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ABSTRACT
Ammonia (NH₃) fluxes from waste treatment lagoons and barns at two conventional swine farms in eastern North Carolina were measured. The waste treatment lagoon data were analyzed to elucidate the temporal (seasonal and diurnal) variability and to derive regression relationships between NH₃ flux and lagoon temperature, pH and ammonium content of the lagoon, and the most relevant meteorological parameters. NH₃ fluxes were measured at various sampling locations on the lagoons by a flow-through dynamic chamber system interfaced to an environmentally controlled mobile laboratory. Two sets of open-path Fourier transform infrared (FTIR) spectrometers were also used to measure NH₃ concentrations for estimating NH₃ emissions from the animal housing units (barns) at the lagoon and spray technology (LST) sites. Two different types of ventilation systems were used at the two farms. Moore farm used fan ventilation, and Stokes farm used natural ventilation. The early fall and winter season intensive measurement campaigns were conducted during September 9 to October 11, 2002 (lagoon temperature ranged from 21.2 to 33.6 °C) and January 6 to February 2, 2003 (lagoon temperature ranged from 1.7 to 12 °C), respectively. Significant differences in seasonal NH₃ fluxes from the waste treatment lagoons were found at both farms. Typical diurnal variation of NH₃ flux with its maximum value in the afternoon was observed during both experimental periods. Exponentially increasing flux with increasing surface lagoon temperature was observed, and a linear regression relationship between the logarithm of NH₃ flux and lagoon surface temperature ($T_s$) was obtained. Correlations between lagoon NH₃ flux and chemical parameters, such as pH, total Kjeldahl nitrogen (TKN), and total ammoniacal nitrogen (TAN) were found to be statistically insignificant or weak. In addition to lagoon surface temperature, the difference ($D$) between air temperature and lagoon surface temperature was also found to influence the NH₃ flux, especially when $D > 0$ (i.e., air hotter than lagoon). This hot-air effect is included in the statistical-observational model obtained in this study, which was used further in the companion study (Part II), to compare the emissions.
from potential environmental superior technologies to evaluate the effectiveness of each technology.

INTRODUCTION

Ammonia (NH₃) is a highly biologically active reduced form of nitrogen, which plays a central role in the nitrogen cycle in the atmosphere, biosphere, lithosphere, and hydrosphere.¹² In the soil, organic nitrogen from once living organisms undergoes mineralization, creating ammonium (NH₄⁺) and NH₃, which exist in aqueous equilibrium. NH₃ also originates from urea, a large component of animal excreta. The urease enzyme, which can be produced by microbial organisms present in waste, breaks urea down into NH₃ and carbon dioxide.³ Depending on ambient conditions such as pH and temperature, gaseous NH₃ can volatilize from soil and liquid surfaces and enter the atmosphere.⁴

As the dominant gas base specie in the atmosphere, NH₃ readily combines with and neutralizes acidic compounds, including sulfuric, nitric, and hydrochloric acid, creating NH₄⁺ aerosols.⁵–⁸ Gaseous NH₃ and NH₄⁺ aerosol undergo wet and dry deposition to the earth’s surface.⁴ The atmospheric deposition of reduced and oxidized nitrogen has received much attention in the past decade because of the harmful effects of excessive nitrogen deposition to nutrient sensitive ecosystems,⁹–¹³ such as eutrophication in coastal and estuarine areas.¹⁴

A significant source of NH₃ emissions (~80%) in the United States is domestic animal waste.¹⁵,¹⁶ NH₃ emissions from extensive commercial swine operations provide a large portion of the atmospheric nitrogen compounds that may deposit and contribute to nitrogen loading to sensitive aquatic ecosystems in North Carolina.¹⁴,¹⁷,¹⁸ The pig population in North Carolina has increased from approximately 3 million to 10 million from 1992 to 1997.¹⁰,¹⁹–²¹ Much of the growth in North Carolina’s swine farms has occurred in eastern North Carolina, with the majority of them concentrated in the coastal plain areas.²² Current estimates indicate that atmospheric NH₃ emitted from North Carolina’s swine facilities accounts for 20.6% of the state’s atmospheric nitrogen emission (~0.33 Tg N yr⁻¹) and 46% of the state’s atmospheric NH₃ emission (~0.15 Tg N yr⁻¹).¹⁰,¹¹,¹³,¹⁴,²³,²⁴ The conventional lagoon and spray technology (LST) is the current system used in North Carolina to manage pig waste. It consists of anaerobic lagoons to store and biologically treat pig waste (~99.5% liquid); effluent from the lagoons are sprayed on surrounding crop fields as a nutrient source.¹⁰ Four distinct components and associated processes of LSTs release NH₃ to the atmosphere: (1) production houses, (2) waste storage and treatment systems such as lagoons, (3) land application through injection or spraying, and (4) biogenic emissions from soils and crops.¹⁰ On the basis of recent estimates,²² the lagoon NH₃ emissions in eastern North Carolina comprise approximately 33% of total swine NH₃ emissions in North Carolina with barns and land applications also emitting approximately 33% each. Quantification of NH₃ emissions sources from swine farms is necessary to assess their potential environmental impacts and help develop prudent control and waste treatment strategies.

The main objective of this study was to measure NH₃ emissions from storage and treatment lagoons and animal houses at two conventional swine farms with LST during several experimental periods of approximately 2 weeks, each representing different seasons, namely cool/cold and warm/hot, to develop the empirical relationships between lagoon NH₃ emission and physicochemical and environmental parameters that mainly control the flux from lagoon, such as lagoon temperature, pH, total Kjeldahl nitrogen (TKN), air temperature, and wind speed. The statistical-observational model was used to estimate the lagoon emissions for the baseline farms with LST for comparison with emissions from water-holding structures at potential environmental superior technology (EST) farms described in the companion papers.²⁵,²⁶ Estimated barn emissions at conventional LST farms are also compared with those from the EST farms in Part II.²⁵

INSTRUMENTATION AND SAMPLING SCHEME

Sampling Sites

NH₃ flux measurements were made during two different seasons at two conventional (i.e., LST) swine farms in eastern North Carolina, their locations shown in Figure 1. The two finishing conventional farms were Stokes and Moore Brothers farms, respectively. Finishing refers to the age and type of pigs. Generally pigs go into these farms weighing approximately 23 kg, they stay in the finishing houses for approximately 18 to 20 weeks, where they are fed and grown to approximately 114 kg before marketing them to processing facilities. Our measurements of NH₃ flux were limited to two 2-week-long periods representing warm and cold seasons.

Stokes farm (35.43 °N, 77.48 °W, 17 m mean sea level [MSL]) is located in Pitt County, North Carolina. Measurement campaigns were conducted from September 9 to 20, 2002 and January 6 to 17, 2003. Four naturally ventilated finishing barns housed 4392 animals with an average mass of 104 kg in the fall season and 3727 animals with an average mass of 88 kg in the winter season. The waste (urine and feces) from the pig houses was flushed periodically (four times a day) with recycled lagoon waste and discharged into a storage lagoon from a single effluent pipe. The storage and treatment lagoon was an anaerobic system with 15,170 m² of lagoon surface area.

Sampling at Moore Brothers farm (35.14 °N, 77.47 °W, 13 m MSL), located near Kinston in Jones County, North Carolina, was conducted from September 30 to October 11, 2002 and January 27 to February 7, 2003. The farm has eight fully slatted finishing houses (pit recharge) with tunnel ventilation system. The eight finishing barns housed 7611 animals with an average mass of 52 kg in the fall season and 5784 animals with an average mass of 67 kg in the winter season. Pit recharge houses are typically flushed once a week. Waste from all the pig barns was flushed out with recycled lagoon waste and discharged into a storage and treatment lagoon from eight effluent pipes, one for each pig barn. The lagoon was an anaerobic system with 17,150 m² of surface area. Table 1 gives a summary of the farm information (lagoon area, number of pigs, pig mass, feed consumption, nitrogen content, and nitrogen excretion) during the two sampling
periods at both conventional farms. NH₃ fluxes were measured from various sampling locations on these storage lagoons during the experimental campaigns at the conventional farms.

Environmental Measurements
During the sampling periods, two Campbell Scientific Instruments model 107 temperature probes measured lagoon waste temperature, and an Innovative Sensors pH probe monitored pH of lagoon waste adjacent to the dynamic chamber system. One of the lagoon temperature probes and the pH probe were suspended from the edge of the wooden platform (used as a floatation device for the chamber) and submerged in the lagoon waste approximately 15 cm below the surface. The other temperature probe was attached to the underside of the platform and monitored water temperature inside the chamber. On a 10-m tall meteorological tower were mounted a Campbell Scientific Instruments CS500 temperature and relative humidity (RH) probe, a LI-Cor 200SZ pyranometer, and a Campbell Scientific Instruments Met One 034A-LC Windset integrated cup anemometer and wind vane. These instruments were used to continuously measure the ambient air temperature, RH, solar radiation, wind speed, and wind direction, respectively. A 50-mL sample of lagoon waste was collected daily from the lagoon flux sampling location (stored near 0 °C), and analyzed for pH, NH₃-N, nitrate (NO₃⁻)-N, and TKN in the Weaver Laboratory of the Biological and Agricultural Engineering Department at North Carolina State University. These parameters have been utilized for the purpose of developing the statistical-observational model for lagoon NH₃ flux at conventional farms (i.e., LST). Ambient NH₃ concentration was also monitored continuously by one of the TEI model 17c chemiluminescence NH₃ analyzers (Thermo Environmental Instruments, Inc.). A sample stream of ambient air travels through ¼-in. Teflon tubing from the top of the 10-m meteorological tower to one of the NH₃ analyzers housed in the mobile laboratory.

Lagoon NH₃ Flux Measurements
NH₃ fluxes from the waste storage lagoons were measured by a dynamic flow-through flux chamber system interfaced to an environmentally controlled mobile laboratory (Figure 2). The on-site measurement period for each season was limited to 2 weeks at the experimental farm sites. NH₃ fluxes from the animal storage waste lagoons, other

Table 1. The summary of lagoon surface area, number of pigs, animal mass, feed consumed, nitrogen content, and nitrogen excretion at the two conventional farm sites (Stokes and Moore farms) during the sampling periods.

<table>
<thead>
<tr>
<th>Farm</th>
<th>Season</th>
<th>Lagoon Surface Area (m²)</th>
<th>Number of Pigs</th>
<th>Average Pig Mass (kg/pig)</th>
<th>Feed Consumed (kg/pig/week)</th>
<th>Nitrogen Content in Feed (%)</th>
<th>Nitrogen Excretion, E (kg-N/week/1000 kg · lm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stokes</td>
<td>Fall (September 9–20, 2002)</td>
<td>15,170</td>
<td>4,392</td>
<td>104.3</td>
<td>12.84</td>
<td>3.14</td>
<td>2.71</td>
</tr>
<tr>
<td></td>
<td>Winter (January 6–17, 2003)</td>
<td>3,727</td>
<td>88.5</td>
<td>12.59</td>
<td>2.52</td>
<td></td>
<td>2.51</td>
</tr>
<tr>
<td>Moore</td>
<td>Fall (September 30 to October 11, 2002)</td>
<td>17,150</td>
<td>7,611</td>
<td>52.3</td>
<td>10.99</td>
<td>3.02</td>
<td>4.39</td>
</tr>
<tr>
<td></td>
<td>Winter (January 27 to February 2, 2003)</td>
<td>5,784</td>
<td>67.0</td>
<td>12.37</td>
<td>2.95</td>
<td></td>
<td>3.90</td>
</tr>
</tbody>
</table>
water-holding structures, and spray fields at the two conventional and several potential EST sites were measured by the dynamic flow-through flux chamber system. The times of spray did not occur close to our measurement periods.

**Dynamic Flow-Through Chamber System.** A flow-through dynamic chamber system with a variable-speed continuous impeller stirrer was used to determine NH$_3$ flux from the lagoon surface$^{22,27}$ The translucent plastic cylindrical chamber, 26 cm in diameter and 46 cm in height (a volume equal to $\sim$24.4 L), was fitted into a circular hole cut into the center of a 1.2- by 1.2-m floating plywood platform, which penetrated the lagoon surface by approximately 7 cm. To create a closed system inside the chamber, a seal was formed between the bottom of the cylinder and the lagoon waste. The cylindrical chamber was lined with a 5-mm thick fluorinated ethylene propylene (FEP) Teflon sheet throughout the inside surface of the chamber. Compressed zero-grade air was used as a carrier gas. The carrier gas was passed through the chamber at a constant flow rate, but these rates often varied from site to site, within the range of approximately 4–10 L min$^{-1}$, set by a mass flow controller and continuously recorded on a data logger. The air inside the chamber was ideally well mixed by a variable speed motor-driven Teflon impeller ranging from speeds of 40–60 rpm for this study. A vent line to the atmosphere was fitted to the sample line of NH$_3$ to prevent overpressurization in the enclosed system and also to ensure that the NH$_3$ analyzers measure steady-state NH$_3$ concentration from the sample stream. Between each daily or diurnal sampling period, the floating chamber system was moved in a random manner to another location on the lagoon within a 10-m radius of the

![Figure 2. Schematic of a flow-through dynamic chamber system interfaced with an environmentally controlled North Carolina State University temperature-controlled mobile laboratory.](image-url)
mobile laboratory. To capture the spatial variability of a lagoon, the mobile laboratory, meteorological tower, and floating chamber system were periodically moved. The NH₃ analyzers were multipoint calibrated before sampling began at each new area and zeroed and spanned every other day.

Chamber Effects. As with any environmental investigation that involves imposing a constructed system on the natural environment, concerns arise in response to the use of the dynamic chamber system. Sensitivity investigations can elucidate the extent to which an experimental system may alter the ambient conditions of the associated microclimate.

To quantify the effect of excessive humidity or moisture in the dynamic chamber system, Roelle introduced visible moisture droplets on the chamber walls and reported NH₃ losses ranging from 6 to 7%. In the study presented here, with compressed air as a carrier gas, moisture can accumulate within the chamber. However, the flow of air through the chamber system consistently inhibited any visible moisture droplets from forming during measurement periods. Thus, NH₃ losses because of moisture in the chamber lie well within the constraints of the humidity experiment and are therefore determined to be minimal.

The vent on the sample line exiting the chamber minimizes the pressure difference between the ambient atmosphere and the air inside of the chamber. To assess the effect of mixing speed on calculated NH₃ flux, the impeller stirrer speed was varied between 20 and 60 rpm during the afternoon of a typical diurnal sampling period. Wind speed at a height of approximately 10 m was continuously monitored during NH₃ flux measurement periods. The corresponding wind speeds at a height of 0.5 m (approximate height of the chamber) may be calculated by the power-law profile expression

\[ \frac{V_f}{V_r} = \left( \frac{z_f}{z_r} \right)^m \]  

where \( V \) is the wind speed at height \( z \), \( V_r \) and \( z_r \) are the reference wind velocity and reference height, respectively; \( m \) is taken to be 0.1 for a smooth water surface in unstable daytime conditions.

Throughout the experimental periods during this study, measured mean wind speeds ranged from 1 to 4 m/sec at a height of 10 m. The calculated wind speeds at a height of 0.5 m (approximate height of the chamber) ranged from 0.7 to 3 m/sec, which was similar to the range of airflow speeds within the chamber measured by a digital hot wire anemometer at various carrier gas flow rates and impeller speeds.

\[ \text{NH}_3 \text{ Analyzer and Flux Calculation} \]. Once the dynamic flow-through chamber system reached steady-state conditions (~30 min of operation), the sample flow from the chamber was introduced into a TEI model 17c chemiluminescence NH₃ analyzer housed in the temperature-controlled mobile laboratory, and NH₃ concentration in the sample flow was measured by the analyzer. Calibrations of the NH₃ analyzer were conducted using a TEI 146 dilution-titration system in conjunction with two different concentrations of calibration gas mixtures of NH₃ (20 and 900 ppmv) in nitrogen and zero-grade air (Machine and Welding Specialty Gases, National Institute for Standards and Technology [NIST] certified). A multipoint calibration typically consists of points at zero, 20, 40, 60, and 80% of the full-scale range. The full-scale range typically equals 20 ppmv for liquid waste surface measurements. The multipoint calibration was conducted before each 2-week field measurement campaign. Zero and span checks were conducted every day of the experiment according to the TEI 17c NH₃ analyzer operator’s manual.

A Campbell Scientific CR21X micrologger (PC208W software) was used as an automated data acquisition system in conjunction with a laptop computer. The CR21X recorded 15-min averaged measurements for NH₃ concentrations of the sample flow from inside the chamber, lagoon pH, lagoon temperature at 15 cm below the lagoon surface, and meteorological parameters. The 15-min averaged NH₃ concentrations in the chamber were used to calculate NH₃ flux on the basis of the mass balance equation. A temperature-controlled mobile laboratory housed all analytical instrumentation for this study. The mobile laboratory consisted of a modified Ford Aerostar van with a 13,500-BTU air conditioner unit. The temperature inside the van was maintained at approximately 20 °C.

**Barn Emission Measurements**

Barn emissions were measured using an open-path Fourier transform infrared (OP-FTIR) spectroscopy system. NH₃ emissions from barn houses were estimated from average NH₃ concentration measured by OP-FTIR and the rated flow rate for the fan size and setting. Operation of fans was monitored to determine when they were on or off during the entire sampling period. Estimated flow rates from naturally ventilated barns were calculated using wind velocity readings. Meteorological parameters (air temperature, RH, solar radiation, wind speed, and direction), ambient NH₃ concentration at 10 m above the ground, and lagoon parameters (lagoon temperature and lagoon pH) were monitored during the flux measurement periods.

**OP-FTIR Spectroscopy**. OP-FTIR spectroscopy allows the measurement of atmospheric concentrations of NH₃ along variable path lengths depending on the placement of the accompanying IR reflective mirrors. When combined with a second spectrometer and network of reflective mirrors, the technology can produce two-dimensional images of atmospheric concentrations of NH₃ passing through a plane. When combined with the release at a known rate of a tracer gas, the technology can be used to estimate NH₃ flux.

Atmospheric NH₃ concentrations were measured using two monostatic OP-FTIR spectrometers (MIDAC Corp.) with a 0.5-cm⁻¹ wavenumber resolution, each consisting of a Michelson interferometer and mercury-cadmium-telluride (MCT) detector. The spectrometers measure NH₃ by reflecting their infrared (IR) beams off the metal retroreflectors (Optiocon, Corp). The instruments have the potential to monitor a wide range of
compounds simultaneously at very low limits of detection (ppb). Compounds are identified by their fingerprint. In the mid-IR region of the electromagnetic spectrum (4000 to 400 cm\(^{-1}\)), absorption usually occurs at several different wavelengths of IR light, which results in patterns or bands. This pattern of energy absorption, called the absorption spectrum, creates a unique fingerprint for each compound and can be used to identify the chemical. To identify a compound, the pattern of absorption bands in sample absorption spectra (number, location, and shape) is comparable with reference absorption spectra. A reference spectrum is created using known concentrations of the pollutant of interest under controlled conditions of temperature and pressure. Although the pattern of absorption is used for identification of compounds, the intensity of the spectral bands is used for quantification. Within constraints, there is a linear relationship between the intensity of the spectral bands and the concentration of the compound.

A calibration cell is used to check the accuracy of an OP-FTIR spectrometer before and after field use. This specially designed glass calibration cell, 0.98 m in length and 4 in. in diameter, is airtight and holds IR-transmitting windows on both ends. The OP-FTIR spectrometer is placed at one end of the cell, and a retroreflector is placed at the opposite end. Known quantities of NIST-traceable calibration gases (sulfur hexafluoride or ethylene) are injected or flowed through the cell and are measured with the OP-FTIR. For the injections, the gas is injected into the cell using a gas-tight syringe (Dynatech Precision Sampling Co.). A Teflon interior-coated stainless-steel pump is placed at one end of the cell, and a retroreflector is placed at the opposite end. Known quantities of NIST-traceable calibration gases are injected or flowed through the cell and are measured with the OP-FTIR. The instruments were set up with an OP-FTIR spectrometer and a 10-in. telescope directed across an open space of the retroreflector. Signal intensity was achieved in the range from 12,000 to 25,000. After the signal intensity was adjusted, an interferogram was obtained using MIDAC Grams/32 software (Galactic Industries Corp.) and was converted mathematically to a single beam. Detector nonlinearity was evaluated by examining the portion of the single beam spectrum at wave numbers below the detector cutoff (650–680 cm\(^{-1}\)). Measurements from fan-ventilated Moore farm were obtained by placing one OP-FTIR spectrometer in front of the fans across the centerline and another OP-FTIR spectrometer along the non-fan side of the houses at the same height. Using known fan flow rates, the measured NH\(_3\) concentration at the centerline was used to calculate emission from the pig house. Factory-calibrated rates (i.e., published fan curves that relate airflow to static pressure) were adopted. The concentrations that were measured were adjusted for the length of the path across the operating fans at each time point. The emission for each barn was then normalized by the total live mass of the pigs in the houses at the time of the sampling.

To calculate the average nitrogen emission from the naturally ventilated houses at Stokes farm, airflow measurements were made by sampling at one location along each of the four sections of the building on the upwind side while the OP-FTIR was deployed. Each location was sampled for 30–60 sec and the high and low readings were recorded for all four locations over a 5- to 7-min period of time. The high and low wind velocity readings were used to calculate the average wind velocity. The curtain opening for each section was measured and the volume of air per second (ventilation rate) flowing through the upwind side of the barn was calculated as the sum of curtain openings times the average wind velocities for the four sections of the building. The net NH\(_3\) concentrations associated with emissions from the building were obtained by subtracting the upwind readings from the downwind readings using the OP-FTIR and then converting the difference to concentrations of NH\(_3\). A moving average was then applied to the concentration data to reduce the effect of wind variations (times when the wind deviated from the predominant direction). Seven consecutive measurements over 3–4 min were used for the average. Emission rate from the building was obtained by multiplying net NH\(_3\) concentration by the corresponding ventilation rate. Emissions were then normalized by the total live mass of swine in the house.

RESULTS AND DISCUSSION

Seasonal and Diurnal Variations of NH\(_3\) Flux

Averages of lagoon NH\(_3\) fluxes, lagoon temperature, lagoon pH, and TKN are summarized in Table 2. There were significant differences in seasonal NH\(_3\) fluxes from storage lagoons at both farms. During the fall season, average NH\(_3\) fluxes were found to be higher than in the winter season for both farms. Between farms there were similar fluxes for both seasons. For Stokes farm, the average NH\(_3\) flux during the fall season was 2349.4 ± 986.4 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\) and ranged from 1602.1 to 9423.5 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\). The average flux during winter season at Stokes farm was 152.7 ± 52.4 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\), and ranged from 22.6 to 288.6 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\). For Moore farm, average NH\(_3\) flux during the fall season was 1685.4 ± 516.4 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\), and ranged from 712.9 to 3495.6 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\). The average NH\(_3\) flux during the winter season at Stokes farm was 370.5 ± 147.1 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\), and ranged from 49.8 to 672.9 \(\mu g\) N \(\cdot m^{-2} \cdot min^{-1}\).

The measured NH\(_3\) fluxes for Stokes and Moore farms during the experimental periods were plotted against corresponding lagoon temperatures and are shown in Figure 3. The measured NH\(_3\) fluxes are distinctly separated in two clusters, and Figure 3 clearly shows their seasonal difference. For both Moores and Stokes farm, a large range of fluxes at low lagoon temperatures can be observed. This phenomenon is discussed in a later section.

The seasonal variation of lagoon NH\(_3\) flux has also been reported in a previous study from a similar type of swine farm storage and treatment lagoon (Farm 10 located in Sampson County) in eastern North Carolina. According to their estimation, the percentage of total yearly flux attributable to the warm season (summer
months) was approximately 60%. Table 2 also summarizes the results of lagoon NH$_3$ fluxes from Farm 10. In September, during the current study’s experimental period, lagoon temperatures ranged from 21.2 to 33.6 °C. These lagoon temperatures were similar to those observed during the May experimental period of Farm 10 (range of 20.4–35.9 °C with an average of 24.7 ± 3.2 °C). For the winter season, experiments from this study and from the Farm 10 study showed similar lagoon temperature ranges (Table 2). Comparing the ranges of seasonal lagoon NH$_3$ fluxes that resulted from this and the Farm 10 study, under similar lagoon temperature conditions, we found that average NH$_3$ fluxes for the similar lagoon temperature conditions agreed well, but there are also some farm-related differences.

Diurnal variation of NH$_3$ fluxes observed during the experimental periods are shown in Figure 4, which depicts the composite hourly averaged lagoon NH$_3$ fluxes measured at the two conventional farms. It also shows seasonal differences in NH$_3$ fluxes during fall and winter periods. Higher NH$_3$ fluxes with more clear and typical diurnal variations were observed during the warm season (fall). Typical diurnal trends of lagoon NH$_3$ flux showed low NH$_3$ fluxes during the morning hours, which increased with time during the early part of the day as the air and lagoon temperatures increased after sun rise, attaining maximum values around late afternoon, and then decreased during the evening hours. This trend was found to follow approximately the diurnal trends of air and lagoon temperatures at the experimental farm sites. Much lower fluxes with little diurnal variation were observed during the cold season (winter).

Dependence of NH$_3$ Flux on Lagoon Parameters.

It has been suggested that lagoon temperature is the most important parameter regulating the loss of NH$_3$ from swine waste storage lagoons to the atmosphere because of its large variation over the course of the year. Results of the study presented here, showing significantly higher NH$_3$ fluxes from the two waste lagoons during the warm season than those during the cold season, support the above statement. According to the previous study, the pH (range from 7 to 8) and TKN (range from 540 to 727 mg-N·L$^{-1}$) are relatively constant in most North Carolina pig waste lagoons. The lagoon pH, TKN, and surface wind speed for two experimental farms during the warm and cold sampling periods ranged from 7.7 to 8.5, from 487 to 815 mg-N·L$^{-1}$ and approximately 0.7 to 3 msec$^{-1}$, respectively (Table 2). Therefore, lagoon temperature, usually varying with season, could be considered as the most important lagoon parameter to regulate NH$_3$ flux.

NH$_3$ Flux versus Lagoon Temperature. Following Aneja et al., a statistical relationship between NH$_3$ flux and lagoon temperature was examined to obtain an statistical-observational model for the two conventional lagoons. An exponential relationship between NH$_3$ flux and lagoon temperature is expected because the liquid-phase mass transfer coefficient of NH$_3$ in water is exponentially related to water surface temperature in the range from 5 to 30 °C, and also because of the dependence of Henry’s Law on temperature. The logarithm of measured lagoon NH$_3$ fluxes at Stokes and Moore farms during both the

<table>
<thead>
<tr>
<th>Experimental Farms</th>
<th>Season Sampling Periods</th>
<th>NH$_3$ Flux (µg-N·m$^{-2}$·min$^{-1}$)</th>
<th>Lagoon Temperature (°C)</th>
<th>Lagoon pH</th>
<th>TKN (mg-N·L$^{-1}$)</th>
<th>TAN (mg-N·L$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stokes</td>
<td>Fall</td>
<td>2349.4 ± 986.4</td>
<td>26.5 ± 1.3</td>
<td>8.1 ± 0.1</td>
<td>561.3 ± 33.3</td>
<td>442.4 ± 18.1</td>
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<tr>
<td></td>
<td>September 9–20, 2002</td>
<td>(1602.1–19432.5)</td>
<td>(24.7–33.6)</td>
<td>(7.7–8.3)</td>
<td>(498–599)</td>
<td>(410–477)</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>n = 599</td>
<td>n = 599</td>
<td>n = 599</td>
<td>N$^a$ = 9</td>
<td>N$^a$ = 9</td>
</tr>
<tr>
<td></td>
<td>January 6–17, 2003</td>
<td>152.7 ± 52.4</td>
<td>7.2 ± 1.0</td>
<td>8.4 ± 0.1</td>
<td>700.0 ± 18.3</td>
<td>560.0 ± 19.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>n = 865</td>
<td>n = 865</td>
<td>n = 865</td>
<td>N$^a$ = 6</td>
<td>N$^a$ = 5</td>
</tr>
<tr>
<td>Moore</td>
<td>Fall</td>
<td>1685.4 ± 516.4</td>
<td>25.0 ± 1.6</td>
<td>8.2 ± 0.1</td>
<td>582.5 ± 135.6</td>
<td>363.5 ± 35.9</td>
</tr>
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<td></td>
<td>September 30 to October 11, 2002</td>
<td>(712.9–34956.5)</td>
<td>(21.2–30.0)</td>
<td>(7.9–8.5)</td>
<td>(487–774)</td>
<td>(316–409)</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>370.5 ± 147.1</td>
<td>7.2 ± 2.5</td>
<td>8.1 ± 0.1</td>
<td>782.0 ± 38.9</td>
<td>635.5 ± 36.9</td>
</tr>
<tr>
<td></td>
<td>January 27 to February 2, 2003</td>
<td>(49.8–672.8)</td>
<td>(1.7–12.0)</td>
<td>(7.9–8.9)</td>
<td>(880–815)</td>
<td>(645–665)</td>
</tr>
<tr>
<td>Farm 10</td>
<td>Summer</td>
<td>4017 ± 987</td>
<td>30 ± 3.3</td>
<td>7.5 ± 0.2</td>
<td>648.1 ± 27.7</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td>December 1–17, 1997</td>
<td>(369–1913)</td>
<td>(8.4–15.3)</td>
<td>(7.9–8.1)</td>
<td>(599–715)</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>305 ± 154</td>
<td>12.1 ± 2.1</td>
<td>7.8 ± 0.1</td>
<td>641.7 ± 39.0</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td>February 1–26, 1998</td>
<td>(90–695)</td>
<td>(8.8–15.1)</td>
<td>(7.7–8.0)</td>
<td>(580–727)</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td>Spring</td>
<td>1706 ± 552</td>
<td>24.7 ± 3.2</td>
<td>7.7 ± 0.1</td>
<td>603.3 ± 48.2</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td>May 16–27, 1998</td>
<td>(851–3594)</td>
<td>(20.4–35.9)</td>
<td>(7.6–7.8)</td>
<td>(540–720)</td>
<td>N/A</td>
</tr>
</tbody>
</table>

Notes: Numbers in parentheses are ranges of the data. n represents the number of data in 15-min average during the sampling periods. N represents the total number of lagoon waste samples for TKN. A lagoon waste sample was collected daily between 12:00 and 13:00 Eastern Standard Time. The water sampling days were: 1) September 10–18, 2002; 2) January 7–10, 14, 15, 2003; 3) October 1–9, 2002; 4) January 27–31, and February 1–5, 2003. 5) Farm 10 (another commercial LST) values are provided for comparison. N/A = not applicable.
experimental periods is plotted against the corresponding lagoon temperatures in Figure 5. Data points \( (n = 2914) \) represent 15-min averages. Lagoon NH\(_3\) flux is found to increase exponentially with lagoon temperature. The best-fitted relationship with \( R^2 = 0.82 \) and \( p < 0.0001 \) is given by

\[
\log_{10}(N) = 0.051T_1 + 1.943 \tag{2}
\]

where \( N \) denotes the NH\(_3\)-N flux expressed in units of \( \mu g\cdot N \cdot m^{-2} \cdot min^{-1} \), and \( T_1 \) is lagoon temperature at \( \sim 15 \) cm below water surface (°C). The slope and intercept of our best-fitted regression line in Figure 5, or the coefficients in eq 2 are comparable with those (slope = 0.048; intercept = 2.1; \( R^2 = 0.78 \)) obtained by Aneja et al.\(^{22}\) from intensive measurements made over a period of 1 yr on Farm 10 in eastern North Carolina. These two empirical models show very good agreement with almost identical fluxes.

A combined effect of chemical and physical processes occurring within a waste storage lagoon was proposed to explain the high NH\(_3\) flux during the warm months.\(^{23}\) The waste sludge at the bottom of the lagoon is chemically broken down and acts as a source of NH\(_3\), and this process accelerates with increase of temperature because of increased bacteria activity in warmer temperatures. The NH\(_3\) produced by the chemical decomposition at the bottom of the lagoon diffuses upward and replaces the volatilized NH\(_3\) from the surface of the lagoon. In the absence of any mechanical or turbulent mixing of the lagoon, buoyancy, molecular diffusion, and mass transfer processes are largely responsible for the transport of NH\(_3\) from the bottom to the upper layers of the lagoon.\(^{23}\) Because higher lagoon surface temperature increases the transfer rate of NH\(_3\) across the liquid-gas interface, higher lagoon surface temperatures during the warm period, coupled with a readily available source of NH\(_3\), are proposed to be responsible for elevated fluxes during the warm season.

**NH\(_3\) Flux and Lagoon pH.** Table 2 shows that the effluent pH continuously measured adjacent to the dynamic chamber system ranged from 7.7 to 8.3 (with an average value 8.1 ± 0.1) during the fall period, and from 8.1 to 8.5 (average 8.4 ± 0.1) during the winter experiment at Stokes Farm.
farm. At Moore farm, it ranged from 7.9 to 8.5 (average 8.2 \pm 0.1) during the fall period and from 7.9 to 8.3 (average 8.1 \pm 0.1) during the winter experiment. Each experimental day, a lagoon waste sample was also taken adjacent to the floating chamber and analyzed for water chemistry (pH and TKN). The daily lagoon pH values ranged from 7.6 to 8.5 during the two experimental periods at both Stokes and Moore farms, and they were plotted against the corresponding lagoon NH\textsubscript{3} fluxes in Figure 6. Relationship between pH and lagoon NH\textsubscript{3} flux revealed no statistically significant correlation between pH and the corresponding flux in Figure 6.

**Figure 4.** Diurnal pattern of NH\textsubscript{3} flux at the Stokes and Moore farms for warm and cold season measurements. Each data point consists of an hourly averaged NH\textsubscript{3} flux (\(\mu g-N-NH_3/m^2/min\)).

**Figure 5.** Log-linear relationship between NH\textsubscript{3} flux (\(\mu g-N-NH_3/m^2/min\)) and the corresponding lagoon temperature (°C) measurement at two conventional farms during the experimental periods. Each data point represents a 15-min average of NH\textsubscript{3} flux.

**Figure 6.** Relationship between pH and lagoon NH\textsubscript{3} flux.
Modeling studies\textsuperscript{23,24} found a positive correlation between lagoon pH and NH$_3$ flux. However, waste lagoon slurry has a high buffer capacity that confines lagoon pH to a relatively narrow range.\textsuperscript{10,22,35}

\textit{NH}_3 \textit{Flux, TKN, and Total Ammoniacal Nitrogen.} TKN includes the sum of nitrogen in NH$_4^+$, organic nitrogen, and oxidized nitrogen in the effluent. Total ammoniacal nitrogen (TAN) is the sum of NH$_3$ and NH$_4^+$.

The amount of TKN and TAN in a conventional pig waste lagoon depends on such factors as animal population and type, animal mass, animal feeding rate, and nitrogen content. In a waste treatment lagoon operating at steady-state conditions, the amount of organic nitrogen within the waste lagoon does not vary significantly.\textsuperscript{22}

A balance exists between the inflow and outflow of nitrogen from the traditional waste treatment lagoon. Table 2 summarizes the average TKN and TAN concentrations at the experimental conventional farm sites during the measurement campaigns. Figure 7 shows a plot of the daily averaged lagoon NH$_3$ flux during the experimental periods (fall and winter) against the TKN of lagoon waste samples collected on the same day from the two experimental farms. Some seasonal differences in TKN concentrations were observed (Figure 7 and Table 2); relatively lower TKN concentrations ranging from approximately 487 to 774 mg-N L$^{-1}$ occurred during the fall period than during the winter period (~681–815 mg-N L$^{-1}$) at both farms. This can be explained by other seasonally dependent characteristics such as lagoon temperature.
Table 1, the nitrogen excretion rates at the farms during the experimental periods were estimated by using mass balance of nitrogen at the farm facility with the farm animal and feeding information (i.e., number of animals, animal mass, feed consumption, nitrogen content in feed). No significant seasonal variations in the estimated nitrogen excretion were found during the experimental periods. Therefore the TKN concentration in lagoon waste might be expected to remain relatively constant over the year. However, increased nitrogen compound losses from the water surface because of high lagoon temperatures (21–34°C) could reduce the TKN concentration during warm periods. In addition, higher precipitation during the preceding spring and summer seasons could dilute the TKN concentration in lagoon waste volume.

TKN concentrations for the fall and winter periods at Stokes farm ranged from 498 to 612 mg-N · L⁻¹ (average 561.3 ± 33.3 mg-N · L⁻¹), and from 681 to 723 mg-N · L⁻¹ (average 700 ± 18.3 mg-N · L⁻¹), respectively. TKN concentrations for the fall and winter periods at Moore farm ranged from 444 to 524 mg-N · L⁻¹ (average 493.3 ± 27.7 mg-N · L⁻¹), and from 680 to 815 mg-N · L⁻¹ (average 782 ± 38.9 mg-N · L⁻¹), respectively.

The correlation between NH₃ flux measurements and effluent TKN concentrations was examined within the individual seasons, because significantly different ranges of TKN concentrations were found. For each season, a positive linear correlation between NH₃ flux and TKN concentration was observed to be statistically significant; and a stronger correlation occurred within the winter period (R² = 0.44, p = 0.005) than during the fall (R² = 0.20, p = 0.01). These linear regression relations are given as

\[ N = 4.49 \text{TKN-333.08, for fall period} \]  \hspace{1cm} (3)
\[ N = 1.67 \text{TKN-995.39, for winter period} \]  \hspace{1cm} (4)

where \( N \) denotes the NH₃-N flux in units of μg-N · m⁻² · min⁻¹.

For all data including both seasons, a much weaker negative correlation is apparent. This may be only a reflection of the much stronger correlation between NH₃-N flux and lagoon temperature.

Figure 8 shows a plot of the daily averaged lagoon NH₃ flux during the experimental periods (fall and winter) against the TAN of lagoon waste samples collected on the same day for the two experimental farms. Seasonal differences in TAN concentrations were observed (Figure 8 and Table 2); with relatively lower TAN concentrations ranging from approximately 316 to 477 mg-N · L⁻¹ occurring during the fall period than those during the winter period (~545–665 mg-N · L⁻¹) at both farms. As discussed for TKN, there were no significant seasonal variations in the estimated nitrogen excretion during the experimental periods. As with TKN, it is hypothesized that higher lagoon temperatures in the fall lead to increased NH₃ loss from the effluent. TAN concentrations for fall and winter experimental periods at Stokes farm ranged from 410 to 477 mg-N · L⁻¹ (average 442.4 ± 18.1 mg-N · L⁻¹) and from 545 to 593 mg-N · L⁻¹ (average 560 ± 19.4 mg-N · L⁻¹), respectively. TAN concentrations for fall and winter periods at Moore farm ranged from 316 to 409 mg-N · L⁻¹ (average 363.5 ± 35.9 mg-N · L⁻¹) and from 545 to 665 mg-N · L⁻¹ (average 635.5 ± 36.9 mg-N · L⁻¹), respectively. TAN concentrations averaged over all effluent samples were found to be 76.2 ± 11.5% of TKN concentrations. Correlation between the TAN concentrations and the NH₃ flux measurements were examined within the different seasons. Positive linear correlation between NH₃ flux and TAN concentration were found for both seasons, although unlike TKN, a stronger correlation was present during the fall period (R² = 0.29, \( p < 0.01 \)) than during the winter (R² = 0.24, \( p < 0.01 \)). These linear
regressions are given as

\[ N = 6.00 \text{TAN-443.9, for fall period} \]  \hspace{1cm} (5) \]
\[ N = 1.44 \text{TAN-624.0, for winter period} \] \hspace{1cm} (6)

where \( N \) denotes the \( \text{NH}_3 \)-N flux in units of \( \mu \text{g-N} \cdot \text{m}^{-2} \cdot \text{min}^{-1} \).

Again for the entire data covering both seasons, a much weaker negative correlation is apparent.

\textit{NH}_3 \textit{ Flux and Environmental Parameters}. The influence of atmospheric environmental (meteorological) parameters, on lagoon \( \text{NH}_3 \) flux was investigated. A second parameter was found to have a significant correlation. This was the environmental parameter known as \( \Delta T \), or the “hot-air” variable, the difference between air and lagoon temperatures (\( \Delta T = T_a - T_l \)). \( \Delta T \) was found to be the second most important parameter influencing the \( \text{NH}_3 \) flux, especially when \( \Delta T > 0 \). Therefore, 15-min averages of \( \log(\text{NH}_3 \text{-N flux}) \) and the corresponding lagoon temperature and air temperatures were statistically analyzed using multiple linear regression. To also account for the farm-related differences in lagoon areas and animal populations, the \( \text{NH}_3 \) flux data used for the multiple linear regression analysis were normalized by animal live mass in metric tons (1000 kg) and adjusted for the lagoon surface area of each conventional farm. We denote this normalized \( \text{NH}_3 \) emission rate by \( F = A \times \text{flux}/1000 \text{ kg} - \text{lm} \), where \( A \) is lagoon surface area at each conventional farm, and \( \text{lm} \) is live animal mass.

Figure 9 shows a plot of \( \log_{10}(A \times \text{flux}/\text{tons}) \) versus the difference (\( \Delta T = T_a - T_l \)) between the air temperature and the lagoon temperature for different ranges of lagoon temperatures. For reference, a vertical line is plotted at \( \Delta T = 0 \), the point at which air and lagoon temperatures are equal. To the left of the \( \Delta T = 0 \) line in Figure 9, the four color categories appear in horizontal bands, with higher emissions corresponding to higher lagoon temperatures (red, cyan) and lower emissions to lower lagoon temperatures (blue, black). This is the expected effect; that is, \( \log_{10} F \) is higher for higher lagoon temperature. This indicates that when \( \Delta T < 0 \) (air temperature < lagoon temperature), \( \log_{10} F \) is determined largely by lagoon temperature (i.e., air temperature has little to no effect on \( F \)). However, the characteristics of the plot to the right of the line \( \Delta T = 0 \) (air temperature > lagoon temperature) are strikingly different for two of the four regimes. Note the hockey-stick-shaped pattern to the plotted points for the lower lagoon temperatures (blue, black). The blue and black points lie in roughly horizontal bands to the left of \( \Delta T = 0 \) (the shafts of the hockey sticks); but at 0 or some small positive \( \Delta T \) the horizontal bands give way to downward trends (the blades of the hockey sticks); that is, \( \log_{10} F \) decreases approximately linearly with increasing \( \Delta T \) for \( \Delta T > 0 \). The statistical significance of the hockey-stick shape (assuming the hinge occurs at \( \Delta T = 0 \)) was assessed by fitting two statistical regression models to the data including the hot-air term \( \Delta T^* \) (\( \Delta T > 0 \)). In one analysis, other variables included in the model were lagoon temperature and a farm indicator; in the second analysis the variables lagoon temperature, farm indicator, season indicator, and their interactions were included. In both analyses, the hot-air variable \( \Delta T^* \) (\( \Delta T > 0 \)) was highly significant (\( p < 0.0001 \) assuming autoregressive errors). It is important to note that both analyses included lagoon temperature in the models, so that the statistical significance of the hot-air variable is not an artifact of very low absolute lagoon temperatures in the lower lagoon temperature regimes (blue, black).

The absence of the hockey-stick shape for the higher lagoon temperatures (red, cyan) are a result of small \( \Delta T \) values for these datasets. For these datasets, \( \Delta T \) is not large enough to determine the presence or absence of the hot-air effect.

In summary, at least for the lower lagoon temperatures (blue, black), if the air temperature is less than the lagoon temperature, then \( \log_{10} F \) is solely dependent on lagoon temperature (independent of air temperature); however, when the air temperature exceeds the lagoon temperature (\( \Delta T > 0 \)), \( \log_{10} F \) decreases by an amount approximately proportional to the amount by which the air temperature exceeds the lagoon temperature (in addition to its dependence on lagoon temperature).

The statistical evidence of the hockey-stick shape for the lower lagoon temperature regimes (blue, black) is substantial. Although the observed hot-air effect was unexpected, it is not without a plausible scientific explanation. The air-lagoon temperature difference, \( \Delta T = (T_a - T_l) \), may be considered as a simple measure of near-surface atmospheric stability, which is found to strongly influence surface-to-air turbulent exchanges, including that of \( \text{NH}_3 \) from a lagoon surface. When \( \Delta T < 0 \), unstable conditions enhance turbulence and enhance fluxes. On the other hand, stable or inversion conditions with \( \Delta T > 0 \) represent weaker turbulence and smaller fluxes. The air-surface temperature difference \( (T_a - T_l) \) is the simplest measure of the stability of the atmospheric surface layer, for which a variety of parameters are used in micrometeorology and air pollution meteorology.\(^{29,36}\) Better measures
include wind shear or wind speed, such as the bulk Richardson number \( R_{ib} = (g \ z_i / T_{0i})(\Delta T + 0.01z_i)/U^2 \), where \( z_i \) of 10 m is the reference height for wind speed \( U \) and air temperature measurements, and \( T_{0i} \) is the surface temperature in absolute units. Thus, \( R_{ib} \) depends on both \( \Delta T \) and \( U \), but in a different manner. Very unstable and convective conditions are characterized by large \( -\Delta T \) and weak winds, near-neutral conditions by small \( |\Delta T| \) and strong winds, and very stable conditions by large \( \Delta T \) and weak winds.

According to the well-known bulk-transfer relations used for estimating surface fluxes, the \( \text{NH}_3 \) flux from a lagoon surface can be expressed as a product of the wind speed at a reference height (e.g., 10 m), the difference in \( \text{NH}_3 \) concentrations at the surface and the reference height, and a dimensionless mass transfer coefficient, \( C_M^* \). Judging from the behavior of the transfer coefficients, for heat and water vapor \( C_M^* \) is expected to depend on atmospheric stability, monotonically decreasing with increasing stability \( R_{ib} \). In particular, \( C_M^* \) decreases rapidly with increasing \( R_{ib} \) for \( R_{ib} > 0 \). This explains the stronger dependence of \( \text{NH}_3 \) flux on \( \Delta T \) for \( \Delta T > 0 \), as shown in Figure 9. The weaker or lack of dependence of flux on \( \Delta T \) for neutral and unstable conditions \( \Delta T < 0 \) can be explained by the opposing effects of wind speed and \( C_M^* \) on flux. Note that, under unstable conditions, as wind speed decreases, \( C_M^* \) is expected to increase.

The dependent variable, \( \log_{10} F \), was found to be mainly dependent on lagoon temperature \( (T_i) \) and the hot-air variable \( D \). A statistical model was created using these variables. The hot-air variable \( D \) was equal to 0 if the lagoon was warmer than the air; and equal to \( \Delta T \) when the air was hotter than the lagoon.

The model also incorporated a farm indicator variable \( (F_i) \) to account for differences in responses to lagoon temperature and \( \Delta T \) between farms. For the purposes of the multiple regression analysis, we set \( F_i \) equal to zero for Stokes farm and \( F_i \) equal to 1 for Moore farm. The statistical analysis also accounted for autocorrelation because most of the data points were in consecutive 15-min intervals and not taken as random samples.

The multiple regression analysis showed a high correlation \( (R^2 = 0.95) \) between \( \text{NH}_3 \) flux and the regressors, which were lagoon temperature \( (T_i) \), hot-air variable \( (D) \), and the farm indicator variable \( (F_i) \). The overall response of \( \text{NH}_3 \) flux between farms differed and was statistically significant \( (t = 646, p < 0.0001) \) as indicated by the farm variable. The multiple regression also shows statistically significant relationships between \( \text{NH}_3 \) flux and lagoon temperature \( (t = 166, p < 0.0001) \), and between \( \text{NH}_3 \) flux and \( D \) \( (t = 55, p < 0.0001) \). The multiple regression equation for lagoon emissions at Stokes \( (F_i = 0) \) and Moore \( (F_i = 1) \) farms is given below.

\[
\log_{10} F = 3.6264 + 0.4782 F_i + 0.0449 T_i - 0.05946 D \quad (7)
\]

Thus, the multiple regression equation for the average lagoon emissions at the two conventional farms is given by

\[
\log_{10} F = 3.8655 + 0.0449 T_i - 0.05946 D \quad (8)
\]

in which both \( T_i \) and \( D \) are expressed in units of °C, and \( F \) in units of kg-N ⋅ min \(^{-1} \) (1000 kg ⋅ l m) \(^{-1} \).

To statistically analyze the possible influence of lagoon pH, a multiple regression analysis was conducted on all data representing 15-min averaged \( F \) or \( \log_{10} F \) and the corresponding 15-min averaged values of lagoon temperature, the hot-air variable \( D \), and lagoon pH at both farms during the four experimental periods. The dependent variable, \( \log_{10} F \), was still found to be mainly dependent on lagoon temperature \( (T_i) \) \( (p < 0.0001) \) and the hot-air variable \( D \) \( (p < 0.0001) \), but its correlation with pH was found to be statistically insignificant \( (p < 0.1098) \). The statistical analysis also accounted for autocorrelation because most of the data points were in consecutive 15-min intervals and not collected as random samples.

**Emissions from Animal Houses**

At the Stokes farm, barn emissions were measured in the winter season with the average \( \text{NH}_3 \) emissions found to be 0.25 kg-N/week/1000 kg ⋅ l m. At Moore farm, two measurement campaigns were performed. In the warm season the emissions were slightly higher than in the cold season. Average emissions were 1.05 and 0.89 kg-N/week/1000 kg ⋅ l m, for warm season and cold season, respectively. Table 3 shows the normalized emissions and compares them to other published studies. It is noted that the natural ventilation conditions at Stokes farm presented additional environmental variables and measurement challenges (wind speed, direction, etc.) as compared with the controlled ventilation system conditions at the Moore farm. However, it can be seen that the emissions from this study are well within the range of emissions from other studies.

**CONCLUSIONS**

\( \text{NH}_3 \) flux measurements during two different seasons (warm and cool) were conducted over pig waste treatment weitere.
lagoons and animal houses at two conventional swine farms in North Carolina. The fall and winter intensive measurements were conducted from September 9 to October 11, 2002 and January 6 to February 2, 2003, respectively. The average lagoon NH₃ fluxes for the two conventional farms during the fall and winter months were 2017 ± 751 and 262 ± 100 μg-N · m⁻² · min⁻¹, respectively. These averages were comparable to those found in the previous study from a typical swine farm (Farm 10) in North Carolina.²²

Typical diurnal variation of lagoon NH₃ flux was observed during the measurement periods. The NH₃ flux increased exponentially with increasing lagoon temperature, and the best-fitted regression relationship between the two is: \( \log_{10}(N) = 0.05171 + 1.943 (R^2 = 0.82 \text{ and } p < 0.0001) \), where \( N \) represents the NH₃-N flux in μg · m⁻² · min⁻¹ and \( T_1 \) is the lagoon temperature in °C. This regression relationship showed very good agreement with that of Aneja et al.²² based on Farm 10 flux measurements.

Relationships between lagoon NH₃ flux and chemical parameters such as pH, TKN, and TAN of lagoon waste were also examined. The lagoon pH stayed in a relatively narrow range from 7.7 to 8.5 during fall and winter seasons. No significant correlation between pH and lagoon NH₃ flux was observed during the experimental periods at either farm. Concentration of TKN and TAN varied significantly with season; lower TKN concentrations (444–612 mg-N) were observed during the winter periods. Increase of NH₃ loss from the water surface because of high lagoon temperature could reduce the TKN concentration during warm periods (i.e., during the fall months). Within both seasons, positive linear correlations between NH₃ flux, TKN, and TAN concentration were observed; these were statistically significant, with a stronger correlation during the winter season for TKN, and a stronger correlation during the fall season for TAN.

The influence of atmospheric environmental parameters on lagoon NH₃ flux was investigated. NH₃ flux was significantly correlated with lagoon temperature and the difference between air and lagoon temperature \( \Delta T = T_a - T_1 \). \( \Delta T \) is considered a measure of near-surface atmospheric stability. The multiple regression equation for the average lagoon emissions at the two conventional farms is

\[
\log_{10}F = 3.8655 + 0.0449T_1 - 0.05946D \quad (9)
\]

in which both \( T_1 \) and \( D \) are expressed in units of °C, and \( F \) in units of kg-N · min⁻¹ (1000 kg · lm⁻¹). Barn emissions were measured for one season at Stokes farm (naturally ventilated) and two seasons at Moore farm (mechanically ventilated). Emissions normalized by live animal mass were found to be comparable to other studies.

The statistical-observational model developed and described herein and the barn emissions protocol is proposed as a valid and objective approach to be used to compare the emissions from potential ESTs to evaluate the effectiveness of such technologies.

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