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Characterizing ammonia emissions from a commercial mechanically ventilated swine finishing facility and an anaerobic waste lagoon in North Carolina

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ABSTRACT

Emissions of atmospheric ammonia–nitrogen [NH_3–N, where NH_3–N = (14/17) NH_3] were measured from a commercial anaerobic swine waste treatment lagoon and from an on–site finishing swine confinement house at the same location. Continuous measurements were made at each potential NH_3–N source for ~1 week during four different seasons. Results presented here represent measurements made for the second year of a multi–year experiment. Barn emissions were estimated to be 2 604±660 g NH_3–N day^{-1}, 1 761±1 087 g NH_3–N day^{-1}, 1 657±1 506 g NH_3–N day^{-1}, and 2 659±1 194 g NH_3–N g day^{-1} in summer, fall, winter, and spring respectively. NH_3–N barn emission factors were calculated to be 1.32±0.32 kg NH_3–N $animal^{-1}$ yr^{-1}, 0.78±0.49 kg NH^3–N $animal^{-1}$ yr^{-1}, 1.55±1.40 kg NH_3–N $animal^{-1}$ yr^{-1}, and 1.35±0.61 kg NH^3–N $animal^{-1}$ yr^{-1} in summer, fall, winter, and spring respectively. Average NH_3–N flux from lagoon was greatest in the summer, >3 943 μg m^{-2} min^{-1}, and lowest in the winter, 981±210 μg m^{-2} min^{-1}. Fall and spring average NH_3–N flux values were >1 383 μg m^{-2} min^{-1} and 1 641±362 μg m^{-2} min^{-1}, respectively.

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1. Introduction

Emissions of trace gases, particularly ammonia (NH_3), from concentrated animal feeding operations (CAFOs) have become a major problem in recent years in North Carolina as changes in crop and livestock production methods are in turn changing emissions of trace gases into the atmosphere (Aneja et al., 2006; US EPA, 2011). In North Carolina, the swine industry has grown significantly over the past two decades and is a major animal agricultural industry in the state, with an inventory of ~10 million animals (daily standing population). Currently, there are more than 1 400 operations with more than 1 000 head, which accounts for over 99% of the state inventory (USDA, 2008), with the majority of the pig farms in North Carolina clustered in the southeastern region of the state. Public concerns about potential environmental, ecological, and health effects of air emissions from CAFOs have increased along with the growth and consolidation of this industry (Aneja et al., 2006).

Ammonia is a by–product of microbial decomposition of the organic nitrogen compounds in manure. It also occurs as absorbed nitrogen in the form of organically bound nitrogen, and as urea in urine (Beline et al., 1998; US EPA, 2001). It is the primary gaseous base in the atmosphere influencing the acidity of precipitation, cloud water, and aerosol species. Ammonia released from near–surface sources (i.e., anaerobic waste lagoons) into the atmosphere generally has a lifetime of ~1–5 days (Warneck, 2000) and may deposit near the source through dry or wet deposition. However, ammonia readily reacts with a variety of acidic atmospheric species, forming ammonium aerosols such as ammonium sulfate, –nitrate, and –chloride, which can have longer atmospheric residence lifetimes (~1–15 days) owing to a decrease in dry deposition velocity (Aneja et al., 1998). Therefore, ammonium aerosols may be transported and deposited further downwind from the source. In eastern North Carolina, an environmental concern that has been associated with ammonium aerosols is their deposition into sensitive aquatic ecosystems, where excessive nitrogen loading may lead to soil acidification, enhanced eutrophication, and the growth of harmful algal blooms, which may cascade to upset ecosystem nutrient balances (Paerl, 1997). In addition, excess levels of ammonia in the air allows for fine particulate matter formation which can cause adverse effects on human health including respiratory problems and lung damage (WHO, 2003; Aneja et al., 2008; Aneja et al., 2009).

Concentrated Animal Feeding Operations in relatively small areas can be a significant source of gaseous NH_3 emissions. Studies on gas emissions from swine housing and manure management systems indicate that ammonia emissions are significant, especially at densely populated animal sites (Aneja et al., 2000; Aneja et al., 2001; Heber et al., 2002; Ni et al., 2002; Lim et al., 2003; Harper et al., 2004). Arogo et al. (2003) reviewed measurement techniques

and studies undertaken to estimate NH_3 emissions from both swine lagoons and animal confinement houses. Walker et al. (2000) estimated that hog operations are responsible for almost 50% of aggregate NH_3 emissions in North Carolina.

There are various factors that may affect the emissions of gases into the atmosphere from CAFOs. These include different types of animal production, farming activities, and climate. Therefore, emissions estimates for one type of CAFO may not translate readily to others. It is therefore important for comprehensive emissions measurements to be made at different types of CAFOs in order to develop accurate emission factors.

An intensive two–year field study was undertaken to investigate and evaluate the variability of ammonia emissions with respect to diurnal and seasonal variations for four seasons, as well as the influence of meteorological and physicochemical factors during each season. Measurements were made at two primary point sources, an on–site waste anaerobic swine lagoon and an animal housing structure, at a farm in eastern North Carolina. Data were collected continuously using in situ measurement techniques for a 1–week period during each season (i.e., fall, winter, spring, and summer) (Aneja et al., 2000). Results for the first year of measurements are presented in Blunden and Aneja (2008) and Blunden et al. (2008). This work presents NH_3 emission estimates for data collected during the second year of the study, thus contributing to a robust data set that provides a reliable estimate of emissions from these commercial sites and provides uncertainties associated with NH_3 emissions. This also allows for accurately quantifying NH_3 emissions and improving NH_3 emission inventories for scientific and regulatory purposes. Accurate emission estimates will allow for improved decision making with respect to improving regional air and water quality by better quantitatively understanding ammonia emissions from these environments.

2. Methods and Materials

2.1. Physiographic location and farm description

The sampling site is located in Jones County, North Carolina, in the southeastern region of the state, where the majority of hog farming operations are located. A schematic of the facility is shown in Figure 1. The site is a ~7 000–head operational commercial swine finishing farm. The on–site waste storage treatment lagoon is ~17 150 m^2, at the normal liquid level. The farm utilizes a conventional "lagoon and spray" technology as its primary means of handling the waste, which is the most widely used method in North Carolina. Waste is flushed directly from the hog barns into the anaerobic lagoon once per week and a portion of the lagoon waste is recycled to recharge the pit. During a flush, the majority of all the liquid was flushed from the building, but it was not power washed. The recharge liquid was all treated lagoon effluent. Waste from the lagoon is also periodically removed and used as spray over seasonal on–site agricultural crops as a source of nutrients. Although the hog waste is flushed into the lagoon daily, the amount entering the system over a one week period is considered insignificant compared to the content of the entire lagoon and therefore the additional waste is not expected to affect measurements. Hog waste was not flushed into the system near the experimental area. In addition, no waste was removed from the system and no mechanical mixing or stirring of the lagoon occurred during the experimental periods.

The farm had eight mechanically–ventilated confinement houses, which were aligned North–South and oriented East–West. Generally, the hogs are rotated out of each house approximately every 18 weeks, weighing ~22–24 kg upon arrival and gaining an average of 5 kg per week (Williams, 2005). The weight gain is assumed to be linear and the weekly animal mortality rate was documented and taken into account for total weight estimations. Each house is cleaned and sanitized between rotations. There are 800–900 animals housed in each barn. A full description of the hog numbers, weights, and number of weeks in rotation for each season during the sampling is provided in Table 1.

Table 1. *Sampling dates and production data for housing emissions in each season*

Season	Sample Dates	Number of Hogs	Number of Weeks in Rotation	Average Weight [a,b]	Total Weight [a,c]
Summer	June 21-27, 2007	885	8	48.7	43 049
Fall	Nov. 2-10, 2007	995	5	34.6	34 428
Winter	Feb. 9-28, 2008	476	21	116.6	55 513
Spring	Apr. 21-26, 2008	875	9	50.6	44 262

[a] *Measured in units of kg*
[b] *Weighted average for pigs brought into the barns on different dates.*
[c] *In general, it is the product of number of hogs and average weight corrected for rotation*

2.2. Barn experimental procedure

One barn, located furthest north on the property, was selected for emission measurements. Five AAA Associates Inc. Maxi–Brute™ fans with plastic shutters (Niles, MI), two 91 cm diameter direct–driven and three 122 cm diameter belt–driven, were located at the west end of the building (see Figure 1). The fans were staged to operate as temperature increased inside the building.

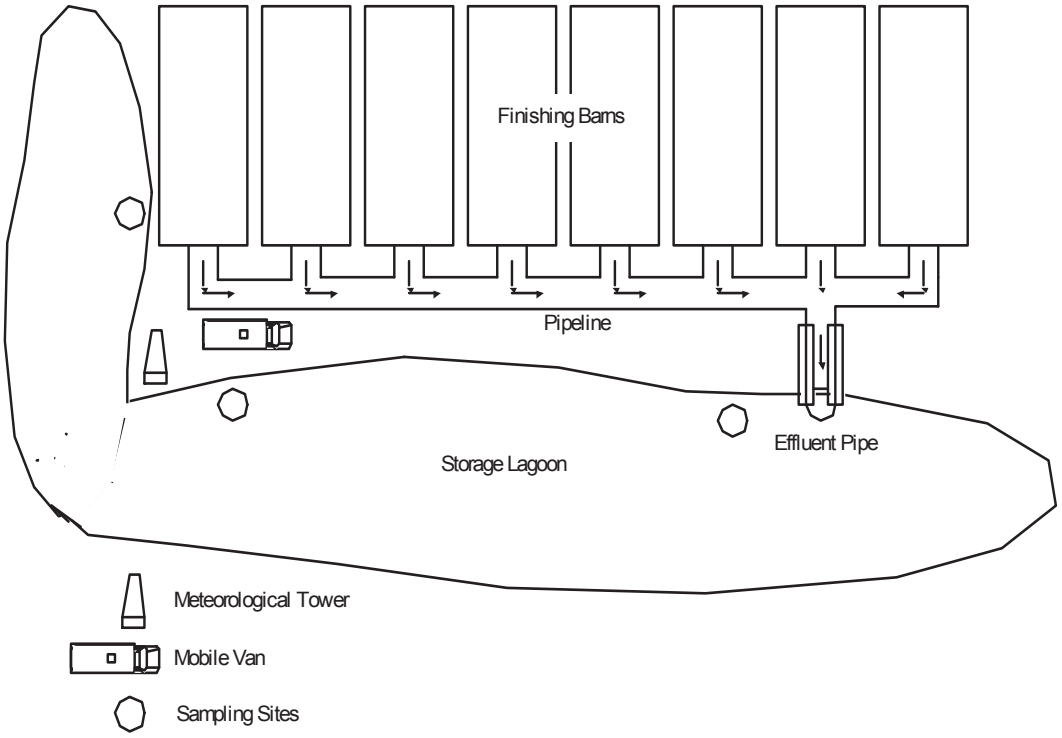
In order to determine fan revolutions per minute (rpm), a Mabuchi VDC motor (Santa Clara, CA) was either mounted to a stainless–steel plate configured to fit over the front the 91 cm fan plate or attached to a cylinder sleeve which fit over the fan shaft of the 122 cm fans. Single analog output wires were connected from each motor to the Campbell Scientific CR10X data logger (Logan, UT) which continuously recorded the measured voltage output every second and averaged the data over a fifteen minute timeframe. Prior to the experiment, each motor was calibrated in the laboratory to obtain voltage outputs at a specific rpm. The rpm for each fan as well as "on/off" times could then be determined and flow rates subsequently calculated. According to manufacturer specifications the direct drive motor on the 91 cm fans is rated at 850 rpm and the 122 cm fan motor is rated at 1 725 rpm. We estimated the "pulley ratio" for the 122 cm fans to be 2.9:1; therefore, the fans should be rotating at ~595 rpms. Blunden and Aneja (2008) determined that the static pressure varied from 0.22–0.32 cm water (21–31 Pa), with an average and median value of 0.28 cm water. This value was subsequently used for further emission rate calculations. The flow rates for each fan size were calculated using the following calculation;

$$FR_C = \left(\frac{FR_M}{RPM_S}\right)(RPM_{Meas}) \quad (1)$$

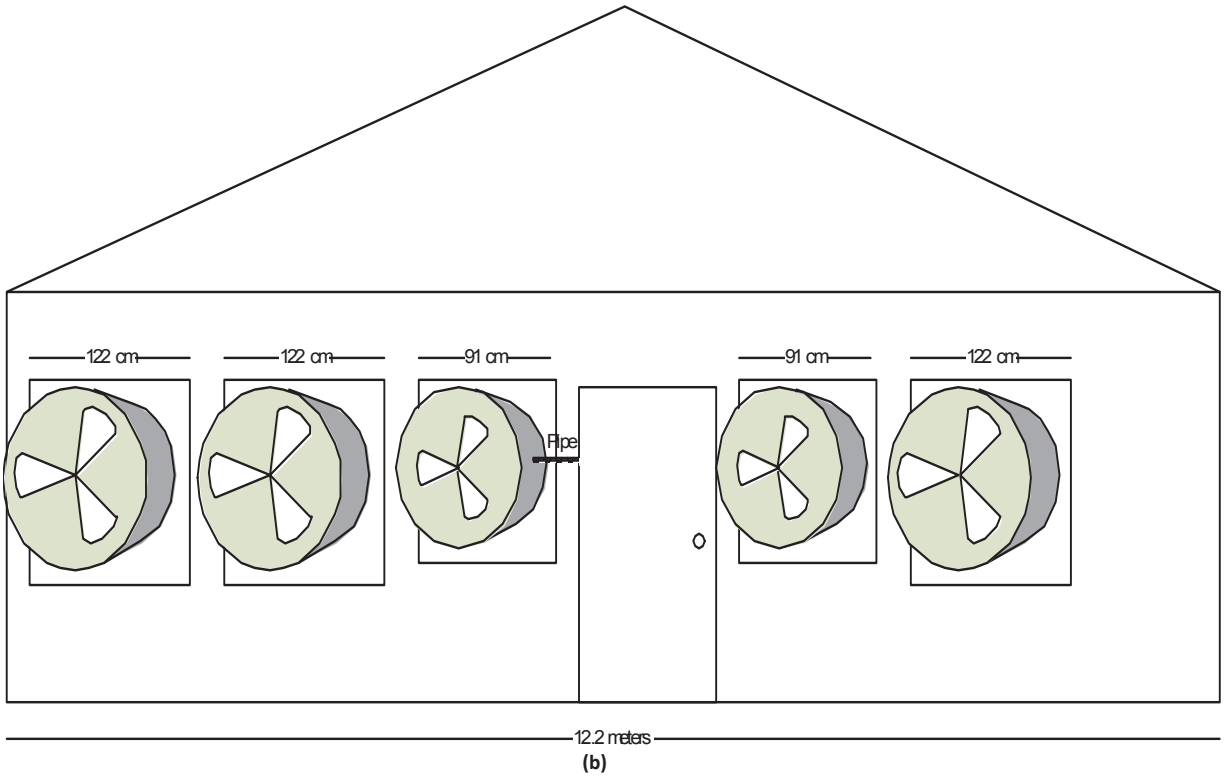
where FR_C is the calculated fan flow rate ($m^3\ min^{-1}$), FR_M is the manufacturers fan flow rate ($m^3\ min^{-1}$) and RPM_S and RPM_{Meas} are the specified and measured revolutions per minute (rpm), respectively. The manufacturer fan flow rate was adjusted to the average static pressure measurements. For example, given a static pressure of 0.28 cm water, manufacturer fan flow rates for the 91 cm fan with shutter and 122 cm fan are ~269 and ~640 $m^3\ min^{-1}$, respectively.

A ¼" o.d., 5/32" i.d. Teflon sample line was inserted inside the chimney of the first 91 cm fan to turn on, between the shutter and fan blade, at roughly half the fan radial distance (see Figure 1). Due

to the nature of the airflow through the building, it is assumed that the gaseous concentrations are uniformly distributed at each fan outlet. The air is drawn through the system in order to deliver the sample to the NH_3 analyzer at a flow rate of ~0.5 L min^{-1}. The barn sampling line was ~5 m in length and the residence time was ~3 seconds. Therefore we observed no condensation in the sampling lines, as the water vapor was continuously flushed out from the sampling lines. Moreover, FEP Teflon was used for the sampling lines, which is recognized to have no hysteresis or wall effects.



(a)



(b)

Figure 1. ***(a)*** *Aerial schematic of study site, a conventional lagoon and spray technology facility and* ***(b)*** *front view of swine confinement house. Not drawn to scale.*

Barn emission rates were calculated using the equation;

$$Emission\ rate = C \times \sum f \quad (2)$$

where C is the gas concentration (μg m^{-3}) and Σf is the sum of the calculated hourly flow rates (m^3 min^{-1}).

Five–minute integrated background samples were collected daily using 6–L electropolished stainless steel SUMMA™ canisters, evacuated to a sub–ambient pressure of <0.05 mm Hg. During sample collection, the valve on the canister was opened slowly over a timeframe of ~4 min and fully opened on the order of 1 min, thus allowing for a ~5 min point sample to be collected. Each sample was drawn through a Teflon tube and delivered to the NH_3 analyzer (temporarily disconnected from the fan outlet sample line). There was a 5–10 minute gap between the time the barn sample line was disconnected and replaced by the background sample line. Data was recorded from the background samples after three minutes, allowing the instruments to stabilize. The background samples collected were on average 0–4% of the concentration values measured for NH_3 at the fan ventilation exhaust and were therefore not considered during emission calculations.

2.3. Lagoon dynamic flow–through chamber system and ammonia analyzer

A dynamic flow–through chamber system with a variable–speed motor–driven continuous impeller stirrer (Aneja et al., 2000) was used to determine ammoniacal nitrogen (NH_3–N) emission from the anaerobic lagoon. The dynamic chamber method offers the ability to examine the flux process in more detail than micrometeorological methods by controlling mixing conditions inside the chamber, and the ability to test treatment effects. Moreover, the chamber technique allows to uniquely relate the flux to physico–chemical properties of the emission location offering the opportunity to parameterize the flux for use in air quality models. The dynamic chamber is a fluorinated ethylene propylene (FEP) Teflon–lined (2 mil thick, ~0.05 mm) open bottom cylinder (0.25 m internal diameter, 0.46 m internal height of chamber above water, 25 L volume), inserted into a floatable platform (0.61 x 0.61 m), which is supported by PVC piping (1.65 m length, 0.15 m diameter) for flotation. When placed over the lagoon, the chamber penetrated the lagoon ~7 cm, effectively forming a seal between the lagoon surface and the air within the chamber. The dynamic chamber was randomly placed on the lagoon surface. A schematic of the dynamic flow–through chamber system is shown in Figure 2.

Compressed zero–grade air (Machine and Welding Supply Company, Raleigh, NC) was used as a carrier gas and pumped through the chamber at a variable flow rate ranging from 5–13 L min^{-1}, controlled by a Model 810–S Mass Trak Flow Controller (Sierra Instruments, Monterey, CA). The air inside the chamber was ideally well–mixed by a variable–speed motor–driven Teflon impeller stirrer ranging from speeds of 40–60 revolutions per minute (rpm). Based on the impeller stirrer design, it is expected that the air flow characteristics inside the chamber at the air–water interface are similar to ambient air, i.e., flow outside the chamber, once steady–state has been reached (Aneja et al., 2000; Aneja et al., 2001). The length of the Teflon tubing (0.40 cm inner diameter) connecting the chamber to a Model 17C chemiluminescence NH_3 analyzer (Thermo Environmental Corporation, Mountain View, CA) was less than 10 m and had a residence time of ~4 seconds. The sample exiting the chamber flowed to the NH_3 analytical instrument: a Model 17C chemiluminescence NH_3 analyzer (Thermo Environmental Corporation, Mountain View, CA). Data collection was initiated once the chamber reached steady state conditions. A vent line was fitted to the exiting sample line to prevent pressurization and was periodically bubble tested to check for under pressurization and/or leaks in the enclosed system.

Multi–point calibrations (80, 60, 40, and 20% of full–range scale) for the Model 17C analyzer were conducted according to the TEI Model 17C instruction manuals prior to each sampling campaign using a TEI Model 146 dilution–titration system (Thermo Environmental Corporation, Mountain View, CA). Each instrument was zeroed and spanned daily during each measurement period.

The suitability of the chamber for measuring NH_3 flux from lagoons is supported by studies by Aneja et al. (2000) and Todd et al. (2001). These studies made NH_3 flux measurements from the same lagoon in 1997 using a chamber system (Aneja et al., 2000) and tracer gas open–path Fourier transform infrared spectroscopy (Todd et al., 2001) respectively. NH_3 flux measurements were similar with fluxes ranging from 5–123 kg NH_3–N ha^{-1} day^{-1} (Aneja et al., 2000) and 7.8–122 kg NH_3–N ha^{-1} day^{-1} (Todd et al., 2001).

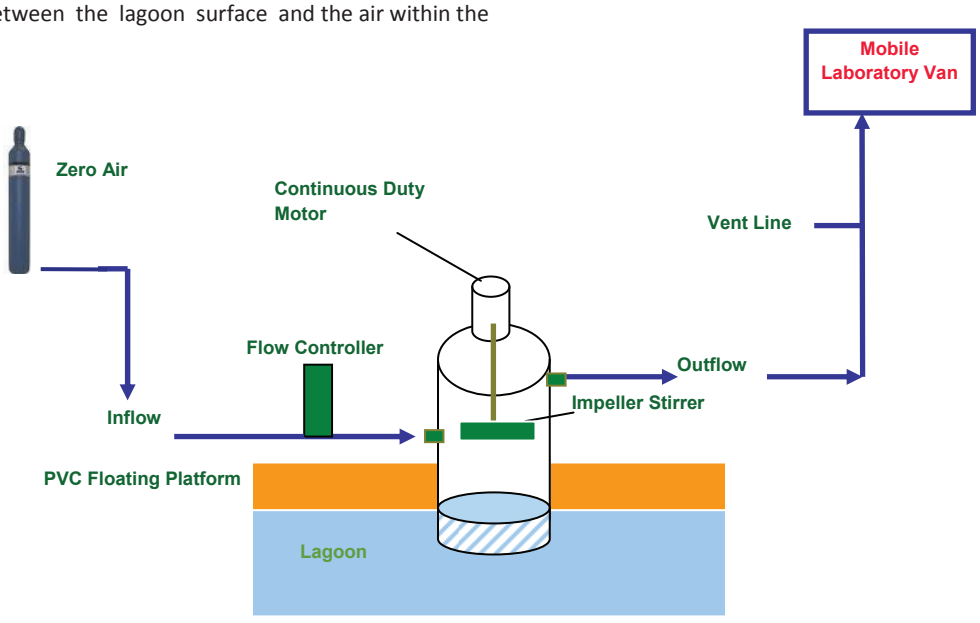


Figure 2. *Schematic of dynamic flow-through chamber system. Not drawn to scale.*

2.4. Lagoon flux calculation

The mass balance for NH_3–N in the chamber is given by;

$$\frac{dC}{dt} = \left(\frac{Q[C]_o}{V} + \frac{JA_L}{V}\right) - \left(\frac{LA_c[C]}{V} + \frac{Q[C]_f}{V}\right) - R \quad (3)$$

where A_L is the lagoon surface area covered by the chamber (m^2), A_C is the inner surface area of the chamber (m^2), V is the volume of the chamber (m^3), Q is the flow rate of carrier gas through the chamber(m^3 min^{-1}), J is the emission flux of the respective gas (μg m^{-2} min^{-1}), [C] is the gas concentration in the chamber (μg m^{-3}), $[C]_f$ is the gas concentration at the outlet of the chamber (μg m^{-3}), L (m min^{-1}) is the loss term by the chamber wall per unit area assumed first order in [C], and R is the chemical production rate in the chamber.

Since compressed zero–grade air was used as the carrier gas, the inlet concentration of NH_3 is $[C]_o$ = 0, and for a well mixed dynamic chamber $[C]_f$ may be assumed to be equal to the gas concentration everywhere in the chamber. The residence time in the chamber is relatively short (ranging from 2–5 minutes, depending on the carrier gas flow rate being used) and therefore no chemical reactions were expected to occur inside the chamber system (R = 0). Finally, at steady state the above equation reduces to;

$$J = [C]_f \left(\frac{LA_c}{V} + \frac{Q}{V}\right) h \quad (4)$$

where h (m) is the height of the chamber measured from the lagoon surface. The value of the total loss term for ammonia was determined by conducting surface loss experiments as proposed by Kaplan et al. (1998), who provided a detailed description of the experiment. Blunden and Aneja (2008) determined during steady–state conditions, the NH_3–N loss term associated with the internal surfaces of the chamber increases as an exponential function [Equation(5)] with ambient temperature (T, °C). This was used throughout the experiment to calculate the loss of NH_3–N to the internal chamber walls and was given by;

$$L_{NH_3-N} = 0.0355\,T - 2.5454\ m\ min^{-1} \quad (5)$$

2.5. Meteorological and lagoon parameters instrumentation

A 10 meter meteorological tower was erected to measure ambient wind speed and direction, air temperature, and relative humidity. A Met One Instruments Model 034–B Windset (Campbell Scientific, Inc., Logan, UT) with an integrated cup anemometer and wind vane mounted on a common shaft was used to measure wind speed at 10 m above the surface. Air temperature and relative humidity (RH) measurements were made at a height of 2 m with a Model CS500–L Vaisala 50Y temperature and RH probe (Campbell Scientific, Inc., Logan, UT) housed in a Model 41303 RM Young 6–plate gill solar radiation shield (Campbell Scientific, Inc., Logan, UT).

A Model CSIM11 pH probe (Campbell Scientific Inc., Logan, UT) was used to continuously monitor near–surface lagoon pH. The pH probe was periodically buffer tested to ensure accuracy and calibration curves were established. Two CS107 (Campbell Scientific Inc., Logan, UT) temperature probes simultaneously measured lagoon surface temperatures inside the chamber as well as 0.3 m outside the chamber. Blunden and Aneja (2008) determined that the difference in lagoon temperature inside and outside the flux chamber are negligible and can be considered to be the same for the purposes of this experiment. The pH and temperature probes were each submerged in the lagoon at a depth of ~6–7 cm beneath the surface.

A Model CR10X data logger equipped with a Model AM 16/32 Channel Relay Multiplexer (Campbell Scientific, Inc., Logan, UT) was used to collect all meteorological and lagoon data and a Model CR23X data logger was used to acquire all gaseous and mass flow data. Data were collected continuously, and averaged and recorded over a 15 minute timeframe. The data loggers and gas analyzers were housed inside a temperature–controlled mobile laboratory (N.C. State University Air Quality Ford Aerostar Mini–Van), maintained at ~21°C.

2.6. Lagoon sample collection

Near–surface lagoon samples (<10 cm depth) were collected daily by hand between 10:00 and 13:00 during each sampling period using sterile plastic 500 mL bottles and submitted to the North Carolina Division of Water Quality (NC DWQ) for determination of total ammoniacal nitrogen (TAN) and total klejdahl nitrogen (TKN). Lagoon samples were also collected during the early morning or late afternoon as well to check for diurnal variations in concentration levels. Samples were collected from the lagoon surface simultaneously near the chamber system and at a randomly chosen location at the lagoon. Lagoon samples collected for TAN and TKN analyses were preserved with sulfuric acid to pH<2, according to NC DWQ specifications. All samples were immediately stored on ice (<4°C) and brought to the NC DWQ laboratory within 48 hours for analysis (Greenberg et al., 1999).

3. Results and Discussion

3.1. Barn emissions

A total of about 388, 564, 496, and 440 fifteen minute averaged barn emission measurements were collected for summer, fall, winter, and spring, respectively. Ventilation rates, barn temperature, and ambient temperature all influence NH_3 emitted from swine finishing buildings (Heber et al., 2002; Arogo et al., 2003; Gay et al., 2003; Heber et al., 2005). Table 2 provides measured fan ventilation rates, temperature at the fan outlet, ambient temperature, and relative humidity. Table 3 lists simple statisitics for measured total emission rate and normalized emission rate for comparison. Emission factors are provided both as g NH_3–N day^{-1} AU^{-1}, where one AU = 500 kg, and also as kg NH_3–N $animal^{-1}$ yr^{-1}. These emission factors were determined based on the production data provided in Table 1. Both types of emissions factors are often provided in related literature e.g. (Arogo et al., 2003; Harper et al., 2004; Blunden et al., 2008).

Table 2. *Average barn measurement statistics for experimental periods during all seasons*

Season	Total Ventilation Rate (m^3 min^{-1}) [a]	Fan outlet temperature (°C)	Ambient temperature (°C)	Ambient relative humidity (%)
Summer 2007				
Average (St dev.)	1 683 (681)	28.0	26.0	67
Range	551-2 370	24.3-33.6	18.2-33.4	29-93
Fall 2007				
Average (St dev.)	343 (128)	20.7	10.2	63
Range	166-991	15.6-24.7	0.5-20.2	26-93
Winter 2008				
Average (St dev.)	313 (252)	18.5	11.6	70
Range	0-1 329	9.1-24.2	1.9-21.3	18-92
Spring 2008				
Average (St dev.)	502 (246)	25.5	16.0	83
Range	234-1 259	21.1-31.2	11.6-28.3	35-94

[a] *Sum of flow from two 91 cm fans and three 122 cm fans*

Table 3. *Statistical summary of total NH_3-N barn emissions, normalized emission rates, and emission factors for each season*

Season	Total emission rate (g day^{-1})	Normalized Emission Rate (g NH_3-N day^{-1} AU^{-1})[a]	Emission Factor (g NH_3-N kg^{-1} day^{-1})	Emission Factor (kg NH_3 animal^{-1} yr^{-1})
Summer 2007, Jun 13-21				
Average (St dev.)	2 604 (660)	30.2 (7.7)	0.06 (0.02)	1.32 (0.32)
Range	1 233-5 702	14.32-66.22	0.03-0.13	0.63-2.85
Fall 2007, Oct 10-20				
Average (St dev.)	1 761 (1 087)	25.6 (15.8)	0.05 (0.03)	0.78 (0.49)
Range	590-6 609	8.6-96.0	0.02- 0.19	0.26-2.95
Winter 2008, Feb 18-26				
Average (St dev.)	1 657 (1 506)	14.9 (13.6)	0.03 (0.03)	1.55 (1.40)
Range	0-8 489	0-76.6	0-0.15	0.00-7.91
Spring 2008, Apr 16-21				
Average (St dev.)	2 659 (1 194)	30.0 (13.5)	0.06 (0.03)	1.35 (0.61)
Range	1 176-9 444	13.3-106.7	0.03-0.21	0.61-4.78

NH_3-N = (14/17) NH_3

[o] AU (Animal Units) = 500 kg live animal weight

Barn air temperature measured at the fan outlet is strongly correlated with ambient air temperature (R^2=0.86, p<0.0001) for all seasons, although barn temperatures are less variable than ambient. As outside temperatures rise or fall, barn temperatures change, causing the ventilation system to respond. A positive correlation was found between barn temperature and ventilation rates (R^2=0.573, p<0.0001). Total ventilation rates increased during summer as temperatures increased and decreased during fall and winter when average barn temperatures were lower; however, it should be noted that the ventilation system was not well correlated with barn temperatures below ~20°C because the fan remains on continuously or kicks on as needed to ensure some airflow through the building at all times.

Figure 3 shows the hourly averaged diurnal emission profile for each season. Each data point has also been averaged over the number of measurements made at that time period. NH_3–N emission rates were highest in the spring and summer, 2 659±1 194 (± represents 1 standard deviation) g day^{-1} and 2 604±660 g day^{-1}, respectively. These rates are attributed to the number and average weight of the swine, and ventilation rates. Total live weight in the spring was 44 262 kg versus 43 049 kg in the summer (see Table 1). Ventilation rates were lower in the spring, averaging 501±247 m^3 min^{-1} compared to 1 683±681 m^3 min^{-1} in summer. Lower ventilation rates allow for gas build up; the build up is released when temperature–dependent fans turn on. Fall NH_3–N emissions averaged 1 761±1 087 g day^{-1} while winter NH_3–N emissions were lowest at 1 657±1 506 g day^{-1}. Although ventilation rates were lower in the fall (343±128 m^3 min^{-1}), animals were in the beginning stage of production weighing a total of 34 428 kg in week five, resulting in lower emissions. A large percentage of winter measurements ranging from 0 g NH_3–N day^{-1} when all fans were off to a short–term maximum of 10–308 g NH_3–N day^{-1} were collected when the primary fan was cycling on and off. Harper et al. (2004) found similar results at a swine finishing operation measuring 2 116 g NH_3–N d^{-1} in the winter and 5 147 g NH_3–N d^{-1} in the summer, suggesting that concentrations and emissions followed the fan cycle as long as cycling was constant. When the primary fan remained on for a longer period of time, ammonia concentrations did not decrease in the same way as when one fan was on intermittently. Turbulent mixing inside the house is likely to be larger in the winter compared to other seasons when some fans were on continously and concentrations inside the building were generally a function of the number of operating fans.

Nighttime spring and summer emissions (between 18:00 and 6:00 Eastern Standard Time (EST)) were approximately twofold higher than emissions measured in the fall and winter. NH_3–N emissions during all four seasons remained relatively constant during the late evening into the early morning hours. During all seasons, emission rates increased after sunrise as ambient and barn temperatures, and thus fan ventilation rates, increased. Spring NH_3–N emissions peaked at approximately 5 500 g NH_3 day^{-1}, just before 12:00 EST, then decreased to about 4 000 g NH_3 day^{-1} between 12:00 and 15:00 EST, when ventilation rates were highest (~1 000 m^3 min^{-1}). Summer emissions were highest about 9:00 EST (3 138±741 g day^{-1}), as emissions built up inside the structure prior to ventilation rate increase. Average temperatures and ventilation rates were substantially lower in the fall and winter; therefore, NH_3–N daytime emissions increased to levels greater than those measured during summer daytime.

Normailized emission rates of 30.2±7.7 g NH_3–N day^{-1} AU^{-1}, 26±16 g NH_3–N day^{-1} AU^{-1}, 14.9±13.6 g NH_3–N day^{-1} AU^{-1}, and 30.0±13.5 g NH_3–N day^{-1} AU^{-1} were found for the summer, fall, winter, and spring respectively. Normalizing emission rates involves removing total live mass as a variable to explain emission rates, allowing for more accurate comparison between seasons and other studies.

An often used comparison is based on emission rates per animal. Emission factors for this study were estimated to be 1.3±0.3 kg NH_3 animal^{-1} year^{-1}, 0.8±0.5 kg NH_3 animal^{-1} year^{-1}, 1.5±1.4 kg NH_3 animal^{-1} year^{-1}, and 1.4±0.6 kg NH_3 animal^{-1} year^{-1} in the summer, fall, winter, and spring, respectively. Emissions per animal were greatest in the winter when the animals were in their final week of rotation and smallest in the fall when swine were in the early stages of production. Results from this study are comparable to those presented in Blunden et al. (2008), which found finishing pigs emitted 0.8 kg NH_3 animal^{-1} year^{-1}, 0.5 kg NH_3 animal^{-1} year^{-1}, 1.7 kg NH_3 animal^{-1} year^{-1}, and 2.4 kg NH_3 animal^{-1} year^{-1} at the same facility in the summer, fall, winter, and spring of 2005, respectively.

3.2. Lagoon emissions

A total of 462 (summer), 493 (fall), 386 (winter), and 460 (spring) fifteen minute averaged lagoon flux measurements were collected for each season. Data were not collected during precipitation events.

The maximum concentration that can be detected by the NH_3 analyzer is 20 000 parts per billion (ppb). During summer, 25% of measured concentrations exceeded the limit, while 1% during fall were above this range. Only data within the range of the NH_3 analyzer was used for analysis.

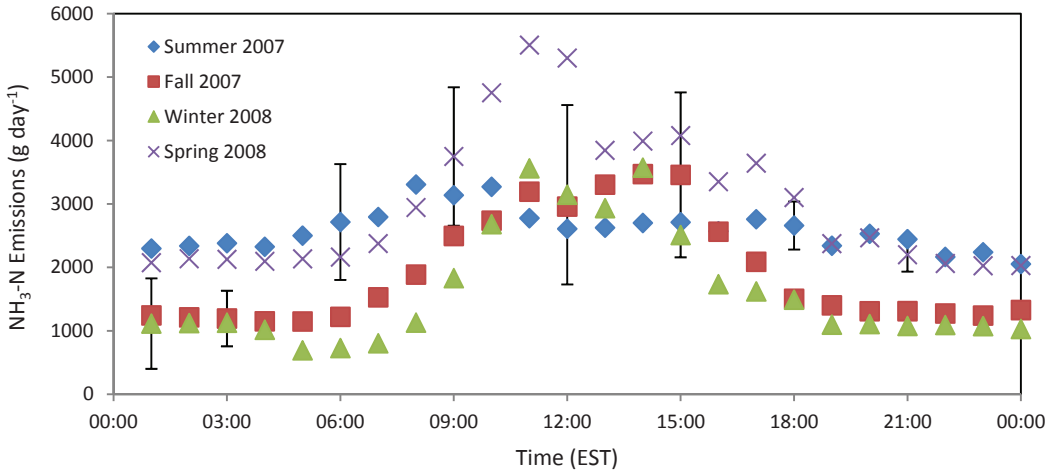


Figure 3. *Seasonally averaged diurnal variations of NH_3-N barn emissions (g day^{-1}). Randomly selected error bars represent ± one standard deviation.*

Table 4 provides statistical data for NH_3–N flux and Table 5 lists measured lagoon surface temperature, pH, TAN, TKN, ambient temperature, and wind speed for each season. Sommer et al. (1991) found lagoon temperature and pH to be the two most important factors that influence NH_3 volatilization.

Table 4. *Average NH_3-N lagoon flux statistical averages during each sampling period*

Season	Sample Dates	Mean (μg m^{-2} min^{-1})	Minimum (μg m^{-2} min^{-1})	Maximum (μg m^{-2} min^{-1})
Summer	Jun. 13-21, 2007	>3 943 [a]	1 875	NA
Fall	Oct. 10-20, 2007	>1 383 [b]	681	NA
Winter	Feb. 18-26, 2008	981 (210) [c]	501	2 818
Spring	Apr. 16-21, 2008	1 641 (362)	1 842	3 860

NH_3-N flux = (14/17) NH_3 flux

[a] NH_3 concentration was above the maximum rage on the ammonia analyzer for 25% of 15-minute averaged data during experimental period; therefore, maximum and average flux values cannot be determined.

[b] NH_3 concentration was above the maximum rage on the ammonia analyzer for 1% of 15-minute averaged data during experimental period; therefore, maximum and average flux values cannot be determined.

[c] Numbers in parenthesis represent 1 standard deviation.

NH_3–N flux. The use of a dynamic–flow through chamber system as an appropriate flux measuring technique has been corroborated by an earlier inter–comparison of NH_3 flux measurements form the same lagoon in 1997 of the chamber system (Aneja et al., 2000) and tracer gas open–path Fourier transform infrared spectroscopy (Todd et al., 2001) respectively. NH_3 flux measurements were similar with fluxes ranging from 5–123 kg NH_3–N ha^{-1} day^{-1} (Aneja et al., 2000) and 7.8–122 kg NH_3–N ha^{-1} day^{-1} (Todd et al., 2001).

Figure 4 shows the hourly averaged diurnal emissions trend for each season. Similar to Figure 3, each hourly averaged data point shown has been averaged over the number of measurement days. Average NH_3–N fluxes were highest in the summer, greater than >3 943 μg m^{-2} min^{-1}, and lowest in the winter, 981±210 μg m^{-2} min^{-1}. Fall and spring average ammonia flux values were >1 383 μg m^{-2} min^{-1} and 1 641±362 μg m^{-2} min^{-1} respectively. Results from this study are slightly lower than those found by Blunden and Aneja (2008) in each season, who determined ammonia emissions to be >4 292, 1 634, 1 290, and >2 495 μg m^{-2} min^{-1} in the summer, fall, winter and spring at the same site using the same chamber system. Lower fluxes in this study may be attributed to differences in average lagoon temperatures, pH, and TAN concentrations.

During all seasons, NH_3–N flux decreased during the night until sunrise. NH_3–N flux generally increased throughout the course of the day peaking in the mid to late afternoon before beginning to decline after sunset. Some points that fall outside the general pattern during the daytime are likely due to disruptions in data collection for routine maintenance in the field.

Table 5. *Statistical parameters for NH_3-N lagoon and ambient air parameters during each sampling period*

Season	Sample Dates	Lagoon Temperature (°C)	Lagoon pH	TAN (mg L^{-1})	TKN-N (mg L^{-1})	Ambient Temperature (°C)	Wind Speed (m s^{-1})
Summer							
Average (St dev.)		24.8 (0.9)	7.6 (0.4)	477 (35)	556 (19)	21.6 (3.5)	1.6 (1.0)
Range	Jun. 13-21, 2007	19.5-28.9	7.2-8.3	440-510	520-590	11.5-34.8	0.0-5.4
Fall							
Average (St dev.)		20.4 (1.2)	7.6 (0.2)	384 (24)	445 (25)	14.8 (5.0)	1.6 (1.5)
Range	Oct. 10-20, 2007	15.0-25.3	7.2-8.0	330-420	390-480	1.8-31.2	0.0-5.6
Winter							
Average (St dev.)		11.5 (0.7)	8.0 (0.1)	544 (11)	595 (48)	8.4 (3.8)	2.3 (1.4)
Range	Feb. 18-26, 2008	9.7-15.4	7.7-8.1	530-560	550-740	-0.6-20.9	(0.0-8.8)
Spring							
Average (St dev.)		19.9 (1.7)	8.0 (0.3)	549 (3)	616 (40)	16.2 (5.4)	2.1 (1.5)
Range	Apr. 16-21, 2008	15.7-24.0	6.1-9.2	540-550	560-690	1.6-29.5	0.0-6.9

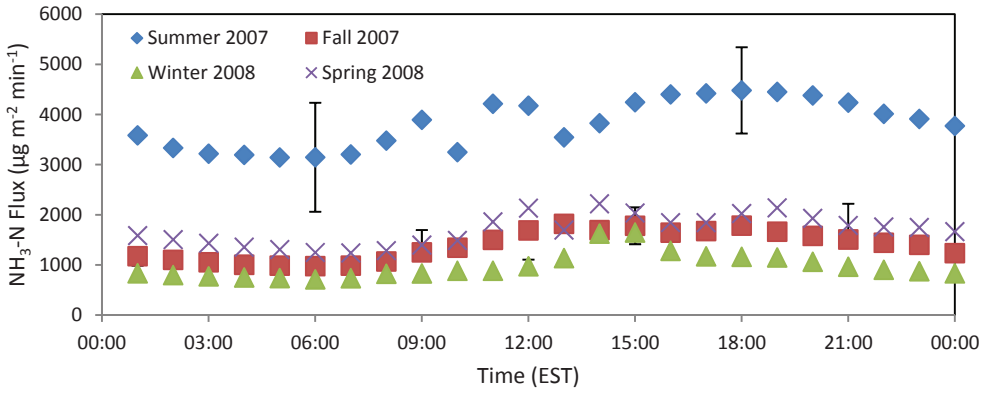


Figure 4. *Seasonally averaged diurnal variations of NH_3-N flux (µg m^{-2} min^{-1}) from the lagoon. Randomly selected error bars represent ± one standard deviation.*

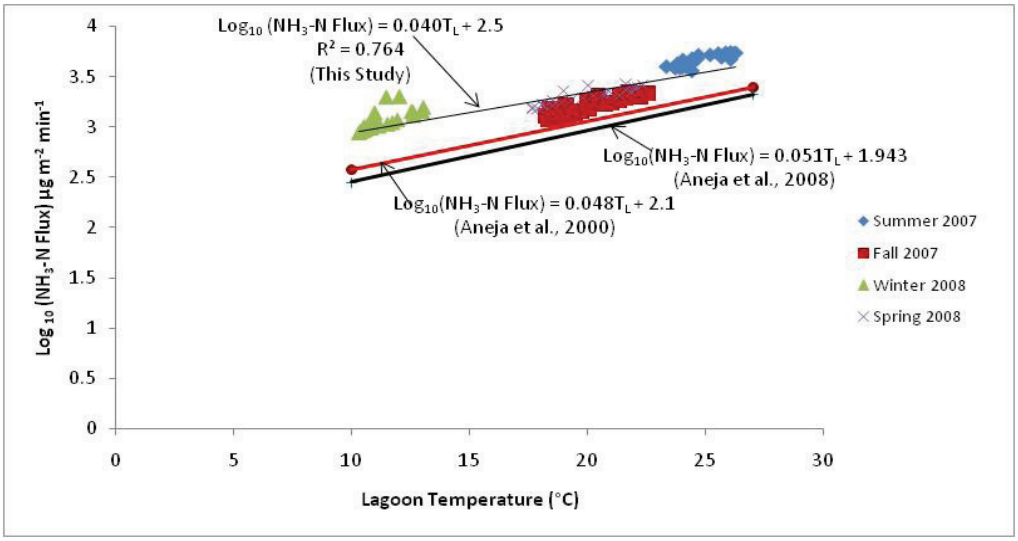


Figure 5. *Log_{10} NH_3-N Flux (µg m^{-2} min^{-1}) plotted against lagoon temperature.*

Lagoon temperature and ammonia flux. Maximum NH_3–N emissions in the summer and minimum NH_3–N emissions in the winter suggest that temperature is an important factor influencing NH_3–N flux from the lagoon into the atmosphere. Seasonally averaged lagoon temperatures measured 24.8 °C in the summer, 20.4 °C in the fall, 11.3 °C in the winter, and 19.9 °C in the spring. Figure 5 shows the exponential relationship between lagoon temperature and NH_3–N flux (R^2=0.764, p<0.0001). Each point in the figure represents an hourly–averaged NH_3–N flux plotted against the simultaneously measured hourly lagoon temperature. This compares well with Aneja et al. (2000) and Aneja et al. (2008), who found a similar relationship and developed predictive models based on lagoon temperature (Figure 5). A predictive model for this study based on observations is given as;

$$Log_{10}(NH_3\text{-}N\ flux) = 0.040T_L + 2.5 \quad (6)$$

The exponential relationship exists between NH_3–N flux and lagoon temperature because the liquid–phase mass transfer coefficients of NH_3 in water are exponential functions of temperature in a range of 5 °C to 30 °C (Ibusuki and Aneja, 1984). Higher temperatures increase NH_3 volatilization rates due to decreased solubility, in turn affecting ammonia/ammonium (NH_3/NH_4^+) equilibrium, which follows Henry's law for dilute systems (Koerkamp et al., 1998).

Because the lagoon is not forcefully mixed, ammonia's main mode of transport is through diffusion and mass transfer processes (Muck and Steenhuis, 1982). Increased NH_3 volatilization rates occur at the surface of the lagoon as temperatures rise causing decomposed NH_3 stored at the bottom of the lagoon to diffuse upward to replenish volatized NH_3. Higher temperatures increase the transfer rate of NH_3 across the liquid–gas interface through these chemical and physical processes.

Lagoon pH and ammonia flux. The pH of the slurry determines whether ammonia exists as the ammonium ion (NH_4^+) and/or unionized molecules of ammonia ($NH_{3(aq)}$). Unionized $NH_{3(aq)}$ in solution increases with pH and only the unionized fraction can volatilize.

Average pH values ranged from 7.5 in the summer to 8.0 in the spring and winter. Average fall pH values were 7.6. The stability of pH throughout the year can be explained by the high buffer capacity of slurry (Olesen and Sommer, 1993). The dissociation of NH_4^+ causes the release of the hydrogen ion (H^+) allowing unionized $NH_{3(aq)}$ to form.

$$NH_4^+(aq) \leftrightarrow NH_3(aq) + H^+(aq) \quad (7)$$

The logarithmic plot of hourly averaged NH_3–N flux versus pH suggests a negative relationship exists between NH_3–N flux and pH with 29% of the variation (R^2=0.29, p<0.0001) explained by pH (Figure 6). This finding is different from numerous published modeling studies that found a positive relationship between ammonia flux and pH based on the equilibrium reaction below, where the pH of the slurry controls the shift in equilibrium (Sommer et al., 1991; Olesen and Sommer, 1993; Aneja et al., 2000; Warneck, 2000):

$$NH_3(aq) + H_2O(aq) \leftrightarrow NH_4^+(aq) + OH^-(aq) \quad (8)$$

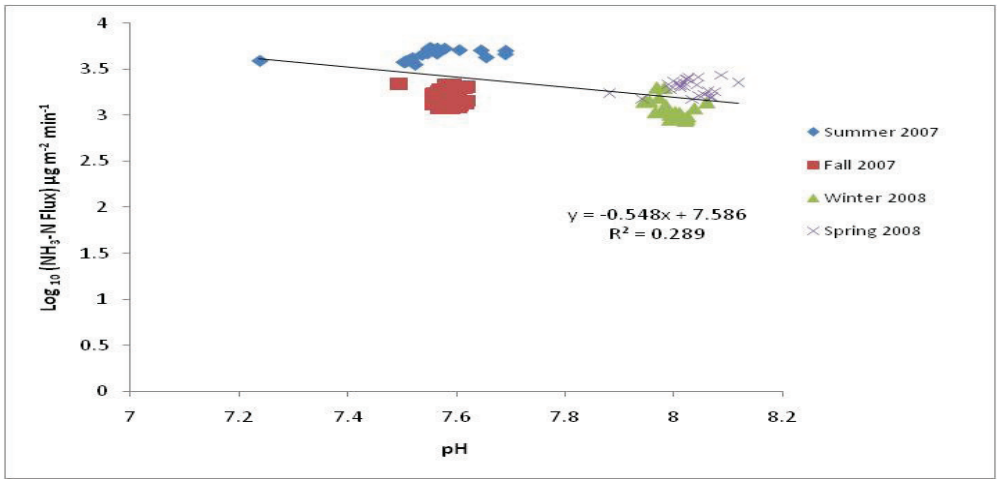


Figure 6. *Log_{10} NH_3-N Flux (µg m^{-2} min^{-1}) plotted against pH.*

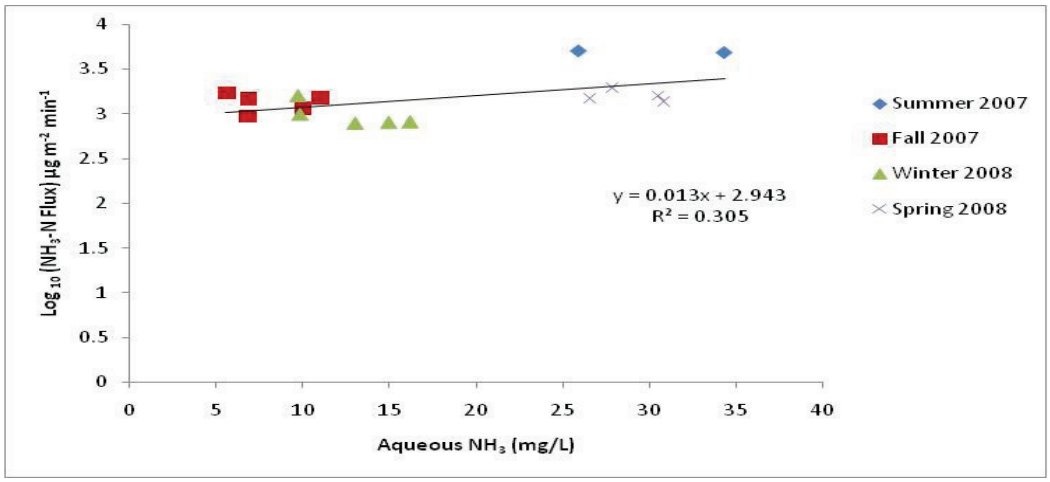


Figure 7. *Log_{10} NH_3-N Flux (µg m^{-2} min^{-1}) plotted against aqueous NH_3 based on TAN concentrations.*

A higher concentration of the hydroxyl ion (OH^-) causes pH to increase shifting the equilibrium to the left releasing more ammonia into the atmosphere. Precipitation events shift equilibrium to the right causing less ammonia to be emitted (Aneja et al., 2009). Blunden (2006) presented a similar correlation between lagoon flux and pH (R^2=0.40, p<0.0001), suggesting that other influential factors (i.e. lagoon temperature, effluent concentration) play a larger role in determining the magnitude of NH_3 flux.

TAN, TKN, and NH_3–N flux. Average concentrations of total ammoniacal nitrogen (TAN) (NH_3+ NH_4^+) were 477 mg L^{-1} as N, 384 mg L^{-1} as N, 544 mg L^{-1} as N, and 549 mg L^{-1} as N in the summer, fall, winter, and spring respectively. Levels of total Kleijdahl nitrogen (TKN) ranged from 445 mg L^{-1} as N in the fall to 616 mg L^{-1} as N in the spring. These results are comparable to those found at similar swine finishing operations e.g. Aneja et al. (2008). Measured concentrations of TAN are useful in determining the amount of unionized NH_3 aqueous concentration in the waste lagoon. Figure 7 shows a positive correlation between average daily NH_3–N flux and aqueous NH_3 concentration (R^2=0.31, p=0.03) when samples and measurements were conducted on the same day. Increasing TAN or TKN concentrations in the lagoon will increase emission of NH_3 (Aneja et al., 2000).

4. Conclusions

Ammonia emissions at a commercial swine finishing facility were measured from a confinement house and a waste storage treatment lagoon for appoximately one week during each season over a one year period to evaluate diurnal and seasonal variations, investigate the influence of meteorological parameters on emissions, and develop emissions factors for use in process based modeling. NH_3–N emissions from the barn were determined to be 2 604±660 g day^{-1}, 1 761±1 087, 1 657±1 506 g day^{-1}, and 2 659±1 194 g day^{-1} in the summer, fall, winter, and spring seasons respectively. Emission factors for swine housing were greatest in the summer and spring measuring 0.061±0.015 g NH_3–N kg^{-1} day^{-1} and 0.060±0.027 g NH_3–N kg^{-1} day^{-1} respectively. Housing emission factors were 0.051± 0.032 g NH_3–N kg^{-1} day^{-1} in the fall and 0.03±0.03 g NH_3–N kg^{-1} day^{-1} in the winter. These results are largely explained by average weights and ventilation rates during each season. A low emission factor in the winter is likely a result of the primary fan turning off and on at cold temperatures—a dynamic that has been largely ignored or disregarded in previous experiments. NH_3–N fluxes from the lagoon were generally in agreement with results found in other studies, measuring >3 943±398 µg m^{-2} min^{-1}, 1 383±283 µg m^{-2} min^{-1}, 981±210 µg m^{-2} min^{-1}, and 1 641±362 µg m^{-2} min^{-1} in the summer, fall, winter, and spring respectively. Relationships found between ammonia flux and average lagoon temperature, pH, and TAN concentrations all suggest that each parameter influences ammonia flux.

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