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Emissions Calculated from Particulate Matter and Gaseous Ammonia Measurements from a Commercial Dairy in California, USA

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Abstract. *Emission rates and factors for particulate matter (PM) and gaseous ammonia (NH₃) were estimated from measurements taken at a dairy in June 2008. Concentration measurements were made using both point and remote sensors. Filter-based PM samplers and optical particle counters (OPCs) characterized aerodynamic and optical properties, while a scanning elastic lidar measured particles around the facility. The lidar was calibrated to PM concentration using the point measurements. NH₃ concentrations were measured using 23 passive samplers and 2 open-path Fourier transform infrared spectrometers (FTS).*

Emission rates and factors were estimated through both an inverse modeling technique using AERMOD coupled with measurements and a mass-balance approach applied to lidar PM data. Mean PM emission factors \pm 95% confidence interval were 3.8 ± 3.2 , 24.8 ± 14.5 , and 75.9 ± 33.2 g/d/AU for PM_{2.5}, PM₁₀, and TSP, respectively, from inverse modeling and 1.3 ± 0.2 , 15.1 ± 2.2 , and 46.4 ± 7.0 g/d/AU for PM_{2.5}, PM₁₀, and TSP, respectively, from lidar data. Average daily NH₃ emissions from the pens, liquid manure ponds, and the whole facility were 143.4 ± 162.0 , 29.0 ± 74.7 , and 172.4 ± 121.4 g/d/AU, respectively, based on the passive sampler data and 190.6 ± 55.8 , 16.4 ± 8.4 , and 207.1 ± 54.7 g/d/AU, respectively, based on FTS measurements. Liquid manure pond emissions averaged 5.4 ± 13.9 and 3.1 ± 1.6 g/m²/d based on passive sampler and FTS measurements, respectively. The calculated PM₁₀ and NH₃ emissions were of similar magnitude as those found in literature. Diurnal emission patterns were observed.

Keywords. Remote sensing, lidar, emissions, particulate matter, ammonia, point sensor, passive sampler.

Introduction

Air emissions of particulate matter (PM) and gaseous ammonia (NH_3) from agricultural operations can be a significant issue for air quality and human health. They are being increasingly investigated for contributions to air pollution budgets. Some air quality regulatory agencies have begun to require air pollution permits for operations that exceed certain sizes. The accurate quantification of agricultural emissions is an important part of identifying their potential air quality impacts. Toward estimating PM and NH_3 emissions from commercial U.S. dairies, a study was conducted in June 2008 in the San Joaquin Valley of California, U.S. The $\text{PM}_{2.5}$, PM_{10} , and TSP concentrations and emissions were characterized using point samplers and an elastic lidar, as reported by Marchant et al. (2011). NH_3 concentrations and emissions were measured using passive samplers and open-path Fourier transform infrared spectroscopy (FTS). Results of these measurements and emissions calculations are herein reported.

Measurements and Methods

The 22.6 ha dairy, shown in Figure 1, was located near Hanford, California and surrounded by agricultural land. There were approximately 1,885 animals on the dairy, totaling 2,335 animal units (AU) based on the definition given in EPA (2001). Milking cows were housed in a combination of open lot and covered free stall pens, calves were housed in individual pens, and all others were housed in open lot pens with an open shelter. Pens had a total area of 13.7 ha, about 65% of the total dairy footprint. Slurry manure passed through a solid separator basin (0.1 ha), with the removed solids stored in windrows and the liquids stored in a lagoon (0.6 ha). Milk cow lanes were flushed with water several times daily; other lanes were scraped weekly. Corrals were scraped as needed, with gathered material stored in each pen for later removal. No corral scraping occurred during the measurement campaign.

Historical wind data showed dominant northwest winds during June, which was confirmed during the study. Instruments were arrayed to measure background concentrations northwest and emission plumes south of the dairy. The facility layout and the adjacent road prevented the placement of point sensors to the southeast. An instrumentation trailer (AQ Trailer) was used for the following: sample preparation, collection, and storage; instrument handling, storage, and servicing; and data storage. Meteorological conditions were recorded with a Davis Instruments Vantage Pro2 Plus¹ (Hayward, Cali.) weather station at 5.0 m at AQ Trailer and two 15.3 m towers instrumented with logarithmically spaced sensors to measure vertical profiles of wind speed (RM Young Gill 3-cup anemometer, Traverse City, Mich.) and temperature and relative humidity (Vaisala model HMP45C, Oulu, Finland), with a Met One Instruments, Inc. Model 024A wind vane (Grants Pass, Ore.) at 15.3 m.

Particle mass distributions were measured using Airmetrics MiniVol PM filter-based samplers (Eugene, Ore.). They were deployed in clusters of either two or three units, individually configured to measure $\text{PM}_{2.5}$, PM_{10} , or TSP. These yield period-averaged mass concentrations. Optical particle size distributions were characterized with Aerosol Profilers Model 9722 (Met One Instruments, Inc., Grants Pass, Ore.), a.k.a. an optical particle counter (OPC). These were co-located with MiniVol clusters around the dairy. The OPCs reported 20 s cumulative counts in eight size bins ranging from 0.3 to 10.0+ μm .

Aglite, a scanning elastic lidar utilizing 3 wavelengths (355, 532, and 1064 nm) developed by Space Dynamics Laboratory in collaboration with the USDA Agricultural Research Service, was placed approximately 800 m west of the dairy. This system, the retrieval method, and its past applications are described by Wojcik et al. (2012). Collected MiniVol, OPC, and meteorological data were utilized in the retrieval to convert the lidar return signal to PM concentrations

throughout the field of view. The lidar mode of operation for emissions quantification during this experiment was a continuous series of upwind and downwind vertical scans, horizontal scans at ~10 m above the dairy, and calibration stares at the upwind reference point. An example set of scans collected at the dairy is shown in Figure 2.

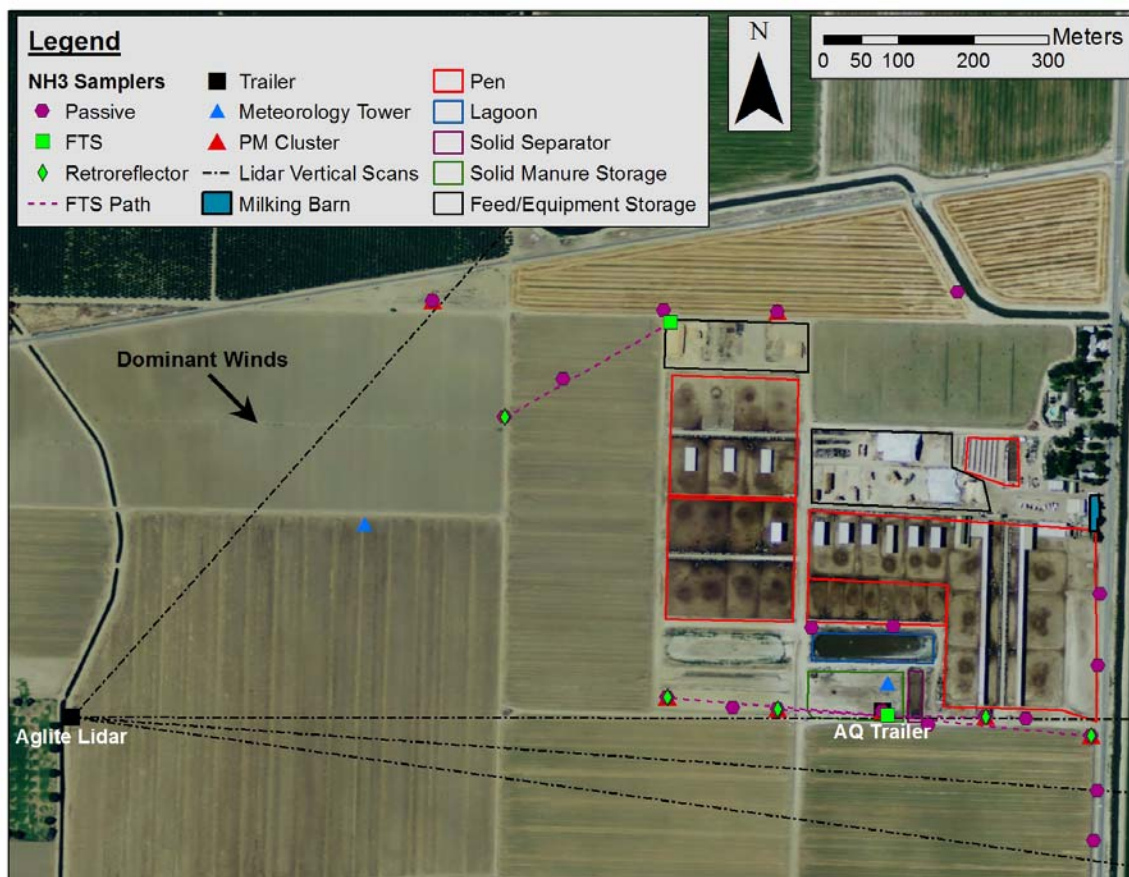


Figure 1. Map showing the dairy and sampling layouts.

Ogawa passive samplers (Ogawa & Company USA, Pompano Beach, Flor.), which operate on the principle of diffusion, loaded with citric acid-coated sample pads were spread around the dairy facility at 23 locations to measure period-averaged NH_3 concentrations. An in-depth description of both the passive sampler and the concentration calculation procedure are provided by Roadman et al. (2003). Exposed samples were analyzed using ion chromatography. In addition, two FTS instruments were deployed to measure NH_3 . Their principle of operation is based on the absorption of energy at different wavelengths by different compounds. The data product is an infrared spectrum which identifies and quantifies the gases present. The upwind FTS instrument, manufactured by Industrial Monitoring and Control Corporation (Round Rock, Tex.), was operated in a monostatic mode with a single beam path to a retroreflector. The downwind FTS was also a monostatic unit, manufactured by MDA (Atlanta, Geor.), but it was set in a scanning system with multiple beam paths to retroreflectors arrayed horizontally and vertically along the southern dairy border.

Samples were collected from mid-day June 13 through June 20. The lidar and OPCs operated nearly continuously through June 20; a lidar component failure late on June 19 prevented further operation. The FTS units collected data from mid-day on June 14 through the end of June 18. The MiniVol samplers and passive NH_3 samplers collected discrete samples due to

their measurement techniques. The MiniVols collected seven samples, a mix of four 23-hr and three 12-hr periods. The passive samplers were deployed for 13 12-hr sample periods. Breaks in sample collection were due to instrument servicing and logistical requirements.

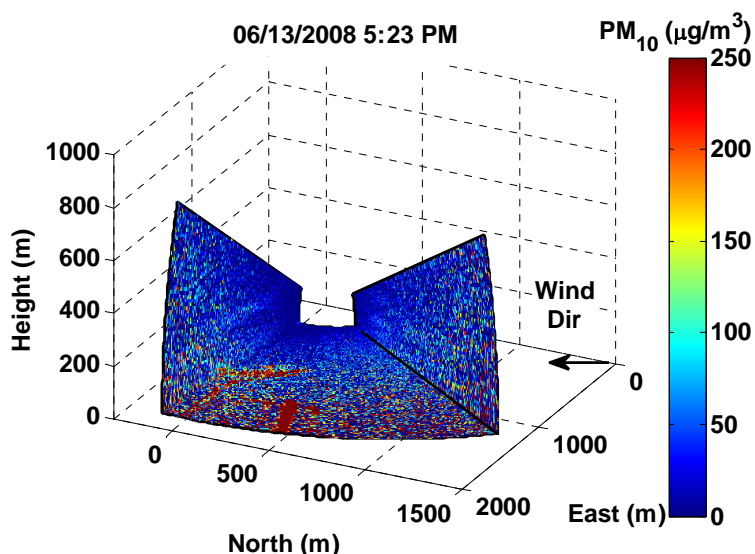


Figure 2. An example Aglite scan set collected at the dairy. The left side vertical scan coincides with the dairy's southern boundary; the dark red area near 500 m north is reflection from trees.

Emission Factor Calculations

PM emission rates were estimated using an inverse modeling technique with AERMOD, an EPA-recommended regulatory Gaussian air dispersion model, coupled with the filter-based mass concentration samples and a flux measurement technique using elastic lidar. NH₃ emissions were estimated through inverse modeling with both passive sampler and FTS data. Inverse modeling is the process of adjusting a user supplied emission rate while comparing the model-predicted levels to the measured concentrations until the best fit is found. The emission rate corresponding to the best fit is the inverse modeling estimate. Sources of PM (pens) and NH₃ (pens, lagoon, and solid separator) at the dairy were modeled as area sources. PM and NH₃ levels resulting from the dairy were calculated by subtracting average upwind concentrations from downwind concentrations. This was done for direct comparison with modeled concentrations as the model does not account for background levels. The lidar flux measurement technique calculates the difference between the PM mass passing through the upwind and downwind scanning planes, which is defined as the average upwind concentration subtracted from each bin in the downwind scanning plane and multiplied by the component of the wind perpendicular to the scanning plane at that elevation. The flux is averaged across the plume and over the period of interest; it is then related to a characteristic of the source, such as area or the number of animals. Meteorological data collected on-site were used in both the inverse modeling and flux techniques. Emission rates were normalized by total animal count and AU to calculate emission factors.

Measured Concentrations and Emissions

Measured PM_{2.5} concentrations ranged from 13.6 to 56.0 µg/m³, PM₁₀ concentrations ranged from 42.3 to 138.6 µg/m³, and TSP concentrations ranged from 69.8 to 246.4 µg/m³. Sample period average concentrations measured by the Aglite lidar in range bins near PM clusters

agreed well with reported PM concentrations. NH₃ levels reported by the passive samplers ranged from 9.2 to 1,248.3 µg/m³, with upwind and downwind averages ($\pm 1\sigma$) of 61.0 \pm 16.0 µg/m³ and 271.2 \pm 194.1 µg/m³, respectively. Background NH₃ concentrations measured by the upwind FTS averaged 46.4 \pm 32.8 µg/m³, while downwind average concentrations ranged from 72.4 \pm 34.1 µg/m³ to 238.7 \pm 88.7 µg/m³. In point sensor and FTS datasets, the highest PM and NH₃ were measured at 2 m above ground level and immediately downwind of the dairy. Lidar measurements were not feasible lower than ~10 m above the ground due to safety concerns.

Diurnal patterns were observed in downwind measurements and calculated emission factors, with the lowest values occurring at sunrise and the highest values around sunset. Mean PM emission factors calculated from these data are presented in Table 1, along with other reported PM₁₀ emission factors. The PM₁₀ values herein reported are generally higher than those from other studies which are from dairies with different housing type and climatic conditions. In addition, the measurement methodologies employed vary. Estimated NH₃ emission factors from both measurement technique datasets are presented in Table 2 with select values from literature. The NH₃ emissions herein reported are much higher than those from other studies. Differences with values calculated in other studies may partially be attributed to varying housing, climatic, and management conditions and emission measurement and estimation methodology.

Table 1. Average (\pm 95% CI) PM emission factors from this study and some reported by others.

Emission Factor	Inv. Modeling (this study) (g/d/head)	Lidar (this study) (g/d/head)	USDA (2000) (g/d/head)	Schmidt et al. (2006) (g/d/head)	Goodrich et al. (2006) (g/d/head)
PM _{2.5}	4.7 \pm 4.0	1.6 \pm 0.2	-	-	-
PM ₁₀	30.7 \pm 18.0	18.7 \pm 2.7	1.8	1.7 (winter) 0.3 (summer)	5 (free stall) 15 (open lot)
TSP	94.0 \pm 41.1	57.5 \pm 8.7	-	-	-

Table 2. Mean (\pm 95% CI) NH₃ emission factors (g/d/head) estimated through inverse modeling from this study, as well as some reported by others.

Emission Source	Passives (this study)	FTS (this study)	Select Reported Values
Pen (g/d/head)	143.4	236.2 \pm 69.1	-
Pond (g/d/head)	29.0	20.3 \pm 10.4	-
Total (g/d/head)	172.4	256.5 \pm 67.7	3.6-43.2 (Arogo et al., 2006), 93.0-100.0 (Flesch et al., 2009)
Pond (g/d/m ²)	5.4	3.1 \pm 1.6	2.3-8.7 (Flesch et al., 2009), 2.6-13.0 (Rumburg et al., 2008)

Conclusions

PM and NH₃ concentrations in and around a U.S. commercial dairy were measured using point and remote sensors. The following average emission factors (\pm 95% CI) were developed from these datasets using an inverse modeling technique: PM_{2.5} -3.8 \pm 3.2 g/d/AU; PM₁₀ -24.8 \pm 14.5 g/d/AU; TSP -75.9 \pm 33.2 g/d/AU; and NH₃ -143.4 g/d/AU from passive sampler data and 207.1 \pm 54.7 g/d/AU from FTS data. PM emission factors estimated from lidar data were 1.3 \pm 0.2 g/d/AU, 15.1 \pm 2.2 g/d/AU, and 46.4 \pm 7.0 g/d/AU for PM_{2.5}, PM₁₀, and TSP, respectively.

These emission factors are higher than most found in the literature, possibly resulting, in part, from differences in housing, climatic conditions, manure and surface management, and measurement and emissions estimation techniques.

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