

# Ammonia Flux Profiles for Various Soil and Vegetation Communities in California

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## ABSTRACT

Atmospheric ammonia was sampled using active denuders co-located with wind instruments on a mast from near the soil surface to a height of 10m. NH<sub>3</sub> flux profiles were calculated from the data for a variety of soil/vegetation communities in central California. Profiles were calculated from samples taken several times during the season at the same location. Sites included rangeland in the Sierra Nevada foothills, various crops and a dairy operation in the San Joaquin Valley. The magnitude and characteristics of the NH<sub>3</sub> flux profiles were compared to similar data from other research outside California. Correlations with air temperature and diurnal differences were similar to those found elsewhere. Some indication of NH<sub>3</sub> absorption by active vegetation was found under circumstances similar to those reported by other researchers. Sampling will continue for an additional year at these and other sites to develop a more complete data set and increase the validity of the correlations between various environmental factors and the NH<sub>3</sub> flux profiles.

## INTRODUCTION

Ammonia is the dominant gaseous base in the atmosphere and a principal neutralizing agent for atmospheric acids. The NH<sub>3</sub> in the atmosphere, along with alkaline soil dust, may control the acidity of precipitation. Volatilized NH<sub>3</sub> may react to form ammonium nitrate or ammonium sulfate and thereby contribute to airborne particulate matter (PM<sub>2.5</sub>). Estimated patterns of nitrogen deposition suggest that, for California locations close to photochemical smog source areas, concentrations of oxidized forms of N dominate, while in areas near agricultural activities the importance of reduced N forms may increase significantly according to Bytnerowicz and Fenn<sup>1</sup>. NH<sub>3</sub> remains one of the most poorly characterized atmospheric trace compounds in terms of overall sources. This situation persists as a result of several factors such as; experimental difficulties associated with NH<sub>3</sub> measurements, rapid gas-to-particle conversion of NH<sub>3</sub> in the atmosphere, the capacity of soils, organic matter, vegetation to act as both sources and sinks for atmospheric NH<sub>3</sub>, and variability in nitrogen fertilizer management and related NH<sub>3</sub> emissions (Langford et al.<sup>2</sup>). Consequently, there is a limited amount of published information from which to develop direct emissions estimates of NH<sub>3</sub> for the state of California in general, and the state's Central Valley in particular. Preliminary measurements of NH<sub>3</sub> background concentrations in the San Joaquin Valley by Fitz et al.<sup>3</sup> estimated February levels of 3-16 µg m<sup>-3</sup> near alfalfa fields. The magnitude and distribution (both regionally and seasonally) of current NH<sub>3</sub> emissions from fertilizer and other agricultural sources is still largely undetermined for the state of California and many other large regions where agriculture is a major land use (Matthews<sup>4</sup>).

A recent report for the California Air Resources Board (ARB) by Potter et al.<sup>5</sup> provided new emissions inventories of ammonia volatilization from surface applied fertilizers and from native soil sources for the state of California. The NH<sub>3</sub> inventory results were based on a combination of field measurements and computer modeling of major nitrogen transformations in the soil that can lead to NH<sub>3</sub> emission fluxes. A year-round sampling campaign was carried out on a series of farms undergoing typical fertilizer applications for commercial crop production. Emissions of NH<sub>3</sub> for a variety of fertilizer types and application methods were mapped for the major crop types in California's four main agricultural valleys. In this same ARB-supported report a comparison of published emission estimates for natural soils showed many missing data values for major vegetation types common to California. Emission

estimates from agricultural soils for periods other than fertilizer application is also very limited. In the absence of a comparable measurement data set to fertilizer applications for NH<sub>3</sub> from other soil sources in California, a computer modeling approach was used to estimate statewide annual emission rates of N-NH<sub>3</sub> from native soils and indirectly from fertilizer N sources in cultivated soils. The modeling system used (called CASA) is based on regional data sets (8-km resolution) from a geographic information system (GIS) developed specifically for the ARB-sponsored research on N-NH<sub>3</sub> emissions. Based on an initial inventory estimate, statewide emissions of NH<sub>3</sub> from soil N sources appear to total just over 30 x 10<sup>6</sup> kg N- NH<sub>3</sub> annually, which includes a large contribution from cultivated soils. The CASA model results suggested other, native soils could be contributing substantially to the statewide emission inventory for emissions of NH<sub>3</sub>.

The emissions estimated by the CASA model are based on well documented relationships between plants and soil microbes known as the Nitrogen cycle. Ammonia is one of the end products of organic matter decomposition by soil fungi and bacteria. Some NH<sub>3</sub> will find its way to the atmosphere and, under certain soil conditions, there may be significant volatile losses. The fate of the NH<sub>3</sub> after it reaches the atmosphere is less well known. The formation of secondary particles, PM<sub>2.5</sub> described above, is certainly one possibility. However, the fate of NH<sub>3</sub> in the micro-climate near active vegetation appears to be complex. The interior structure of the typical plant leaf is adapted to absorb atmospheric CO<sub>2</sub> for photosynthesis. The diffusive characteristics of NH<sub>3</sub> in the atmosphere are likely to be similar to CO<sub>2</sub> suggesting the possibility of NH<sub>3</sub> absorption as well. Harper et al. (1983)<sup>6</sup> found NH<sub>3</sub> losses to the atmosphere from tropical pastures in Australia after fertilization and at various other times through the season. NH<sub>3</sub> losses were correlated with high air temperatures and solar radiation levels. Absorption by vegetation was observed during the same study correlated with dawn and dusk periods. Harper et al. (1996)<sup>7</sup>, on a temperate grassland in Georgia, found emissions after fertilization but again measured NH<sub>3</sub> absorption by the vegetation. He estimated 6% of the N in the vegetation was absorbed as atmospheric NH<sub>3</sub> in the cooler part of the growing season and 11% in the summer. An earlier investigation by Harper and Sharpe<sup>8</sup> on irrigated corn in Nebraska used <sup>15</sup>N labeled fertilizer to show both emission and absorption of NH<sub>3</sub> by the crop at various times through the season. Some absorption of <sup>15</sup>N labeled NH<sub>3</sub> by crops that had not been fertilized was detected; indicating the possibility that emission of NH<sub>3</sub> from fertilized plots was absorbed by nearby vegetation. NH<sub>3</sub> absorption was correlated with air temperature, solar radiation, soil moisture, soil N levels but primarily with NH<sub>3</sub> concentration in the atmosphere. High atmospheric NH<sub>3</sub> resulted in absorption regardless of the other factors. Absorption of NH<sub>3</sub> has been reported in other research, notably Porter et al.<sup>9</sup>, where corn seedlings were placed in an atmosphere spiked with labeled <sup>15</sup>NH<sub>3</sub>. They found absorption of up to 30% of NH<sub>3</sub> in a 24 hour period.

The somewhat contradictory findings cited above illustrate the complexity of NH<sub>3</sub> transport in the soil-plant-atmosphere system. The data presented below are from a variety of soils and vegetation communities in California. The same sites were sampled at various times of the year. Sampling was continuous over several days and included separate diurnal samples. The results, to date, suggest both emission and absorption correlated with many of the same factors seen in the previous research. Additional sites have been established and further data is to be collected and analyzed.

The active denuder methodology developed at CSU Fresno for the study by Potter et al.<sup>5</sup> was used for the sampling portion of this project. This methodology represents an established technique in air quality studies, and it satisfies the inventory development requirement for continuous sampling of soil gas emissions over relatively long time periods. Denuders and anemometers were co-located at 0.5, 1, 2, 4, and 10 meters above the soil surface, mounted on a portable mast. The denuder is a 47 mm disk of glass fiber filter paper, treated with citric acid (5% in 95% ethanol) and dried. A commercially available, 12 volt air sampling pump is used to pull air through the denuder disk at a regulated rate of 1 to 5 liters per minute. Lower air flow rates were used for long sampling period of several days. The higher air flows were selected when the sampling period was to be limited to a few hours. Filter samples were refrigerated and taken to the Graduate Laboratory

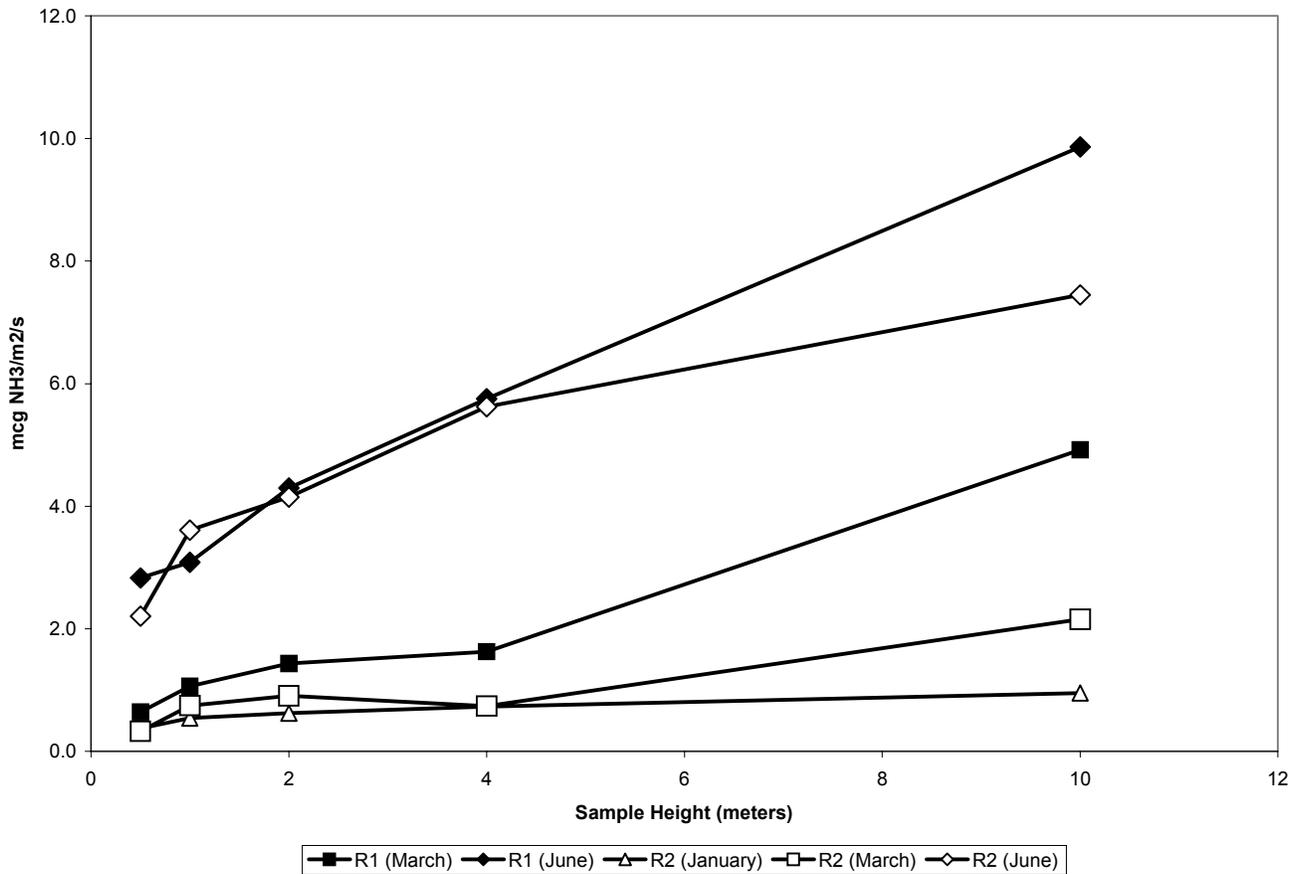
of the CSU Fresno College of Agricultural Science and Technology, to be analyzed by project personnel. The  $\text{NH}_4$ -Citrate was extracted from the denuder with distilled water and analyzed with a spectrophotometer. The amount of ammonia on the denuder disk is reported in  $\mu\text{g NH}_3$ . The concentration of  $\text{NH}_3$  in the air at the sampling point was determined by dividing the amount of ammonia on the disk by the volume ( $\text{m}^3$ ) of air pumped through the denuder in the sampling period to derive concentrations in units of  $\mu\text{g N-NH}_3 \text{ m}^{-3}$  air at the sampling point. There was a considerable wind profile over the 10m height of the sampling mast. Wind speeds at 10m were typically higher than the 0.5m sampling point by a factor of 5 or more. The wind speed was significantly different even between the 0.5m and 1.0m sampling elevations. The wide range in wind speed over the sampling profile required the conversion of the concentration in  $\mu\text{g NH}_3 \text{ m}^{-3}$  to a flux in  $\mu\text{g NH}_3 \text{ m}^{-2} \text{ sec}^{-1}$ . The  $\text{NH}_3$  flux was calculated by multiplication of the concentration by the wind speed in m/s.

Sampling locations were selected from cropland, range, natural vegetation and dairies. Five crops, three categories of natural vegetation and a dairy were sampled during the first year of the study. Permanent and annual agronomic crops were sampled several times from the beginning of the crop season through harvest. Two different grazing sites were sampled every 3 months. A conifer forest in the Sierra Nevada and a coastal forest have been sampled once and will be again in 2003. The dairy sampling was conducted as part of a long term project at a facility near Hanford in the San Joaquin Valley. The results reported below are for sampling sites on foothill rangeland, a field with a barley crop that was followed by a corn crop, and a silage sorghum crop around the dairy. These were all in central California, within 100 km of Fresno. Sampling at these and the other locations are continuing through 2003. The data reported here are selected as examples of the preliminary conclusions. Results of the complete sampling program and the conclusions from it will be available in 2004.

## RESULTS AND DISCUSSION

A correlation between air temperature and  $\text{NH}_3$  levels was anticipated. One of the reasons for sampling the same site at various times through the year was to verify that relationship. The magnitude of the flux and the characteristics of the vertical profile did appear to correlate with the average air temperature. The San Joaquin Experimental Range is a 2000 Ha field site administered by CSU Fresno for the US Forest Service. Two sampling sites were designated and sampled every few months beginning in January of 2002. Some of the data is shown below in Figure 1.

**Figure 1.** Ammonia flux profiles for two rangeland sites located 40 km north of Fresno at an elevation of 300m. Each line represents an average of 3 to 5 days of continuous sampling. The two sites were about 200m apart. R1 (closed graph symbols) was an open grassland with no trees or brush within 50m of the mast and R2 (open symbols) had a higher level of vegetation; a mix of grass, brush and trees.

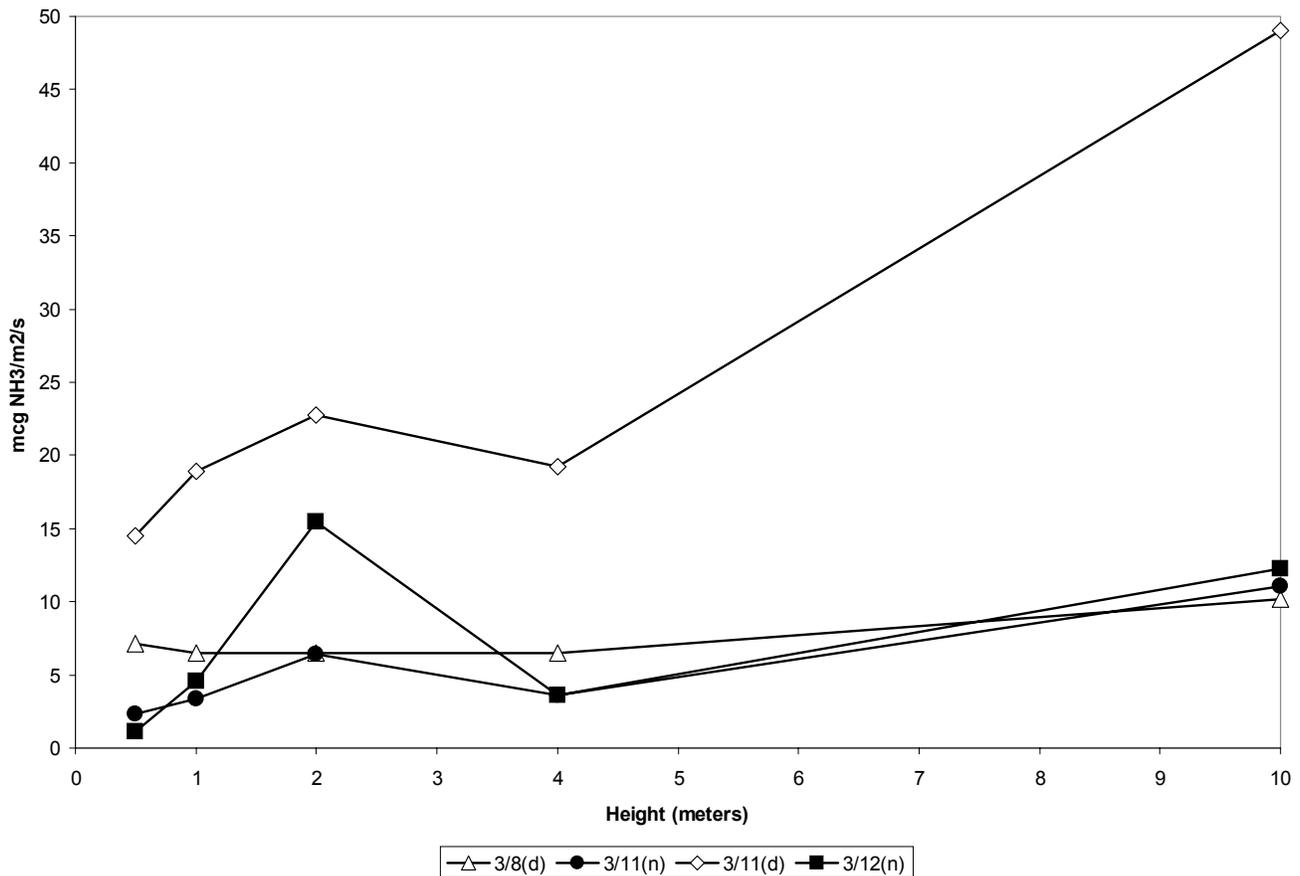


The average air temperature during the sampling in January was 8.6 C. In March the average was 14.6, and in June the temperature averaged 24.0 C. The lowest  $\text{NH}_3$  values were in January, the highest in June. There were two sampling sites and both sets of flux profiles appear to be correlated with temperature. Site R1 was in an open meadow of annual grasses. Site R2 had brush and trees enclosing a small area of grasses. It is likely that the air temperature, particularly in the shade, near the soil surface was lower at R2. The  $\text{NH}_3$  flux values were higher for the R1 site sampled at the same time as the R2 site, particularly at 10m. The difference may be due in part to temperature. The R1 site had no tall vegetation within 50m of the mast compared to the R2 site where there were 20m trees within 5m of the mast. Further sampling may be able to distinguish other correlations with environmental factors between the two range sites.

A second relationship between an environmental factor and atmospheric  $\text{NH}_3$  flux profiles is the distinct, diurnal difference. In each sampling episode to date, at least one day of collection was divided into a day and night sample. Nearly every one of those diurnal sample pairs showed considerably more

NH<sub>3</sub> during the day compared to the night sample. Figure 2 is data from a field planted to barley. There were three periods of sampling from the seedling stage in November of 2001 to the harvest in March, 2002. The average of the flux profiles for each sampling period showed the same correlation with air temperature as the rangeland in Figure 1. Figure 2 shows the diurnal differences for the last sampling in early March.

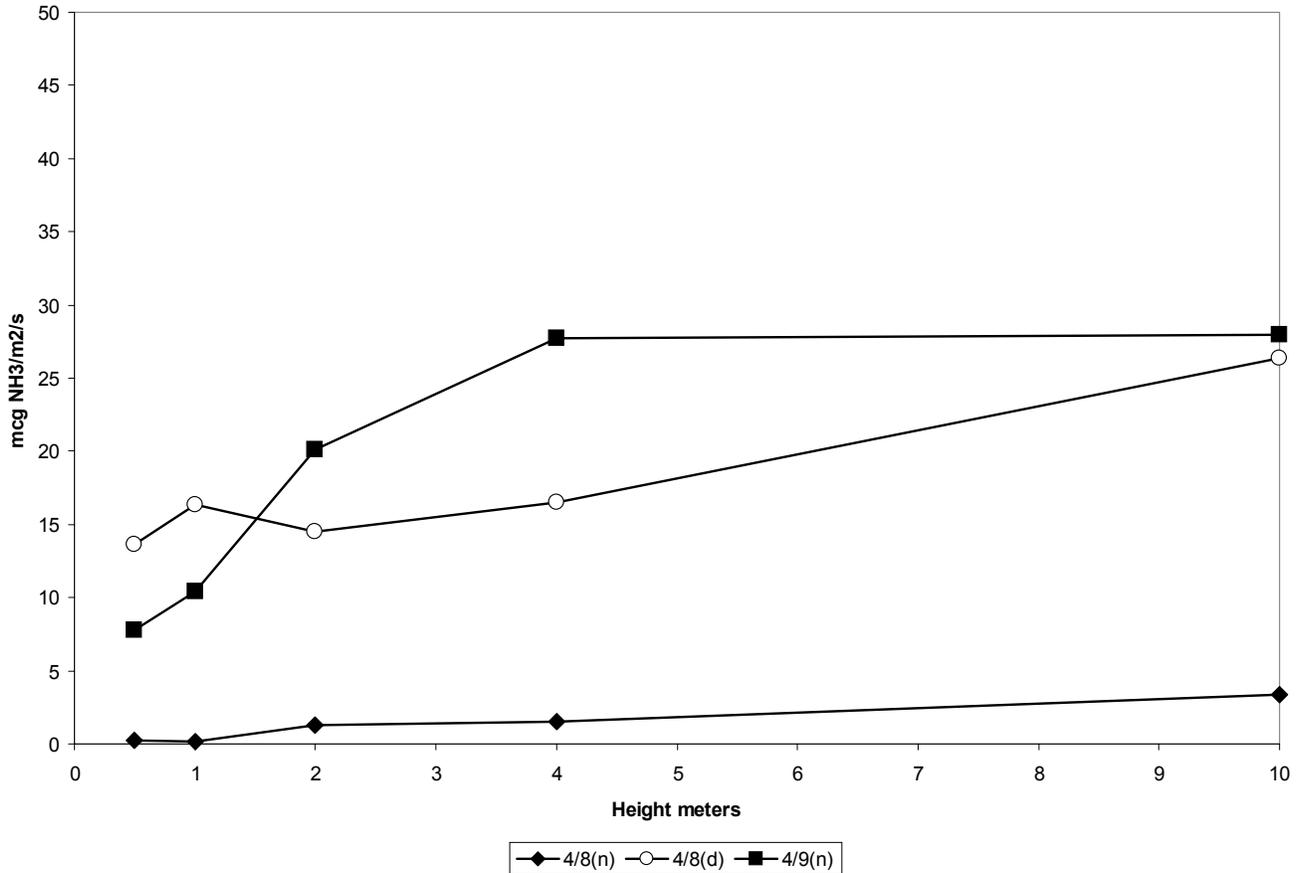
**Figure 2.** Ammonia flux profiles over a barley crop just prior to cutting for silage. Plant height was about 1m. Day time samples are designated by (d) in the legend and an open symbol on the graph. Night data is shown as (n) and a closed symbol.



The two night samples are lower in magnitude and show less profile difference over the sampling heights. The day sample for March 11 is typical of day samples for most of the vegetation types in the study. The magnitude is considerably higher and there is a more pronounced gradient of flux values between the 10m sample and the 0.5m sample. The other day sample, March 8, appears to violate the postulated correlation. Its profile is more characteristic of a night sample. March 11 was a typical spring day in the San Joaquin Valley. The average temperature was 19.9 C, relative humidity was 41%, with a light wind. March 8 was a cool day (13.5 C) near the end of a storm event that dropped 12mm of rain. The air temperature during the storm was more characteristic of night time levels. It is also likely the moisture in the air reduced the NH<sub>3</sub> values. Increased atmospheric moisture, either elevated humidity or precipitation, has been linked to lowered NH<sub>3</sub> levels in previous work.

The barley was cut for dairy feed a few days after the dates shown in Figure 2. The field was immediately replanted to corn that emerged in late March. By the second week in April, the corn was about 30cm tall and beginning a period of rapid growth. The sampling system was set up at the same location as in the previous crop of barley.

**Figure 3.** Ammonia flux profiles from a silage corn crop following the barley crop in Figure 2. The plant height was about 40 cm. The sampling coincided with a fertilizer application and an irrigation.

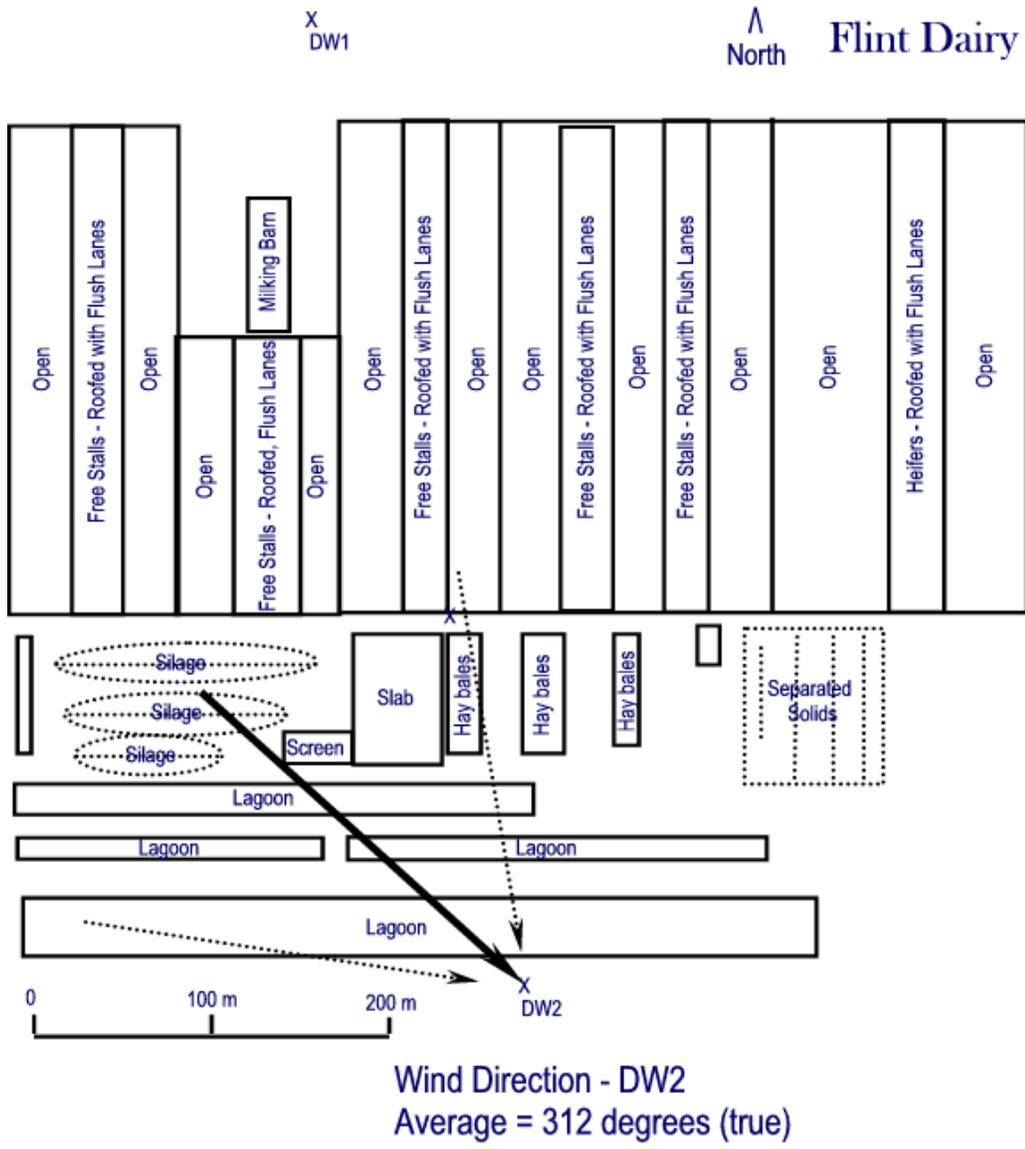


The diurnal differences seen in Figure 2 are also apparent for the April 8 day and night samples in Figure 3. The following night sample, April 9 shows higher flux values and a more pronounced profile than the previous day sample. A heavy application of composted manure was applied to the corn during the day on April 9. The application was followed by an irrigation. The combination of the broadcast compost and the applied water appears to have caused a considerable emission of NH<sub>3</sub> in the field. This pattern is consistent with other fertilizer applications monitored in the previous project of 1999-2000.

The correlation of air temperature and diurnal conditions with NH<sub>3</sub> flux profiles was expected from the experiences of previous research. Additional data from another season and several other crops will, hopefully improve the precision of the relationships shown above. The simultaneous NH<sub>3</sub> emissions and absorption by active vegetation that was suggested in Harper's research also appears to apply to the data collected for this study. Many of the flux profiles shown in the previous figures indicate a gradient of NH<sub>3</sub> from the atmosphere to the soil/vegetation surface. While the transport pathways in the microclimate from the surface up to 10m are likely to be complex and variable for different times of the day and season; it is possible to speculate that the vegetation is acting as a net absorber of NH<sub>3</sub> when the flux profile shows a steep gradient from the ambient atmosphere toward the surface. Both Harper and Porter's work concluded absorption of NH<sub>3</sub> by the plant from the air was related to the amount of atmospheric NH<sub>3</sub> surrounding the plants. Harper<sup>8</sup> noted several factors that sometimes correlated with absorption but stated the concentration of NH<sub>3</sub> in the air was the most consistent factor affecting NH<sub>3</sub> absorption and found it would supersede the other factors. Atmospheric NH<sub>3</sub> concentrations found by Harper to result in measurable absorption were less than most of those monitored at 10m in this study. The ability of a vegetation/soil community to both emit and absorb atmospheric NH<sub>3</sub> may be illustrated by data from another sampling location of this project. A dairy near

Hanford was chosen for a series of ROG samples, though a variety of constituents including NH<sub>3</sub> were collected. A diagram of the dairy is shown in Figure 4.

**Figure 4.** Diagram of a dairy near Hanford, CA. Ammonia flux profiles were monitored at DW1, DW2 and DW3. The arrows show the typical wind direction (solid arrow) and variation (dotted arrows)



The three sampling locations at the Flint Dairy were:

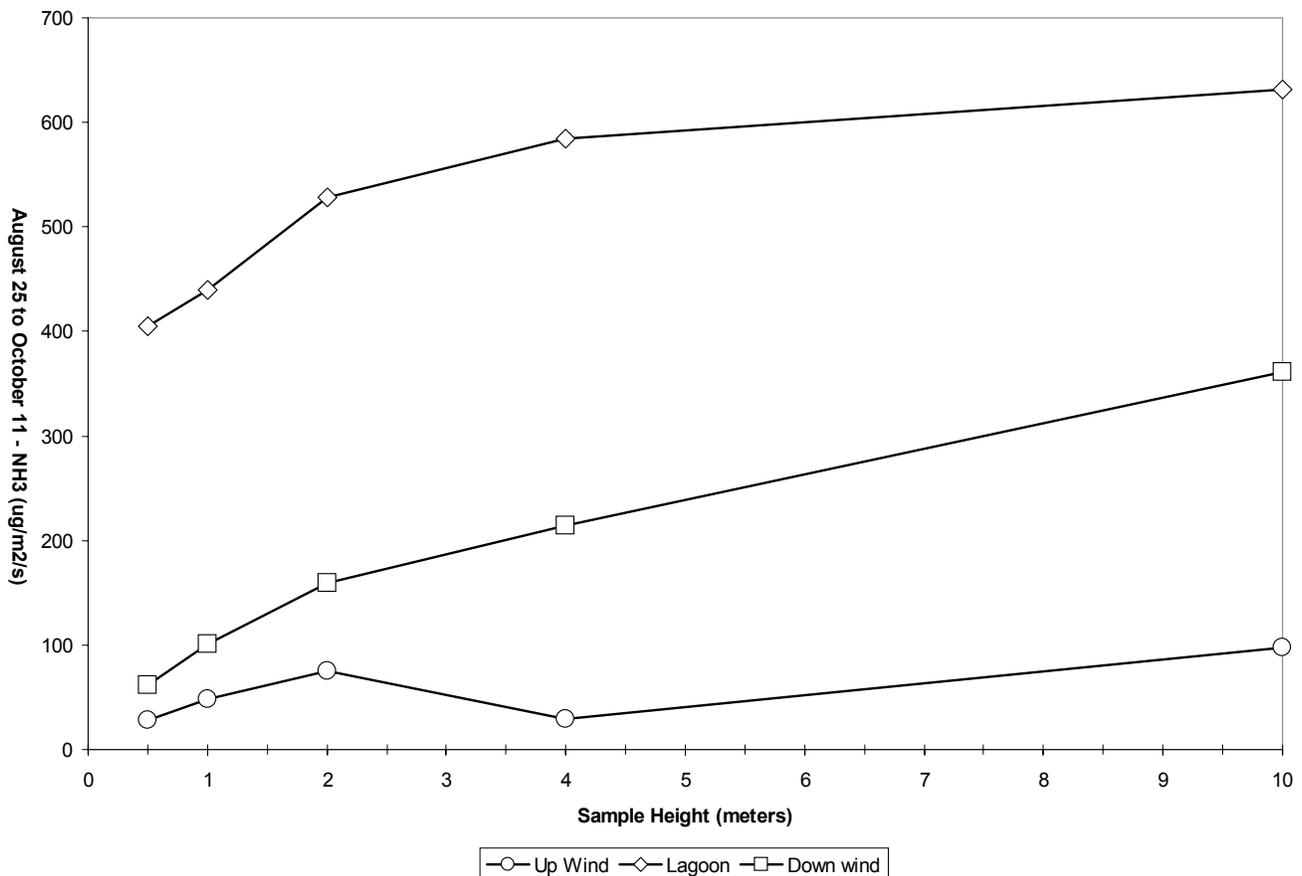
DW1 – upwind of the dairy’s confined animal feeding operation (CAFO). The immediate fetch (400m) upwind from DW1 was a field planted to sorghum. The field was part of the Flint Dairy operation. There were fields of various permanent and annual crops for 3km upwind, at which point there were several other dairies.

DW2 – immediately downwind of the CAFO, along the edge of the main lagoon. The normal wind direction passed through the animal holding/feeding area, the milking barn, feed storage and effluent treatment screens and lagoons before reaching DW2.

DW3 – downwind of the CAFO across 300m of a field planted to the same sorghum crop as that upwind of DW1

The CAFO at the Flint Dairy was surrounded by fields that were planted with the sorghum crop described above. The crop was about a week past emergence (plant height = 10cm) when the sampling began on August 25. The sampling for NH<sub>3</sub> was continuous from that time until October 11 when the crop was harvested (plant height = 150cm) to make silage. The average of the NH<sub>3</sub> flux profiles at each location, for the seven weeks that the crop was grown are shown in Figure 5. Note the magnitude of the flux values on the “Y” axis of the graph. The ammonia levels associated with this CAFO can be several times that of the rangeland and field crops shown in Figures 1, 2 and 3. The sorghum crops surrounding the dairy are grown to feed the dairy herd. Effluent from the free-stall areas where the animals are fed is washed into the lagoons and used to fertilize and irrigate the surrounding crops. The nutrients in the effluent are recycled by harvesting the crops as feed for the animals

**Figure 5.** Ammonia flux profiles from the dairy shown in Figure 4. This data is the average of continuous sampling from late August when the surrounding crops were planted until early October when the crops were harvested.

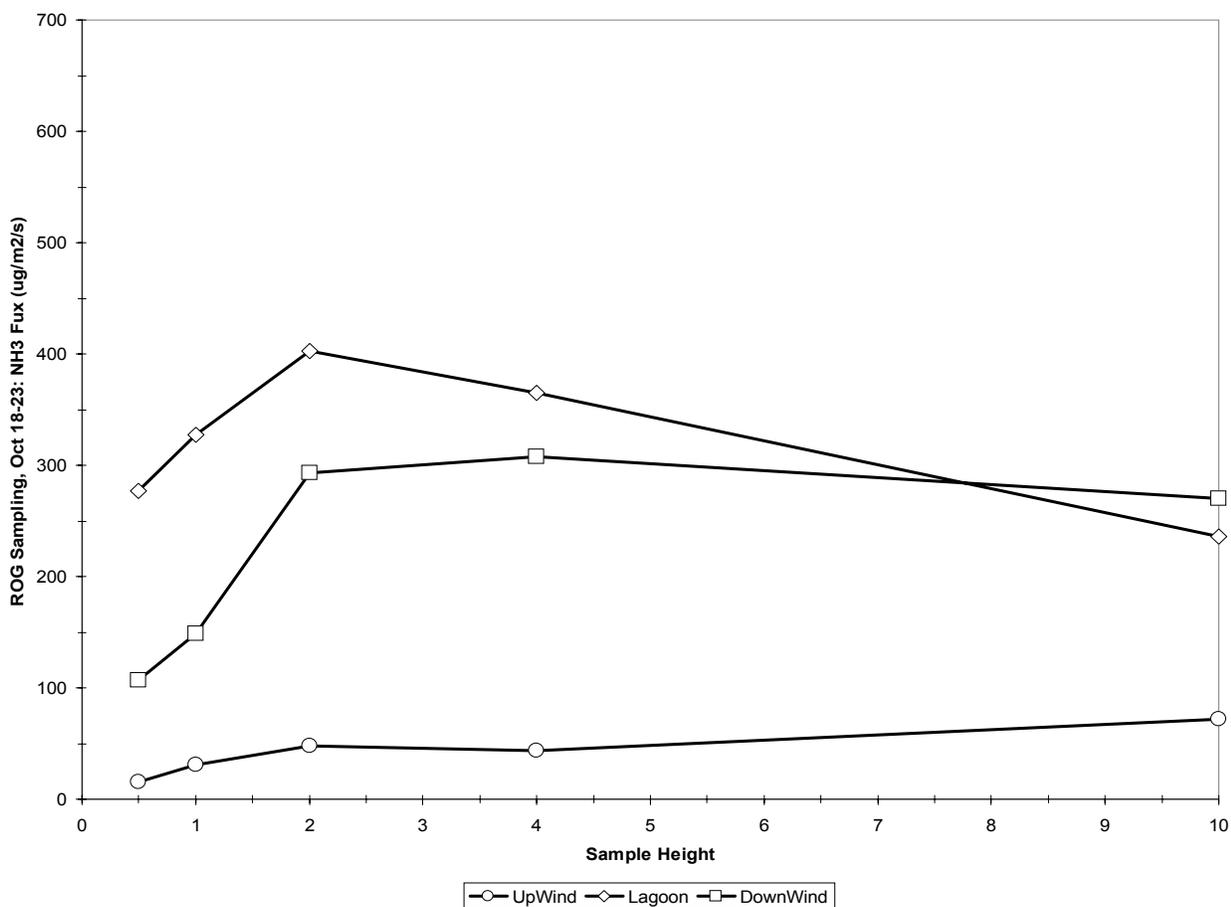


The NH<sub>3</sub> flux profile for the DW1 (upwind location) resembles those found for similar crops in central California. The sorghum is not unlike the barley and corn crops shown in Figures 2 and 3; and the flux profiles are similar in both magnitude and shape. The Lagoon sampling location, DW2, shows a significant increase in NH<sub>3</sub> magnitude, which is to be expected from the 2000 Holsteins located between the two sampling points. Most of the NH<sub>3</sub> is probably from catalysis of urea by urease in the soil of the free stall and open areas of the CAFO. It is also possible that some NH<sub>3</sub> emission comes from other

areas of the dairy but emission from the main lagoon is not indicated by the flux profile at DW2. The sampling site was located 5m from the downwind edge of the lagoon. Any significant NH<sub>3</sub> emission from the lagoon should produce a positive flux gradient from the lowest sampling level toward the ambient atmosphere sampled at 10m. The flux levels at 0.5m and 1.0m are about 2/3 of those at 4m and 10m. The short fetch across the 30m width of the lagoon is not likely to affect the flux at the higher sampling elevations but should influence the NH<sub>3</sub> concentration in the air at 1m and below. The fact that the surface fluxes are significantly less than those found higher may indicate the water at the surface of the lagoon is absorbing NH<sub>3</sub> emitted by the animals further upwind.

Absorption of NH<sub>3</sub> from the air by a surface across which the air passes may also be indicated by the NH<sub>3</sub> flux profile at DW3, the sampling site downwind from DW2. The typical wind direction as shown in Figure 4 is directly from DW2 to DW3. The primary influences on the NH<sub>3</sub> profile between those two points would be various dispersion mechanisms in the atmosphere and emission/absorption of NH<sub>3</sub> by the surface. The magnitude of the DW3 NH<sub>3</sub> flux at 10m is over six times greater than that sampled at 0.5m and the flux gradient is remarkably uniform with elevation. It would be difficult to suggest significant NH<sub>3</sub> emission took place as the air traveled 300m from DW2 to DW3. The NH<sub>3</sub> level at 10m decreases by about half between DW2 and DW3, most likely due to atmospheric dispersion. The fluxes close to the surface, however, are nearly down to those monitored at the upwind site, DW1, which could be considered background. The surface over which the air passed from DW2 to DW3 was the sorghum crop. The high ambient NH<sub>3</sub> levels would, according to Harper, indicate the absorption of atmospheric NH<sub>3</sub> by the leaves of the vegetation. The strong flux gradient in Figure 5, at DW3, from the ambient atmosphere to the vegetation surface supports that indication.

**Figure 6.** Ammonia flux profiles associated with the dairy in Figure 4. These samples were taken after the sorghum crops surrounding the dairy were harvested. The fields were disked and the soil was dry.



Further sampling at these sites was done after the crops surrounding the dairy were harvested. The fields were disked after cutting and picking up the sorghum. The resulting surface was relatively smooth and dry through the following sampling period from October 18 to 23. The field procedure was primarily designed to collect dairy ROG samples for this period. However, the  $\text{NH}_3$  sampling systems were still in place to provide wind speed and direction data so denuders were also used to determine  $\text{NH}_3$  profiles to compare with the ROG results. The  $\text{NH}_3$  flux profiles for the three locations of this later sampling program are shown in Figure 6. The results of the  $\text{NH}_3$  sampling can be compared with those shown in Figure 5 to evaluate the difference between flux profiles over a crop and profiles over bare soil.

The  $\text{NH}_3$  data from this ROG sampling program is not precisely comparable to that from the previous seven weeks of sampling even though the locations and sampling equipment were the same. The ROG samples and corresponding  $\text{NH}_3$  samples were taken during selected, 2-hour periods when the wind was consistently from the direction indicated in Figure 4 at a reasonably consistent speed. The data in Figure 6 was collected from mid-day through late evening on each of three days (October 18, 21 and 23). The flux profile differences between the sampling locations can still be evaluated and compared, in a general way, with those from the previous period when the crop was growing.

The flux profile at the upwind location, DW1, is very similar to that from the preceding several weeks. The field immediately upwind from DW1 was cut along with the others surrounding the dairy but most of the land upwind from that field still had active vegetation. It would appear that the presence or absence of a crop immediately upwind from the dairy had little effect on the upwind profile as long as there were other fields of vegetation further upwind. The lagoon flux profile was different from the average at DW2 during the previous seven weeks. The lower flux values at 10m may be due to the lower temperature in mid October compared to that of August and September in the area. The differences in sampling procedure may also have affected the measured profile at DW2 in Figure 6. The profile at DW3, across the harvested field from DW2 is similar in magnitude and characteristics to DW2. The DW3 flux profile was much closer to the background levels found at DW1 in the sampling during the crop season so it would appear the change from active vegetation to bare soil between DW2 and DW3 may have affected the profile. If  $\text{NH}_3$  absorption by the growing crop had a significant effect on the profile at DW3 then that effect should have disappeared during the later sampling period shown in Figure 6, as it seems to have done. There is still a gradient toward the surface in both profiles below 2m that may indicate absorption by the lagoon surface as postulated from the data in Figure 5 and absorption by the soil surface between DW2 and DW3. Dry deposition of  $\text{NH}_3$  on soil and other surfaces is a process that has been suggested in previous research. The soil surface between DW2 and DW3 may also be capable of  $\text{NH}_3$  absorption but it appears that it is not as effective as an actively growing crop. A positive gradient of  $\text{NH}_3$  fluxes from the surface to the atmosphere was expected in this data since the decomposition of the residue from the harvested crop should release considerable  $\text{NH}_3$ . The lack of that expected gradient is most likely due to the fact that the ROG sampling followed the harvest by only a week. The soil microbes require a period of time to respond to a change in environment such as that caused by harvest of the crop and so were not yet rapidly decomposing the residue.

## CONCLUSIONS

No definite conclusions can be drawn from the data at this point in the study. A number of the initial assumptions appear to be on the way to confirmation and the significance of some others may be greater than was originally assumed. It certainly appears that atmospheric  $\text{NH}_3$  is higher during the day compared to night. The increase in  $\text{NH}_3$  emissions with higher air temperatures suggested by Harper and others is also consistent with the data collected in this research. The balance between  $\text{NH}_3$  emissions and absorption, particularly in relation to actively growing vegetation is less clear. The characteristic shape of most flux profiles in this and the previous work done in the San Joaquin Valley suggests a net absorption of  $\text{NH}_3$  near the surface. Knowledge of the N cycle in effect, particularly for

cultivated, fertilized crops would suggest there is a significant amount of  $\text{NH}_3$  produced by soil microbes. The fate of the  $\text{NH}_3$  produced in the soil is complex and not well documented but emission of a portion of it to the atmosphere is almost a certainty, as modeled by CASA. The intriguing suggestion, originally from Harper and others is the fact that  $\text{NH}_3$  absorption by vegetation is as viable a fate for atmospheric  $\text{NH}_3$  as is the hydrolysis by rain and dew, dry deposition, and the combination with  $\text{NO}_x$  and  $\text{SO}_x$  to form secondary  $\text{PM}_{2.5}$  particles; the reason this study was commissioned.

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