 25.8% reduction in NH_3 emissions and a 13.1% reduction in N_2O emissions using membrane cover technology. This study also mentions the use of superphosphate, an additive that has been previously documented as reducing the loss of nitrogen and certain GHG emissions, including NH_3, CH_4, and H_2S. The incorporation of such additives can be prominent. For instance, Yuan et al. [37] highlighted that superphosphate and dicyandiamide substantially reduced NH_3, CH_4, and N_2O emissions, while phosphogypsum could also curtail NH_3 and CH_4, but at the cost of increased N_2O emissions. Similarly, Jiang et al. [38] found that superphosphate not only facilitated the composting process but also significantly decreased NH_3 emissions during the thermophilic phase, in contrast to bentonite, which increased NH_3 emissions, while the effect of superphosphate was further impacted by the addition of salts, calcium dihydrogen phosphate or calcium sulfate, and free phosphoric or sulfuric acids [39]. Citrogypsum, a by-product of citric acid synthesis, could also be used to reduce NH_3 emissions [40]. In addition, the combined use of acidification and biochars could also effectively mitigate NH_3 losses [41]. Ma et al. [42] reported findings that further support our observations. Their research identified a decline in NH_3 and N_2O emissions by 11.8% and 26.4%, respectively, using the ePTE-TEX$_{comm}$ membrane, and a remarkable reduction in N_2O emissions by 68.4% with the ZT membrane. This decline in emissions was also associated with an improved bacterial community, suggesting a possible biological dimension to the emissions reductions observed. No additional additives were cited in the study, implying that the membranes' impact was primary in achieving these results. Li et al. [5] focused on the composting of kitchen waste and found a 48.5% and 44.1% reduction in NH_3 and N_2O emissions, respectively, with membrane-covered composting. Their study also attributed the reduction in NH_3 emissions to adsorption by the condensed water layer under the inner membrane and the reduction in N_2O emissions to micro positive pressure in the reactor promoting oxygen distribution similarly.

On alternative strategies, Zuokaitė et al. [43] explored the use of natural covers like wood bark, sawdust, peat, and a grass layer, presenting potential alternative or complementary solutions in scenarios where membrane covers may not be feasible. Ermolaev et al. [44] also provided a different perspective, suggesting that broader variables like temperature, moisture content, mixing frequency, and the amount of added waste could significantly influence gas emissions during composting. Szymula et al. [45] reported the effective use of natural sorbents like given biochars, bentonites, zeolites, and—to a lesser extent—perlites in the effective reduction of NH_3 emissions.

3.6. Reduction of Emissions Inside the Covered Piles Compared to Uncovered Piles

To gain a deeper insight into the efficiency of the covers, a comparative analysis was conducted to gauge the emission reductions of the covered piles (ePTE-TEX$_{comm}$ and ProfiCover®) against the uncovered pile for inside measurements. The results of this analysis are illustrated in Table 3.

The results of the comparative analysis showed the reduction percentages of various emissions inside the piles of the covered piles (ePTE-TEX$_{comm}$ and ProfiCover®) compared to the uncovered pile. Inside measurements indicated that both covers resulted in reductions in emissions of NH_3, CO_2, CH_4, N_2O, and propane equivalent. The ProfiCover® semi-permeable membrane showed greater reductions of NH_3, CO_2, CH_4, and N_2O compared to the ePTE-TEX$_{comm}$. Specifically, the ProfiCover® reduced CO_2 emissions by approximately 70%, which was higher compared to the reduction by ePTE-TEX$_{comm}$ of around 60%. On the other hand, the ePTE-TEX$_{comm}$ reduced NH_3 emissions by around 63%, which was higher than the reduction by ProfiCover® of around 53%. This divergence in performance implies a potential trade-off in emission reduction efficacy between the two covers. Nevertheless, the implication is that the use of either cover substantially decreases emission production, thereby enhancing the compost quality and increasing nitrogen retention in the compost, which are pivotal for the environmental sustainability and efficiency of the composting process.

4. Discussion

While the primary focus of this analysis was on the emissions of NH_3 and GHGs, it is noteworthy to consider the broader implications of membrane covers for composting processes. Furthermore, not all studies exclusively reported reductions in gas emissions with added materials or alterations to the composting process. For instance, Wei et al. [46] reported that the incorporation of cornstalks led to decreased hydrogen sulfide emissions but had negligible effects on NH_3. This underscores the importance of specific cover materials and their interactions with composting substrates. Studies by Ma et al. [42,47,48] and Li et al. [49] also highlighted the role of membrane covers in gas emissions and bacterial community succession during composting. These works emphasize the multifaceted impacts of membrane covers, spanning beyond just emission reductions and massively influencing microbial community dynamics. All these findings are also consistent with studies that investigated the effect of coverings on gaseous emissions from composting. For example, Bernal et al. [50] explored the impact of covering composting material with zeolite minerals. Their study found that the combination of mixing pig slurry with easily degradable straw and covering the composting material with zeolite minerals led to a marked reduction in NH_3 emissions. This suggests that the physical barrier provided by covers can play a pivotal role in preventing the direct release of NH_3 into the atmosphere [50]. Similarly, Chadwick [51] emphasized the potential of compacting and covering manure heaps in reducing NH_3 emissions. The research indicated that such practices are particularly effective when the manure contains high ammonium–N contents. This aligns with the present findings, reinforcing the idea that covering composting piles can be a crucial strategy in environmental management [51] and capturing nitrogen as an essential nutrient. A study by Sun et al. [35] also demonstrated the advantages of using a semi-permeable membrane-covered composting system. It highlighted a reduction not only in NH_3 production and emissions but also in GHG emissions. This suggests that certain covers might offer dual benefits by optimizing the composting process and further reducing harmful emissions. Furthermore, Berg et al. [52] investigated the efficacy of different materials for covering liquid manure storage facilities. Their findings highlighted the importance of maintaining a lower pH value to effectively reduce NH_3 emissions, emphasizing the role of environmental conditions in conjunction with covering techniques [52]. The consistent findings across different studies underscore the importance of using covers in composting or manure storage practices.

The comparative analysis of emission reductions for covered and uncovered compost piles corroborates the findings from previous research that investigated the effect of cover technologies on gas emissions during composting. For instance, the substantial reductions in CH_4 emissions echoed the findings of Sun et al. [35] and Fang et al. [36], where membrane-covered composting systems were found to significantly mitigate CH_4 emissions. Similarly, the observed decrease in NH_3 emissions conforms with findings across several existing studies, underscoring the efficacy of cover technologies in reducing NH_3 volatilization. Hou et al. [14] found that mitigation measures, such as covering, can help reduce NH_3 emissions. Soto-Herranz et al. [29] studied the reduction of NH_3 emissions from laying hen manure in a closed composting process using gas-permeable membrane technology and observed that the emission rate inside the membrane decreased compared to the uncovered condition, which aligns with our findings.

Statistical testing using Levene's test confirmed the homoscedasticity of the data, ensuring the robustness of these findings. The Mann–Whitney U test indicated significant differences in emissions between the inside and outside of both compost piles, corroborating the mitigative capabilities of the semi-permeable membrane covers. This aligns with prior research advocating for the advantages of compost covers. While both covers effectively reduced NH_3 and CO_2 emissions compared to uncovered compost piles, their impact on other GHGs, notably, CH_4 and N_2O, was somewhat limited, calling for further research. It is also worth noting the potential of these covers to diminish VOCs, as indicated by the decreased propane equivalent emissions. In summation, this investigation reinforces the idea that the application of membrane technologies like ePTE-TEX$_{comm}$ and ProfiCover® plays a consequential role in ameliorating environmental impacts, particularly in reducing detrimental emissions such as NH_3 and CO_2 during large-scale organic manure composting. Furthermore, the reduction in emissions, as shown in our study, suggests an increase in nitrogen retention within the compost, which is instrumental for improving compost quality and the environmental sustainability of composting processes. Although our study did not delve into the specifics of nitrogen retention mechanisms, the referenced papers provide insights into potential pathways such as reduced NH_3 volatilization and filtrate leaching, as well as the transformation of organic nitrogen into NH_4^+ through mineralization processes [42]. Ma et al. [42] further support our results; they observed that membrane covers significantly reduced gas emissions compared to an uncovered control. The membrane cover created a controlled environment that minimized the release of gases, leading to a more efficient composting process. Cao et al. [31] demonstrated that a membrane-covered composting system decreased NH_3 and H_2S emissions and reduced the loss of total nitrogen from the compost pile. Li et al. [33] also found that combined membrane-covered systems improved the aerobic composting process and reduced gas emissions. Cao et al. [31] examined the effects of membrane-covered technology on compost quality and nitrogen-containing gas emissions during aerobic composting. The study highlighted that the membrane-covered sample had significantly lower emissions compared to the control sample. Specifically, the membrane-covered sample had a germination index of 50% and 80% approximately 2 and 9 days earlier, respectively, than the control sample. This indicates that the membrane cover facilitated faster composting and reduced the release of nitrogen-containing gases.

The disparity in emission reduction efficacy between the ePTE-TEX$_{comm}$ and ProfiCover® suggests that different cover manufacturing technologies may exhibit varying levels of performance in mitigating specific emissions, mainly due to the pore size of the semi-permeable membrane used. This is consistent with the variance in emission reductions reported in the papers that employed different cover technologies and additives. For example, Cao et al. [31] reported a reduction in N_2O and CO_2 emissions of 68.4% and 1.56%, respectively, using membrane cover technology, which presents a contrasting degree of CO_2 emissions reduction when compared to our findings.

Addressing sometimes exceedingly high standard deviations, other authors reported that data on gaseous emissions from full-scale composting plants are related to the composting technology and waste characteristics [53]. A study carried out in commercial composting plants in Denmark where methane emissions were measured over a one-year period showed that methane emissions were significantly affected by factors including the type of feedstock and composting technology, treated feedstock mass, operational patterns, and season [54]. We identified in our study that the possible inadequate mixing and initial inhomogeneity of the manure before building the piles can be a reason for the high variability of obtained concentrations, highlighting the importance of the massive impact of operational conditions on any measurements conducted on composting biomass, resulting in a wide gap between laboratory-scale modeling and on-site performance. Therefore, an exhaustive sampling campaign or online measuring system would be necessary to obtain representative and reliable data for a single plant, even if this is not a feasible way to obtain information.

5. Conclusions

This study's comprehensive evaluation of emission reductions underscores the significant efficacy of compost covers, particularly the ePTE-TEX$_{comm}$ and ProfiCover®, in mitigating gaseous emissions during the composting processes, especially during the thermophilic phase of organic manure composting. A comparative analysis between inside and outside emissions revealed that both covers played a pivotal role in emission containment. The ePTE-TEX$_{comm}$ manifested a reduction in most gaseous emissions, with decreases of 90.8% for NH_3 and 59.6% for CO_2. Other gases, including N_2O and propane equivalent, experienced reductions of 23.1% and 44.8%, respectively. On the other hand, the ProfiCover® presented even more compelling reductions. Emissions of NH_3 and CO_2 plummeted by 93.3% and 85.9%, respectively. CH_4, contrasting with the ePTE-TEX$_{comm}$, observed a significant reduction of 55.6%, attributed to the different semi-permeable membranes used. Concurrently, N_2O and propane equivalent followed, with substantial reductions of 56.7% and 84.5%, respectively.

When comparing the emission reductions inside the covered piles to those inside the uncovered pile, the ProfiCover® stood out, with reductions of 52.6% for NH_3, 69.6% for CO_2, a noteworthy 95.0% for CH_4, 63.9% for N_2O, and 82.4% for propane equivalent. The ePTE-TEX$_{comm}$, not far behind, showcased notable reductions as well. The values were 62.79% for NH_3, 60.5% for CO_2, 91.4% for CH_4, 58.3% for N_2O, and 75.5% for propane equivalent.

Further research should investigate integrating membrane cover technologies with additional emission control strategies like biofilters or amendments to develop an optimized and comprehensive solution. More studies are needed to focus explicitly on optimizing cover design, materials, and implementation practices to enhance the mitigation impact, especially on GHGs beyond CO_2. Long-term and life cycle assessments of membrane covers should be performed to evaluate sustainability implications and impacts on compost quality over time. Cost–benefit analyses would provide helpful information for industrial facilities considering adopting these technologies. Exploring the use of alternative sustainable materials for membrane covers could be worthwhile to reduce environmental impacts. Developing functionalized "smart" membrane materials could present opportunities to selectively control gas transport and modulate emissions. Communication and knowledge-sharing between researchers and industry partners are key to translating these technologies into widespread adoption. Overall, membrane covers show promise as an effective tool for reducing certain harmful emissions from organic manure composting, but ongoing research and development focused on optimization and integration with other methods is important.

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