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Research article

Methane emission during on-site pre-storage of animal manure prior to anaerobic digestion at biogas plant: Effect of storage temperature and addition of food waste

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ABSTRACT

This study investigated the temperature dependency of CH_4 emission from pre-storage of animal manure prior to anaerobic digestion at 15, 20, 25 and 30 °C using lab-scale anaerobic digesters. The manure was added and removed daily to simulate the pre-storage process at biogas plants. CH_4 emission accounted for 1–46% of total CH_4 potential from pig manure (PM) and 1–2% of that from cattle manure (CM) at the investigated temperatures, with significant increases above 25 °C. Addition of food waste (FW) reduced the CH_4 emission when storage temperature was 20 °C or lower for PM and 25 °C or lower for CM due to volatile fatty acid accumulation and lower pH (< 5.5) but emissions increased with higher storage temperatures.

1. Introduction

Intensified livestock production is an important source of GHG emissions (CO_2, N_2O and CH_4) and contributes to environmental issues (Gerber et al., 2013). Compared to CO_2, CH_4 emission is more critical due to the higher global warming potential (Møller et al., 2004). In Denmark, anaerobic digestion (AD) of animal manure is highly promoted with the ambitious target of using 50% of the total manure for energy production by 2020 (Regeringen, 2011). Treating animal manure with AD can produce non-fossil energy and reduce the CH_4 emission from the spontaneous AD of animal manure during storage and field application (Kebreab et al., 2006; Sommer et al., 2004). Protocols for estimating CH_4 emissions from manure have been proposed by the Intergovernmental Panel on Climate Change (IPCC, 2016a). According to the guideline, when storing manure or mixing manure with other substrates prior to feeding on-site at the biogas facility, the CH_4 emissions from these activities need to be assigned to the biogas process (Liebetrau et al., 2017). CH_4 emission during the pre-storage process depends on the storage temperature, manure composition (e.g. organic matter degradability, ammonia concentration and pH) and the methanogenic community as modified by storage conditions and pretreatment (Chen et al., 2008; Elsgaard et al., 2016; Witarsa and Lansing, 2015; Zeeman, 1994). The temperature dependency of CH_4 emission can be described by Arrhenius-derived models, and is affected by storage substrate and adaptions (Elsgaard et al., 2016). Low storage temperatures will reduce CH_4 emission by decreasing the activity of methanogens but also that of other bacteria implied in methanogenic fermentation (Le Mer and Roger, 2001). There are extensive literature regarding CH_4 emissions response to storage temperatures. For instance, CH_4 emissions during manure storages have been observed at temperatures of 5 °C, but reach the maximum emission in the mesophilic temperature range (30–37 °C) (Cullimore et al., 1985). The default CH_4 emission factors given by IPCC is divided into three regions (cool, 10–15 °C; temperate, 15–25 °C and warm, > 25 °C) according to the annual temperature (IPCC, 2006b). In this study, storage temperatures between 15 and 30 °C were chosen as the on-line monitoring of storage temperature at a full-scale biogas plant in Denmark covered this range on a yearly basis (data not shown).

Beside CH_4 emission estimation, researchers have also focused on reduction of CH_4 emissions by manure acidification. Manure acidification using concentrated acids to reduce CH_4 emissions and sulphuric acid has been suggested for economic reasons as well as its fertilizer value (Ottosen et al., 2009; Petersen et al., 2012). Most methanogenic bacteria function in a pH range between 6.7 and 7.4, but optimally at pH 7.0–7.2 (Lay et al., 1997), while the optimum pH of hydrolysis and acidogenesis has been reported as being between pH 5.5 and 6.5 (Ward et al., 2008). The commercial system for slurry acidification will bring pH to 5.5, which inhibits both CH_4 production and sulfate reduction potentials. However, manure acidification will also lead to negative effects on AD since the growth and activity of sulfate

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Abbreviations

AD	Anaerobic digestion
B_0	Maximum or ultimate methane yield ($mLCH_{4}.g_{VS}^{-1}$)
B	Cumulative methane yield ($mLCH_4. g_{VS}^{-1}$)
CM	Cattle manure
FW	Food waste
GHG	Greenhouse gas
HRT	Hydraulic retention time
PM	Pig manure
OLR	Organic loading rate
TS	Total solids
SRB	Sulfate reducing bacteria
SMY	Specific methane yield
VFA	Volatile fatty acid
VS	Volatile solids

reducing bacteria (SRB) are encouraged using H_2SO_4 acidified manure (Moset et al., 2012). SRB themselves can compete with methanogens for electron sources such as acetate or hydrogen, and with obligate hydrogen producing bacteria for propionate (O'Reilly and Colleran, 2006). Hydrogen sulfide (H_2S) produced by SRB can result in an inhibition of anaerobic digestion or even total failure (Sutaryo et al., 2013). Food waste (FW) has low pH and high biodegradability, which generates large concentrations of volatile fatty acid (VFA) during AD (Zhou et al., 2018). Co-digestion of animal manure and FW have been widely reported as feasible for biogas production (El-Mashad and Zhang, 2010; Marañón et al., 2012). During the pre-storage process, addition of FW to manure could also reduce the pH and generate high concentrations of VFAs at anaerobic condition, which has inhibitory effect on the CH_4 emission by acting as an uncoupling agent of the cell membrane potential and thereby arrest microbial metabolism (Barret et al., 2013; Ottosen et al., 2009). Moreover, it has another possibility that higher CH_4 emissions are experienced after co-storage with FW due to the higher biodegradability, especially when stored at higher temperature. With this situation, the biogas plant could change the strategy to collecting the emitted CH_4 from the pre-storage tank as part of the biogas production. Therefore, it is essential to determine the temperature-dependent effect of co-storage with FW to see if this will have a positive or negative impact and the best strategies to either separate or mix manure with co-substrates can be selected.

Most of the published works regarding CH_4 emission from storage have been evaluated based on batch test results. However, manure storage at biogas plants tends to operate on a semi-continuous add-and-remove basis and homogenous mixing process also required when material is pumped into the reactor, thus the storage tank is comparable to an anaerobic digester with a very different biochemical environment compared to that of a batch process. This work used daily-fed anaerobic digester reactors to simulate full-scale manure storage tanks and their emission during short-term anaerobic pre-storage over a longer period to account for the possible acclimation of the microbial community over time. The ultimate CH_4 yield (B_0) after storage was also measured to determine the CH_4 losses during pre-storage.

2. Material and methods

2.1. Substrate

Pig manure (PM) and cattle manure (CM) were collected from animal housing storage tanks located at the research center, Aarhus University Foulum (8830 Tjele, Denmark) twice per month from February to May 2017. The cattle were fed with a mixture of maize silage and first cut grass (50%*w/w*) while pigs with the weight of 30–109 kg were fed with commercial pig feeds (DLG a.m.b.a, Denmark) and wheat straw (100 g animal^{-1}. d^{-1}). The animal housing storage tanks were agitated daily and emptied every two days. After collection, both CM and PM were stored using 40 L sealed buckets (fully filled with manure) at ambient temperature.

Food waste (FW) was collected from grocery stores (COOP and Dansk Supermarked) at Midtfyn (Fyn, Denmark). All collected FW were homogeneously crushed and mixed by a specialized company (NT environment, 5750 Ringe, Denmark) prior to the experiment. Collected samples (CM, PM and FW) were stored at −18 °C in a freezer and thawed before analysis. Characteristics of CM, PM and FW are listed at Table 1.

Table 1
Characteristics of cattle manure, pig manure and food waste.

Substrate	TS (%)	VS (%)	pH	VFAs (mg.L^{-1})	NH_4-N (g.L^{-1})
Cattle manure	8.2–8.8	6.6–7.3	6.6–6.9	7500–9800	1.5–1.9
Pig manure	1.2–1.5	0.8–1.2	6.2–6.7	2000–6000	0.6–1
Food waste	7.13	6.02	3.61	4700	0.2

2.2. Pre-storage test

The experiment lasted 95 days from February to May 2017 and was divided into three periods. The first period measured the emission of PM and CM separately. The second and third periods measured emission of manure mixed with food waste. The manure with food waste experiment was divided by two periods because the manures were stored at ambient temperature before being used for the pre-storage test. It was assumed that this temperature will affect CH_4 emission during the test, especially when the daytime temperature was higher (≥15 °C) (King et al., 2011). Because the 15 °C treatment was conducted in a room with heating but not cooling, the 3rd period ended when room temperature exceeded 15 °C due to high outside temperatures. The details of the three periods are shown below:

1) Days 1–52. Average climatic temperature 2.8 °C.

100% animal manure (PM and CM were examined separately);

2) Days 53–70. Average climatic temperature. 6.3 °C.

75% (*w/w*) PM or CM with 25% (*w/w*) FW, the maximum ambient temperature during most of the daytime was lower than 15 °C;

3) Days 71–97. Average climatic temperature 8.2 °C.

75% (*w/w*) PM or CM with 25% (*w/w*) FW, the maximum ambient temperature during the daytime was higher than 15 °C.

Eight lab-scale digesters with 15 L working volume were used to simulate the on-site pre-storage tanks at a biogas plant. In a real biogas plant, the retention time (HRT) for manure storage varied between few days to weeks based on the structures of main feedstock and treating capacities. In this study, the tanks were manually fed daily by an amount based on the hydraulic retention time (HRT) of 7 days, with a corresponding mass of material removed to maintain constant tank mass. Four storage temperatures (15, 20, 25 and 30 °C) were chosen to simulate temperatures that have been recorded at a commercial biogas plant (Maabjerg BioEnergy, Holstebro, Denmark). During the experiment, emitted gases were collected with 20L Tedlar ® sampling bags with double polypropylene septa. Gas emission was measured daily by connecting the bag to a tube with inlet to a column filled with acidified water (pH < 2) and gas volume was calculated by the water displaced until the two pressures (column and sampling bag) were equal. Pre-