The Effect of Biofuel Production on Swine Farm Methane and Ammonia Emissions

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Methane (CH_4) and ammonia (NH_3) are emitted to the atmosphere during anaerobic processing of organic matter, and both gases have detrimental environmental effects. Methane conversion to biofuel production has been suggested to reduce CH_4 emissions from animal manure processing systems. The purpose of this research is to evaluate the change in CH_4 and NH_3 emissions in an animal feeding operation due to biofuel production from the animal manure. Gas emissions were measured from swine farms differing only in their manure-management treatment systems (conventional vs. biofuel). By removing organic matter (i.e., carbon) from the biofuel farms' manure-processing lagoons, average annual CH_4 emissions were decreased by 47% compared with the conventional farm. This represents a net 44% decrease in global warming potential (CO_2 equivalent) by gases emitted from the biofuel farms compared with conventional farms. However, because of the reduction of methanogenesis and its reduced effect on the chemical conversion of ammonium (NH_4^+) to dinitrogen (N_2) gas, NH_3 emissions in the biofuel farms *increased* by 46% over the conventional farms. These studies show that what is considered an environmentally friendly technology had mixed results and that all components of a system should be studied when making changes to existing systems.

MITIGATION OF TRACE GAS EMISSIONS has become an important consideration in the design and management of animal feeding operations (AFOs). Although the trend toward increased size of AFOs may increase unit animal production efficiency, both in terms of energy consumption and emissions due to urine and manure management, the consolidation of large numbers of animals can result in a large, localized source of greenhouse and air-quality gases, including methane (CH_4), ammonia (NH_3), and odors. The conversion of animal manure to biofuels (e.g., methanol) is often promoted as an environmentally beneficial management system, with the potential to reduce greenhouse gas emissions (e.g., Wulf et al., 2006; Ghafoori et al., 2006; Brown et al., 2007). Biofuel production systems are designed to remove organic matter (carbon [C]) from the manure stream to produce hydrocarbon fuels. In traditional manure management, this C would ultimately be emitted to the atmosphere as carbon dioxide (CO_2) and CH_4 during decomposition. In theory, biofuel management leads to a direct reduction in greenhouse C gas emissions by diverting the products of manure decomposition, that is, CH_4 with a global warming potential (GWP) equivalent of approximately 25 times CO_2, to a fuel source that is consumed for its energy with its byproduct C emitted as CO_2 (with a GWP of 1).

During urine and manure management processing, however, complex decomposition interactions occur between C and nitrogen (N) compounds. Much of the N that enters into manure processing lagoons is converted to environmentally benign dinitrogen (N_2) gas (Harper and Sharpe, 1998; Harper et al., 2000, 2001, 2004) by microbial and/or chemical denitrification, which reduces the potential for NH_3 emissions. Harper et al. (2000, Table 2) found interesting correlations between emissions of NH_3, CH_4, nitrous oxide (N_2O), and CO_2 from the urine and manure-processing lagoons, suggesting that manipulation of the management system to reduce emissions of one constituent may affect the emissions of another. This study showed that in urine and manure lagoons with a high rate of methanogenesis, there was a significant

amount of chemical conversion of ammonium (NH_4^+) to N_2; however, when CH_4 production (methanogenesis) decreased, smaller N_2 emission rates coincided with higher rates of N_2O production instead of N_2 production. Methanogenesis is only possible at extremely low O_2 concentrations, and under these conditions N_2 production is thermodynamically more favorable than nitrate (NO_3^-) production.

In a series of six geographically widely spaced swine operations in North Carolina, Harper and De Visscher (unpublished data) showed a relationship between CH_4 production and conversion of NH_4^+ to N_2. They found that as CH_4 emissions increased, N_2 emissions increased by a 4:1 CH_4/N_2 ratio (i.e., for a four CH_4 emissions unit increase, N_2 emissions increased by one unit). Amon et al. (2005) and Clemens et al. (2006) found higher NH_3 emissions from biogas-effluent manure slurry than from untreated manure, which they explained by higher NH_4–N content and pH of the effluent. These studies are consistent with the work of Strik et al. (2006), who found an increase in NH_4–N content of manure with time spent in a biogas reactor, and that of Loria et al. (2007), which suggested biogas production increases the NH_4–N content of manure slurry. These latter two studies did not evaluate N_2 production. Because of these relatively new insights, there was concern that removal of the organic matter (and decreasing lagoon methanogenesis) from lagoon urine and manure processing systems may reduce the NH_4^+-to-N_2 conversion and increase the amount of NH_3 emissions from the farms associated with biofuel production. Could the implementation of a biofuel system as a component of the urine and manure processing strategy for AFOs also have negative consequences? Accordingly, the purpose of this study is to evaluate CH_4 and NH_3 gas emissions from farms that are nominally identical, except in regard to manure management, and to determine if the reduction of CH_4 by a biofuel production system affects the total nutrient cycling in the animal production system.

Materials and Methods

Biofuel Facility

The focus of this study was a biofuels facility constructed to capture biogas from digested pig manure and for conversion of the biogas into biomethanol and biodiesel, a process whereby C in the animal manure is converted to a usable and less-polluting fuel for internal combustion engines. As a result, the manure C, which is aerobically or anaerobically decomposed in normal manure-management systems and generally lost to the atmosphere as CO_2 and CH_4, is converted to a usable product.

This biofuel facility is part of a multifarm swine complex in the semiarid Central Great Basin of the United States. Farms are spread over several kilometers along a broad valley (elevation 1500 m), separated from each other by several hundred meters. Liquid manure is collected from 12 12,000-animal finishing farms (144,000 animal total, Fig. 1A) and conveyed to a central treatment plant. The manure is first concentrated, by means of gravity thickeners, and then conveyed to two covered earthen digester tanks, where it is heated to 35°C for undergoing bacterial processing ("digestion"), resulting in the biogas production. The digester effluent, digester sludge, and gravity thickener supernatant are conveyed back to the farm manure
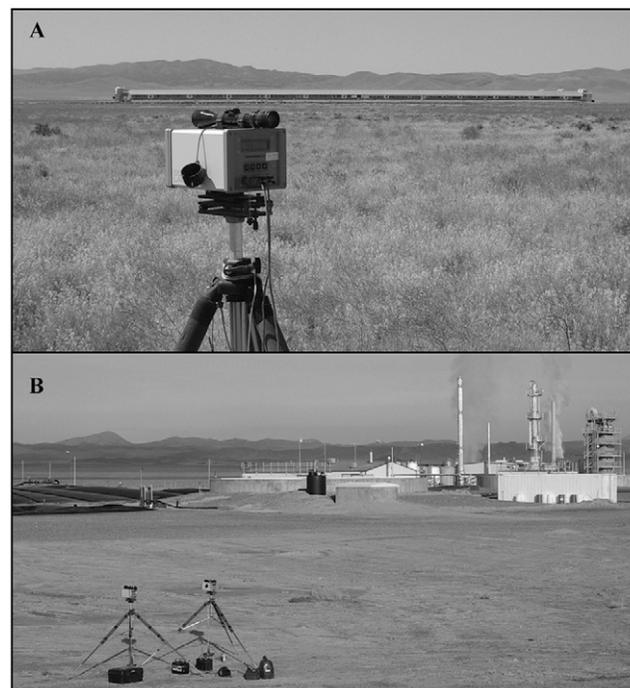
Fig. 1. (A) Open path laser unit located downwind of a farm. (B) Two laser units located at the biofuel facility (B). The laser reflectors are not visible in these pictures.

lagoons where the lagoons operate at the same hydraulic loading rate as they would without the biofuels plant, but with a much-decreased solids loading rate due to the extraction of manure C for methanol production. The biogas is collected and conveyed to a biomethanol conversion plant on the site (Fig. 1B). Biomethanol produced at the facility is trucked from the site as a liquid for conversion to biodiesel at a location remote from the swine production facility.

The Swine Farms

Methane and NH_3 gas emissions were measured from three farms at the production complex. These farms were nominally identical 12,000 animal "finisher" farms. Each consisted of three closely spaced and joined barns with adjacent primary and secondary open-air manure lagoons. The "conventional" farm (Control) uses traditional manure treatment: liquid manure is transferred from the barns into lagoons, where evaporation and decomposition maintain long-term manure equilibrium at the farm such that no manure removal (other than that occurring naturally, i.e., unimpeded venting of C and N compounds to the atmosphere) is performed. Water temperature data were collected using HOBO Water Temp Pro data loggers (Onset Computer Corporation, Bourne, MA). Lagoon pH was measured with pH probes on-site and on samples of the effluent collected twice a month, and frozen, for subsequent pH and NH_3/NH_4^+ analysis. The two "biofuel" study farms (BF1 and BF2) were converted from the traditional manure-management system to the biofuel system as described above.

Emissions measurements were also made at the biofuel plant site. Because the gravity thickeners expose manure to the air, there will be gas emissions from these locations. The rest of the biofuel site is enclosed (i.e., sealed off from the atmosphere), the intent being to eliminate all other emission points.

Emission measurements were conducted from 30 January to 23 February and 27 June to 14 July 2005.

Inverse-Dispersion Technique

The inverse-dispersion technique uses an atmospheric dispersion model to infer the emission rate that best explains the observed downwind gas concentration under existing meteorological conditions, namely, wind direction and speed, temperature stratification, and (consequent) degree of turbulent mixing (Flesch et al., 2004). Consider an area source emitting tracer gas at a uniform but unknown rate Q (g m^{-2} s^{-1}) and assume an average tracer concentration C (g m^{-3}) measured in the plume of dispersing gas. The dispersion model predicts the ratio of the concentration at the measurement location (or more specifically, the increase in atmospheric concentration above background levels attributable to the source) to the emission rate, $(C/Q)_{sim}$. The emission rate may be computed as

$$Q = \frac{(C - C_b)}{(C/Q)_{sim}} \qquad [1]$$

where C_b is the background tracer concentration. This technique is well suited for "ideal surface layer problems" (see Flesch et al., 2004), that is, horizontally uniform terrain where the wind and turbulence can be described by well-known functions of height. In these cases, the wind statistics needed to predict $(C/Q)_{sim}$ can be inferred from the friction velocity, u_*, the Monin–Obukhov stability length, L, the surface roughness length, z_0, and the average wind direction, β. These primary meteorological properties can be measured with a three-dimensional sonic anemometer. The averaging interval for this type of measurement is ideally in the range of 10 to 60 min.

The terrain around the farm complex is ideal for application of idealized dispersion models: flat and uniform with a sparse coverage of low sagebrush and grass extending in all directions (Fig. 1A). But the farms can complicate an idealized dispersion calculation because farm structures can create wind complexity and the exact spatial distribution of emissions from the barns and lagoons are unknown. (For a compound emission source, e.g., a farm with barns and urine and manure lagoons, one must make assumptions about the relative distribution of the component emissions.) However, Flesch et al. (2005b), McGinn et al. (2006), and Gao et al. (2010) suggested that if the concentration and wind measurements are made far enough downwind of the farm, the emission calculations will be insensitive to these complications. With regard to estimating how far downwind to measure concentrations, Flesch et al. (2005b) suggested that the crucial distance scales are the height of the largest wind obstacle, h, and the maximum distance between the source components, x_s (e.g., the distance between the centers of the barn and lagoon). They recommended concentration observations be made more than $10h$ downwind of a farm and more than $2x_s$. This technique has been used in a number of animal feeding operations (e.g., Flesch et al., 2005b; McGinn et al., 2006; Harper et al., 2009, 2010).

Concentration and Wind Observations

Methane and NH$_3$ concentrations were measured with open-path lasers (GasFinders, Boreal Laser Inc., Edmonton, Canada). The path-average gas concentration was measured between the laser and a distant retroreflector and processed to give 15-min averages (C). Lasers were calibrated on-site using calibration tubes flooded with gas standards. Three NH$_3$ lasers and a single CH$_4$ laser were used. Background concentrations of CH$_4$ and NH$_3$ were taken as C_b = 1.75 and 0 µL L^{-1}, respectively (corroborated when the wind brought air with no farm-gas sources upwind over the laser paths).

The lasers were placed so their paths would be downwind of the emission sites for the prevailing southwest winds. Figure 2 shows the configuration of the sites and laser locations for summer measurements. Configurations for winter measurements were slightly different due to a shift in predominant wind direction between seasons. At the three farms the lasers paths were approximately 200 m downwind of the barns and/or lagoons at the nearest point (Fig. 1A). This was more than $25h$ from the barns and, depending on the farm, was $1.3x_s$ to $6x_s$ downwind (where x_s is the distance separating barns from lagoons). Ammonia emissions were measured from the three farms concurrently using the three NH$_3$ lasers. With only one CH$_4$ laser during summer, we first measured CH$_4$ emissions from the control farm and then moved to one of the treatment farms. During winter, lasers were available to measure downwind concentrations for both farms simultaneously. All lasers were used at the biofuel site (Fig. 1B). The NH$_3$ lasers were positioned so their paths made up three sides of a square
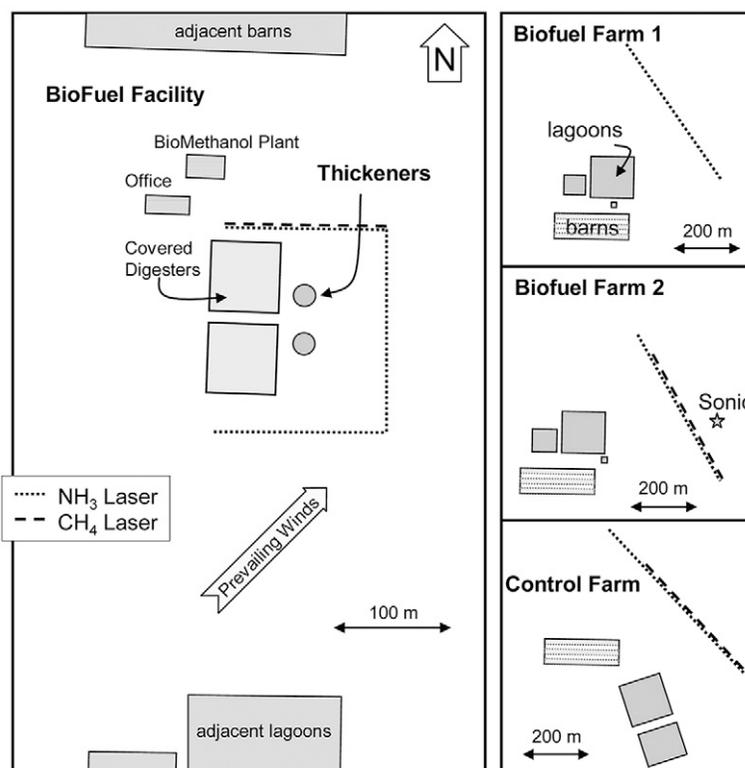
Fig. 2. Summer measurement locations at the four emission sites.

around the two open-air thickeners. In theory, this allowed us to make emission measurements for a wide range of wind directions. The single CH_4 laser during summer was placed northeast of the thickeners. At the closest location, the laser paths were 60 m from the thickeners.

A three-dimension sonic anemometer (CSAT-3, Campbell Scientific, Logan, UT) provided the wind information for the dispersion calculations. The anemometer was placed at height of 2 m. Wind velocity and temperature signals were sampled at a frequency of 16 Hz. The necessary wind statistics (u_*, L, z_0, β) were calculated for 15-min intervals to match the C observations, as described in Flesch et al. (2004). The sonic observations also provided velocity standard deviations (in each of three-dimensions) that were used in the dispersion model.

The sonic anemometer was placed to measure the ambient winds (unaffected by farm structures) at a location central to the study sites: approximately 400 m from the first treatment farm, 1.5 km from the second treatment farm, and 8 km from the control farm. Given the homogeneous character of the landscape (Fig. 1A, flat terrain, low shrub desert flora, no obstacles), we assumed the winds measured at the sonic location were representative of the conditions at the four sites. It should be noted that because of the distance between the farms (and the anemometer), the measured wind statistics for any one 15-min analysis period may not be applicable (concurrently) at all the sites. This adds uncertainty to our individual 15-min emission values. However, over many observations we assume the wind regime between the sites is nearly identical, and our average emission calculations are more accurate.

Backward Lagrangian stochastic Application Details

Following Flesch et al. (2005a), we used a backward Lagrangian stochastic (bLS) dispersion model to calculate $(C/Q)_{sim}$ (WindTrax, Thunder Beach Scientific, Nanaimo, Canada). Thousands of trajectories are calculated upwind of the laser path for each 15-min observation period (e.g., 250,000). The important information is contained in the trajectory intersections with ground ("touchdowns"), and we compute

$$\left(C/Q\right)_{sim} = \frac{1}{N}\sum\left|\frac{2}{w_0}\right| \qquad [2]$$

where N is the number of computed trajectories, w_0 is the vertical velocity at touchdown, and the summation covers only touchdowns within the source. (The units of Q are g m^{-2} s^{-1} in this equation. Hereafter, we multiply the areal emission rate by the source area and report Q as an area-integrated emission rate with units of kg h^{-1}.) The touchdowns map the concentration "footprint", that is, the ground area where emissions influence concentration.

Each farm is represented as three surface area sources: the two lagoons and the area outlined by the barns (Fig. 2). Each area is assumed to have the same areal emission rate so that touchdowns in any of these areas are counted equally in Eq. [2]. Equating the areal emissions rates in this manner represents an approximation. The two lagoons may have somewhat different emission rates, while the barn is not, in fact, an area source (emissions occur from vents on the walls) and, even

if treated as such, may have an effective area source strength differing from that of the lagoons. However, following the arguments of Flesch et al. (2005a), we assume that with C measured sufficiently far downwind, the inferred mean emission rate from the complex is insensitive to the correctness of the approximation. Emissions at the biofuel site were assumed to originate exclusively from the two thickeners, which, having identical manure input rates, were assumed to have equal areal emission rates.

The bLS technique to estimate emission rate depends on a good description of atmospheric transport, which is known to be difficult in extreme conditions. Following Flesch et al. (2005a), we eliminated measurements during periods (i) where $u_* \leq 0.15$ m s^{-1} (low wind conditions), (ii) where $|L| \leq 10$ m (strongly stable/unstable atmosphere), and (iii) where $z_0 \geq 1$ m (associated with uncertainty regarding the proper wind profile). For some wind directions, the farm plumes only "glanced" the path of the lasers, giving more uncertain Q estimates. To avoid these problems we removed periods (iv) where the laser touchdowns do not cover some portion of all the source areas (e.g., at least 50% of the barn and lagoons, or both thickeners).

The bLS technique for calculating emissions has been tested in a number of tracer release studies, conducted in a variety of terrain settings and source configurations. Some of these verification studies are listed in the Appendix. One concludes that with careful use of the technique (e.g., proper equipment siting, data filtering, data averaging), the expected accuracy of the emission calculations should be approximately ±10%.

A problem in this study was a lack of nighttime data suitable for emission calculations, either because of laser alignment problems, unsuitable wind directions, or wind statistics that did not meet the analysis criteria (e.g., low winds). The summer control farm, in particular, had little data that conformed to the selection criteria during the periods from 2300 to 0800 h. To obtain sufficient data to create daily emission curves we relaxed the nighttime data criteria, for the summer control farm only, to include data with an atmospheric stability of $|L|$ from 5 to 10 m for nighttime periods (see Fig. 3C, open circles, for the relaxed data criteria periods). As explained in Flesch et al. (2005b), we thus anticipate greater errors than ±10% (Appendix) for these periods.

Results and Discussion

Biofuel Production Site

Manure slurry from the swine farms is transported and injected into sealed digesters at the biofuel production site. Before injection into the digesters, there was a potential for CH_4 and NH_3 emissions from the gravity thickeners (Fig. 1B and 2), which had an open surface.

Methane emissions should be small at the biofuel site. However, we found some periodic evidence of fugitive CH_4 emissions around the digester pits (either from leaking pipes, digester covers, or leakage from the soil around the pits). We omitted these emission periods on the principle that they are not representative of a well-designed and maintained system. Thus, in our study, we assume CH_4 emissions from the biofuel site are negligible (compared with the farm sites) and can be ignored.
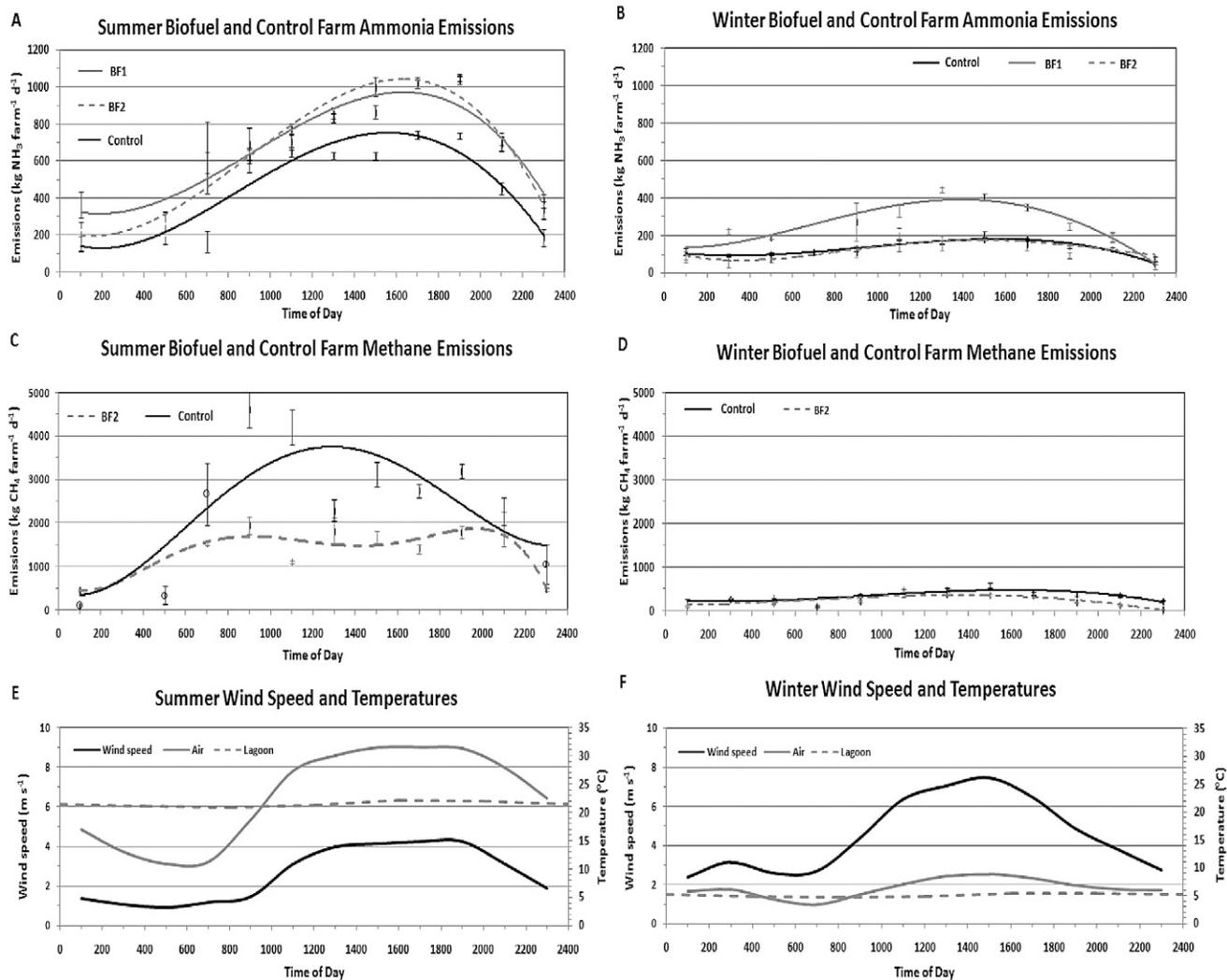
Fig. 3. (A, B) Average summer and winter NH$_3$ emissions, plotted versus time of day, for biofuel (BF1, BF2) and control farms in the Central Great Basin (error bars are one standard error for each 2-h period). **(C, D)** Average summer and winter CH$_4$ emissions for biofuel and control farms. **(E, F)** Average summer and winter wind speed plus air temperature (height = 2 m) and lagoon temperature (depth = −0.05 m) at the farm site.

On-site NH$_3$ measurements showed average summertime emissions were 2.6 kg NH$_3$ h^{-1}, equivalent to 0.4 kg NH$_3$ h^{-1} for each of the six farms providing organic matter to the site. This was only 2% of the farm emissions during this period. Thus, like methane emissions, these additional NH$_3$ emission rates were ignored when calculating representative emissions from the biofuel system.

Farm Sites

Figure 3 gives the average diurnal emissions relationship for NH$_3$ and CH$_4$ during summer and winter for two biofuel farms and one control farm, along with average wind speed plus air and lagoon temperatures. These emissions were calculated by grouping the available data (15-min observations) according to the time of day, and averaging the data in 2-h blocks throughout the 24-h day. The objective was to create a properly weighted daily average emission rate (i.e., our 15-min observations were not evenly distributed over the day). The curves in Fig. 3 were obtained from these 2-h block averages, with the error bars showing the standard error of the 15-min observations within each block.

The annual emissions were estimated by averaging the daily emission rates from the summer and winter periods. Previous studies (Harper et al., 2004, 2009; Zhu et al., 2010) showed that an annual estimate obtained from the winter and summer averages adequately estimates the annual average, and the addition of a transitional season (three total seasons) did not significantly change the average annual emissions.

Methane and Ammonia Emissions

Comparison between Biofuel Farms

We anticipated that the two biofuel farms, which are nominally identical in management, would have similar emission rates. This was observed to be the case for NH$_3$ during summer (Fig. 3A) as the two biofuel treatment farms showed no significant difference in NH$_3$ emissions ($p > 0.05$). However, there was a small difference during winter (Fig. 3B, $p < 0.01$). The difference between the two biofuel farms' emissions during winter was perhaps due to differences in lagoon solution temperature (average wintertime temperatures for the BF1 and BF2 farms were 6.4 and 5.4°C, respectively.). We believe the lagoon temperature for BF1 farm was higher since it is about 1 km closer

to the biofuel production site (effluent thermal absorbtion by the soil during return transmission). Ammonia emissions showed diurnal variability during both measurement seasons, but, as anticipated, the winter emissions were a fraction of the summer emissions. Comparison of annual emissions showed no difference between the biofuel farms ($p > 0.05$).

Comparison between Biofuel and Control Farms

Comparisons between the average of the two biofuel farms and the control farm NH_3 emissions (Table 1) showed significantly higher emissions from the biofuel farms during summer (188 kg NH_3 farm^{-1} d^{-1}, +38%, $p < 0.02$) and winter (62 kg NH_3 farm^{-1} d^{-1},+48%, $p < 0.01$). Although there was no significant difference (Table 1) in annual lagoon NH_4^+ concentrations between the biofuel and control farms ($p > 0.05$), the annual lagoon pH was significantly higher in the biofuel farms ($p < 0.05$). This higher pH allows for a greater dissociation of NH_4^+ to NH_3 and therefore the potential for larger NH_3 emissions (Harper, 2005; Koelliker and Kissel, 1988). Comparison of CH_4 emissions between the biofuel and control farms showed significantly smaller CH_4 emissions from the biofuel farm during both summer (1552 kg CH_4 farm^{-1} d^{-1}, –49%, $p < 0.05$) and winter (108 kg CH_4 farm^{-1} d^{-1}, –32%, $p < 0.05$). Both winter NH_3 and CH_4 emissions were only a fraction of summer emissions. When we take the average of winter and summer emission rates to estimate annual emissions, we find that the biofuel farm had annual emissions of CH_4 that were 47% lower but NH_3 emissions that were 46% higher, with the differences being mostly due to the differences in summer emissions. This study showed that on an annual basis, the biofuel farms had CH_4 and NH_3 emissions differences (Table 1) greater than the 10% level of uncertainty we believe exists in the bLS technique (Appendix).

We found the expected result that CH_4 and NH_3 emissions were correlated with wind speed and air temperature (Fig. 4A–D). Studies (Harper et al., 2000, 2004) have shown that increasing wind speeds lead to increasing emissions, on a short-term or diurnal basis, from ponds and naturally ventilated sources. For example, at the control farm (Fig. 4A and 4C), emissions were weakly correlated with wind speed within seasons (r^2 ranging from 0.01 to 0.46). When comparing winter and summer emissions with air temperature, however, we see that long-term emissions are better related to air temperature ($r^2 \geq 0.80$ for both gases). Over long periods

of time, the air temperature will be a surrogate of the solution temperature (Fig. 3E and 3F) in both the lagoons and barns. Studies have shown for NH_3 (Harper et al., 2000, 2004) that solution temperature affects the dissociation between NH_4^+ and NH_3 in solution and the diffusion of NH_3 in solution, both of which will alter the emission rate. (NH_3 is a diffusive gas influenced by the physical and chemical factors of solution concentration [NH_4^+], solution hydrogen ion concentration [pH], turbulence [wind speed], and solution temperature. For a discussion of the relationship of physical and chemical factors to NH_3 emissions, see De Visscher et al., 2002; Harper, 2005.) Temperature has a similar effect on emissions from the barns because of the amount of barn air exchanged to maintain comfort of the animals. The effect of temperature on CH_4 emissions has a greater effect on biological activity than on the physical chemistry of gas in solution and transport since CH_4 has five orders of magnitude less solubility in water than NH_3.

GWP Comparison between the Biofuel and Control Farms

There was a 47% decrease in average annual CH_4 emissions at the biofuel farm compared with the control farm; however, average annual NH_3 emissions increased by 46% compared with control farm emissions. These changes in emissions can be used to look at the effect of biofuel production on the GWP of a farm system.

The annual GWP decrease, when considering that methanol (from methane, GWP of 25, 100-yr time horizon) is consumed as biodiesel and emitted as CO_2 (GWP of 1), was 45% compared with control farm emissions. However, NH_3 emissions were increased on the biofuel farms, which is a precursor of indirect nitrous oxide (N_2O, GWP = 298, 100-yr time horizon) emissions. Because a fraction of the NH_3 will, on redeposition on the soil environment elsewhere, be transformed into N_2O (assuming an emissions factor of 1% of deposited NH_3/NH_4^+, Intergovernmental Panel on Climate Change default), an annual NH_3 increase of 144 kg NH_3 d^{-1} would result in a combined net reduction in GWP of 44% for this biofuel management system. By removing the C, and reducing methanogenesis and global-change gas emissions (Harper et al., 2000, 2004; Harper and De Visscher, unpublished data) from the manure-processing system of swine production, the

Table 1. NH_3 and CH_4 emissions from biofuel production and control farms.

Farm type	Season	Lagoon surface concentration	Lagoon surface pH	NH_3 emissions	CH_4 emissions
		mg NH_4^+-N L^{-1}	[H$^+$] = 1 × 10^{-pH}	kg NH_3 farm^{-1} d^{-1}	kg CH_4 farm^{-1} d^{-1}
Biofuel farms	Summer	1818	8.21	692 ± 59†	1651
Control farm	Summer	1753	8.13	504	3203
% difference	Summer	–	–	+38%	–49%
Biofuel farms	Winter	1827	8.34	190 ± 24†	232
Control farm	Winter	1855	8.16	128	340
% difference	Winter	–	–	+48%	–32%
Biofuel farms	Annual	1823	8.27	460	942
Control farm	Annual	1804	8.14	316	1772
% difference	Annual	–	–	+46%	–47%

† Standard error.

processing of manure NH_3/NH_4^+ to N_2 gas was detrimentally causing an *increase* in NH_3 emissions (air-quality gas) by 46%. This increase in NH_3 emissions creates a potential for additional NH_4^+ particulates and haze production compared with conventional production practices. What had been intended to function as an environmentally friendly technology had (according to our observations) mixed results.

Conclusions

We have seen encouragement to develop "manure-to-fuel" technologies for farming systems, including removal of organic C from animal manure processing systems (e.g., lagoons) for fuel production. As we document in this study, manure-to-fuel production has the benefit of reducing greenhouse gas emissions from animal production systems. It appears, however, that it can also have unforeseen consequences. With the manure removed from the processing system (reduction of methanogenesis), NH_3 emissions were increased. Other studies also have shown that as methanogenesis decreases, the rate of conversion of NH_4^+ to (harmless) N_2 undergoes a parallel decrease. These studies show that we should be aware of the potential for countervailing interactions (in terms of release rates to the atmosphere of diverse species) when modifying animal manure management systems.

Fig. 4. Emission rates of CH_4 and NH_3 plotted versus ambient wind speed and air temperature (height = 2 m) from the control farm.

Appendix

Table A1 summarizes several tracer studies on the accuracy of the bLS analysis technique for calculating emissions (Q). Accuracy is indicated by the gas recovery, which is the percentage ratio of the bLS calculated emissions to actual emissions (i.e., $Q_{bLS}/Q_{release} \times 100$). These studies had an average recovery of 98% with a standard deviation of 5%. We conclude that for a good site with appropriate instrument placement and data filtering (as discussed in detail in these studies), one can expect a nominal bLS accuracy of $100 \pm 10\%$ (± two standard deviations—a span that includes 95% of a Gaussian distributed population). This would be the accuracy of an average of multiple measurement periods. For a single observation (e.g., one 15-min value), these results suggest a higher ±42% uncertainty (twice the average of the within-study standard deviation). This period-to-period uncertainty is due to uncertainties in the bLS model, uncertainty in the idealized representation of the wind,

noisy C observations, and so on, uncertainties that are reduced by appropriate averaging.

Table A2 compares nine studies where trace-gas emissions were determined using the bLS analysis procedure and alternative techniques (true emissions were unknown). The alternative techniques include the integrated horizontal flux and flux-gradient micrometeorological techniques and a SF_6 ruminant tracer technique (SF_6).

Acknowledgments

Table A1. Tracer studies using the backward Lagrangian Stochastic (bLS) analysis procedure. These studies evaluated emissions in both ideal wind conditions (homogeneous terrain) and in winds disturbed by obstacles, from either single or multiple sources, and with either point or line-average concentration (C) measurements. Each study was based on many measurement periods (typically 15 min each), and the average gas recovery and standard deviation of the recovery are presented. Well-described data-filtering criteria were used in each study to reduce bLS errors. For studies with wind obstacles, the downwind distance (M) between the C measurement and the upwind obstacle is given in terms of the obstacle height (h).

Gas	Recovery ± SD	Site characteristics	Reference
	%		
CH_4	102 ± 22	Grass, no obstructions, line-average C	Flesch et al. (2004)
CH_4	98 ± 20	Grass, obstructions ($M > 5h$), line-average C	Flesch et al. (2005a)
CH_4	93 ± 14	Grass, no obstructions, multi-source,† line-average C	Gao et al. (2008)
CH_4	98 ± 11	Grass, obstructions ($M > 10h$), multisource,‡ line-average C	Gao et al. (2008)
CH_4	106 ± 16	Grass, no obstructions, line-average C	McBain and Desjardins (2005)
CH_4	99 ± 20	Grass, obstructions ($M > 5h$), line-average C	McBain and Desjardins (2005)
SF_6	100 ± 29	Whole-farm, dairy ($M > 9h$), line-average C	McGinn et al. (2006)
CH_4	86 ± 17	Whole-farm, dairy ($M > 9h$), line-average C	McGinn et al. (2006)
CH_4	103 ± 16	Grass, no obstructions, line-average C	Gao et al. (2009a)
CH_4	99 ± 30	Grass, no obstructions,§ line-average C	Gao et al. (2009b)
CH_4/CO_2	99 ± 29	Wheat stubble, no obstructions, line-average C	Loh et al. (2009)
CH_4	98 ± 18	Barn source ($M \geq 10h$), line-average C	Gao et al. (2010)
Average	98 ± 5		
Average SD (within-studies)	21 ± 6		

† Results for trial C1 are given here—simplest of the multisource layouts studied. Recovery is calculated based on the summation of all emission sources.

‡ Results for trial C2 are given here—simplest of the layouts studied, but with obstacles placed around the sources. Another trial in this study used higher obstacles ($M \sim 5h$), and the recovery was reduced to 0.75. Recovery is calculated based on the summation of all emission sources.

§ Reported recovery in this study was 109 ± 45. The high standard deviation (45%) was due to the experimental design (4 laser lines, solving for C_b [background tracer concentration] and Q [uniform gas emission rate]) which led to ill-conditioned solutions. We report the recovery calculated when periods with very large uncertainties are removed (as discussed in Gao et al., 2009b).

Table A2. Studies comparing the backward Lagrangian stochastic (bLS) analysis procedure with alternative techniques.

Gas	Differences	Site characteristics	Reference
Metolachlor	bLS 6% higher than IHF†	Bare soil, no obstructions, point C‡	Flesch et al. (2002)
CH_4	bLS ≈ 10% higher than IHF	Cows in pasture (enteric emissions), line-average C	Laubach and Kelliher (2005a)
CH_4	bLS 14% higher than IHF	Cows in pasture (enteric emissions), point C	Laubach and Kelliher (2005b)
CH_4	bLS 3 and 18% higher than IHF (2 sensor configurations, all fetches)	Cows in paddock (enteric emissions), point and line-average C	Laubach et al. (2008)
Chemical	bLS 2% higher than IHF	Short grass, point C	Flesch et al. (1995)
NH_3	bLS 22 to 36% higher than FG§	Commercial beef feedyard, point C	Todd et al. (2007)
NH_3	two bLS configurations were +10% and −5% of IHF calculation	Field emissions of manure slurry, point C	Sanz et al. (2009)
SF_6	bLS 7% less than SF_6	Cows in small pens (enteric emissions), line-average C	McGinn et al. (2009)
NH_3	For all measurements bLS averaged 19% higher than IHF. For cumulative emissions, bLS was within 5% of IHF.	Urea volatilization from wheat crop, 25-m radius circular plot, point C.	Turner et al. (2010)

† IHF, integrated horizontal flux.

‡ C, concentration.

§ FG, flux-gradient.

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