

## **A Passive Flux Denuder for Evaluating Emissions of Ammonia at a Dairy Farm**

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### **IMPLICATIONS**

The measurement of ammonia emission rates from fugitive sources traditionally has required expensive air sampling methods to measure concentration and wind speed. The passive flux denuder has been shown to be a simple and effective method to measure these emissions rates.

The sampling equipment is inexpensive, no electricity or batteries are needed to collect samples, and the sampling can be done with inexperienced personnel. This approach will facilitate local agencies in measuring emission rates from such fugitive emission sources. The affected industries can also easily monitor the effectiveness of control strategies to mitigate these sources.

## **ABSTRACT**

There is significant interest in developing economical methods to quantify ambient concentrations of gaseous pollutants. Passive samplers can provide an inexpensive alternative to direct sampling. Conventional denuder technology has been developed to measure semi-volatile pollutants, but active sampling inherently requires electrical power and pumps. The idea of comparing active sampling with a passive configuration, specifically for ammonia flux, came from the need to estimate ammonia emissions from area sources in California. Passive and active denuders were collocated at a dairy farm at the California State University, Fresno, Agricultural Research Facility. In this project, ammonia measurements were made for the dairy farm lagoon as it underwent acidification, and for the dairy farm as a whole. Comparisons were made of the flux measurements obtained directly from the passive flux denuder and those calculated from an active filterpack sampler and wind velocity. The results show significant correlation between the two methods and invite further investigation into characterization of the passive flux denuder response. With further evaluation of this technique, it is possible that a larger inventory base for ambient ammonia emissions, can be developed more economically than by using active samplers.

## **INTRODUCTION**

Ammonia ( $\text{NH}_3$ ) is an important trace constituent of the gaseous composition that make up the lower troposphere. It is the dominant gaseous base and is responsible for determining the level of atmospheric acidity<sup>1</sup>. The role that ammonia plays in neutralizing acidic aerosols has led to many studies concerning health effects of atmospheric aerosols<sup>2,3</sup>. Higher local emissions of  $\text{NH}_3$  result in the enhancement of aerosol formation that ultimately result in the following:

- 1) Formation of ammonium nitrate  $\text{NH}_4\text{NO}_3$  by reacting with  $\text{HNO}_3$ .
- 2) Formation of sulfate as the increase in pH enhances the rate of oxidation of dissolved sulfur dioxide by ozone<sup>4</sup>.
- 3) Formation of either ammonium sulfate ( $\text{NH}_4\text{HSO}_4$ ), or ammonium bisulfate ( $(\text{NH}_4)_2\text{SO}_4$ ) by neutralization of sulfuric acid<sup>5</sup>.
- 4) Formation of ammonium chloride by reacting with  $\text{HCl}$ <sup>6</sup>.

$\text{NH}_3$  has a lifetime usually of 1-5 days<sup>7</sup> but once transformed to ammonium aerosol has a lifetime that increases by at least a factor of three<sup>8</sup>. This allows for more widespread perturbation of soil acidity as well as direct deposition to plants resulting in excess nitrogen. Excess nitrogen is known to enhance the replacement of slow-growing plant species with fast-growing grass species<sup>9</sup>, causing adverse effects on many types of vegetation. In coastal areas excess nitrogen alters the growing frequencies of both toxic and non-toxic phytoplankton, seriously affecting this delicate ecosystem<sup>10</sup>. Increased production of ammonium aerosol also enhances light scattering, resulting in visibility degradation<sup>11</sup>.

It is known that the largest contributor of ammonia to the global budget is from domestic animal waste, as those from dairy, cattle, and swine farms<sup>12</sup>. Although there is now increased concern about  $\text{NH}_3$  emissions from mobile sources, there is still a strong need to evaluate domestic waste sources. California is ranked highest in milk production, producing 26 billion pounds of milk and cheese<sup>13</sup>. In 1998, livestock cash receipts totaled \$6.85 billion, due mainly to increased milk and cream sales<sup>13</sup>. While the growth of this industry has resulted in significant economic returns for the region, there is the issue of effective manure management. As much as 40% of feed dry matter fed to the animals can be excreted in the manure, requiring a significant

amount of labor and capital investment for manure management and disposal<sup>14</sup>. In dairy operations, manure is commonly handled as an effluent stream of liquid or slurry manure by means of a hydraulic flushing – lagoon storage – irrigation system. A major problem associated with the manure management is the loss of ammonia produced during the decomposition of manure in storage lagoons.

Between 80 and 200 different gases have been identified during the decomposition of manure<sup>15</sup>. Of these gases,  $\text{NH}_3$  is of primary concern because of: (a) the health issues related to particulate matter (PM) formation and (b) the loss of important plant nutrients from the manure. For example,  $\text{NH}_3$  is volatilized from lagoons, whereby the valuable ammonium ion ( $\text{NH}_4^+$ ) is lost as  $\text{NH}_3$ <sup>16</sup>. Volatilized  $\text{NH}_3$  can react in the atmosphere to produce ammonium nitrate or ammonium sulfate and thereby contribute to airborne particulate matter (PM). The California Air Resources Board (ARB) has developed preliminary emissions inventories for  $\text{NH}_3$  from most potential sources in the state<sup>17</sup>, all of which were reported with high initial uncertainties for source contributions. In the urban environment of California, ammonium nitrate accounts for between 30 and 60 percent of the fine aerosol mass. The Federal government and the State of California have established ambient air quality standards for particulate matter with aerodynamic diameters of both 10 microns or less ( $\text{PM}_{10}$ ) and 2.5 microns or less ( $\text{PM}_{2.5}$ ), which invites further concern of the overall ammonia contribution to the fine aerosol mass.

As a result of the health, environmental, economic and regulatory concerns, there is a need to quantify  $\text{NH}_3$  emissions at dairies. Since  $\text{NH}_3$  is one of the by-products of microbial degradation of manure and organic matter, a major location for this gas emission in a dairy operation is the lagoon, where the manure is stored and can be subjected to aerobic or anaerobic conditions<sup>18</sup>. In addition, gaseous emissions from these lagoons are influenced by climatic factors such as wind

speed, temperature, humidity and precipitation<sup>15,19</sup>, as well as, the pH of the dairy effluent<sup>18</sup>. For example, acidification of cattle slurry applied to grasslands has been used to reduce NH<sub>3</sub> emission rates<sup>20</sup>. The objective of the current study was to compare the NH<sub>3</sub> fluxes determined by passive flux denuders with those determined by an active sampling technique.

## **EXPERIMENTAL**

### **Active Sampling**

Filter packs were used to collect ammonia gas<sup>21</sup>. These consisted of a 47mm diameter Teflon front filter to remove particulate ammonium followed by a glass fiber filter treated with citric acid (5% in 95% ethanol and dried) to adsorb gaseous ammonia. A commercially available 12-volt air sampling pump was used to pull air through the filter pack at a nominal flow rate of 4 liters per minute. The treated filters were extracted with distilled water, reacted with Nessler's Reagent, and the absorbance was read on a spectrophotometer at 425nm. The amount of ammonia on the filter was determined in µg NH<sub>3</sub>, after correction for blank filters, using a calibration curve relating absorbance to ppm NH<sub>4</sub><sup>+</sup>-N in solution. The colorimetric procedure involving the use of the spectrophotometer is the method commonly used among dairy operators in the region for determining ammonium concentrations in the dairy effluent used for irrigating fields<sup>22</sup>.

The ammonia flux was determined by multiplying the denuder concentrations by the average wind speed at a given location. Anemometers were collocated with filter packs at 1, 2, and 5 meters above the soil surface to measure wind speed and direction. Finally, to be consistent

with flux data reported from the passive denuders (described later) the  $\mu\text{g N-NH}_3 \text{ m}^{-2} \text{ s}^{-1}$  values converted to  $\text{ng N-NH}_3 \text{ cm}^{-2} \text{ min}^{-1}$ .

## Passive Sampling

***Fabric Denuders.*** Diffusion denuders have been developed to sample air pollutants that are semi-volatile such as ammonium nitrate, which is a solid particle in equilibrium with ammonia and nitric acid vapor. These devices selectively and irreversibly remove the gas phase components while allowing particles to pass unattenuated. Diffusion denuders were first constructed as tubing bundles<sup>23</sup>, progressed to annular geometries<sup>24</sup>, and then to honeycomb structures<sup>25</sup>. Fitz and Motallebi recently reported the successful use of fabric denuders to selectively remove nitric acid<sup>26</sup>. The denuder sampling approach they proposed is based on diffusion batteries to remove very fine particles. The use of cloth fabric as the denuder substrate is analogous to the wire screen denuders for collecting very fine particles in diffusion batteries. Cloth, with a typical thread size of 100  $\mu\text{m}$  spaced on centers of 250  $\mu\text{m}$ , leaving an open grid of 150  $\mu\text{m}$  (typical dimensions for cloth), has been shown to efficiently remove gaseous species when coated with a suitable adsorbent. To estimate removal efficiency, comparisons to theoretically calculated values using the theory developed for wire diffusion screens were conducted. Fabric denuders were first evaluated by looking at the removal efficiency of  $\text{HNO}_3$  by  $\text{Na}_2\text{CO}_3$  coated examples<sup>26</sup>. Fitz and Motallebi observed that they could estimate denuder efficiency by estimating the fractional penetration of the gas and assume that the denuder is treated with a chemical that quantitatively removes a target gas, when the gas in question comes in contact with the denuder surface<sup>26</sup>. They concluded that two denuders made of a cotton fabric, 47 mm in diameter, were required in series to ensure a collection of over 95% of the  $\text{HNO}_3$  acid in ambient air when sampling up to 10 L/min.

*Ammonia Removal by Fabric Denuders.* To build upon this work and possibly apply this type of denuder to ammonia measurements, work was done to investigate the efficiency of ammonia removal by using phosphoric acid as the coating material<sup>27</sup>. Denuders were evaluated in the laboratory by passing known concentrations of ammonia (NH<sub>3</sub>) through them at various temperatures and humidities. Ammonia concentrations were generated using commercial permeation tubes with specified emission rates. Gas mixtures in the range of 10-40 ppb were generated at relative humidities of 10-80%. Concentration measurements, used to determine penetration, were made before and after the denuder. The concentration of ammonia was monitored by reducing NH<sub>3</sub> to NO by passing the sample through a high temperature stainless steel converter and then measuring the NO concentration with a commercial chemiluminescent NO analyzer. Denuders coated with 2% phosphoric acid and with 9% phosphoric acid were tested for collection of ammonia. Table 1 shows a summary of the results of testing phosphoric acid coated fabric denuders. Overall, the collection efficiency of a single 9% phosphoric acid denuder is better than 90%, and two denuders used in series would have better than 99% collection. This denuder is therefore suitable for field sampling of ambient ammonia.

### **Construction of a Passive Sampler Using Fabric Denuders**

The passive flux sampler design is based on the approach used by Schjoerring<sup>28</sup>, but, instead of using denuder tubes, the design was modified to use 47mm fabric denuders. Like filter types of active samplers, an open-face Savillex Teflon filter holder (Minnetonka, MN) is used to locate a pair of fabric denuders. However instead of an outlet to a pumping system, an open outlet is used. This open outlet was made from a section of machined 50mm PVC pipe. The other end of the PVC pipe is fitted with a similar pair of denuders, so there is symmetry on both sides of the

center axis through the PVC pipe. No flow restriction device was necessary. Sample collection depends on the wind flowing through the assembly. The amount of ammonia collected, therefore, is proportional to the wind speed, direction, and ammonia concentration. To measure the flux at a point, two such samplers are needed: one on an east-west axis, the other on a north-south axis.

Figure 1 shows a schematic of this sampler in a north-south axis orientation.

An experiment was conducted to examine the flow abatement through the passive denuder assembly. A variable speed blower was used to simulate wind speeds from 2 to 7 m sec<sup>-1</sup>. The laminar flow output from the blower was sufficiently large enough in diameter so as to encompass the front surface area (47 mm) of the denuder assembly. The simulated wind speed was measured with a Dwyer Wind Meter. The pressure drop inside between the pair of north-to-south or east-to-west fabric denuders was calibrated for different flows by using a manometer. This allowed determining the flow versus pressure response of the denuder assembly. This could be related to the bulk air speed past the denuder. Measurements were then made of the pressure drop inside the denuder assembly for various air speeds generated by the blower. A plot of the two in Figure 2 shows that the flow through the denuder is linearly reduced by approximately a factor of 11. Since these tests were done under ideal conditions, where the air was blown directly on the front face of the denuder assembly and neglecting the turbulent and directional changes in field conditions, this can only be used as an approximation. Further work needs to be done to investigate the flow abatement through the denuder as a function not only of wind speed, but also of turbulence and wind direction.

## Field Measurements

The California State University, Fresno, site is a medium-sized dairy operation (500 head). It provided an opportunity to conduct controlled measurements and to study emissions from an adjacent dairy lagoon. This also provided the best opportunity to compare the two types of sampling techniques. The inclusion of emissions from both the dairy farm as well as the lagoon was to eventually provide emission estimates for total dairy operation (to be reported elsewhere). The investigation of the lagoon was scheduled to coincide with CIT's (Center of Irrigation Technology) investigation into lagoon acidification practices to investigate dairy lagoon emissions from non-acidified lagoons, and acidified lagoons.

Six sets of passive denuders were used to collect samples at the dairy farm. Since the prevailing winds are from the northwest, the sampling tower was selected to be approximately 100 meters downwind of the dairy with passive denuders at 1m, 2m (two at this elevation) and 5 m. The collocated sample set was used to estimate the overall precision of the measurement. An upwind sample and another sample 100 meters downwind from the tower, both at 1m, completed the six sample sets. A total of 48 denuders were required for each sampling day, with two days sampled. The lagoon was located due north of the dairy and at a distance far enough from the dairy farm itself to be considered independent.

Four sets of passive denuders were used to sample the lagoon. Since the prevailing winds are also from the northwest, the sampling tower was directly southeast of the primary lagoon. The samplers, both active and passive, as well as individual meteorological sensors were set at 1m, 2m and 5 m. An upwind sample at 1m completed the four sample sets. A total of 32 denuders were collected for each of three sampling days. To minimize flow variation through the passive

denuders, sampling was conducted during the time of day when winds were consistently from a single direction. This was generally the case for the five sampling episodes as sampling times were kept between three and four hours during the early afternoon when the winds were the most stable. Two field blanks were taken for each sampling period. Table 2 lists the climatic conditions during monitoring of  $\text{NH}_3$  emissions at the dairy lagoon.

### **Passive Denuder Analysis**

The collected denuder samples were stored in airtight vials at  $0^\circ\text{C}$  and later analyzed in the laboratory by the indophenol method<sup>29</sup>. The denuders were first extracted with deionized water. Then reagents were added and the mixtures were allowed to react at room temperature for 45 minutes after which the absorbance at 635 nm of each sample was measured repeatedly using a Beckman DU-600 spectrophotometer. Any sample exhibiting an absorbance greater than 1 was diluted and measured again. The calculation of the actual ammonia nitrogen concentrations in the samples was done using a calibration curve based on an ammonium chloride standard solution. To generate this calibration curve, six standard solutions of ammonium chloride with concentrations in the range of 0–1 mg N/l were treated the same way as the denuder extracts. All denuder ammonia measurements were corrected for field blank ammonium.

Table 3 shows the data from one of the experiment sets. The indophenol methods result in the final concentration per denuder (micrograms per denuder). For the passive sampler, the horizontal ammonia flux was determined by adding the net amount of ammonium that was collected by each denuder and dividing this sum by the denuder's collection surface area. From the sample time (for this data set 240 min) and the active surface area of the denuder ( $13.9 \text{ cm}^2$ ) the total flux through the denuder for each of the 4 directions was calculated. The strongly biased directions for example the north and west for the dairy at one meter, show high denuder

efficiencies, greater than 90%. The directions opposite to the primary wind direction show much lower efficiencies and clearly have components of diffusion as well as turbulence. The lowest value of ammonia flux, which for this data set occurred for the dairy 5 m from the south, of 1.0 ng/cm<sup>2</sup>/min, can be used as a reasonable approximation for the diffusion component (D). Over the sampling days the diffusion component, determined this way, had a variance of 1.0 ng/cm<sup>2</sup>/min. Therefore the total net ammonia flux is the sum of the fluxes from the four directions (*i*) minus the diffusion component in each.

$$F_{NH3} = \sum(F_{NH3})_i - 4D \quad (1)$$

Figures 3 and 4 are plots showing the contribution to the total ammonia flux from each direction for the two sampling periods at the dairy farm. Figure 4 is a plot of the data from Table 2. The dairy farm was located northwest to the sampling tower and winds came from the northwest with average speeds of 1.5 to 2.3 m/s. The average directions were between 295 to 305 degrees. The plots show that there is almost equal contribution to the total flux from the north and west denuders. There is a markedly strong gradient to the ground as the net flux contribution from the five-meter elevation is significantly lower. Since the wind speed, direction and temperature were fairly consistent for the two days, then the close agreement of the two data sets indicate that emission rates from the dairy are fairly consistent under similar climatic conditions.

## RESULTS

The data from both the active and passive samplers were tabulated and are shown in Table 4. Figure 5 shows a plot of the 15 data points scaled differently by a factor of 20. The trend is quite consistent between the two. By plotting the data against each other, the plot in Figure 6 shows that they are significantly correlated with an  $r^2$  of 0.773. The average difference between the two

is 19.9 with a standard deviation of 4.1.

Although this ratio exceeds the predicted laboratory results of approximately 11, many factors could bias the results of the passive sampler being low. First, at the lagoon most of the wind came from the northwest at approximately 290 to 300 degrees, and was not directly into the face of the denuders. If we assume just a cosine function on the wind direction, this would increase the ratio from 11 to approximately 15.5. Also, the passive denuders and active samplers were both sampled for a period of approximately 4 hours for each of the measurements. Since the flux from the active sampler represents an average of the concentration multiplied by an average of the wind speed, there are likely discrepancies from this as the passive sampler measures flux directly. There are also the effects of turbulence that need to be further investigated on the passive sampler. Finally, filter packs tend to be biased high since ammonium nitrate particulate collected on the filter are very likely to be partially volatilized.

To estimate the error for the passive samplers, two sets of passive samplers were collocated at the 2 m height for two of the sample measurements. The individual 16 fabric denuders from the primary denuder set used for the flux analysis were compared with the collocated set as shown in Figure 7. The individual denuders show a very strong correlation with  $r^2=0.984$ . The offset of approximately 13% between the two is likely from inconsistencies in the phosphoric acid coating and sample analysis. The variance due to diffusive effects was found to be approximately  $1 \text{ ngNH}_3/\text{cm}^2/\text{min}$ , so that the data on Figures 4 and 5 for the passive samples are  $F_{\text{NH}_3} \pm (.065(F_{\text{NH}_3})+1)$ . Active samples were tested and shown to have cumulative errors based on the flow measurement, wind speed measurement and inconsistencies of the citric acid coating and filter analysis. Overall there is no consideration to diffusive variance and the data on Figure 4 for the active samples are  $F_{\text{NH}_3} \pm 0.08(F_{\text{NH}_3})$ .

The northwest bias of the individual denuders is observed from the plots in Figures 8 and 9. The stronger western component is due to the location of the tower and geometry of this particular dairy lagoon. The tower was located southeast of the secondary lagoon, but the primary lagoon was located due west of the secondary lagoon and therefore expected to increase the western component of the  $\text{NH}_3$  flux. Figures 8 and 10, pre and post acidification plots of the net ammonia flux, indicate that the net flux has been abated quite significantly by the acidification process. Before the addition of any acid, the average pH in the primary lagoon was 7.5, but it dropped to 6.3 on the day following the addition of approximately 1500 gallons of concentrated sulfuric acid. Overall, there was a decrease in the ammonia fluxes from the lagoon during the experiment, as shown in Figure 11. This is consistent with that of ammonia emission-pH trends reported in the literature<sup>30,31</sup>. A pH of greater than 7.5 in the lagoon favors  $\text{NH}_3$  volatilization<sup>18</sup>. It has also been reported that acidification of cattle slurry has been shown to reduce  $\text{NH}_3$  emission rates<sup>20</sup>.

Ammonia ( $\text{NH}_3$ ), which is in equilibrium with ammonium ( $\text{NH}_4^+$ ) in the wastewater, is the end product of nitrogen degradation in the storage lagoon. The loss of  $\text{NH}_3$  to the atmosphere is in response to any differences in the  $\text{NH}_3$  partial pressures between that in equilibrium with the liquid phase and that in the ambient atmosphere. Hence the climatic and environmental factors that affected the  $\text{NH}_3$  partial pressure of the ambient atmosphere would have ultimately influenced the  $\text{NH}_3$  fluxes during the three days of the experiment. Table 2 shows the average values for wind speed, wind direction, air temperature, and relative humidity (RH), as well as the pH, temperature, and dissolved oxygen (DO) in the wastewater during the sampling at the lagoon.

In addition to the changes in pH of the wastewater in the lagoon, the minor climatic changes in the parameters listed in Table 2 may also account for the differences in the ratio of  $\text{NH}_3$  fluxes determined by the two methods observed during the three days of monitoring at the lagoon. For example, the relatively highest  $\text{NH}_3$  flux observed at the 5 m location during the acidification was probably influenced by the fact that the highest average wind speed was also recorded at this location. Following the acid injection, the lagoon was aerated for approximately two hours. The primary purpose for the aeration was to maintain a slightly aerobic layer. The aerobic surface, evident by the slight increase in DO was expected to control the odors associated with gases such as ammonia and hydrogen sulfide produced at the bottom of the anaerobic lagoon<sup>32</sup>. For example, under aerobic conditions the  $\text{NH}_3$ , produced within the sub-surface anaerobic layers, will first be converted to  $\text{NH}_4^+$  by heterotrophic bacteria and then into nitrate and nitrite by autotrophic bacteria. Hence, the nitrite and nitrate will remain solution and the amount of  $\text{NH}_3$  emitted from the lagoon will be reduced.

## **CONCLUSION**

The passive flux denuder has shown to have the potential to be an accurate and economical method to measure ammonia fluxes from area sources. This information can be used with dispersion models to determine the overall emission rates for these types of sources. Additional research is needed to determine the cause of the discrepancy between the flux determined by the active and passive samplers.

**Table 1.** Phosphoric acid coated fabric denuder efficiency.

	<b>Nominal Conc. ppb</b>	<b>Temp C</b>	<b>RH %</b>	<b>Duration days</b>	<b>Flow rate L/min</b>	<b>Denuder Coating (w/w)</b>	<b>Collection efficiency %</b>
<b>NH<sub>3</sub></b>	<b>38</b>	<b>21</b>	<b>17</b>	<b>0.9</b>	<b>2</b>	<b>2% phosphoric</b>	<b>66</b>
<b>NH<sub>3</sub></b>	<b>38</b>	<b>20</b>	<b>20</b>	<b>1.8</b>	<b>2</b>	<b>9% phosphoric</b>	<b>90</b>
<b>NH<sub>3</sub></b>	<b>40</b>	<b>38</b>	<b>20</b>	<b>2.7</b>	<b>2</b>	<b>9% phosphoric</b>	<b>95</b>
<b>NH<sub>3</sub></b>	<b>15</b>	<b>21</b>	<b>20</b>	<b>1.1</b>	<b>2</b>	<b>9% phosphoric</b>	<b>100</b>
<b>NH<sub>3</sub></b>	<b>15</b>	<b>21</b>	<b>80</b>	<b>7.0</b>	<b>2</b>	<b>9% phosphoric</b>	<b>100</b>
<b>NH<sub>3</sub></b>	<b>13</b>	<b>38</b>	<b>22</b>	<b>4.9</b>	<b>2</b>	<b>9% phosphoric</b>	<b>100</b>

**Table 2.** Climatic conditions during monitoring of NH<sub>3</sub> emissions at the dairy lagoon.

Description	Ambient Atmosphere					Wastewater at Surface		
	Height (m)	Speed (m/s)	Direction (degrees)	Temp. (C)	Relative Humidity (%)	pH	Temp. (C)	Dissolved Oxygen (%)
Pre -acid				26.1	32.6	7.5	24.0	0.3
Pre-acid	1	1.12	292.9					
Pre-acid	2	2.01	291.6					
Pre-acid	5	2.10	297.0					
During acid				25.9	35.9	6.9	23.9	0.3
During acid	1	1.83	300.7					
During acid	2	2.91	297.1					
During acid	5	3.09	303.2					
Post acid				24.3	42.6	6.3	22.8	0.7
Post acid	1	0.94	240.6					
Post acid	2	1.25	237.0					
Post acid	5	1.12	242.9					

**Table 3.** Set of dairy 2 passive denuder data showing total NH<sub>3</sub> flux calculation.

Sample ID Field	Ammonia Concentration ug/denuder	Denuder efficiency	Total Flux Through Denuder ng/cm2/min	Total Ammonia Flux ng/cm2/min
4NF	111.6	94%	28.6	27.5
4NB	7.4			
4SF	10.1	57%	4.2	3.2
4SB	7.6			
4EF	4.1	38%	2.6	1.6
4EB	6.7			
4WF	116.9	93%	30.2	29.2
4WB	8.8			
<b>DAIRY 1 METER</b>				<b>61.5</b>
5NF	98.6	92%	25.6	24.6
5NB	8.2			
5SF	10.2	52%	4.7	3.7
5SB	9.5			
5EF	3.8	32%	2.9	1.8
5EB	8.1			
5WF	106.9	93%	27.5	26.5
5WB	7.7			
<b>DAIRY 2 METER</b>				<b>56.6</b>
6NF	112.6	94%	28.6	27.6
6NB	6.6			
6SF	7.5	52%	3.4	2.4
6SB	6.9			
6EF	6.3	36%	4.2	3.2
6EB	11.3			
6WF	130.5	93%	33.7	32.7
6WB	9.8			
<b>DAIRY (Co-located) 2 METER</b>				<b>65.8</b>
7NF	56.2	100%	13.5	12.4
7NB	-0.1			
7SF	3.0	69%	1.0	0.0
7SB	1.3			
7EF	22.7	78%	7.0	5.9
7EB	6.3			
7WF	87.2	99%	21.1	20.1
7WB	0.8			
<b>DAIRY 5 METER</b>				<b>38.5</b>

**Table 4.** Comparison of active and passive denuder data.

<b>Count</b>	<b>Description</b>	<b>Height (meters)</b>	<b>Active (ngNH<sub>3</sub>/cm<sup>2</sup>/min)</b>	<b>Passive (ngNH<sub>3</sub>/cm<sup>2</sup>/min)</b>	<b>Ratio</b>
1	Pre-acid	1	991.7	52.5	18.9
2	Pre-acid	2	1026.4	59.1	17.4
3	Pre-acid	5	592.6	40.7	14.6
4	During acid	1	1031.2	70.2	14.7
5	During acid	2	946.9	63.2	15.0
6	During acid	5	804.1	42.3	19.0
7	Post acid	1	509.5	19.2	26.6
8	Post acid	2	391.5	15.1	25.9
9	Post acid	5	242.7	10.2	23.8
10	Dairy 1	1	1418.3	61.5	23.1
11	Dairy 1	2	1094.8	56.6	19.4
12	Dairy 1	5	839.2	38.5	21.8
13	Dairy 2	1	1798.9	82.3	21.9
14	Dairy 2	2	1282.2	85.1	15.1
15	Dairy 2	5	903.5	26.8	33.8

**Figure 1.** North-south passive sampler using 4 fabric denuders.

**Figure 2.** Flow abatement through the passive denuder assembly.

**Figure 3.** plot showing ammonia flux from the dairy versus wind direction (first day of measurements).

**Figure 4.** plot showing ammonia flux from the dairy versus wind direction (second day of measurements).

Figure 5. Comparison of active and passive denuder data for the 15 sample points.

**Figure 6.** Regression plot of active and passive denuder data.

**Figure 7.** Comparison of collocated passive denuder data.

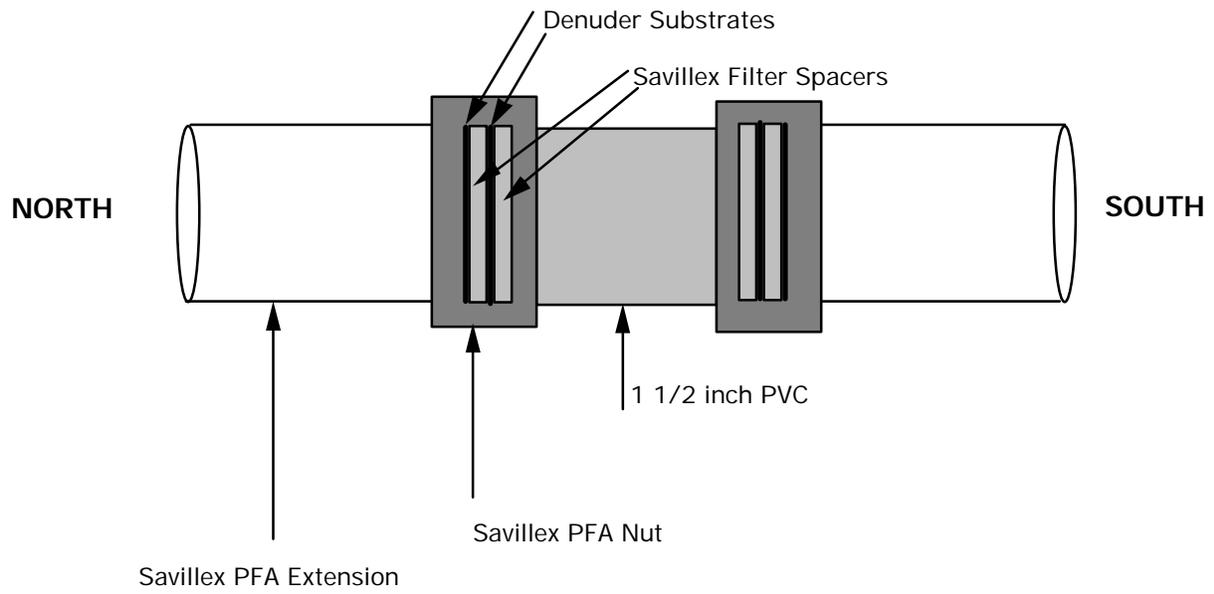
**Figure 8.** plot showing ammonia flux from the dairy lagoon versus wind direction (pre-acidification).

**Figure 9.** plot showing ammonia flux from the dairy lagoon versus wind direction (during acidification).

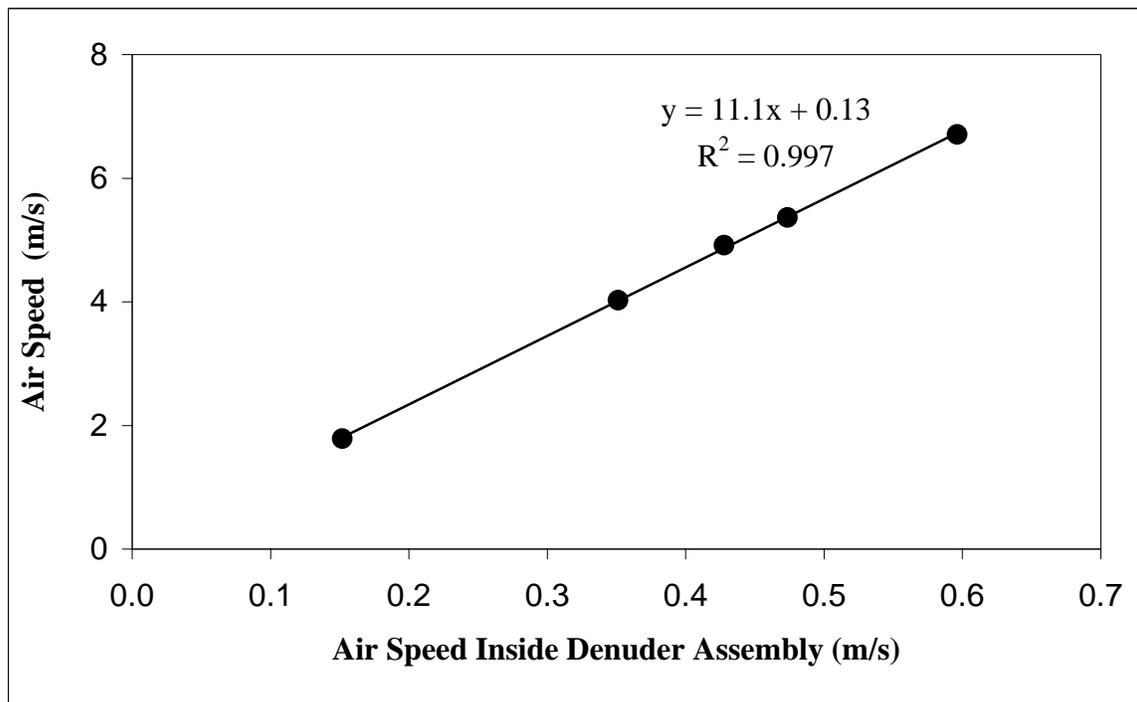
**Figure 10.** plot showing ammonia flux from the dairy lagoon versus wind direction (post acidification).

**Figure 11.** Ammonia fluxes measured with active samplers before, during and after acidification of lagoon.

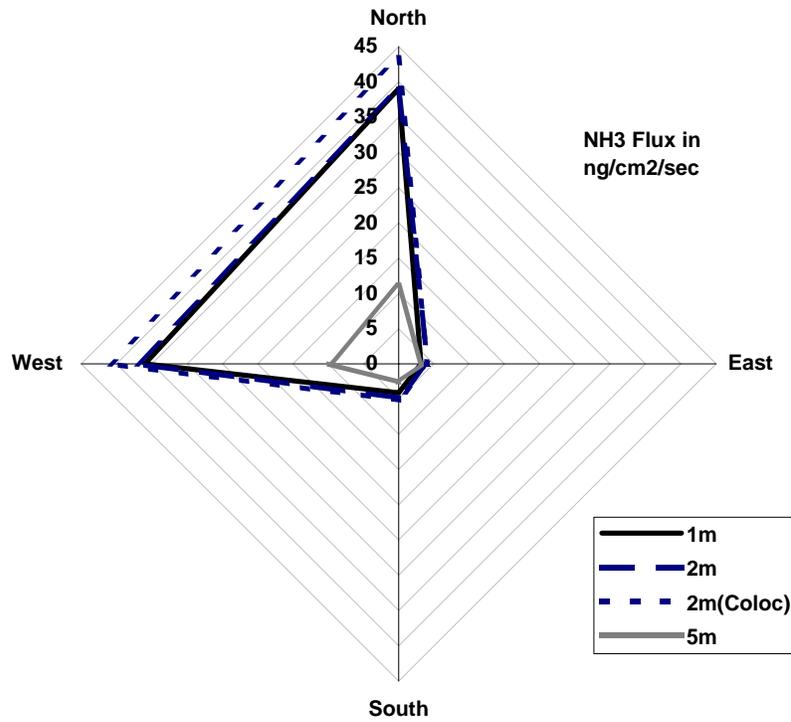
**Figure 1.** North-south passive sampler using 4 fabric denuders.



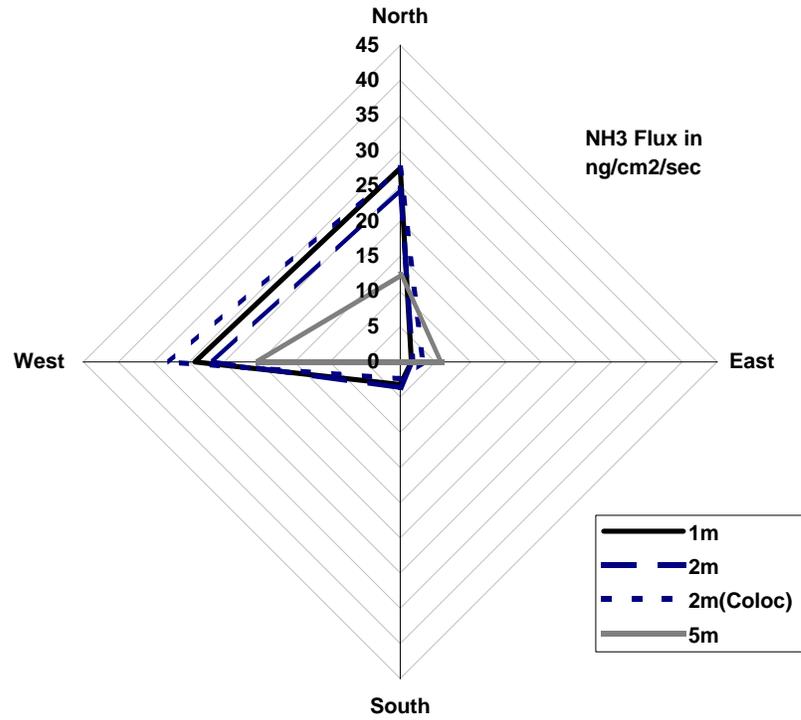
**Figure 2.** Comparison of air speed outside and inside the passive denuder assembly



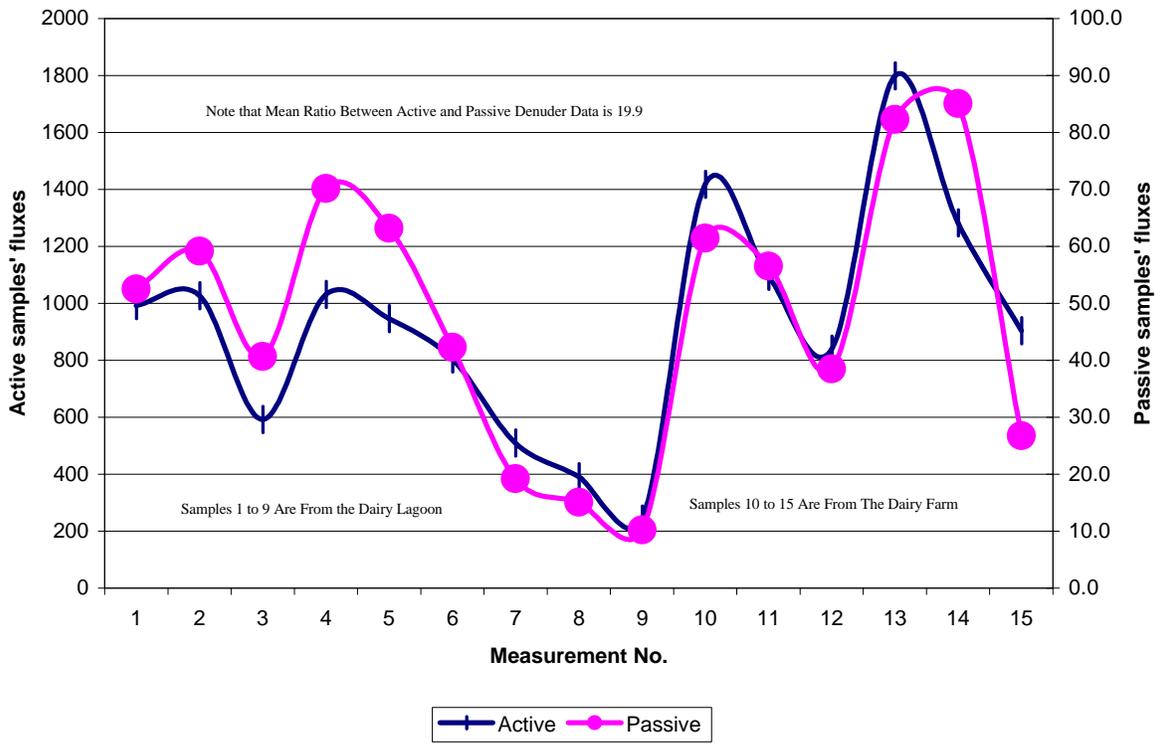
**Figure 3.** Plot showing ammonia flux from the dairy versus wind direction (first day of measurements).



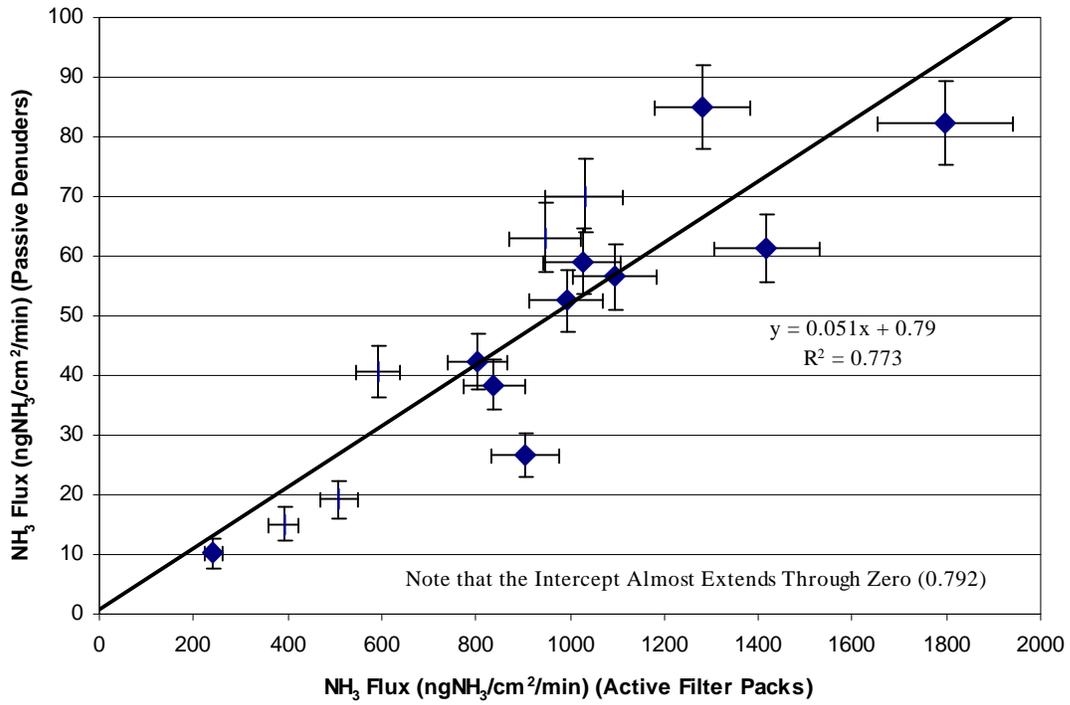
**Figure 4.** Plot showing ammonia flux from the dairy versus wind direction (second day of measurements).



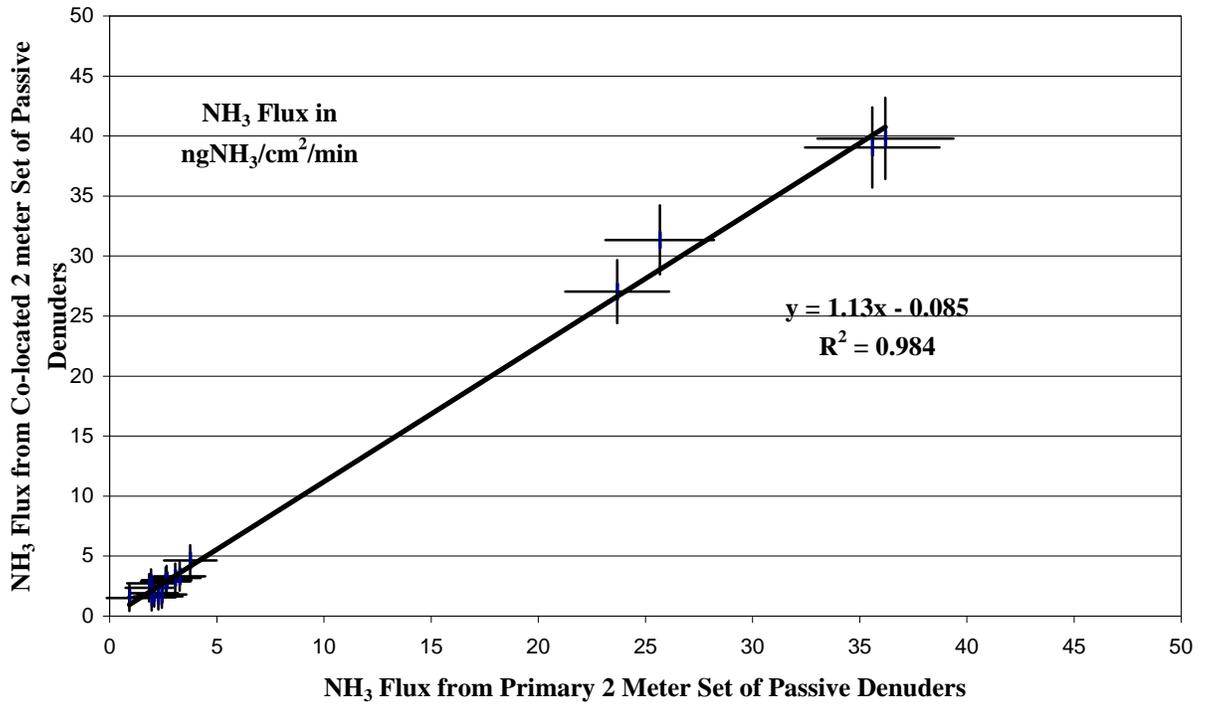
**Figure 5.** Comparison of active filter pack and passive denuder flux data for the 15 sample points.



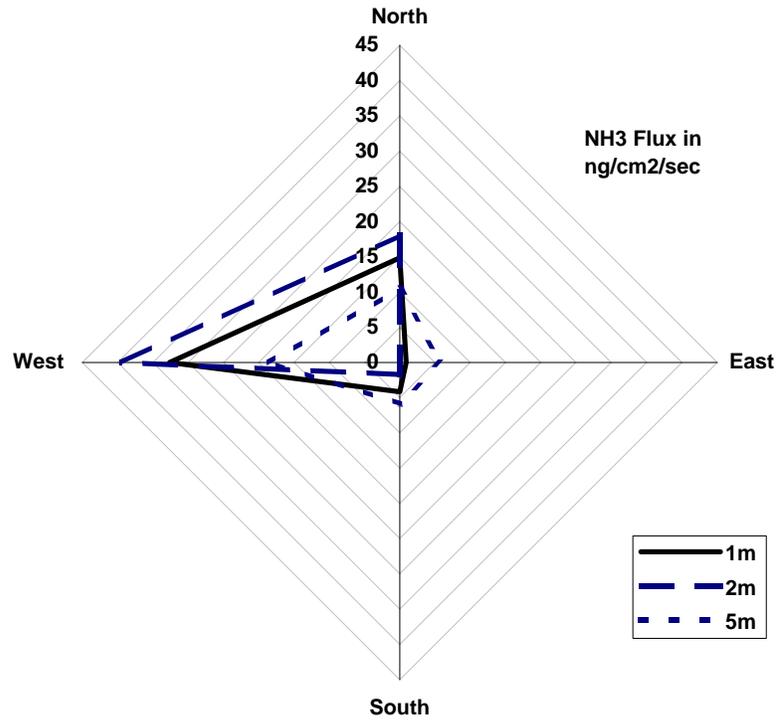
**Figure 6.** Regression plot of active filter pack and passive denuder flux data.



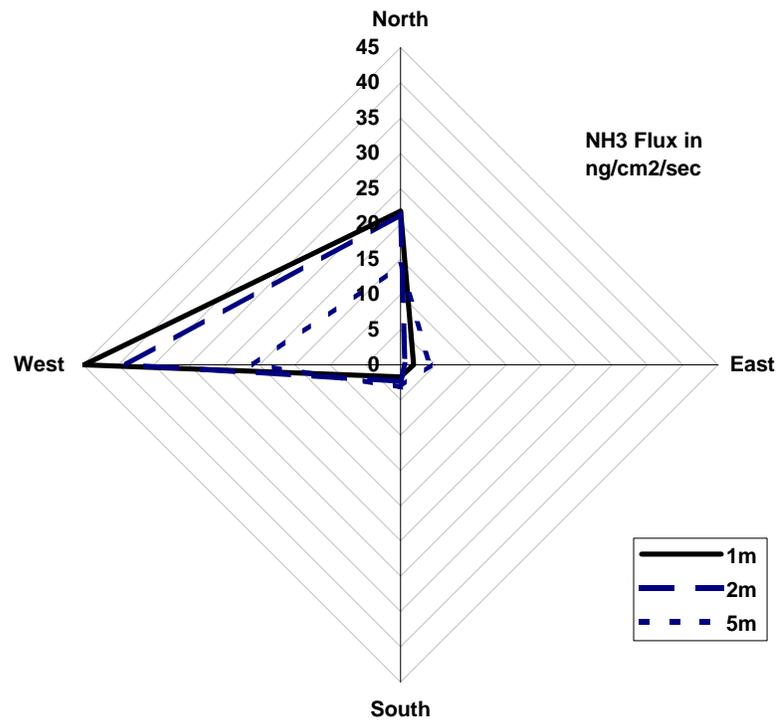
**Figure 7.** Comparison of collocated passive denuder data.



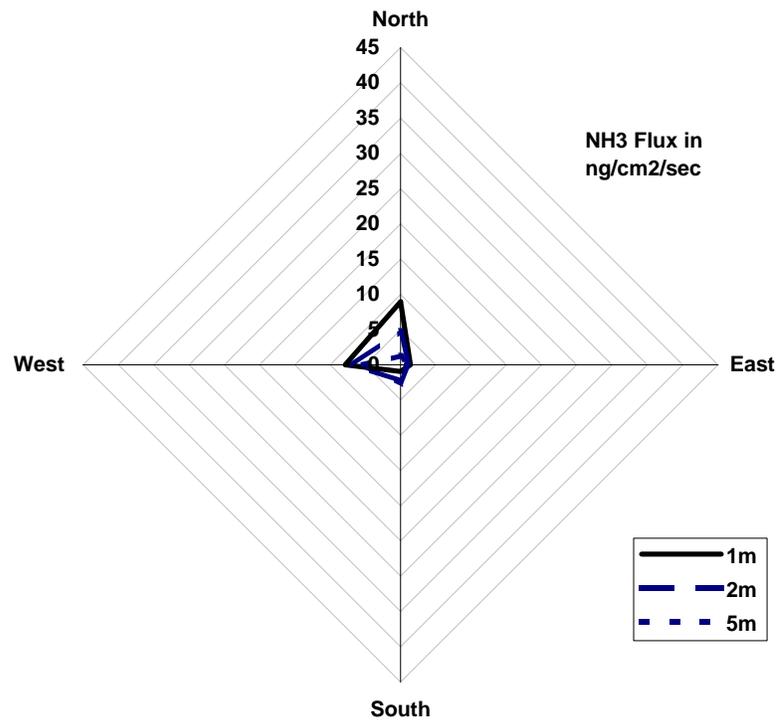
**Figure 8.** Plot showing ammonia flux from the dairy lagoon versus wind direction (pre-acidification).



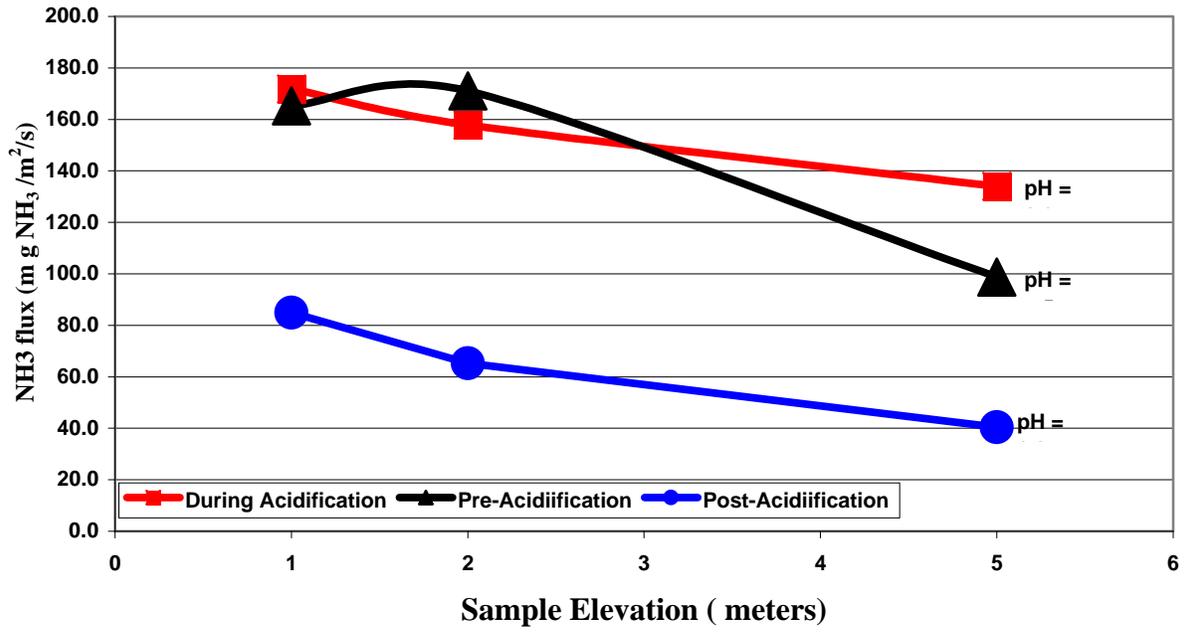
**Figure 9.** Plot showing ammonia flux from the dairy lagoon versus wind direction (during acidification).



**Figure 10.** Plot showing ammonia flux from the dairy lagoon versus wind direction (post acidification).



**Figure 11. Ammonia fluxes measured with active samplers before, during and after acidification of lagoon.**



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## **ACKNOWLEDGMENTS**

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## **ABOUT THE AUTHORS**