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A process-based approach for ammonia emission measurements at a free-stall dairy

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Abstract. A protocol using flux chambers was employed to determine ammonia emission rates from different low level area sources (LLAS) including free stalls, open lots, manure composting areas, lagoons and separated solids in a central Texas dairy. Data including ammonia emissions from these sources were collected for summer and winter seasons of 2003. Ammonia concentration measurements were made using chemiluminescence-based analyzers. The estimated emission rates for the facility were $24.7 \pm 25.4 \text{ kg.day}^{-1}$ for winter and $63.1 \pm 31.1 \text{ kg.day}^{-1}$ for summer. This difference was due to temperature, loading rate of dairy waste, and bacterial activity of LLAS. The uncertainty analysis showed that 9.4% of ammonia sampling uncertainty was attributed to ammonia sensors, calibration gas impurity and air flow controllers. In winter, the compost and the free-stall contributed about 77% to the total emission rates for the facility. But in summer, 65% of overall ammonia emissions were contributed by two lagoons at the dairy. These results suggest that seasonally dependent best management practices may be needed to reduce annual average ammonia emissions from free stall dairies.

Keywords. air quality, ammonia, dairy, emission rate, flux chamber

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Introduction

Ammonia (NH_3) emissions to the atmosphere are environmentally important, not only because of their role in the chemistry of air pollutants present in the atmosphere, but also because of undesirable ecological effects as N compounds are subsequently deposited from the atmosphere back to land (Phillips et al., 2000). When combined with nitric acid, ammonia forms aerosol nitrate, which contributes significantly to total particulate matter (PM). The role that ammonia plays in neutralizing acidic aerosols has led to many studies concerning health effects of atmospheric ammonia. Asman and Janssen (1987) and Brost et al. (1988) described the results of this acid-ammonia combination as following:

1. Formation of ammonium nitrate (NH_4NO_3) by reacting with nitric acid (HNO_3).
2. Formation of either ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) or ammonium bisulfate by neutralization of sulfuric acid (H_2SO_4).
3. Formation of ammonium chloride (NH_4Cl) by reaction with hydrochloric acid.

In both Europe and United States, the largest sources of ammonia emission are livestock and poultry operations and they account for an estimated 70-90% of total emissions. Cattle including dairy cows are the largest of livestock sources contributing to ammonia emissions (Battye et al., 1994; USEPA, 2000; Pain et al., 1998; Hutchings et al., 2001).

Emission flux is used to determine emissions from the agricultural feeding operations (AFOs). Additionally, flux is the source term in many dispersion models that estimate ambient concentrations of contaminants (Dupont, 1987).

Flux chambers may be used to measure gaseous emissions, especially ammonia, from the low level areas (LLAs). These sources may include lagoons, compost piles, manure storages, open lots and animal buildings. A majority of ammonia emissions come from AFOs, so more research should be conducted to improve methods for accurately measuring emission rates of ammonia from low level area sources (LLAS).

The quantification of ammonia emissions from low level area sources in a dairy is needed to understand which sources contribute the highest to the overall ammonia emissions during winter and summer conditions. Ammonia emission concentrations are converted to emission rates to develop source specific ammonia emission control strategies. The objective of this study was to use a flux chamber protocol that provides direct estimation of ammonia emission fluxes and rates from low level area sources (LLAS) in a central Texas dairy.

Materials and Methods

A free-stall dairy in central Texas was chosen to develop a process-based approach for an ammonia emission measurement protocol. An aerial photo of the site showing LLAS at the dairy is presented in Figure 1.

A total of 1840 lactating cows and 250 dry cows were housed at the dairy during this study. Cows were housed on the dairy in open lots and free stalls. The open lot included centralized feeding and watering areas and free standing shelters for relief from severe weather conditions.

Accumulated manure in the free-stall barns was removed by flushing four times a day (7am, 1pm, 7pm, 1am). There were three free-stall barns located on this dairy and each free stall was flushed in series from north to south. The slurry was then transported into a solids separator system for liquid-solid separation. The separated liquid portion was transported to the first cell (lagoon 1) of the anaerobic lagoon. The effluent from lagoon 1 was conveyed to lagoon 2 with a

pipe outlet. Lagoon 2 also accepted runoff from two other open-lots. Open-lots housed lower milk producing cows or non-lactating cows. Each open-lot was an un-paved, confined area with access to feed bunkers and water tanks. Manure produced in the open-lot was removed by scraping using tractor mounted blades. The rate of manure production was generally higher near feed bunkers and water tanks. The scraped manure was stockpiled and either land applied or composted on-site. The composted separated solids were used as bedding for the free stall barns.

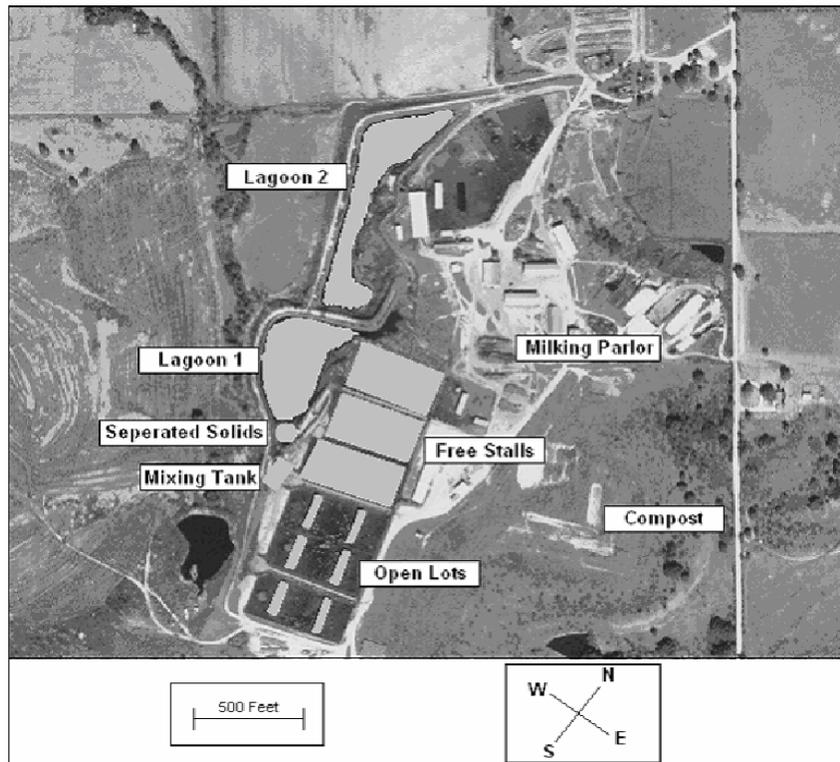


Figure 1. An aerial view of the dairy facilities.

The surface areas of the free stalls, open lots, separated solids, lagoon one, lagoon two, and compost site were measured to estimate ammonia emission rates for LLAS.

Isolation Flux Chamber Sampling Protocol

Isolation flux chambers have been used to measure emission fluxes of volatile organic compounds (VOCs) and inorganic gaseous pollutants from a wide variety of sources (Eklund, 1992). The basic design of the flux chamber includes a hemispherical top and a cylindrical skirt. Odotech Incorporated supplied the hemispherical top for use in this research (Odotech Inc. Montreal, Canada). The custom-fabricated stainless steel skirt and the top were joined by a set of wing nuts and sealed using a gasket (Figure 2). The dome contained four symmetrical holes with stainless steel fittings. A tubing inlet located at one of the stainless steel fittings allowed for the flow of sweep air into the chamber. A fitting on the top of the hemisphere allowed for the pollutant stream to be conveyed to a measurement device. Two of these holes were used to connect the flux chamber to Teflon[®] and low density polyethylene (LDPE) tubing used to move

the sweep air (contaminant free zero grade air) and sampling air (polluted air from flux chamber) to and from the flux chamber for purging and sampling, respectively (Mukhtar et. al, 2003).

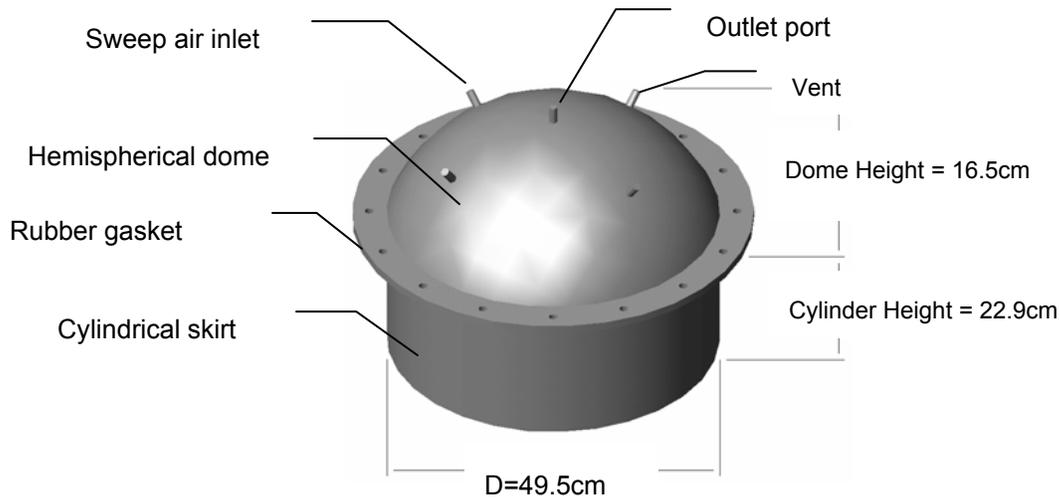


Figure 2. Schematic of isolation flux chamber.

Given the dimensions in figure 2, the incoming sweep air flow rate of 5 L/min for winter sampling and 7L/min for summer sampling, it required approximately 30 minutes to achieve 3.5 residence times (Equation 1) for purging the chamber followed by a 30-min ammonia sampling period.

$$t = R_t \cdot \frac{v}{Q} = \frac{3.5 \cdot \left(\frac{\pi d^2 \cdot h}{4} + \frac{\pi d^3}{12} \right)}{7 \frac{l}{min}} = 32.25 \text{ min} \dots \dots \dots \text{eq.(1)}$$

where:

t = residence times ($R_t=3.5$) in chamber.

v = volume of chamber (cm^3)

d = chamber diameter (cm)

h = chamber height (cm)

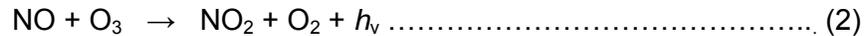
Q = flow rate into chamber (L/min)

Sweep air with zero hydrocarbons was introduced into the chamber. Air sample was drawn from the chamber at a flow rate of 2 L/min. Both flows were maintained using mass flow controllers (MFC) at standard conditions (25 °C, 1 atm and 0% RH).

Measurement of Ammonia Concentrations

Two chemiluminescence-based analyzers (Model 17C, Thermo Environmental Instruments, TEI, Massachusetts) were used to measure ammonia for real time sampling procedure. The

operating principle of the analyzer has been reported by Mukhtar et al. (2003). The principle of chemiluminescence is based on the reaction of nitric oxide (NO) with ozone (O₃) which is given in equation (2).



An air sample is drawn into the analyzer by an external vacuum pump. This air sample mixes with ozone (O₃) which is generated by an internal ozone creator in a reactor chamber of the analyzer. This reaction produces nitrogen dioxide (NO₂), oxygen (O₂) and other nitrogen compounds, in equations (3) and (4), reactions occur in reaction chamber as follows:



Ammonia concentrations are determined by subtracting NO_x signals from the N_t signal in either parts per million (ppm) or parts per billion (ppb).

The TEI analyzers were calibrated using known concentrations of ammonia (NH₃) and nitric oxide (NO), certified standard gases guaranteed by the manufacturer (Praxair, Inc., Danbury, CT) to be within ± 2% accuracy. Each cylinder was connected to a mass flow controller (MFC) (Aalborg, Inc. Orangeburg, New York). All MFCs were calibrated by manufacturer. Mass flow controllers were necessary to regulate the amount of air supplied to the static mixer box and to the analyzer. In this calibration process mixture inflow rate of 2 L/min to the analyzer was maintained. The remaining air mixture was vented to the atmosphere.

On-site measurements, during the winter and summer of 2003, were conducted by using a mobile laboratory. The mobile laboratory included ammonia analyzers, air flow mixing devices, a multiplexer system including MFCs, a zero air generator (Model 737-12, AADCO Instruments, Village of Cleaves OH), gas cylinders and power generator for electricity.

For the winter data, only one TEI was used. Thus, it required at least 65 minutes to generate a flux reading. In the summer of 2003, several notable improvement were made to increase number of samples taken and improve data logging efficiency using new multiplex controls, a modified LabVIEW program (National Instruments, Austin, TX) for data logging and a zero air generator for sweep air and for purging the TEI analyzers. Six chambers and the multiplexer allowed increased sample collection efficiency by minimizing the chamber set-up time between samples. More importantly, the chambers were lowered pneumatically on the area source only during sampling protocol limiting potential gas accumulation if the chamber sat on an emitting surface for longer periods. A detailed description of these improvements is described by Boriack et al. (2004a).

Ammonia Flux and Emission Rate Calculations

Measured ammonia concentrations were converted into mass concentration (C_{mass}) using equation (5). The mass concentration was converted into emission flux using equation (6). The volumetric flow rates used were 5 L/min for winter and 7 L/min for summer sampling and the “foot print” area of the chamber was 0.192m². To estimate the emission rates, mass concentrations and emission fluxes values must be known. Emission rates were calculated for each individual LLAS using equation (7).

$$C_{mass} = 1000 \times \left(\frac{P}{RT} \right) \times C_{ppm} \times MW_p \dots\dots\dots (5)$$

where:

- C_{mass} = mass concentration ($\mu\text{g}/\text{m}^3$)
- R = universal gas constant (0.08206 atm-L/gmol-K)
- T = absolute temperature (K)
- P = pressure (atm)
- C_{ppm} = corrected volumetric NH_3 concentration (ppm)
- MW_p = molecular weight of pollutant ($\text{NH}_3 = 17.03 \text{ g/mol}$)

Once the concentration in mass per volume is determined, equations 6 and 7 are used to calculate ammonia flux and rate, respectively:

$$EFl_{\text{NH}_3} = \frac{C_{mass} \times V_{fc}}{A_{FC}} \dots\dots\dots (6)$$

where

- EFl_{NH_3} = NH_3 gas emission flux ($\mu\text{g}/\text{m}^2\text{-s}$)
- V_{fc} = volumetric flow through the flux chamber (m^3/s)
- A_{FC} = area of flux chamber (“footprint”, m^2)

$$ER = EFl \times A_{sc} \dots\dots\dots (7)$$

where:

- ER = Emission rate, $\mu\text{g}/\text{s}$.
- A_{sc} = Area of source (LLAS), m^2 .

Uncertainty Analysis

Uncertainty analysis was performed on ammonia sampling process. The first order Taylor series technique was used by Boriack et al. (2004b) to calculate uncertainty of this sampling procedure. The uncertainty analysis was limited to three major components of the ammonia sampling system including the analyzer, analog inputs and outputs, calibration gases and MFC. The uncertainty was found to be 9.4% for a 40 ppm ammonia stream analyzed with a sensor (TEI) set to a 0-50ppm span.

Results and Discussion

Two studies were conducted in 2003 at the same free stall dairy. First study was in the winter of 2003. Twenty nine samples were collected using isolation flux chamber method to determine the emission rates of ammonia from different low level area sources (LLAS) of dairy. Data from NH_3 concentrations (in ppm) have been corrected for NH_3 adsorption through the flux chambers. The procedure for accounting for adsorption losses has already been described

(Capareda, et al., 2004). Results of ammonia concentration and calculated emission rates of winter season are shown in Table 1.

Table 1. Ammonia concentrations and emission rates for 2003-winter.

| LLAS | Number of Samples | Concentration (ppm) | Concentration ($\mu\text{g}/\text{m}^3$) | E. Flux ($\mu\text{g}/\text{m}^2/\text{s}$) | Area (m^2) | ER (kg/day) | LLAS Temp ($^{\circ}\text{C}$) | Ambient Temp ($^{\circ}\text{C}$) |
|---|-------------------|------------------------------|--|---|-----------------------|---|----------------------------------|-------------------------------------|
| Compost | 3 | 17.4 \pm 23.5 ^b | 12120 | 5.3 \pm 7.1 ^b | 21000 | 9.5 \pm 12.9 ^b | 30.1 | 8.5 |
| Free Stall | 5 | 36.4 \pm 23.3 | 25354 | 11.0 \pm 7.0 | 9790 | 9.3 \pm 6.0 | 6.4 | 6.3 |
| Dry Open Lot | 3 | 6.5 \pm 8.8 | 4527 | 2 \pm 2.7 | 26000 | 4.4 \pm 6.0 | -1.0 | -1.0 |
| Wet Open Lot | 4 | 14.1 \pm 5.4 | 9821 | 4.3 \pm 1.6 | 1400 | 0.5 \pm 0.2 | -1.0 | -1.0 |
| Separated Solids | 2 | 9.3 \pm 7.9 | 6478 | 2.8 \pm 2.4 | 110 | 0.03 \pm 0.02 | 3.6 | 3.7 |
| Lagoon 1 | 6 | 2.0 \pm 0.5 | 1393 | 0.6 \pm 0.2 | 14000 | 0.7 \pm 0.2 | 8.7 | 16.7 |
| Lagoon 2 | 6 | 0.4 \pm 0.3 | 279 | 0.1 \pm 0.1 | 16000 | 0.2 \pm 0.1 | 9.5 | 13.0 |
| Statistic | 29 ^a | | | | 88300 ^a | 24.7 ^a \pm 25.4 ^b | 8.0 ^c | 6.6 ^c |
| ^a Summation | | | | | | | | |
| ^b 95% confidence interval (CI) | | | | | | | | |
| ^c Average | | | | | | | | |

The average ambient temperature during winter conditions was 6.6 $^{\circ}\text{C}$ (Table 1). The highest average ammonia concentration was measured from the free-stall (36.4 ppm) and the lowest concentrations were detected from two lagoons (≤ 2 ppm). The lagoons did not have a significant contribution to the overall emission rates due to minimal biological activity as a result of low temperatures in the winter season. The compost and the free-stall areas contributed nearly 77% to the total ammonia emission rate for this facility while the dry open-lot contributed an additional 17%. The overall estimated emission rate of ammonia was $24.7 \pm 25.4 \text{ kg}\cdot\text{day}^{-1}$ for this facility. The area of free-stalls was less than one half of that of the dry-open lots, but they contributed the highest to the overall ammonia emission rate. This was due to the higher density of cows in the free stall barn resulting in greater amounts of manure and urine (waste) accumulation. Additionally, higher free stall barn temperatures as compared to open lot also contributed to higher ammonia emissions from waste (Table 1). High temperature of actively composting piles resulted in increased ammonia emissions and higher emission rates resulted from large area of the compost site. The wet open-lot had higher ammonia concentrations than the dry open lot but as a result of its small area it contributed little to the overall emission rate.

The second study was conducted in the summer of 2003. Fifty five samples were collected with isolation flux chamber methods to determine the emission rates of ammonia from the same sources plus the crowding area (adjacent to the milking parlor) of the dairy. Additionally, for better understanding of ammonia emissions from the free tall barn, sampling was conducted on the feed and non-feed sides and from bedding and watering areas of the barn. During the summer sampling, an improved automated-air flow control device (multiplexer) was used. The multiplexing procedure allowed controlling and regulating all MFCs automatically and

simultaneously. Ambient air, source, and chamber temperatures and chamber relative humidity were measured and recorded using HOBO sensors and data-loggers (Onset Computer Corporation, Pocasset, MA). The flux chambers were covered with cylindrical covers on the sides and on top to minimize potential over heating of the chamber when exposed to the environment. This insulation kept inside temperature of the chamber similar to the ambient temperature (Table 3).

During summer, ammonia concentrations ranged from 1.9 ppm at the composting site to 74.0 ppm on the feeding side of the free-stalls. Results of ammonia concentration and estimated emission rates are shown in Table 2.

Table 2. Ammonia concentrations and emission rates for 2003-summer.

| LLAS | Number of Samples | Concentration (ppm) | Concentration ($\mu\text{g}/\text{m}^3$) | Flow (L/min) | E Flux ($\mu\text{g}/\text{m}^2/\text{s}$) | Area (m^2) | ER (kg/day) |
|---|-------------------|----------------------------|--|--------------|--|-----------------------|------------------------------|
| Compost | 11 | 1.9 \pm 1.6 ^b | 1321 | 7.10 | 0.81 \pm 0.7 ^b | 16600 | 1.17 \pm 0.97 ^b |
| Freestall | 14 | | | | | 9790 | |
| Non-feed | 5 | 57.5 \pm 50.5 | 33349 | 7.09 | 20.53 \pm 23 | 2700 | 4.79 \pm 5.4 |
| Feed | 5 | 74.0 \pm 72.4 | 51574 | 7.09 | 31.75 \pm 31 | 3090 | 8.48 \pm 8.3 |
| Bedding | 2 | 2.4 \pm 22.2 | 1698 | 7.09 | 1.05 \pm 9.5 | 3800 | 0.34 \pm 3.1 |
| Water Area | 2 | 21.7 \pm 84.4 | 15113 | 7.09 | 9.30 \pm 36.2 | 200 | 0.16 \pm 0.63 |
| Open Lot | 8 | 4.8 \pm 3.9 | 3317 | 7.10 | 2.05 \pm 1.7 | 38000 | 6.72 \pm 5.5 |
| Crowding Area | 4 | 9.6 \pm 8.2 | 6690 | 7.03 | 4.06 \pm 3.4 | 925 | 0.32 \pm 0.3 |
| Separated Solids | 4 | 3.7 \pm 7.2 | 2428 | 7.09 | 1.50 \pm 2.9 | 109 | 0.01 \pm 0.03 |
| Lagoon 1 | 8 | 32.8 \pm 7.1 | 22878 | 7.10 | 14.09 \pm 3.0 | 19200 | 23.4 \pm 5 |
| Lagoon 2 | 6 | 28.1 \pm 2.9 | 19588 | 7.10 | 12.07 \pm 1.3 | 17000 | 17.72 \pm 1.9 |
| Statistic | 55 ^a | - | - | - | - | 101624 ^a | 63.1 ^a \pm 31.1 |
| ^a Summation | | | | | | | |
| ^b 95% confidence interval (CI) | | | | | | | |

Difference in ammonia emissions occurred due temperature variations, waste loading rates and biological activity. For instance, emission concentrations from both lagoons were ≤ 2 ppm in the winter season (Table 1) while summer ammonia concentrations were 32.8 ppm and 28.1 ppm from lagoon 1 and lagoon 2, respectively (Table 2). This increase in ammonia emissions from lagoons was attributed to increased volatilization of ammonia due to much higher lagoon temperatures in summer. Despite the higher compost pile-surface temperatures in summer than those in the winter season, ammonia concentrations from compost were lower in summer than those in winter (Table 1). During summer ammonia measurements, most compost piles had already gone through an active heating cycle (the differences between pile and ambient temperatures were only 9.8 °C during summer sampling as compared to 21.6 °C during winter sampling) and bacterial activity of composting was reduced, resulting in lower ammonia volatilization. Generally, free-stall ammonia concentrations in summer were higher than those from winter due to higher summer temperatures.

During summer, feed area of free-stall had the highest concentration followed by the non-feed side, water area and bedding (Table 2). The feed side of the barn had the most amount of dairy waste accumulation, resulting in the highest ammonia emissions. Waste around water tanks was diluted due to water spillage by cows in the vicinity, resulting in lower ammonia emissions than those from feed and non-feed sides. Bedding was composted separated solids with most nitrogen tied up in organic matter and very little ammonia volatilization, hence the lowest ammonia emissions were measured from the bedding area.

The overall estimated summer emission rate of ammonia was $63.1 \pm 31.1 \text{ kg day}^{-1}$ for this facility. It is noticeable that 65% of overall ammonia emission rates were contributed by two lagoons during the summer sampling. The free-stalls contributed an additional 22% to the overall ammonia emission rates.

Table 3. Weather data of sampling location in 2003-summer.

| Sampling Site | Barometric Pressure | LLAS Temp. | Chamber Temp. | Ambient Temp. | Chamber RH |
|------------------------|--------------------------|-------------------------|--------------------------|-------------------------|------------------------|
| | [kPa] | [°C] | [°C] | [°C] | [%] |
| Compost | 97.36 ±0.07 ^a | 43.17 ±7.1 ^a | 39.13 ±1.8 ^a | 33.34 ±1.6 ^a | 36.00 ±29 ^a |
| Free Stall | | | | | |
| Non-Feed side | 97.2 ±0.07 | 25.79 ±3.16 | 30.12 ±2.05 ^a | 33.38 ±1.33 | - |
| Bedding | 97.3 ±0.0 | 33.91 ±56.1 | 33.18 ±5.4 | 34.60 ±0.2 | - |
| Feed side | 97.20 ±0.05 | 27.02 ±2.78 | 31.09 ±2.48 | 33.34 ±3.14 | - |
| Water Area | 97.00 ±0.0 | 23.79 ±2.07 | 31.47 ±4.4 | 34.53 ±2.76 | - |
| Open Lot | 97.15 ±0.05 | 30.63 ±3.5 | 35.30 ±3.1 | 33.27 ±1.43 | 64.00 ±27 |
| Crowding | 97.14 ±0.03 | 21.54 ±1.0 | 24.20 ±1.0 | 25.62 ±1.0 | 73.00 ±2 |
| Separated Solid | 97.56 ±0.22 | 34.01 ±5.2 | 32.66 ±4.7 | - | - |
| Lagoon 1 | 97.05 ±0.2 | 29.48 ±1.2 | 29.68 ±1.8 | 29.61 ±2.3 | 87.00 ±9 |
| Lagoon 2 | 97.24 ±0.08 ^a | 28.42 ±0.7 | 27.71 ±2 | 26.67 ±1.9 | 89.00 ±11 |

^a 95% confidence interval (CI)

Low level area source surface, ambient and chamber temperatures and chamber relative humidity values are presented in Table 3. Chamber relative humidity data was unavailable for several LLAS due to limited number of humidity sensors. The average ambient summer temperature was 31.5 °C. No condensation was observed inside the chamber because LLAS, chamber and ambient temperatures were nearly the same during summer measurements.

Conclusions

The quantification of ammonia emissions from low level area sources in a dairy is needed to understand which sources contribute most to the overall ammonia emissions during winter and summer conditions. Ammonia emission concentrations are converted to emission rates to develop source specific ammonia emission control strategies. Two consecutive seasonal studies were conducted to estimate ammonia emission rates at a dairy in Central Texas in

2003. The overall and individual component ammonia emission rates were estimated with an improved flux chamber protocol. The estimated emission rate for the facility described in this study was 24.7 ± 25.4 kg day⁻¹ for the winter and 63.1 ± 31.1 kg day⁻¹ for the summer. This difference occurred due to seasonal changes in the temperatures, dairy waste loading rates, and the biological activity of LLAS. It is necessary to consider both management practices and climate conditions to determine ammonia emissions from the animal feeding operations. Long-term studies should be conducted to examine the impact of management practices on reducing ammonia emissions from animal feeding operations.

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