Effects of anaerobic digestion and aerobic treatment on the reduction of gaseous emissions from dairy manure storages

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Abstract: Effects of anaerobic digestion and aerobic treatment on the reduction of gaseous emissions from dairy manure storages were evaluated in this study. Screened dairy manure containing 3.5% volatile solids (VS) was either anaerobically digested or aerobically treated prior to storage in air-tight vessels. Anaerobic digestion was carried out using a mesophilic anaerobic sequencing batch reactor operated at a hydraulic retention time of 20 days and an organic loading rate (OLR) of 1 gVS/L/day. Aerobic treatment was achieved using an aerobic reactor operated at a hydraulic retention time (HRT) of 10 days and an OLR of 2 gVS/(L·d). The treated manure was put into the storage on a daily basis for a period of 180 days. All the gases produced during this period were captured and analyzed for methane, carbon dioxide and volatile organic compounds (VOCs). Untreated manure was stored and analyzed in the same way as the treated manure and used as a control for comparison. The results show that low amounts of gases were produced during the first 84 days of storage in both treated and untreated manure, but increased significantly after this time point. The generally expected positive impacts of anaerobic and aerobic treatment on the reductions of methane and VOCs were confirmed in this study. However, the effects of anaerobic and aerobic treatment varied over the time of storage, especially for VOCs. The results of this study indicate that to achieve significant reductions in VOC emission the storage time of anaerobic digester or aerobic reactor effluent should be limited to no more than 100 days.

Key words: anaerobic digestion, aerobic treatment, methane, volatile organic compounds, dairy manure
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1 Introduction

Anaerobic digestion and aerobic treatment have been used, or are being adopted, on livestock farms to provide treatment for animal manure prior to storage. These methodologies have the potential to significantly reduce gaseous emissions from manure management systems. Previous research evaluating the air quality impact of anaerobic digestion and aerobic treatment technologies on animal manure focused mainly on odor control. The effectiveness of aeration on odor control for various types of animal manure has been well documented in the literature[1,2]. Previous research results[3-5] have indicated that anaerobically treated manure had lower odor intensity than untreated manure. Zhang et al.[6] evaluated the odor characteristics of untreated and anaerobically treated swine and dairy manure over a three-month anaerobic storage period and found that untreated manure had strong odors with high concentrations of volatile fatty acids (VFAs), hydrogen sulfide (H_2S) and methyl mercaptan (CH_3SH) in the emitted gases, while anaerobically digested manure had lower odors, VFAs and low or no detectable sulfur gases. Most of the research regarding volatile organic compound (VOCs) emissions from confined animal feeding

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operations has been focused on compounds associated with odors [7]. However besides causing odors, VOCs also contribute to the formation of ozone by reacting with nitrogen oxides in the air in the presence of sunlight[8]. The vast majority of confined animal feeding operations in California are located in the San Joaquin Valley which is in extreme nonattainment of current state and federal ozone standards. Reducing the emissions of VOC from livestock operations is an important task with respect to the reduction in ozone formation, as well as odor control. Several recent studies on the measurement of VOC emissions from dairies have been reported in the literature[8-11]. In light of the increased need for greenhouse gas emission reduction, it is important to quantify the effects of various treatment technologies for the reduction of methane and nitrous oxide.

Gaseous emissions from animal manure occur in two major steps: the production of gas in the manure and the release of the gas from the manure. In the present study focus was put on the production of methane and VOCs from treated and untreated manure during storage, as the production of the gasses was directly correlated to the emission of the gases. Reductions in the mass production of gasses in manure storage translate directly into the reductions in their emission. The objectives of this study were to determine and compare the total mass production of methane and VOCs during the storage of treated and untreated dairy manure, and to assess the impact of anaerobic digestion and aerobic treatment technologies for the reduction of gaseous emissions from manure storages. As a reference, the production of carbon dioxide from the stored manure (treated and untreated) was also measured and was presented. The changes in microbial populations within the manure storages as a result of manure treatment via anaerobic digestion and aerobic treatment were also examined and have been presented in a recent article by McGarvey et al.[12]. For this study, the gases produced from the anaerobic treatment processes were collected and combusted for energy production. The gases produced from the aerobic treatment process were released into the atmosphere. The impact of the gases released during the anaerobic or aerobic treatment processes was not the focus of this study.

2 Materials and methods

2.1 Experimental set-up and design

The experimental set-up as shown in Figure 1 was used to provide the anaerobic or aerobic treatment of dairy manure prior to the storage and to compare the treated manure with the untreated manure for their gaseous emissions over a six-month storage period. System 1 was designed to provide anaerobic treatment prior to the manure storage. System 2 was used as a control for System 1. System 1 employed an anaerobic digester with a 4L working volume, operated at (35±1)℃, with a hydraulic retention time (HRT) of 20 days, and a volatile solids (VS) loading rate of 1 g/(L·d). The digester was operated as a completely mixed digester, mixed for three minutes every hour and was fed with 200 mL prepared dairy manure once a day after 200 mL effluent was removed. In System 2 (control for System 1), the same amount of untreated manure was pumped into the storage vessel each day as in System 1. System 3 was designed to provide aerobic treatment prior to the manure storage. System 4 was used as the control for System 3. System 3 employed an aerobic reactor with a 2 L working volume operated at room temperature (20±2)℃, with a five day HRT, and a dissolved oxygen concentration of 2 mg/L. The aerobic reactor was fed once a day with 400 mL prepared dairy manure after 400 mL effluent was removed. Compressed air was supplied into the reactor via air diffusion stones installed at the bottom of the reactor at a rate controlled to maintain the DO at 2 mg/L, which was monitored using a DO probe and meter. In System 4 (control for System 3), the same amount of untreated manure was pumped into the storage vessel each day as in System 3. The conditions of both anaerobic digestion and aerobic reactor were designed to reduce the VS in the dairy manure by approximately 40% based on previous research by Li and Zhang[13,14]. The actual performance of the anaerobic digester and aerobic reactor showed an average of 55% and 43% of VS reduction, respectively, in the dairy manure over a six month period of operation.

Over a six month period, all four systems were fed from the same dairy manure prepared in the laboratory from fresh manure collected from the UC Davis dairy farm. The fresh manure was mixed with water and screened with a sieve with openings of 2 mm×2 mm to remove large particles, diluted with water to achieve the desired TS content of 4.4%, and placed into a feed tank maintained at 4℃. The TS of 4.4% was chosen because it is similar to the level observed in manure collected on a typical commercial dairy using flush and scrap waste removal systems.

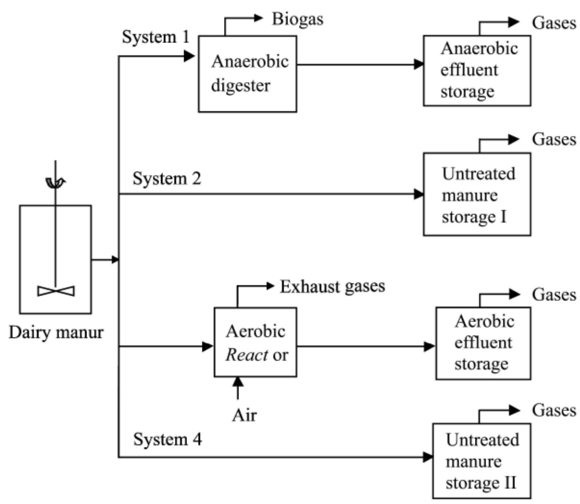

Figure 1 Experimental set-up for dairy manure treatment study

The treated and untreated control manures were placed in separate 80 L storage vessels. All four storage vessels were sealed air tight, and flushed filled with nitrogen gas to create an anaerobic environment similar to those encountered in anaerobic waste ponds found on commercial dairies. During the experiment, a 10 L tedlar bag was connected to the top of each storage vessel through a vent port to collect all of the gasses emitted from the waste. The gas volume in the bag and the headspace of the storage vessel were measured weekly. At the time of measurement, the two volumes were added to arrive at the total gas phase volume. Gas samples were analyzed for the concentrations of methane, carbon dioxide and VOCs. The mass of a particular gas (e.g., methane) present in the gas phase was calculated by multiplying the total gas phase volume by the concentration of that gas (e.g., methane). At the time of measuring the gas phase volumes, liquid volume in the storage vessel was also measured. The concentrations of the individual gases in the liquid phase were determined using Henry's Law, assuming equilibrium between the liquid and gas phases. The mass of the gas (e.g., methane) in the liquid phase was calculated by multiplying the liquid volume by the concentration in the liquid. The masses in the liquid and gas phases were added together to provide the total mass of the gas at the time of measurement. The amount of gas produced over the period of measurement was calculated by subtracting the mass at the previous measurement time from the mass at the current measurement time. The cumulative mass of a particular gas at a given time is normalized based on the amount of TS in the dairy manure that was fed into the system. The performance of the anaerobic digester and the aerobic reactor were monitored by taking samples from the influent and effluent of each reactor twice a week over, six month period and analyzed for TS, VS and BOD_5 using the standard methods[15]. The biogas production in the anaerobic digester was measured daily using a wet tip meter (http://wettipgasmeter.com/) and measured weekly for methane and hydrogen using a gas chromatograph equipped with thermal conductivity detector.

2.2 Chemical analysis

The methane and carbon dioxide contents of the gas samples were measured using a GC/TCD (5890 Hewlett-Packard Company, Wilmington, DE, USA). An Alltech carbosphere 80/100 column with dimensions of 6'× 1/8' SS was used. Helium was used as a carrier gas at a flow rate of 82 mL/min. The temperatures of the oven, injector and detector were 100ºC, 120ºC and 120ºC, respectively. The concentrations of VOCs in the gas samples were determined using the Gas Chromatography/Mass Spectrometry (GC/MS) system which consisted of a Varian CP-3800 Gas Chromatograph with built-in high performance sample concentrators and the Varian Saturn 2200 Ion Trap Mass Spectrometer (Varian, Inc., Palo Alto, CA, USA). The system meets the exact requirements for the US EPA TO-14 and TO-15 Methods[15]. The samples were trapped on a cryotrap at -130℃ and then purged with ultrapure N_2 to flush any sample remaining in the tubing or valving onto the cryotrap. After purging, the cryotrap was rapidly heated to 200℃ to transfer/desorb the contents and retrap them on the cryo-focus trap at -130℃. The cryo-focus trap was rapidly heated up to 200℃ to inject the sample onto a CP-Select 624 CB capillary column (60 m×0.32 mm× 1.8 μm). Helium was used as the carrier gas in constant flow mode at 2.0 mL/min. The GC oven temperature program was as follows: initial temperature -30℃ hold 5 min; ramp 5℃/min to 200ºC. The GC/MS was calibrated using a TO-15 calibration mix of 62 components with each at 100 ppb from Scott Specialty Gases (Plumsteadville, PA, USA).

The liquid samples taken from the feed tank, effluent from the anaerobic digester and the aerobic reactor, and the storage vessels were measured for total solids (TS), volatile solids (VS) and suspended solids (SS), biochemical oxygen demand (BOD_5), and other elements as shown in Table 1 using Standard Methods (APHA, 1995). All the analysis was performed in duplicate and reported as the average.

3 Results and discussion

The TS and BOD_5 of influents and effluents from the reactors (anaerobic digester and aerobic reactor) are shown in Table 1. The results of other analyses (nutrients and microbial populations) are presented in McGarvey et al.[12]. Aerobic treatment resulted in a 32% TS reduction, a 43% VS reduction and a 77% BOD_5 reduction in the dairy manure. Anaerobic treatment resulted in a 43% TS reduction, a 55% VS reduction and an 87% BOD_5 reduction as compared to the feed manure. The biogas yield of anaerobic digester was measured to be 0.32 L/gVS fed and average methane content in the biogas was 64.3%.

Table 1 Chemical analysis results of the feed and effluent of anaerobic and aerobic treatment reactors (values in the table are averages of 12 measurements)

Parameter	Dairy manure used as feed for treatment reactors	Anaerobic reactor effluent	Aerobic reactor effluent
TS/mg • L^{-1}	44,000	25,000	30,000
VS/mg • L^{-1}	35,200	15,900	20,100
BOD_5/mg • L^{-1}	14,900	1,900	3,500

The cumulative productions of methane, carbon dioxide and VOCs over time during the storage of treated and untreated manure are shown in Figures 2, 3, and 4, respectively. Gas production was reported as mg gas per gram of total solids in the dairy manure feed material and was calculated from the total cumulative amount of gas produced over the period divided by the total amount of total solids in the manure that was fed into the system. The data show that the productions of methane and carbon dioxide were low during the first 84 days; however, after this time point gas production increased quickly. Among the 51 VOCs included in the standard method of US EPA TO-15[16], the following compounds were detected in the gas samples taken from the manure storage vessels: acetone, 2-propanol, chloromethane, acetaldehyde, ethyl acetate, benzene, toluene, methyl iso-butyl ketone, trans-1,3-Dichloropropene, and 2-Hexanone. The major compounds emitted were acetone, 2-propanol, acetaldehyde, benzene and toluene. Also many other VOC compounds were likely produced from the dairy manure, but were not analyzed in this study.

The reduction in the amount of gasses produced (methane, carbon dioxide, and VOCs) from the dairy manure storage after 112 and 180 days as a result of aerobic or anaerobic treatment, compared to their respective controls (untreated manure storage), are shown in Table 2. Generally speaking, anaerobic treatment was more effective than aerobic treatment for the reduction of methane and VOCs production from manure storages. However, the effects of anaerobic and aerobic treatments varied over time; for example, after 112 days the anaerobic digester effluent storage had 63.13% less methane while the aerobic digester effluent storage had 22.26% increase in methane production. The latter was not expected but may be explained by the fact that the aerobic reactor effluent contained more VS than the effluent of anaerobic digester. It is also plausible that

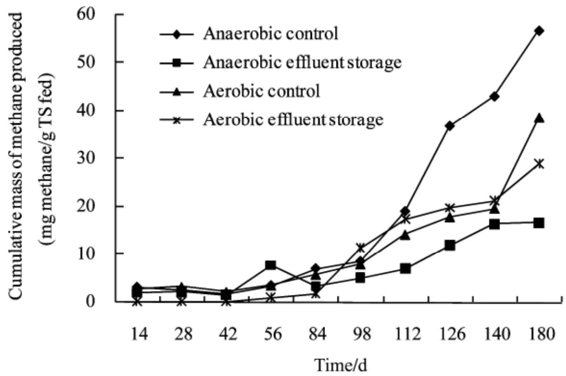

Figure 2 Cumulative methane production during storage of treated and untreated manure

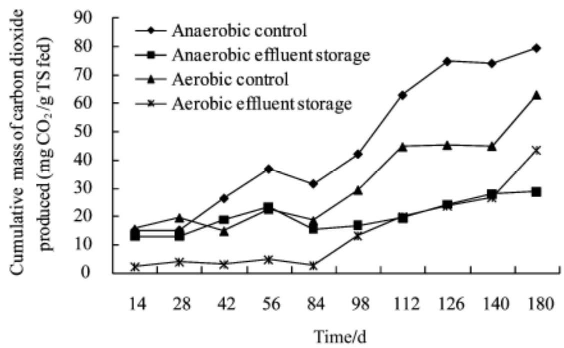

Figure 3 Cumulative production of carbon dioxide during storage of treated and untreated manure

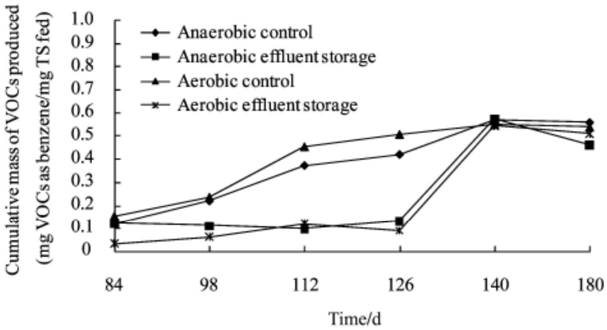

Figure 4 Cumulative VOCs production during storage of treated and untreated manure

the release of organic compounds from the breakdown of aerobic bacterial cells formed in the aerobic reactor contributed to the methane production. The VOC productions from the anaerobic and aerobic effluent storages are 74.05% and 73.98% lower, respectively, than those produced in the untreated manure storages (controls). Therefore, significant reductions in the production of methane and VOCs from the manure storage were achieved as a result of anaerobic treatment prior to storage. Significant reductions in VOC production from the manure storage were achieved as a result of aerobic treatment, but an increase in methane production from the manure storage was observed toward the 180 day storage.

After 180 days, the anaerobic digester effluent storage continued to show significantly less methane production (70.31%) and the aerobic reactor effluent storage showed a 24.58% reduction, as compared to the untreated manure storage (control). However, both the anaerobic digester and aerobic reactor effluent storages experienced significant production increases in VOC during the period from 112 to 180 days, diminishing the effects of these treatments. Compared to untreated manure storages (controls), anaerobic digestion and aerobic treatment resulted in 17.27% and 5.26% reductions in the cumulative production of VOCs in the storages. The increased production of VOCs after 112 days could be due to the decomposition of complex organic compounds in the dairy manure that were not degraded in the treatment reactors. This indicates that there is a need for limiting the storage time of manure after anaerobic and aerobic treatment if significant VOCs reductions are desired.

Table 2 Reduction (%) in production of different gases during dairy manure storage as a result of anaerobic and aerobic treatments as compared to their respective control

Gases	Storage period (days)	Anaerobic treatment	Aerobic treatment
Methane	112	63.13	-22.26*
	180	70.31	24.85
Carbon dioxide	112	68.95	55.17
	180	63.74	31.04
VOCs	112	74.05	73.98
	180	17.27	5.26

*indicates the increase of methane production.

4 Conclusions

The effects of anaerobic and aerobic treatment on the production of methane, carbon dioxide, and VOCs in manure storages were evaluated in this study. Generally speaking, very small amounts of gases were produced during the first 84 days of storage in both treated and untreated manure storages, with most of the gases being produced afterwards. The results of this study showed the positive impact of anaerobic and aerobic treatment on the reductions of methane and VOCs, except that the aerobically treated manure showed higher methane production in the storage than the control under the period of 98-140 days. This effect of aerobic treatment on the methane emissions needs to be further investigated. The effects of anaerobic and aerobic treatment varied with time during the storage, especially for VOCs. The results of this study indicate that to achieve significant reductions in VOC emissions from aerobic and anaerobically treated waste, the storage time for the effluent under the tested conditions should be limited to approximately 100 days.

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