Scaling: Field Experiments and Measurements

Observing Air Quality from Space: From Research to Decision Support

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Abstract

Air pollution is a major cause of respiratory and cardiovascular disease. The principal pollutants of concern are ozone and aerosols, which are produced in the atmosphere by chemical reactions involving nitrogen oxides (NO_x = NO + NO_2), volatile organic compounds (VOCs), carbon monoxide (CO), and sulfur dioxide (SO_2). These precursors have both anthropogenic (principally combustion) and natural sources. Aerosols are also directly emitted to the atmosphere by combustion, dust suspension, and sea spray.

High concentrations of ozone and aerosols originating from human industrial and agricultural activity are a threat to public health in most densely populated regions of the world. Surface air measurement networks have insufficient coverage to monitor population exposure, and are generally lacking in the developing world. Ozone and aerosol formation depend in complex and nonlinear ways on the concentrations of precursors, for which little in situ observational data are available. Management decisions for air quality require emission inventories for the precursors, and these are generally constructed by a "bottom-up" approach involving application of emission factors to activity rates. Such inventories are often uncertain by a factor of 2 or more, particularly in the developing world. Air quality management also requires better understanding of the long-range transport of pollution, including on intercontinental scales.

Recent advances in tropospheric remote sensing have stimulated research interest in the application of satellite observations to air quality issues. Observations of NO_2 and formaldehyde from a variety of space-borne instruments have been used to place new top-down constraints on sources of NO_x and VOCs and their interannual variability and trends. Observations of CO from satellite missions have also been used to constrain CO sources and to track the intercontinental transport of pollution plumes. Combined observations of ozone and CO from the NASA Aura Mission are offering constraints on intercontinental transport of ozone pollution. Aerosol optical depth (AOD) observations from the other NASA Earth Observing System satellites have been used to infer surface air concentrations of PM, providing new insights in particular in the developing world where few in situ data are available. Assimilation of these measurements of AOD observations, and those of trace gas abundances as well, are being used in near-real-time air quality analyses and forecasts.

These research advances have come under notice by the air quality management community, which is increasingly seeing satellite observations as a crucial part of a hierarchy of observations and models needed for future air quality management, both for monitoring and for improving understanding of source-receptor relationships and of the underlying processes. This paper will review how satellite assets already in space are being exploited for characterizing, understanding and predicting air quality, and the limitations of these current missions for which air quality applications were in many cases incidental to the original mission goals. The paper will also present some of the considerations for developing a strategy for future space missions with increased capability for air quality applications. Some of the key considerations in the design of new instruments are the spatial and temporal resolution required to effectively monitor air quality from space, which in turn informs the consideration of the various potential orbital vantage points. An approach that has been used to address the issue of spatial resolution by calculating the spatial length scales of trace gases within the boundary layer and free troposphere using both in-situ aircraft data from five different NASA aircraft campaigns and simulations with an air quality model will be presented.

Measurement and Analysis of Ammonia and Hydrogen Sulfide Emissions from an Anaerobic Swine Waste Treatment Lagoon and Confinement Building in North Carolina

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Abstract

Emissions of atmospheric ammonia-nitrogen (NH_3-N, where NH_3-N = (14/17)NH_3) and hydrogen sulfide (H_2S) from a commercial anaerobic swine waste treatment lagoon and a fan-ventilated swine confinement building located next to the lagoon were measured over a one year period. Continuous simultaneous measurements were made at the lagoon using a dynamic flow-through chamber system for ~1 week during four seasons in an effort to examine diurnal and seasonal variability and the respective relationships of NH_3-N and H_2S emissions to lagoon physiochemical properties. During the fall and spring, average NH_3-N fluxes were 1634 ± 505 and 2495 ± 465 $\mu g\ N\ m^{-2}\ min^{-1}$, respectively. The lowest fluxes were observed during the winter where average flux values were 1290 ± 246 $\mu g\ N\ m^{-2}\ min^{-1}$. The lowest fluxes for H_2S were also observed during the winter season, ~0.0 $\mu g\ m^{-2}\ min^{-1}$. Average fluxes increased slightly during the fall (0.3 ± 0.1 $\mu g\ m^{-2}\ min^{-1}$) and spring (0.5 ± 1.0 $\mu g\ m^{-2}\ min^{-1}$) and were observed at highest flux values during the summer (5.3 ± 3.2 $\mu g\ m^{-2}\ min^{-1}$). Generally, the lagoon emissions for H_2S were ~3-4 orders of magnitude less than NH_3-N.

Continuous simultaneous NH_3-N and H_2S emissions from a swine confinement house were also made for ~1 week period during four seasons. Normalized NH_3-N emission rates were highest in winter (33.5 ± 22.1 g N day^{-1} 500kg live animal $weight^{-1}$ (LAW)) and lowest during fall (11.6 ± 7.2 g N day^{-1} 500kg LAW^{-1}). Normalized H_2S emissions were lowest in summer and fall (1.4 ± 0.7 and 1.7 ± 0.5 g day^{-1} 500kg LAW^{-1}) and were highest during the winter and spring seasons (4.2 ± 2.2 and 4.3 ± 1.0 g day^{-1} 500kg LAW^{-1}).

Introduction

Changes in livestock production methods in the U.S. are in turn changing emissions of trace gases (e.g., sulfur and nitrogen species) into the atmosphere (Aneja et al, 2006). In North Carolina, for example, large-scale commercial operations have emerged over the last few years, dramatically increasing the number of hogs in a geographically concentrated area. In this state, there are currently over 1400 operations with more than 1000 head, accounting for almost 99% of the state inventory (http://www.nass.usda.gov/QuickStats/). Subsequently, emissions of potentially harmful gases such as ammonia, (NH_3) and hydrogen sulfide (H_2S) from confined animal feeding operations (CAFOs) have become a major problem in recent years. Public concerns about potential environmental and health effects of air emissions from CAFOs have increased in parallel with the growth and consolidation of this industry.

Ammonia released from near-surface sources (i.e. waste treatment lagoons, confinement houses) into the atmosphere has a relatively short lifetime of ~1-5 days (Warneck, 2000) and may deposit near the source through dry or wet deposition. However, ammonia can also participate in atmospheric reactions (e.g. gas-to-particle conversion) once airborne, forming ammonium aerosols such as ammonium sulfate, -nitrate, -chloride, which tend to have longer atmospheric residence lifetimes (~1-15 days) owing to a decrease in dry deposition velocity (Aneja et al., 1998) and therefore may be transported and deposited further downwind from the source. An environmental hazard in eastern North Carolina associated with gaseous ammonia and ammonium aerosols is deposition into sensitive coastal river systems where nitrogen loading may lead to enhanced eutrophication and soil acidification, which may in turn upset plant nutrient balances near sources (Paerl, 1997).

Hydrogen sulfide is a colorless, potentially lethal gas released from swine manure (U.S.EPA, 2001). It is produced as manure decomposes anaerobically, resulting from the mineralization of organic sulfur compounds as well as the reduction of oxidized inorganic sulfur compounds such as sulfate by sulfur-reducing bacteria (U.S. EPA, 2001). The U.S. Center for Disease Control (CDC) warns that brief exposures to high concentrations (> 500 ppm) can cause unconsciousness or death (ATSDR, 2004). With a low odor threshold ranging from 0.0005 to 0.3 ppm (ATSDR, 2004), it is also one of the primary gases released from swine facilities that is associated with odor complaints due to it's characteristic "rotten egg" smell. Donham et al. (1982) reported that hydrogen sulfide appeared to be the main toxic substance in liquid manure associated with death and illness for people with acute exposure to gases emanating from liquid manure. Campagna et al. (2004) have reported a correlation between elevated ambient H_2S concentrations and hospital visits for respiratory diseases .Potential negative health effects of overall emissions from hog farms have been identified in production workers and neighboring residents that include respiratory illnesses such as bronchitis and asthma, and increased psychological stress (Schiffman, 1998). However, it is noted that H_2S is just one component of many odorous and potentially harmful gases that have been identified in hog farm emissions.

Significant efforts have been devoted to understanding emissions of ammonia from waste storage treatment lagoons (Harper et al., 2000; Aneja et al., 2000, 2001a, b). Arogo et al. (2003) have reviewed measurement techniques and studies undertaken to estimate NH_3 emissions from both swine lagoons and animal confinement houses. Emissions of ammonia (Zhu et al., 2000; Heber et al., 1997, 2000; Wood et al., 2001, Schmidt et al., 2002) and hydrogen sulfide (Heber et al., 1997; Zhu et al., 2000; Ni et al., 2002; Schmidt et al., 2002) from swine confinement houses have been estimated. To date, few studies have reported H_2S emissions from waste storage treatment lagoons (Zahn et al., 2001; Lim et al., 2003), none from farms in the southeastern U.S. Various factors, such as farming activities (i.e., feed composition, animal density) and regional climates, may affect the emissions of gases into the atmosphere. Emissions estimates generated for one set of conditions or for one type of CAFO may not translate readily to others. It is therefore important for comprehensive emissions measurements to be made from different types of operations.

The primary objective of this research initiative was to investigate and evaluate the variability of ammonia and hydrogen sulfide emissions with respect to diurnal and seasonal variations as well as the influence of meteorological and physico-chemical factors. Data presented here was collected continuously for about a one-week period from an anaerobic swine lagoon and about one week from a swine finishing barn during each season (i.e., fall, winter, spring, summer) for one year at a commercial hog farm in eastern North Carolina. Generally, the barn measurements were made immediately prior to following the lagoon experimental, except during the fall season where lagoon measurements were made in fall 2004 and barn measurements were made in fall 2005.

Methods and Materials

Physiographic Location and Farm Description

The experimental site is a commercial swine finishing farm, operating under steady-state conditions, located in Jones County in the southeastern Piedmont region of the state. The on-site waste storage treatment lagoon is 3.06 Ha (30,630 m^2) and the farm maintains eight fan-ventilated confinement houses with ~800-900 animals housed in each barn. Each barn has a shallow manure collection pit which is emptied once a week. The farm utilizes a conventional "lagoon and spray" technology as its primary means of handling effluent. Effluent is flushed directly from the hog barns into the storage lagoon where it is treated via natural microbial processes. The stored wastewater is used to recharge the barn pits, and also periodically irrigated over on-site agricultural crops for nutrient enrichment purposes.

Dynamic Flow-Through Chamber System

A flow-through dynamic chamber system with a variable-speed motor-driven continuous impeller stirrer (Kaplan et al., 1988; Kim et al., 1994; Chauhan, 1999; Aneja et al., 2000) was employed to determine NH_3-

N and H_2S flux from the lagoon. The translucent plastic cylindrical chamber, 0.25 m internal diameter, 0.46 m internal height of chamber above water or soil, and volume $0.025m^3$, is fitted into a circular hole cut into the center of a 0.61 x 0.61 m floating ½" thick ultra-high molecular weight (UHMW) polyethylene platform. The chamber penetrated into the lagoon ~7 cm, creating an internally closed system. The platform is supported by 0.15 m diameter by 1.68 m length PBC piping for floatation, designed to sit a few centimeters above the lagoon in order to minimize experimental effects and simulate atmospheric conditions at the lagoon surface as accurately as possible. A schematic of the chamber system is shown in Figure 1.

The chamber was lined internally with a 2 mil fluorinated ethylene propylene (FEP) Teflon sheet to reduce chemical reactions and build up of temperature inside the chamber. Compressed zero-grade air was used as a carrier gas and pumped through the chamber at a variable flow rate of ~0.009-0.012 m^3 min^{-1} utilizing a Model 810-S Mass Trak Flow Controller (Sierra Instruments, Monterey, CA). The in-flowing air was carried through a ¼" o.d., 5/32" i.d Teflon FEP sample line to the chamber. The air inside the chamber is ideally well-mixed by a variable-speed motor-driven Teflon impeller stirrer ranging from speeds of 40-60 rpm for this study.

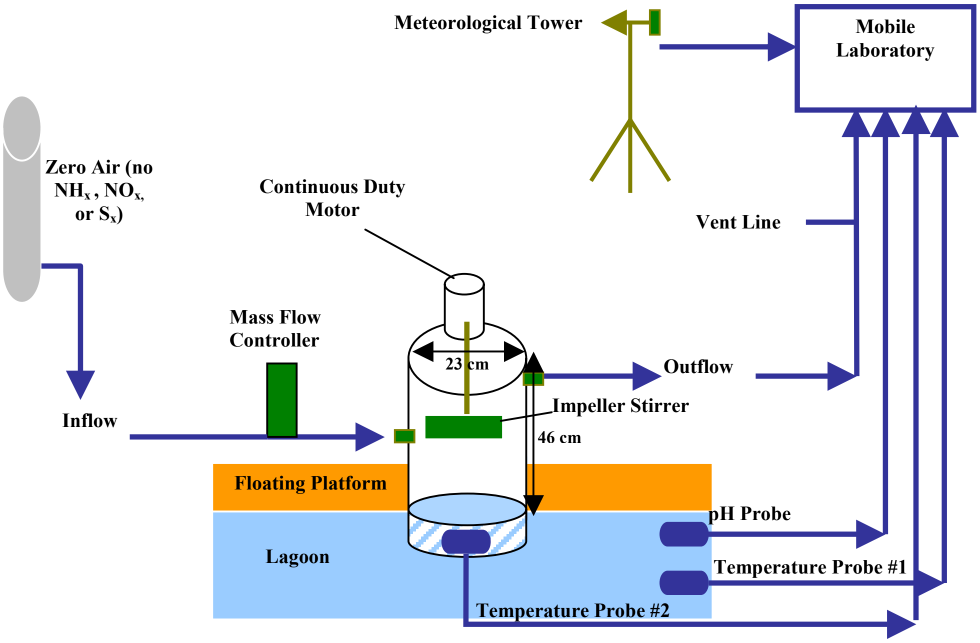

Figure 1. Schematic of dynamic flow-through chamber system configured to measure emissions from a swine waste treatment lagoon

A vent line was fitted to the exiting sample line to prevent pressurization and was periodically bubble tested to check for under pressurization and/or leaks in the enclosed system. The entire closed system was lined with Teflon inside the chamber and stainless steel fittings in order to minimize chemical reactions with sample flow. The sample exiting the chamber travels through the sample line and is then split in order to deliver the sample simultaneously to the different gas analysis instruments.

Lagoon Flux Calculation

In order to calculate NH_3-N and H_2S fluxes for this research experiment, the following mass balance equation was used for the dynamic flow-through chamber system:

$$\frac{dC}{dt} = \left(\frac{q[C_{air}]}{V} + \frac{JA}{V}\right) - (C)\left(\frac{LA_w}{V} + \frac{q}{V}\right) - R \qquad (1)$$

where C gas concentration inside the chamber

C_{air}	concentration in carrier air
q	flow rate of compressed air through the chamber
V	volume of the chamber
A	surface area of the lagoon covered by the chamber
A_w	inner surface area of the chamber of inner and upper wall surfaces
L	total loss of gas in the chamber per unit area due to reaction with inner and upper walls of the chamber
h	internal height of the chamber
J	emission flux per unit area
R	gas phase reactions inside the chamber

Since zero-grade air was used as the carrier gas, C_{air} was assumed to equal zero and gas phase reactions, R, was also assumed to be zero. Since the air inside the chamber was assumed to be well mixed by the impeller stirrer, C was assumed to be constant within the chamber. At steady-state conditions, the change of concentration with respect to time $\left(\frac{dC}{dt}\right)$ was zero. Therefore equation (10) can be simplified as:

$$\frac{J}{h} = C_{eq}\left(\frac{LA_w}{V} + \frac{q}{V}\right) \tag{2}$$

Loss term, L, is determined experimentally while equilibrium-state gaseous concentration (C_{eq}), flow rate (q), and chamber dimensions (V and h) are all measured. Kaplan *et al.* (1988) has devised a method for calculating loss term by calculating the slope of the plot of $-\ln\left[\frac{C_{eq} - C(t)}{C_{eq} - C_o}\right]$ versus time (t). For this experiment, C_o is the initial equilibrium state gas concentration measured by the chamber system at a constant flow rate ($m^3\ min^{-1}$). C_{eq} is the measured gas concentration at a second equilibrium state at an increased or reduced flow rate into the chamber system. C(t) depicts gas concentration at any time, t, during the transition between the first and second equilibrium states. L is determined by:

$$L = \left(slope - \frac{q}{V}\right)\left(\frac{V}{A_w}\right) \tag{3}$$

Lagoon Sample Collection

Lagoon samples were collected daily during each initiative using sterile plastic 500 mL bottles and submitted to the North Carolina Division of Water Quality (NC DWQ) for total ammoniacal nitrogen (TAN), total Klejdahl nitrogen (TKN), and total sulfide analysis. Lagoon samples collected for TAN and TKN analyses were preserved with sulfuric acid to pH < 2 and samples collected for sulfide analyses were preserved with 6N sodium hydroxide and 2N zinc acetate to pH > 9, according to NC DWQ specifications. All samples were immediately stored on ice (<4°C) and brought to the NC DWQ laboratory within 48 hours for analysis.

Barn Emission Experimental Design

Five ventilation fans, two 92 cm (36”) diameter direct driven and three 122 cm (48”) diameter belt-driven, were located at the west end of the building. The fans were staged to operate as temperature increased inside the building. In order to accurately determine fan flow rates, a Mabuchi VDC motor (Santa Clara, CA) was either (1) mounted to a stainless-steel plate configured to fit over the front of the 92 cm fans or (2) attached to a cylinder sleeve which fit over the fan shaft of the 122 cm fans. Single analog output wires were connected from each motor to a Campbell Scientific CR10X data logger which continuously recorded the measured voltage output. Prior to the experiment, each motor was calibrated in the laboratory to obtain voltage outputs at a specific rpm. The rpm for each fan, as well as “on/off” times, could then be determined and flow rates subsequently calculated. According to manufacturer specifications the direct drive motor on the 92 cm fans is rated at 850 rpm and the 122 cm belt-driven fans are rated at 1725 rpm. However, fan belts may become loose over time and the rpms decrease, thus affecting the flow rate (Janni et al., 2005). Static pressure inside the building was monitored using a Model PX655 Omega pressure transducer.

Taking into account static pressure, the flow rates for each fan size were calculated using the following calculation:

$$CalculatedFlowRate = \left(\frac{ManufacturerFanFlowRate}{SpecifiedRPMs} \right) (MeasuredRPMs) \qquad (5)$$

Due to the nature of the airflow through the building, it is assumed that the gaseous concentrations are uniformly distributed at each fan outlet. A ¼" o.d., 5/32" i.d. Teflon® sample line was inserted inside the chimney of the first 92 cm fan to turn on, between the shutter and fan blade, at roughly half the fan radial distance. The air is drawn into the lines via vacuum pumps from each gas analyzer and the sample line is split in order to deliver the sample simultaneously to the different analysis instruments.

Barn Flux Calculation

It is assumed that the concentration is uniform across the barn at the fans. Therefore, the emission rate is calculated by

$$J = C_g * \sum F \qquad (6)$$

where		
	J	gas emission rate from swine barn
	C_g	gas concentration at fan exhaust
	F	flow rate of each fan

Gas Analyzers and Meteorological Instrumentation

A Model 450C pulsed fluorescence H_2S/SO_2 analyzer and a Model 17C chemiluminescence NH_3 analyzer (Thermo Environmental Corporation, Mountain View, CA) were used to measure the concentration of ammonia and hydrogen sulfide, respectively. Multi-point calibrations (80, 60, 40, and 20% of full-range scale) for the Model 450C and Model 17C analyzers were conducted in the laboratory prior to each sampling campaign. During the field study, zero and span checks for H_2S and NH_3-N concentration were conducted daily.

A 10 meter meteorological tower was erected to measure ambient wind speed and direction, air temperature, relative humidity, and solar radiation. Wind speed and direction were measured at 10 m above the surface and air temperature, relative humidity, and solar radiation measurements were made at 2 m height.

At the lagoon, a Model CSIM11 pH probe (Campbell Scientific Inc., Logan, UT) was used to continuously monitor near-surface (6-7 cm depth) lagoon pH. Two CS107 (Campbell Scientific Inc., Logan, UT) temperature probes simultaneously measured the near-surface lagoon temperatures inside the chamber as well as 0.3 m outside the chamber to ensure there are no significant differences. To measure barn exhaust temperature, a CS107 temperature probe was placed in front of a 91 cm outlet fan.

A Model CR10X data logger equipped with a Model AM 16/32 Channel Relay Multiplexer (Campbell Scientific, Inc., Logan, UT) was used to collect all meteorological, lagoon, and static pressure data and a Model CR23X was used to acquire all gaseous and mass flow data. Data was collected every second, and averaged and recorded over a 15 minute timeframe. The data loggers and gas analyzers were housed inside a temperature-controlled mobile laboratory (N.C. State University Air Quality Ford Aerostar Mini-Van), maintained at ~21°C (~70°F).

Results

Lagoon Flux

The NH_3 and H_2S analyzers were programmed to collect data at 20,000 (maximum concentration range) and 1,000 ppb, respectively. Table 1 summarizes the NH_3-N and H_2S fluxes from the lagoon for each season as well as lagoon temperature and pH, both of which are expected to have a significant effect on emission rates (Aneja et al., 2000; Zahn et al., 2002). The lowest average daily fluxes for both NH_3-N and

H_2S were observed during the winter experimental period, 1290 µg-N m^{-2} min^{-1} and ~0.0 µg m^{-2} min^{-1}, respectively. During the winter season, only 2.3% of the H_2S concentration values were above the minimum detectable limit (1 ppb) for the analyzer and so the flux value was considered to be negligible. The highest observed average daily H_2S flux values, 5.3 µg m^{-2} min^{-1}, were observed during the summer. Generally, almost 87% of the total H_2S flux was observed during the summer season.

Table 1. Statistical summary table of average daily NH_3-N and H_2S lagoon fluxes, lagoon temperature, and pH for each season

Season	Sample Dates	NH_3-N Flux[a,b]	H_2S Flux[b]	Lagoon Temperature[c]	Lagoon pH
Fall	Oct 26 – Nov 1, 2004	1634(505)[d]	0.3(0.1)	18.2(1.7)	8.09(0.03)
		1110-3305[e]	0.0-0.8	15.7-23.3	8.02-8.15
Winter	Feb 15-21, 2005	1290(246)	~0.0(0.2	11.5(1.4)	8.12(0.03)
		867-2027	0.0-1.2	9.1-14.5	8.02-8.20
Spring	Apr 14-19, 2005	2495(465)	0.5(1.0)	15.1(1.7)	8.10(0.02)
		1565-3216	0.1-7.1	12.4-19.8	8.03-8.15
Summer	Jun 14-18, 2005	4294[f]	5.3(3.2)	29.7(1.4)	7.99(0.05)
		3864-?	0.8-13.3	26.9-32.5	7.90-8.10

[a] NH_3-N flux = (14/17) NH_3 flux

[b] Units of flux are µg m^{-2} min^{-1}

[c] Measured in °Celcius

[d] Numbers in parenthesis represent one standard deviation

[e] Represents range of values

[f] NH_3 concentration was above maximum range on the ammonia analyzer and so maximum and average flux values are unknown.

For NH_3-N, the concentration exceeded the maximum value (20,000 ppb) on the analyzer during most of the summer experiment. The average daily flux value of 4294 µg-N m^{-2} min^{-1} includes maximum measured concentrations and is reported only to demonstrate the dramatic increase during the warmest months. This should not be considered a true flux value. The actual flux is assumed to be higher, but the extent of the increase is unknown. Aneja *et al.* (2000) reported a range of ammonia flux values of 2358-8526 µg-N m^{-2} min^{-1} from another hog lagoon in North Carolina during the summertime.

Lagoon samples were collected daily throughout each experimental period at the surface near the experimental chamber system and simultaneously at a randomly chosen location at the lagoon. The samples were compared to check for differences in concentration levels between the fixed chamber location and other random locations where effluent samples were collected. Using SAS Statistical Software Version 8.2 (SAS Institute, Cary, NC), an independent samples t-test was performed (n = 23) and it was determined that there was no significant difference (p-value > 0.05) in concentration (mg/L) for any of the lagoon samples (sulfide, TAN, TKN). It is therefore assumed that the concentration levels are relatively uniform throughout the lagoon surface and that the emissions measured at the chamber locations represent the average emissions from the entire lagoon surface. The average daily emission from the 3.06-ha lagoon during the summer is calculated to be more than 189 kg day^{-1} for NH_3-N and 230 g day^{-1} for H_2S.

Barn Emission Rates

A Teflon® sample line was inserted inside the chimney of the first 92 cm fan to turn on in series in order to measure NH_3-N and H_2S gas concentrations at the fan ventilation exhaust. Concentration measurements for NH_3-N were highest during the winter and spring, 8.9 and 8.4 ppm, respectively. The higher concentration values during the winter may likely be attributed to less airflow through the building since the ambient and barn temperatures were generally cooler and the fans are staged to turn on as temperatures increase. A buildup of the gas therefore occurs in the building with fewer fans operating. During the spring, the average hog weight was ~88 kg as compared to 38-58 kg during the other seasons when measurements were made. It is expected that the large concentrations measured in the spring are due to the age and weights of the animals housed in the barn. The highest average concentration for H_2S (673 ppb) also occurred during the winter. This value is well below the 500 ppm concentration level at which the CDC reports that severe health risks may occur but well within the odor threshold range (ATSDR, 2004).

Table 2. Statistical summary table of total and normalized average daily NH_3-N and H_2S barn emission rates for each season

	Total Emission Rate[a]			**Normalized Emission Rate**[a,b]		
Season	**Mean**[c]	**Minimum**	**Maximum**	**Mean**[c]	**Minimum**	**Maximum**
NH_3-N						
Winter 2005	3260 (2156)	54	8930	33.5 (22.1)	0.6	91.7
Spring 2005	4508 (1652)	1550	14514	30.5 (11.2)	10.5	98.2
Summer 2005	1643 (796)	628	8935	24.2 (9.2)	9.2	131.6
Fall 2005	890 (552)	299	3027	11.6 (7.2)	3.9	39.4
H_2S						
Winter 2005	407 (212)	0	911	4.2 (2.2)	0.0	9.4
Spring 2005	482 (142)	100	859	4.3 (1.0)	0.7	5.8
Summer 2005	83 (49)	2	287	1.4 (0.7)	0.0	4.2
Fall 2005	134 (40)	1	252	1.7 (0.5)	0.0	3.3

NH_3-N flux = (14/17) NH_3 flux

[a] Units of flux are g day^{-1}

[b] AU (Animal Units) = 500 kg Live Animal Weight

[c] Numbers in parenthesis represent one standard deviation

Total and normalized emission rates for both NH_3-N and H_2S are given in Table 2. Calculated total emission rates for both NH_3-N and H_2S were highest during the spring, 4508 and 482 g day^{-1}, respectively. Emissions were lowest during the fall season for NH_3-N (890 g day^{-1}). The average emission rate for each season was normalized by 500 kg LAW, thus removing the total live mass as a variable to explain emission rates. Normalizing the data also provides an effective means to compare emission rates with other studies. The normalized emission rates ranged from 11.6-33.5 g day^{-1} for NH_3-N and 1.4-4.3 g day^{-1} for H_2S. It is likely that other factors such as climatic conditions, animal activity, and/or length of time the animals have been housed in the barn may affect the emission rates.

Conclusions

Emissions of atmospheric NH_3-N and H_2S from a commercial anaerobic swine waste treatment lagoon and a fan-ventilated swine confinement building located next to the lagoon were measured over a one year period. Continuous simultaneous measurements were made at the lagoon using a dynamic flow-through chamber system and at the barn ventilation exhaust for ~1 week each during four seasons in an effort to examine diurnal and seasonal variability.

Lagoon flux was highest for both NH_3-N and H_2S during the summer sampling initiative. The NH_3-N measurements surpassed the instrumentation maximum value (20,000 ppb) during most of the summer experiment. Generally, the NH_3-N emissions were ~3-4 orders of magnitude greater than H_2S. The average daily emission from the 3.06-ha lagoon during the summer was more than 189 kg day^{-1} for NH_3-N and ~230 g day^{-1} for H_2S. The lowest fluxes were measured during the winter.

At the barn fan exhaust, the highest emission rates from the barns were measured during the winter and spring for both NH_3-N and H_2S. The lowest emission rates were measured during the fall for NH_3-N and summer for H_2S.

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Gas Exchange between Canopy and Atmosphere

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Abstract

The paper gives brief descriptions of an inverse Lagrangian model of gas transport in plant canopies and multi-layer models of canopy gas exchange. The inverse Lagrangian model allows the inference of sites of gas exchange in the canopy and their source and sink strengths from measured profiles of gas concentration and statistics of the canopy turbulence. The second model analyses gas exchange in terms of resistances to transport across boundary-layers at leaf and foliage surfaces, and in the case of bi-directional gas fluxes, resistances to further transport through stomata to leaf interiors. Gas diffusion through stomata is controlled by stomatal resistance and the difference between the gas concentrations at the leaf surface and that in the sub-stomatal cavity, known as the compensation point. Coupling of both models is seen as a powerful way to study gas exchange in plant canopies and examples are given of their combined use to study pathways of NH_3 exchange between canopy and atmosphere.

Introduction

Understanding, modelling and measuring the exchange of trace gases between agricultural sources and the atmosphere can be complicated by the fact that the gases may have both sources and sinks within the canopy. Examples include CO_2, NH_3, NO, NO_2 and SO_2. Early attempts to infer source/sink distributions of scalars within crop and forest canopies relied on a classical flux-gradient relationship appropriate for the air-layers above the canopy, in which the flux was calculated as the product of a height-dependent turbulent diffusivity, K, and the mean local concentration gradient of the scalar. The subsequent demonstration that classical K-theory is inappropriate in the canopy space, e.g., Denmead and Bradley (1987), led to the development of alternative approaches. Some of these require high-frequency measurements of scalar concentrations, not yet possible for most of the trace gases of interest to the Workshop. Others, though, rely on mean concentration gradients and the paper is concerned primarily with the application of one of these known as Inverse Lagrangian (IL) analysis which is based on a Localized Near Field (LNF) theory due to Raupach (1989a). It assumes that the scalar concentration profile within the canopy is the result of contributions from both local and distant sources. Transport in the near field close to an emitting surface such as a leaf is described by a Lagrangian dispersion model and that in the far field, say >1 canopy height downwind, by turbulent diffusion following K-theory. Inputs to the analysis are the mean scalar concentration profiles above and within the canopy, certain statistics of the turbulence and an index of atmospheric stability which can have a large influence on the results through its effect on far-field diffusion. The analysis allows us to identify the sites of gas exchange in plant canopies in a non-disturbing continuous way. It is a micrometeorological alternative to cuvettes and chambers. Detailed descriptions of the technique and its practical application have been given by Raupach (1989b) and Denmead et al. (2005). A brief outline is given below.

The paper describes different applications of the IL approach. Most have been concerned with inferring sources and sinks of heat, water vapour and CO_2 within the canopy (Denmead and Raupach, 1993; Denmead et al., 2000; Leuning, 2000; Katul et al., 2001; and Denmead et al., 2005). However, others have examined source-sink distributions for trace gases: ammonia (Harper et al., 2000; Nemitz et al., 2000a; Denmead et al., 2005), methane (Leuning et al. 2000), and radon (Simon et al., 2005). The studies of NH_3 exchange are perhaps of most interest to the Workshop. They reveal a variety of emission patterns and raise the question of how much of the NH_3 released from a ground source (fertiliser, slurry or manure) can be recovered by the canopy as the gas diffuses to the atmosphere. The same question arises for other reactive trace gases such as SO_2 and NO. The paper examines this question as well as the coupling of IL analysis and multi-layer models of canopy gas exchange to understand emission processes for reactive gases.

Inverse Lagrangian Dispersion Analysis

The semi-quantitative description here is a précis of developments by Raupach (1987, 1989a,b). In extensive, uniform plant canopies in steady atmospheric conditions, the conservation equation for scalars becomes

$$S(z) = dF/dz \qquad [1]$$

where S is the source or sink strength for the scalar, z is height above the ground and F is the vertical flux density of the scalar. On integration, Eq. [1] leads to

$$F(z) = F_g + \int_0^z S(z)dz \qquad [2]$$

where F_g is the flux density of the scalar to or from the ground.

The forward Lagrangian approach to turbulent transfer in plant canopies aims to predict the mean concentration profiles of scalars from given source distributions. It does so by tracking an ensemble of "marked particles" carrying the scalar. Turbulent motions in plant canopies are coherent over finite length and time scales so that fluid particle velocities exhibit persistence over significant time intervals. The typical persistence time for the vertical velocity of a wandering element is the Lagrangian time scale T_L. In the near-field close to the source, particle trajectories depart little from straight lines. The mean depth of the cloud generated by the source, σ_z, grows linearly with time, t, and is calculable from a Lagrangian description of the turbulence:

$$\sigma_z(t) \rightarrow \sigma_w t \qquad [3a]$$

with σ_w being the standard deviation of the vertical velocity w. In the far-field some distance from the source, persistence breaks down and particle trajectories resemble random walks, as in a diffusion process; cloud depth increases as $t^{1/2}$.

$$\sigma_z(t) \rightarrow \left[2\sigma_w^2 T_L (t - T_L)\right]^{\frac{1}{2}} \qquad [3b]$$

The near-field extends about one canopy height downwind of a point scalar source such as a leaf.

It is evident that the scalar concentration at any point in the canopy results from both near- and far-field influences, so that the actual concentration C can be split into a far-field component C_f that obeys gradient diffusion, and a near-field component C_n that does not.

$$C(z) = C_f(z) + C_n(z). \qquad [4]$$

Raupach (1989a) shows how both C_f and C_n can be related explicitly to the source density profile in the canopy provided one knows σ_w and T_L. The far-field diffusivity is given by

$$K_f = \sigma_w^2 T_L. \qquad [5]$$

Following K-theory,

$$F(z) = -K_f(z)\frac{\partial C_f}{\partial z}, \qquad [6]$$

which, on integration, leads to

$$C_f(z) = C(z_R) - C_n(z_R) + \int_z^{z_R} \frac{F(z)}{K_f(z)}dz, \qquad [7]$$

where $C(z_R)$ is the concentration at a reference level z_R well above the canopy. Raupach (1989a) derives a mathematical expression for C_n involving S, σ_w and T_L which then enables concentrations to be related to source strengths and statistics of the turbulence, specifically σ_w and T_L. In practice, σ_w can be measured directly, but usually, T_L will be inferred from models or theoretical considerations. Raupach (1989a,b) discusses the various assumptions made in the analysis and the sensitivity of the analysis to its parameters, while various forms for the profiles of σ_w and T_L are described in Denmead et al. (2005). To summarise, once σ_w and T_L are known, then C_f and C_n, hence C_z, can be found through LNF analysis for any given source distribution.

The inverse problem is to find the source distribution from a given, measured concentration profile. Raupach (1989b) treats the problem in discrete form. The canopy is divided into m source layers, each with a uniform source density S_j, where $j = 1,\ldots,m$. For simplicity, any flux from the ground is assumed to be lumped in with the source from the lower layer ($j = 1$), although Raupach (1989b) states that this is not a necessary simplification. Scalar concentrations are measured at n heights z_i, the top height coinciding with the reference level z_R well above the canopy, where $C = C_R$. Concentrations at any level are influenced by the sources at all levels. Raupach (1989b) notes that the two sets of heights can be fixed quite independently and need not coincide in any way.

The first step in the analysis is to consider the concentration profile resulting from the uniform release of a scalar tracer in source layer j with source density s in that layer and zero elsewhere. From the developments for the forward problem, the resulting concentrations c_i can be found (Eq.4 and 7 and Raupach's (1989a) mathematical expression for C_n alluded to above). The c_i define a dispersion matrix D with elements D_{ij} given by

$$D_{ij} = \frac{c_i - c_R}{s\Delta z_j} \qquad [8]$$

Each D_{ij} is the concentration at measurement height i produced by a uniform source of unit strength extending through layer j, but zero elsewhere. As Raupach (1989b) observes, the matrix **D** can be calculated by any theory that solves the forward problem and, in particular, by LNF theory given the turbulence parameters σ_w and T_L. Once **D** is known, the concentration profile generated by any source profile S_j is given by the superposition of solutions. Thus

$$C_i - C_R = \sum_{j=1}^{m} D_{ij} S_j \Delta z_j \;. \qquad [9]$$

Leuning et al. (2000) outline simplified procedures for calculating the D_{ij}.

Raupach (1989b) noted that by choosing $m = n$ so that S_j is sought in the same number of source layers as there are concentration values C_i, Eq.[9] becomes a set of m linear equations which can be solved for the source profile S_j. He pointed out, however, that because small errors in C_i and the turbulence parameters can produce large errors in S_j, it is necessary to include redundant concentrations so that source densities are sought in m layers from n concentrations with $n>m$. He developed a least squares procedure to minimize errors in S_j.

Leuning (2000) introduced additional parameterizations to account for the effects of stable and unstable atmospheric conditions on σ_w and T_L. Stability has been shown to have quite large effects on the analysis. Leuning (2000), for instance, showed that in nighttime stable conditions, applying stability corrections reduced the predicted net fluxes of water vapour and CO_2 from a rice crop by >50% and gave results which agreed very well with independent measurements of the fluxes by eddy correlation, while Denmead et al. (2005) found increases of 30 to 40% in predictions of the net fluxes of heat, water vapour and CO_2 from a wheat canopy through applying Leuning's corrections in daytime unstable conditions, and similarly good agreement with eddy correlation measurements. Denmead et al. (2005) concluded that the stability correction scheme of Leuning (2000) is both adequate and necessary.

Denmead et al. (2005) discuss practical considerations in the application of inverse Lagrangian analysis. These include:

- the need for extensive, horizontally uniform canopies,
- the need to have more concentration measurements than source layers (8 conentration measurements and 4 source layers seems to be a reasonable sampling scheme),
- the need for correct specification of the T_L profile (the most successful are forms of the T_L profile in which T_L decreases with depth in the canopy),
- the need to take account of atmospheric stability,
- instrumental needs (using just one instrument to make the same measurement at all levels wherever possible).

Gas Exchange in the Canopy

There are many multiple-resistance analog models of canopy gas exchange which can be used to predict or measure source-sink distributions. Of particular interest here are models for bi-directional fluxes. Relevant examples are models of NH_3 exchange due to Sutton et al. (1995), Asman et al. (1998) and Nemitz et al. (2000b). These envisage NH_3 diffusing from some reference point above the canopy to the surface of a leaf through an aerodynamic resistance R_a and a leaf boundary-layer resistance R_b (using the terminology of Sutton et al. (1995)). They invoke the concept of an ammonia compensation point, χ_s, originally proposed by Farquhar et al. (1980). At the single leaf level, the compensation point is the NH_3 concentration within the leaf sub-stomatal cavity that is in equilibrium with the $[NH_4^+]$ in the leaf apoplast, governed by temperature and pH. If the NH_3 concentration in the air surrounding the leaf, $\chi(z)$ is higher than the compensation point, NH_3 is taken up by the leaf by diffusion through stomata with resistance R_s; if it is less, NH_3 is lost from the leaf by the same pathway. The flux of gas to or from the leaf is described by

$$F_g = (\chi_s - \chi(z))/(R_a(z) + R_b + R_s). \qquad [10]$$

The resistances R_a and R_b can be determined from wind and temperature profiles and R_s can be measured with a diffusion porometer.

Ammonia can also be adsorbed by dew and thin water films on leaf cuticles and subsequently desorbed. Cuticular leaf exchange is generally modeled as diffusion to or from the leaf surface across the leaf boundary layer and a cuticular resistance R_w. The concentration of NH_3 at the leaf surface is assumed to be zero. The concentration of NH_3 about the leaf, χ_c, is given by

$$\chi_c = \frac{\chi(z)(R_a(z) + R_b)^{-1} + \chi_s R_s^{-1}}{(R_a(z) + R_b)^{-1} + R_s^{-1} + R_w^{-1}}, \qquad [11]$$

and the canopy flux F_c by

$$F_c = -\frac{\chi_a - \chi_c}{R_a(z) + R_b}. \qquad [12]$$

Nemitz et al. (2000) describe how to extend this one-layer leaf model to a multi-layer canopy model with emission from a ground source. The extension will not be detailed here.

The same model is applicable to the exchange of other trace gases relevant to air quality. Conrad and Dentener (1999) list typical compensation points for soil and vegetation for selected trace gases. As well as NH_3, the list includes CO, OCS, NO, NO_2. We can add to these CO_2, for which the compensation point depends on whether the plant has a C3 or a C4 pathway, and SO_2 and O_3 for which the compensation points are generally assumed to be zero (Baldocchi et al., 1989; Asman et al., 1998). The necessary changes for those gases for which no compensation points exist are obvious.

Bi-directional Ammonia Fluxes

It is evident from the foregoing sections that coupling micrometeorological analyses of source-sink distributions in canopies with multi-layer models of gas exchange can provide a powerful tool for studying processes of canopy exchange, since it can identify not only the exchange sites and their source-sink strengths, but also the soil and plant processes involved. This is particularly useful for bi-directional fluxes. Published studies of NH_3 exchange provide some good examples. Harper et al. (2000), Nemitz et al. (2000a,b) and Denmead et al. (2000, 2005) have used both IL analysis and canopy gas exchange models to study pathways of NH_3 exchange in plant canopies. These investigations reveal various source-sink patterns for NH_3 emission and transport and permit an evaluation of the ability of plants in the canopy to trap NH_3 emitted from the soil.

Harper et al.(2000) examined the emission of NH_3 following overhead sprinkler application of dairy effluent to a dense corn crop 3.3m high with a leaf area index (LAI) of 5. The expectation was that most of the ammonia emission would be from the soil surface and that because of the high concentrations generated in the canopy space following the application of effluent, there could be substantial recapture of the emitted NH_3 by the foliage. The IL analysis indicated that although very high concentrations occurred in the canopy after the application (in excess of 400 $\mu gN\ m^{-3}$ immediately afterwards), very little NH_3 was recaptured. In one observation period, for instance, some 17% of the NH3 emitted from the soil was recaptured in the first hour after application, but the soil emission decreased subsequently and the foliage became a net NH_3 source. Overall, only 21% of the net emissions came from the soil and 79% from the foliage. Apparent compensation points calculated from Eq. [10] exceeded published values for corn by one to two orders of magnitude. It was concluded that NH_3 losses from the plants were predominantly from the foliage rather than by diffusion from leaf interiors. This suggests that ammonia-releasing residues can remain on the leaf surface for many days after effluent application.

Nemitz et al. (2000b) examined NH_3 exchange in a canopy of oilseed rape, 1.38m tall with an LAI of 5.3. Their IL analysis indicated that the litter layer on the ground surface was a source of NH_3, but all the NH_3 emitted from the litter was recaptured in the lower half of the canopy. The exchange of NH_3 with the atmosphere was governed by the top 0.5m of the canopy, which constituted a second source. The NH3 compensation points for leaves in this layer, calculated from measurements of apoplastic $[NH_4^+]$ and pH, indicated that no significant stomatal emission should have occurred, suggesting that the seed cases in the layer were the source of the NH_3 emitted to the atmosphere.

Denmead et al. (2005) report an analysis of emissions of NH_3 from a crop of sugarcane for several days after urea fertilizer was spread onto a trash mulch on the soil surface. The crop was 0.8m tall with an LAI of only 1.4. The IL analysis indicated that the foliage recaptured only 6% of the N emitted from hydrolysis of the urea despite the development of quite high NH_3 concentrations within the canopy, typically around 100 $\mu gN\ m^{-3}$. The resistance analog model of canopy gas exchange confirmed the rates of leaf uptake inferred from the IL analysis when the rates were small ($<1\ \mu gN\ m^{-2}\ s^{-1}$), but predicted much higher uptake rates than the IL analysis when they were $>2\ \mu gN\ m^{-2}\ s^{-1}$. The authors suggest that the discrepancy could be due to the very high rates of NH_3 uptake creating non-equilibrium conditions in the leaves because of the slow translocation of the absorbed NH_3. It should be noted that the concentrations of NH_3 within the canopy and the rates of leaf uptake observed here were orders of magnitude higher than those reported by Nemitz et al. (2000a). The ability of the canopy to recapture substantial amounts of NH_3-N emitted from fertilizer or plant residues at the ground is an important question in agricultural air quality that is still undecided.

It should be said that in all three of the examples considered above, the IL analysis predicted net canopy exchanges of NH_3 that were in excellent agreement with independent micrometeorological measurements in the air layer above the canopy, but this is not its primary purpose. The real benefit of the analysis is its ability to provide detailed information on the magnitude and distribution of scalar sources and sinks within the canopy in a non-disturbing, continuous way. When coupled with multi-layer canopy models of gas exchange, it provides a valuable tool for understanding and determining the many pathways by which reactive trace gases, and NH_3 in particular, are exchanged between canopy and atmosphere.

Conclusions

Coupling the IL analysis with canopy gas exchange models has demonstrated the existence of several different pathways for NH_3 exchange in the canopy, some expected, some not. They include long-lasting volatilization from residues of effluent sprinkled on the foliage, volatilization from fertilizer and plant residues at the ground surface; some of the canopy acting as an NH3 source, some of it as a sink; losses from foliage elements other than leaves, such as seed cases,; and absorption by canopy foliage. Just how efficient foliage absorption is in recapturing NH_3 volatilized from sites within the canopy remains an open question. The evidence is that recovery can be as much as 100% when volatilization rates (and canopy gas concentrations) are small, but it decreases as volatilization rates increase. Similar questions arise for other reactive gases such as SO_2, NO and NO_2: how much of the SO_2 deposited on canopies is absorbed by the foliage, how much reaches the ground; are chamber measurements of NO and NO_2 emissions from the ground good indicators of how much is emitted to the atmosphere? The models described here provide very useful tools for answering such questions.

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The Surface Boundary Layer of a Cattle Feedlot: Implications for Long-term Measurement of Ammonia Fluxes

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Abstract

Emissions of ammonia and other traces gases from cattle feedlots can impact air quality at local and regional scales. Unfortunately, there is uncertainty in current estimates of feedlot emissions, much of which stems from inadequate field measurements. Micrometeorological techniques are appealing choices for measuring trace gas losses because they: (1) integrate over large areas, (2) do not disturb the surface or affect aerodynamic transport, and (3) can conceptually be operated for long time periods on a near continuous basis. Regrettably, many micrometeorological methods are ill-suited for work at feedlots because of the potential for spatial variation in fluxes across the surface and inadequate upwind fetch. Furthermore, it is difficult to evaluate the potential of different methods because the characteristics of the surface boundary layer above feedlots have not been studied in detail. In this project, sonic anemometers and fast response gas analyzers were used measure the turbulent regime above a large commercial feedlot in Kansas. Data showed how weather, topography, pen surface conditions, and other factors affected the surface boundary layer. Analyses included the effect of advection arising from non-homogenous emissions as well as errors induced by sloping or hilly terrain. Results from the boundary layer study were used to evaluate the feasibility of different flux measurement techniques, including: the modified Bowen ratio, flux profile methods, conditional sampling, and relaxed eddy accumulation. Partitioning of ammonia flux into gaseous and aerosol components was included in the analysis. A central goal was to predict which technique, in theory, would be best for obtaining near continuous ammonia flux data over long time periods (e.g., one year). Development of these techniques is needed to improve measurement and modeling of the feedlot nitrogen balance. Determining the fate and transport of all nitrogen used in the feedlot operation is central to quantifying long-term environmental impacts.

An Integrated Approach to Measuring Emissions from Confined Animal Feeding Operations at the Whole Facility Scale

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Abstract

Agricultural operations produce a variety of particulates and gases that influence air quality. Agriculture, through wind erosion, tillage and harvest operations, burning, diesel-powered machinery and animal production operations, is a source of particulate matter that can enter human lungs and cause pulmonary problems. Animal production operations can be a source of gaseous emissions such as ammonia, odor-causing volatile organic compounds, hydrogen sulfide, greenhouse gases (methane, nitrous oxides) and airborne pathogens. These emissions can negatively impact human health, property values, and the environment. The presence of buildings and other structures often make whole facility measurement capability a requirement for understanding the source strength and characteristics. The ability to use standoff methods to determine the movement and concentrations of emissions on a whole facility basis opens new capabilities for model development and verification.

An integrated system to measure whole facility emission was designed to characterize the complex structures and temporally dependent emission rates often associated with production operations. This approach combines state of the art standoff measurement techniques with standard point source monitoring equipment to provide the calibrated, high spatial and temporal frequency data required to develop and validate the models required for emission reduction and regulation. This effort includes the design, construction and operation of a new multi-wavelength lidar developed to map and track particle emissions. The lidar incorporates a laser emitting simultaneous, pulsed NdYAG laser radiation at 355, 532 and 1064 nm at a pulse frequency of 10 kHz. The system also includes open path FTS measurements for integrated chemical concentrations, and state-of-the-art point measurements of turbulence, particulate and gas concentrations.

This approach was evaluated in a multidisciplinary atmospheric study at a swine production farm in Iowa. Aerosol plumes emitted from the facility were prominent phenomena, and their variations with temperature, turbulence, stability and feed cycle were studied, using arrays of particle samplers and turbulence detectors. Other lidar measurements focused on air motion as seen by long duration scans of the farm region. Successful operation of this system confirms the value of the multidimensional approach for the determination of agricultural emissions in the complex terrain often accompanying production facilities.

Lidar Studies

Emissions of gases and particulates from animal production facilities are of concern because of the potential environmental and health implications. However, the measurement of the emission rate of different gases or particulates has proven to be difficult because of the differences among facilities, variations within a facility caused by differences in building management (ventilation fans), and the interactions among buildings caused by the altered wind flow regime induced by buildings in close proximity to one another. Observations of ammonia concentration observed at different distances and heights above the ground surface adjacent to a 400 head grow-finish swine production facility throughout a production cycle revealed large variations within a day and among days (Hatfield and Pfeiffer, 2005). Much of this variation could not be explained by simple differences in wind speed or wind direction. This prompted a study that utilized a lidar system to measure the dispersion of particulates from this facility coupled with detailed measurements of the turbulence components of wind (Eichinger et al., 2005, Unpublished data).

The use of the lidar system to measure the complete air volume above this facility began to reveal the complexity of air flow and dispersion surrounding production facilities. It was observed that the vertical velocities of wind were 1-5 m s-1, which is much larger than expected. The turbulence regime surrounding the buildings was extremely variable depending upon the wind direction. These observations revealed how quickly a plume of particulates was emitted and dispersed from the facility (Eichinger, W., 2005, Unpublished data). These observations also began to reveal that single point observations located adjacent to a facility may not be capturing the air volume required for an accurate measurement of the emission rates.

These preliminary studies have prompted the development of a more sophisticated lidar system to provide a measure of the particulate size fraction and potentially the gaseous composition of the air volume surrounding an animal production facility. To more accurately quantify the emissions from animal facilities it is necessary to quantify the turbulence regime in order to understand the rate and pattern of dispersion and couple these mechanisms with an improved estimate of the composition of the atmosphere. The goal of these studies is to describe the development of an improved method to quantify the emission rates and dispersion patterns from animal production facilities.

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New Measurement Protocol for the Determination of NH_3 Emission Factors from Animal Houses in the Netherlands

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Abstract

The Dutch government uses ammonia emission factors, which are determined according to standardized measurement protocols, to assign year-averaged ammonia emission levels to specific animal housing systems and animal categories. This protocol relies on intensive measurements at a single location, and therefore provides a large amount of data that can be used to study the factors controlling the ammonia emission processes. However, the costs of these measurements are usually large. In addition, this approach does not take into account the effect of farm management on the ammonia emission that apperars to be a major variation source and restricts the accuracy of the current single location based protocol.

To deal with these problems, a new measurement protocol has been proposed, which is based on measurements on a larger number of animal houses with the same housing system. In addition, continuous measurements are no longer required. Instead, daily averages could be used, which decreases the requirements for the equipment used in the measurements. Besides, the proposed measurement protocol requires only a limited number of measurement days per location, equally distributed (but at random) over the whole year, to take seasonal effects into account.

Introduction

The deposition of ammonia (NH_3) and ammonium (NH_4^+) can cause a large number of environmental problems, including soil acidification, eutrophication, nitrogen leaching into groundwater and loss of biodiversity (Heij and Schneider, 1991; Bobbink *et al.*, 1992). To reduce NH_3 emissions, emission ceilings have been assigned for all EU member states (EU, 2001). In order to comply with the emission reduction assigned to the Netherlands, development of new measures is needed, which should be supported by fast and accurate measurements to arrive at new estimates of the NH_3 emission factors from each agricultural source.

Agriculture is, by far, the most important contributor to the total NH_3 emission in the Netherlands, with emissions from animal housing and after the application of manure being of particular importance (Sliggers, 2001). To determine the yearly NH_3 emission from specific animal housing systems and animal categories, NH_3 emission factors are usually applied. The measurement protocol used to establish these emission factors (Anonymous, 1996) relies on intensive measurements at a single location. This sampling scheme provides useful information to study the factors that influence NH_3 emissions. However, it only takes variation within an animal house into account, and neglects the possible importance of management variation between different farm locations with the same housing system, which has proven to be an important variation source for odor (Ogink and Klarenbeek, 1997). Besides, costs of these measurements are high, which restricts the number of new animal housing systems that can be measured and therefore limits the development of new and better housing systems.

To deal with these problems a new measurement protocol has been proposed. The objective of this paper is to summarize the basic pillars on which this protocol has been funded.

Methods

To quantify and assess the importance of the different sources of variance associated with NH_3 emission measurements from animal houses, a statistical analysis was performed. First of all, all available datasets were classified according to the following aspects:

- Animal category
- Housing system (per animal category)

- Measurement method and strategy used for the measurements

Then, the emission data (daily averages) were transformed to natural logarithmic values to obtain a normal distribution with dispersion independent of the emission levels (De Boer and Ogink, 1994). The transformed data were analyzed with the REML (Residual Maximum Likelihood) directive of the GENSTAT 6.1 (6th edition) statistical package, to quantify the following sources of variance:

- Variance between farms (σ^2_b)
- Variance within a farm (σ^2_w)
- Variance of measurement equipment (σ^2_m)

Finally, the total variance (σ^2_t) associated with measurements according to a specific sampling scheme was determined by using a model developed by Ogink and Klarenbeek (1997) for odor emissions:

$$\sigma_t^2 = \frac{\sigma_b^2}{k} + \frac{\sigma_w^2}{k \cdot l} + \frac{\sigma_m^2}{k \cdot l \cdot m}$$

With k the number of farms, l the number of measurements (sampling days) within a farm, and m the replicate number of measurements at a sampling day.

Seasonal effects and effects of growth cycles on emission patterns were investigated by expressing daily emissions as a percentages of the average emission (over year or growth cycle), and studying the relative patterns in time.

Results and Discussion

Analysis of all available datasets for fattening pigs and sows shows that not only the variation within the animal house, but also the variation between farms is of importance (Mosquera and Ogink, 2004). In contrast, the accuracy of the measurement equipment used for the measurements seems to have relatively low importance. In order to improve the accuracy of the emission measurements (more accurate emission factor), it is advised to increase the number of locations to be measured rather than to increase the number of measurements in a particular location or to improve the accuracy of the measurement equipment. Therefore, continuous measurements are no longer necessary, and the number of measurement days per location can be largely reduced (Vranken *et al.*, 2004). Besides, since the requirements for the equipment can be reduced, alternative methods (Mosquera *et al.*, 2002, 2005) can be applied. Since some factors affecting NH_3 emissions from animal houses follow a diurnal pattern, use of daily averages is highly recommended. Analysis of growth cycles of fattening pigs and emission showed consistent strong linear relationships. The available emission data were not sufficiently even distributed over the seasons to allow for a clear identification of seasonal effects on emissions. However we surmise that such effects do exist, and may differ between animal categories In order to take potential seasonal effects into account in sampling strategies, a year-round sampling strategy is advised.

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