First field Demonstration of a Novel Gas-Phase Photo-oxidation Method for Mitigation of Methane, Ammonia, and Odor from Agricultural sources

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Abstract

There is significant climate and environmental benefit to mitigating the environmental impacts of livestock and biogas production. Methane is a strong greenhouse gas that leads to global warming, ammonia emissions pollute groundwater, and odor is a serious local problem which is often regulated. There is a growing focus on the reduction of global methane emissions, including public pledges by countries and food companies. Although some solutions exist for ammonia and odor removal, no scalable method effectively treats methane from enteric fermentation or integrates the mitigation of all three pollutants. It was found that MEPS, utilizing UV-light, chlorine gas and a NaOH scrubber, could remove 98 %, 94 %, and 80 % of methane, ammonia, and hydrogen sulfide (respectively) from the air in a pig barn. Tests in a dairy barn found a quantum yield of 1.54%, corresponding to a specific energy input of 0.5 kWh/g_{CH_4}, when operating at a methane removal efficiency of 51 %. While there is much room for optimization of the MEPS process, this work demonstrates an important step in developing scalable technology for eradicating low concentration methane sources from agriculture.

Synopsis

For the first time, a process removing methane, ammonia, and odor from cattle and pig barn air was demonstrated. the goal is to reduce agriculture's global warming impacts.

Introduction

Extensive efforts are underway to limit carbon dioxide emissions and decarbonize the energy supply.[1] In contrast, technology for reducing methane emissions is less well-developed, despite methane's total climate impact since the industrial revolution being 2/3 as large as that of carbon dioxide,[2] as well as methane's contributions to tropospheric ozone pollution.[3] A new report from the National Academies of Sciences, Engineering and Medicine has examined Atmospheric Methane Removal and concluded that there are no commercially available technologies that operate at methane concentrations

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below 1000 ppm.[4] An analysis by Abernethy et al.[5] showed that 3/4 of all methane emissions occur at these dilute concentrations, including emissions from cattle, wastewater treatment plants, coal mine ventilation and waste disposal. Recent work has shown that many methane mitigation strategies are orders of magnitude away from being feasible at scale,[6] and major breakthroughs in the performance of photocatalysis and biofilter technology would be required for these systems to have a large mitigation impact.[5,6]

Chemically, the oxidation of methane is well understood. Its oxidation is highly exothermic, with the heat of combustion of methane being 890 kJ/mol.[7]

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \qquad (1)$$

Despite methane oxidation being exothermic, there is a remarkable kinetic barrier. The C-H bond in methane is the least reactive of any hydrocarbon, resulting in methane having the longest atmospheric lifetime of any hydrocarbon, around nine years.[8] This kinetic barrier may be overcome with catalysts, however current candidates have their individual drawbacks. High temperature catalysts are generally performance limited by the Sabatier principle, and biofilters are performance limited by rate, conditions, and avoidance of ammonia.[6,9,10] Photocatalysts are still orders of magnitude away from being economically and energetically feasible[5,6]

To reduce agricultural methane emissions, the addition of 3-NOP to feed has been shown to lower emissions from enteric fermentation in livestock, with methane emission reductions of 13-32 %.[11–13] However, the long term effect of the feed additive has been questioned, as they seem to lose effectiveness over time,[14] but these additives could still play an important role in lowering near term methane emissions. Setting aside the issue of societal acceptance,[15] the limited ability of feed additives to mitigate enteric methane emissions drives research into more comprehensive methods for reducing methane from agriculture. Manure is, along with enteric fermentation, the largest contributor to methane emissions from livestock,[16] with some of the solutions for lowering emissions being acidification[17] and frequent removal of manure.[18] An alternative to avoiding the production of methane from manure is to enhance it and use it as biogas, however methane leaks during biogas production have been shown to be significant, and can negate any climate benefits of biogas production.[19–22]

The negative environmental impacts of livestock production extend beyond methane emissions and the derived impacts thereof.[23] Odor compounds from cattle, pigs and biogas plants adversely impact human health and the environment. Odor arises from a complex mix of volatile organic compounds (VOC) and reduced sulfur gases including hydrogen sulfide. Ammonia emissions have received particular attention and regulation. Ammonia reacts with atmospheric acids to form fine particulate matter linked to respiratory disease and mortality. It damages ecosystems by forming acid and causing eutrophication of waterways, and where microbial processes can also lead to increased nitrous oxide formation. Ammonia can also react with hydroxyl radicals directly in the atmosphere, leading to the formation of nitrous oxide and further exacerbating global warming.[24] With approximately 50 % of ammonia emissions originating from animals in confined feeding operations, the eradication of both ammonia and methane from these point sources would positively impact global greenhouse gas balances in the atmosphere.[25]

A new technology involving catalytic chain reactions of gas phase radicals, the Methane Eradication Photochemical System (MEPS) has been introduced previously[26] and demonstrated under laboratory conditions. This system, based on chlorine atoms, was shown to destroy methane with an energy input of 2.1 kWh/g_{CH_4}, and a space velocity of 0.33 min^{-1}, for 50 ppm methane at a removal efficiency of $\sim$50 %. The idea is based on earlier work on hydroxyl radical reactors, so-called gas phase advanced oxidation.[27–30]

In the current study, we present results for

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the performance of the MEPS process to both oxidize methane, but also remove ammonia and odor compounds in a real world setting. For the first time, this technology was tested on air samples live in the field. Air was processed from a biogas plant, a dairy barn, and a pig barn, and the process was run under continuous conditions. Operational performance, as well as process yields and energy use were measured and varied. Finally, we report on the impact of the MEPS technology on the mitigation of other co-pollutants (such as ammonia and odor compounds) from these sources.

Methods

The MEPS laboratory-scale prototype is a photoreactor that uses chlorine radicals (chlorine atoms, $Cl^{\bullet}$) to initiate the breakdown of CH_4. The process is shown in Figure 1. The methane-laden air is drawn from the specific environment through 40 mm PVC piping. The air is initially led through a counter-flow water scrubber to remove any large particles and physical debris. Subsequently, chlorine gas, produced by electrolysis of NaCl, is added to the airflow, using a 'CHLORINSITU IV Compact'-system from Prominent.[31] After the addition of molecular chlorine gas, the air flows into the photoreactor. The reactor has a size of 0.25 m^3 (1 m x 0.5 m x 0.5 m) and is lined with polished aluminum for UV reflectivity, which is covered with a 125 μm film of PFA (perfluoroalkoxy polymer) for chemical resistance. The reactor is illuminated at both ends with LEDs (light emitting diodes) with a peak emission wavelength at 368 nm, shown in Figure. 2. Each end of the reactor has eight printed circuit boards containing 9 LEDs each, totaling 144 LEDs. 260 W of power is applied in total, with a 35 % electrical power to photon efficiency,[32] and powered by a variable DC supply. Following the photoreactor, the air travels into a sodium hydroxide (NaOH) counterflow scrubber that removes residual molecular chlorine and hydrochloric acid (HCl), which is produced as a by-product of methane oxidation. The chlorine-free airflow is then vented. To avoid the possibility of chlorine leakage, the system is run at under-pressure, with air being drawn in by a fan (Lindab CK-250-C), capable of pulling up to 270 L/min through the system. Calculated average air residence times in the photo-reactor were in the range 57-127 s.

The reactor performance was evaluated in the field at the Aarhus University agricultural research center in Foulum, Denmark. Experiments were conducted to remove methane from exhaust air in three separate environments: a dairy barn (2 cows) with an experimental point-extraction system, a pig barn (26 pigs) - detailed description by Dalby et al.,[33] and the ventilation air of a biogas production plant (sampled after its odor biofilter), as earlier described by Sadegh et al.[34] The prototype was placed inside the barns and the ventilation room, allowing for real time processing of the air streams. The system operated for several hours at a time, and sensor data was recorded at one-minute intervals.

Measurements

13 experiments were conducted, 3 in a dairy barn, 4 in a pig barn, and 6 at a biogas plant. The experiments varied in length from 10 to 62 minutes, the average values for all experiments can be found in the supporting information, Table S3. The inlet and outlet of the system were monitored for concentrations of CH_4 and background compounds. Methane was measured through real-time tunable diode laser spectroscopy (TDLS) using an Axetris LGD compact-A CH4 sensor (10 ppb resolution). The inlet methane concentration was adjusted to correct for a small leak of air into the system by comparing the inlet and outlet concentrations when no chlorine or UV light was applied in the reactor (control). Average values were calculated for each experiment, excluding the first and last two data points due to system response delays. Some experiments were discarded from the barn experiments as methane concentrations were unstable due to variable animal behavior. The identification and quantification of low concentration odor related compounds in the pig barn air and MEPS

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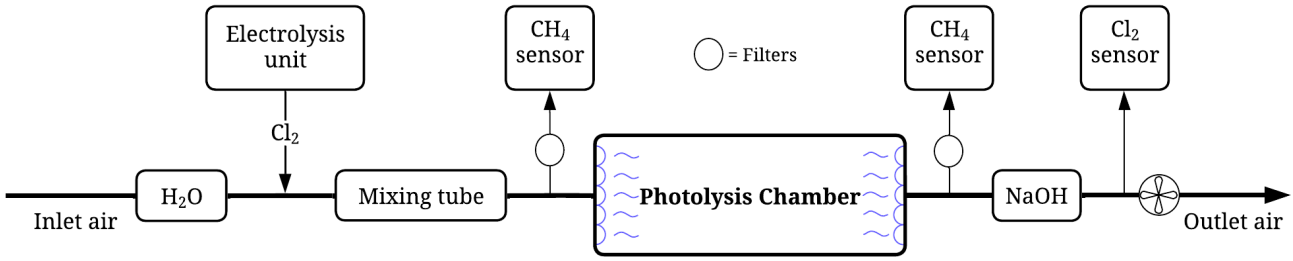

Figure 1: A schematic of the experimental setup. The filters are a combination of charcoal filters, humidity filters, and particle filters, to protect the gas sensors. H_2O and NaOH refer to wet scrubbers with the corresponding working fluid.

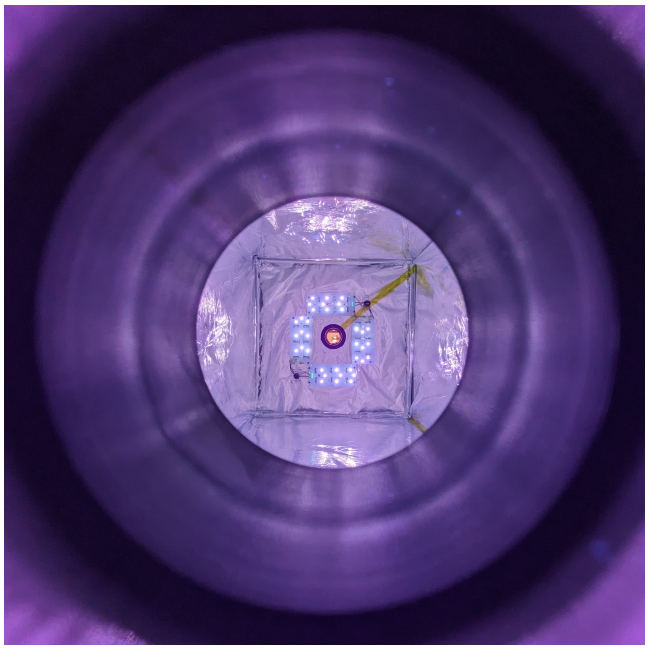

Figure 2: Visual representation of the photoreactor, as seen through the inlet.

reactor was performed using an online Proton Transfer Reaction - Quadrupole Mass Spectrometry (PTR-MS) (Ionicon Analytik, Innsbruck, Austria). PTR-MS is a highly sensitive method based on the ionization of gaseous compounds by protonated water, H_3O^+, in a drift tube and subsequent detection of the ionized species by a quadrupole mass spectrometer, detecting ions based on their mass-to-charge ratio (m/z). The voltage, temperature, and pressure of the drift tube were set at 600 V, 75 °C, and 2.1 mbar. The E/N number was approximately 145 Td in all experiments. The concentrations have been corrected for rate constant contributions as described in the Supporting Information, using rate constants from Cappellin et al,[35] along with calculations by the method proposed by Su.[36] The PTR-MS measured odorants with a dwell time of 0.2 seconds and the detection limit for the different compounds ranged from 0.02 to 1 ppb, depending on the compound.[37] Air was pumped into the PTR-MS through a $1/8^{th}$ inch PTFE tube. An electrochemical cell, a 'Membrapor Chlorine Gas Sensor Cl2/C-200', was used to monitor chlorine in the exhaust from the MEPS system, to ensure any emissions were below 1 ppm. Chlorine safety sensors with alarms were also placed near the system.

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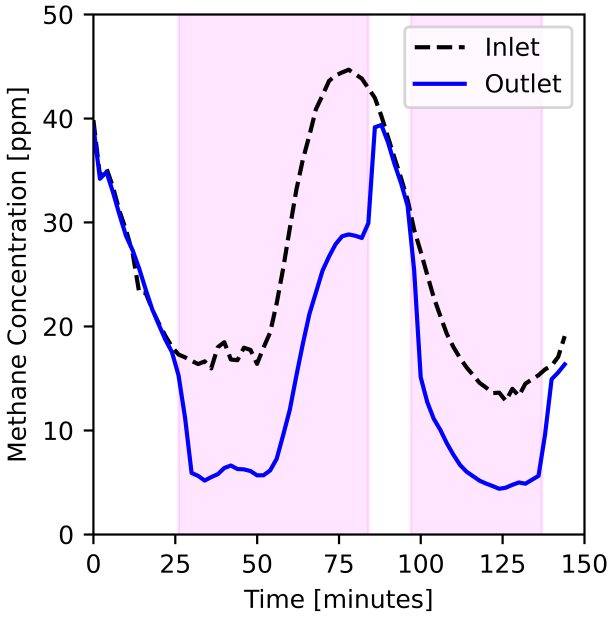

Figure 3: Methane concentration as a function of time during two consecutive experiments at the biogas plant. Pink shading indicates when the lights in the reactor were turned on, showing the removal of methane in the outlet air stream as compared to the inlet stream.

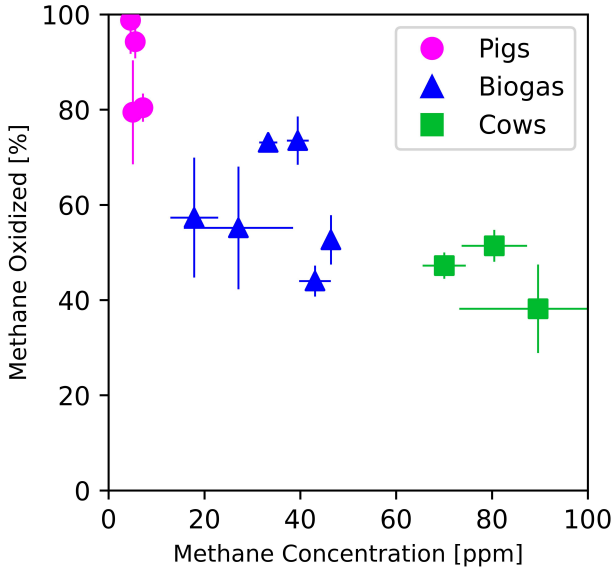

Figure 4: Methane oxidation yield as a function of initial methane concentrations in the air streams from different sources. The uncertainties marked are 1σ.

Results and Discussion

Methane Oxidation

The MEPS reactor was capable of removing methane from all three air sources via the addition of both chlorine gas and UV light simultaneously to the air streams. Figure 3 shows data from a typical experiment, displaying how the combination of light and chlorine in the air stream removed methane. The removal of methane is shown by the difference between the inlet and outlet concentrations (Figure 3).

The amount of methane that was oxidized under various conditions and for the different air sources is displayed in Figure 4, as a function of the initial methane concentration. Due to the nature of field experiments, initial methane concentrations were not controlled, and therefore the system was operated with a wide range of initial methane concentrations. Between 38 and 52 % of inlet air methane was oxidized from dairy barn ventilation air, over a concentration range of 70 to 90 ppm. A high level of methane oxidation was achieved in pig barn air (80-100%), at low initial methane concentrations. Methane oxidation from biogas plant air ranged from 44-73%, with initial concentrations ranging from 18-46 ppm. Even with co-pollutants present, the oxidation of methane seems effective.

Apparent Quantum Yield

While Figure 4 may appear to show that the MEPS is more efficient at removing methane at lower concentrations, it is necessary to investigate the apparent quantum yield (AQY) and the energy efficiency of this methane removal to better understand MEPS as a cost and energy effective process for methane removal. The AQY is one of the main metrics used to evaluate methane mitigation technologies initiated by photons.[5] AQY is calculated by comparing the number of photons added to the system with the number of methane molecules oxidized, as shown by equation 2, where $n_{\mathrm{CH_4}}$ is the number of methane molecules oxidized, e is the energy efficiency of the LEDs (35 %), P_{input} is the power input to the LEDs, λ is the wavelength of the LEDs, h is Planck's constant

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and c is the speed of light. This gives a measurement of the energetic efficiency of the process, and allows comparison between different reaction conditions and other methane oxidation processes.

$$AQY = \frac{n_{CH_4}}{e \cdot P_{input} \cdot \frac{\lambda}{hc}} \quad (2)$$

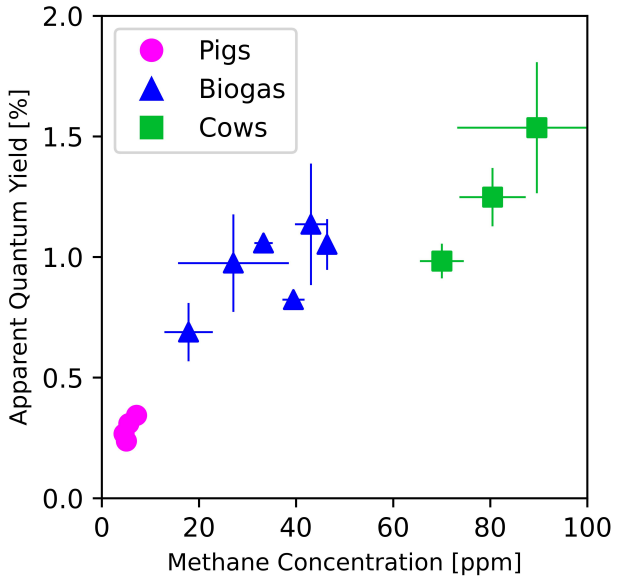

Figure 5: Apparent Quantum Yield as a function of methane concentration. The Uncertainties shown are 1σ.

The highest AQY for the MEPS was found to be 1.54 %, as shown in Figure 5, meaning that 1.54 % of the photons generated by the LEDs, oxidized a methane molecule. To the best of our knowledge, this is by far the highest apparent quantum yield for methane oxidation ever recorded, and is the first within the range of 9 ± 8 % set as the goal by Abernethy et al. although further development is warranted.[5,38,39] The theoretically highest achievable AQY is calculated from the number of chlorine radicals that can be created from a single photon, 2, and the reaction efficiency of chlorine radicals oxidizing methane (2.5 chlorine radicals/methane molecule), due to chlorine reacting with the products of the initial oxidation,[26] not accounting for wall reactions or reactions with co-pollutants the maximum achievable AQY is therefore 80 %. Thus there is significant space for optimization of the MEPS process from the current setup. The highest AQY was achieved in the dairy barn, and at the highest concentration of methane encountered in the field. This is consistent with increased reaction rate at high methane concentration and lesser impact of side reactions with other constituents. Light intensity was held relatively constant throughout all of the experiments, leading to a non-optimized light to chemical reactants ratio. Future improvements to the process may include optimization of light and thus energy inputs into the system, especially with respect to inlet methane concentrations.

Energy Efficiency

To compare our results with those of the literature, we look at the energy efficiency of the process. The energy efficiency is calculated using the energy used to power the LEDs, the electrolysis (chlorine production), pumps for circulation of water in the scrubbers, and the fan pulling air through the system. The total energy consumption was then compared to how much methane was removed, as shown in Figure 6.

The results of this study are shown in comparison with the results reported by Krogsbøll et al.[26] in 2023. Due to improvements in the reactor design including reactor geometry, lamp placement and lining, we see improved energy efficiency at equivalent removal efficiencies, and higher removal efficiencies at equivalent energy inputs. The best energy efficiency found was 0.50 kWh/g_{CH_4}, at a methane concentration of 90 ppm. This is comparable to theoretical best case costs of thermo-catalytic methane oxidation (estimated from the literature at 0.51 kWh/g_{CH_4} for 100 ppm methane concentrations).[40] Even at this small scale, the technology already shows cost competitiveness with best case scenarios for thermo-catalytic methane oxidation. Three main factors to investigate before implementation would be the required carbon intensity of the electricity used, along with the societal and individual cost of methane emissions, all of which would depend on the application site.

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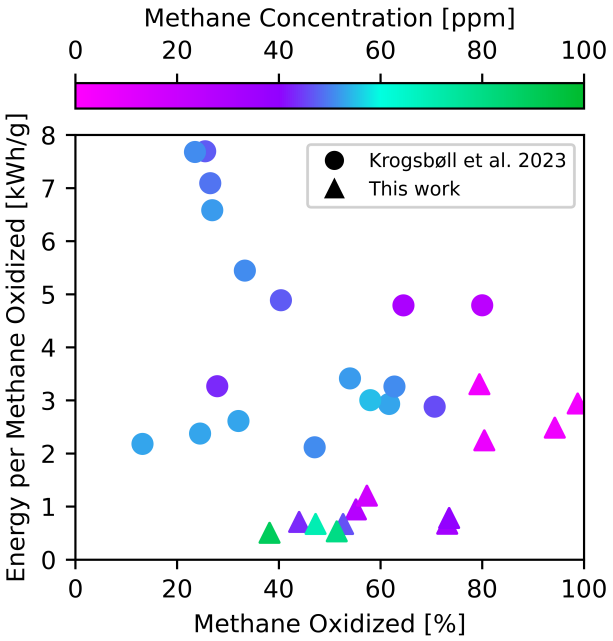

Figure 6: Energy efficiency as a function of oxidation capacity, as the methane concentration is varied (shown by the color). The results of this work are compared to the results published by Krogsbøll et al.[26]

While this demonstration of the MEPS technology shows promise as an effective and scalable method for methane oxidation in low methane concentration environments, there remains significant space for the improvement of the process and process optimization. In the current prototype reactor, chlorine concentration, light intensity, and residence time (the three primary control variables) were not controlled in relation to methane concentration, and thus further work can be done to optimize these inputs to reduce energy and resource use, while maximizing methane oxidation. In theory, these variables could be controlled based on varying concentrations of methane in the inlet streams for optimal reaction kinetics. In larger scale deployments of the MEPS technology, the physical dimensions of the photoreactor itself will increase the average photon pathlength, leading to more efficient chlorine radical formation per unit light, further increasing the AQY and energy efficiency. Due to the dimensional limitations of the current prototype, these gains can first be realized at larger scales. We expect that scaling up the MEPS technology will lead to reductions in specific energy usage and improved process optimization, and thus improve both process economics and the energy efficiency of the system.

Removal of Ammonia, Hydrogen Sulfide and VOC

Livestock production contributes to the emission of malodorous volatile compounds, which create nuisance for farming communities.[41] These emissions are primarily from microbial decomposition of organic material and proteins found in manure, along with H_2S from microbial sulfate reduction.[37] Studies have characterized emissions from livestock farming, including pig barns.[37,42] This study measured a comprehensive spectrum of odor compounds as a function of time using PTR-MS in a pig barn and evaluated the influence of MEPS on these emissions.

Table 1: Key odorants and their odor threshold values (OTVs).[42,43]

m/z	Assignation	OTV [ppb]
35	Hydrogen Sulfide	0.41
49	Methanethiol	0.07
60	Trimethylamine	0.032
63	Dimethyl sulfide	3
89	Butanoic acid	0.19
103	C_5-carboxylic acids	0.037/0.078
109	4-Methylphenol	0.054
118	Indole	0.3
123	4-Ethylphenol	1.6
132	3-Methyl-1H-indole	0.0056

The concentrations of the odorants decreased significantly upon activation of the system (Figure 7), demonstrating its efficiency in mitigating strong livestock odorants. The prototype removed over 70 % of all above mentioned compounds. Notably, highly odorous compounds like butanoic acid, carboxylic acid, and 4-methylphenol with concentrations significantly exceeding their OTVs, were reduced by over 90% (Figure 7). Ammonia and hydrogen sulfide are two of the most harmful livestock

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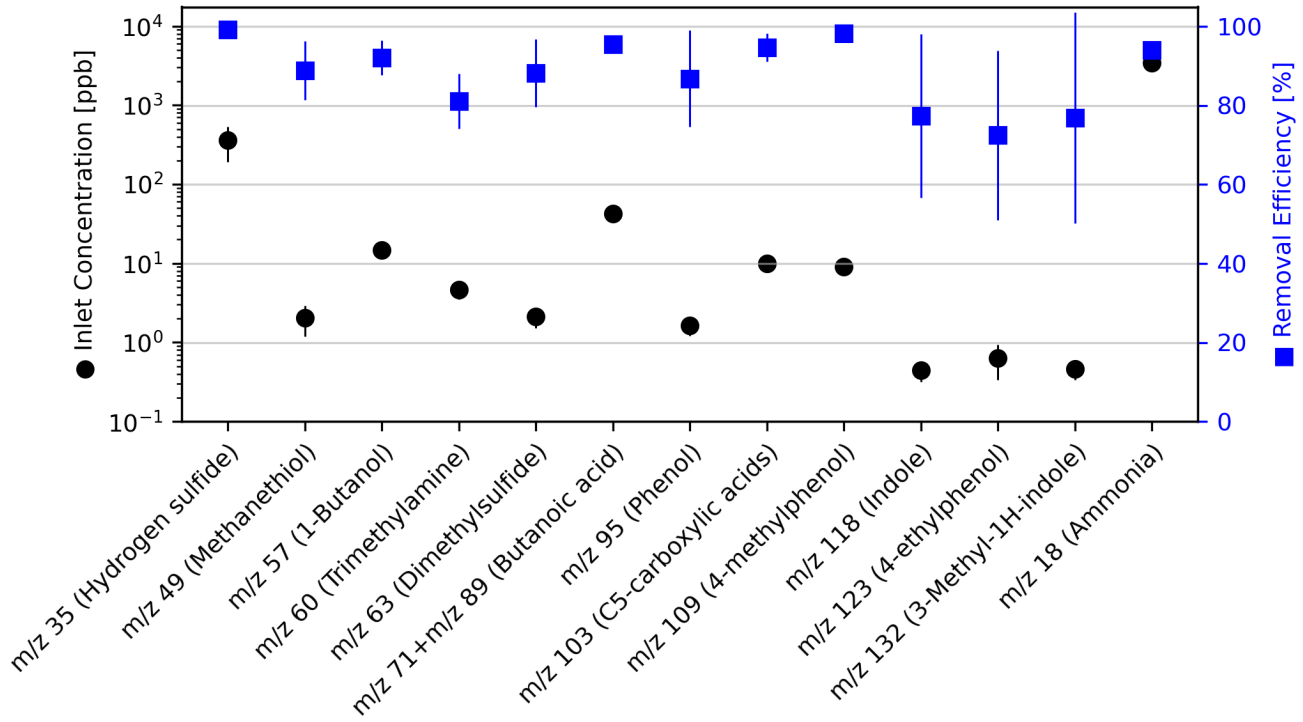

Figure 7: The effect of MEPS on malodorous emission in the pig barn. The average concentration over a 10-minute time period on the inlet air compared to a 10-minute average of the concentration after MEPS, the first two measurements for each period omitted due to a response delay. Uncertainties shown are 1σ.

emissions, for ammonia mainly due to eutrophication, secondary PM, soil acidification, biodiversity loss, and secondary GHG production, whereas H_2S is odorous and contributes to sulfate particles.[41] They are products of anaerobic microbial decomposition of protein and sulfur compounds in manure.[37] Inlet concentrations of ammonia (∼3460 ppb) and hydrogen sulfide (∼360 ppb) far exceed their OTVs (1500 ppb for ammonia).[42] Comparable concentrations are reported in the literature.[42,44] The system showed excellent removal efficiencies of 94 % for ammonia and 99% for hydrogen sulfide (Figure 7). The ammonia is removed partially from the scrubbers and partly from the photoreactor. It was not possible to distinguish the removal of hydrogen sulfide from the NaOH scrubber compared to the photoreactor; however, NaOH has formerly been used to remove H_2S.[45] As can be seen in the supplementary information, other compounds were also significantly removed, although their higher OTVs may make them of lesser concern. While the system effectively mitigated odorants, it led to increased concentrations of certain compounds. The compounds which increase in concentration are not found to have a GWP or strong odor. Chloroacetone is found to be toxic to humans, but its concentration is 100 times below the threshold limit values (TLV) for 8 hours exposure,[46] and is therefore not a concern. Chloroamines are found to be produced at concentrations up to 25 ppb, which is well below the limit set for the working environment in a swimming pool of 0.3 mg/m^3.[47] Compounds such as m/z 45 (acetaldehyde), m/z 55 (butanal), m/z 87 (2,3-butanedione), and m/z 129 (octanone) showed minimal changes in concentration (SI). Acetaldehyde and 2,3-butanedione are strong odorants with OTVs of 1.5 ppb and 0.05 ppb,[42] respectively, and inlet concentrations of 10–20 ppb and 2–8 ppb.

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Implications

Although this field test has focused on ventilation air from a dairy and pig barn and biogas plant, the method is general and could be used for other sources of dilute methane, including ventilation air from coal mines, landfills, and wastewater treatment plants. As a class of reactor, gas phase radicals are able to overcome obstacles in converting light energy to chemical reactivity due to the lack of phase change. Photocatalysts have a quantum efficiency of 0.001%.[5] In contrast the current study achieved a quantum yield of up to 1.54%, and the theoretical limit is much higher, potentially 80%. One reason behind this high efficiency is the formation of two radicals when Cl_2 absorbs one photon of light. Furthermore the reaction rate of methane with chlorine is 16 times quicker than the reaction of methane with OH.[48] We believe that the main loss of energy in the system is wall loss, as the optical depth of the system is longer than the dimension of the reactor. The optical pathlength of the chlorine can be calculated by equation 3 using the concentration (100 ppm) and the absorption cross section (acs) of $1*10^{19}$ cm^2 to be 40 m.[48–50]

$$l_{Cl_2,368nm} = \frac{1}{Conc_{Cl2} * acs_{Cl_2,368nm}} \quad (3)$$

Thus, the performance is expected to improve as the system is scaled up. Generally, the data demonstrates the system's high removal efficiency across diverse pollutants, even with significant variations in inlet concentrations.

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An early edition of this paper was submitted to the ChemRxiv preprint server for community feedback.[51] The graphical abstract was designed with Freepik.

Conflict of Interest

Ambient Carbon Methane Holding ApS owns patent WO2022053603A1(2022).[52]

Author Contributions

MK, HSR, AF and MSJ designed the experiments. MK, HSR and NF conducted the experiments. MK, MR, NF, NW and AF did the data analysis. All authors contributed to writing the article.

Supporting Information Available

Supporting Information can be downloaded as a separate PDF.

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