

Ammonia Flux Profiles and ROG sampling at California Dairies

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ABSTRACT

Three dairies in the Central Valley of California were selected to be sampled for ammonia flux profiles and Reactive Organic Gasses (ROG) from the fall of 2002 through the spring of 2004. Sampling locations at each dairy included an upwind site and sites downwind of significant dairy operations such as the animal containment, feed storage areas, the lagoon and surrounding cropland. Ammonia was sampled using active denuders co-located with wind instruments from the soil surface to a height of 10m. Some indication of NH₃ absorption by active vegetation was found under circumstances similar to those reported by other researchers. The ROG and methane samples were collected over a period of two hours in canisters and analyzed by GCMS and GCFID. The ROG sample data was used to evaluate a dispersion model to calculate emission fluxes from the dairies. Preliminary results from the first samples analyzed with the model are slightly less than current emissions estimates used in California. The work will continue through 2004 and final results will be available in about a year.

Introduction

An air quality dilemma is developing in the Central Valley of California. The process of meeting federal air quality standards has been complicated by state legislation with higher standards and accelerated timelines that will be difficult for the state's agricultural industry to meet. A significant part of the problem is the fact that the actual emissions from operations that are assumed to be among the largest sources, i.e. dairies, are not well documented. Some components of the existing emissions inventory are based on research of questionable relevance to California conditions. Of particular concern are emission factors for the particulate matter precursor ammonia and the long list of ozone precursors known as reactive organic gasses (ROG) from the wide variety of agricultural practices in the state. The need for "sound science" upon which to base emission factors needed for the baseline inventory has only recently become apparent to the state and local air quality agencies and the agricultural industry. Dairy production is one of the most valuable components of California agriculture. Dairy operations are also estimated to be the largest agricultural source of ROG and a major ammonia source based on current and possibly questionable emission factors. The uncertainty regarding the existing emissions along with the magnitude of the estimated values is the heart of the dilemma that will only be resolved by research to either confirm the current factors or produce better ones.

Air quality research has been conducted at the CSU Fresno Center for Irrigation Technology since 1998. The initial project was a study of ammonia emissions related to commercial fertilizer applications to various crops in California. The study was a cooperative effort funded by the California Air Resources Board to provide data for their ammonia inventory. CSU Fresno was responsible for the collection of field data and Chris Potter of the Ames Research Center – NASA modeled regional emission factors from the field data. Results from that project were presented at this conference in 2002 and the project report was submitted that same year. A second project, supported by the ARB was begun in 2001 to sample ambient ammonia levels related to various crops and natural vegetation to establish variability due to seasonal weather, soil type and other environmental factors. Some results of that study were presented at this conference last year. The rise in interest related to dairy emissions has driven the priority of this research project toward the development of methods to sample and model ROG and ammonia emissions from the wide variety of agricultural operations associated with milk production and the disposal of waste from dairies and the related crop production operations. The application of the ammonia sampling methodology developed for the previous projects was easily converted to the sampling of ammonia at

dairies. The development of collection and analytical methods for ROG has proven to be much more difficult. The modeling of emission factors from the collected field data was the capability that required the most development. The process used in the previous fertilizer project was not applicable so a new method had to be found and validated. This paper will focus on the selection and validation of methods to sample ROG and model emission factors based on those samples. These sampling/modeling procedures will then be used to determine ROG and ammonia emission factors and the effect on them of seasonal weather and changes in dairy practices.

The ammonia sampling methodology was developed for the previous studies of fertilizer application and ambient ammonia fluxes from various vegetation covers. It consists of a filter treated with citric acid (5% in ethanol) through which air is drawn at 1 to 5 liters/minute by a pump. These sampling units are battery powered and mounted at various heights on a 10m mast. The sampling period varies from 2 to 24 or more hours. The sampling periods for the data presented here were either 2 hours to match the ROG sampling periods or about 12 hours, diurnally, to monitor daily variability. The filters are taken to the lab, stored under refrigeration and analyzed for ammonia by a simple spectro-photometric method. The method is described in more detail in the papers presented at the '02 and '03 conferences (Krauter, 2002 and Krauter, 2003). The ROG sampling was done in 6 liter summa canisters according to published methods (EPA-TO-15, 1999). The analysis of the samples discussed in this paper was done at the Oregon Graduate Research Institute by Dr. R. Rasmussen. Subsequent samples are being analyzed at CSU Fresno by gas chromatography (Varian Saturn CP3800) with a cryogenic concentration unit and a mass spectrometer (Varian 2100T) according to published methods (EPA-TO-15, 1999).

Model Selection

The model selected for this project was the Industrial Source Complex Short Term version 3 (ISC-STv3). It is a steady state Gaussian plume model that can be used to predict downwind concentration from area sources (EPA, 1995). ISC-STv3 is used to calculate 1-hour average concentrations at receptor locations placed anywhere around the source. The traditional inputs for the model include the relative placement of sources and receptor locations, as well as recorded meteorological conditions and known emission fluxes. The output from the model is the predicted concentration at the selected receptor locations. In order to predict concentrations from area sources, the source is broken into multiple finite-length line sources. The finite-length line source equation results from the integration of the point source equation in the crosswind direction as shown in equation 1.

$$\text{Equation (1)} \quad C = \frac{Q_A}{2u\pi} \int_x \frac{V}{\sigma_y \sigma_z} \left(\int_y \exp \left[-0.5 \left(\frac{y}{\sigma_y} \right)^2 \right] dy \right) dx$$

where

- C = concentration of pollutant ($\mu\text{g}/\text{m}^3$)
- Q_A = pollutant emission rate ($\mu\text{g}/\text{s}\cdot\text{m}^2$)
- V = vertical term used to describe vertical distribution of the plume
- σ_y, σ_z = Pasquill-Gifford plume spread parameters based on stability class
- u = average wind speed
- X = upwind direction
- Y = cross wind direction

The integration of this equation results in equation 2.

$$\text{Equation (2)} \quad C = \frac{2q}{\sqrt{(2\pi)\sigma_z u} } \left\{ \exp \left[-\frac{(z-H)^2}{2\sigma_z^2} \right] + \exp \left[-\frac{(z+H)^2}{2\sigma_z^2} \right] \right\} (G_2 - G_1)$$

where

C = concentration of pollutant ($\mu\text{g}/\text{m}^3$)

q = emission rate ($\mu\text{g}/\text{m}/\text{s}$)

σ_z = Pasquill-Gifford plume spread parameter based on stability class (m)

u = average wind speed at pollutant release height (m/s)

H = emission height (m)

Z = Receptor height (m)

G_2, G_1 = Gaussian distribution function to account for edge effects

The resulting equation is the infinite length line source equation with a correction for the edges of the source. An infinite line source placed perpendicular to the wind direction has no change in concentration with crosswind movement. A finite length line source must account for the edges of the source where concentration does diminish with crosswind movement. Therefore the correction for edge effects is a function of the crosswind distance from the end of each line source to the receptor (Y) and the horizontal plume spread parameter (σ_y). Since the horizontal plume shape is represented by a normal distribution, equation 3 can be used to determine the fractional portion of the area under a normal curve. This value is used as a scalar to decrease the predicted concentration.

$$\text{Equation (3)} \quad G = \frac{Y}{\sigma_y}$$

where:

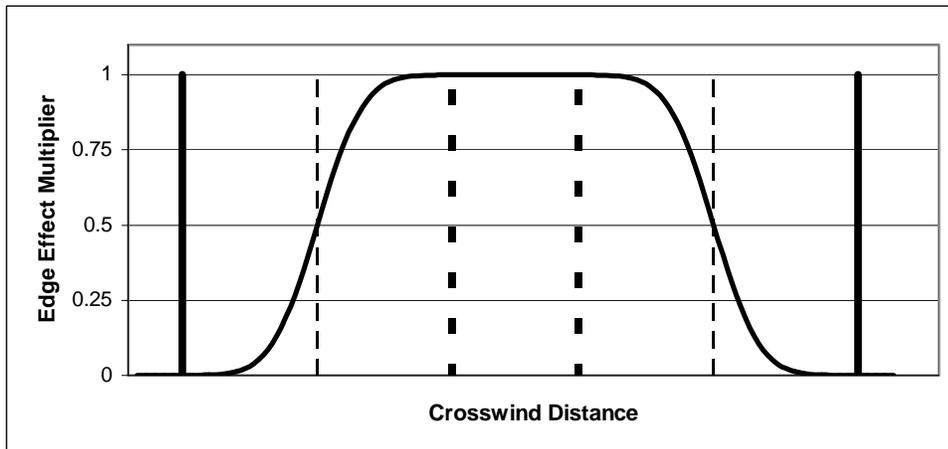
G = number of standard deviations

Y = crosswind distance between receptor and edge of line source

σ_y = horizontal plume spread standard deviation

The edge effect phenomenon is due to the horizontal dispersion of the pollutant. When the receptor is more than $3.8 \sigma_y$ from the edge of a line source, the edge effects are accounted for in ISC-STv3. The result of edge effects produces the plume shape shown in figure 1.

Figure 1. The magnitude of edge effects in the crosswind direction is shown here. The dashed lines represent a position directly downwind of the end of the line source. The peak concentration is reached at a distance of $3.8 \sigma_y$ towards the center of the field. There is no contribution to the concentration when the receptor is placed greater than a distance of $3.8 \sigma_y$ outside the edge of the field.



This process of dividing the area source into multiple line sources is similar to the algorithm used in the Point Area and Line Sources 2.0 model (PAL) (EPA, 1995). The summation of the predicted concentration from each line source is the resulting concentration at the receptor location. In order to determine the modeled concentration at any receptor location, the number of line sources used is increased until the predicted concentration with N line sources converges with that predicted with N-1 line sources. The difference between ISC-STv3 and PAL is the method used for evaluation of the integral as well as the specific limits used to determine convergence. These changes were made in order to optimize the computing time used to determine the concentration (EPA, 1995) but yield the same results. ISC-STv3 can also handle more variations in the configuration of area sources while PAL was limited to strictly North-South East-West orientations (Peterson and Rumsey, 1987). The method used by ISC-STv3 also allows for the placement of receptors at any location in or around area sources. The only limitation on placement of receptors is the upwind distance to the nearest line source. The limitation is due to the calculation of the σ_z parameter. When the upwind distance from source to receptor approaches zero, this value approaches zero, yielding inconsistent results. Therefore, ISC-STv3 limits the minimum downwind distance, from source to receptor, to 1 meter.

The Gaussian model is typically used to determine a concentration at a given location due to a known emission flux. In this case, the concept is used in reverse; we want to determine an emission flux, from a known concentration. Due to the fact that equation 1 cannot be solved directly for flux, it is necessary to use a multi-step process. This is done using the proportional nature of flux, Q_A , to the predicted concentration. The goal is to determine a flux that will predict the measured net concentration when used in ISC-STv3 with the actual meteorological conditions present during sampling. The process can be most simply described for a single area source. First, the source receptor orientation is modeled using an emission flux unit (EFU) of $1 \mu\text{g}/\text{m}^2/\text{s}$. Then the ratio of the modeled concentration to the measured net concentration is used to scale the EFU. If $1 \mu\text{g}/\text{m}^2/\text{s}$ is chosen as the initial EFU, the required flux to match the measured concentration will be equal to the ratio of measured to predicted concentration. The units of the flux can then be adjusted to those required for the given operation.

In the case of multiple area sources with different emission fluxes it is necessary to have a receptor for each area source emitting at a different rate. Each receptor is then used to determine an emission flux for an individual area source. The dairy used as an example here has two identified sources with different fluxes (see figure 2). There were three sampling locations setup to handle this configuration. Location DW1 was setup as a background receptor, upwind from the dairy to determine ambient concentrations of the target pollutants. The DWx sampling site was directly downwind of the freestall and exercise pens. The DW2 site was downwind of the large lagoon. The first step in determining the two different emission fluxes was to calculate the flux from the freestall areas as described above. In this case the freestalls and exercise pens were assumed to have uniform fluxes. Once the flux from the freestall/exercise pens was determined, it could be used to calculate its contribution to the receptor at the edge of the lagoon. The net concentration at the edge of the lagoon was determined by subtracting the upwind concentration and the freestall/exercise pen concentration from the measured concentration at the lagoon site. The same process could then be used to determine the flux from the lagoon.

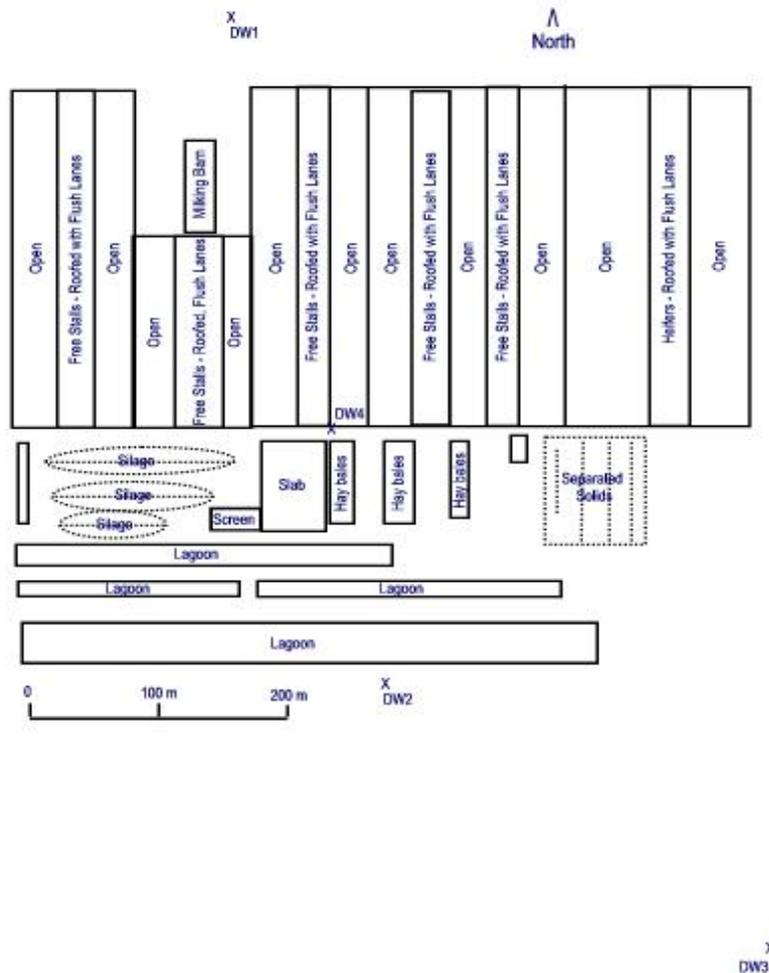
Dairy Sampling

This dairy was one of three selected for a series of ammonia and ROG samples that began in October of 2002. The dairies were chosen to be medium sized, about 2000 head, built or renovated at least 3 years but no more than 10 years ago and equipped with the usual free stall management system. Free stall dairies have feeding lanes and bedding stalls that the animals move into by choice. The free stalls are arranged in long, sloping concrete surfaces that can be flushed several times each day to remove waste. The waste is flushed into a pit to allow sand to separate then the light solids are separated from the liquid, usually by a screen system. The separated solids, primarily undigested fibers from the animal feed, are dried and used as either bedding in the free stalls or soil amendments on surrounding cropland. The liquid from the separation process still contains a considerable load of very fine solids. It usually goes to

another settling basin and then to a large storage lagoon. Water from the lagoon is used for subsequent flushing cycles and will eventually become irrigation water on adjacent cropland to recycle the nutrients.

In addition to the free stalls which are usually covered for shade in summer and warmth in winter, the animals have a larger, open exercise area that is usually bare soil. This particular dairy is near Hanford in Kings County, CA. It and two others in Fresno and Merced counties have been sampled every three months beginning in the summer of 2003. The objective is to determine seasonal variability for emission fluxes of ammonia and ROG for average sized dairies with the most common waste management systems.

Figure 2. A diagram of the dairy near Hanford, CA where ROG and ammonia samples were taken for modeling of emission fluxes.



The initial sampling of this dairy was done in October of 2002 as a trial of the sampling systems. At the time it was selected, no model had been chosen. This monitoring of ammonia and ROG differs from later work in that the sampling equipment was rented and the analysis was done at an outside lab. The research objectives included the development of a sampling protocol, acquisition of equipment and the establishment of the analytical capability to handle the samples in our own laboratory. All of those objectives have been met but the analytical system has only recently begun to produce usable results. Consequently, the modeling has been limited to the data from the initial sampling in October, 2002.

Since no model had been selected, the location of sampling sites for this first attempt was changed for each of the three days of sample collection to anticipate the needs of a variety of possible models. There were four sampling periods of two hours on each of the three days. Each period included an upwind sample (DW1) and a sample at the downwind edge of the dairy where the lagoon was located (DW2). A third sample was taken but its location was different for each day. On the first date, October 18, the third sample was co-located with DW2 but at 10m instead of the usual 2m height. On the second

sampling date, October 21, the third sample was located in the center of the dairy downwind of the free stall area but upwind from the lagoon. That proved to be the proper location for the ISC-STv3 modeling program. The third sampling date was October 23 and the third sampling site was 300m downwind from the lagoon across an uncropped field. Ammonia samples were taken at the same time and location as the ROG canister samples. Data from the ROG samples with the results of the modeling are shown in Table 2 for the emissions from the free stall and exercise areas and in Table 3 for the lagoon. Table 1 shows the sampling dates, weather conditions and the ambient concentrations of methane and ROG or total non-methane hydrocarbons (TNMHC) from the DW1 sampling site.

Table 1. Sampling dates, meteorology and ambient concentrations of methane and ROG.

Sampling Periods			Upwind Concentrations					Meteorological Conditions			
Date	Start	End	CH ₄ ppbv	Degree F	Degree K	CH ₄ µg/m ³	TNMHC µg/m ³	Wind Speed (m/s)	WD degrees	WD Stdev	Stability Class
18-Oct-02	1243	1416	2283	67.4	292.8	1520.1	60.2	1.41	305.5	79.1	2
18-Oct-02	1554	1754	2410	72.0	295.3	1591.0	54.5	0.94	306.1	25.9	4
18-Oct-02	1815	2005	2819	63.7	290.8	1890.4	107.8	0.76	322.0	11.0	6
18-Oct-02	2030	2225	2662	59.9	288.7	1798.0	78.0	1.19	323.2	8.1	6
21-Oct-02	1248	1440	2375	74.5	296.8	1560.4	44.5	1.26	343.5	29.9	2
21-Oct-02	1522	1722	2178	75.5	297.3	1428.3	51.4	2.20	320.0	14.0	4
21-Oct-02	1737	1940	5171	67.4	292.8	3443.1	139.7	0.95	312.1	5.3	6
21-Oct-02	2005	2155	2721	62.1	289.9	1830.4	75.7	0.23	318.4	5.1	6
23-Oct-02	1205	1347	2162	65.8	291.9	1443.9	82.5	1.22	344.2	25.7	2
23-Oct-02	1428	1630	2144	69.9	294.2	1420.8	46.9	0.77	311.0	27.1	2
23-Oct-02	1737	1940	2316	60.9	289.2	1561.4	63.1	1.74	288.3	5.7	6
23-Oct-02	2010	2210	2302	56.6	286.8	1565.0	54.5	2.01	312.3	8.3	6

Table 2. Modeled emissions of methane and ROG (TNMHC) from free stalls and exercise areas.

Sampling Periods			Free Stall / Exercise Areas Sampled DW4 Concentrations				Modeled Concentrations		
Date	Start	End	CH ₄ µg/m ³	TNMHC µg/m ³	Net CH ₄ µg/m ³	Net TNMHC	Free Stall µg/m ³	Exerc. µg/m ³	Modeled Conc
21-Oct-02	1248	1440	2444.3	173.2	883.9	128.7	154.0	72.3	113.2
21-Oct-02	1522	1722	3913.5	234.3	2485.1	182.9	174.3	114.0	144.2
21-Oct-02	1737	1940	11969.7	402.9	8526.6	263.2	693.1	552.1	622.6
21-Oct-02	2005	2155	9601.2	227.2	7770.8	151.5	734.1	513.7	623.9

Sampling Periods			Methane Emissions						
Date	Start	End	Measure/Mod eled	µg/m ² -s Freestall	µg/m ² -s Exerc.	kg/day	kg/1000hd- day	kg/hd/yr	lb./hd/yr
21-Oct-02	1248	1440	7.8	39.1	39.1	295.2	147.6	53.9	118.5
21-Oct-02	1522	1722	17.2	86.2	86.2	651.6	325.8	118.9	261.6
21-Oct-02	1737	1940	13.7	68.5	68.5	517.6	258.8	94.5	207.8
21-Oct-02	2005	2155	12.5	62.3	62.3	470.7	235.4	85.9	189.0

Sampling Periods			Reactive Organic Gas Emissions Measured as Total Non-methane Hydrocarbons (TNMHC)						
Date	Start	End	Measure/Mod eled	µg/m ² -s Freestall	µg/m ² -s Loaf	kg/day	kg/1000hd- day	kg/hd/yr	lb./hd/yr
21-Oct-02	1248	1440	1.1	5.7	5.7	43.0	21.5	7.8	17.3
21-Oct-02	1522	1722	1.3	6.3	6.3	48.0	24.0	8.8	19.3
21-Oct-02	1737	1940	0.4	2.1	2.1	16.0	8.0	2.9	6.4
21-Oct-02	2005	2155	0.2	1.2	1.2	9.2	4.6	1.7	3.7

Table 3. Modeled emissions of methane and ROG (TNMHC) from the lagoon and downwind edge of the dairy.

Sampling Periods			Free Stall / Exercise Areas Sampled DW4 Concentrations				Modeled Concentrations		
Date	Start	End	CH4 $\mu\text{g}/\text{m}^3$	TNMHC $\mu\text{g}/\text{m}^3$	Net CH4 $\mu\text{g}/\text{m}^3$	Net TNMHC	Free Stall $\mu\text{g}/\text{m}^3$	Loaf $\mu\text{g}/\text{m}^3$	Lagoons
21-Oct-02	1248	1440	3532.4	63.5	1972.0	19.0	15.9	28.4	126.8
21-Oct-02	1522	1722	4311.9	98.6	2883.6	47.2	16.7	28.7	153.9
21-Oct-02	1737	1940	N/A	N/A	N/A	N/A	N/A	N/A	N/A
21-Oct-02	2005	2135	21937.5	251.3	20107.1	175.6	63.3	126.5	631.8
23-Oct-02	1205	1347	1484.2	72.0	40.3	-10.5	16.5	29.6	131.0
23-Oct-02	1428	1630	1487.2	56.8	66.4	9.9	7.0	11.7	158.0
23-Oct-02	1737	1940	2389.2	110.7	827.9	47.6	N/A	N/A	370.5
23-Oct-02	2010	2210	3683.3	97.2	2118.3	42.7	19.7	28.2	319.5

Sampling Periods			Methane Emissions					
Date	Start	End	Measure/ Modeled	$\mu\text{g}/\text{m}^2\text{-s}$	kg/day	kg/1000hd day	kg/hd/yr	lb./hd/yr
21-Oct-02	1248	1440	1.96	19.64	36.64	18.32	6.69	14.7
21-Oct-02	1522	1722	1.73	17.30	32.27	16.14	5.89	13.0
21-Oct-02	1737	1940	N/A	N/A	N/A	N/A	N/A	N/A
21-Oct-02	2005	2135	1.16	11.59	21.63	10.81	3.95	8.7
23-Oct-02	1205	1347	-5.50	-55.05	-102.71	-51.36	-18.74	-41.2
23-Oct-02	1428	1630	-27.14	-271.41	-506.40	-253.20	-92.42	-203.3
23-Oct-02	1737	1940	2.89	28.86	53.85	26.92	9.83	21.6
23-Oct-02	2010	2210	1.97	19.74	36.83	18.41	6.72	14.8

Sampling Periods			Reactive Organic Gas Emissions Measured as Total Non-methane Hydrocarbons (TNMHC)					
Date	Start	End	Measure/ Modeled	$\mu\text{g}/\text{m}^2\text{-s}$	kg/day	kg/1000hd day	kg/hd/yr	lb./hd/yr
21-Oct-02	1248	1440	0.15	1.50	2.79	1.40	0.51	1.1
21-Oct-02	1522	1722	0.31	3.07	5.72	2.86	1.04	2.3
21-Oct-02	1737	1940	N/A	N/A	N/A	N/A	N/A	N/A
21-Oct-02	2005	2135	0.28	2.78	5.19	2.59	0.95	2.1
23-Oct-02	1205	1347	-0.08	-0.80	-1.50	-0.75	-0.27	-0.6
23-Oct-02	1428	1630	0.06	0.63	1.17	0.58	0.21	0.5
23-Oct-02	1737	1940	0.13	1.28	2.40	1.20	0.44	1.0
23-Oct-02	2010	2210	0.13	1.34	2.49	1.25	0.46	1.0

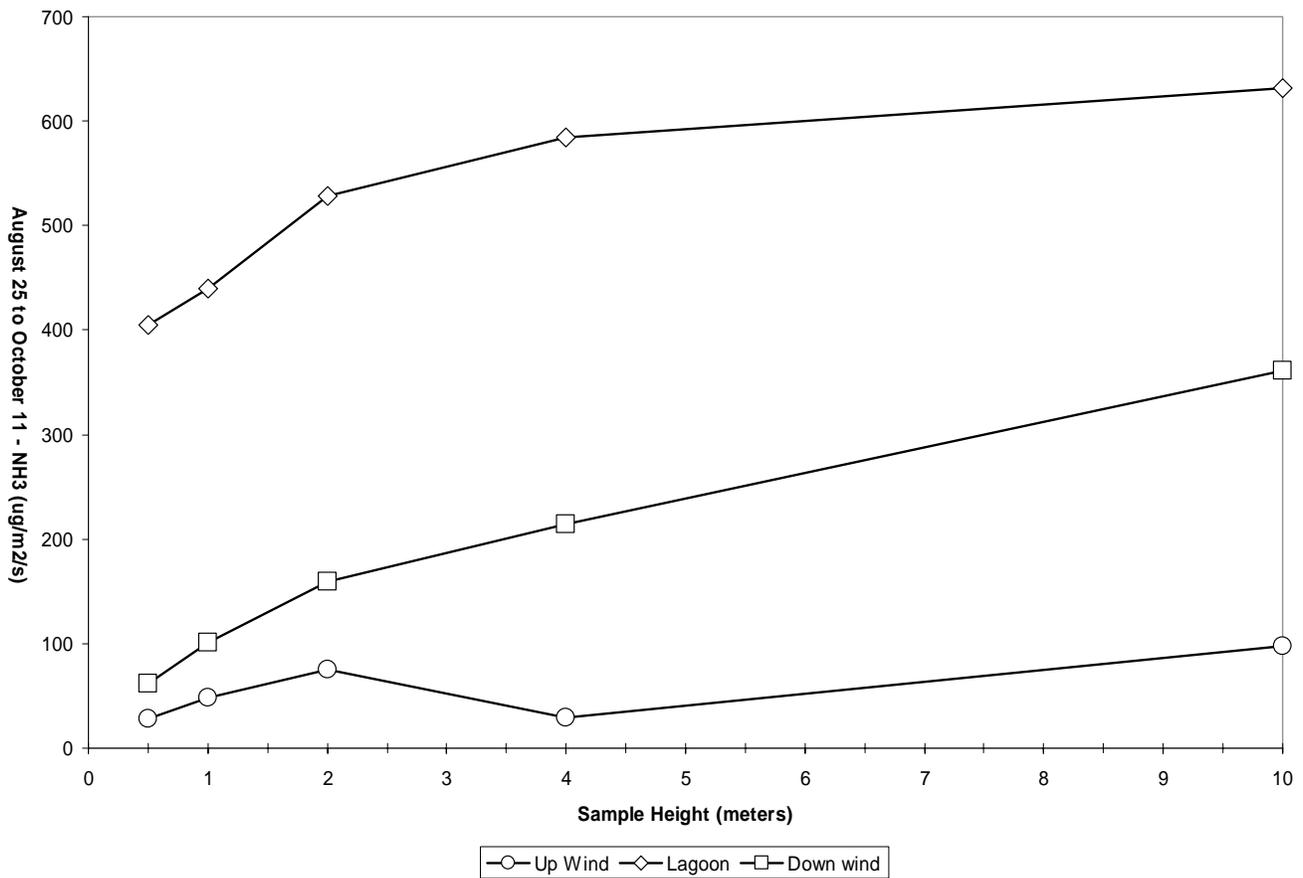
These results should be considered as examples of emissions from one dairy for a single sampling event rather than representing dairy emissions in general. They are presented here only to show the working results of the model that will be used with a succession of samples from this and two other dairies at various times of the year. With that caveat firmly in mind, it is possible to draw some conclusions from these results of the model. The methane emissions for the free stall areas shown in Table 2 are somewhat lower than but still reasonably close to the tentative estimate for dairy cow methane emissions in California, 135 kg./head/year. It may be that methane emissions are higher during the summer and the average of the seasonal samples will be closer to the published values. The methane emissions from the lagoon shown in Table 3 are quite low but are consistent on the October 21 date. The negative values that appear for methane on October 23 are likely to be problems with the model rather

than evidence that the lagoon is a methane sink. The ISC-STv3 model requires an upwind and downwind sample difference to calculate a net concentration. On the 21st, the free stall sampling site, DW4, was ideally located as an upwind lagoon value. There was no sample from DW4 on the 23rd so the ambient site, DW1 was used. The fact that there was another emitter, the free stall area, between the lagoon and its upwind site may have prevented the model from calculating the correct emission for some sampling periods on the 23rd.

The ROG emissions from the model are also interesting and reasonably close to current estimates but should not be construed as anything other than examples. Emissions from the free stalls are near the 5.8 kg./head/year (12.8 lb./head/year) that is currently used by the California ARB as the total ROG emission from a dairy cow. The higher values during the day may correlate with feeding and exercise when animals are more likely to ruminate and excrete waste from which ROG can volatilize. Temperature or other environmental conditions may also be part of the apparent diurnal variability. ROG emissions from the lagoon are significantly lower than those from the free stalls which is contrary to some current estimates. It should be pointed out that the lagoon sampled at this dairy is the third in a series, is very large and is diluted with considerable amounts of fresh water prior to its use as flush or irrigation water. Further sampling and modeling at this and other dairies will be needed before ROG emissions from lagoons can be confidently evaluated.

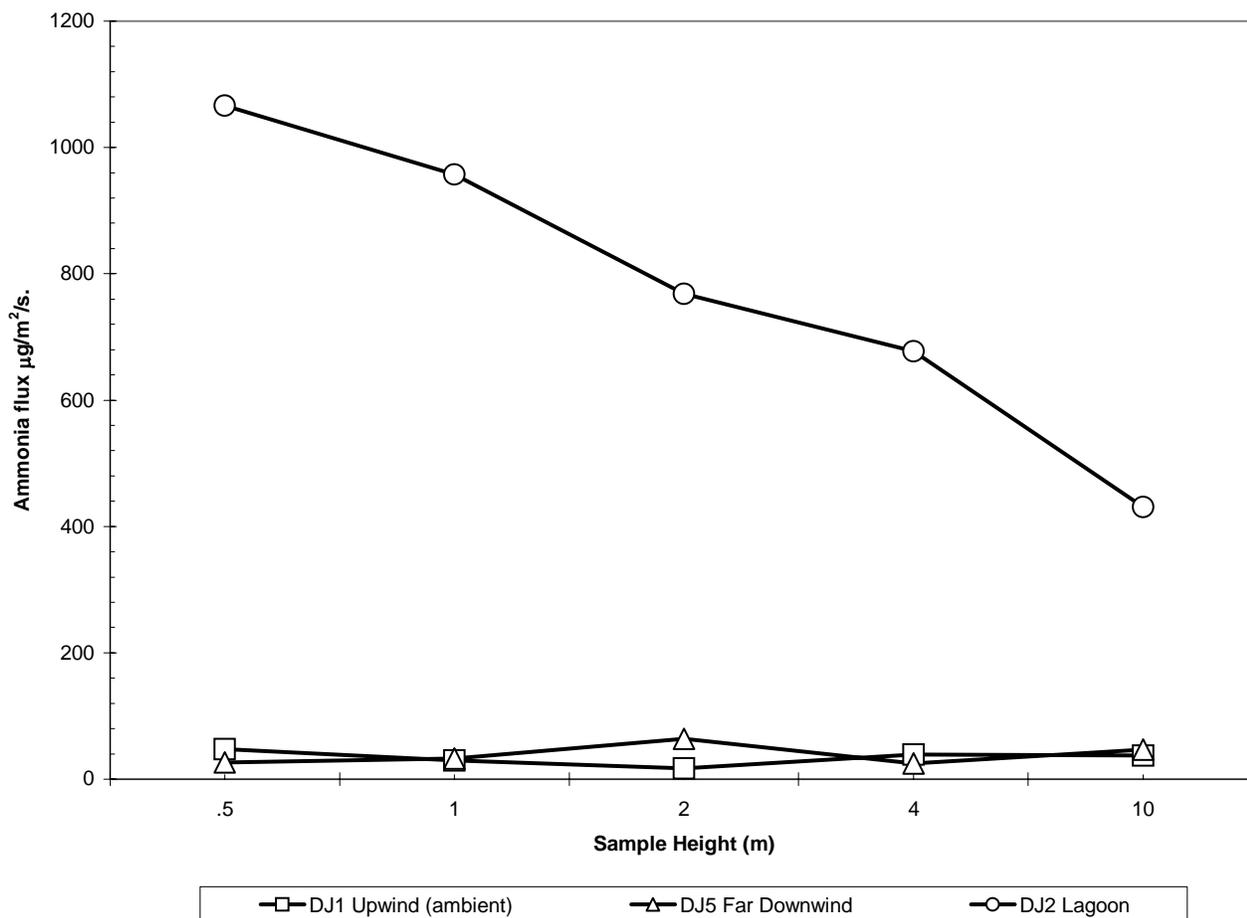
Ammonia data from these sampling periods at this dairy were presented at last year's conference. At that time, there was evidence that the considerable emission of ammonia from the lagoon or other dairy facilities was significantly attenuated by the crop growing in the field downwind of the dairy. That data is shown in figure 3.

Figure 3. 2002 Ammonia flux profiles from the sites DW1, DW2 and DW3, shown in Figure 2.



The downwind sample was 285m from the lagoon across a field of forage that grew from to a height of 1.5m by the end of the 8 weeks of sampling. Further ammonia sampling has been done at this and the other dairies to further investigate the magnitude of ammonia absorption by vegetation. Data from the Merced County dairy for March, 2004 is shown in figure 4.

Figure 4. Ammonia flux profiles for March 12, 2004 at a dairy in southern Merced County, CA. Site DJ1 was upwind of the dairy to measure ambient conditions. Site DJ2 was on the downwind edge of the lagoon. Site DJ5 was approximately 500m downwind of the dairy across a field of mature forage.



The considerable ammonia flux from the lagoon shown as the profile for DJ2 (Lagoon) is completely attenuated by the time the air passes over the 500m of crop surface to DJ5 (Far downwind). The flux values for DJ5 are not significantly different from the ambient values at DJ1 (Upwind). Figure 4 is an example of ammonia monitoring at these sites from March 12 -19, 2004. The data from the other sampling dates was identical to this shown in the figure. Modeling of the ammonia emissions will be attempted using the ISC-STv3 modeling program later this year to further evaluate absorption of ammonia by vegetation.

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Key words:

Emissions Inventory

ROG

Ammonia

Dairy

