PERMEABLE SYNTHETIC COVERS FOR CONTROLLING EMISSIONS FROM LIQUID DAIRY MANURE

A. C. VanderZaag, R. J. Gordon, R. C. Jamieson, D. L. Burton, G. W. Stratton

ABSTRACT. *Liquid manure storages emit greenhouse gases (GHGs) and ammonia (NH_3), which can have negative effects in the atmosphere and ecosystems. Installing a floating cover on liquid manure storages is one approach for reducing emissions. In this study, a permeable synthetic cover (Biocap ™) was tested continuously for 165-d (undisturbed storage + 3-d agitation) in Nova Scotia, Canada. Covers were installed on three tanks of batch-loaded dairy manure (1.3 m depth × 6.6 m^2 each), while three identical tanks remained uncovered (controls). Fluxes were measured using steady-state chambers. Methane (CH_4), carbon dioxide (CO_2), and nitrous oxide (N_2O) were measured by absorption spectroscopy, and NH_3 was measured using acid traps. Results showed covered tanks consistently reduced NH_3 fluxes by approximately 90%, even though a surface crust formed on controls after about 50 days. Covers continued to reduce NH_3 flux during agitation. Covered tanks also emitted significantly less CO_2 and N_2O than the controls (p-value <0.01). However, CH_4 fluxes were not reduced, and therefore overall GHG fluxes were not substantially reduced. Short-term trends in CH_4, CO_2, and N_2O flux provided insight into cover function. Notably, bubble fluxes were a key component of CH_4 emissions in both treatments, suggesting the covers did not impede CH_4 transport.*

Keywords. *Air quality, Emissions, Floating cover, Liquid manure storage.*

In many livestock production systems, manure is handled as a liquid and stored in tanks or lagoons until land-applied. These storage systems emit greenhouse gases (GHGs) including methane (CH_4), carbon dioxide (CO_2), and nitrous oxide (N_2O) (van der Meer, 2008), and ammonia (NH_3) (McGinn et al., 2008). Reducing emissions is important for addressing environmental concerns and for improving agricultural carbon (C) and nitrogen (N) conservation. Installing floating covers on stored liquid manure is one way some producers are trying to achieve this environmental goal. Covers are intended to provide a resistance to gas mass transfer from liquid to air, and to function as a biofilter (Miner and Suh, 1997), whereby microorganisms convert undesirable gases into more innocuous forms. Covers can be added to existing farm-infrastructure, and therefore have potential to be widely used. Synthetic covers are durable and unlike natural covers such as straw, do not impede pumping of the slurry. Permeable materials allow precipitation to seep though, eliminating the need for water removal on the cover. Permeable geotextile covers also have relatively low capital costs compared to some other permeable materials and impermeable covers (Nicolai et al., 2004).

A recent review (VanderZaag et al., 2008) identified that information about the effect of permeable synthetic covers on GHG emissions from manure storages was limited to a single study (Zahn et al., 2001). Furthermore, effects on NH_3 emissions were uncertain because some studies found NH_3 emissions were reduced (Miner et al., 2003; Portejoie et al., 2003), while others observed increased emissions (Clanton et al., 2001). Efficacy changes with time were also unclear, improving in some studies (Zahn et al., 2001; Miner et al., 2003) but worsening in others (Clanton et al., 2001; Bicudo et al., 2004). Whether gases are temporarily trapped in the liquid and subsequently released during agitation remains unclear (Bicudo et al., 2001).

Therefore, this study was conducted to assess the effect of a permeable synthetic cover (Biocap™) on GHG and NH_3 fluxes from stored liquid dairy manure. A research approach was chosen that exposes manure and covers to environmental conditions and agitation while allowing replication and frequent flux measurements. The specific objectives were to: (i) determine changes in CO_2, CH_4, N_2O, and NH_3 fluxes, (ii) characterize the effect of agitation, and (iii) evaluate short-term (minutes – hours) and long-term (days – months) flux trends.

METHODS

SITE DESCRIPTION

Liquid dairy manure was stored in six concrete tanks (surface area of 6.6 m^2 each, fig. 1a, b) at the Nova Scotia Agricultural College (NSAC) in Truro, Nova Scotia, Canada. Fresh manure from the NSAC dairy unit was loaded into the tanks to 1.3 m-depth (8.6 m^3) on 6 May 2008. No additional manure was added during the study. The next day, floating

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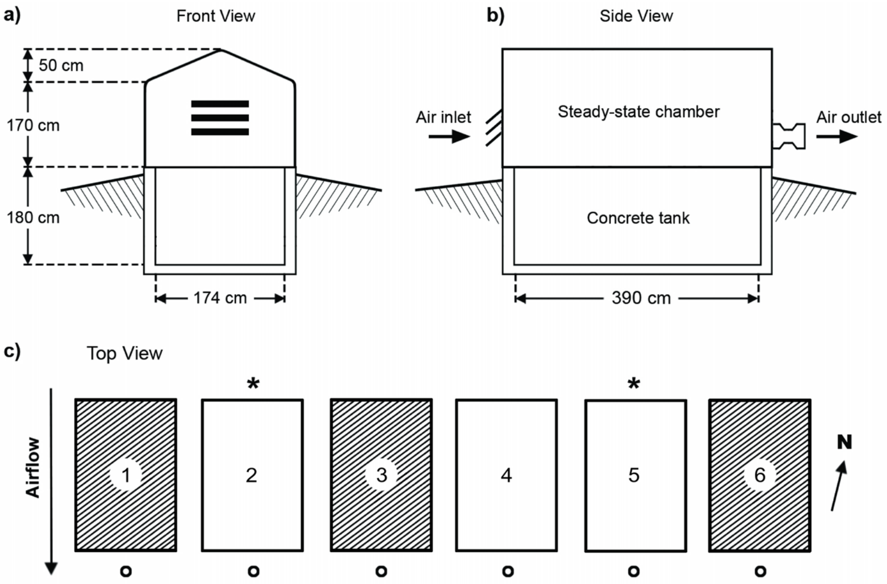

Figure 1. Diagram of the research site showing a cross-section of one manure storage tank and steady-state chamber from the front (a) and side (b), and a top view (c) of all six tanks indicating inlet (*) and outlet (o) air sampling locations. Tanks with BioCap™ covers are shaded, and control tanks are unshaded. All tanks were agitated at the end of the study. The space between tanks was 120 cm.

covers were installed in three tanks (fig. 1c). Commercially available Biocap™ covers (Baumgartner Environics, Olivia, Minn.) were used. These are a composite of approximately 1-mm acrylic-polyester geotextile-fabric adhered to 18 mm of permeable polyethylene foam (recycled, cross-linked closed cell foam). The covers were custom-made to fit tightly, and each had a small removable section (0.12 m^2) to allow access for manure sampling and agitation. The other three tanks were controls, which did not receive a synthetic cover, but were allowed to develop a natural crust.

Flux monitoring was conducted from 12 May through 28 October 2008 using steady-state chambers that exclude precipitation. To maintain an approximately neutral water balance (precipitation = evaporation) and provide a surface disturbance similar to rainfall, sprinklers inside each chamber were operated twice per week (30 mm wk^{-1}) through August, and once per week thereafter (15 mm wk^{-1}). A flow meter was used to ensure all tanks received the correct amount of water. Water was taken from a groundwater well (pH 7.9, nitrate-N <2.3 mg L^{-1}, Fe <0.02 mg L^{-1}, Mn < 0.02 mg L^{-1}, sulfate 43 mg L^{-1}). To monitor the water balance, freeboard was measured continuously in tank 3 (covered) and tank 4 (control) using SR50 sonic ranging sensors [Campbell Scientific (Canada) Corp., Edmonton, AB], and confirmed by manual measurements in all tanks.

Manure was agitated at the end of the study using three remote-controlled electric trolling motors (25-kg thrust, providing up to 70-W m^{-3} manure; Johnson Outdoors Inc., Racine, Wis.). First, tank-pairs were agitated intermittently for 8 h per day on three consecutive days, during which time the Biocap™ covers remained on. Then, covers were removed and intermittent agitation continued for two days (table 1).

MANURE SAMPLES AND CHARACTERISTICS

Monthly manure samples were taken at the near-surface, middle, and bottom of each tank and were refrigerated and analyzed according to recommended methods (Peters et al., 2003). Total ammoniacal N (TAN = NH_3-N + NH_4 $^+$-N) content was determined by distillation. Total Kjeldahl N (TKN) was determined by acid digestion. Total-C (TC) was determined using the Dumas method of combustion in a CNS analyzer (LECO Corp., St. Joseph, Mich.). Dry matter (DM) content was determined by drying manure samples (approximately 20 g) at 105°C, and volatile solids (VS) were then determined by loss-on-ignition at 550°C. The pH was measured potentiometrically using an electrode (Accumet; Fisher Scientific Inc., Waltham, Mass.). The E_H (Redox potential) of each sample was determined on-site, before refrigeration, with a calibrated electrode (Orion Star; Thermo Fisher Scientific Inc., Waltham, Mass.).

Table 1. Agitation schedule.[a]

Day[b]	Agitation[c]	Day	Agitation (cover removed)
1-3	T1, T2	4-5	T1
4-6	T3, T4	7-8	T3
7-9	T5, T6	10-11	T6

[a] There were insufficient mixers to agitate all tanks simultaneously, so each tank-pair was agitated simultaneously for 3 d (8-h d^{-1}). Afterwards, covers were removed and agitation continued in those tanks for 2 d.

[b] T = Tank; Day 1 = 18 Oct.

[c] Biocap™ covers on in T1, T3, and T6.

To measure crust thickness, an arrow-shaped probe was inserted through the crust, then rotated 90° and lifted until the shoulders of the probe-head met the bottom of the crust (modified from Smith et al., 2004). An average was calculated from five measurements along a central transect in each tank.

ENVIRONMENTAL MEASUREMENTS

Environmental parameters were recorded every 60 s using a data-logger [CR1000; Campbell Scientific (Canada) Corp., Edmonton, AB] that calculated hourly and 24-h averages. Air temperatures inside each chamber were measured by three shielded copper-constantan thermocouples suspended approximately 30 cm above the manure. Manure temperature was measured in each tank at 5 cm below the surface and 10 cm above the bottom. Net radiation was measured inside chamber 2 and 5 (described later) at 1.50-m height using net radiometers [Q-7.1; Campbell Scientific (Canada) Corp., Edmonton, AB]. Periodic manual measurements of manure, crust, or cover surface temperatures (depending on the treatment and presence of a crust) were taken with a non-contact infrared thermometer (42500; Extech Instruments, Waltham, Mass.).

FLUX MEASUREMENTS

Steady-State Chambers

Six steady-state flux chambers (fig. 1a, b) were installed on the tanks and remained in place at all times except during manual measurements (e.g., manure and crust sampling) and when agitators were installed or removed. Chambers were made with transparent greenhouse plastic (0.15-mm Super Durafilm 4; AT Plastic, Edmonton, AB) on aluminum frames. Fresh air entered the chambers through three vents and exited through a 35-cm diameter exhaust fan (Leader Fan Industries, Toronto, ON). Exhaust fan speed was set to provide a nominal air-exchange rate of 2 to 3 times per min, and it was consistent among chambers and through time. Airspeed near the manure surface was measured periodically with a hot-wire anemometer at 16 locations, ranging from 0.5 to 1 m s^{-1}. Exhaust ducts had a venturi shape to promote laminar airflow in the narrow section, where exhaust airspeeds were measured every 60 s using cup anemometers (7911, Davis Instruments, Hayward, Calif.). Hourly averages were recorded by a data-logger [CR10X; Campbell Scientific (Canada) Corp., Edmonton, AB]. Flux densities were calculated using the steady-state equation (Livingston and Hutchinson, 1995):

$$F = \frac{Q}{A}(C_o - C_i) \tag{1}$$

where

F = flux density (mg m^{-2} s^{-1})

Q = airflow rate (airspeed in the venturi × cross-sectional area of the venturi, m^3 s^{-1})

A = surface area of the manure tank (m^2)

C_i = gas concentration in the inlet air (mg m^{-3})

C_o = gas concentration in the outlet air (mg m^{-3}).

Inlet air was sampled at two points, 1.7 m above ground, 0.3 m in front of tanks 2 and 5 (fig. 1c). These samples were assumed to represent the inlet air of all chambers, so in calculations C_i was the average. For all gases, outlet air was sampled at the center of each exhaust duct. The chamber setup was tested before the study using a mass recovery of N_2O (Crill et al., 1995). A known mass of N_2O was added at the chamber inlets using a mass flow controller and certified standard gas (Air Liquide Canada Inc., Montreal, QC) while N_2O in exhaust air was monitored using 10-Hz data with the trace gas analyzer described later. The average recovery was 97%.

Despite their advantages, steady-state chambers alter the enclosed environment (Livingston and Hutchinson, 1995; Cole et al., 2007). As a result, absolute fluxes measured in this study have an uncertain relationship with the actual flux magnitudes that would occur without chambers. We assume relative flux versus time and treatment are representative of actual differences. Although measured fluxes are reported, trends and treatment differences are the focus of this analysis.

CH_4, N_2O, and CO_2 Measurement

Air from each sampling location (two inlet, six outlet) traveled through 25 m of polyethylene tubing (3.2 mm i.d.) to a valve box where air from one of eight sites was directed to a high-flow air dryer and then to one of two tunable diode laser trace gas analyzers (TDLTGA, Campbell Scientific, Logan, Utah) that measured the CH_4 and N_2O concentration. While cycling sequentially through the eight sites, air was constantly drawn through all valves and tubing by sending air from the remaining valves directly to the vacuum pump (bypassing the analyzers). Airflow in each sample tube was set to 0.9 L min^{-1} by an orifice at the intake (D-12-BR, O'Keefe Controls Co., Turnbull, Conn.). Certified reference gases (Air Liquide Canada Inc., Montreal, QC) were used in the TDLTGA reference cell. Reference gases that bracket the measurement range were used for span calibrations. Concentration data, parameters, and diagnostics from the TDLTGA were recorded by a data-logger [CR5000, Campbell Scientific (Canada) Corp., Edmonton, AB] that also controlled valves and recorded an average concentration from each location every 4 min (i.e. one cycle of eight sites at 30 s per site). When switching between sites, data were omitted during the sample crossover period. The average coefficient of variation (CV) for ambient samples during typical operation was 2.5% for CH_4 and 0.5% for N_2O.

The CO_2 concentration at each sampling location was determined using a similar set-up with the following differences: no external air dryers were used, an infrared gas analyzer (Li-Cor 6400; LI-COR Biosciences, Lincoln, Nebr.) with a N_2 reference gas measured CO_2 concentration at each site for 45 s on an 8-min cycle. The average CV for ambient samples during typical operation was 1%.

Due to power outages, equipment repairs, and maintenance, data were not obtained from: 18-19 May, 12-13 June, 20-21 July, 31 July to 7 August for CH_4; 21-27 May, 27 June to 2 July, 17-21 July, 2-4 August for N_2O; and 8-12 June, 11-15, 26-29 July, 8-17 September, 14-15 October for CO_2.

NH_3 Measurement

Air from each sampling location traveled through 25 m of polyethylene tubing to an ammonia trap. Sample air was bubbled through 100 mL of 0.005 M H_3PO_4 (Chantigny et al., 2004) using a dispersion tube (Ace Glass, Vineland, N.J.). Airflow in each tube was regulated by a 3-L min^{-1} orifice (O'Keefe Controls Co., Turnbull, Conn.) between the suction pump and an airflow meter (Gallus 2000, Actaris Metering Systems, Greenwood, S.C.). All sample locations were monitored simultaneously using eight traps. For practical reasons, a sampling interval from 0830 h to 0830 h (the next day) was used to measure daily average NH_3 flux. Samples were typically obtained three days per week except during agitation when samples were obtained each day. The CV for ambient samples measured in the same week was 5% to 30%.

After a 24-h sampling period, a 13-mL subsample from each trap was immediately refrigerated in a capped plastic tube. The aqueous NH_4^+-N concentration was determined by the phenate method using a Technicon AutoAnalyzer II

(Technicon Instruments Corp., Tarrytown, N.Y.). The aqueous concentration was used to calculate the average NH_3-N concentration in sample air:

$$C_{air} = C_{aq} \times V_{aq} / V_{air} \tag{2}$$

where

V_{aq} = trapping-solution volume (m^3)

V_{air} = sample-air volume (m^3)

C_{aq} = NH_4^+-N concentration in the trapped liquid ($mg\ m^{-3}$)

C_{air} = NH_3-N concentration in air ($mg\ m^{-3}$), which is either C_o or C_i (eq. 1) depending on sample location.

DATA ANALYSES

Data processing and flux calculations were performed using MATLAB® (The Mathworks Inc., Natick, Mass.). Covers were randomly assigned using adjacent tank-pairs as blocks to minimize potential effects of spatial variability and micro-climates. Since flux measurements were taken across time from each manure tank, repeated measures analysis was used to compare fixed effects of cover, time, and agitation. The random effects of tank and block were also included in the model. This analysis was performed on daily average data, using PROC MIXED (SAS Institute, 2008). Repeated measurements were not equally spaced (due to data gaps), so measurements closer in time were more correlated than those that were farther apart. Covariance structures suitable for unequally spaced data were selected based on fit-statistics (Littell et al., 1998; Littell et al., 2006). Regression analysis and tests on non-repeated sample means were conducted using JMP® (SAS Institute, 2007).

A key assumption in flux calculations was that ambient samples represent all inlet air. When valid, the difference between ambient samples should be zero. Thus, for each gas, the concentration difference between concurrent ambient samples was calculated along with a 5-period running standard deviation (SD) of the differences. Consecutive differences not bound by 0 ± 2SD were flagged and associated data were checked and manually removed. This procedure caused <2% of CH_4, N_2O, and NH_3 data to be removed. However, many CO_2 data were removed especially during nights with low wind. This can be explained by advection of CO_2-rich air (due to respiration) from the surrounding landscape during stable atmospheric conditions. Similar problems occur in eddy covariance CO_2-flux measurements (Baldocchi, 2003). Removing these data should not bias results because CO_2 flux did not exhibit a diurnal trend in either treatment.

RESULTS AND DISCUSSION

ENVIRONMENTAL PARAMETERS

Manure Temperature

Altering manure temperature by covering can affect fluxes through the rate of microbial gas production (Conrad, 1996). The covers had a dark surface that heats up due to insolation. However, insulating material lining the cover will reduce the transfer of heat to the manure. To investigate the potential warming effect on the manure in the tanks, infrared surface temperature was measured. The geotextile surface of covered tanks were always significantly warmer than the surface of control tanks, especially on sunny days. For example, at mid-day on 20 June, the average IR-temperature for covered tanks was 50 ± 5°C compared to 29 ± 4°C for controls, but manure temperature at 5-cm depth was similar (17.1 ± 0.5°C compared to 16.1 ± 0.5°C). Late in the year, and on cloudy days, thermocouple data showed covered tanks had warmer near-surface manure temperatures on a daily basis (p-value <0.05; fig. 2), presumably due to insulation. This was evident in hourly thermocouple data, for instance at mid-day on 25 September, the temperature at 5 cm in covered tanks was 16.6 ± 0.3°C compared to 12.8 ± 0.5°C in control tanks.

Water Balance

The covered tanks had less evaporative losses, leading to a progressive depth increase (fig. 3). In total, 2800 L (422 mm) of water was added to each tank with sprinklers. Overall, control tanks had approximately neutral water balances, indicating a 2.6-mm d^{-1} evaporation rate; whereas depth increased in covered tanks, indicating a significantly lower evaporation rate of 1.4 mm d^{-1} (p-value <0.01; fig. 3, right-panel). Reduced evaporation observed in the present study may result from reduced convection, perhaps further reduced by particles in the manure plugging pores in the cover. Less evaporation would reduce available freeboard and increase transport costs.

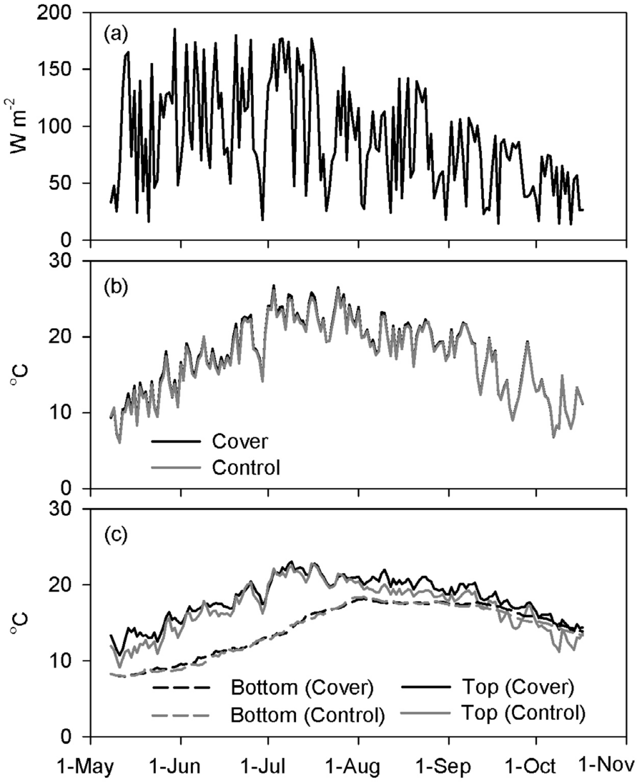

Figure 2. Daily average environmental parameters at the site: (a) net radiation, (b) air temperature (treatment average), and (c) manure temperature measured near the top and bottom of each tank (treatment average).

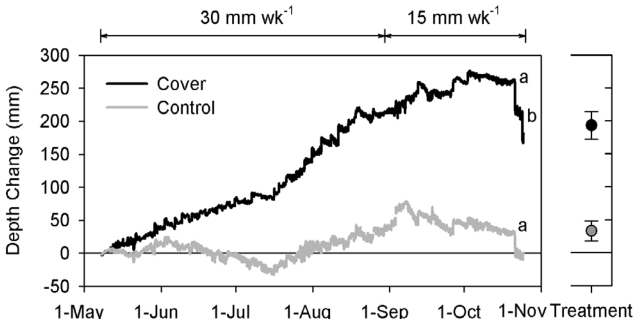

Figure 3. Cumulative depth change in tank 3 (cover) and tank 4 (control), measured hourly with sonic ranging sensors. Positive depth changes imply precipitation exceeds evaporation. Precipitation was simulated using sprinklers inside each chamber, supplying the rate shown above. Labels indicate the start of agitation (a), and cover removal (b). The right panel (treatment) shows overall depth change for each treatment based on freeboard measurements at the start and end of the study (three tanks each; mean ± standard deviation).

MANURE CHARACTERISTICS

Manure analyses are summarized in table 2. Only results from top and bottom sample locations and the first and last sample dates are shown. Changes with time were gradual for all parameters, except TAN and TKN where most of the decrease occurred in May and June. Samples from mid-depth were similar to samples from the top. No significant treatment differences were observed for concurrent samples of any parameter at any depth, suggesting covers did not alter manure characteristics. Significant changes through time were observed within each treatment. This included decreasing all forms of N (confirming N-loss implied by flux measurements), increasing pH (favors higher NH_3 flux), and increasing E_H (less favorable to CH_4 production, though still below the +50 mV threshold for methanogenesis; Conrad, 1996).

FLUXES DURING UNDISTURBED STORAGE

CH_4, CO_2, and N_2O Fluxes

There was no significant difference in CH_4 fluxes between treatments (table 3). Significant changes did occur with time, indicating a lag-phase of approximately 50 d before CH_4 flux increased exponentially (fig. 4). A lag is expected for fresh manure stored in clean tanks (without inoculum; van der Meer, 2008) and was comparable to the delay reported by Massé et al. (2008). There was a significant difference in CO_2 emissions between treatments (table 3). The treatment × time interaction was also significant, reflecting that covers initially reduced CO_2 fluxes 20% to 35%, but after a crust formed on control tanks the covers no longer had an impact on CO_2 flux (fig. 4, table 4). Fluxes of CH_4 and CO_2 peaked simultaneously, coinciding with crust formation (fig. 4). Presumably, biogas (CH_4 + CO_2) production exceeded diffusion, causing bubbles that carried particles to the surface (Misselbrook et al., 2005). Bubbles were also visibly lifting particles in covered tanks; however, particles remained submerged due to the cover and positive water balance.

Table 3. Significance levels (*p*-values, or ** for *p*-value < 0.01) of the main effects and interactions on daily average flux of methane (CH_4), carbon dioxide (CO_2), nitrous oxide (N_2O), and ammonia (NH_3) using repeated measures.

Source[a]	CH_4	CO_2	N_2O	NH_3
Treatment	0.51	**	**	**
Time	**	**	**	**
Agitation	**	**	0.66	0.08
Treatment × Time	0.23	**	**	**
Treatment × Agitation	**	**	0.37	0.17
Agitation × Time	**	**	0.61	**
Treatment × Agitation × Time	**	**	0.48	**

[a] Data obtained after covers were removed during agitation are not included.

Table 2. Manure characteristics at the start (9 May) and end (17 October, prior to agitation) of the study.[a]

		Control[b]		Cover[b]	
	Time	Top	Bottom	Top	Bottom
Dry matter (DM, %)	Start	2.2 (0.3)^{ab}	7.7 (0.4)^{ab}	2.2 (0.3)^{b}	7.3 (0.3)^{ab}
	End	2.5 (0.5)^{ab}	5.2 (1.1)^{ab}	1.9 (0.2)^{b}	4.2 (1.6)^{ab}
Volatile solids (% of DM)	Start	70 (2)^{b}	87 (1)^{b}	71 (2)^{b}	85 (2)^{b}
	End	75 (7)^{b}	87 (3)^{b}	70 (2)^{b}	86 (6)^{b}
Total carbon (%)	Start	0.9 (0.1)^{b}	3.4 (0.2)^{ab}	0.9 (0.1)^{b}	3.2 (0.1)^{ab}
	End	1.0 (0.3)^{b}	2.3 (0.5)^{ab}	0.8 (0.1)^{b}	1.8 (1.7)^{ab}
Total ammoniacal N (mg L^{-1})	Start	1730 (287)^{a}	2230 (455)^{a}	1650 (144)^{ab}	2173 (140)^{ab}
	End	1013 (31)^{a}	1063 (15)^{a}	1053 (45)^{a}	1117 (61)^{a}
Total Kjeldahl N (mg L^{-1})	Start	2213 (136) ^{a}	2597 (169) ^{a}	2213 (127) ^{a}	2547 (189) ^{a}
	End	1590 (60) ^{a}	1727 (6) ^{a}	1550 (142) ^{a}	1783 (140) ^{a}
pH	Start	6.8 (0.0)^{a}	6.7 (0.1)^{a}	6.8 (0.0)^{a}	6.7 (0.1)^{a}
	End	7.5 (0.1)^{a}	7.5 (0.1)^{a}	7.5 (0.0)^{a}	7.5 (0.0)^{a}
E_H (mV)	Start	-178 (47)^{a}	-147 (9)^{a}	-202 (10)^{a}	-145 (30)^{a}
	End	-39 (18)^{a}	-51 (3)^{a}	-46 (3)^{a}	-45 (21)^{a}

[a] Samples were taken below the cover or crust, and at the bottom. The mean of three tanks in each treatment is shown with standard deviation in parentheses.

[b] There were no significant differences between treatments at the same time and sampling depth. For each parameter, superscripts indicate significant differences (*p*-value < 0.05) between the start and end in the same sampling location ('a'), and between depths at the same time in the same treatment ('b').

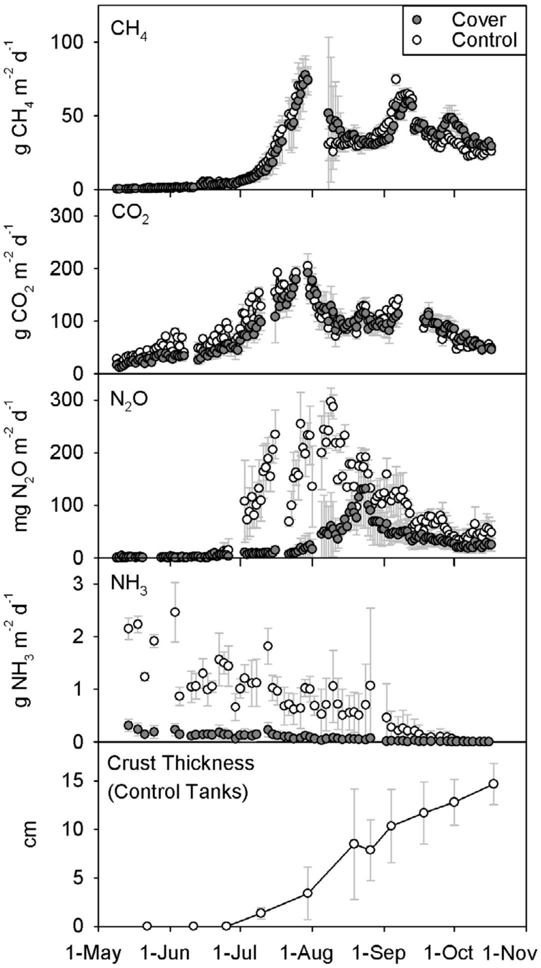

Figure 4. Daily average gas fluxes for each treatment and crust thickness on the controls during undisturbed storage (circles represent the average of 3 replicates in each treatment, whiskers are the standard deviation).

N_2O was emitted in control tanks about 1 month before fluxes were observed in covered tanks (fig. 4). A neutral or negative water balance favors N_2O production in surface crusts because of microbial activity in aerobic microsites (Sommer et al., 2000). Fluxes from covered tanks did not begin until late July, suggesting the covers were not as conducive for producing and emitting N_2O. For N_2O, effects of treatment, time, and treatment × time were all significant (table 3), reflecting flux-reductions provided by covers after crust development (48% to 93%; table 4).

NH_3 Emissions

Ammonia flux significantly declined with time for both treatments (fig. 4; table 3), a trend observed in other batch-loaded studies (Xue et al., 1999; Misselbrook et al., 2005). A concurrent decline in TAN confirmed that N was lost, which would lead to lower NH_3 emissions. The cover treatment had significantly lower fluxes (table 3) and provided about 90% flux-reductions for most of the study (table 4) despite declining fluxes and crusts on control tanks. Surface resistance is one potential reason why the Biocap™ covers reduced NH_3 fluxes. Another is that the covers reduced evaporation, therefore diluting the manure and decreasing TAN concentration near the surface of covered tanks. Lower flux-reductions in October (30%) coincided with cool temperatures, low fluxes, and the crusts on control tanks reaching a maximum thickness of 14 cm.

EMISSIONS DURING AGITATION

In both treatments, agitation led to significant increases in 24-h fluxes of CO_2 and NH_3, decreased N_2O, and had no effect on 24-h fluxes of CH_4 (fig. 5). During agitation, hourly fluxes of CH_4 (in both treatments) and CO_2 (control treatment only) exhibited similar trends to what was observed in a previous study (VanderZaag et al., 2009) — spiking 2- to 5-times higher than summer maximums. However, the spikes were offset by low fluxes when mixers were off (approximately zero flux for CH_4). Thus, there was little change in the 24-h average flux of CH_4. The possibility that agitation caused unsuitable conditions for methanogenesis was examined by frequent pH and E_H measurements, but no significant changes were observed. The N_2O fluxes declined in control tanks presumably because crusts were destroyed, eliminating N_2O production sites (Sommer et al., 2000). In comparison, our previous study found agitation had no effect on N_2O flux because N_2O emissions had already declined to zero before agitation started (VanderZaag et al., 2009). Ammonia fluxes from covered tanks were similar to pre-agitation levels in October; whereas fluxes from controls increased significantly. Thus, covers continued to provide high NH_3 flux-reductions (94%; table 4) demonstrating that surface resistance was effective even when manure was agitated. When the covers were removed, substantial increases in NH_3 and CO_2 fluxes were observed (fig. 5). In just two days, NH_3 lost from previously covered tanks was approximately 20% of total losses with the covers on (165 d). Thus, leaving the covers in place during agitation maintains NH_3 flux-reductions. It also suggests that covers were enhancing resistance to CO_2 transport.

OVERALL COVER EFFICACY

Overall, during 162 d of undisturbed storage and 3 d of agitation (with covers), Biocap™ covers provided significant ($p < 0.05$) emission reductions of CO_2 (15%), N_2O (68%), and NH_3 (89%) as shown in table 4. Total GHG emissions from both treatments were dominated by CH_4 emissions (converted to CO_2-equivalent global warming potential; CO_2e). If CO_2 emissions are excluded from the GHG-total, then there was no difference between treatments (table 5). If CO_2 emissions are included, covers reduced GHG emissions by 2.5% (*p*-value <0.05). Including indirect N_2O emissions does not change these conclusions (i.e. 1% of NH_3-N emissions; Solomon et al., 2007; van der Meer, 2008). The observation that covers reduced three of four gases, but did not substantially reduce total GHG emissions confirms that decreasing CH_4 emissions is imperative for liquid manure.

CONTEXT FOR EMISSIONS

Fluxes of CH_4 after the lag-phase (monthly averages: approximately 23 to 35 g CH_4 m^{-3} d^{-1}) were comparable to fluxes from stored dairy manure (approximately 16 to 56 g CH_4 m^{-3} d^{-1}; Sneath et al., 2006). The cumulative CH_4 emissions were approximately 3 kg CH_4 m^{-3} (4400 L CH_4 per m^{-3} manure). This corresponds to 0.14 L CH_4 g^{-1} VS (assuming top, middle, and bottom samples each represent

Table 4. Average gas flux from each treatment (three replicates; standard deviation in parentheses), and flux-reduction provided by the cover treatment. Total emissions are shown at the bottom of the table.

		CH_4 Flux (g CH_4 m^{-2} d^{-1})			CO_2 Flux (g CO_2 m^{-2} d^{-1})			N_2O Flux (mg N_2O m^{-2} d^{-1})			NH_3 Flux (mg NH_3 m^{-2} d^{-1})		
Period	Crust[a] (cm)	Control	Cover	% Redn.[b]	Control	Cover	% Redn.	Control	Cover	% Redn.	Control	Cover	% Redn.
9-31 May	0 (0)	1 (0)	1 (0)	0	38 (2)	25 (3)	34	1 (1)	1 (2)	0	1,885 (60)	221 (94)	88
June	0 (0)	3 (0)	3 (1)	0	62 (4)	40 (8)	35	3 (4)	3 (1)	0	1,278 (130)	143 (82)	89
July	2 (1)	33 (5)	29 (9)	12	148 (4)	118 (19)	20	154 (14)	11 (8)	93	997 (264)	121 (66)	88
Aug.	8 (4)	32 (4)	36 (7)	-13	106 (1)	109 (12)	-3	174 (16)	72 (12)	59	692 (555)	55 (22)	92
Sep.	11 (3)	46 (2)	44 (3)	4	100 (9)	95 (14)	5	85 (21)	41 (17)	52	183 (228)	17 (11)	91
1-17 Oct.	14 (1)	26 (2)	35 (3)	-35	56 (6)	60 (7)	-7	44 (19)	23 (11)	48	10 (4)	7 (0)	30
Agitation	0	28 (4)	29 (5)	-4	241 (100)	78 (15)	68	15 (5)	13 (3)	8	666 (362)	41 (35)	94
Totals[c]	Days	(g CH_4 m^{-2})			(g CO_2 m^{-2})			(mg N_2O m^{-2})			(mg NH_3 m^{-2})		
Undisturbed	162	3,950	4,043	-2	14,560	12,682	13	13,579	4,307	68	139,714	15,458	89
Agitation	3	84	87	-4	723	234	68	45	39	13	1998	123	94
Total:	165	4,034	4,130	-2	15,283	12,916	15	13,624	4,346	68	141,712	15,581	89
Total: kg CO_2e m^{-2}		100.9	103.3		15.3	12.9		4.1	1.3				

[a] Crust thickness is the average (standard deviation) of control tanks.

[b] % Reduction = $([F_{control} - F_{cover}] / F_{control}) \times 100$; where $F_{control}$ and F_{cover} are the flux in control and covered tanks, respectively.

[c] Calculated by multiplying treatment-average flux for each period by the number of days in each period and summing. Emissions were converted to CO_2 equivalent (CO_2e) on a 100-yr time horizon using CH_4 = 25 and N_2O = 298 (Solomon et al., 2007).

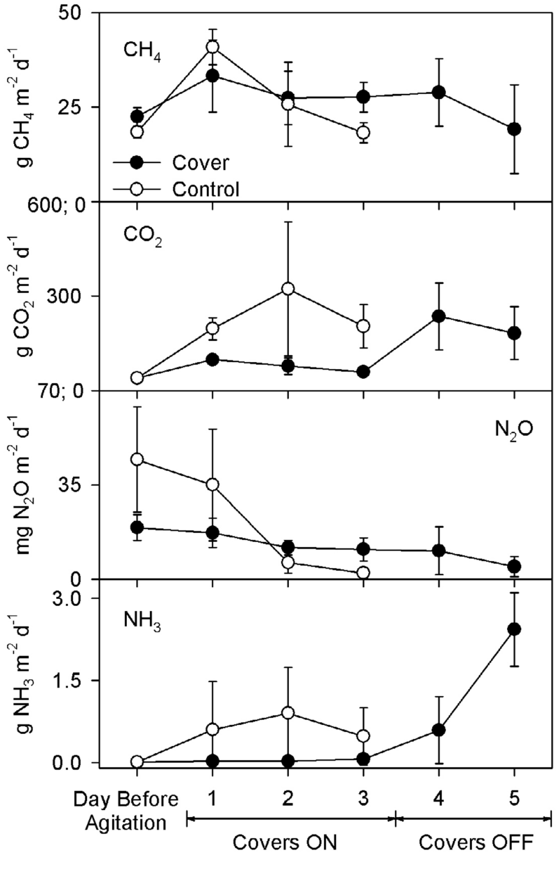

Figure 5. Daily average flux from covered and control tanks before and during agitation. Circles represent treatment means, whiskers the standard deviation. On days 1-3, covered tanks were agitated with covers in-place; whereas, on days 4 and 5 covers were removed and agitation continued. Control tanks were agitated for 3 days.

Table 5. Cumulative GHG emissions during 162 d of storage and 3 d of agitation.[a]

	Control	Cover	Reduction[b]
CH_4	100.9 (1)	103.3 (2)	n.s.
N_2O	4.1 (0.1)	1.3 (0.3)	68.5% *
GHG total	105.0 (1.4)	104.6 (1.7)	n.s.
CO_2	15.3 (0.2)	12.9 (1.1)	15.5% *
GHG total including CO_2	120.3 (1.2)	117.5 (1.2)	2.5% *

[a] Values are the mean (standard deviation) of three tanks in each treatment (kg CO_2e m^{-2}). The total GHG emissions are shown with and without including CO_2 (since it is not a net contribution to atmospheric CO_2.

[b] % Reduction = $([F_{control} - F_{cover}] / F_{control}) \times 100$; where $F_{control}$ and F_{cover} are the flux in control and covered tanks, respectively. Only statistically significant reductions are shown (n.s. for p-values > 0.05, * for p-values < 0.05).

1/3 of the tank) and a methane conversion factor (MCF) of 55% using a B_0 value of 0.24 L of CH_4 per g of VS in the manure (Zeeman and Gerbens, 2000). This MCF is higher than default values for manure tanks in cool and temperate climates (39% and 45%; IPCC, 2000). The discrepancy could be because the IPCC defaults are average values and do not account for the warm-season monitoring period, modified climate inside the chambers, and batch-loading used in the present study.

Fluxes of CO_2 were consistent with expectations for anaerobic breakdown of organic matter, as evidenced by the approximately 50:50 CO_2-C:CH_4-C ratio (after CH_4 lag-phase; Conrad, 1996).

For N_2O, the maximum 4-min flux from a control tank was comparable to the maximum flux measured at mid-day from crusted dairy slurry (893 vs. approximately 950 mg N_2O m^{-2} d^{-1}; Sommer et al., 2000). Pre-crust NH_3 fluxes from the controls were higher than one lab-scale chamber study (up to 0.75 g NH_3 m^{-2} d^{-1}; Xue et al., 1999), lower than another (3.6 to 6 g NH_3 m^{-2} d^{-1}; Sommer et al., 1993), and lower than field-scale emissions (daily average: 3.6 to 8.6 g NH_3 m^{-2} d^{-1}; McGinn et al., 2008).

Reductions of CH_4 and NH_3 flux can be compared to previous studies on permeable synthetic covers, but no reports were found on CO_2 or N_2O emissions. Some studies used geotextile covers, and others used Biocap™ covers. The only study on CH_4 flux found no effect initially, but after one month, the covered section of a lagoon had significantly higher fluxes than an uncovered section of the same lagoon. This increase was attributed to higher methanogenesis, although methanogenesis was not measured directly (Zahn et al., 2001). In our study, however, these treatment differences and trends were not observed. Thus, more research is needed to determine whether permeable synthetic covers tend to increase CH_4 production or perhaps could be designed to reduce CH_4 emissions by hosting methanotrophs (Petersen and Miller, 2006). Fluxes of NH_3 were reduced more (and more consistently) in our study than in previous field-studies on swine lagoons (17% to 54%, Zahn et al., 2001; 29% to 45%, Bicudo et al., 2004) and a pilot-scale study using dairy manure (geotextile did not reduce NH_3 flux; Clanton et al., 2001). A potential explanation for the enhanced performance in our study is that covers maintained 100% buoyancy, whereas others observed sinking — at least partially caused by snow accumulation (Bicudo et al., 2004).

SHORT-TERM FLUX TRENDS

Short-Term Trends in CH_4 and CO_2 Fluxes

Daily and monthly averages might suggest CH_4 fluxes from covered and uncovered tanks were nearly identical. This was not the case, however, on a short timeframe (fig. 6). Fluxes of CH_4 and CO_2 (fig. 7) were strongly influenced by short-term events, a characteristic that has been previously noted for CH_4 (Husted, 1994; Kaharabata et al., 1998; Park et al., 2006; Sneath et al., 2006). The flux-trend from any tank consisted of two main components: a baseline flux, presumably due to diffusion; and intermittent bursts, presumably due to bubble flux (ebullition). In control tanks, bubbles were periodically seen emerging through cracks in the crusts; whereas in covered tanks bubbles were not visible — even at the edges. Despite similar flux trends, there was no correlation within or between treatments. For example, high CH_4 flux observed in one tank did not predict concurrent high fluxes elsewhere. This suggests the cycle of gas production, bubble accumulation, and release, was independent of treatment and external factors. The covers might be expected to trap bubbles underneath and force more CH_4 to move by diffusion. However, data indicate that transport was still sporadic. Accounting for bubble flux is essential and may explain variability in previous studies where intermittent, short-duration measurements were taken (e.g. Husted, 1994; Sommer et al., 2000; Laguë et al., 2005). Our study suggests short measurements (minutes to hours) are inadequate for assessing CH_4 fluxes from liquid manure. Consider data from 15 to 18 August (fig. 6). A treatment comparison at one instant could show the cover was reducing CH_4 flux from -1400% to +87%. Even 20-min averages yield a range from -149% to +64%. In comparison, the treatment effect was -9% when 4-min data are averaged over 3 days. Thus, to capture the net production rate, CH_4 flux measurements must be frequent and long enough to average over stochastic transport processes — "snapshots" are inadequate. Another implication is that a wide flux range should be expected, so outlier removal should be done carefully. Care is also warranted for high-frequency measurements, since spike-removal algorithms could remove meaningful data.

Short-Term Trends in N_2O Flux

N_2O fluxes showed a diurnal trend. For example, from 15 to 18 August (fig. 6), the coefficient of determination (r^2) between N_2O flux and chamber air-temperature-squared (T_{air}^2) was 0.88 for covered manure and 0.91 for the control (both regressions, $p < 0.001$; data during simulated rainfall was removed). Two implications are: (i) short, mid-day flux measurements (e.g., Sommer et al., 2000) will tend to overestimate the daily average N_2O flux, and (ii) cooler surface temperatures should reduce N_2O flux, thus shaded storages and reflective covers may be advantageous.

Fluxes During Rain Events

Flux events during rainfall have little effect on overall emissions, but give insight into gas production and transport (fig. 6). For example, on 7 July, CH_4 emissions from the control (tank 4) spiked when sprinklers were on. This was likely a result of bubbles released from particles at the surface (the crust was <2 cm thick, so the physical disturbance was noticeable). In contrast, CH_4 flux in covered tanks dropped to zero; presumably, because water acted as a sealant while percolating through the cover. Fluxes of N_2O also dropped in both treatments and then rapidly returned to previous trends, suggesting lower fluxes were due to restricted transport, not decreased production.

CONCLUSION

Our results, from batch-loaded, pilot-scale dairy manure tanks frequently monitored for six months, show tanks with a Biocap™ floating cover emitted significantly less CO_2 and N_2O than controls. However, CH_4 emissions were not reduced, and since CH_4 represents the largest portion of total GHG emissions, total GHG emissions were not reduced. Thus, permeable covers designed to reduce CH_4 fluxes are needed. NH_3 fluxes were consistently reduced by approximately 90%, even though a crust formed on the undisturbed controls after about 50 days (which reduced fluxes in the control tanks). Excellent flux-reductions were also observed during agitation. Removing covers before agitation, however, led to greater losses of CO_2 and NH_3. Thus, being able to agitate manure below floating covers is beneficial, and may be preferable to materials that disintegrate during mixing.

Short-term (4-min) CH_4 flux data showed that bubble fluxes were a key component of fluxes from both covered and uncovered storages. This observation suggests that the Biocap™ cover does not substantially impede CH_4 transport. Moreover, bubble fluxes emphasize that brief measurement "snapshots" are inadequate for accurately measuring CH_4 fluxes from liquid manure, or even for making valid comparisons among treatments. The N_2O flux from Biocap™-covered and naturally crust-covered storages both had a diurnal trend that was strongly correlated with air temperature.

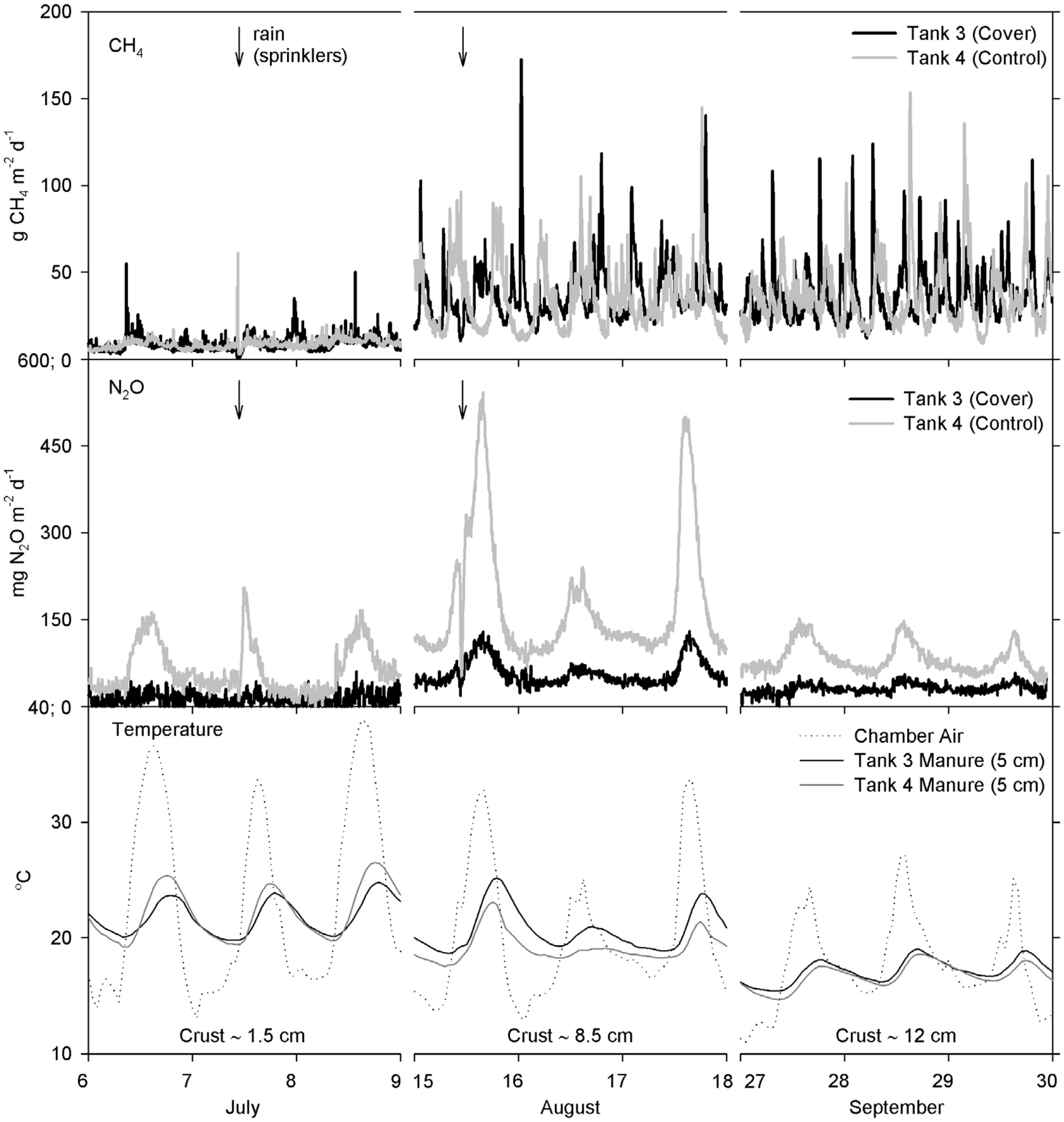

Figure 6. Fluxes of CH_4 and N_2O (4-min data, reported as d^{-1} for ease of comparing with other figures) are shown in the top two panels for tank 3 (covered) and tank 4 (control). The bottom panel shows average air temperature in all chambers (measured 30 cm above manure), manure temperature measured approximately 5 cm below the surface, and the approximate crust thickness (treatment average). These data are shown for three days in July, August, and September. Vertical arrows indicate simulated rain events (via sprinklers in the chambers).

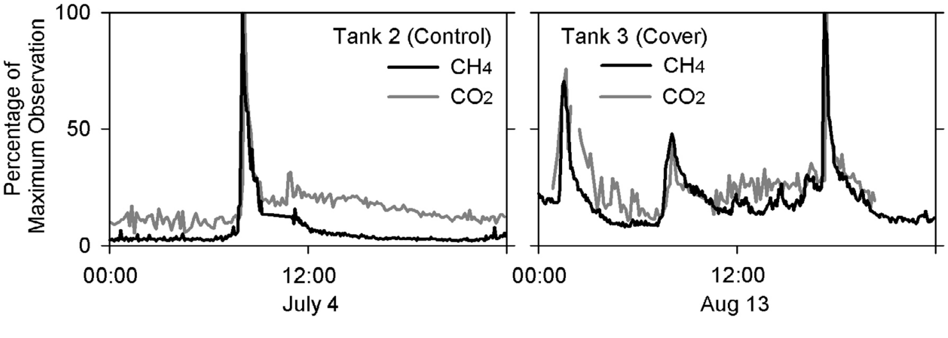

Figure 7. Fluxes of CH_4 (4-min data) and CO_2 (8-min data) showing bubble fluxes. Fluxes are shown from a control tank (left) and a covered tank (right), and each is normalized by dividing by the maximum flux of each gas.

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