



NH₃ Emission from Pig Husbandry in Relation to Ventilation Control and Indoor Air Cooling

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Abstract

Animal husbandry is known as one of the main contributors to agricultural NH₃ emission, which might lead to acidification and eutrophication of soils and waters. NH₃ is released indoors mainly from urine and stored excrements. Thereby, the NH₃ release is related to animal activity, pen fouling, ventilation, indoor temperature and humidity. The influence of ventilation and indoor air climate on the diurnal and seasonal emission rate of NH₃ was investigated in this research work. Different ventilation control strategies were tested in a research pig facility over a total of four fattening periods. Throughout this time, NH₃ concentrations were measured continuously by NDIR spectroscopy, indoors, as well as in the incoming and exhaust air. The ventilation rate was determined using calibrated measuring fans. The ventilation strategies featured adiabatic indoor air cooling, and either an increased, activity controlled, or reduced, CO₂ and temperature controlled ventilation rate. At days with mean daily outside temperatures above 14 °C, indoor air cooling resulted in a reduction of the diurnal maximum temperature by about 4 to 5 K, the ventilation rate was reduced by about 22 %. The combination of indoor air cooling and CO₂ control did furthermore reduce the ventilation rate by about 33 %, at mean daily outside temperatures above 14 °C, and by about 20 % for lower outside temperatures.

The mean NH₃ emission rate of the reference ventilation differed significantly between mean daily outside temperatures below and above 14 °C. Nevertheless, highest mean NH₃ emission rates, 130 g d⁻¹ LU⁻¹ and 120 g d⁻¹ LU⁻¹, occurred during the two fattening periods in early and late spring. At mean daily outside temperatures below 14 °C, NH₃ emission were furthermore increased significantly for ventilation with evaporative indoor air cooling, in average by about 10 % to 14 %. In contrast, no significant differences in NH₃ emission occurred between the different ventilation strategies at warm outside temperatures above 14 °C. Hence, in order to reduce NH₃ emission by adiabatic indoor air cooling, an optimized fogging control, regarding complete evaporation of the water, was required. On a yearly basis (330 fattening days), the mean measured NH₃ emission rate ranged at 5,1 kg NH₃ pig place⁻¹ year⁻¹.

Introduction

Main sources of NH₃ in animal husbandry systems are urine and excrements. Beside feed composition, mainly the storage duration and management of excrements in the stable, the indoor air climate, fouled surfaces, as well as the animals themselves exert an influence on the release of NH₃. Ventilation control directly influences the ventilation rate, the indoor temperature, and humidity, but also the fouling behaviour of the pigs, and thus the emission of NH₃. The influence of the indoor air climate on pen fouling and hence on the NH₃ emission rate was defined by Ni (1998) as floor factor and was calculated using the indoor temperature and the weight of the animals as input variables. The indoor temperature and air velocity exerts furthermore a dampened influence on the slurry temperature and thereby increases or lowers the release of NH₃ from the slurry storage (Berckmans et al. 1994). Nevertheless, high air velocities and turbulences might also increase the NH₃ release, caused by a dilution effect on the air above the slurry surface (Hartung, 1995). The quantification of the effects of different influencing variables which are contrary to each other and/or are related to each other, like the ventilation rate and the indoor temperature, can in general be difficult. Although the ventilation rate and NH₃ emission were clearly positive correlated ($R^2 = 0.729$) during investigations of Ni (1998) in mechanically ventilated pig facilities, no or even negative correlations between the ventilation rate and the NH₃ emission rate were found by Gallmann (2003) in a natural ventilated building for fattening pigs, in which the ventilation rate was not controlled by the indoor temperature. High variations in measured emission rates have their origins furthermore in different regions,

facilities, ventilation systems, seasons, management, as well as in the varying emission course throughout the fattening period (Aarnink, 1997; Groot Koerkamp et al. 1998; Ni, 1998; Gallmann, 2003). The potential to lower NH₃ emission by ventilation control strategies can be estimated with up to 35 %, based on investigations on ventilation effects in mechanically and natural ventilated buildings (Keck, 1997; Ni, 1998; Gallmann et al., 2003). However, the verification of such a reduction potential in mechanically ventilated buildings during long term measurements is hardly performed. Thus, the aim of this investigation was to study the influence of evaporative indoor air cooling in a facility for fattening pigs on the emission of NH₃, and to compare the effects of different ventilation strategies, which use additional control variables (animal activity and CO₂ indoor concentration), and hence feature either an increased or a lowered ventilation rate.

Methods

Research Facility

Investigations on ventilation control, indoor air cooling, and NH₃ emissions were carried out in two separately compartments (54 pigs each, 0.9 m² pig⁻¹) at the research facility for fattening pigs (Hartung, 2001), University of Hohenheim. Fattening started at an average weight of 25 to 30 kg per pig. The measurements were performed until the pigs reached an average weight of approximately 105 kg. Each compartment featured two pens, equipped with a slotted concrete floor and a slurry pit underneath each pen (Figure 1). The pens were subdivided into a lying and feeding area and an excremental area, characterized by a ratio of the slotted floor area of 14 % and 6 %, respectively. Feeding was supplied either with a sensor liquid feeding system or an ad libitum mash feeder. The feed composition and protein content of the food was equal for both feeding systems and was adapted in fattening week 2, week 5, week 8, and week 11. Straw was supplied for occupation. The slurry was sampled in the slurry pits during fattening, and was completely remove after each fattening period (Haeussermann et al., 2004).

Ventilation System and Tested Ventilation Control Strategies

The mechanical ventilation system was designed as under-floor extraction, and was equipped with one separately controllable ventilation fan per compartment. Fresh air was supplied via two air inlet pore channels per compartment, each arranged centrally above the animal area. The ventilation rate was controlled either by temperature (Reference and Strat. B), temperature and animal activity (Strat. A), or by CO₂ indoor concentration and temperature (Strat. C) (Table 1).

Table 1: Overview on ventilation control strategies and control input variables

ventilation control strategy	Reference - no humidifying -	Strat. B - humidifying -	Strat. A - humidifying -	Strat. C - humidifying -
control variables*	Temperature ¹	Temperature ^{1,2} Humidity ²	Temperature ^{1,2} Humidity ² Animal activity ^{1,2}	CO ₂ indoor concentration ¹ Temperature ^{1,2} Humidity ²

* control input for: ¹ventilation rate ²fogging system

For each of these ventilation strategies, the set temperature (T_{set}) decreased non-linearly from 25 °C to 16 °C during the fattening period. Using Strat. A, the ventilation rate was controlled by temperature but was increased during short-termed periods when the group animal activity was high. In contrary, the ventilation rate was kept on minimum at Strat. C until either the maximum value for the CO₂ indoor concentration (2000 ppm) or the maximum value for the indoor temperature ($\Delta T_{set} = 3$ K) was exceeded. The four ventilation strategies were distributed randomly on the two compartments and on the four fattening periods. At the first two fattening periods, each strategy was tested throughout three-week long measuring sections, once per compartment per fattening period. At the last two fattening periods, the ventilation strategies were tested twice per compartment per fattening period, but throughout half the length of a measuring section (10/11 days) at each time (Haeussermann et al., 2004).

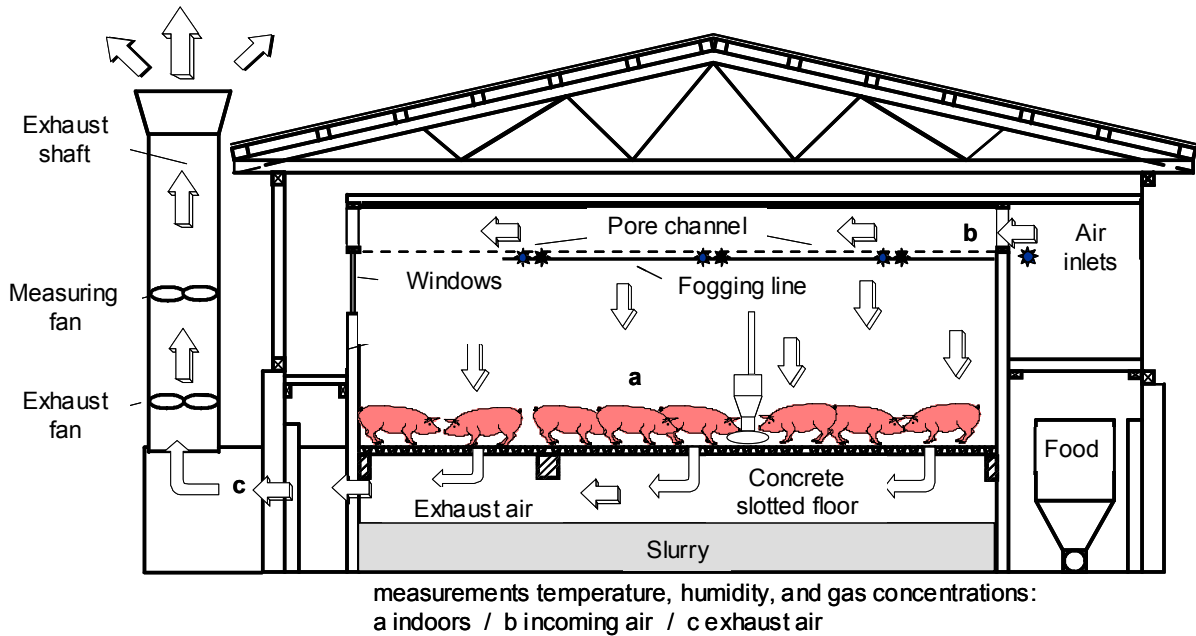


Figure 1. Vertical projection (west-east), research facility for fattening pigs, University of Hohenheim (modified, adopted from Hartung, 2001, Gallmann 2003)

Fogging System and Control

Two separately controllable fogging lines per compartment were used to cool down and to humidify the air (Figure 2). They were placed one inside the compartments (3 nozzles per pen) and a second one in front of the air inlets (2 nozzles per inlet). Water supply was 885 ml per minute per compartment when both fogging lines were operated. The pressure of the pump was fixed at 7 MPa. During the investigation, fogging was operated in a pulse-cycle, set on four minutes fogging-on and three minutes fogging-off. A fogging cycle started either when the indoor temperature rose to more than 1.5 K above T_{set} (Strat. B, Strat. C, and Strat. A), when the indoor humidity dropped below 50 % (Strat. B, Strat. C, and Strat. A), or by the signal of the group animal activity (Strat. A). Maximum indoor humidity was set on 80 % with a hysteresis of 10 %, minimum indoor temperature for fogging equalled T_{set} .



Figure 2. Air humidifying at the incoming air and indoors using separately controllable fogging lines with four and six high pressure nozzles respectively per compartment

Measurements

During a total of four fattening periods, outside and indoor temperature, relative humidity, and the group animal activity were measured continuously with one average value per 15 min (Table 2). NH₃ and CO₂ concentrations indoors, as well as in the incoming and in the exhaust air were measured at the same frequency using NDIR spectroscopic gas analyzers. For the gas analysis, the air was sucked through condensation protected Teflon tubes into the measuring chamber of the gas analyzers, placed in a temperate room outside the compartments. The air sampling was rotated every 150 sec between the six sampling positions (indoor air, incoming air, exhaust air of compartment 1 and compartment 2). An average value of the respective sampling position was calculated from the last 30 sec, while the first 120 sec were used to compensate maximum gas transport times as well as rise and decay times of the measuring instruments in relation to the high differences in gas concentrations between the sampling points. A correction of the measured NH₃ concentration was performed subsequently to the measurements according to the water vapour cross sensitivity of the gas analyser and the water content of the air at the respective sampling position (Brose, 2000). Parallel to the gas measurements, the ventilation rate was determined using a calibrated measuring fan (Table 2). The calculation of the NH₃ emission rate took into account the difference in the gas concentration at the exhaust and incoming air, the ventilation rate, as well as the animal weight, calculated as Livestock Units (1 LU = 500 kg). The strategies were compared by their mean values and inter-quartile ranges (25th and the 75th percentiles of the measured mean daily values).

Table 2. Measured variables, measuring instruments, frequencies and accuracies

	Measuring instrument	measuring range / accuracy	sampling rate	measuring frequency
Ventilation rate	measuring fan MULTIFAN	200 – 10000 m ³ /h +/- 20 m ³ /h	1 sec	15 min
Air temperature	resistance (PT 100) Hygroclip, ROTRONIC	- 40 to + 85 °C +/- 1 °C	1 sec	15 min
Relative humidity	capacitive Hygroclip, ROTRONIC	0 – 100 % +/- 1 %	1 sec	15 min
Animal activity	passive infrared sensors (Pedersen and Pedersen, 1995)	0 – 5 V (scale)	1 sec	15 min
CO ₂	NDIR – Spectroscopy Unor 610, MAIHAK	0 – 10000 ppm +/- 10 ppm	1 sec	15 min
NH ₃	NDIR – Spectroscopy Binos 4b, ROSEMOUNT	0 – 100 ppm +/- 1 ppm	1 sec	15 min

Results and Discussion

NH₃ emissions averaged at 130 g d⁻¹ LU⁻¹ and 120 g d⁻¹ LU⁻¹ during the two fattening periods in spring and at 97 g d⁻¹ LU⁻¹ and 94 g d⁻¹ LU⁻¹ in the summer period and in the winter period, respectively. The total variation of the mean daily NH₃ emission rate was in a range of 54 to 181 g d⁻¹ LU⁻¹ or 5 to 26 g d⁻¹ pig⁻¹ and thus comparable to the upper range of mean values reported in literature for fattening pigs, kept on fully or partly slatted floors (23 to 150 g d⁻¹ LU⁻¹ or 5 to 20 g d⁻¹ pig⁻¹; Aarnink (1997); Keck (1997); Groot Koerkamp et al. (1998); Ni (1998); Hahne et al. (1999); Heber et al. (2000); Navarotto et al. (2000); Rom and Dahl (2002); Vranken et al. (2002); Demmers et al. (2003); Gallmann (2003); Guarino et al. (2003); Guingand (2003); Van den Weghe et al. (2005). Throughout the whole investigation period, an average emission rate of 5,1 kg NH₃ animal place⁻¹ year⁻¹ was measured (330 fattening days per year).

The mean diurnal course of the NH₃ emission was mainly influenced by the animal activity, the ventilation rate, and the indoor temperature. Throughout the year, an increase of the daily NH₃ emission rate was pronounced seasonally especially during spring and autumn. In order to compare the ventilation control strategies, days with mean daily outside temperatures below and above 14 °C were considered separately.

Effects of the Ventilation Strategies at Mean Daily Outside Temperatures Below 14 °C

During days with mean daily outside temperatures below 14 °C, only minor influences of the different control strategies on the indoor air temperature and ventilation rate occurred. Average values as well as the

total variation of the values were mainly predetermined by the control settings (Table 3 and Table 4). However, the ventilation rate was significantly lowered for the CO₂ controlled ventilation strategy (Strat. C), in average by about 20 % compared to the Reference (Table 3). Thus, the mean indoor temperature of Strat. C was increased by about 1 K (Table 4). At mean daily outside temperatures below 14 °C, water fogging was mainly used for increasing the relative humidity on an inter-quartile range of 51 % to 61 % (Strat. B and Strat. A), compared to 44 % to 52 % (Reference). When using Strat. C, however, water fogging was additionally applied to support indoor air cooling.

Table 3. Ventilation rate [m³ h⁻¹ pig⁻¹] at mean daily outside temperatures below >T1< and above 14 °C >T2<]

	Reference	Strat. B	Strat. C	Strat. A
mean >T1<	45^b	46^b	36^a	46^b
Inter-quartile range	31 – 51	34 – 55	29 – 40	34 – 56
mean >T2<	119^c	93^b	80^a	92^b
Inter-quartile range	120 – 131	65 – 127	57 – 100	67 – 117

^{a, b, c} means with different superscript letters differ significantly (p < 0,05)

Table 4. Indoor temperature [°C] at mean daily outside temperatures below >T1< and above 14 °C >T2<]

	Reference	Strat. B	Strat. C	Strat. A
mean >T1<	19,2^a	18,8^a	19,9^b	18,9^a
Inter-quartile range	16,6 – 21,0	17,3 – 20,2	18,6 – 21,4	17,9 – 20,1
mean >T2<	24,5^c	20,4^a	21,8^b	20,4^a
Inter-quartile range	21,3 – 27,5	18,4 – 22,2	19,4 – 23,2	18,6 – 21,9

^{a, b, c} means with different superscript letters differ significantly (p < 0,05)

Considering NH₃ emissions during mean daily outside temperatures below 14 °C, the three ventilation strategies based on humidifying (Strat. B, Strat. A; Strat. C) featured significantly, in average 10 % to 14 % higher NH₃ emissions when compared to the reference strategy (Table 5). As the evaporation rate of the fogged water depends largely on indoor temperature and humidity (Haeussermann et al., 2005), an incomplete evaporation of the fogged water during cold and humid or during moderate warm days was likely to cause humid and subsequent fouled surfaces. The relation between pen fouling and the release of NH₃ or the NH₃ emission rate is reported among others in Aarnink (1997), Ni (1998) and Gallmann (2003). According to Aarnink and Elzing (1998), an increase in pen fouling causes an increase in the percentage rate of NH₃ released from the floor surface, from 30 % towards up to more than 40 % of the total NH₃ release.

Table 5. NH₃ emission rate [g d⁻¹ LU⁻¹] at mean daily outside temperatures below >T1< and above 14 °C >T2<]

	Reference	Strat. B	Strat. C	Strat. A
mean >T1<	99^a	113^b	109^b	113^b
Inter-quartile range	84 – 113	87 – 142	82 – 125	90 – 140
mean >T2<	114^b	110^b	113^b	110^b
Inter-quartile range	86 – 131	77 – 138	100 – 121	79 – 128

^{a, b, c} means with different superscript letters differ significantly (p < 0,05)

Effects of the Ventilation Strategies at Mean Daily Outside Temperatures Above 14 °C

Clear influences of the ventilation control strategies on the ventilation rate and indoor air temperature occurred at mean daily outside temperatures above 14 °C. Here, the ventilation rate was reduced in average by about 22 % when using the fogging system (Strat. B and Strat. A). The combination of adiabatic indoor air cooling and a partly CO₂ controlled ventilation rate (Strat. C) did furthermore result in a reduction of the mean ventilation rate by about 33 % (Table 3). Comparing mean daily outside temperatures below and above 14 °C, the indoor temperature was approximately 1.5 K to 5.3 K higher at the latter, whereof the total difference was depending on the ventilation strategy (Table 4). Due to evaporative indoor air cooling (Strat. B, Strat. C, and Strat. A), diurnal temperature peaks were lowered in maximum by about 7 K, in average in >T₂< by about 4 to 5 K. When using the fogging system, the inter-quartile range of the relative indoor humidity was increased on 64 % to 82 % (Strat. B, Strat. A, and Strat. C).

Based on the significantly reduced ventilation rate and indoor temperature, a reduction of the NH₃ emission rate was expected for those ventilation strategies which cool the indoor air by fogging of water. However, no significant differences occurred between the mean NH₃ emission rate of the different ventilation control strategies at mean daily outside temperatures above 14 °C (Table 5). A seasonal influence on the NH₃ emission rate was indicated by the significant differences between mean daily outside temperatures below and above 14 °C of the reference ventilation. However, an increase of the NH₃ release, caused by incomplete evaporation of the fogged water, and subsequently fouled surfaces, has to be assumed also for mean daily outside temperatures above 14 °C. Thus, the reduction of the NH₃ emission rate due to the lowered ventilation rate and lowered indoor temperature can be estimated approximately with 10 % to 14 %, but has to take into consideration all multiple influences. Resulting from the investigations, the reduction of the indoor temperature and ventilation rate was sufficient to counteract an increased NH₃ emission rate, caused by incomplete evaporation and fouled surfaces. Nevertheless, the reduction of emitting NH₃ from animal facilities by evaporative indoor air cooling would require furthermore an optimisation of the fogging control with regard to an continuously complete evaporation of the water.

Conclusions

The influences of ventilation control (ventilation rate and indoor temperature), and adiabatic indoor air cooling on the emission of NH₃ were investigated in a research pig facility throughout four fattening periods. The following conclusion were drawn:

- The yearly emission rate averaged on 5,1 kg NH₃ animal place⁻¹ year⁻¹ (330 fattening days);
- The mean diurnal course of the NH₃ emission rate was mainly influenced by the animal activity, the ventilation rate, and the indoor temperature;
- Throughout the year, an increase of the mean daily NH₃ emission rate was pronounced seasonally especially during spring and autumn;
- NH₃ emissions were increased for ventilation strategies with humidifying at mean daily outside temperatures below 14 °C. For warmer outside temperatures, however, this increase was counteracted by the clear reduction of the indoor temperature and ventilation rate;
- The potential to reduce NH₃ emissions from mechanically ventilated pig facilities by the reduction of the indoor temperature and ventilation rate can be estimated with 10 % to 14 % during mean daily outside temperatures above 14 °C;
- Nevertheless, in order to realize this reduction potential for NH₃ emissions, using evaporative indoor air cooling, an optimisation of fogging control, with regard to an continuously complete evaporation of the water was required.
- Due to the effect of the fogging system and incomplete evaporation of water on the NH₃ emission rate, no estimation of separate effects of temperature or ventilation rate can be concluded from these investigations.

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Emissions of Particulate Matter (PM₁₀) from Housing Systems for Fattening Pigs

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Abstract

Indoor and exhaust concentrations of aerosols with an upper particle size limit of 10 µm (PM₁₀) were measured discontinuously during two fattening periods. The investigation were performed in a research pig facility, equipped with a mechanical under-floor ventilation system. Continuous measurements included indoor temperature, humidity, ventilation rate, and animal activity. Indoor and exhaust concentrations of airborne dust were sampled discontinuously during full day periods. For the measurements, calibrated scatter light photometers (TSI, Model DustTrak 8520) has been used. Measuring frequency was 1 minute. Thereby, the correlation between the indoor and exhaust PM₁₀ concentration was generally high, featuring an R²_{adj} of 0.82. Nevertheless, for low ventilation rates during winter, only 65 % of the aerosols, measured indoors, were transported to the outside, while for high ventilation rates in summer, the percentage between indoor and exhaust concentrations was clearly increased on 95 %. Similarly, the mean PM₁₀ emission rate was increased from 4.2 g d⁻¹ LU⁻¹ in winter towards 7.2 g d⁻¹ LU⁻¹ during the summer period.

Introduction

The release of dust particles in animal husbandry is influenced mainly by feeding operations, used bedding materials, animals (particles from skin and excrements), as well as the kind of ventilation and management (Aarnink et al., 1999; 2004; Hartung, E. et al., 2004; Keck et al., 2004). It is well known that aerosols and bioaerosols in animal husbandry can cause respiratory health problems (Donham et al. 1995, Seedorf and Hartung, J. 2002). Thus, possibilities to influence the indoor dust concentration in pig husbandry has been investigated intensively during the last years (CIGR, 1994; Pedersen et al., 2000). Nevertheless, only few information about ranges of particulate emissions in relation to season, husbandry systems or regions exists. High ventilation rates during summer are accompanied in general with low indoor concentrations but increased emission rates, especially of inhalable and PM₁₀ particle size fractions (Takai et al., 1998; Aerts et al., 2004; Jacobson et al., 2004; Koziel et al., 2004).

Methods

Indoor and exhaust concentrations of PM₁₀ aerosols were measured discontinuously during two fattening periods (Haeussermann et al., 2006). The investigations were performed in a research pig facility (Hartung, E., 2001), equipped with a mechanical ventilation system (Figure 1). The two compartments of the research facility featured two pens each, 27 pigs per pen, and were equipped with a slotted concrete floor and a slurry pit underneath each pen. Fresh air was supplied via two air inlet pore channels per compartment, the outgoing air was extracted under-floor (Figure 1). Feeding was supplied either liquid, using a sensor feeding system (20 feeding times per day), or ad libitum using a mash feeding system. Straw was offered via an occupation equipment.

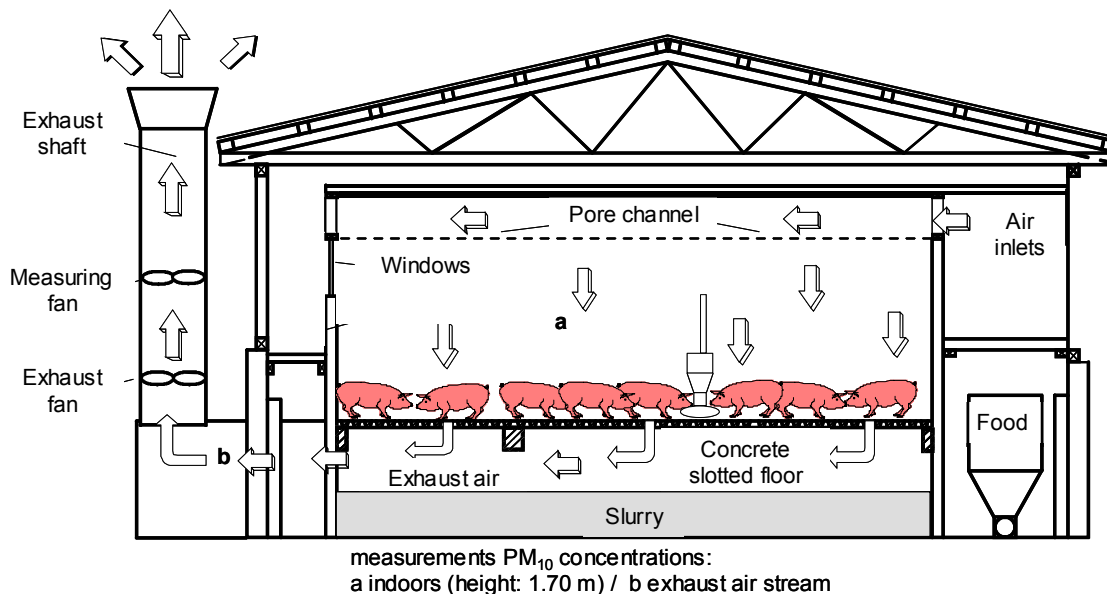


Figure 1. Vertical projection of the research facility for fattening pigs (modified, adopted from Hartung, E. 2001; Gallmann, 2003)

Measurements

Continuous measurements included ventilation rate, indoor temperature, humidity, and animal activity (Table 1). Airborne PM₁₀ particles (particles < 10 µm) were monitored discontinuously during full day periods, simultaneously at one representative measuring point indoors as well as in the exhaust shaft (Figure 1). For the PM₁₀ measurements, calibrated scatter light photometers were used (Table 1). The measuring frequency for dust, ventilation rate, indoor temperature, humidity, and animal activity was one average value per minute. The average weight of the animals was determined every three weeks and was interpolated for time periods in between.

Table 1. measured variables, measuring instruments, frequencies and accuracies

	Measuring instrument	measuring range / accuracy	measuring frequency
Ventilation rate	measuring impeller MULTIFAN	200 – 10000 m ³ /h +/- 20 m ³ /h	1 min
Air temperature	resistance (PT 100) Hygroclip, ROTRONIC	- 40 to + 85 °C +/- 1 °C	1 min
Relative humidity	capacitive Hygroclip, ROTRONIC	0 – 100 % +/- 1 %	1 min
Animal activity	passive infrared sensors (Pedersen and Pedersen, 1995)	0 – 5 V (scale)	1 min
PM ₁₀	Scatter light photometer TSI, DustTrak 8520 TM	0,001 – 100 mg m ⁻³ +/- 0,001 mg m ⁻³	1 min

Results

The correlation between the indoor and exhaust PM₁₀ concentration was generally high, featuring an R²_{adj} of 0.82 (0.89 and 0.61 in winter and summer, respectively, Figure 2). Nevertheless, for low ventilation rates during winter, only 65 % of the aerosols, measured indoors, were transported to the outside, while for high ventilation rates in summer, the percentage between indoor and exhaust concentrations was clearly increased on 95 %. Similarly, the mean PM₁₀ emission rate was increased from 4.2 g d⁻¹ LU⁻¹ in winter

(mean ventilation rate: $291 \text{ m}^3 \text{ h}^{-1} \text{ LU}^{-1}$) towards $7.2 \text{ g d}^{-1} \text{ LU}^{-1}$ during the summer period (mean ventilation rate: $735 \text{ m}^3 \text{ h}^{-1} \text{ LU}^{-1}$) (Table 2). Beside ventilation, the level of the emission rate was influenced to a high range by the animal weight and animal activity. Nevertheless, relations between influencing variables and the PM_{10} emissions, as well as between indoor and exhaust concentrations, differ at different housings and husbandry systems. Hence they have to be investigated separately for each individual system.

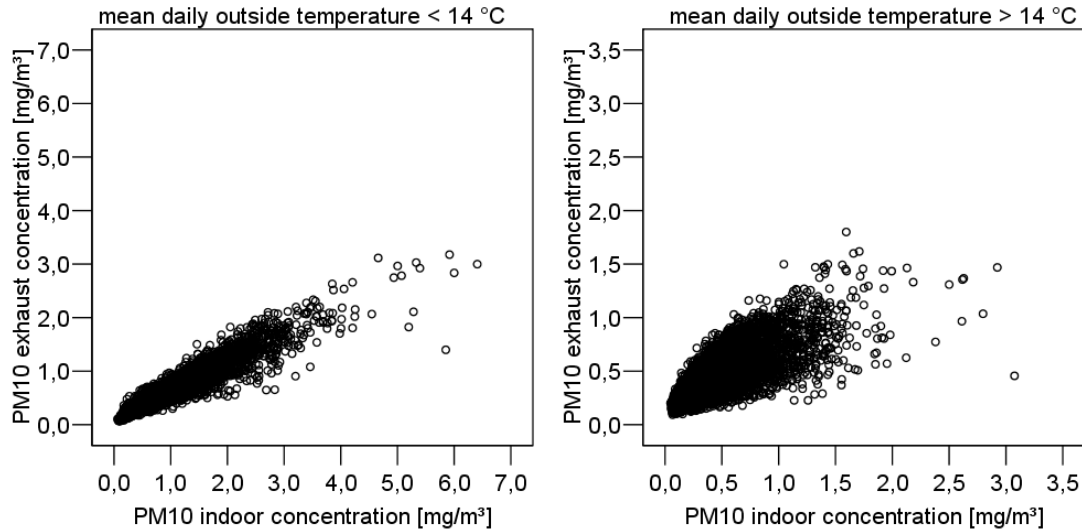


Figure 2. range diagrams of the PM_{10} indoor and exhaust concentration during the winter and summer period (mean daily outside temperature below and above $14 \text{ }^\circ\text{C}$, respectively)

Table 2. Average ventilation rate, PM_{10} concentration and PM_{10} emission during the winter and the summer period

	Ventilation rate [$\text{m}^3/\text{h}/\text{LU}$]	PM_{10} indoor concentration [mg/m^3]	PM_{10} exhaust concentration [mg/m^3]	PM_{10} emission rate [$\text{g}/\text{d}/\text{LU}$]
Winter period ($t_0 < 14 \text{ }^\circ\text{C}$)	291 (235 – 372)	1.03 (0,57 – 1,26)	0,58 (0,37 – 0,79)	4,2 (3,3 – 5,2)
Summer period ($t_0 > 14 \text{ }^\circ\text{C}$)	735 (598 – 854)	0.44 (0,35 – 0,56)	0,43 (0,30 – 0,67)	7,2 (5,5 – 9,5)

t_0 : mean daily outside temperature (): total range (mean values separate measuring periods)

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Using Physical Models to Simulate Emissions from Anaerobic/Facultative Swine Manure Stabilization Ponds

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Abstract

Researchers at Oklahoma State University (OSU) have simulated emissions from swine manure stabilization ponds under controlled conditions using a pilot bioreactor consisting of four 270 liter, 3.66 m deep columns that recreate environmental conditions found in anaerobic/facultative lagoons. The pilot bioreactor was equipped with both head-space and wind-tunnel air sampling devices. When fed manure produced by hogs eating a typical corn-soybean diet and programmed to simulate late summer conditions, the pilot bioreactor showed that 65% of applied carbon was emitted as CH₄ and CO₂, and 72 % of applied nitrogen was emitted as NH₃. There was a marked difference in the CH₄:CO₂ ratio between night and day emissions, with higher ratios occurring during the night. NH₃ emissions greatly increased during daylight hours once the temperature of the liquid surface began to rise.

Introduction

Most manure handling systems found on swine farms in the southern United States use anaerobic stabilization ponds – commonly called lagoons -- to treat liquid manure. Effluent is recycled to the buildings to remove manure, and excess effluent is irrigated to cropland. Stabilization ponds are low labor, easily managed treatment systems, and are very efficient at removing organic matter from the waste stream (Hamilton et al, 2002). The overall effect of lagoon treatment is to covert organic matter to gases. Emitted gases are byproducts of the carbon, nitrogen, and sulfur cycles of the pond ecosystem, and include the problematic gases: methane (CH₄), carbon dioxide (CO₂), hydrogen sulfide, oxides of nitrogen, and ammonia (NH₃).

The complexity of making measurements of full-scale stabilization ponds is a major factor in the limited availability of lagoon emission data in the literature. The difficulty arises from two hindrances: the ability to study cause and effect relationships under controlled conditions, and the ability to accurately measure emission of gases over the entire lagoon surface (Hamilton et al, 2006). Researchers at Oklahoma State University (OSU) have attempted to overcome these hindrances by constructing a pilot bioreactor consisting of four 270 l, 3.66 m deep columns that recreate environmental conditions found in anaerobic/facultative lagoons. By precisely controlling the environmental (heating, lighting, mixing) and operational conditions of the column section, cause and effect relationships can be explored. The problem of surface measurement is reduced by assuming that the layered biological communities found in lagoons can be recreated in the 3.66 m of column depth, and that the gases emitted from the top of this column represent emissions from the total lagoon surface.

Results and Discussion

The model pilot bioreactor used in this study is made of four insulated PVC pipes, 0.305 m (1 ft) in nominal diameter, and 3.66 m (12 feet) in depth. Hamilton (1998) gives a complete description of bioreactor design. Two sets of two columns are coupled with the same lighting and heating systems to achieve two replicated sets of conditions from the four columns. Columns are labeled A, B, C, and D; and are usually operated as two, two-column sets (AB and CD). The pilot facility was inoculated with effluent and sludge from a facultative lagoon located at the OSU Swine Research Center (OSU-SRC) in the fall of 1998, and has been programmed ever since to repeat the heating, mixing, and lighting patterns experienced by a typical anaerobic/facultative stabilization pond under late-summer conditions at 35° N latitude in Oklahoma. Swine manure fed to the pilot bioreactor was collected from pigs housed in metabolic chambers fed a fortified corn-soybean. Two distinctly different lagoon systems have been modeled using the pilot bioreactor.

Single-Cell Anaerobic Stabilization Simulation

Two single cell anaerobic lagoons, operated at organic loading rates typical of lagoons found on commercial swine farms have been monitored on an annual basis for a study of sludge accumulation (Hamilton, 2004). These lagoons, labeled OK1 and OK2 were used to set organic loading rates on the pilot scale simulator. EC and pH of the full sized stabilization ponds were used to compare the response of loading rate and hydraulic balance of the pilot-scale bioreactor to full-sized lagoons. Both of the full-sized lagoons used in this study remained red or violet-red in color year round, which indicates the presence of photosynthetically active, purple sulfur and/or purple non-sulfur, anaerobic bacteria. Scum layers of windblown algae also frequently occurred in downwind corners of the lagoons. The bioreactor columns also maintained a red to red-violet color under a thin layer of algae. Algae were removed from the surface every day before feeding.

Table 1. Geometric data and organic loading rates of one pilot-scale bioreactor column and single cell anaerobic stabilization ponds (from Hamilton et al, 2006).

Facility	Dimensions at Maximum Drawdown Level (m)				Side-slope	Volumetric Loading Rate (g VS/m ³ -day)	Surface Loading Rate (kg VS/ha-day)
	Depth	Dia.	Width	Length			
Pilot Bioreactor	3.66	0.305	-	-	-	25.2	935
OK1	2.22	-	36.3	65.1	2.7:1	67.6	1,160
OK2	2.29	-	37.4	61.0	3.5:1	61.0	922

Emission rates of CH₄ and CO₂ from the lagoon simulator were measured using a closed chamber system. The atmosphere immediately above the liquid surface was covered by a modified version of chamber used by Ball et al. (1999) for soil analysis. The chamber used in this study was a 0.2 m tall, 0.09 m diameter polypropylene (Nalgene) cylinder, held in a floating position by a block of 0.15x0.15 m-square Styrofoam. Working headspace of the gas collecting cylinder was 800 ml. Samples were taken from the floating chamber on a frequent basis and analyzed on a gas chromatograph. Daily mass of CH₄ and CO₂ emitted was calculated by multiplying the molar emission rate by the time interval over which it was measured, and summing masses for all the measurement intervals for the day.

Average values of CH₄+CO₂ gas emission rate and average values for molar CH₄:CO₂ ratios are plotted against time of day in Figure 1. CH₄+CO₂ emission was relatively constant at 2 mmoles/m²-min, with a slight increase after feeding, and a slight decrease after the lights were turned off. Mixing produced a spike in gas emission at 7:00. There was a remarkable difference in CH₄:CO₂ ratio between day and night. The ratio of CH₄ to CO₂ dropped from 1.8 to 1.2 almost immediately after the lights turned on, and returned to 1.8 after the lights went off.

Daily carbonaceous gases masses emitted are given in Table 2. Average mass of CH₄+CO₂-C emitted was 2,400 mg/day. The mass of total carbon applied to the column was 3,750 mg/day; therefore, approximately 65% of the applied carbon is accounted for in CH₄+CO₂ emission.

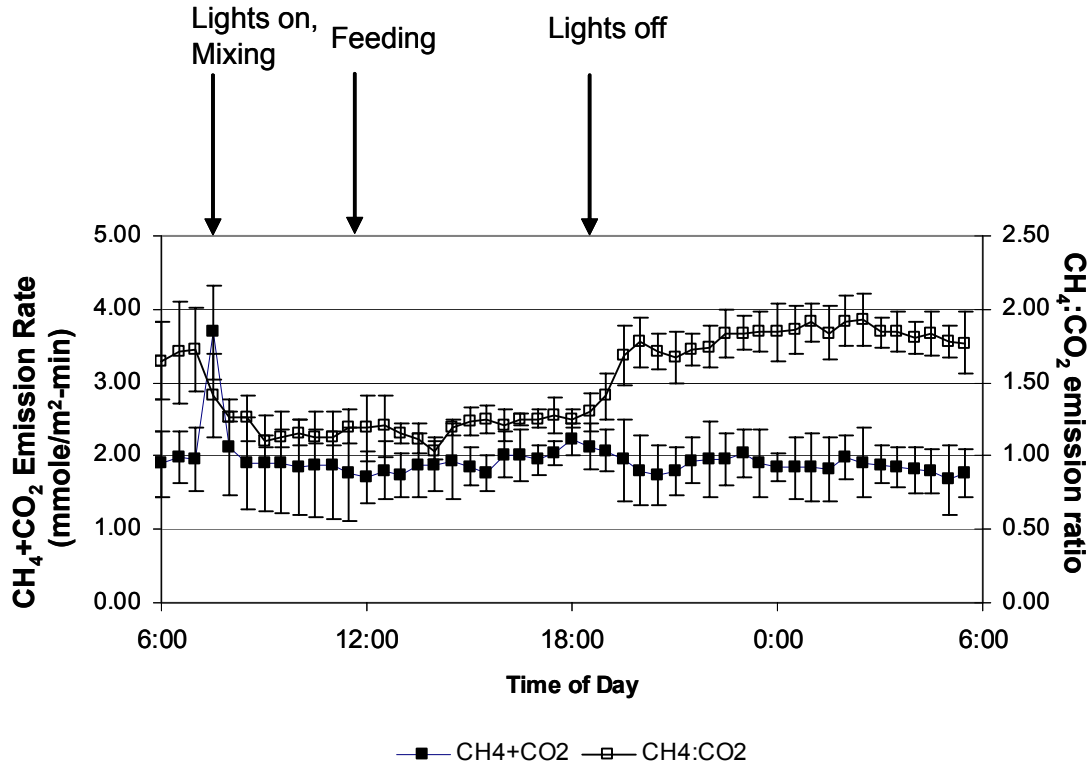


Figure 1. Emissions of carbonaceous gases from pilot reactor simulating single-cell, anaerobic stabilization ponds (from Hamilton, 2006).

Table 2. Masses of Methane, carbon dioxide, and total carbon emitted from pilot reactor simulating single cell anaerobic swine manure stabilization ponds during 24 hour test periods (from Hamilton et al, 2006).

	June 22/24	July 17-18	August 10-11
Moles emitted (mmole/day)			
CH ₄ -C	140	125	91
CO ₂ -C	94	89	61
CH ₄ +CO ₂ -C	234	214	152
Masses emitted (mg/day)			
CH ₄	2,200	2,000	1,450
CO ₂	4,100	3,900	2,700
CH ₄ +CO ₂ -C	2,750	2,550	1,800
Masses emitted from an equivalent lagoon surface (kg/ha-day)			
CH ₄	300	280	200
CO ₂	580	550	380
CH ₄ +CO ₂ -C	390	360	260

Facultative Stabilization Pond Simulation

The stabilization pond located at OSU-SRC differs from lagoons typically found on commercial hog farms in Oklahoma. First, the surface organic matter loading rate of the OSU-SRC lagoon is considerable lower than those found at commercial facilities (Table 3). Secondly, the physical set-up of the OSU-SRC waste stabilization pond makes it function somewhere between a single-cell and a two-cell lagoon. Manure enters the smaller of two cells. This is called the “Sludge Side” because settled manure solids are retained in the cell by the 1.35 m high, submerged dike. Liquids are free to pass into a larger “Clear Side”. There is considerable mixing of liquids between the two cells due to wind action and the water level increase of the sludge side during loading. In effect, the lagoon acts like a two-cell lagoon for manure solids, and a single cell-lagoon for lagoon liquids. The pilot bioreactor was set-up so that liquids are allowed to pass from the outside side column (A or D), which simulates the sludge side, to the inside column (B or C), which simulates the clear side. Only the outside column was fed. Liquids taken 2 m deep from both columns were recycled to the top of the two-column set at 7:00 each day.

EC and pH of the pilot-scale bioreactor was comparable to the full-sized stabilization pond. Both the bioreactor and the full-sized pond remained red or violet-red throughout the experiment. Furthermore, community DNA extracted from the bioreactor columns had a 70% match to community DNA extracted from the analogous positions in OSU-SRC pond (Ball et al, 2006).

Table 3. Loading rates on the OSU-SRC lagoon and a two-column set of the pilot-scale bioreactor.

Facility	Volumetric Organic Loading Rate “Clear Side” only (g VS/m ³ -day)	Surface Organic Loading Rate across Total Surface (kg VS/ha-day)	Surface Nitrogen Loading Rate across Total Surface (kg TKN/ha-day)
Pilot Bioreactor	24	169	26
OSU-SRC Stabilization Pond	77	355	27

Methane, carbon dioxide, and ammonia emissions were measured by placing wind tunnel on top of each column. Air speed across the top of the 0.071 m² column section was 0.2 m/s. NH₃ gas concentrations (ppb by volume) were determined by sampling the wind tunnel flow with a chemiluminescence ammonia analyzer. A gas chromatograph was placed in line with the ammonia analyzer to determine CH₄ and CO₂ concentrations.

The 26 kg TKN/ha loading rate given in Table 3 translates to 360 mg of N applied to the two-column set each day (25 mmoles/day). Emissions of carbonaceous gases and NH₃ from the simulated facultative stabilization pond are given in Table 4. From this table, approximately 72% of applied nitrogen was captured as NH₃ from the facultative pond.

Table 4. Average mass of carbonaceous and ammonia gasses emitted (mmole/day), simulated facultative stabilization pond under late summer conditions (from Carter and Hamilton, 2005).

	CH ₄	CO ₂	Total C	NH ₃
Sludge Side	110	90	200	11.0
Clear Side	50	30	80	7.6
Total	160	120	280	18.6

The ratio of CH₄ to CO₂ did not exhibit as great a difference between night and day as was exhibited in the simulation of the single-cell anaerobic lagoons, and CH₄+CO₂ emissions were higher than would be expected for the lower VS loading rate. These discrepancies may be explained by the constant air flow across the surface of the simulator. The actual situation is likely to be a combination of both results in that air flow across the surface of a stabilization pond will cycle between periods of calm and wind. NH₃ emissions showed a profound difference between night and day, and appear to coincide with the temperature of the liquid surface. As temperature rises, so does emission of NH₃.

Conclusions

Simulation of anaerobic/facultative stabilization ponds under late summer conditions show that 65 % of applied C and 72 % of applied N can be accounted for in the emission of CH₄, CO₂, and NH₃. CH₄ emissions from the simulated stabilization ponds were comparable to those of similarly loaded swine manure treatment lagoons (Hamilton, et al, 2006; Sharpe and Harper, 1999).

The pattern of CH₄ to CO₂ ratio may be attributed to a combination of four processes: (i) aerobic and anaerobic photosynthetic organisms, and aerobic bacteria living symbiotically with aerobic photosynthetic organisms, produce higher volumes of CO₂ during daylight, (ii) the small, but appreciable amount of oxygen produced by algae during daylight suppresses CH₄ production, (iii) higher volumes of CO₂ are produced as complex organic matter is converted to acetate by both aerobic and anaerobic bacteria, and (iv) excess CO₂ and H₂ are converted to CH₄ by methanogenic bacteria once acetate reserves become depleted. NH₃ emission patterns tend to be less complex, and are tied to surface temperature of lagoons.

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Reducing the Risks of Pesticide Exposure via Atmospheric Transport

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Abstract

The fate and transport of agriculturally-significant organic pollutants in the atmosphere is an important national issue. Volatile components and residues bound to dusts may rise into the atmosphere, travel long distances, and be deposited far from the point of origin through various deposition processes, risking contamination of sensitive ecosystems. Examining the environmental fate of agricultural pesticides is complicated by their sporadic and intense application to soils and crops throughout the growing season. Scientists at USDA-ARS have worked with their partners to conduct fundamental research on the atmospheric transport and deposition of semi-volatile organic pesticides at field, watershed, and regional scales. Results analysis of these will assist in identifying the major volatilization pathways from which more effective management practices to mitigate pesticide volatilization can be developed.

Introduction

Research concerning the environmental fate and transport of semi-volatile organic pollutants has focused largely on urban/industrial chemicals such as polychlorinated biphenyls (PCBs) and polyaromatic hydrocarbons (PAHs). In many cases, these chemicals are present at equilibrium in the atmosphere, and their fate may be predicted using models based on partition coefficients. The study of the environmental fate of agricultural pesticides is complicated by their sporadic and intense application to soils and crops throughout the growing season. Agricultural pesticide residues are rarely present at equilibrium concentrations within the environmental compartments of soil, plants, air and water. They are structurally more complicated and have more reactive sites compared with industrial pollutants; thus, they are frequently more polar and will degrade more readily and at variable rates. Long term studies have been conducted on the atmospheric transport and deposition of non-fumigant organic pesticides at field, watershed, and regional scales and a number of new phenomena have been documented.

Methods

Air samples were obtained using a high volume sampler with a glass-fiber filter followed by polyurethane foam plugs. Vapor concentrations were measured periodically for 24 h in the regional studies and continuously for up to 120 h after pesticide application usually in a logarithmic profile above the soil surface in the field studies. Rain samples collected on an event-basis and extracted on-line using SPE cartridges. All samples were analyzed by GC/MS or by LC-MS-MS as were appropriate blanks and spikes. In nearly all studies, weather towers were located near samplers.

Results and Discussion

Several field studies were conducted to examine the processes that affect pesticide volatilization and deposition. While losses from the soil to the atmosphere were greatest within the first 24 hours after application, volatilization continued to be an important loss mechanism. The magnitude and duration of post-application pesticide volatilization was enhanced by high soil moisture and solar radiation (temperature), frequent light precipitation events, and low organic carbon soils (Prueger et al., 2005). These volatilized residues were readily transported to adjacent riparian areas and captured by the plant canopy.

Subsequently, precipitation events washed off the plant canopy and delivered the residues directly to surface waters via the riparian ecosystem streams, bypassing mitigation by the riparian area. These observations are being incorporated into tools for predicting the effectiveness of riparian mitigation function (Angier et al., 2002; 2004).

Studies of regional deposition via precipitation have shown that the Henry's constant of the pesticide will strongly influence its persistence in the atmosphere. Chemicals with low Henry's constants will be readily scavenged, and frequent precipitation events after application will result in increased wet deposition loads. Chemicals with higher Henry's constants will be less efficiently scavenged and loads will correspond to total rainfall during the entire growing season. In a four year study of the Choptank Watershed on the Chesapeake Bay, wet deposition was shown to contribute up to 20% of the annual pesticide loadings to the Bay. In addition, the net gas-exchange flux for some compounds was found to be both positive and negative, i.e., pesticides can be both deposited to and volatilized from the Bay waters depending on the time of year. Particle phase deposition also contributed to the total atmospheric contributions; however, traditional particle phase partitioning, as described by the Junge-Pankow model, cannot explain the higher pesticide concentrations observed on the particles (Goel et al., 2005; Kuang et al., 2003).

In South Florida, the Everglades and Biscayne National Parks are in close proximity to agricultural fields. The effects of high humidity, warm temperatures, low soil organic carbon, frequent pesticide application and crop irrigation, as is the case for South Florida, create a unique situation whereby atmospheric concentrations of pesticides would be expected to be quite high. Indeed, air concentrations in South Florida ranged from 1 to 3 orders of magnitude higher than is typically observed in other places in the country. Pesticide concentrations in rain were also higher. However, pesticide concentrations in canal waters were below levels of concern suggesting that either degradation in the water or a net pesticide flux from the water to the air may be occurring (Hapeman et al., 2003; Harman-Fetcho et al., 2005).

Ecosystems at high altitudes and located downwind from agricultural regions are particularly vulnerable to atmospherically-derived pesticide residues. Large quantities of pesticides are used in the San Joaquin Valley. Warm, moist prevailing winds from the coastal mountains remove volatilized residues from the agricultural fields and deposit them in the much cooler Sierra Nevada Mountains. This long range transport has been implicated as contributing to the amphibian population declines. In addition, the colder temperatures will decrease pesticide degradation rates increasing the pesticide residue exposure times for especially for aquatic species (Fellers et al., 2004; Lenoir et al., 2000; Sparling et al., 2001).

Overall, pesticide residues are present in the atmosphere throughout the year. Deposition occurs continuously via particulate and gas phase deposition. The new knowledge acquired over the course of these investigations will be useful in the development of more effective management practices to mitigate pesticide volatilization. Prevention of volatilization is the most efficient way reduce atmospheric deposition loads and to lessen the impact of agricultural activities on their surrounding ecosystems.

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Assessment of Crop Loss Due To Cumulative Air Pollution Load in South West MP, India

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Abstract

The present research is unique in the sense; as for the first time the cumulative air pollution load in an area was taken into account for the crop loss and not the individual pollutants, that too in field conditions at 5 sites. In an attempt to identify the extent of the distribution of SPM, SO₂, NO_x and O₃ at certain sites in South West M.P. Air monitoring following standard protocol (APHA,1977) was carried out. The areas were classified in two types on the basis of class of pollutant i.e. one where particulate pollutants were higher i.e. Nayagaon-Khor and Nimbaheda and another where gaseous load was prevalent i.e. Dewas, Nagda and Pithampur. Crop samples were also collected from same sites for analysis of biomass, yield and calorific value. The annual SPM loads in the area ranged between 204 - 459 mg/m³ at about 1km and 79 - 154 mg/m³ at 5km. The new industrial areas however had lower load but the cement zone of Nayagaon-Khor and Nimbaheda retains as high as 259 mg/m³ at 5km. As regards SO₂, the annual average was between 5 - 44 mg/m³ with about 36 - 44 mg/m³ in most of the regions at 1km distance except the cement zone (5mg/m³). The levels remain 15 - 20 mg/m³ between 4 - 5 km and up to 5 mg/m³ at 7 - 8 km in prevailing direction of wind. Almost similar conditions existed for NO_x where annual average concentration ranged up to 52 mg/m³ with a minimum level of 17 mg/m³ between 4 - 5 km. The range of O₃ levels remained between 9 - 32 mg/m³ at 1km in prevailing wind direction. But the range of cumulative load of all these pollutants was too high in the host zone of 1 - 2 km around the industrial pockets i.e. 218 - 618 mg/m³ and remained as much as 302 mg/m³ at about 5 km. In prevailing wind directions these levels extend up to 6 - 8 kms or more. Quantitatively and qualitatively crops were affected due to cumulative pollution load and percentage reduction in biomass at all sites ranged between 6 - 25% while in the yield 10 - 35% reduction was observed. The calorific value also indicates the impact of cumulative pollution load. About 5 - 35% decrease was noted. Also regression analysis among all these parameters with cumulative pollution load indicates a direct relationship.



Characterization and Abatement of Air Emissions from Egg Production

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Introduction and Objectives

Air pollutants including ammonia (NH₃), particulate matter (PM), odor, and pathogens, emitted by animal production represent risks to the health and well-being of animals, workers and neighbors, and to the global environment (NRC, 2003). This project, funded by the 2004 USDA-NRI program, will provide information about air emissions from layer facilities, since egg production is known to emit significant amounts of PM, ammonia and odor. Specifically, the objectives of the monitoring are to:

1. Determine whether belt battery (BB) barns emit less air emissions than high rise (HR) barns and establish emission factors for each type of barn.
2. Quantify effects of litter composting and wet scrubbers on air emissions.

Test Methods

Emission measurements will be conducted for 12 months at an egg production facility that has BB barns, HR barns, and chicken manure composting sheds. One on-farm instrument shelter (OFIS) will monitor two BB barns and the other OFIS will monitor two HR barns. A portable ammonia monitoring unit (PAMU) will monitor wet scrubber performance for three months.

A multi-point gas sampling system in each OFIS will continuously cycle gas analyzers through six or seven sampling locations in each barn. Real-time PM monitors will measure PM₁₀ and PM_{2.5}. Weather, barn temperature and humidity, bird and worker activity, and operation of fans, feeders, lights, and manure belts will be recorded along with PM and gas concentrations every 60 s. Pathogen (E. Coli, Salmonella) and odor (European olfactometry) emissions will be measured monthly. Manure sampling and nitrogen (N) balance analysis will be conducted to analyze ammonia N losses.

Differential static pressure across walls with ventilation fans and conveyor openings will be measured continuously. A vibration sensor will be attached to the shroud of each exhaust fan to monitor individual fan operation. Fan airflow rate will be estimated based on barn static pressure, fan operation time, and the relationship between static pressure and fan airflow rate. Emissions will be calculated by multiplying the concentration differences between inlet and exhaust by barn ventilation rate. Continuous data will be acquired at 1 Hz and a 60-s average recorded every minute.

Air at selected continuous or nearly continuous exhaust fans will be sampled for 10 min per measurement cycle and ambient air will be sampled for 30 min/day to measure NH₃ and carbon dioxide (CO₂) concentrations. PM₁₀ and PM_{2.5} concentrations will be measured continuously at one fan exhaust in each of the BB and HR barns (Figs. 1 and 2). Each month, pathogen and odor concentrations will be sampled, and manure N will be analyzed to calculate the N balance.

The characteristics of each barn and compost shed are given in Table 1. Each BB barn has eight tiers (5 upstairs and 3 downstairs) in each of six rows of cages. Each barn is equipped with 24 end-wall fans (divided into four groups of six fans each located in the four corners), and 12 fans in each sidewall. The diameter of these 48 fans are 122 cm. A 122-cm single-speed fan in the NE conveyor belt annex and a 92-cm fan in the SW annex are used to keep the manure belts warm with exhaust barn air (Fig. 1). The gas sampling locations are allocated to the fans associated with ventilation stages 1 and 2, which are distributed along the length of the building.

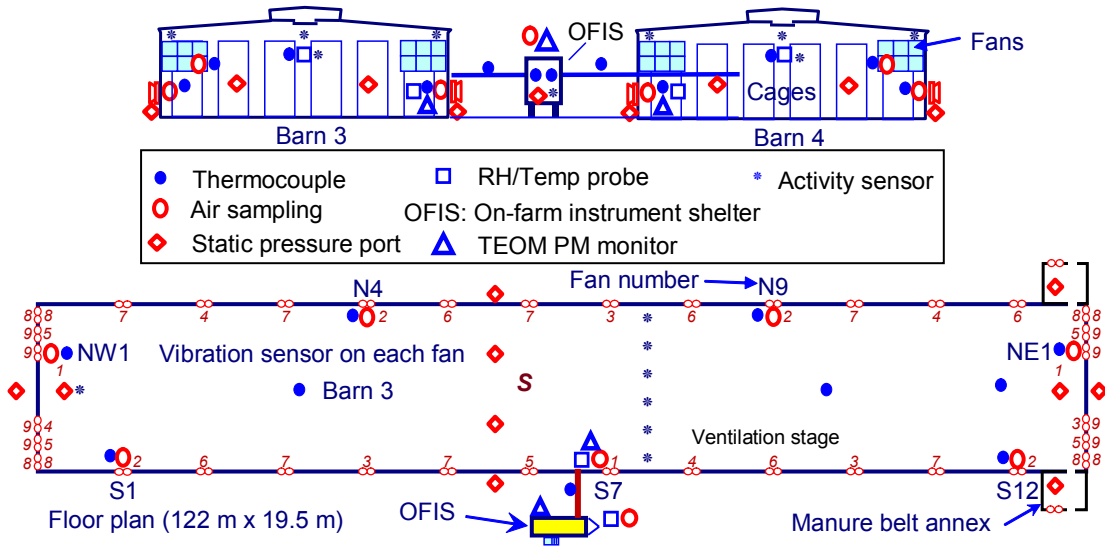


Figure 1. End view and floor plan of BB barns and OFIS. Barn 4 floor plan not shown.

Each HR barn has 8 rows of 8-tier cages. The barns are equipped with ridge baffle air inlets and sixteen, 122-cm diameter exhaust fans in each sidewall of the manure pit (Fig. 2). The fan staging strategy proposed to accommodate the gas sampling locations has ventilation stages 1-3 controlling the three sampled fans in each pit sidewall.

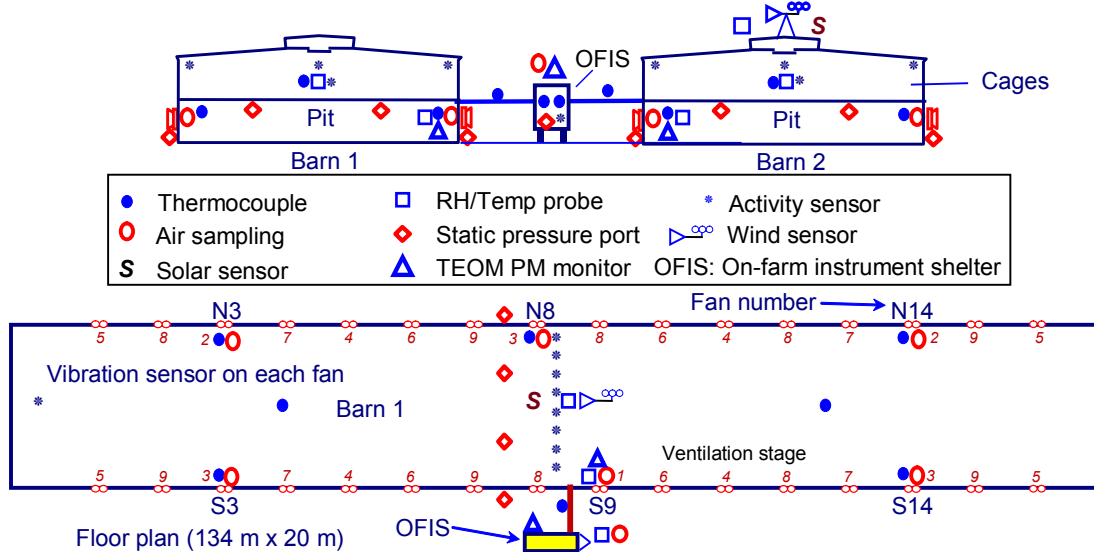


Figure 2. End view and floor plan of HR barns and OFIS. Barn 2 floor plan not shown.

The compost piles in the compost shed are turned every 3 d. There are four single-speed 92-cm diameter ventilation fans in the east endwall. A wet scrubber will be installed at the exhaust of one of these fans to test its ammonia removal efficiency. Ammonia and carbon dioxide concentrations will be sampled at the inlet (fan exhaust) and outlet of the wet scrubber.

Measurement Innovations

Fan airflow measurement is a challenge, especially in large barns with 30 to 100 fans. Based on the concept of using a commercial vibration detector to monitor fan operation (Ni *et al.*, 2005), a new low-cost (about \$28 each) vibration sensor was developed and tested by the Ohio State University. It provides low-voltage on/off fan signals, is weatherproof, and is more reliable than the commercial detector.

Purdue University developed a new multi-point gas sampling system (GSS) that is more reliable and easier to maintain than previous designs. The GSS temperature is higher than ambient temperature to reduce risk of condensation and decrease gas adsorption and desorption at wetted surfaces. Additional sensors in the new GSS allow better monitoring and control.

Data acquisition and control software for barn air emission measurements, AirDAC, written in LabVIEW (National Instrument, Austin, TX), has been significantly improved. AirDAC is now more user-friendly and allows easy modification of configurations such as sampling locations, and records all configuration and parameter changes in a file.

A detailed Quality Assurance Project Plan (QAPP) for this project is being written in conjunction with the QAPP for the National Air Emission Monitoring Study (NAEMS) (Heber 2006), which has many common standard operating procedures.

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Table 1. Characteristics of Barns at Egg Production Site.

Descriptive Parameters	Barns 3-4	Barns 1-2	Shed 2
# buildings at site	4	4	2
Building type	BB	HR	Hoop
Year of construction	1996	1975	1996
Building orientation	E-W	E-W	E-W
Spacing between buildings, m	18.3	12.8	18.0
Width, m	19.5	19.8	18.9
Length, m	122	134	107
Ridge height, m	8.5	7.5	4.6
Sidewall height, m	6.1	6.7	3.6
Barn inventory	180,000	138,000	-
Number of tiers of cages	8	6	-
Manure collection method	Belts	Loader	-
Manure accumulation time, d	4	365	60
External storage	Shed	-	-
Duration of external storage, d	100	-	-
Number of air inlets	12	8	4
Inlet type	Ridge baffle	Ridge baffle	Door
Inlet control basis	Temperature	Temperature	-
Walls with fans (N,S,E,W)	N,S,E,W	N,S	E
Number of exhaust fans	50	32	4
Largest fan diameter, cm	122	122	91
Smallest fan diameter, cm	70	122	91
# ventilation stages	6	8	-
# temperature sensors	16	8	-
Summer cooling	Air	Misters	-



Nitrous Oxide Fluxes from Loamy Sand Soil in Northeast Germany - Seasonal Variations and Influence of Nitrogen Fertilization, Precipitation, and Crop Types

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Abstract

N₂O flux measurements have been carried out at an experimental field since 1999. Different types and levels of fertilization were applied to annual plants (hemp, rape, rye, sorghum, triticale) and perennial plants (grass, poplar, willow). The mean of annually accumulated emission of N₂O-N from all measuring spots was 1.4 kg N₂O-N ha⁻¹ y⁻¹ and the mean annually fertilizer-induced N₂O-N emission from all fertilized sites was 0.6 % for the period from 1999 to 2005. The mean nitrogen conversion factor for perennial crops was lower (0.3 %) than that for annual crops (0.8 %). Several enhanced N₂O emission spots with maxima above 1000 µg N₂O m⁻² h⁻¹, lasting for several weeks, were observed at sites with 150 kg N ha⁻¹ y⁻¹ fertilization in the course of the measurements. These local peak emissions were mainly responsible for the dependence of the nitrogen conversion factor on fertilization level. Annual N₂O emissions are correlated to annual precipitation. Soil nitrate concentrations and N₂O emissions show similar seasonal variations. Usually during the winter season, increased N₂O fluxes due to freeze-thaw cycles were measured for a few days. Additionally, four non-fertilized sites of fallow land were studied in 2002, the year with the highest precipitation of the study period. The comparatively high emission rate of 5.3 kg N₂O-N ha⁻¹ y⁻¹ of this fallow land should be the result of three emission enhancing factors: mineralisation of plant cultivation residues in the soil (hemp in 2001), soil cultivation in 2002 (plowing and harrowing to get a “black fallow”), and higher denitrification rate due to increased water-filled pore space in 2002.

Introduction

Nitrogen fertilizing is one of the main sources of anthropogenic contribution to the global dinitrogen oxide (N₂O) emission as shown by Bouwman (1990), who found a linear relationship between N₂O emissions and fertilizer input. In soil, N₂O is produced predominantly by two microbial processes, the oxidation of ammonium (NH₄⁺) to nitrate (NO₃⁻; called nitrification) and the reduction of NO₃⁻ to gaseous forms NO, N₂O, and N₂ (called denitrification). The rate of N₂O production depends on the availability of mineral N in the soil (e.g. Granli and Bøckman, 1994; Bouwman, 1996). The conversion factor (ratio of N₂O-N emission to input of fertilizer-N) ranges from 0.25 % to 2.25 % depending on soil type (IPCC, 1996 and 2000). When the cultivation of crops is assessed with regard to greenhouse gas abatement, this conversion factor plays a significant role, e.g. the nitrogen fertilizer-induced emission of N₂O-N may counterbalance the carbon dioxide (CO₂) advantage of biofuels (in case of high nitrogen fertilizer application and conversion factor > 2 %), since N₂O as a greenhouse gas contributes to global warming 296 times more effectively than CO₂ (IPCC, 2001).

This aspect, to know more details on this possibility of counterbalancing the CO₂-advantage of biofuels, was the motivation to start a long-term study on N₂O soil emissions, the results of which are presented here. In 1993, the German Federal Environment Office (Umweltbundesamt: UBA) published an ecological balance of rapeseed oil and of rapeseed oil methyl ester (UBA, 1993). This publication led to an intensive discussion about the emission of N₂O and the quantities of nitrogen fertilizer that are really necessary for the cultivation of renewable raw materials. This UBA study reached the controversial result that, with regard to the impact on the climate, rapeseed oil as a diesel substitute does not provide substantial advantages as compared with diesel fuel. The environmental effects of the fertilizer-induced N₂O emissions are an important buttress of the argumentation chain, since a fertilizer-induced N₂O-N emission of 2% to

3% of the annual quantity of N fertilizer applied was assumed to be the probable range. Therefore, several institutions in Germany started studies with the aim of better quantification of the nitrogen conversion factor under climatic conditions of Central Europe.

Later on, in a second UBA study on the assessment of the use of rapeseed oil/RME in comparison with diesel fuel (UBA, 1999), the conversion factors recommended by the IPCC (1996, 2000) were employed. N₂O formation remained as a negative factor in the environmental balance. With regard to the greenhouse effect, the CO₂ balance was now positive, but only additional CO₂ credits due to the comprehensive utilization of rape biomass give biofuel a real advantage over diesel fuel. In addition, nitrous oxide contributes to the depletion of ozone in the stratosphere and thus exerts a negative influence on the environmental balance of biofuels. Since N₂O release is controlled by many factors, significant deviations may occur depending on the location.

Agronomic practices such as tillage and fertilizer applications can significantly affect the production and consumption of N₂O because of alteration in soil physical, chemical, and biochemical activities. Following N-fertilizer applications, an increase in N₂O flux rates has been observed in field and laboratory experiments (e.g. Jackson, 2003; Kaiser et al., 1998; Mulvaney et al., 1997). N₂O emission from croplands at site scales occurs essentially with great spatial and temporal variability (Dobbie and Smith, 2003; Hellebrand et al., 2003; Veldkamp and Keller, 1997). The annual pattern of temporal variation of N₂O emissions is determined in the temperate regions by the seasons and weather conditions, since soil N₂O emissions are regulated by temperature and soil moisture and so are likely to respond to climate changes (Frolking et al., 1998).

In addition to this background variability, agricultural management such as tillage and fertilizing schedule may enhance N₂O emission (Hénault, 1998a and 1998b). Tillage could cause immediate changes in microbial community structure reported by Jackson et al. (2003), thus produce large N₂O emissions at the beginning of crop season. Spatial variability is mainly caused by heterogeneity in soil properties and agricultural management. Additionally, there are different short time emission peaks lasting for hours or days and weeks, the source of which is not explicitly known (Brown et al., 2002; Dobbie and Smith, 2003; Hellebrand et al., 2003; Van der Weerden et al., 1999; Veldkamp and Keller, 1997).

Nitrification and denitrification processes may be stimulated after the application of nitrogen fertilizer (e.g. Dobbie et al., 1999; Firestone and Davidson, 1989; Freney, 1997; Mosier, 1994). The balance between the two processes contributing to the N₂O emission will vary with climate, soil conditions and soil management (Skiba and Smith, 2000). Soil cultivation and precipitation, affecting the soil air exchange rate, should also influence nitrification and denitrification, which are aerobic and anaerobic processes, respectively. Numerous authors studied the emission of N₂O dependent on soil type, fertilization and crop species (e.g. Flessa et al., 1998; Dobbie et al., 1999; Hénault et al., 1998a). There are still uncertainties regarding the soil specific conversion factor, especially the influence of precipitation, soil moisture, temperature, soil nitrate concentration and other variables. Since the N₂O emission factor depends on local conditions, the main aim of this study was to determine this factor and its typical variability for the cultivation of annual and perennial crops on sandy soils under climatic conditions of North-East Germany.

Methods

Experimental Field

The N₂O flux measurements have been performed since 1999 in an experimental field with various crops cultivated for the production of biofuels. The experimental field was established in 1994. A sufficient homogeneity of the soil is reflected by means and standard deviations (sd) of 40 soil samples (soil horizon 0 to 30 cm): clay content 6.2 % (sd: 1.3), organic carbon content.

Table 1. Plants at the columns of the experimental field since 1999.

Each column consists of 4 differently fertilized sites: A, B, C, and D.

Column Period	1	2	3	4	5	6	7	8	9	10
10/1998-03/1999	G*	W*	P1	P2**	P3	F	T	R1	T1*	F*
04/1999-09/1999	G*	W*	P1	P2**	P3	H1	T	R1	T1*	H2*
10/1999-03/2000	G*	W*	P1	P2**	P3	R1	T	H1*	F*	H2
04/2000-09/2000	G*	W*	P1	P2**	P3	R1	T	H1*	Ra*	R1
10/2000-03/2001	G*	W*	P1	P2**	P3	T2	T	R4*	F	G,C
04/2001-09/2001	G*	W*	P1	P2**	P3	T2	H1	R4*	H1	G,C
10/2001-03/2002	G	W*	P1	P2**	P3	R2*	F*	Rc*	H1	G,A,C
04/2002-09/2002	G	W*	P1	P2**	P3	R2*	F*	Rc*	H1	G,A,C
10/2002-03/2003	G	W*	P1	P2**	P3	T2*	Rb*	R2*	H1	G,A,C
04/2003-09/2003	G	W*	P1	P2**	P3	T2*	Rb*	R2*	H1	G,A,C
10/2003-03/2004	G	W*	P1	P2**	P3	Rb*	R3*	T2*	H1	G,A,C
04/2004-09/2004	G	W*	P1	P2**	P3	Rb*	R3*	T2*	Co	S
10/2004-03/2005	G	W*	P1	P2**	P3	F*	T2*	R2*	F	F
04/2005-09/2005	G1	W*	P1	P2**	P3	H1*	T2*	R2*	S*	SF*
10/2005-03/2006	G1	W*	P1	P2**	R5*	R6*	R7*	R2*	F	F

*: Columns with one measuring spot per site A, B, C, and D.

**: Columns with two measuring spots per site A, B, C, and D.

A: Alfalfa (*Medicago sativa*)C: Clover (*Trifolium repens*)Co: Corn (*Zea mays*)

F: Fallow land

G: Grass (orchard grass: *Dactylis glomerata* L.)G1: Sudan grass (*Sorghum sudanense*)H1: Hemp (*Cannabis sativa* L.) var. Felina 34H2: Hemp (*Cannabis sativa* L.) var. Fedrina 74P1: Poplar (*Populus maximowiczii* x *P. nigra*) var. Japan 105 with grassP2: Poplar (*Populus maximowiczii* x *P. nigra*) var. Japan 105P3: Poplar (*Populus maximowiczii* x *P. trichocarpa*) var. NE 42 with grassRa: Rape (*Brassica napus* L. spp. oleifera Metzg.) var. LicosmosRb: Rape (*Brassica napus* L. spp. oleifera Metzg.) var. ExpressRc: Rape (*Brassica napus* L. spp. oleifera Metzg.) var. ArtusR1: Rye (*Secale cereale* L.) var. AmiloR2: Rye (*Secale cereale* L.) var. MatadorR3: Rye (*Secale cereale* L.) var. AvantiR4: Rye (*Secale cereale* L.) var. HacadaR5: Perennial Rye (*Lolium parvifolium* L.) var. WaldiR6: Perennial rye (*Secale montanum* L.) var. PermontaR7: Rye (*Secale cereale* L.) var. RPM 1S: Sorghum (pearl millet: *Pennisetum glaucum* (L.) R. Br.) var. Rona 1SF: Sunflowers (*Helianthus annuus* L.) var. KW0411T: Topinambur (Jerusalem artichoke: *Helianthus tuberosus* L.)T1: Triticale (X *Triticosecale* Wittm.) var. AlamoT2: Triticale (X *Triticosecale* Wittm.) var. ModusW: Willow (*Salix viminalis*) with grass

0.9 % (sd: 0.14) and pH value 6.0 (sd: 0.34). Soil texture of the topsoil was classified as loamy sand (Hellebrand and Scholz, 2000). The weather means at the Potsdam Weather Service station (about 10 km away) between 1951 and 1980 were 8.6 °C for air temperature and 595 mm for precipitation. In the period of this study (1999-2005), the mean temperature increased (mean of the last seven years: 9.7 °C) and the mean annual precipitation decreased (mean of the last seven years: 573 mm; DWD, 2006).

The field was subdivided into 40 sites (624 m² each). Different plant varieties (Tab. 1) or plant combinations were arranged as columns (four sites each, labeled as A, B, C, and D) with a distance of 6 m

between each column. Grass (column 1) was mowed two to three times every year. The short rotation wood (or “field wood”: poplar and willow; column 2 to 5) was periodically harvested (every two to four years). The crops in columns 6 to 10 were mostly annual plants, which were rotated or planted according to actual needs. The different types and levels of fertilization were applied in four rows, perpendicular to the columns. There were sites with different levels of nitrogen input (fertilizer: calcium ammonium nitrate; A: $150 \text{ kg N ha}^{-1} \text{ y}^{-1}$; B and C: $75 \text{ kg N ha}^{-1} \text{ y}^{-1}$) supplemented by PK-fertilizer (A), wood ashes (B), and straw ashes (C) and sites without fertilization (D).

Gas flux Measurements

In 1997 and 1998, FT-IR gas analysis was applied (Hellebrand and Scholz, 2000). FT-IR gas analysis gave variation coefficients of 7% for N_2O and CH_4 flux measurements at atmospheric mixing ratios and of up to 20% for NH_3 concentration determination below 1 ppm. Therefore, the accuracy was insufficient to get precise nitrogen conversion factors. Nevertheless, it could be shown that no NH_3 was emitted, although calcium ammonium nitrate was applied as fertilizer.

Since 1999, gas flux measurements have been performed four times a week by means of closed chamber technique and an automated gas chromatograph (GC). The sealing rings (Y profile, sealing by water level) for the gas flux chambers (cover boxes) were embedded in the soil. One ring was put on each measuring spot at the sites A to D of the columns with different crops. The gas flux chambers had a volume (V) to area (A) ratio of $V/A = 0.315 \text{ m}$ (volume 0.064 m^3 , inner diameter 0.509 m). Fluxes were usually measured in the morning. Two evacuated gas samplers (100 cm^3 bottles with Teflon sealing and vacuum taps) were connected to each box. The first was filled when the box was put on the water-sealed ring on the soil and the second one after about 60 minutes enclosure time. The samplers were then connected with the automated GC-injection control system. The GC was fitted with an electron capture detector (ECD). The operating temperature for the ECD was about $300 \text{ }^\circ\text{C}$ (sometimes manual shifted up to $\pm 5 \text{ K}$ to get identical peak sizes for the maximum concentrations of CO_2 and N_2O of the calibration gases mixture) and the column temperature was $65 \text{ }^\circ\text{C}$. Both the pre-column (length 1 m) and the main column (length 3 m) were packed with Porapak Q (80/100 mesh). In one computer-controlled run up to 64 samples could be analyzed. The N_2O detection limit was 5 ppb (5×10^{-9}). At atmospheric mixing ratio, the coefficient of variation was 1.2% for N_2O measurements. For each level of fertilization, the N_2O emission factor was calculated by taking the difference between the mean values of the fertilized sites and the non-fertilized sites.

CO_2 and N_2O , both of them generated in the soil, have nearly equal diffusion constants. The easily measurable CO_2 served for the evaluation of linearity and mixing homogeneity of the measuring chamber (measurements at several heights in the closed chamber). Studies on the concentration increase of CO_2 in a flux chamber on bare soil and on grass sites demonstrated that the increase in concentration was linear (R^2 between 0.9973 and 0.9986) during measurement periods of 60 minutes (Hellebrand and Scholz, 2000).

Soil Ion Measurements

Since 2003 soil samples were drawn from sites with the three fertilizer levels (A: $150 \text{ kg N ha}^{-1} \text{ y}^{-1}$; B: $75 \text{ kg N ha}^{-1} \text{ y}^{-1}$ and D: no fertilizer) and different crops (columns 2, 4, 6, and 7; Tab. 1). The samples were taken from the 0-30 cm soil depth close to the corresponding measuring rings. The water content of the soil samples was obtained by gravimetric determination of weight loss when soil samples were dried at $105 \text{ }^\circ\text{C}$ for 24 h. The concentration of mineral nitrogen (ammonia nitrogen $\text{NH}_4^+\text{-N}$ and nitrate nitrogen $\text{NO}_3^-\text{-N}$) and other ions was determined by ion chromatography after extracting by deionized water and filtering. In the present paper, only the course of concentration of soil nitrate is considered. The detailed evaluation of all the soil ion concentrations will be published elsewhere.

Results and Discussion

Fertilizer-Induced N₂O Emissions

The emission of N₂O followed the expected pattern of fertilizer-induced emissions (FEE in Fig. 1 and Fig. 2, fertilization date in Tab. 2). These enhanced emissions were detectable at all fertilized sites after fertilizing and lasted from four to eight weeks. We also found temporarily and spatially limited high fluctuations throughout the entire study since 1999. N₂O emission peaks over 1000 $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ were observed from few measuring spots (Fig. 2). These findings are in accordance with other studies (e.g. Augustin et al., 1998; Röver et al., 1998). Because measurements were taken four times a week, the "regularities" (induced emissions) and fluctuations could be studied with sufficient temporal resolution at different crop sites on a relatively homogeneous sandy soil. Except during the few freeze-thaw cycles (FTC in Fig. 1 and Fig. 2), the N₂O emission rate usually dropped to less than 30 $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ between October and March.

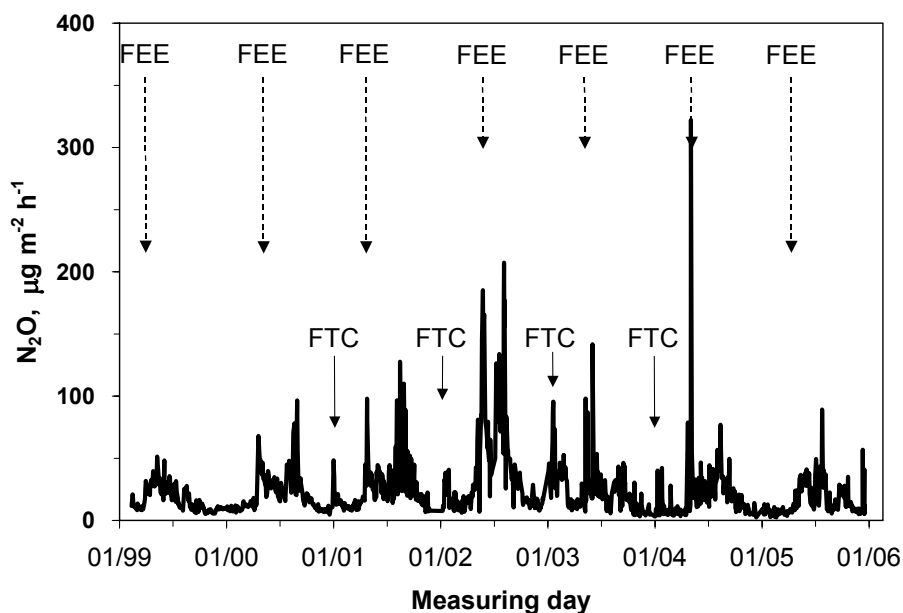


Figure 1. Time series of N₂O emissions from all sites of the experimental field.

Daily mean values of emission rates from all measuring spots,
 FEE: Fertilizer enhanced emissions, fertilisation usually in April, see Table 2,
 FTC: Freeze thaw cycle enhanced N₂O emissions, usually in January.

Table 2. Date of fertilizer application and fertilization rates.

Year	1999	2000	2001	2002	2003	2004	2005
Fertilization							
1*	March 26	April 14	April 17	April 9	April 8	April 21	April 18
2**	May 7	May 5	May 7	May 13	May 14	May 6	May 10
3***	May 18	May 25	May 14	May 21	June 2	May 18	May 19

1* : A: 50 kg N ha⁻¹ y⁻¹; B: 50 kg N ha⁻¹ y⁻¹; C: 50 kg N ha⁻¹ y⁻¹; D: -

2** : A: 50 kg N ha⁻¹ y⁻¹; B: 25 kg N ha⁻¹ y⁻¹; C: 25 kg N ha⁻¹ y⁻¹; D: -

3*** : A: 50 kg N ha⁻¹ y⁻¹; B: -; C: -; D: -

Other Enhanced N₂O Emissions

No clearly frost-induced emission rates were found in January 2000 and 2005. Frost-induced N₂O-emissions are related to the course of temperature and could be dependent on several factors such as groundwater level, water-filled pore space, soil pH, soil nitrate content, soil texture, and soil structure (Flessa et al., 1998; Röver et al., 1998; Mogge et al., 1996). However, the frost-induced emissions we measured at a couple of the sites are small compared to the total annual N₂O emission budget from the sandy soil of the experimental field.

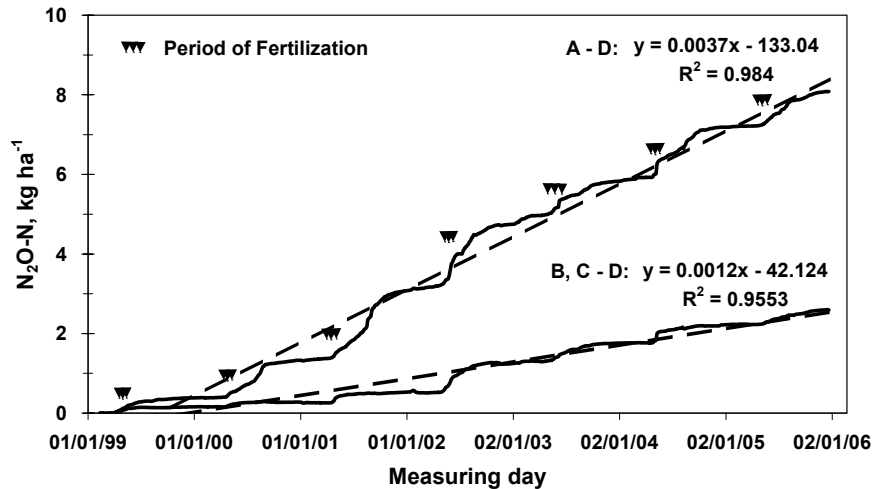


Figure 3. Accumulated mean emission differences.

The regression line of the mean emission differences between A-sites (150 kg N ha⁻¹ y⁻¹) and D-sites (non-fertilized) has a threefold slope compared to the mean emission differences between B/C-sites (75 kg N ha⁻¹ y⁻¹) and D-sites.

There were several unusual high emission periods at fertilized sites within the course of the measurements since 1999. In 2000, emission rates between 400 and 800 $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ were measured at the rape site with fertilization level of 150 kg N ha⁻¹ y⁻¹ from the end of July till the harvest in the beginning of September. In 2001, very high N₂O emission rates of up to 1370 $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ were observed from one poplar measuring spot with fertilization level of 150 kg N ha⁻¹ y⁻¹ over a period from July till October. There were high emission rates between 350 and 900 $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ from all fertilized rape sites for several weeks in July 2002 and in July 2004. Since most of these enhanced emissions were observed at A-sites (150 kg N ha⁻¹ y⁻¹), the conversion factor increases with the fertilization rate (Tab. 3 and slope of regression lines in Fig. 3).

Additionally, four non-fertilized sites of fallow land were measured in 2002. High emission rates between 200 and up to 1480 $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ were observed from the sites of the fallow land during the period from end of May 2002 till middle of August 2002. This fallow land, which was cultivated and fertilized the years before, even had the highest annual emission rate of 5.3 kg N₂O-N ha⁻¹ y⁻¹ although no fertilizer was applied during the year of study. As the fallow land was only ploughed and harrowed in March but no drilled (“black fallow”), the high precipitation in 2002 could have caused enlarged anaerobic zones (increased water-filled pore space) within the topsoil resulting in enhanced N₂O emissions.

Table 3. Annual precipitation, mean N₂O-N emissions and mean nitrogen conversion factor at differently fertilized rows A, B, C, and D.

Year	Annual Precipitation, mm	N ₂ O-N Emission, kg N ₂ O-N ha ⁻¹ y ⁻¹				N ₂ O-N Conversion factor, %		
		Mean of all sites A	Mean of all sites B, C	Mean of all sites A, B, C	Mean of all sites D	Mean of all sites A	Mean of all sites B, C	Mean of all sites A, B, C
1999	406.1	1.11	0.84	0.88	0.71	0.26	0.20	0.22
2000	537.8	1.92	0.88	0.98	0.80	0.75	0.17	0.37
2001	626.8	2.47	0.86	1.09	0.74	1.15	0.32	0.59
2002	762.5	2.80	1.34	2.15	1.10	1.13	0.86	0.95
2003	428.3	1.92	1.23	1.40	0.84	0.72	0.64	0.66
2004	630.2	1.78	0.86	1.15	0.48	0.86	0.70	0.75
2005	617.1	1.61	0.95	0.85	0.54	0.71	0.48	0.56
1999-2005	572.7	1.94	1.13	1.41	0.86	0.72	0.55	0.60

Table 4. Mean N₂O-N emissions and conversion factors from sites with perennial crops and annual crops (differently fertilized rows A, B, C, D).

Crop	N ₂ O-N Emission, kg N ₂ O-N ha ⁻¹ y ⁻¹				N ₂ O-N Conversion factor, %		
	A	B	C	D	Mean sites A	Mean sites B, C	Mean sites A, B, C
Grass (3 years)	1.30	0.97	1.23	1.04	0.17	0.08	0.11
Willow (6 years)	1.04	0.75	0.97	0.57	0.31	0.39	0.36
Poplar (7 years)	1.72	0.68	0.89	0.50	0.82	0.38	0.52
Mean Perennial crops	1.35	0.80	1.03	0.70	0.43	0.28	0.33
Rye (6 years)	2.68	1.38	1.67	0.93	1.16	0.79	0.91
Triticale (4 years)	2.09	1.70	1.44	0.74	0.90	1.11	1.04
Hemp (2 years)	1.29	0.78	0.99	0.67	0.41	0.29	0.33
Rape (4 years)	3.82	1.66	2.02	1.40	1.61	0.59	0.93
Mean Annual crops	2.48	1.38	1.53	0.94	1.02	0.70	0.80

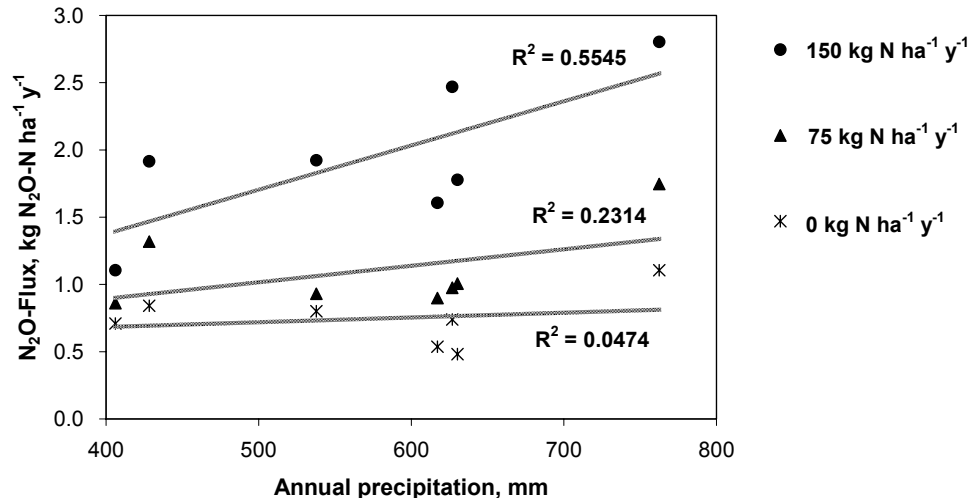


Figure 4. Correlation between mean annual total N₂O emissions from differently fertilized sites and annual precipitation.

The mean annual N₂O emissions and the mean nitrogen conversion factor of the differently fertilized rows A, B, and C had maxima in the period 2001/2002 (Tab. 3). There is a clear correlation between annual precipitation and annual total N₂O emissions. This correlation increases with the level of fertilization (Fig. 4). An obvious difference exists between N₂O emission rates from sites with perennial crops and annual crops (Tab. 4). The emissions from sites with annual crops were about 50 % higher than from sites with perennial crops. This is true, independent on fertilization level. Considering the conversion factor, it was about twice for sites with annual crops compared to perennial crops (Table 4). Although this phenomenon is not fully understood, changes in physical structure by soil tillage may alter biological activity and thus N₂O emission over the crop season (Jackson, 2003; Kaiser et al., 1998; Mulvaney et al., 1997).

Soil Nitrate and N₂O Emissions

The correlation between soil nitrate and N₂O emissions was much lower than initially expected. The close relationship between N₂O generation and extractable soil nitrate (denitrification) and ammonia (nitrification) is well understood (e.g. Bremner, 1997). The seasonal change of soil nitrate concentration and N₂O fluxes shows similarities (Fig. 5), but due to the temporal and local fluctuations of N₂O emissions and of nitrate concentrations, the correlation might depend on the locations and schedule of soil sampling compared to the N₂O flux measurements. The soil samples were taken outside the measuring rings (in order not to disturb the soil surface) in distances of 30 to 50 cm, neither synchronous nor daily but only weekly. There is nearly no correlation between daily flux measurements and weekly nitrate concentration measurements ($R^2 = 0.03$), whereas a slight correlation exists for the monthly means ($R^2 = 0.20$). This is interpreted as a result of the high dynamics of N₂O flux, which can considerably vary at the same measuring ring in the course of one week. On the other hand, the monthly means reflect a general tendency. Therefore, the correlation increases, as both quantities show similar seasonal changes (Fig. 5).

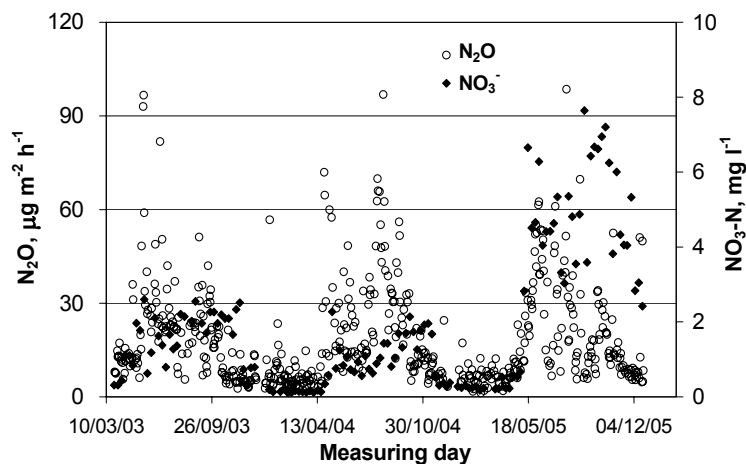


Figure 5. Seasonal change of mean soil nitrate concentration and mean N₂O fluxes

N₂O Emission and CO₂ Mitigation of Energy Crops

The mean N₂O-N emission factor was approximately 0.6 % for all A-, B-, and C-sites (0.7 % for A-sites and 0.5 % for B- and C-sites) for the year period from 1999 to 2005 (Tab. 3). Additionally, the emission factor shows variation depending on crop type (annual or perennial) and year (precipitation). Due to the local enhanced N₂O emissions from several measuring spots at A-sites, the mean of the emission factor increased with the nitrogen fertilization level (annual means of induced emissions; sites A to B, C; Tab. 3) and annual crops emitted more N₂O than perennial crops (Tab. 4). Reason could be variations in biological activity in dependence on type and level of soil fertilization. This is being examined at present. The results measured here are at the lower part of the range of the N₂O emission factor, which is recommended by IPCC (1996, 2000) for the fertilization-based N₂O inventories. Thus, it can be stated that the emission of N₂O is comparatively low on the sandy soils of our experimental field. The CO₂ advantage of energy crops will not be reduced by nitrogen fertilizing as long as fertilizing results in an adequately higher biomass yield (Scholz and Ellebrock, 2002). This result is also true for other crops, cultivated on sandy soils as source for renewable vegetable raw materials, if excessive fertilizing is avoided.

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Carbon Monoxide from Composting due to Thermal Oxidation of Biomass: An Additional Pathway for CO in Agricultural and Forest Ecosystems

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Abstract

Carbon monoxide (CO) influences tropospheric chemistry and, in particular, tropospheric ozone (O₃) concentration. The major sources of CO are incomplete combustion and photochemical oxidation of atmospheric hydrocarbons. Other sources include emissions from green plants, dry soils, and degrading plant matter. Studying trace gas releases from composting processes with a high-resolution FT-IR spectrometer, we observed emissions of carbon dioxide (CO₂) and CO from heaps of fallen leaves, during rotting of green waste, and windrow composting of livestock waste, and on a laboratory scale, during rotting experiments from non-sterilized as well as from sterilized dead plant matter in a temperature and ventilation controlled substrate container. For composting of green waste, emission rates were calculated from ratios of the concentration differences between compost air and background air and by evaluation of the carbon balance of the heaps. Emissions from windrows of farm waste were measured using gas flux chambers. In laboratory experiments, the plant material was placed in a closed substrate container. In case of experiments with sterilization, the closed container was exposed to 136°C for three hours. After cooling down, the container was ventilated via 0.2 µm filters for concentration analysis. The concentration of CO was up to 120 ppm in compost piles of green waste, up to 10 ppm during composting of livestock waste, and up to 160 ppm during laboratory experiments. The flux ratio of CO-C to CO₂-C varied between 10⁻⁴ and 10⁻³ during composting and was up to 10⁻² in experiments with sterilized plant matter. The mean CO flux rates were ~20 mg CO m⁻² h⁻¹ for compost heaps of green waste, and 30 to 100 mg CO m⁻² h⁻¹ for fresh dung-windrows. Arrhenius-type plot analyses resulted in activation energies of 52 kJ mol⁻¹ for thermochemically produced CO from sterilized compost substrate and 65 kJ mol⁻¹ for CO from unsterilized compost substrate, i.e. a temperature raise from 5°C to 20°C triplicates the thermal CO production. Though globally and regionally not an important source, thermochemically produced CO emissions could affect local air quality near large composting facilities.

Introduction

The background for CO studies at the Potsdam Institute of Agricultural Engineering

The reunification of Germany in 1990 led to an evaluation of all scientific institutions of the former GDR by the German Science Council with recommendations for future research directions (Wissenschaftsrat, 1991). "Substance cycling and energy fluxes in agriculture" was a new research direction for the Institute of Agricultural Engineering Potsdam-Bornim. With it, the new task for agricultural engineering was now the design of technologies and procedures aiming at greenhouse gas abatement. At the same time (1993), a new federal administrative regulation as part of the German "Waste Avoidance and Management Act", called "Technical Instruction on Municipal Waste", was passed (BMU, 1993). Composting became an important way for municipal waste processing by this instruction. Composting has been accepted by the public as an environment friendly way of organic waste processing, but in 1993 there were few figures on the emissions of ammonia (NH₃), nitrous oxide (N₂O), and methane (CH₄) during composting available. Therefore, it was decided to launch research on emissions during composting on a laboratory scale, measurements at pilot plants, and analysis of leaching and emissions from compost heaps in the frame of landscape management and conservation. To analyze the concentrations of NH₃, N₂O, CH₄, and other gases

in compost-air, a high resolution FT-IR measuring system was chosen for laboratory studies (Hellebrand and Kleinke, 1996; 1997). Additionally, this system could be used for gas analysis from field measurements by gas sampling with commercial gas bags (Linde, 2006).

During initial evaluation of the recorded FT-IR spectra from compost-air, we observed previously unidentified absorption lines near the intensive CO₂-absorption band (2060-2100 cm⁻¹, Figures 1 and 2). The detailed analysis showed that these lines reflected CO in the mid to high ppm range, which was not expected for green waste compost-air. The compost heaps were located in a clean-air area, far away from traffic lines. The findings of our first evaluation were included in a publication on composting (Hellebrand, 1998). A more detailed search on the sources of CO during composting started, the results of which are presented in this contribution.

CO on a Global Scale

CO is known to the public as an air pollutant and toxic gas. The sources for the atmospheric load results mainly from incomplete combustion of fossil fuels and biomass (1350 Tg; IPCC, 2001a) and photochemical oxidation of atmospheric hydrocarbons (1230 Tg; IPCC, 2001a). Vegetation (150 Tg; IPCC, 2001a) and Oceans (50 Tg; IPCC, 2001a) contribute on a smaller scale. CO is not a greenhouse gas, because its contribution to the infrared absorption in the 10 μm infrared window of the atmosphere is too low. However, emissions of CO have indirect effects on climate through enhanced levels of tropospheric O₃ and CH₄ as a result of its reaction with hydroxyl-radicals HO• (Crutzen, 1974; Liu et al., 1987; Fuglestvedt et al., 1996; Daniel and Solomon, 1998). Therefore, the Working Group I of the Intergovernmental Panel on Climate Change included CO in its reports.

The mean atmospheric CO abundance is approximately 80 ppb, with an atmospheric lifetime of one to three months (IPCC, 2001b). The Northern Hemisphere contains almost twice as much CO as the Southern Hemisphere, mostly due to anthropogenic emissions. Atmospheric CO trends are sensitive to the time period considered, e.g. for the period 1991 to 1999, the CO trend was -0.6 ppb y⁻¹, and for 1996 to 1998, the trend was positive by +6 ppb y⁻¹ (IPCC, 2001b). The most important sink for CO is the reaction with HO• (Logan et al., 1981), with a smaller sink via soil uptake (Table 2.6 in IPCC, 1995). The latter results from soil microbial processes (Conrad and Seiler, 1980; Conrad et al. 1981; Conrad and Seiler, 1985a; Scharffe et al., 1990; Conrad, 1996), whereas simultaneously occurring CO production in soils has been assigned to thermochemical oxidation of organic matter (Conrad and Seiler, 1985a; 1985b; Seiler and Conrad, 1987). Although the global budget of CO is fairly well understood, there are some uncertainties associated with the CO fluxes from natural sources.

Direct and Indirect Biogenic Sources of CO

CO from biogenic sources has different pathways – direct CO-release from certain plants (Bauer et al., 1979, and reference therein), production or consumption by specialized microorganisms (Radler et al., 1974; Conrad and Seiler, 1980; 1985a; Conrad, 1988; 1996; Scharffe et al., 1990; King, 1999), generation by photochemical and thermochemical oxidation of biomass (McConnell et al., 1971; Fischer and Lüttge, 1978; Lüttge and Fischer, 1980; Conrad and Seiler, 1985b; Tarr et al., 1995; Kanakidou and Crutzen, 1999; Schade et al., 1999), and by oxidation of CH₄ and of non methane hydrocarbons (NMHC) of biogenic origin, such as isoprene (McConnell, 1971; Crutzen, 1974; Novelli et al., 1992; Miyoshi, et al. 1994).

The production of CO from plant litter (Sanhueza et al., 1994; Zepp et al., 1996 and 1997; Hellebrand, 1998; Sanhueza et al., 1998) in the dark is most likely caused by thermochemical oxidation processes, and has previously been studied in more detail (Schade, 1997; Schade et al. 1999). Thermochemically (or thermally induced) CO production from grass and leaf litter obeys the Arrhenius law with activation energies mostly between 60 and 90 kJ mol⁻¹, with strong evidence that CO is not produced biologically (Schade, 1997; Schade et al. 1999). The authors estimated global CO emissions due to thermochemical oxidation of organic carbon from plant litter of 40 Tg CO (Schade and Crutzen, 1999). Uncertainties are related to the fact that thermal CO production does not only depend on temperature, but also on type of plant litter and moisture content. Increasing moisture content generally promotes CO emission.

Methods

FT-IR Measuring System

Trace gas analysis was carried out with a high resolution FT-IR spectrometer (Spectrum™ 2000, PERKIN ELMER). The system was equipped with two heated, long path multi pass gas cells (FOXBORO® - Invensys Process Systems). The temperature of the cells was kept at 80 °C to avoid condensation at the gold coated mirrors of the cells. The gas cell with optical variable path length of up to 20 m had a measuring volume of 5.4 dm³ and was used for quantification when samples with at least this volume were available. The second gas cell with a fixed optical path length of 7.2 m (48 passes with a base of 0.15 m) had a volume of only 0.5 dm³ and was used in experiments with limited sample size or to achieve sufficient time resolution for continuous flow measurements with changing gas composition.

The FT-IR spectrometer used here operates as a so-called one-beam system. To get a pure sample spectrum, at first the background spectrum (empty gas cell) must be measured. Then a sample spectrum is obtained by a second measurement with gas cell filled. To minimize stochastic and systematic errors the FT-IR spectrometer with its beam channels and detector housings was purged with dry, CO₂ free air (BALSTON® FT-IR Purge Gas Generator; -73°C dew point of cleaned air). Separate MCT-detectors for each gas cell were used to optimize sensitivity. Generally, spectra were recorded with 64 scans.

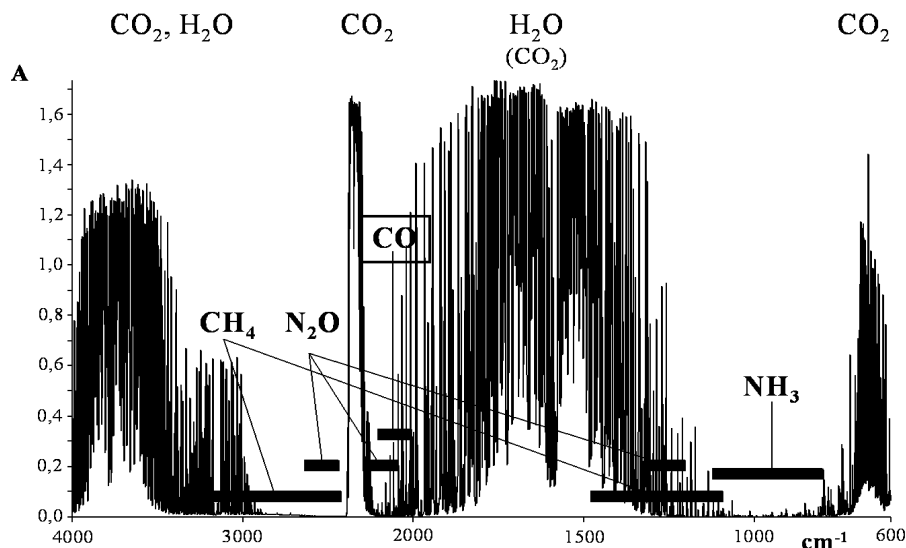


Figure 1. FT-IR spectrum of ambient air between 4000 cm⁻¹ and 600 cm⁻¹ (absorbance to wave number; measured with 20 m long path gas cell). The IR wave bands of environmentally relevant gases are indicated.

A constant range (4000 cm^{-1} to 600 cm^{-1}) and fixed point intervals (0.05 cm^{-1}) were applied for calibration and for all measurements with quantitative evaluation. The detection limit was better than 10 ppb for CO in nitrogen (20 m gas cell; 0.2 ppm for 7 m gas cell). Because of IR-line overlapping in case of compost air with concentrations of CO_2 above 5 Vol.-%, N_2O above 300 ppm, and saturated water vapor pressure (at ambient temperature) the detection limit for CO deteriorated by a factor of 5 to 10 dependent of compost air composition (Figures 2 and 3). The systematic error in the determination of CO concentrations due to the above interferences was estimated to be less than 20 % for CO concentrations below 1 ppm (20 m cell) and $\sim 10\%$ for CO above 10 ppm. The error depends on the type of gas and the concentration span to be evaluated (line positions and changes in line intensities), on calibration and spectrum evaluation, and on measurement procedure. Errors of the FT-IR-measurements of gas samples, caused by electronic noise, gas handling, and other stochastic sources were estimated to be in the range of 5 %. Furthermore, the additional error due to gas sampling by flux chambers reduces the accuracy in the determination of emission rates.

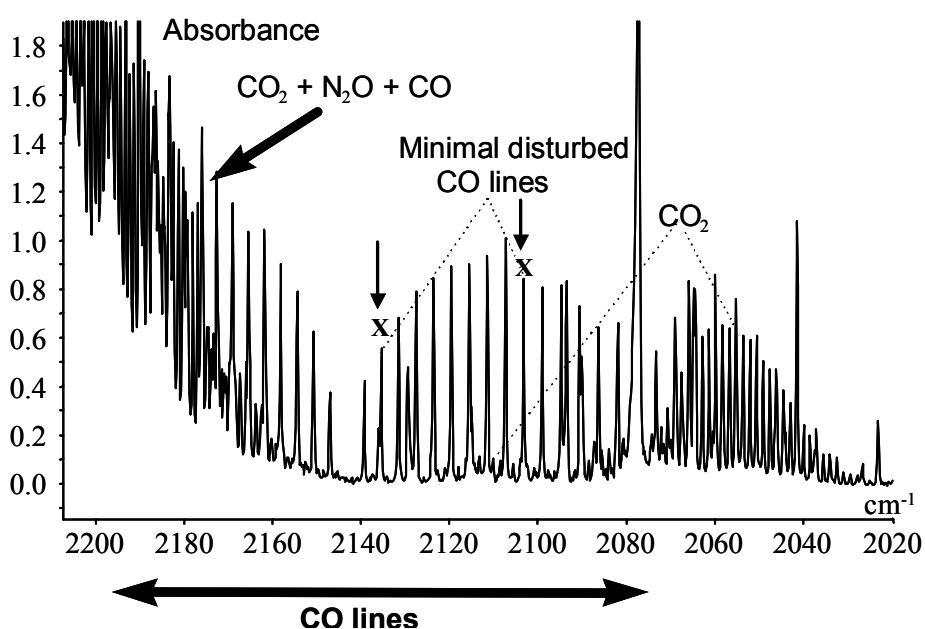


Figure 2. Minimal disturbed FT-IR lines (marked by X in figure) of air from substrate container during laboratory scale composting of hay (mixed herbage) from landscape conservation, 70 % moisture content (w.b.) and amended by calcium ammonium nitrate. Measurement in absorbance mode, resolution 0.2 cm^{-1} , temperature $35\text{ }^\circ\text{C}$, aeration rate $50\text{ cm}^3\text{ min}^{-1}$. CO: 173 ppm; CO_2 : 19.1 Vol.-%; CH_4 : 1.7 ppm; N_2O : 281 ppm; NH_3 : 0.9 ppm.

FT-IR Calibration and Concentration Measurements

Spectrum Quant+ software (PERKIN ELMER) was applied for concentration evaluation of the FT-IR spectra. The software operates with chemometric procedures (PLS: partial least-squares) based upon inputs from calibration spectra. This multi regression method was developed by Wold (1966). A cross-validation was used. For base line corrections, first derivatives were chosen. The spectra had to contain the range of gas concentrations of the gas (or gases) to be evaluated and, if possible, different concentration levels of disturbing gases like water vapor or others. Secondly, during the development of the calibration method, the most suitable wave number ranges for the calibration procedure had to be put in, or in other words, non-interesting and disturbing spectrum regions were to be "blanked". In the calibration methods developed here, 15 to 30 standards (calibration spectra) were measured with a resolution of 0.2 cm^{-1} and a point interval of 0.05 cm^{-1} .

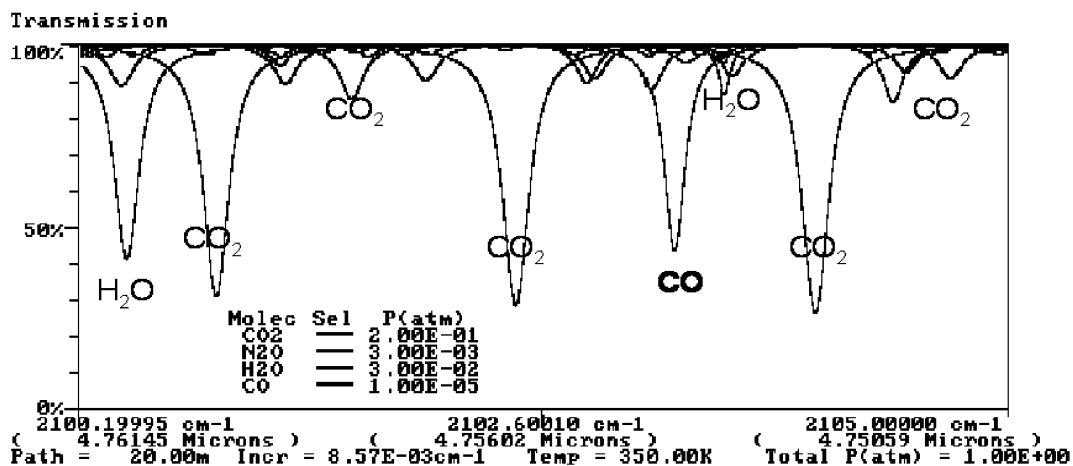


Figure 3. HITRAN simulation of spectral lines between 2100.2 cm⁻¹ and 2105.0 cm⁻¹. The CO line at 2103.2 cm⁻¹ is nearly undisturbed (CO: 10 ppm; CO: 20 Vol.-%; N₂O: 3000 ppm; H₂O: 22 g m⁻³ – 100% humidity at 24°C).

For multi-gas analysis, the concentration distribution with the maximum of mutual linear independency was calculated. Then, specific instrument calibration mixtures (Linde HiQ® Specialty Gas) were applied or, in most of the cases, the gas mixtures were produced by using volume calibrated 10 dm³ plastic bags (Linde Plastigas®; Linde, 2006) filled with nitrogen or filled with ambient air of known composition and by adding definite quantities of pure gases by micro-liter syringes through the septum of the bags. It was found that the best calibration methods were achieved using the PLS1 algorithm of the software and blanking all spectra parts except regions with no or only weak overlapping spectral lines. These lines were determined by qualitative measurements with variable concentrations and by spectra simulation using USF-HITRAN-PC (Figure 3; HITRAN, 1992). It was found that the best accuracy could be achieved when the standard lines and sample lines did not differ in the magnitude of absorbance. Although it is possible to develop a calibration method, which works for e.g. CO₂ from 200 ppm till 20 Vol.-%, usually strong deviations between predictions and real concentrations will be measured on the low or high end of the concentration range. Therefore, for evaluation of high variable gas concentrations in the sample, e.g. from compost air, intensity adopted calibration software tools had to be applied.

CO-Flux Measurements During Laboratory Compost Studies

For laboratory experiments, the plant material (1 kg sun dried mixed herbage from landscape management and conservation) was wetted up to a moisture content of ~70 weight-% by soaking with two liters of water and waiting for 24 h. Composting studies were performed with nitrogen amendments, specific CO studies at a later date without any amendment except water. The wetted plant material was then placed into a temperature-controlled, ventilated chamber applying a constant air exchange rate (Figure 4). Substrate temperature was monitored with thermocouples in the center, top, and bottom of the chamber. Experiments were carried out to study emission rates of different gases as influenced by substrate temperature, moisture content, and nitrogen content, and chamber aeration rate. The latter was manually regulated via a peristaltic pump and viscosity flow meters and the exact rate was measured by a drum gas meter (Ritter®, Figure 4). The composition of the input air and of the air leaving the chamber was analyzed by FT-IR. Gas production rates were calculated on the base of volume flow rates and concentration differences.

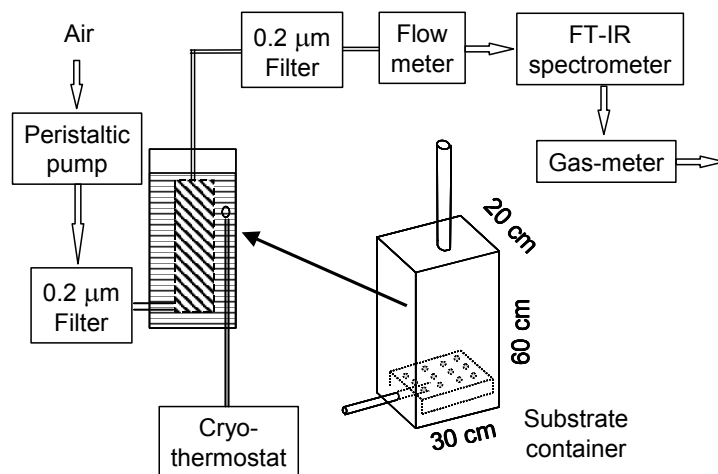


Figure 4. Scheme of laboratory gas measurements.

CO-Flux Estimates at Green Waste Compost Heaps

For the composting of green waste, a typical, trapezoidal compost heap was made of mixed green waste from landscape management and conservation. The initial dimensions of the compost heap were 11 m x 7 m x 3 m high. The initial mass of the compost heap was 14800 kg with a carbon content of 4300 kg. Its mass was measured by means of a trailer and two axle balances with a resolution of 20 kg and an accuracy of 50 kg at maximum load of 25 t. Mass measurements were repeated after heap turnover 32 and 70 days after start and at the end of composting at 194 days. The carbon content of the material was analyzed by means of three mixed samples of the compost material using a TOC analyzer. The initial carbon mass decreased to 730 kg after rotting. For gas sampling, compost air from inside the heap was sampled by a small diaphragm pump through inserted polyethylene pipes at nine different measuring points into gas bags each of 10 dm³ volume at each measuring day (Figure 5).

CO-Flux Measurements at Dung-Windrows

The CO flux from dung windrows was calculated from concentration increases inside static gas flux measuring chambers, which had had a volume V of 0.189 m³ and a bottom area A of 0.292 m². Preliminary studies using CO₂ sensors with data loggers (Testo AG, Model 950) and a photo-acoustic multi-gas monitor (Brüel & Kjær, Model 1302) indicated that a sampling time of 10 min was sufficient for a flux measurement under most circumstances. (Hellebrand and Kalk, 2000; 2001).

Results and Discussion

CO from Green Waste Compost Heaps, from Dung Windrows, and from Plant Litter

The experimental results indicated that CO production depends on the availability of oxygen. Initially, similar maximum CO concentrations were observed at all measuring tubes after onset and after turnover of the compost heap. After significant progress of the composting process, highest CO concentrations were observed at measuring tubes 1, 4, and 7 in the upper layer of the compost heap, and lowest CO levels were measured at the bottom part of the heap (pipes 3, 6, and 9 in Figure 5), consistent with a typical aeration, i.e. O₂ availability gradient inside a compost heap. The mean of the concentrations measured at the nine tubes was utilized for the determination of the total emissions in connection with weighing and analysis of the rotting compost material. Over the six months of composting, 3570 kg carbon were released as CO₂-C and 1.7 kg as CO-C (approximately 0.04 % referring to the initial carbon mass or around 0.05 % with reference to CO₂-C). In relation to the surface, the mean CO flux was 20 mg CO m⁻² h⁻¹ (Hellebrand, 1998).

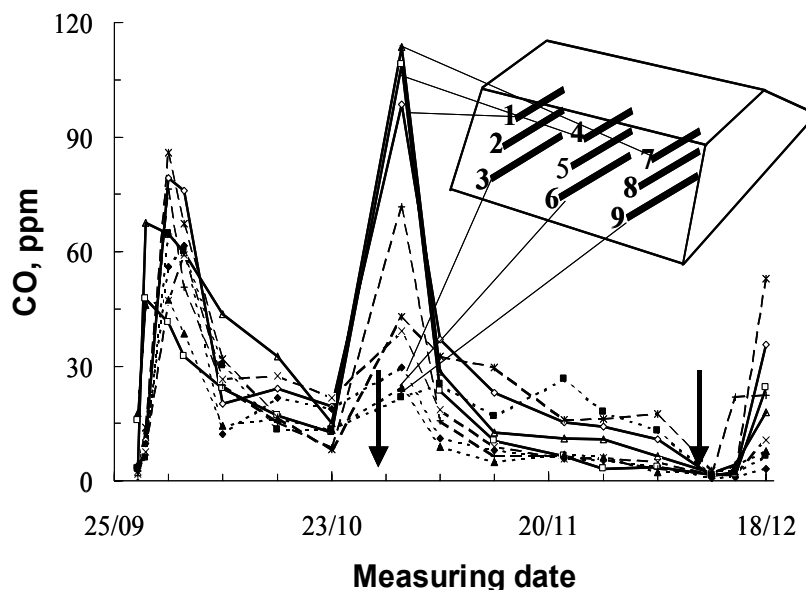


Figure 5. Concentration of CO at the different measuring tubes and scheme of compost heap with measuring tubes (mixed herbage from landscape conservation on bare soil). Arrows indicate dates of turning the compost.

Similar emissions were measured during composting of farm waste. At fresh windrows of animal waste, the flux rates were between 30 and 100 mg CO m⁻² h⁻¹. After a period of one to two weeks, emissions dropped to very low levels, and during early spring and late fall, uptake was occasionally recorded when the ambient CO mixing ratio was high. The accumulated CO emissions during composting of farm waste were calculated by interval integration. Per composting period, the total fluxes were between 1.6 and 3.7 g m⁻². Six periods between seven weeks and up to three months of windrow composting of farm waste were evaluated. The mean emission from all periods was 3 g m⁻² and a mean value of 4×10⁻⁴ was calculated for the ratio between total CO-C and CO₂-C fluxes.

Enhanced CO levels were also observed during several test measurements of air sampled from heaps of fallen leaves and heaps of mowed grass in different parks of Potsdam. Air from the center of the heaps was taken via polyethylene pipes and gas bags using a small diaphragm pump. The measured concentrations ranged from 3 to 22 ppm.

CO from Sterilized and Non-Sterilized Hay (Mixed Herbage from Landscape Conservation)

After evidence of CO emissions from composting had been established, the influence of temperature (Figure 6) and aeration rate on emissions was analyzed by laboratory measurements using hay from landscape management and conservation (sun dried mixed herbage). The results were puzzling at first, as they showed an initial maximum of CO after incubation, dropping to a nearly constant but much lower level for about four days, then rising slightly again.

An explanation may lie in the way the experiment was performed, not allowing sufficient aeration (constant rates between 25 and 150 cm³ min⁻¹) to keep the substrate aerobic while drying it slowly at the same time. If CO-emissions are indeed of thermochemical instead of biochemical origin, the previously established emission dependence on O₂ availability and wetness (Schade, 1997) predicts a higher initial emission when the material is wet and has not consumed the chambers O₂ significantly yet. Emissions then drop because the composting becomes partially anaerobic due to the low aeration rate. As the material dries out, a more

aerobic environment is slowly reestablished and CO emissions start increasing again, albeit at lower levels as the material is drier.

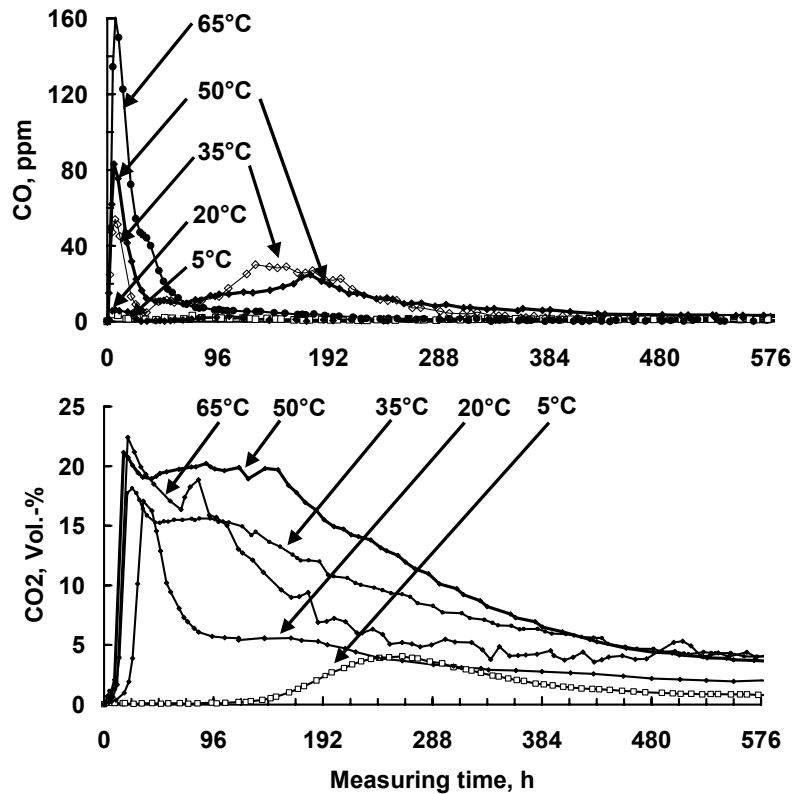


Figure 6. Temperature influence on CO and CO₂ concentration trends in the air from degrading of hay (mixed herbage) from landscape conservation, 70 % moisture content (w.b.), ventilation rate 25 cm³ min⁻¹ kg⁻¹ substrate.

To test the thermochemical CO emission assumption, the filled substrate container, separated by bacteria-tight 0.2 µm filters from ambient air, was sterilized in an autoclave at 136 °C for three hours. The subsequent measurements of CO₂ inside the chamber (previously 20 Vol.-% versus now 0.2 Vol.-% (Figure 7)) proved the effectiveness of the sterilization, while, at the same time, nearly unchanged CO levels evidenced thermochemical oxidation as the dominant CO emission process.

The level of CO and CO₂ inside the chamber was dependent on temperature. An Arrhenius plot of calculated formation rates after the minimum (mean values of the periods from 96 h to 144 h) resulted in activation energies of ~65 kJ mol⁻¹ (CO) and ~43 kJ mol⁻¹ (CO₂) for the samples without sterilization (Figure 8). In experiments with sterilization, the Arrhenius plot of production rates (mean values of the periods from 48 h to 144 h) gave activation energies for the thermochemical oxidation of ~52 kJ mol⁻¹ for CO and ~30 kJ mol⁻¹ for CO₂ (Figure 9). These findings are in line with the results obtained by Schade (1997; Schade et al., 1999).

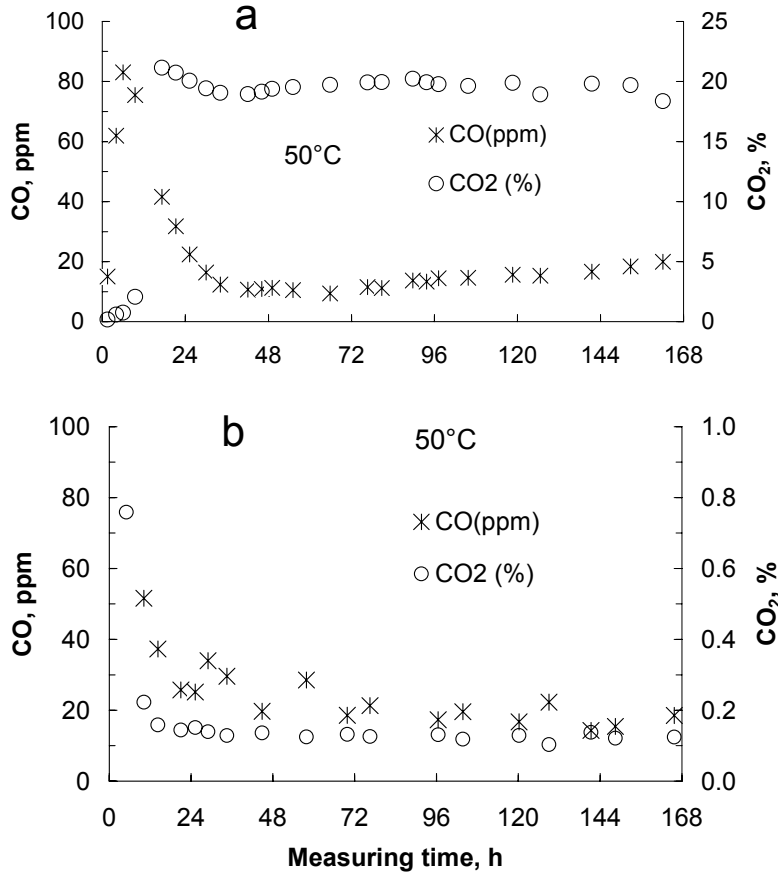


Figure 7. CO and CO₂ concentration trends in the air from degrading of hay (mixed herbage) from landscape conservation, 70 % moisture content (w.b.), temperature 50 °C, ventilation rate 25 cm³ min⁻¹ kg⁻¹ substrate. a) without sterilization b) sterilized for 3 h at 136°C

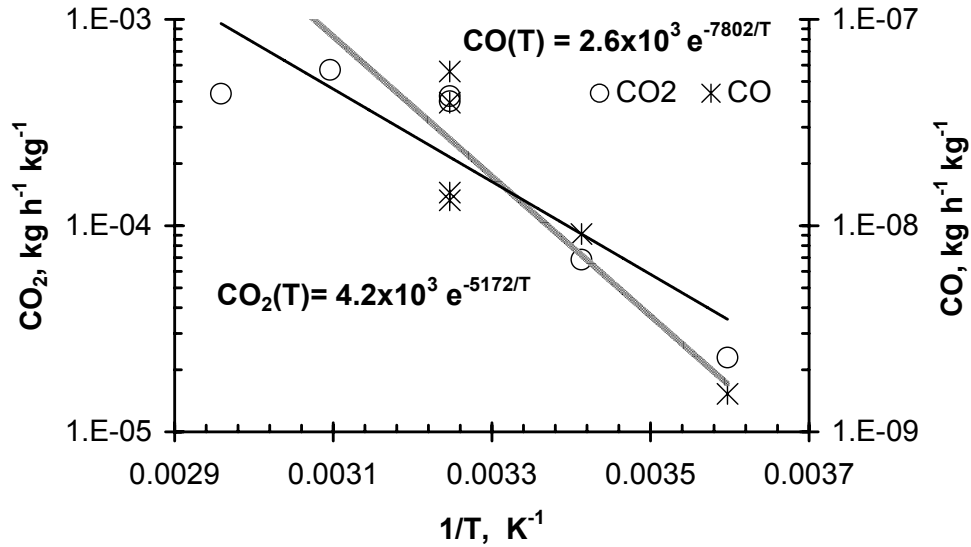


Figure 8. Production rates of CO and CO₂ between 278 K and 338 K plotted against inverse temperature. Non-sterilized substrate, 70 % moisture content (w.b.), ventilation rate 25 cm³ min⁻¹ kg⁻¹ substrate.

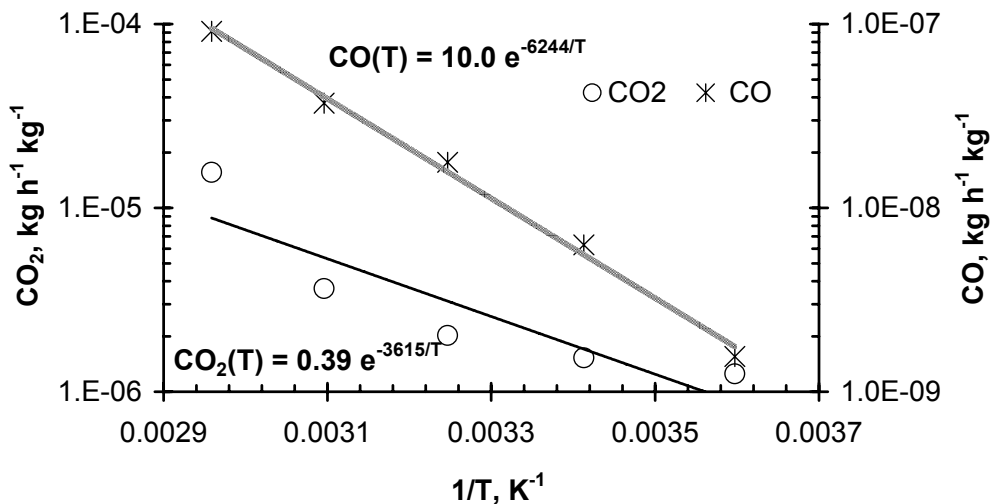


Figure 9. Production rates of CO and CO₂ between 278 K and 338 K plotted against inverse temperature. Sterilized substrate, 70 % moisture content (w.b.), ventilation rate 25 cm³ min⁻¹ kg⁻¹ substrate.

Conclusions

Very high concentrations of CO have been observed inside dedicated composting and other heaps of degrading plant matter. Flux chamber measurements confirmed that CO is emitted to the atmosphere from the composting process and its ratio to simultaneously emitted CO₂ lies between 0.1 and 1 % CO-C/CO₂-C. Furthermore, our laboratory measurements confirmed that the production process of CO is independent of microbial activity in the substrate, but is promoted by increased temperatures and wetness, and needs

oxygen. These results are consistent with the conclusions of Conrad and Seiler (1985b) and Schade (1997, Schade et al., 1999), and reconfirm that the slow decomposition of organic material is a source of atmospheric CO. The results also confirm that, in principle, CO emissions can be calculated from the activation energy and the pre-exponential factor calculated from Arrhenius plots based on laboratory measurements (Schade and Crutzen, 1999). Previously estimated global CO emissions from this process ($\sim 40 \text{ Tg CO y}^{-1}$) were low compared to the dominant anthropogenic sources of CO, such as fossil fuel combustion. However, they may often occur in regions that receive comparatively low inputs of anthropogenic CO emissions, such as in the tropics.

The specific composting processes evaluated here are commonly carried out in Germany for a multitude of organic wastes, including yard wastes and municipally collected household biowastes. Composting facilities in Germany that use the more common aerobic composting process handled an annual throughput of roughly 7200 Gg (1Gg = 1 kiloton) of fresh waste in 2003 (Statistisches Bundesamt, 2005). Based on a wetness of 50% and an assumed average carbon mass loss during the composting process of another 50%, we estimate that between 2 and 20 Gg CO-C may be emitted into the atmosphere from biowaste composting in Germany. That is, compared to dominant road traffic emissions of 1700 Gg CO per year (UBA, 2005), a relative amount of less than 1%, and therefore likely not contributing significantly to tropospheric ozone formation. Although negligible on this basis, CO emissions from the composting facilities may contribute significantly to local air pollution under stagnant meteorological conditions.

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Comparing Field Odor Assessment Methods with an Atmospheric Dispersion Model for Calibrating Setback Estimation Tools for Livestock

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Abstract

Downwind odor assessment to develop setback estimation or siting tools for new livestock facilities is a high priority in several states. The University of Nebraska is currently developing an Odor Footprint Tool (OFT) to assist livestock producers and the general public in understanding the impact that odors have on downwind residents. Air dispersion modeling is a promising approach for assessing the area near livestock facilities for odors. A Gaussian plume dispersion model, AERMOD (AMS/EPA Regulatory Model) was used to predict downwind concentrations for the OFT. Calibration of this procedure was conducted by human receptors using two different methods, on-site intensity and mask scentometry. There was only a weak relationship between mask scentometry and field intensity ($R^2=0.51$). The experiments were conducted at cattle feedlots and swine lagoons. Several statistical tests were used for assessing model performance, it appears that the mask scentometer technique agrees better with model predictions than field intensity. Field techniques to ground truth models are needed to better understand and predict odor impact to communities.

Introduction to Field Odor Assessment

Field olfactometry is defined as the use of a technique or device to determine dilutions to threshold (D/T) of an odor in the ambient atmosphere. The individuals making such measurements have been referred to by a number of names, including nasal rangers, field sniffers, (field) odor assessors, and (field) odor surveyors. Scentometers were first developed in the late 1970's by Barneby and Sutcliffe and are a low-cost (about \$500 per unit) portable instrument (Sheffield, 2004). The scentometer dilutes ambient air with air that has been "cleaned" by a charcoal chamber. Disadvantages of the instrument include a lack of control of inhalation rates by different panelists, the discomfort of glass inhalation tubes, the odor fatigue caused by poor nasal sealing to the glass ports, and the inability to prevent olfactometry fatigue between measurements.

The mask scentometer (Henry, 2004; Sheffield et al., 2004), as shown in Figure 1, is functionally similar to a conventional box scentometer. It is comprised of a ¼-face respirator mask with two cartridges. The first cartridge is fitted with a charcoal filter having a plenum and two ¼-inch holes for drawing air into the mask. The second cartridge is fitted with a dial mechanism that is rotated to change the ratio of filtered air to ambient (odorous) air to established dilution ratios of 170, 31, 15, 7, and 2. The advantage of the mask scentometer is that it minimizes odor fatigue, allows for one-handed operation, and can be easily operated over long periods of time without fatigue. Users sniff the air that is drawn through the two sets of ports and mixed within the mask chamber, generally going from the highest dilution setting to lower dilution ratios in order to determine the dilutions to threshold. Users adjust the dial setting every 5-8 seconds, allowing a D/T measurement to be taken every 30 seconds.



Figure 1. Mask Scentometer

The field intensity method was developed by the University of Minnesota, adapted from ASTM Standard E 544-99 “Standard Practices for Referencing Suprathreshold Odor Intensity.” The method references field intensity measurements to a standardized n-butanol scale. The scale used was adapted from the static scale method of 12 concentrations ranging from 10 to 10,000 ppm n-butanol, reduced to 5 levels so that the odor intensities can be committed to memory by assessors. When measurements are made in the field, assessors wear a ¼-face respirator mask between intensity assessments. In the typical scenario, 60 intensity assessments can be made in 10-15 minutes.

Sheffield et al. (2004) conducted a 6-week study of five odorous sources in Idaho to quantify the variability of field and laboratory olfactometry methods and their correlation to odor intensity, as well as ammonia and hydrogen sulfide concentrations. The comparison of four olfactometers indicated that the use of laboratory olfactometry (dynamic, triangular forced-choice) or Nasal Ranger[®] field olfactometers resulted in the least amount of variability across odor sources. Significant differences with poor correlation were found between odor intensity methods and the use of Tedlar bags for storage of ambient air samples.

The University of Minnesota employed trained panelists as field sniffers and using the field intensity data they collected to scale INPUFF-2 model results in the development of the Odor From Feedlots Setback Estimation Tool (OFFSET). The procedure implemented was a modified version of the one used in Germany by Hartung and Jungbluth (1997). Seven individuals were deployed perpendicular to a centerline marked off from 25 to 400 meters from the source and spaced 5 to 20 meters apart. They were provided with stopwatches, charcoal filter masks, and clipboards with data sheets. Field intensity measurements were assessed once every 10 seconds for a 10-minute period. The masks were removed only long enough (a few seconds) to assess the intensity of the ambient odor (Jacobson, 2000).

Since the early 1980s air dispersion models have been studied to predict odor concentrations downwind from livestock sources (Janni, 1982; Caeny and Dodd, 1989; Mejer and Krause, 1985; Lorimer, 1986; Omerod, 1991; McPhail, 1991; Gassman, 1993; Chen et al., 1998). Only a few studies exist comparing the results of dispersion models to field odor panelists (Li et al. 1994; Hartung and Jungbluth, 1997, Zhu et al., 2000; Guo et al., 2001).

Methodology

Assessments were made at a manure treatment lagoon from and two cattle feedlots. Sites were selected that were topographically level surrounding the source, were isolated from other odor sources, and had few or no obstructions surrounding the site within 500 meters. This was deemed to be most desirable for dispersion modeling and to minimize unwanted odors from other sources.

The data set represents downwind odor data from a lagoon assessed on two different days - once in the fall of 2003 and once in the spring of 2004 - and two different feedlots one assessed in the spring of 2004 and

the other in the fall of 2003. At least 7 and up to 10 individuals were used to form transects downwind of the facility on the days of the experiments.

Lagoon

The lagoon treated manure from 1,250 swine finishers, 800 sows and gilts, and 800 nursery pigs. The anaerobic lagoon was typical of many Nebraska lagoons and was uniquely situated 1/2 mile to the east of the production barns. This allowed the researchers to isolate the lagoon odors from other odors from the facility or neighboring odor sources. The lagoon was situated just south of an east-west gravel road, and was surrounded by cropland with little topographical relief surrounding the facility in all directions.

The flat terrain allowed transects to be set up to the north of the facility on a fall day when the prevailing wind was from the southwest. Transects were established at 111, 153, 198, and 246 meters from the lagoon. The day of the assessment was warm and sunny, with no cloud cover and a very gentle and shifting breeze. Wind speeds during these assessments were between 1-3 m/s (A and B stability class).

On the day of the spring assessments, the wind was from the northeast instead of the south. Transects were established downwind at 58, 73, 103, and 134 meters. Atmospheric conditions during the day would be described as sunny with 4-7 m/s wind speeds (A and B stability class).

Feedlot

The capacities of the studied feedlots were 4,200-head and 1,600 head, and both were located in central Nebraska. Terrain near the facility had very little topographical relief. The width of the source was several times greater than the width of the sniffer transect. Odor monitoring was conducted on a warm, sunny and windy spring day and a cold overcast day. Wind speeds during the warm assessments were between 3-5 m/s (and B and C stability) and 5-9m/s (and D stability class) for the colder assessments. Transects were established downwind at 150, 265, 390, and 504 meters for the warmer feedlot assessments and at 106, 308, and 505 meters for the colder feedlot assessments.

Assessment Technique

Individuals, or field sniffers, were recruited who could refrain from drinking caffeinated drinks, eating spicy foods, and wearing perfume and cologne on the days of participating in the study. Field sniffers attended a daylong training seminar that instructed them in the use of the mask scentometer and how to assess odor intensities. Mask scentometer readings or "Mask DT's" are taken by turning a dial on the mask through a series of notches that correspond to increasing dilution ratios of ambient air (odorous air) to air that has been cleaned with a carbon filter (clean air) (Henry, 2004). When the dilution first reaches the point at which the sniffer can recognize the odor, this DT is recorded. This DT is considered to be the same as an Odor Unit (OU), which can be used directly for model comparison.

Next, sniffers recorded odor intensities. Sniffers were trained to correlate livestock odors with a reference odorant, n-butanol. Sniffers used the Minnesota 10- step n-butanol scale ranging from 0 to 5 in 1/2-step increments, where 1 is considered barely noticeable and 5 is considered very annoying. Sniffers removed their masks briefly to take this measurement between breaths. Odor intensity was then converted to D/T and compared to model predictions for use in scaling and calibrating models.

Experimental Set-up

A weather station was placed on the facility the day before a sniffing event by a researcher. It was located, if possible, in the plume where the sniffers were expected to locate the next day. The weather station was instrumented to record, net solar radiation, temperature, relative humidity, wind speed, wind direction, solar heat flux, and barometric pressure.

On the day of the experiment, sniffers calibrated them selves at a location away from the plume near the site with n-butanol. They also calibrated themselves against each other, by subjecting themselves to varying levels of the target odor (near outer fringes of plume) and agreeing among themselves that they were all reporting intensity and scentometer readings consistently. They would dawn their masks before entering the plume. First, mask scentometer measurements were taken every 30 seconds, evaluating every DT within the range of the instrument, if no odor was detected, a 0 was recorded. Mask scentometer

measurements were taken for 15 minutes. Next, field intensity measurements were taken every 15 seconds for 15 minutes. The sniffers were instructed to become familiar with the target odor and make their assessment on the target odor, this was done to minimize the influence of background odors that at low concentrations can produce a false positive. Sniffers collected intensity data every 15 seconds for 15 minutes, and dilution to threshold (DT) from a mask scentometer every 30 seconds for 15 minutes. Each sniffer was stationed at a location in the transect for a 30 minute period before moving to another location. Sniffers were randomly located in each transect, and no sniffer was ever in the same position in the transect on the same day. The mask allowed sniffers to collect two odor measurements sequentially. After a sniffer was notified to start taking measurements, they began taking mask scentometer assessments. After they finished, they waited for the odor intensity signal start, and then collected odor intensity measurements. When they finished with these they would move on to the next transect.

Sniffers recorded measurements on a data sheet. Stopwatches were used that could be set to chime at a set interval. A lead sniffer was identified and synchronized his watch with the weather station clock so measurements would correspond to modeled data. The lead sniffer would then start all of the sniffers at the same time. Sniffer responses were arithmetically averaged for each assessment, therefore the average for an assessment represents the average of 30 measurements for the mask scentometer and 60 measurements for field intensity. In total there were over 4,500 mask scentometer assessments and 9,000 field intensity assessments conducted over the course of the study.

Emissions

All sites studied were area sources and measured using a stainless steel wind tunnel device. The tunnel, shown in Figures 2 and 3 was constructed according to plans from Schmidt and Bicudo (2002) originally designed by Jiang et.al. (1995), consisting of an inlet PVC stack, blower, expansion chamber, air filter, pressure gauge, tunnel body, mixing chamber, outlet PVC “T” and two gas sampling ports. Wind tunnels are portable, open-bottomed enclosures that are placed over the emitting surface. Ambient or filtered air is drawn or blown through the tunnel to mix with and transport the emissions away from the emitting surface. This is intended to simulate the convective mixing and transport process present above the emitting surface.



Figure 2. Collecting Emissions with a wind tunnel from a lagoon



Figure 3. Collecting Emissions with a wind tunnel from a feedlot

Emissions were estimated by collecting a bag sample and having that sample analyzed by an olfactometry lab. The reported dilution to threshold was then converted to a standard emission rate adjusted for height and wind speed of 1 meter height and 1 meter per second. The emission rates were then adjusted for each model run for varying wind speed and stability class. It is accepted that atmospheric stability affects odor emission rate. This is because stability affects both the slope of the wind speed profile and the degree of turbulence. Odor concentrations measured using wind tunnels were adjusted to a standard 1m/s and 1 meter high standard emission rate (Table 1), using a procedure outlined in Smith and Watts (1994).

Hence, for the wind tunnel data, the odor emission rate varies with tunnel wind speed according to:

$$EV = EM * (u_T/u_M)^{0.63}$$

Where,

EV = Odour emission rate at tunnel wind speed, u_t

EM = Base odour emission rate measured at tunnel wind speed, u_m

u_T = wind tunnel speed (m/s)

u_M = Tunnel wind speed for measurements, i.e. 0.3 m/s.

0.63 = adopted exponent (for feedlots), 0.5 for lagoons

The formula to convert wind speed at 10 m (for example) to wind speed at 0.125 m (i.e. tunnel speed) in neutral stability is:

$$U_T = U_{10} * \text{LN}(0.125/Z_{0.2}) / \text{LN}(10/Z_{0.1})$$

Where,

U_t = wind tunnel speed (m/s) at 0.125 m

U_{10} = wind speed (m/s) at 10 m

$Z_{0.1}$ = surface roughness for anemometer at 10 m (0.3 m)

$Z_{0.2}$ = surface roughness for pond (0.03 m).

Kaye and Jiang (1999) outline a procedure (Table 1) to adjust emissions for use in AUSPLUME for stability and wind speed. The procedure is theoretical and not substantiated with experimental data, but is reasonably consistent with Ormerod (1991 and 1994).

Table 1. Adjustments made to emission rates based on wind speed and stability class (Kayle and Jiang(1999))

Wind Speed Category	Speed Rate (m/s)	Median Wind Speed (m/s)	Stability Class					
			A	B	C	D	E	F
Relative Odour Emission Rate (%)								
1	0-0.6	0.3	86%	86%	80%	72%	46%	30%
2	0.6-1.2	0.9	149%	149%	139%	125%	80%	52%
3	1.2-1.8	1.5	192%	192%	180%	161%	104%	67%
4	1.8-2.4	2.1	227%	227%	213%	190%	123%	79%
5	2.4-3.0	2.7	257%	257%	241%	216%	139%	90%
6	>3.0	6.5	399%	399%	374%	335%	216%	139%

Emissions collection from area sources such as lagoons and feedlot surfaces are difficult to perform. In this study, the samples were in the lower ranges of detection capable by the laboratories used and were in the range of 8-34 DT. Another issue with respect to the olfactometry results is the lower detection limit (LDL) of the olfactometer. The LDL is the lowest value of the air quality characteristic which can be distinguished from a zero sample. Most olfactometers have a detection rate of 20-50 OU/m³ (Gostelow et al., 1993). Given that some of the olfactometry results used to determine emission rates are below the LDL of the olfactometers, there is a large degree of uncertainty concerning the emission rates of the sources studied. The emission rates used for each of the assessments is shown in Table 2.

Table 2. Emissions used for Individual Model Runs

Location	Run	Time	Emission OU/m ² s	Stability class	Wind speed m/s
Lagoon 1					
	1	Met tower failure			
	2	1:50-2:20	4.7	B	2.7
	3	2:30-3:02	3.5	B	1.5
	4	3:09-3:40	2.7	A	0.9
Lagoon 2					
	1(DT)	10:05-10:39	8.9	B	5.4
	1(I)	10:05-10:39	8.8	A	5.2
	2(DT)	11:27-12:00	9.8	B	6.5
	2(I)	11:27-12:00	9.4	B	6.1
	3(DT)	10:47-11:20	9.7	B	6.4
	3(I)	10:47-11:20	9.1	B	5.6
	4(DT)	10:05-10:39	7.0	A	3.3
	4(I)	11:13-11:39	8.2	B	4.6
	Random	14:47-15:03	8.4	B	4.8
Feedlot 1					
	1	9:53-10:35	15.1	D	8.6
	2	11:16-11:47	14.8	D	8.3
	3	12:05-12:30	13.6	D	7.3
	4	2:00-2:31	12.3	D	6.2
	5	2:49-3:19	12.6	D	6.4
	6	3:32-5:02	11.2	D	5.3
Feedlot 2					
	1	10:57-11:27	2.3	C	3.2
	2	11:41-12:11	2.7	C	4.4
	3	12:14-12:50	3.0	C	4.9
	4	13:02-13:38	2.8	C	4.4

Modeling

The adjusted emission rates were then used to model each individual assessment session. The source or sources were created in AERMOD as area sources. On-site meteorological data was collected at 1-minute time steps, and applied to the model. AERMOD and most models only accept 1 hour time step meteorological data, so 1-minute time step data was modeled as hourly data and assumed to be 1-minute predictions without any adjustment.

Relationship Between Field Intensity and Mask Scentometer

The first analysis completed was to determine if a relationship existed between the field intensity method and mask scentometry (Mask DT). It was assumed originally that only one method would be needed to develop the scaling factors for the dispersion model, but if both methods produced the same result, then one could be abandoned for the superior method.

Field data from assessments was processed for each assessment event. Each time a sniffer collected data over the 15 minute period the data was averaged and a maximum and minimum value was determined. It is assumed that the odors were similar in magnitude during the 15-minute assessment period, and the on-site meteorological data was queried to determine if wind speed and direction would have changed substantially to invalidate this assumption. All measurements were taken during consistent conditions and were in the A-B atmospheric stability classes as defined by the SRDT method (USEPA, 2000). It is assumed that the odors during the assessment period were relatively consistent and back-to back measurements can be compared. Individual responses were converted to the actual DT observed, and the individual intensity responses were converted to DT as per Jacobsen et al (2000). The results reflect the average of the DT's for an assessment period. It should be noted that taking the average of the intensities converted to DT will yield a different arithmetic result. The equation for converting swine intensities to DT is shown below:

$$DT_{swine} = 9.429(e^{1.085I})$$

$$DT_{beef} = 8.367(e^{1.078I})$$

Where *I* is the intensity of the odor as determined by a trained assessor.

The conversion for the mask scentometer was also done according to Table 3.

Table 3. Mask Scentometer Settings

Dial Position	Mask Scentometer D/T
1	170
2	31
3	15
4	7
5	2

The calculated field intensity DT and mask scentometer (mask DT) by the same sniffer at a given location downwind of the source emission were compared. A statistics analysis was completed and the results are shown in Table 4. As can be seen, the mean of the mask DT is almost an order of magnitude less than the mean response from the field intensity or field DT.

Table 4. General Statistics of Mask DT and Field Intensity DT

Mask Scentometer DT		Calculated Field Intensity DT	
Mean	5.1	Mean	58.2
Standard Error	0.97	Standard Error	8.0
Median	1.8	Median	17.9
Mode	0.1	Mode	14.3
Standard Deviation	9.0	Standard Deviation	73.5
Sample Variance	80.7	Sample Variance	5,395.8
Kurtosis	16.9	Kurtosis	5.1
Skewness	3.7	Skewness	2.1
Range	56.8	Range	391.1
Minimum	0	Minimum	8.4
Maximum	56.8	Maximum	399.4
Sum	434	Sum	4,944
Count	85	Count	85
Confidence Level (95.0%)	1.9	Confidence Level (95.0%)	15.8

The standard deviation and sample variance are much less with the mask DT, suggesting that there is less variability with this method than the field intensity method. The mean suggests that the Mask DT is approximately an order of magnitude less than calculated field intensity, which has some implications for modeling because typically model results have been factored up to match intensity assessments by field sniffers (Koppolu et al., 2004).

Next the dataset was analyzed using a t-test assuming unequal variances. The results of a t-test using Microsoft EXCEL are shown in Table 5. The P critical two tail is less than 0.01 indicating that a significant difference between Mask DT and calculated Field Intensity DT. A single factor ANOVA produced the same result.

Table 5. Statistical t-test Results, Mask and Field Intensity DT's

	Mask DT	Field Intensity DT
Mean	5.10	58.17
Variance	80.72	5,395.83
Observations	85	85
Hypothesized Mean Difference	0	
Degrees of freedom	87	
t Stat	(6.61)	
P(T<=t) one-tail	1.47E-09	
t Critical one-tail	1.66	
P(T<=t) two-tail	2.95E-09	
t Critical two-tail	1.99	

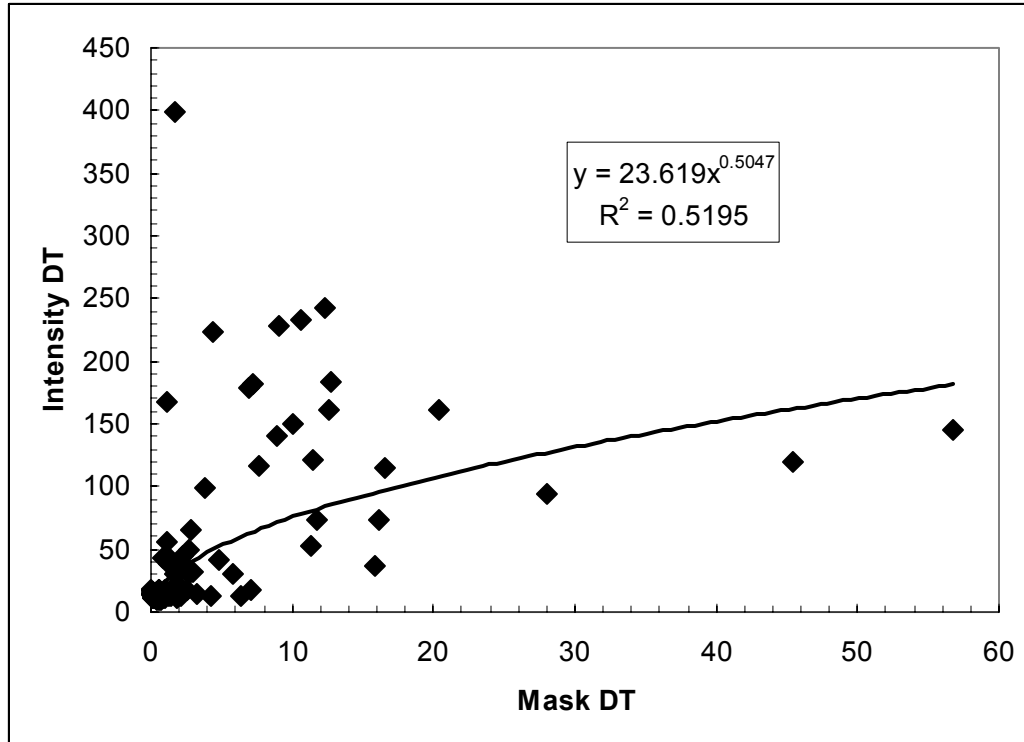


Figure 4. Field Intensity DT versus Mask DT

Figure 4 shows a scatter plot of the calculated Field Intensity DT versus Mask DT. The relationship between the two is best represented with a power relationship and produces a coefficient of correlation of 0.52 (R^2) indicating that there is not a very good relationship between the two methods. A more robust analysis of the relationship between mask scentometry and field intensity is presented in Henry (2005).

Henry (2005) also reported that a relationship between peak and mean measurements may exist for the mask scentometer ($R^2=0.75$), but that this did not hold true for field intensity ($R^2=0.33$). Although peak to mean ratio's appeared to be similar for both field methods, scatter for field intensity was more severe than for Mask DT. Finally, Henry (2005) reported that an analysis of individual data revealed that some sniffers provide a consistent peak to mean ratio, while others do not. Additionally some individuals may be very consistent when using the mask scentometer, but not when reporting field intensity, and vice versa. Some were good at both and some at neither. Henry (2005) suggested that that individual sniffer sensitivity be assessed in future work with field assessors and that a similar type of analysis be performed between assessors to ensure consistent results are being reported in any future work that will be used to ground-truth air dispersion models.

Comparison of Model Results to Field Assessments using Statistical Tests

Statistical tests were used to evaluate model performance. In this study, these included model bias, fractional bias, normalized mean square error, root mean square error, skill_v and skill_r by Wilmott (1981), Pielke (1984) and Hurley (2000). Common measures of ascertaining model performance include the mean, standard deviation, coefficient of correlation, and the coefficient of variation as statistical measures.

Model Bias (MB):

$$MB = \frac{1}{n} \sum_{i=1}^n (O_i - P_i) \quad \text{Where: Ideal value for MB} = \text{zero}$$

N = Number of observations
 O_i = Observed
 P_i = Predicted

Fraction Bias (FB):

$$FB = 2 \frac{\bar{O} - \bar{P}}{\bar{O} + \bar{P}} \quad \text{Where: Ideal value for FB is between } \pm 0.67; \pm 2 \text{ is good}$$

\bar{O} = Mean of observed values
 \bar{P} = Mean of predicted values

Normalised-Mean-Square Error (NMSE):

$$NMSE = \frac{\overline{(O_i - P_i)^2}}{\bar{O}\bar{P}} \quad \text{Where: Ideal value for NMSE} = \text{zero}$$

O_i = Observed
 P_i = Predicted
 \bar{O} = Mean of observed values
 \bar{P} = Mean of predicted values

Root-Mean-Square Error (RMSE)

$$RSME = \frac{1}{n} \left[\sum_{i=1}^n (O_i - P_i)^2 \right]^{1/2} \quad \text{Where: Ideal value for RMSE} = \text{zero}$$

N = Number of observations
 O_i = Observed
 P_i = Predicted

Skill_r

$$\text{Skill}_r = \frac{\sqrt{\frac{1}{n} \sum_{i=1}^n (P_i - O_i)^2}}{O_{std}} \quad \text{Where: Ideal value for Skill}_r < 1$$

N = Number of observations
 O_i = Observed
 P_i = Predicted
 O_{std} = Standard deviation of observed values

Skill_v:

$$\text{Skill}_v = \frac{P_{std}}{O_{std}} \quad \text{Where: Ideal value for Skill}_v = 1$$

N = Number of observations
 O_i = Observed
 P_i = Predicted
 O_{std} = Standard deviation of observed values

Table 8 shows the results of the aforementioned statistical tests. While good agreement for all tests is well beyond the range for ideal, with the exception of Fractional Bias, in most cases the mask method is closer to ideal than the intensity method. This suggests better agreement may be expected from model predictions when compared to mask scintometer assessments (and related devices) than field intensity. It should be

noted that there is a considerable amount of variability with respect to individual assessors and the oflactometry used to determine the emission rates of the sources.

Table 8. Statistics for comparison of model predictions to Mask Scentometer and Field Intensity Assessments

Test	Ideal	Lagoon 1		Lagoon 2		Feedlot 1AM		Feedlot 1PM		Feedlot 2	
		Mask	Intensity	Mask	Intensity	Mask	Intensity	Mask	Intensity	Mask	Intensity
Model Bias	0	-1.8	11.7	5.2	105	31	86	4.1	52	4.6	32
Fractional Bias	±0.67 to ±2	-1.6	1.5	0.9	1.9	1.4	1.8	0.7	1.8	1.6	1.9
Normalized Mean Square Error	0	10.8	22.7	2.6	59	15.1	34	6.6	38	69	133
Root Mean Square Error	0	0.5	5.5	1.5	25	11	24	2.7	15	2.3	8.3
Skill_r	1	8.7	0.9	1.3	1.5	1.1	1.3	1.0	1.2	1.1	1.4
Skill_v	1	6.5	0.07	0.2	0.01	0.06	0.03	0.08	0.03	0.03	0.01

Comparison of Model Results to Field Assessments using Scaling Factors

For the development of separation distance tools, the scaling factor or peak to mean ratio, is used to “factor” the emission rate of the source for separation distance tools, such as OFFSET (Odor From Feedlot Setback Estimation Tool) and the OFT (Odor Footprint Tool). Measured assessments (both field intensity and mask) were compared to predicted ambient odor levels from AERMOD. These comparisons involve calculating scaling factors (peak to mean ratios) to adjust relatively short-time interval (seconds) odor intensity measurements in the odor plume to AERMOD predictions which are based on relatively long time period (hourly) averages. Depending on the source type (point, area, volume) and the type of facility being modeled Koppolu (2002) reported results indicated that scaling factors in the range of 0.2 to 3900 may be needed to adjust AERMOD predictions to short-term odor measurements.

Scaling factors or peak to mean ratios, a more thorough discussion of the assumptions behind peak to mean ratios is presented in Koppolu (2002), but for the purposes of this paper they are used synonymously. Tables 6 shows the scaling factors for every experiment, each experiment included 3-4 cross sections at distances listed previously. Table 7 show the scaling factors for the different area source types, lagoons and feedlots. Such scaling factors are useful in determining appropriate emission values for OFFSET and the OFT. In general, scaling factors for the mask scentometer are less than those for the field intensity method.

Table 6. Scaling Factors (Peak to Mean Ratios)

	Lagoon 1		Lagoon 2		Feedlot 1AM		Feedlot 1PM		Feedlot 2	
	Mask	Intensity	Mask	Intensity	Mask	Intensity	Mask	Intensity	Mask	Intensity
Average Scaling Factor	0.2	3627	3.3	47.4	6.0	18.2	2.2	18.3	88	136
Standard Deviation of Scaling Factor	0.2	16,525	2.9	55.6	10.0	16.5	4.4	12.6	354	244
	Swine Lagoons						Cattle Feedlots			
	Mask		Intensity		Mask		Intensity			
Suggested Scaling Factors	1.8		1837		32		58			

Conclusions

Trained field sniffers evaluated ambient odor levels using a mask scentometer and calculated field intensity downwind from a cattle feedlot and swine lagoon. There was a significant difference observed between the method of assessing ambient odors downwind from livestock facilities using a mask scentometer and a field intensity assessment method adapted from the ASTM Standard E-544-99 “Standard Practices for Referencing Suprathreshold Odor Intensity” by the University of Minnesota. It appears that the use of a mask scentometer returns a result an order of magnitude less than the same person who performs a field intensity assessment when done in the field back-to-back. The standard deviation and sample variance of the dataset suggests that there is less variability in the mask scentometer data than the calculated field intensity method.

Scaling factors were determined for two area sources, lagoons and feedlots. Mask scentometer scaling factors for anaerobic lagoons (not to include manure storages) and feedlots were 1.8, and 32 respectively. Field intensity scaling factors for anaerobic lagoons were found to be 1,837 and 58 respectively. The smaller scaling factors indicate a closer agreement to model predictions. Several statistical tests were used to evaluate the agreement between the field methods and model predictions. While none of the tests, except fractional bias, reported ideal results, mask scentometer and AERMOD predicted results were in general closer to ideal values than field intensity assessments. This suggests that the mask scentometer, and similar devices operated in the same fashion and with a similar technique (many consecutive measurements over a given period of time) has better agreement with model predictions. This research suggests that AERMOD odor predictions are in better agreement to mask scentometer assessments than field intensity assessments. This finding has substantial implications to the ground-truthing of atmospheric dispersion models.

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A Novel Concept for Modeling Odor Impact from Feedlots

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Abstract

Emissions from feedlot operations are known to vary by environmental conditions. Estimating the emissions from feedlot sources for modeling odors is a difficult challenge. A new approach using a feedlot hydrology model to derive necessary inputs for a feedlot emissions model has been developed. A feedlot pad emissions model that predicts emissions as a function of pad temperature, pad depth, and pad moisture content has been developed. A second model that predicts the emissions from sediment basins and holding ponds and is a function of days since rain, inflow volume, inflow ratio (pond volume), and temperature has been developed. Additionally, appropriate emission values for manure and compost storage areas have been investigated. The use of a feedlot hydrology model to estimate the variables for use in the emissions model allows for variable emissions for existing or proposed sites to be predicted over a long time horizon. The application of this process can be used to model odor impact from feedlots with atmospheric dispersion models. Such an application allows for variable emissions based on climate changes, management, and site-specific conditions. A possible application of this process is the development of simple separation distance tools for small to medium sized feedlots. The feedlot emissions predictions used in the model were based on an extensive set of odor samples collected by a wind tunnel over a range of environmental conditions. A brief explanation of the methodology is presented.

Introduction

Determining appropriate separations between livestock facilities and communities and residences has been an intensely studied issue for two decades in Australia. Estimating the odor emissions from animal production facilities is not an exact science. Most separation distance guidelines, setback tools, or odor criteria have been established using empirical formulae or average annual emissions with wind tunnels or flux hoods and dispersion models. Under dry conditions, well managed feedlots can be characterized by low odor emitting sources per unit area, but with vast size comparatively. Feedlot odor emissions under wet conditions may be 1-2 orders of magnitude higher. Feedlot sources are inherently difficult to collect emissions from because of the low odor concentrations experienced, and the complex range of factors influencing emissions. Finally, other production area sources such as manure storage areas, compost piles, holding ponds, and sediment basins have generally not been previously considered as sources in the modeling of odors from feedlots.

Background

Emissions from feedlots can be separated into several different sources, each with their own characteristics and with variables that define their emission characteristics. The major odor sources at feedlots determined in this study include:

- Feedlot pad or surface emissions: The production area where the cattle are fed. The feedlot pad will have the largest area, will be increasing in pad thickness slowly, followed by a rapid decrease as cleaning occurs. Pad emissions will be most influenced by temperature, depth of manure, and moisture content.
- Sediment basins: Sediment basins are used to collect runoff from the feedlot production areas, temporarily hold runoff from rain events and slowly release them to holding ponds. Sediment basins are characterized as being wet and full of solid laden liquids for several days after a runoff event, and being dry the rest of the time. Sediment basin emissions are influenced by inflow/volume ratios, days since rain, and temperature.

- Holding ponds: Holding ponds are permanent, usually lined structures that store runoff water until it is appropriate to land apply the effluent to cropland. Typically they have a wetted surface year round, unless they are pumped dry. Traditionally they are allowed to fill to their capacity and pumped to their lowest level. Holding ponds emissions are influenced by inflow/volume ratios, days since rain, and temperature.
- Manure storage areas: These are area where manure from the feedlot surface or pad is collected and stored until land application. These areas are characterized by being open areas of piled manure. Manure pad emissions are relatively stable, but are influenced by disturbance to the crusting layer, if present.

The primary odor emission source from feedlots comes from pad emissions. Holding ponds and sediment basins collect and contain the contaminated runoff from the pads and can be a source of emissions. Other sources such as manure storage and compost areas, feed storage areas, drains and collection channels, feed processing areas, and mortality storage areas are assumed to be less significant, or minor, because of their relatively small footprint and odor characteristics.

Additionally, land application activities and sites can be a significant source of odors during manure application events. However, management of these activities are solely at the discretion of the operator, and can be managed to minimize or maximize odor impact to neighboring residents. Because of the large influence that management plays in the impact of odors from these activities, and the very short time horizons these event could impact receptors, it is not considered a reasonable source to include in this process.

Method

Currently feedlot odor emissions used for modeling of odors and setback estimation tools are assumed to be constant (either an annual average or constant) in contrast to previous work and use of models by the industry. Previous work has ignored emission contributions from other production area sources such as holding ponds, sediment basins, and manure storage areas. Recent work by Nicholas et al. (2004a) has shown that feedlot emissions are a function of temperature, moisture (rainfall and runoff), surface area, and time since last event. An odor emissions model has been developed by Feedlot Services Australia Pty. Ltd. (FSA) for open feedlots that predicts the hourly emissions from feedlot pads, holding ponds, sediment basins, and manure storage areas (Atzeni et al. 2004, Nicholas et. al. 2004ab, Hudson et al., 2005). The model is comprised of two parts, the first part estimates pad moisture content, pad thickness, and pond volume using MEDLI, a feedlot hydrology model (Gardner and Davis, 1998), and a nutrient excretion model, BEEFBAL (McGahan et al., 2002). The second component uses the output from MEDLI and a meteorological file for the site to estimate the hourly emissions from production area sources. The model generates an hourly emissions file that can be used in AUSPLUME (Lorimer, 1986) or another suitable odor dispersion model to predict the hourly transport of the plume of odors from the feedlot. An outline of the process presented is shown in Figure 1 below. The results of the emissions model, along with on-site meteorological data, are input to an atmospheric dispersion model to develop odor contours (and eventually odor impact at receptor locations) near a feedlot (Nicholas et al., 2005c, D'Abreton, 2005ab). An example of the results from this process is shown in Figure 2.

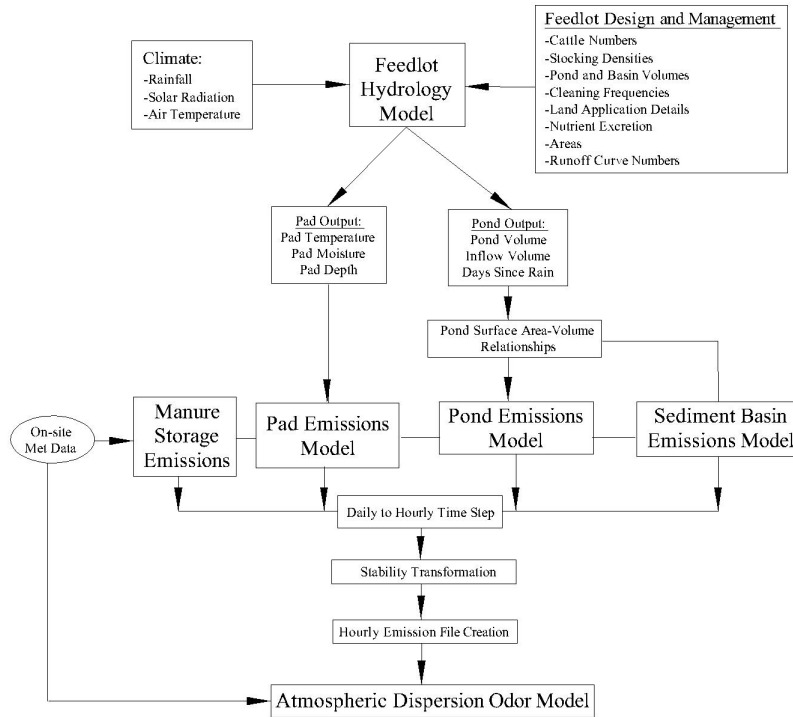


Figure 1. Process of using hydrology model to develop inputs for emissions model and then using an atmospheric dispersion model to predict odor impact

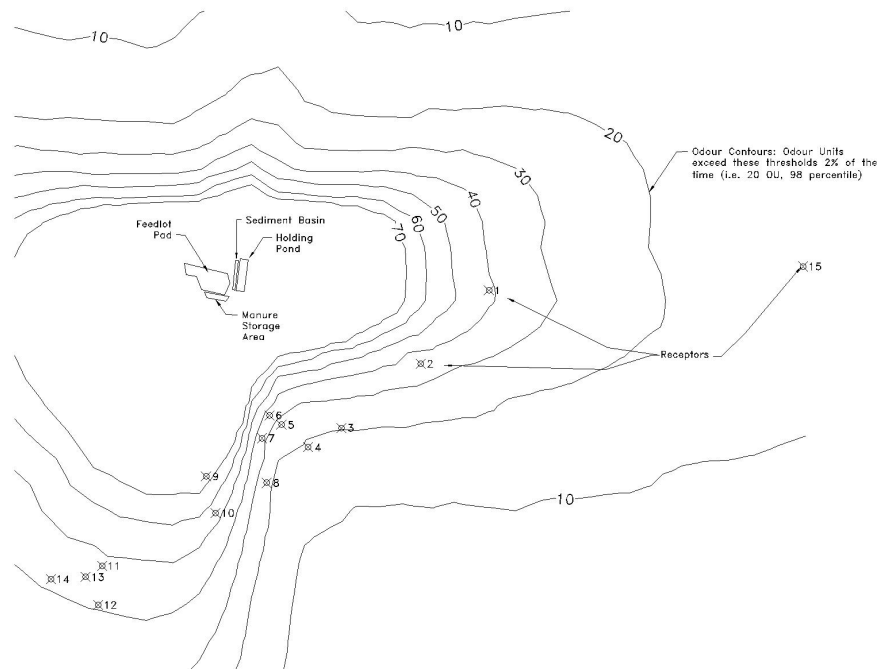


Figure 2. Example of output from AUSPLUME, showing source, receptors, and odor contours for a feedlot.

Conclusion

A process has been developed using a feedlot hydrology model to generate the needed input variables for a feedlot emissions model. A feedlot emissions model was developed from wind tunnel data that provides varied emissions based on environmental conditions for feedlot pads, holding ponds, sediment basins, and manure storage areas. The feedlot emissions model is capable of providing hourly variable emissions for feedlot pads, holding ponds, sediment basins, and manure and compost storage areas. The emissions data can then be converted to a file that can be used in an atmospheric dispersion model for evaluating odor risk for a specific site.

Using a feedlot hydrology model to derive the parameters needed to predict the variable emissions from feedlot production facilities could be used to predict the odor impact from new facilities and expansions. Additional data is needed to confirm that the feedlot pad emissions model reasonably predicts the pad emission rate for a more varied set of environmental conditions. Further work is needed to verify or ground-truth the use of the odor assessment process described above, possibly by using trained assessors, neighborhood surveys, or odor diaries. Such studies would further validate the robustness of this approach.

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Monitoring of Air Quality and Atmospheric Depositions in Background Areas in Denmark

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Abstract

Since 1989 concentrations and depositions of eutrophying air pollutants have been monitored in Denmark within the framework of the Danish Background Air Quality Monitoring Program. The program is based on a combination of measurements and model calculations with regional and local scale models. The results have shown that the average depositions of nitrogen to Danish marine waters are 10 kg N ha^{-1} and hence the atmospheric depositions contribute significantly to the large anthropogenic supply of nitrogen to the Danish marine waters. This excess supply of nitrogen leads to regular episodes with oxygen deficiencies followed by death of fish and benthic fauna. Moreover, the average annual depositions to Danish natural ecosystems are about 16 kg N ha^{-1} thereby exceeding the critical loads of many of the sensitive ecosystems. Since 1989 the total deposition of nitrogen has decreased by about 23%. This decrease is due to reductions in emissions of ammonia and nitrogen oxides in Denmark and throughout the rest of the European countries.

Introduction

During the last decades the air quality and the deposition of various air pollutants have been monitored in the Danish Background Air Quality Monitoring Program. The overall goal with the program is to determine the present state of the air quality in background areas in Denmark and to quantify the deposition of air pollutants to Danish marine waters and natural ecosystems. Moreover, it is the aim to measure long-term trends in concentrations and depositions in order to evaluate effects of national and international efforts to reduce air pollution. Finally, it is the aim to fulfil the national and international requirements for background monitoring in Denmark. The program is based on a combination of measurements at a network of stations and model calculations with both regional scale Eulerian models and local scale Gaussian plume models. The measurements are used for determination of the actual state of the air quality and deposition and the long-term trends. The model calculations are mainly used to estimate the depositions to larger geographical areas and the influence of the various sources of air pollutants and their origin. Moreover, local scale model calculations are also used for determination of nitrogen deposition to selected natural ecosystems.

The program covers eutrophying and acidifying air pollutants, heavy metals, ozone, and selected organic compounds as pesticides, PAH and nitrophenols. However, at present the main focus is on eutrophying nitrogen compounds originating from both national and international emissions of ammonia and nitrogen oxides. The deposition of these compounds has large impact. The air pollution originating from agricultural activities related to the large production of livestock in Denmark is of special interest. The annual Danish emission of ammonia was 80,000 tons nitrogen in 2003 (Illerup et al., 2005) corresponding to roughly 15 kgN per capita. Denmark is therefore one of the European countries with the largest emission of ammonia per capita. This paper presents some of the main results from monitoring of air pollution with nitrogen compounds during the last 17 years.

Methods

The network of measurement stations consist of 6 stations for measurements of the concentration of nitrogen gases and particles and 8 stations for measurements of the wet deposition of ammonium and nitrate (Figure 1). All the stations are situated in Danish background areas in order to obtain measurements representing the background levels for that area rather than the impact of a local strong source.

The diurnal sampling of nitrogen gasses and particles is carried out with the Danish filter pack sampler which is a slight modification of the filter pack sampler used in EMEP (EMEP, 2004). The filter pack

sampler is normally used for sampling of the sum of ammonia and particulate ammonium together with the sum of nitric acid and particulate nitrate (referred to as total nitrate). However, parallel measurements using denuders and filter packs have shown that the filter pack can give a reasonable separation between ammonia and particulate ammonium under Danish conditions. Hence the results are given separately for ammonia and particulate ammonium. The diurnal concentrations of nitrogen dioxide are measured using NO_x -monitors at two stations and a NO_2 -sampler based on sampling on potassium iodide coated glass filters on one station (EMEP, 2004).

The wet deposition of ammonium and nitrate is sampled on half-monthly basis using standard bulk samplers comparable to the samplers used within EMEP (EMEP, 2004). Double or triple sampling is used at all the stations in order to reduce uncertainties especially from contamination of the samples.

Aqueous extracts of the filters and the precipitation samples are analyzed for the nitrogen compounds using ion chromatography and automatic spectrophotometric techniques.

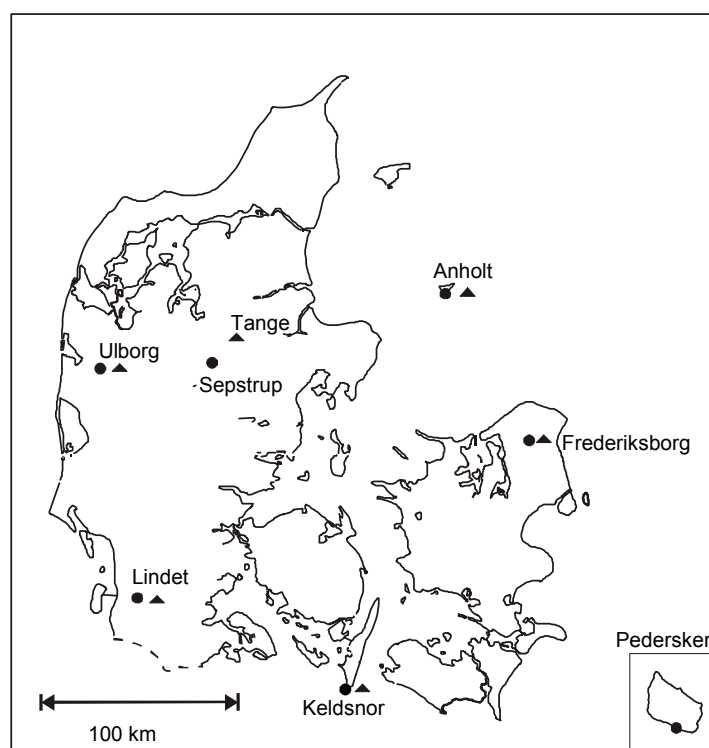


Figure 1. Measurement stations in the Danish Background Air Quality Monitoring Program used for measurements of nitrogen containing gasses and particles (▲) and wet deposition of ammonium and nitrate (●).

The regional scale model calculations of the nitrogen depositions are carried out with the Danish Eulerian Hemispheric Model (DEHM) which is a three dimensional Eulerian model covering the Northern Hemisphere (Christensen, 1977; Frohn, 2004). DEHM contains three nests with increasing resolution in order to make calculations for the Danish marine waters and land surfaces with a resolution of 16.67 km x 16.67 km. The meteorological input data are calculated using NERI's version of the meteorological mesoscale model MM5v2 (Grell et al., 1995). Emissions are a combination of several sources, where source and distribution in time and space is described by Hertel et al., (2002). The emission inventories for Europe are based on data from EMEP with a geographical resolution of 50 km x 50 km (EMEP, 2005). The annual total emissions for Danish sources are produced at NERI and the geographical distribution of the

emissions of NH₃ is based on an emission inventory with a geographical resolution of 1 km x 1 km (Illerup et al., 2005; Ambelas Skjøth et al., 2006).

NERI has also developed a local scale model OML-DEP for detailed calculations of the nitrogen deposition to selected natural ecosystems. The model is based on the Danish OML model, which is a Gaussian dispersion model (Olesen et al., 1992). This OML-DEP model can handle point and area sources and has been formulated for calculation of the dry deposition of ammonia using the Surface depletion principle (Horst, 1977). The model domain used in connection with the monitoring program is 16 km x 16 km with a geographical resolution of 400 m x 400 m. The model takes into account the medium to long-range transport of ammonia from the regional scale model DEHM. The local scale model uses a subset of meteorological input data generated by MM5v2 for use in the regional scale model. The local land use affects the deposition via associated turbulence and deposition velocity. The calculation on local scale is based on a special emission inventory for the Danish emissions of ammonia with a geographical resolution of 100 m x 100 m and division into area and point sources (Gyldenkærne, 2005).

Results and Discussion

The model calculations have shown that the nitrogen deposition to the Danish marine waters on average was about 10 kg N ha⁻¹ in 2004 (Figure 2). The deposition is highest in the southern part of the Danish marine waters due to areas with high emissions of nitrogen oxides and ammonia in the countries south of Denmark. The total input of nitrogen to the Danish marine waters is about 100,000 tons nitrogen per year. It has been calculated that the atmospheric depositions to the inner Danish waters (Kattegat and Belt Sea) contribute with up to one third of the total nitrogen load from surrounding land and atmosphere (Ærtebjerg et al., 2003). Hence the atmospheric deposition contributes significantly to the general supply of nitrogen to the inner Danish waters. The large supply of nitrogen has been one of the main reasons for the regular episodes with oxygen deficiencies and subsequent death of fish and benthic fauna, which have been observed in these waters during the last decades (Ærtebjerg et al., 2003).

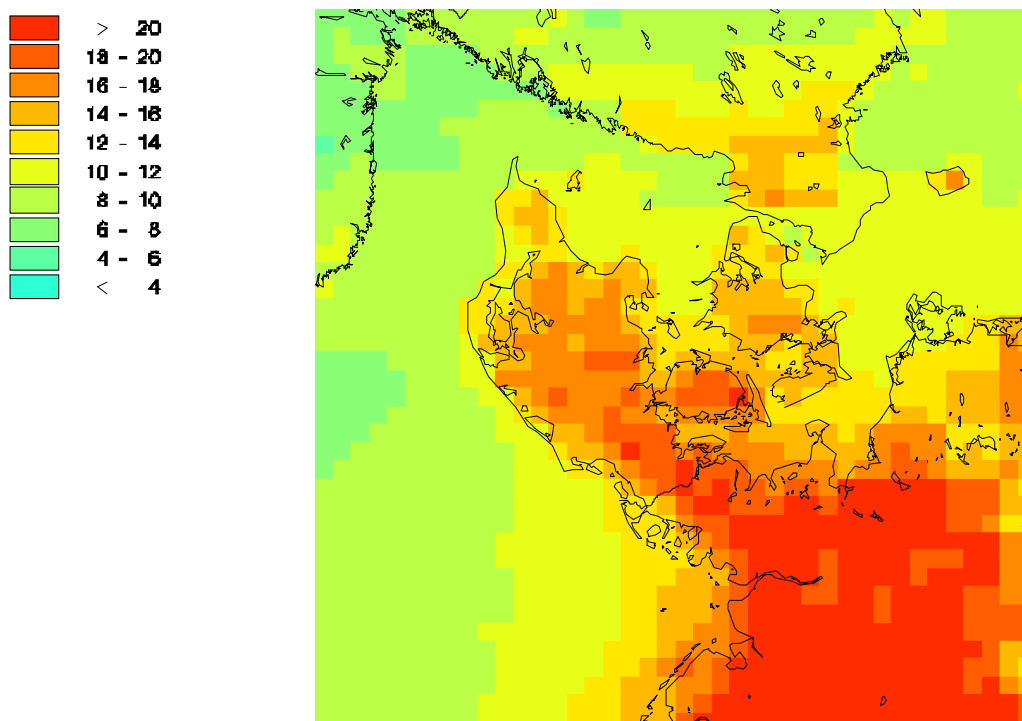


Figure 2. Nitrogen deposition in 2004 to Danish marine waters and land surface calculated using DEHM. The units are kg N ha⁻¹.

The model calculations have shown that the nitrogen deposition to the Danish land surfaces on average was about 16 kg N ha⁻¹ in 2004 (Figure 2). The highest depositions are seen in the south western part of

Denmark due to high density of animal husbandry, high precipitation and short distance to major emission areas in the countries south of Denmark. Hence the deposition is above the critical loads for many of the sensitive Danish ecosystems e.g. heath and bogs (Bak, 2001). This finding is in agreement with the observation of a decline of the coverage and quality of these ecosystems in Denmark (Bach et al., 2005).

As an example, Figure 3 shows the results from local scale model calculations of the deposition of ammonia to Randbøl Heath which is situated in the central part of Jutland (the peninsula in the western part of Denmark). The total deposition of nitrogen to the heath is 14-15 kg N ha⁻¹ of which 2-3 kg N ha⁻¹ is due to ammonia and 12 kg N ha⁻¹ is due to medium to long range transport of nitrogen containing compounds. The critical load for nitrogen deposition to heath in Denmark is 10-15 kg N ha⁻¹ (Bak, 2003). During the last 17 years there has been a significant decrease in the nitrogen deposition (Figure 7) and it is therefore most likely that the critical load has been exceeded for decades at Randbøl Heath.

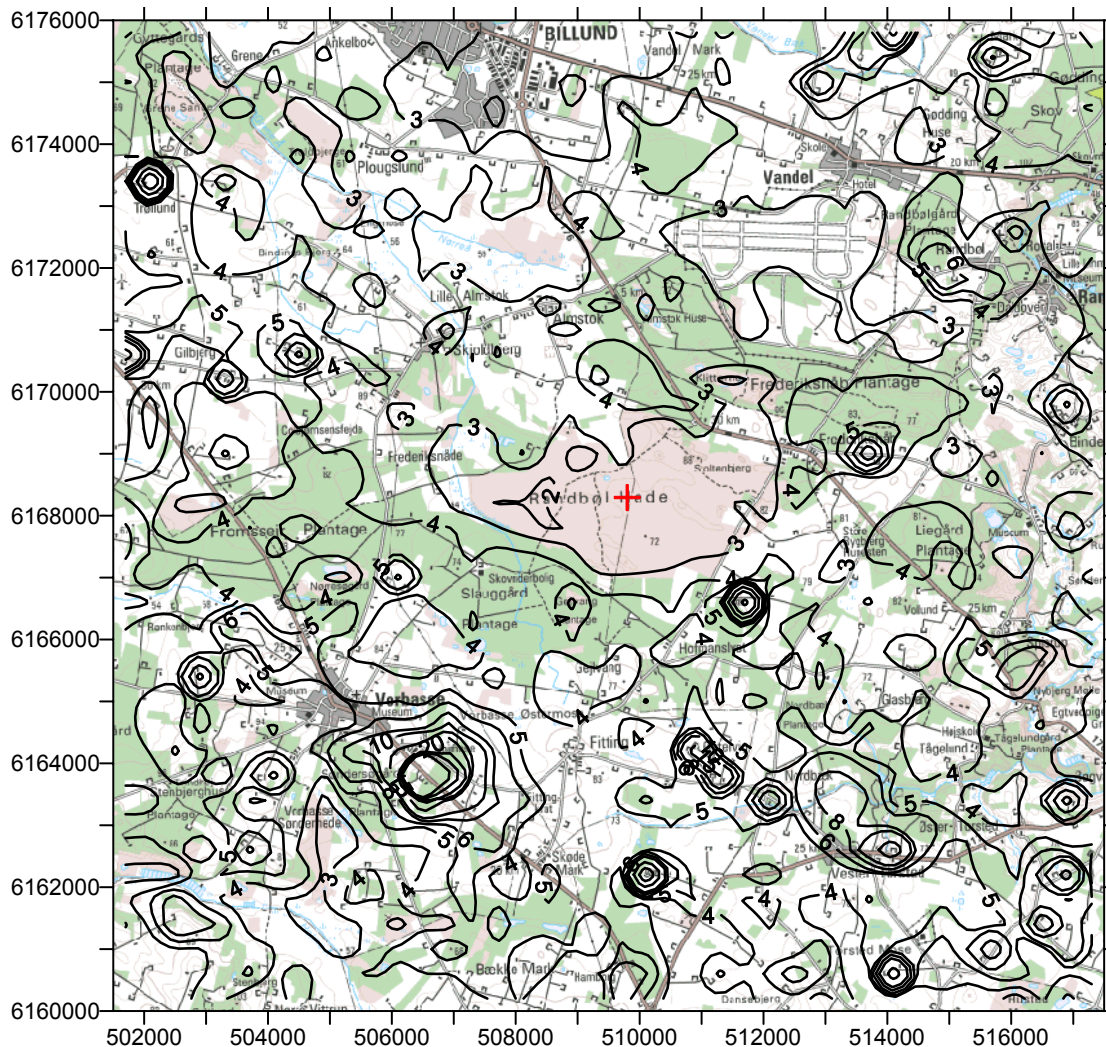


Figure 3. Model calculation of ammonia deposition to Randbøl Heath (cross in the middle). The calculations are carried out with the local scale model with a geographical resolution of 400 m x 400 m. The numbers on the axis corresponds to the UTM 32 coordinates in meters. The numbers on the contour lines corresponds to the ammonia deposition and is given in kg N ha⁻¹.

Recently the last fifty year's change in the vegetation at Randbøl Heath has been characterized on the basis of air photographs (Degn, 2006). This work has shown that the coverage with heath plants and other dwarf bushes has decreased from 90% in 1954 to less than 15% in 2005. Simultaneously the coverage with

nitrophilous grasses (*Molinia coerulea* and *Deschampsia flexuosa*) has increased and taken over the heath. The high atmospheric deposition of nitrogen at Randbøl Heath is believed to be one of the major reasons for this change (Degn, 2006).

Model calculations have shown that air pollution from Danish sources is responsible for about 45 % of the total nitrogen deposition in Denmark. Of this 40% comes from agriculture and only 5% from various combustion processes (Figure 4). The remaining parts (55%) comes from long range transport of reduced and oxidized nitrogen from agriculture and combustion sources in other countries; mainly from countries south and west of Denmark. It is therefore evident, that the problem with high nitrogen supply to Danish marine waters and natural ecosystems has to be solved on an international level and that agriculture in Denmark as well as the other European countries is the main source of atmospheric nitrogen deposition in Denmark.

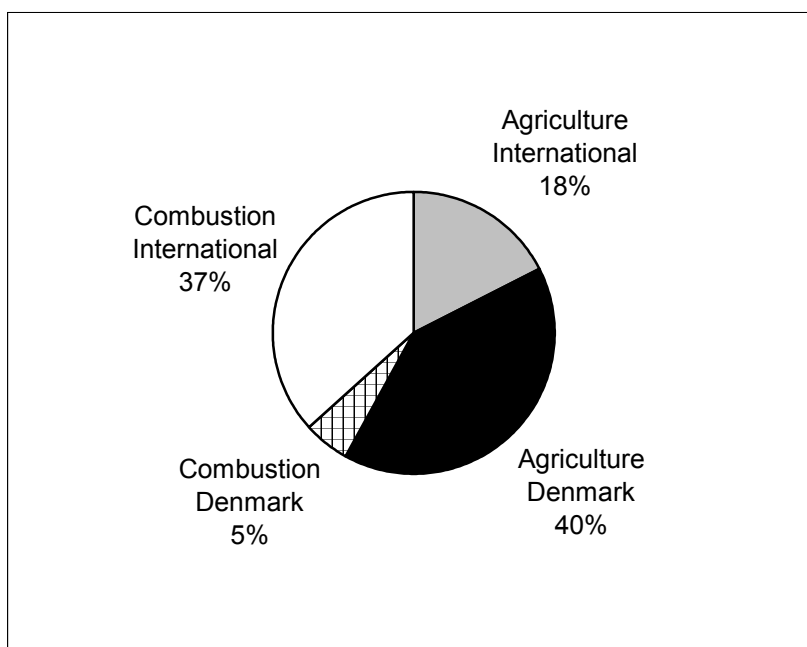


Figure 4. The contribution of Danish and international sources to the deposition of nitrogen in Denmark in 2004. The deposition is also divided in nitrogen due to agriculture and combustion processes.

The measurements have shown that the concentrations of particulate ammonium and total nitrate have decreased significantly at all the stations (Figure 5). The average decrease since 1989 is 42 and 29 % for particulate ammonium and total nitrate, respectively. The long-term trend for ammonia varies considerably from station to station due to varying influence from local sources. There is a tendency for a decrease in the long-term trend of the ammonia concentration at all six stations. However, the decrease is only significant at one of the stations. On average the decrease is about 20 % since 1989.

The observed pattern with a small decrease in primary ammonia and a large decrease in secondary particulate ammonium may be explained by the interaction with the atmospheric sulfur compounds. Since 1989 there has been a statistical significant decrease in sulfur dioxide (90 %) and particulate sulfate (60 %) in Denmark (Ellermann et al., 2005). As a consequence the atmospheric lifetime of ammonia may have increased since the chemical conversion from ammonia to particulate ammonium has been slowed down due to the decrease in the reaction with particulate sulfate. This can explain the observations of a small decrease in ammonia and a large decrease in particulate ammonium.

The wet deposition of both nitrate and ammonium has decreased since 1989 with 12 and 29 %, respectively (Figure 6). Averaged over all the stations the change is statistically significant for ammonium but not for nitrate. However, there is a tendency for a decrease in wet deposition of nitrate at all stations. This decrease

in wet deposition has been measured despite, that there has been an increase in the precipitation at five out of six stations.

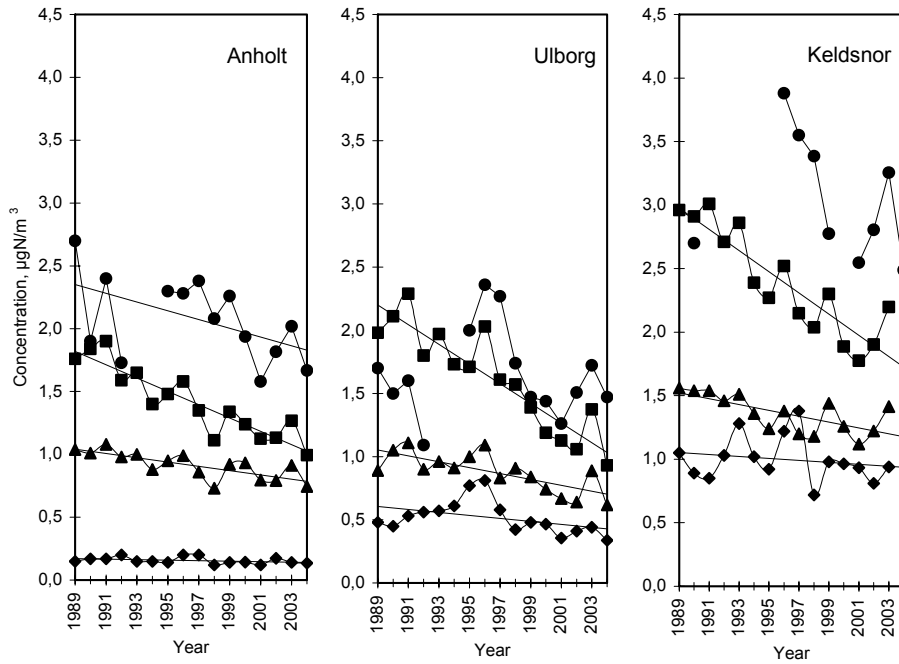


Figure 5. Long-term development in nitrogen concentrations at three of the Danish main stations: Anholt, Ulborg, and Keldsnor. () Nitrogen dioxide, () particulate ammonium, () sum of nitric acid + particulate nitrate, () ammonia.

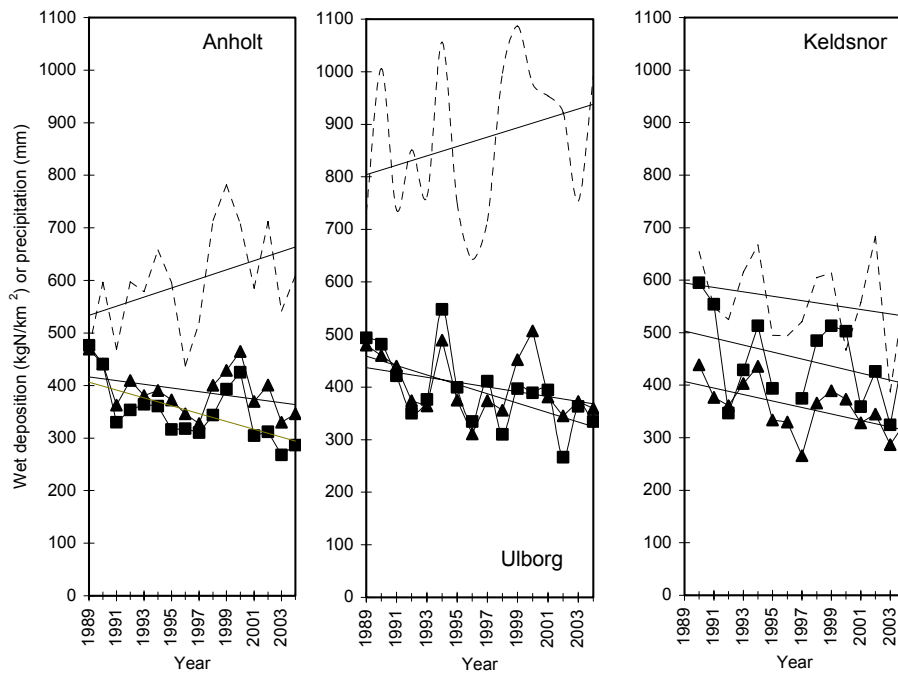


Figure 6. Long-term development in wet depositions of nitrogen at three of the Danish main stations: Anholt, Ulborg, and Keldsnor. Precipitation (dashed line), () ammonium, () ammonia.

nitrate.

The long-term trends in both concentrations and wet depositions of nitrogen can readily be explained by the reductions in the emissions due to international and national action plans (EU directive for National Emission Ceilings, Gothenburg Protocol, Danish Action Plans for the Aquatic environment etc.). In Figure 7 it is shown that the relative decrease in the nitrogen deposition in Denmark follows the relative decrease in the emission of nitrogen in the European Union and Denmark. Approximately half of the nitrogen depositions can be attributed to Danish sources and half to long range transport. It is therefore evident that the efforts in order to reduce emissions in Denmark as well as in the rest of the European countries have lead to significant changes in the nitrogen deposition.

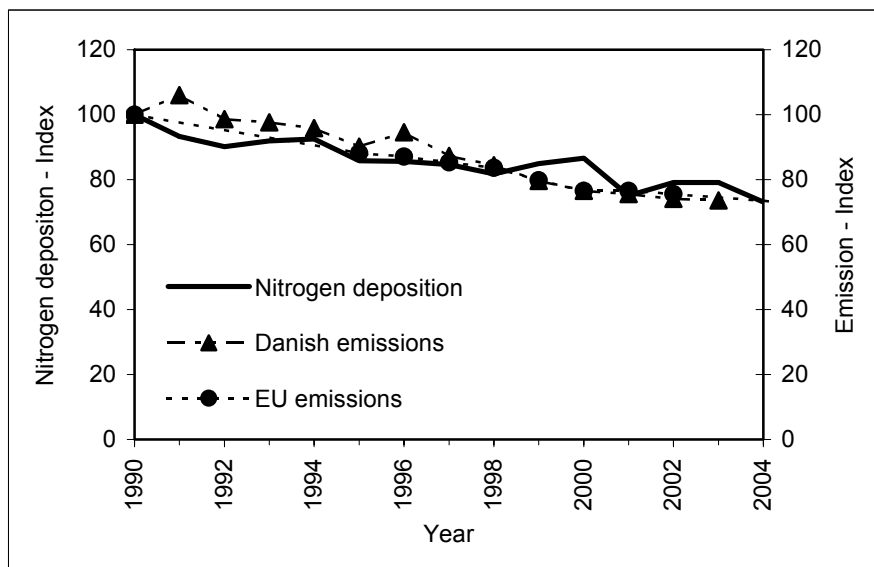


Figure 7. Comparison between the long-term trends in nitrogen deposition and emissions from Danish and European sources (EU). The trend in nitrogen deposition is calculated as the average for the six main monitoring stations. Deposition and emissions are given relative to 1990. (Illerup et al., 2005; EMEP, 2005)

Conclusions

Long-term monitoring of the air quality and depositions to Danish marine waters and natural ecosystems has documented that the nitrogen deposition in Denmark is high compared to critical loads for many of the sensitive Danish ecosystems, and the deposition contributes to the eutrophication of the Danish marine waters. Moreover, the monitoring program has shown that it is beneficial to use a combination of measurements at fixed stations and model calculations in order to get a better understanding of the observations and the processes determining the size of the nitrogen deposition in Denmark. Finally, the measured decrease in the long-term trend for the nitrogen deposition has shown that the work with reduction of emissions has lead to significant results. However, further reductions in the emissions may still be needed in order to protect the Danish nature and ensure high bio diversity in the country.

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Effects of Technologies for Reducing Ammonia Pollution from Agriculture: Outline of an Environmental-Economic Framework

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Abstract

The aim of the poster is to present a joint methodology for applying models and data for supporting policy decisions and advisory services with respect to handling ammonia emissions from livestock production and the depositions resulting from these emissions. The full chain of pressures and effects encompassing the link between production activities and welfare effects is subject to analysis enabling quantitative cost-benefit analysis. The possibilities for integrating spatial agricultural data with geographically targeted models of costs and benefits are discussed including links to models of emissions, transport, and localized loads and exposure. The components of the socio-economic analysis are discussed in terms of cost analysis and benefit transfer enabling CBA of site specific changes in air emissions and the resulting welfare effects. By developing and analyzing scenarios both reflecting differences in abatement technologies and localization of the livestock production, a range of costs and benefits can be explored for different abatement technologies that will assist policy makers in targeting policies. The poster describes the methodological framework and the current status of knowledge and identifies issues for further research.

Introduction

Agricultural livestock production and the resulting air emissions may cause welfare losses because of odour problems to neighbors and ammonia depositions to protected nature areas (EEA, 1995). Therefore, odour problems are addressed in the Third Aquatic Action Plan and ammonia emissions are subject to regulation internationally in the EU National Emission Ceilings Directive and Gothenburg protocol and in the National Ammonia Action Plan.

However, the scale of the problems is a result of a complex interaction between the size, location and production technology of livestock farms, emissions and transport, and the exposure of the recipients (urban settlement and protected nature areas). In Denmark agricultural husbandry production is intense in some regions, and local ammonia emissions add significantly to the deposition of transboundary nitrogen. Total nitrogen deposition varies between county averages of 17-24 kg N ha⁻¹ y⁻¹ but may be as high as 100-200 kg N ha⁻¹ y⁻¹ near large husbandry farms. The effects on odour problems are realized but the techniques for quantifying these are yet to be applied and validated in practical use. With respect to the effects on nutrient poor nature sites the deposition of nitrogen exceeds the critical loads of many nitrogen poor ecosystems. In 1999 the critical loads were projected to be exceeded for all Danish bogs and oligotrophic lakes (*Littorelletalia uniflorae*), 94% of coniferous forests, 63% of deciduous forests, 57% of dry grasslands, 42% of inland heath lands and 7% of coastal dune heaths (Bak et al., 1999).

One of the major policy questions with respect to nature protection in Denmark and northern Europe is the possibility for reducing ammonia loads. The complexity of the emission and deposition patterns and the dispersed location of nitrogen sensitive nature areas pose a demanding task for policy analysis. This is because significant contributions to loads derive both from local, national and international sources as well as the varying critical loads between different locations.

The reduction of eutrophication and acidification effects from air pollutants was included in the form of explicit targets in the recent impact assessment of the Thematic Strategy for Air Pollution by the EU Commission (EU Commission 2005). Given the coarse resolution of the RAINS model's grid cells (50*50 km) and the site-specific character of potential benefits from emission reductions, no attempt was made to quantify the benefits in monetary terms.

In order to support policy decisions and give recommendations to cost-efficient abatement strategies, this chain needs to be quantitatively modeled, encompassing the site-specific aspects of the problem. Therefore,

development of analytical tools linking economic models, emission models, and models of deposition and critical loads is needed to identify cost-efficient strategies for complying with the international agendas for protecting nature and biodiversity.

Methods

Agricultural Data

The site specific nature of both the emission sources and the recipients, *in casu* the nutrient poor ecosystems, implies that policy analysis needs to be site specific in order to identify cost-efficient strategies for reducing ammonia loads from agriculture. Therefore, the starting point of the analytical framework is a site-specific description of the agricultural production with emphasis on livestock production in terms of the location of stables, storage facilities for manure and the crop areas where manure is applied. This is done based on the in a GIS by using digitized maps of the location of agricultural field blocks based on the General Agricultural Register (GAR); and location of farm buildings and husbandry based on the General Husbandry Register (GHR). Both agricultural registers are geo-referenced and developed in order to control farmers eligibility to receive EU hectare and husbandry premiums. For each farm unit data on land use and livestock hold are recorded in the register, and the initial ammonia emission from each farm is estimated as part of the Danish emission inventories (Mikkelsen et al., 2005).

Cost Estimates and Emissions Reductions

Based on the geographically distributed agricultural data it is possible to identify technologies relevant for reducing ammonia emissions. The technologies are analyzed with respect to their financial economic costs and effects on ammonia emissions (NH₃-N). Effects on ammonia emissions are estimated based on practical tests, typically performed by the Danish extension center as part of the identification of Best Available Techniques (EU Commission, 2003; for a Danish reference see www.lr.dk). The costs are estimated in annualized values comprising both investments, maintenance and running costs and benefits in terms of increased nitrogen content in the manure and, subsequently reduced need for fertilizer input (formula 1).

$$AC_i = IC_i + MC_i + RC_i - NV_i$$

where

AC_i is the annual abatement costs of technology *i*,

IC_i is the annualized investment costs (interest and depreciations) of technology *i*,

MC_i is the maintenance cost of technology *i*,

RC_i in the annual running costs (input and labor) of technology *i*,

NV_i in the benefits in terms of increased nitrogen content in the manure of technology *i*.

In table 1 examples of abatement technologies derived from an ongoing analysis are given.

By relating the abatement technologies to the different types of livestock production it is possible to evaluate the consequences of different scenarios in terms of costs and reduces ammonia emissions. As the farm data are geo-referenced the scenarios can be developed to reflect both the type of livestock production, the location of the production and the various abatement technologies, thus, addressing a wide number of policy-relevant dimensions. One example of using the framework in policy analysis are reported on Schou et al. (2006) demonstrating the effect of establishing buffer zones to reduce ammonia loads to nature areas.

Table 1. Financial economic costs and emissions reductions resulting from abatement technologies in stables.

	DKK/kg N	DKK/AU ¹	NH ₃ -N red.
Acidification of manure from slaughter pigs	55	615	70%
Cooling of manure from slaughter pigs	69	688	40%
Chemical air cleaning in pig stables.	42	367	64%
Acidification of manure from dairy cows	119	433	50%
Drying of manure from hens	37	247	60%

1. AU: Animal Unit; One AU correspond to a yearly production of 100 kg N in manure.

Modeling of Deposition Patterns

However, in order to quantify the full chain of driving forces and effects related to agricultural ammonia emissions modelling of transport and deposition of ammonia needs to be included. At NERI a regional scale Eulerian model DEHM-REGINA is applied for calculating atmospheric background depositions of nitrogen to Danish nature. The local scale plume model OML-DEP is applied for computing nitrogen depositions from single farms (barns, storage tanks and evaporation from fields in connection with manure application) to the nearby area within a few km from the farm. These two models are operated as routine tools within the Danish background monitoring programme, and furthermore they are combined within the DAMOS (Danish Ammonia Modelling System) model system for assessment of total nitrogen loads in the vicinity of livestock farms (Hertel et al. 2006). The national Guideline for regulating ammonia from livestock farms in Denmark is currently under revision, and the future Guideline will be based on OML-DEP and DEHM-REGINA and the DAMOS system. Local ammonia emissions are calculated from Agricultural field blocks based on GAR; and location of farm buildings and livestock is based on GHR. Standard emission factors are obtained from a Danish norm system. A methodology for deriving a detailed seasonal variation in emissions has been developed and applied in the models (Ambelas Skjøth et al., 2004; Gyldenkerne et al., 2005). Application of the detailed seasonal variation has improved the model performance considerably. Model calculations have shown that typically a little more than 20% of the ammonia emission from local livestock farms is deposited within a radius of 2km from the farm.

Benefits of Reduced Deposition and Odour Problems

Knowing the changes in depositions and the exceedence in the critical loads for various nature types and the exposure of odour to individual households, the last step in a CBA is to estimate the monetary benefits from the changed effects. The empirical work on valuation of non-marketed goods does not enable valuation of such site-specific effects. Therefore research is needed both in terms of sampling of primary data to be used for estimating preferences for changes in biodiversity protection and changes in exposure to odour from livestock production, and in terms of developing benefit methods for generalized use of the results. One example related to odour problems could be undertaking a case study using stated preference and hedonic pricing methods for estimating neighbors' perceptions and valuation of odour problems from pig production. Results from the stated preference method in terms of willingness to pay for avoiding odour problems can be transferred to the case study. This is done by adjusting the primary results to the study area based on information about the socio-economic background of the population such as average income, percent of newcomers from urban areas, percent of residents with personal relations to animal farms, etc. The hedonic pricing results can be implemented in a regionally differentiated hedonic price model, which describe odour impact on house prices corrected for other housing and local characteristics. Together with information on house prices and house locations the hedonic price model allows the estimation of welfare losses from odour impacts for individual households in the different scenario settings.

Conclusions

The overall aim of this poster is to establish a joint framework for applying models and data for supporting policy decisions and advisory services with respect to reduce eutrophication of nitrogen poor nature types and handling odour problems from livestock production. The framework is designed to facilitate the use in practical policy analysis and will be demonstrated by analysis of costs and benefits of technology scenarios for abating odour problems from livestock production in a case study area. The system should be based on a GIS and couple spatial agricultural data with models of emissions, transport, and localized loads, and further, include socio-economic components enabling CBA of site specific changes in air emissions and the

resulting effects. Analysis should focus on developing and analyzing a range of scenarios both reflecting differences in abatement technologies and localization of the livestock production, a range of costs and benefits are explored which will assist policy makers in targeting policies to yield the most welfare improvements.

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Footprints on Ammonia Concentrations from Emission Regulations

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Abstract

Ambient ammonia concentrations are primarily related to agricultural activities. In this paper a simple ammonia emission model is presented and used for analyzing 15 years of ambient ammonia concentrations from Denmark. The emission model is used for explaining changes in observed concentrations as a result of changed agricultural practice enforced by national regulations.

Introduction

Ammonia is a waste problem in areas with large agricultural activity. The surplus of fertilizer associated with agricultural activities affects rivers, coastal waters, terrestrial ecosystems, and may e.g. cause an irreversible death of our forests (Nosenga, 2003). Effects of high nutrient loads are already seen in USA and Europe (Nosenga, 2003) and recently it has been estimated that 60% of the European ecosystems have exceedances of their critical level of nutrient load (Bak, 2001).

Reduction initiatives such as the Gothenburg protocol aims at reducing emission of air pollutants into the atmosphere. Depending on reduction methods and constraints laid on farming practice, changes may be observed in ambient concentrations as a result of the applied reduction strategies. Here we present and explain the air quality changes in Denmark related to Danish agriculture during a 15 year transition period from a period with almost no regulations to a period with a vast amount of regulations.

Two thirds of the Danish land areas are applied for agriculture, and the environmental problems associated with the surplus of fertilizer have been observed for decades. The surrounding coastal waters had already in the 1980s frequent algae blooms, followed by low oxygen levels at the bottom waters: In extreme cases causing a subsequent death of fish and benthic fauna (Conley et al., 2002, Ærtebjerg et al., 2003). As a consequence, three national aquatic action plans have been implemented in Denmark. The main purpose of these action plans is to reduce the load of (either input of ?) nitrogen to the marine waters, ground water, lakes and streams through the implementation of a large number of ammonia reduction methods (Grant et al. 2004). Regulations have gradually been introduced during the 1990s and have forced the Danish farmers to change their practice especially with regard to the handling of manure. This paper presents an ammonia emission parameterization for the different agricultural sources. The model will be used for explaining variations and trends in ambient ammonia concentrations in Denmark during 15 years. Finally the results are discussed and conclusions will be drawn.

Measurements

Diurnal concentrations of several atmospheric components including NH_3 have been measured during the period 1989-2003 and reported annually as part of the Danish national air quality monitoring program (e.g. Ellerman et al., 2003, 2004, 2005) initiated to follow the impact of the national action plans. Measurements of the ammonia concentration are carried out using the filter pack method as described by Ambelas Skjøth et al. (2004). Five monitoring stations are land-based and one is located in the middle of the Kattegat Sea (Ellermann et al., 2006).

The measured seasonal pattern of ammonia concentrations at the land based background stations has been changing during the entire measuring period. A change, which until now, has remained unexplained. Here we present the results from the station located at Tange (56° 21' N, 9° 36' E). The time series from the other land based stations show a similar seasonal pattern, although the annual trend is not as significant as seen in Tange.

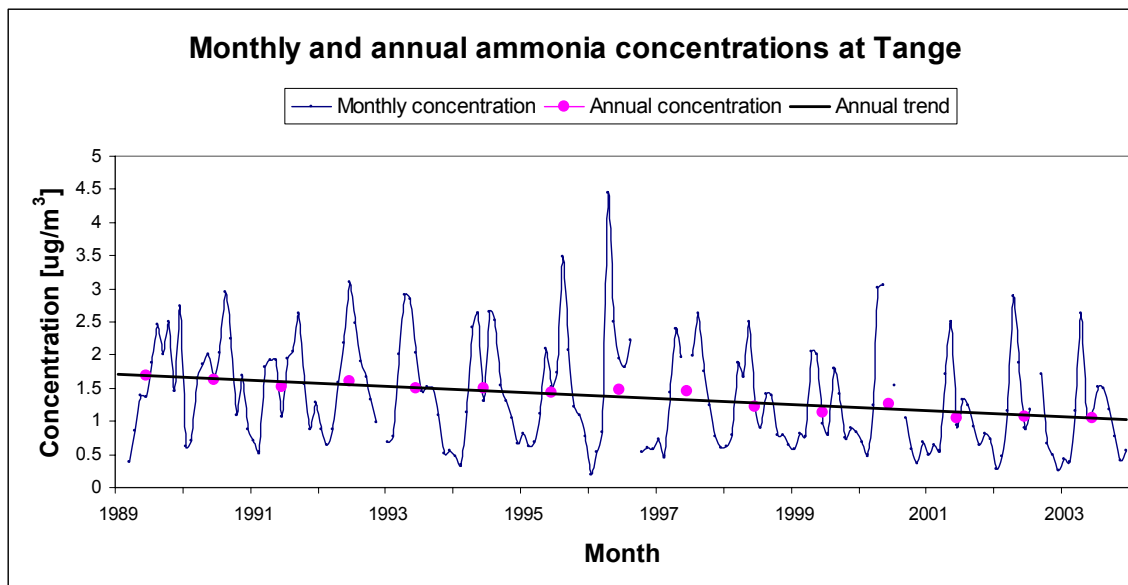


Figure 1. Monthly and annual ambient ammonia concentrations measured at Tange.

The measured seasonal pattern and the annual ambient mean concentration of ammonia has changed during the years 1989-2003. During the first years, the concentration is generally high during summer and moderate peaks are seen during spring and autumn. The last years the summer and autumn level has decreased while the spring level has increased. Furthermore, there is a decreasing trend in the annual ammonia concentrations, where the trend at the station near Tange is statistical significant on a 95 % confidence level.

Description of the Emission Model

The emission model has been developed with the requirements of on the one hand being relatively simple but on the other hand also be able to describe the main variations in ammonia emission during the year. Furthermore the parameterization should be able to handle changing agricultural production methods and finally it should be possible to implement the parameterization in local and regional scale models.

Therefore, to fulfil these demands, a parameterization based on several submodels have been developed (Gyldenkerne et al., 2005, Ambelas Skjøth et al., 2004). Each sub model handles a group of ammonia sources within the agriculture. These main groups can separated into buildings, grown crops, application of manure and mineral fertilizer, grazing animals and other sources. Figure 2 shows an example of the temporal variation of ammonia from these groups.

The model takes into account differences in volatilization of ammonia due to temperature differences. Therefore, the temporal variation of ammonia differs between different types of buildings (Figure 2 left). Furthermore, application of manure and artificial fertilizer is applied several times during the year (Figure 2 right), where application time depends on plant growth. Therefore, date of application changes between years and geographical location.

Model results

The emission model requires an overall annual emission inventory and local meteorological data. Data may be obtained from measurements or as in this case, results from the non-hydrostatic numerical weather prediction model MM5 (Dudhia, 1993), which is applied as meteorological driver for the hemispheric transport-chemistry models at NERI (Heidam et al., 2004).

In Denmark the geographical location of each agricultural building such as stables and storage and the number of animals associated to the location are stored in central databases. Furthermore, information about individual fields associated to the production facilities is also available. Each production facility has due to the legislation introduced in the early 1990s estimates of the input and output of nitrogen during

each of the stages in the production line. Therefore, it is possible to produce highly detailed annual ammonia emission inventories, which may be sorted into different sectors of the agricultural production (Ambelas Skjøth et al., 2004). A graphical example of these inventories is given in Figure 3.

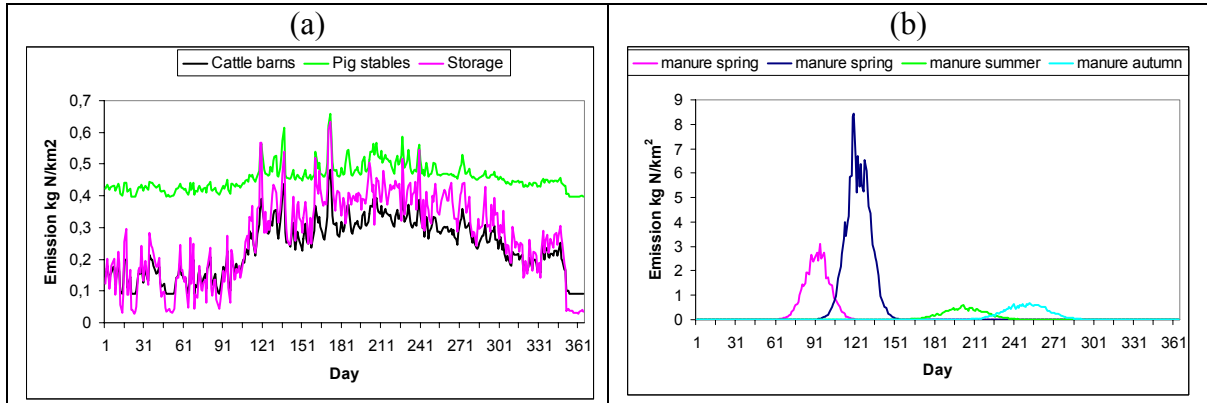


Figure 2. Temporal variation of NH₃ emission from different source groups in the NH₃ parameterization. Calculations are in [kg N/km²] for the Tange area. Emission calculations are from buildings and storage(a) and from field applications of manure(b).

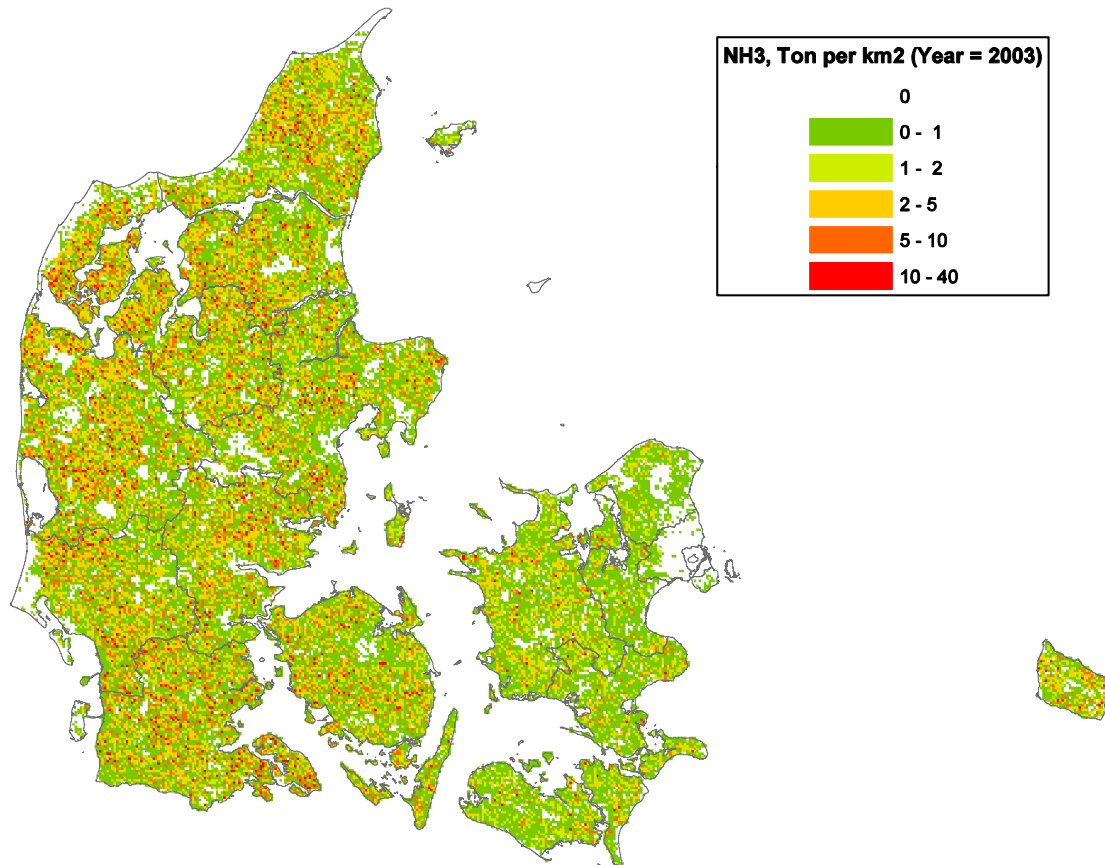


Figure 3. Geographical distribution of the annual NH₃ emission [Ton N/km²] from Danish agriculture distributed in a 1 km x 1km grid for the year 2003.

Figure 3 shows the geographical distribution of the total annual emission for the year 2003 on a 1 km x 1 km grid. Similar inventories have been made for 12 different agricultural categories for the years 1985 - 2003. The areas with high emissions marked by red spots are scattered throughout the country and the western part of the country generally has a higher emission density than the eastern part. The annual national emission for the different agricultural categories for the years 1985, 1990, 1995, 2000 and 2003 are presented in Table 1.

There is no central statistical information concerning application methods of manure. Therefore questionnaires among 15% of all Danish farmers are used as a basis for information about application methods in Denmark during the years. Use of mineral fertilizer at field level is based on a method described in Ambelas Skjøth et al., (2004), by combining the national consumption of mineral fertilizer within the agricultural industry, the plant need of fertilizer at field level subtracted by the applied amount fertilizer from husbandry, which again depends on the available manure at farm level.

The emission is distributed in categories (Table 1): emission from buildings, storages, grown crops, emission due to application of manure and mineral fertilizer. Remaining emission categories are grassing animals and straw treated with ammonia. The table shows, that especially the emission from application of manure during summer and autumn has decreased during the entire period in the range 90% or more.

Table 1. Annual ammonia emissions in Denmark, where the emissions are distributed into different sectors used by the emission model (Gyldenkærne et al. 2005).

Water Test	1985 [kTon N]	1990 [kTon N]	1995 [kTon N]	2000 [kTon N]	2003 [kTon N]
Pig houses	19.787	18.674	17.979	18.578	18.578
Cattle barns	10.926	10.614	9.104	9.598	9.598
Storage	13.936	12.298	11.125	10.176	9.136
Crops	13.174	13.010	11.125	11453	11.476
Manure early spring	6.722	6.042	6.120	5.941	4.330
Manure in crops	5.491	6.680	11.296	14.183	14.129
Manure summer	8.106	7.396	3.207	1.834	512
Manure autumn	6.232	4.826	1.806	1.114	640
Manure late autumn	13.256	10.306	4.156	1.429	640
Mineral fertilizer	7.870	8.662	7.606	5.582	4.437
Grassing animals	2.591	2.413	2.500	2.370	2.257
Ammonia treatment	6.551	10.210	6.647	2.471	661
Traffic	0.053	0.106	0.993	1.828	1.989
Total	114.695	111.236	94.196	86.556	80.391

Before the year 1997, there is no detailed statistical information on farm level with geographical coordinates. However, information on agricultural areas, animal type and animal number is available on municipality level for each year. These numbers has been used to redistribute the annual ammonia emission during the period 1985-1996. Therefore, inventories for the years 1985-1996 has lower spatial accuracy than later years. However, the lower quality only affects the emission model to a very small degree as the study area applies emissions from an area of 20 km x 20 km surrounding the measuring site (Gyldenkærne et al., 2005), which is roughly twice the size of an average Danish municipality.

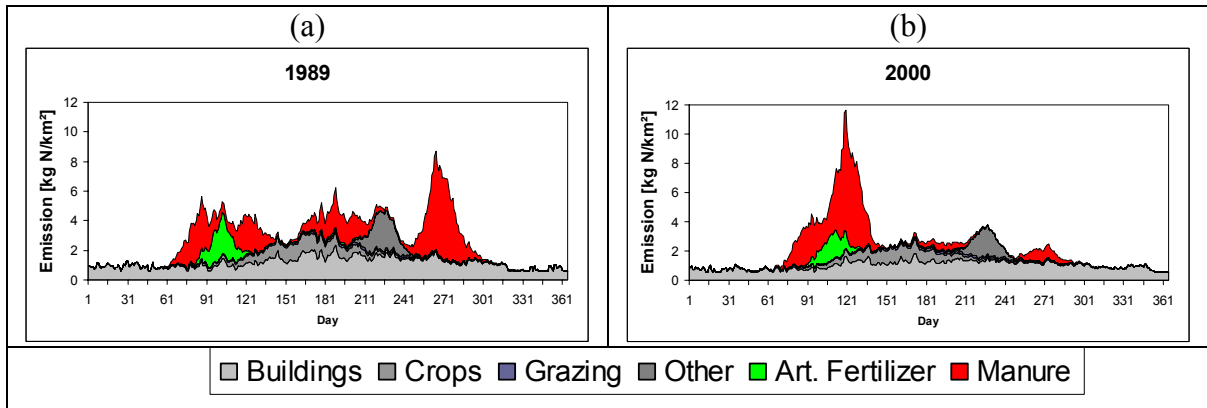


Figure 4. Calculated diurnal ammonia emission [kg N/ha²] from different agricultural sectors during the year 1990(a) and 2000 (b) for the Tange area.

Figure 4 shows an example of the calculated temporal development of the accumulated ammonia emission in the Tange area. Each color represents the sectors displayed in Table 1 generalized into 6 main groups: buildings, crops, grazing animals, other sources, artificial fertilizer and application of manure. In general the overall accumulated level has decreased from 1990 to 2003. Especially the emission resulting from the summer and autumn application of manure has decreased. Contrary the emission from the spring application has increased. Figure 5 shows the calculated relative change in monthly emission during the entire period 1989-2003 and the corresponding relative change in ammonia concentrations measured in the Tange area.

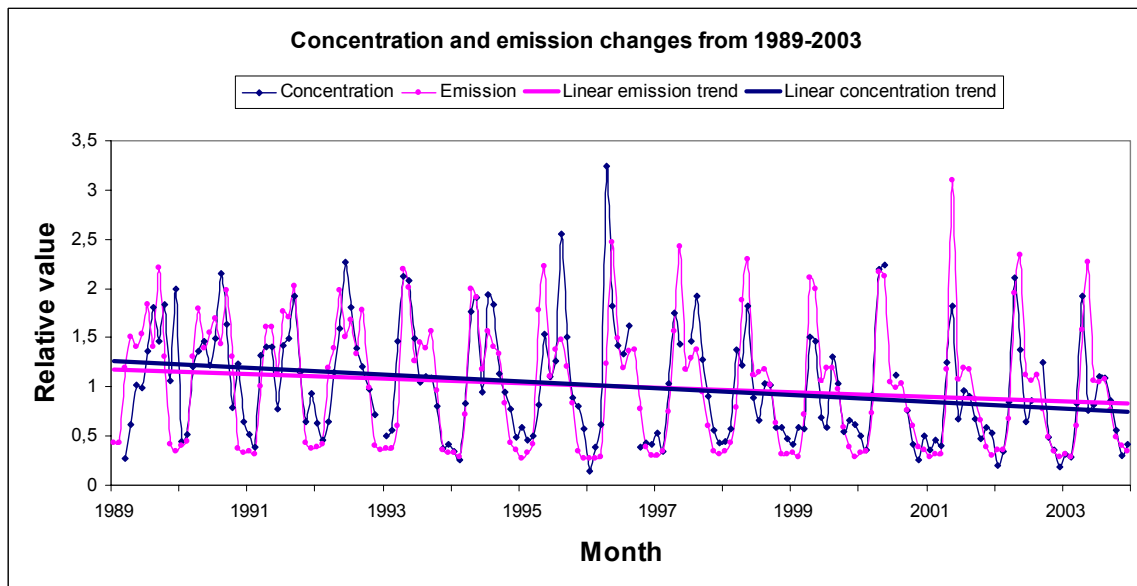


Figure 5. Calculated relative monthly ammonia emission (pink) and relative monthly ammonia concentrations (blue) for the Tange area during the period 1989-2003.

A linear trend for each time series have been applied in Figure 5. Both trends are decreasing and almost identical.

Discussion and Conclusion

A highly detailed NH₃ emission inventory based on information about the exact location of individual buildings and distribution of the number of animals within the Danish livestock has been produced for this study. The inventory has been applied by the emission model and diurnal emission calculations are performed at the measuring sites, where the results for the Tange station are discussed in detail.

For the year 1990 the emission model shows a relatively high emission level from spring to autumn (Figure 4). In 2003 several sources have been reduced significantly, where especially the contribution from manure application in the summer and autumn has been reduced (Figure 4). This decrease occurred gradually during the period 1985-2003 (Table 1). During the same period the emission from manure applied in crops during late spring have increased by more than a factor of two.

On a monthly basis a gradual change in the observed seasonal pattern of ammonia concentrations is seen (Figure 5). This change in pattern is also reflected in the results from the emission model. During the period 1989-1995 emissions as well as concentrations have shown relatively large peaks in spring and autumn. After 1995 the autumn top decreases and the spring top increases in both modeled emissions and concentration measurements.

The change in seasonal pattern (Figure 5) is a result of the Danish regulations aiming at reducing the nitrogen loss, which have forced the farmers to change general practice within the entire chain of food production. One important result is that the annual amount of nitrogen lost to the atmosphere has decreased by about one-third (Table 1 and Figure 1). Another important result is that an increasing fraction of the manure is applied during spring (Table 1 and Figure 5). Within the atmosphere, a direct consequence of the legislations is a change in the seasonal variation and a decreasing overall annual level (Figure 5).

A direct consequence of the Danish legislation is that storage requirements have increased at the same time as special demands for manure application have been enforced. Furthermore, the number of Danish farms are decreasing thus increasing the average farm size due to merging of farms. The larger farms and storage capacities decrease the number of point sources throughout the country, but increase the emission from the remaining point sources. This change increases the NH₃ gradients around the point sources and the environmental problems around these new larger ammonia hot spots. Thus care must be taken in the local regulation of the farms. This is currently being implemented with a new set of tools in the regulating of the agricultural production in Denmark. The core in the new tools is the Danish Ammonia Modeling System (Hertel et. al., 2006), which is a combination of several state-of-the-art air pollution models handling nitrogen and ammonia on local as well as regional scale.

A numerical study by Gilliland et. al (2003), showed that changing the temporal variation in NH₃ emissions over the USA changes where and when the deposition of nitrogen compounds occurs; Probably due to a seasonal differences in prevailing winds and chemical composition of the atmosphere. Therefore, applying a seasonal emission variations representing actual emission patterns probably redistributes the large scale nitrogen deposition. Therefore careful model scenarios must accompany emission reduction initiatives: These scenarios must use state-of-the-art numerical air pollution models with advanced chemical schemes and deposition modules like the EMEP (Touvinen et al., 2004) model or DEHM-REGINA (Frohn et al., 2002) model including the expected variations in the amount and pattern of emitted NH₃. However, the model results and measurements presented here also show, that applying significant reductions of ammonia emissions as seen in Denmark will have an immediate a similar effect on the ambient concentrations of ammonia.

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Atmospheric Emissions from a Deadstock Cremation Unit

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Abstract

The disposal of normal on-farm mortalities or deadstock is receiving more attention by both farmers and regulators alike due to issues of biosecurity and fear of recycling prion related diseases. One method for deadstock disposal is the use of small on-farm cremation units. This method is currently under review in Ontario and, as part of the review process, the environmental performance of the technology needs to be verified. As such, a commercially available, on-farm deadstock cremation unit was stack tested for various air emissions at the Arkell Research Station at the University of Guelph in September of 2005. The unit consisted of a primary chamber with a maximum rated capacity of 386 kg (850 lbs) and a secondary chamber located in line with the stack with a 0.3 second retention time. Diesel consumption for the primary and secondary burners is rated at 11.4 litres/hr (3.0 gallons/hr) and 2.5 litres/hr (0.65 gallons/hr), respectively. The stack testing protocol was designed, using established stack testing methodologies, to sample for combustion gases (O₂, CO, SO₂, and NO_x), particulate matter, acid gases, speciated metals, speciated volatile organic compounds, and speciated semi-volatile organic compounds (dioxins and furans, PAHs, PCBs, chlorobenzenes, etc.). To ascertain the level of variability in the data, tests were conducted using two different animal species (swine and poultry) with triplicate tests conducted for each animal species. This resulted in a total of six separate cremation runs being characterized for this specific unit. Results from this unit are compared to similar results from two other deadstock cremation units tested with the same protocol in 2003.



Air Quality and Emission Factors in Turkey Production

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Abstract

Airborne contaminants are factors of air quality in and emissions from the stables. Concentration of gases and particulate matter may influence health and welfare of the individuals living or working in these structures. Emissions may lead to environmental impacts. Particularly the emissions of ammonia and PM fractions must be considered in national and international emission inventories. For this purpose emission factors and activity data, for example the animal numbers, must be known.

Inside buildings, air quality is determined by monitoring the concentration of ammonia, carbon dioxide, total dust and PM10 with on-line techniques.

Emissions are investigated by measuring concentration of the contaminants and flow rates in the exhaust of forced ventilated stables. Dust concentration is measured with the gravimetric method. Fractions are calculated from an additional particle size analysis using a pre-separator, light diffraction and light scattering techniques.

The still running study is carried out in naturally and forced ventilated turkey houses. No influence of birds age, but of season and lighting procedure could be observed. In the average, concentrations remain below limit values. Total dust ranges below 4 mg/m³, PM10 below 1 mg/m³. Ammonia concentration normally not exceed 20 ppm.

Emissions factors depend on the age of the birds and the kind of litter. Aim of the project is to give answers to the questions of animal health and welfare, farmers health and welfare and load of the environment.

Introduction

Turkey meat becomes more and more popular and therefore consumption and production is steadily increasing. Mainly by economic reasons naturally ventilated stables are favoured in Germany but forced ventilated stables also exist. In some regions with high animal densities higher environmental loads are prevented using exhaust air cleaning techniques for forced ventilation.

In general airborne contaminants are factors of air quality in stables and environmental load by emissions from the stables. Concentration of gases and particulate matter may influence health and welfare of the individuals living or working in these structures. Emitting mass flows may lead to environmental impacts. Particularly the emissions of ammonia and PM fractions must be considered in national and international emission inventories (UN ECE 1979, 1999). Emission factors, which are required for inventories, are related to the number of animal places. Total national emissions will be calculated, compared with national ceilings and a necessity of reduction derived.

This paper gives methods and results of comprehensive investigations of the air quality in a naturally and forced ventilated turkey barn. Concentration of ammonia, carbon dioxide and total dust were measured with on-line techniques with respect to different periods of time over several periods of fattening. Dust was measured as total and PM10 fraction on-line mainly using a dust monitor. In some cases gravimetric samples were taken.

Emission measurements were carried out in the exhaust flow of the forced ventilated stables. Air flow was detected by the climate control computer which was calibrated by a grid measurement using a hot wire anemometer. Dust concentration was measured with the gravimetric method and isokinetic sampling. Fractions like PM10 could be calculated from the complete particle size distributions. Main parameters of this still running study are birds age, ambient climate conditions and different kinds of litter.

Methods and Materials

Stables

The investigations were carried out on farm level in a naturally ventilated cock house with respect to air quality inside only, figure 1, and in forced ventilated barns which belong to the experimental set-up of the FAL considering the comprehensive task.



Figure 1. View into the naturally ventilated barn (Hinz et al. 2004)

The experimental set-up covers 12 single compartments as described in a former study (Hinz et al., 1999). Always two compartments form one unit with respect to the ventilation system. While in the previous study environmental enrichments for the birds were in the focus, in the present study different types of litter are investigated. A direct comparison of the results from both houses is difficult because the stocking densities were similar but not the stocking numbers.

Measuring Equipment

Concentration of NH_3 and CO_2 were measured using an opto-acoustic multi-gas monitor at one central position in each stable. Contrary to dust there are no special requirements for measuring gas concentrations in the exhaust air flow. The probes were taken in a middle area of the ducts. Flow rate was monitored by the climate control computer.

Two targets are to distinguish measuring dust and PM concentration: air quality inside with respect to human and animal health and welfare and emission flow through the outlet ducts with respect to the environment. Measurements must consider particle size fractions. Definitions e.g. from CEN, ISO and US-

EPA classify different particle fractions. They depend on effects and targets from total dust down to the ultra fines PM_{0.1}, which were found to be hazardous to individuals and are part of global transport.

Human health-related particle size fractions according to ISO 7708 (ISO 1995) are the thoracic and the respirable (risk group) fractions. Concerning ambient air, US-EPA defined PM₁₀ and PM_{2.5} (US-EPA 1987). Curves following the different definitions are given in figure 2.

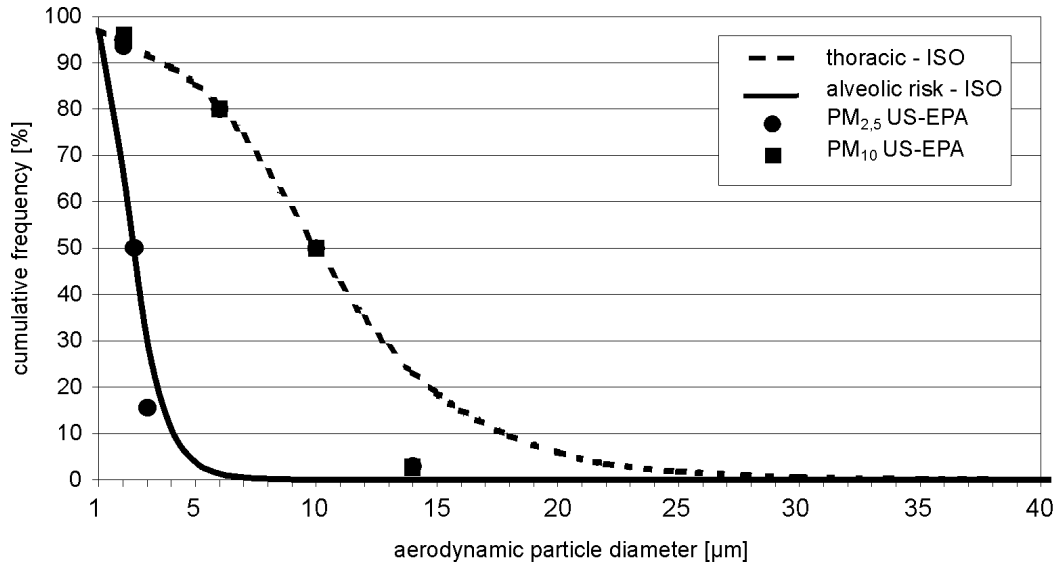


Figure 2. Fractions of PM acc. to EN ISO 7708 and US-EPA

The curves describe virtual particle separators simulating the corresponding parts of the breathing tract. They are characterized by their shape and by the 50 % value of separation and penetration. That means e.g. that 50 % of all particles with an aerodynamic diameter of 2.5 µm are alveolar while 50 % will be separated in the upper parts. The index numbers 10 and 2.5 in the US-EPA definitions mean the cut-off diameter of particle separators that the total suspended particulates (TSP) have to pass. For this particle size the separation efficiency is 50 %. In this respect PM_{2.5} and PM₁₀ are comparable with the respirable (risk group) respectively the thoracic fraction but the different conventions show differences in the ranges of particle diameters concerned. Thoracic considers diameters up to 40 µm but PM₁₀ ends at 15 µm, figure 2. PM_{2.5} rejects 94 % of 3 µm particles, 50 % of 2.5 µm particles and 16 % of 2 µm particles. Samplers with same cut off diameters but different shaped penetration curves will collect different fractions of PM.

TSP in ambient air means particles with settling velocities below 0.1 m/s respectively aerodynamic diameters smaller than 57 µm.

Total dust related to emission flows (often also used as TSP) may contain all sizes of dust particles up to a limit of 500 µm.

To compare results of particle concentration measurements the used definitions must be given. The definitions above are only related to size without any consideration of substances and materials. Particle dispersions from agriculture and nature are composed by various matter of different origin. They can include inorganic matter e.g. from soil but also organic matter from plants or animals as well as dead and alive microorganism like germs, fungi, viruses, bacteria or parts of these e.g. endotoxins. The definition of these so called bio-aerosols doesn't reflect to particle size in a specific way and will be not taken into account with the following.

Depending on the definitions and regulations particle size sensitive measurements are required. Table 1 gives possible ways for this purpose.

Table 1. Particle size-selective measurements

Aerodynamic separation	Conventional filter weighing
	Online electronic balance
	Online optical detection
Sampling total dust	Online size selective optical detection
	Conventional filter weighing with additional particle size analysis

Measurements inside the buildings followed the conditions at working site, that means all samples were collected with a sampling velocity of 1.25 m/s through a circular slot, independent from the following method of mass detection.

Inside a building the conventional gravimetric filter procedure was used as reference of total dust. The sampler was running in a high volume mode. Glass fibre filters used were pre-conditioned in a climate chamber at $20^{\circ} \pm 5^{\circ}\text{C}$ and $50\% \pm 5\%$ relative humidity for 24 hours before each weighing.

The TEOM instrument is mainly used for on-line monitoring of total dust. Later on a pre-separator is installed to monitor directly PM10 with a PM10 pre-separator. The heart of the TEOM is its tapered element oscillating micro balance. Particles are collected and weighed in a cumulative way. With the differences of weight in certain steps of time actual data of concentration are calculated. Averaging time depends on level of concentration. This conservative mass-related principle gives reliable results, but it must be considered that the element is heated up to 50°C . This influences the results if particles contain volatiles like VOC.

The Grimm scattered light monitor (SLM) detects the light impulses scattered by each single particle crossing the measuring volume. The instrument discriminates and counts all signals and calculates all wanted fractions from this raw data using new or previous definitions. Light intensity depends on the size, structure, the refractive number of a particle and the wavelength of the optical source. Fig. 3 gives scattered light intensity versus particle size for a constant wavelength λ but two different refractive numbers n_1 and n_2 .

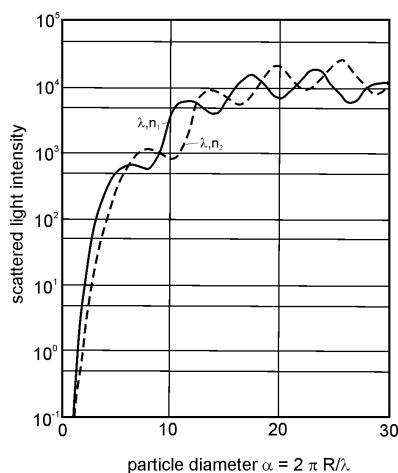


Figure 3. Scattered light intensity versus particle size for refractive numbers n_1 and n_2 .

For this reason a calibration of the opto-electronic device is a mandatory regulation using just that material which will be measured. Otherwise an optical equivalent diameter can be given only. Fig. 3 shows this as principle limitation of such instruments for larger particles depending on n (but also on λ) no clear correlation between size and light intensity exists caused by periodical fluctuations. Most of those instruments count the number of large particles in an open upper class. With an increasing number, mass or volume information without correction becomes faulty. Hence follows that no information about TSP is available for size distributions with a high content of larger particles.

The major advantage of the instrument is its simple handling to get very quick on-line information about the influence of location or process parameters and, as mentioned above, the possible consideration of nearly all particle size definitions.

Determination of emission flows requires simultaneous measurements of air flow rate and contaminants concentration. To ensure representative samples and to avoid changes in particle size distribution by the sampling procedure itself, the so called isokinetic condition must be kept. That means equal velocities in the duct and the probe. In figure 4 the scheme of particle emission measurements in the ducts of selected compartments of the turkey house is given.

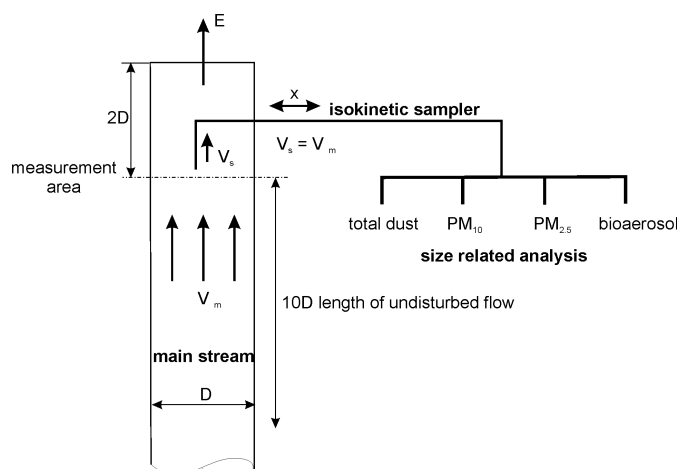


Figure 4. Scheme to measure PM emissions

A high-volume sampler with a pre-separator was used. Its fractional efficiency follows the previous fine dust definition according to the convention of Johannesburg with a cut-off diameter of $5 \mu\text{m}$ and a penetration limit for particles larger than $7.07 \mu\text{m}$.

To get knowledge about the content of the finer particle a scattering light monitor was installed in the flow behind the cyclone separator. To determine the complete size distribution up to the largest particles the separated coarse fraction was analysed e.g. with light diffraction methods. The complete size distribution is required if PM10 or PM2.5 shall be given as percentage from total dust (TSP) as often done in the past when creating emission inventories.

Air flow was monitored as mentioned above for ammonia emissions.

Results and Discussion

Results will be presented separately for air quality inside and the emissions.

Inside the buildings, naturally and forced ventilated, for all investigated contaminants influences of the season and courses of a day could be observed. Generally the concentration is higher in winter than in summer whereby more than 90 % ranged below 20 ppm for ammonia and 4 mg/m^3 for total dust. Differences were observed in the daily courses. With quite constant values over the day ammonia showed

its maximum at 6am, while PM concentrations (TSP, PM10) had two peaks, one in the evening and an other in the morning. This resulted from a higher activity of the birds at that times. Concentrations were nearly constant for weekly averages. No dependency on birds age was found in the naturally ventilated barn.

Emissions were measured from the forced ventilated stable only. In the running study the birds are standing on different kinds of litter which may influence birds' health and behaviour, but also resulting emission rates.

Figure 5 shows emissions from the compartments with chopped straw and wood shaves.

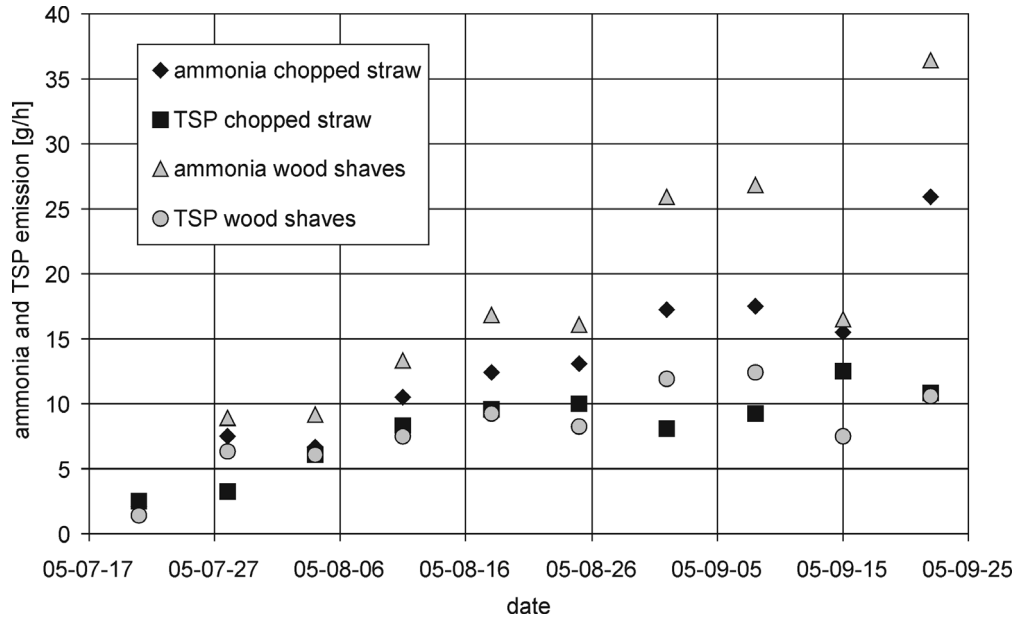


Figure 5. NH₃ and TSP emissions from turkey barns littered with chopped straw or wood shaves

An increase of emissions with birds age can be observed, but more distinct for the chopped straw and ammonia. The results reflect to animal places because the number of birds in the compartment was taken to be constant neglecting the losses. The magnitude of emissions factors and their ranges are given in table2.

Table 2. Emission factors for NH₃ and TSP

Contaminant	Emission factor [kg animal ⁻¹ a ⁻¹]
NH ₃	0.08 – 1.125
TSP	0.05 – 0.69
PM10	0.0125 – 0.17

These values are calculated on the base of 1h- averages measured in certain steps of a fattening period. They must be seen as first estimations.

Particle emissions are related to the separated fraction of the cyclone which was measured gravimetrically. This coarse fraction represented the total mass up more than 90 %. Particle size distribution of this coarse fraction showed only a small content of PM10, given for one example in figure 6. 50 % of total mass result from particles larger than 53 μm .

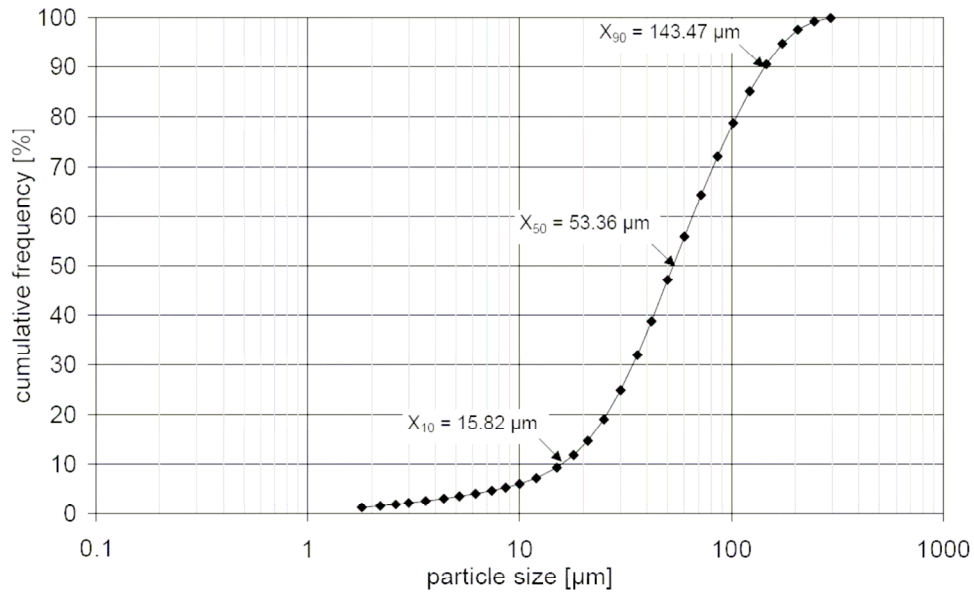


Figure 6. Particle size distribution of the TSP forming coarse fraction from the cyclone pre-separator.

Conclusions

Investigations were carried out in a naturally ventilated turkey house on farm level and in separated compartments of an experimental set-up with forced ventilation.

Concentrations of ammonia and PM were monitored inside the building with respect to farmers and animals' health and welfare.

Emissions were estimated by measuring airborne concentrations in and the air flow through the exhaust ducts of the mechanical ventilation system.

Total dust ranges below 4 mg/m³, PM10 below 1 mg/m³. Ammonia concentration normally do not exceed 20 ppm.

Emissions range between 0.08 kg animal⁻¹ a⁻¹ and 1.125 kg animal⁻¹ a⁻¹ for ammonia respectively from 0.0125 kg animal⁻¹ a⁻¹ to 0.17 kg animal⁻¹ a⁻¹ for PM10.

Particle size distribution was measured in the range of 0.5 μm up to some hundred μm particles.

Considering the conditions of the turkey houses there was no remarkable impact to farmers and animals health and welfare as well as the environment.

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Integrating Lidar and Atmospheric Boundary Layer Measurements to Determine Fluxes and Dynamics of Particulate Emissions from an Agriculture Facility

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Abstract

Lidar technology offers the ability to quantify concentrations of small particulates in the atmosphere in certain ranges of time and space. While this is a valuable tool to visualize the behavior of plumes emitted from the surface, the actual flux of particles cannot be estimated from such data alone. To determine the mass flux of particles, the concentrations must be properly integrated with wind and turbulence properties.

The goal of this study is to utilize a model that uses wind and particle density information to calculate the flux of particles from an animal facility near Ames, Iowa. The model is a simplified conservation equation for particle density in the atmosphere. This approach essentially quantifies fluxes in and out of a box centered over the facility and estimates the surface source by assuming conservation of mass.

In addition, we hypothesize that distinct turbulence structures will sometimes interact with the intermittency of the surface emission from the buildings, resulting in episodic changes in emission fluxes from the site. A second objective involves documenting how intermittent the emission plumes are and how they are connected to periodic large scale turbulence events.

Lidar data of particle size and density in the vicinity of the site were collected during an intensive field campaign lasting nearly 2 weeks. In addition to the lidar data, turbulence data were measured at several levels on each of three towers, located upwind, inside and downwind of the source area.

The model requires measurements of the vertical profiles of both concentrations of particulates and the mean horizontal wind. The concentrations were measured using the lidar, while winds were measured using a combination of cup anemometers and sonic anemometers. This allows the emission fluxes to be calculated during 15 to 30 minute periods when winds are consistent.

Flux calculations await the final calibration of the lidar returns using measured particle densities. Flux estimates will be made when distinct plumes are observed under steady-state wind conditions.

Current results are presented showing evidence of episodic plumes of CO₂ in response to intermittent vertical motions of turbulences.

Introduction

Agricultural facilities and operations result in emission of various particles and gases to the atmosphere. Among these are small particles. The emissions of small particulates from such facilities need to be quantified in order to determine the effects of such operations on air quality. Unfortunately, the size and heterogeneous nature of such facilities makes it impossible to make any simple direct measurements of emission rates.

The emergence of lidar allows measurements to be made of the spatial distribution of particles including size and density. The USU lidar was developed exactly for this purpose and allows the spatial distribution of particle density to be quantified for the atmosphere in the vicinity of an agricultural facility. However, lidar measurements of particle density will not yield the actual rate of emission from the site. This requires an integration of particle density data from the lidar with wind and turbulence information.

Determining the surface flux of particulates in this way still poses a challenge. The animal facilities especially are quite heterogeneous and variable in size. The sources have a spatially complex pattern and have distinct effects on wind and turbulence. The typical micrometeorological approaches used to estimate surface fluxes are also problematic in these cases, since they are predicated on a uniform surface or source region of adequate spatial extent.

In this case we propose using a simplified form of the conservation equation for particle density of the air to estimate the emissions for a confined animal facility near Ames, Iowa. The simplified conservation equation can be written as:

$$\frac{\partial \overline{\rho_p}}{\partial t} + u \frac{\partial \overline{\rho_p}}{\partial x} = \frac{\partial}{\partial z} (\overline{w' \rho_p'}) \quad (1)$$

where ρ_p is the density of particles, u is the horizontal velocity, w is the vertical velocity, t is time, and x is distance along the mean horizontal wind direction. The primes in the last term denote the instantaneous deviation of the value from the temporal mean. The last term describes the change with height of the vertical flux of particulates. In simple terms, the equation states that changes of the vertical particle flux with height are related to lateral transport or horizontal advection of particles by the mean horizontal wind. The plumes from an individual facility will only in fact rise up to a finite distance, depending upon the source strength and intensity of the turbulence. At some height z_r , the average particle density will not be affected by the local source region being studied.

Noting the above, and assuming steady-state conditions, equation (1) can be integrated to yield:

$$\int_0^{z_r} u \frac{\partial \overline{\rho_p'}}{\partial x} dz = (\overline{w' \rho_p'})_{sfc} \quad (2)$$

We now have an expression relating the flux at the surface with the horizontal changes of particle density.

$$\text{Surface Flux} = \int_0^{z_r} \overline{u \frac{\partial \rho}{\partial x}} dz \quad (3)$$

Solving this equation requires knowledge of the vertical profiles of mean horizontal wind and particle density. Essentially, equation (3) expresses the surface flux as the difference between the flux into and out of the sides of a controlled volume. Equation (3) also calculates the surface source as the difference between the lateral fluxes in and out of the control volume. A similar approach was used to estimate water vapor fluxes from irrigated patches on the landscape by Hipps and Zehr (1995) and Prueger et al. (1996).

Methods

The experiment was conducted at an animal facility near Ames, Iowa. The study was conducted in late August and early September of 2005. The animals were confined in separate buildings spaced over the site. A 20-meter tower was located in the middle of the facility, and there were 7-meter towers both upwind and downwind of the site. Measurements were made at three heights on the large tower and two heights on the smaller towers. At each location, high frequency measurements of 3-dimensional wind, air temperature, water vapor density, and CO₂ density were made. All measurements were made at 10 Hz. The 3-D sonic anemometer (Model CSAT3 – CSI, Logan, UT) was used for measurements of wind and temperature, while the LiCor Model 7500 was used to measure water and CO₂ densities. In addition, a vertical array of cup anemometers was mounted on each 7-meter tower.

The lidar and other air quality measurements during this experiment are described elsewhere in these workshop proceedings. These measurements allow the actual particle density at certain size ranges to be computed for each spatial element mapped by the lidar.

The first step is to document periods when plumes were observed and the lidar was scanning in an appropriate pattern to measure the upwind and downwind faces of the control volume. The setup and a conceptual picture of this approach are depicted in Figure 1.

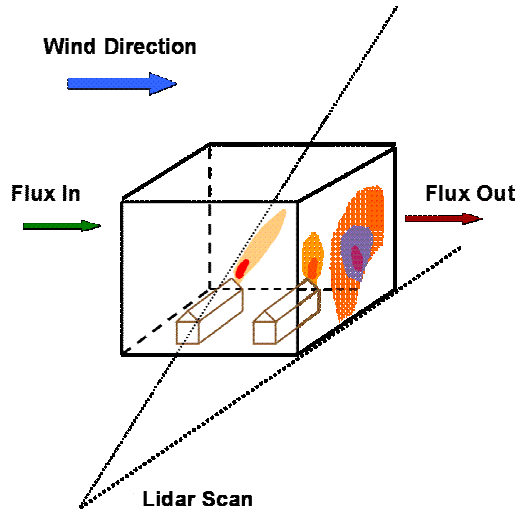


Figure 1. Representation of the approach used to calculate surface emissions.

When distinct plumes of emission were observed during fairly steady-state conditions, the wind and lidar data can be input into equation (3) to solve for the average surface emission. These calculations will be completed for all periods during the experiment that meet the above criteria.

Periods will be identified when distinct turbulence structures are present and related to fluctuations in plume activities. The intermittent nature of both the turbulence and emission will be conducted by examining the time series of turbulence data and plume behavior as measured by the lidar.

Preliminary Results

Currently the lidar data are being calibrated to measured particle concentrations to produce images with actual density units. These must be produced before the flux calculations can be made. However, the turbulence measurements of vertical wind and CO₂ density can be used to look at some of the intermittent behavior of CO₂ from the facility.

Some connections between episodic turbulence structures and emission can be observed by looking at the vertical wind and CO₂ density time series. Figure 2 illustrates a case in which there is a strong coherence between large updrafts of vertical wind and CO₂ density at the 12-meter level on the tower in the middle of the complex. There are about 15 minutes of data plotted.

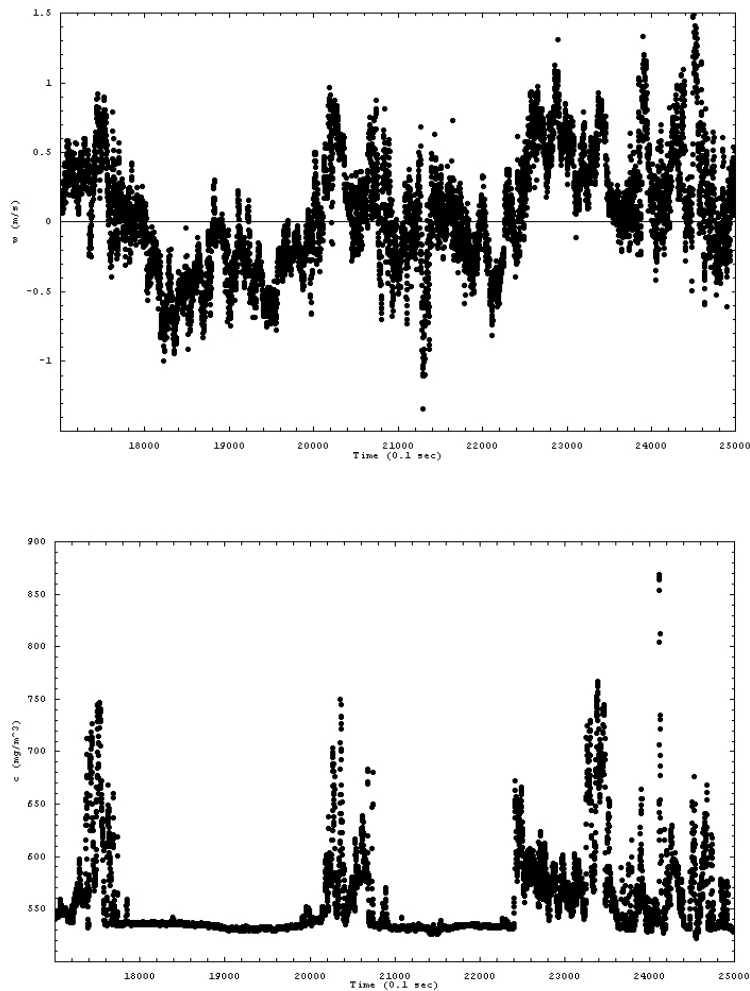


Figure 2. Time series of vertical velocity and CO₂ density at a height of 12 meters above sources.

The data are from a period in the morning when convection was just starting to couple the lowest surface layer with the air above. Note that the CO₂ remains near constant during periods of either weak vertical motion or downward vertical gusts. The source of the CO₂ is the buildings below, and under these conditions, it does not reach the sensor. However, periodically strong updrafts associated with passage of a large eddy bring up plumes of larger CO₂ density.

Future analyses will examine the coherence between plume emissions and transport with distinct turbulence events and structures. This will indicate how emissions are affected by periodic large scale eddies sweeping the source area. The implications for modeling the transport of particulates will be addressed.

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Quantifying Aerosol Mass Remotely from Real-Time Lidar Measurements

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Abstract

The purpose of this project was to develop a technique to measure remotely in real-time, aerosol mass emitted from agricultural operations. Lidar measurements of plumes emitted from field preparation and spray operations are used as examples of the technique. This paper demonstrates the integration of lidar images of aerosol plume cross sections to determine suspended concentrations. These integrations are combined with aerosol size distributions to determine the volume distribution of aerosols in the plume over time.

Introduction

Agricultural operations act as aerosol sources in a variety of ways. Two such sources are aerial spraying and field preparation (figure 1). To date, little is known about the dynamic concentrations, fluxes and transport distances of such aerosols (Holmén et al. 1998). Aerosol plume characteristics, such as concentration, are typically predicted via models such as AgDrift (Teske et al. 2002). Traditional methods of measuring non-point source aerosols are via samplers in the field (Clausnitzer and Singer 1996) or downwind (Hoffmann and Kirk 2005) which are only useful for small areas or spatially regular plumes. In an effort to overcome these limitations remote lidar measurements of agricultural plumes have been combined with traditional point sampling to measure the extent of plume movement and concentration across the plume area (Holmén et al. 2001a; Holmén et al. 2001b). This work presents a new calibration technique that allows the remote measurement of plume concentration via lidar. It shows results of the calibration from both an aerial spraying operation and a traditional cotton field preparation operation. Both studies took place near Las Cruces, New Mexico in the spring of 2005.



Figure 1. Agricultural aerosol sources. Top: Aerial spray operations for vector control, Bottom: Dust generation from field preparation

Methods

Measurements

Near dawn on April 27, 2005, the New Mexico State University Cessna T188C aircraft applied oil based tracer in five single-pass swaths to simulate vector control operations. Each pass of the aircraft was monitored remotely by the University of Connecticut elastic backscatter lidar. The lidar is capable of taking cross sectional measurements of the relative plume concentrations by slicing through the plume vertically.

Traditional farming practices for cotton were performed at the New Mexico State University Leyendecker Plant Science Research Center. The lidar was used to measure the particle concentration above the field as generated from the farming operations. In this study, the lidar took horizontal scans across the field at several different heights. This results in a three dimensional volume image of the plume. Figures 2 and 3 show a representative area and volume lidar images of the spray plume and dust plume respectively

Particle size distributions for the aerial spray were determined from wind tunnel calibrations of the spray nozzle configuration. Emission rates were the application rate for a unit length of aircraft movement. Particle size distributions of dust from the field operations were determined by sampling with cascade impactors and real-time mass monitors. Emission rate was determined from the maximum sampling measurement adjusted for unit length of tractor movement.

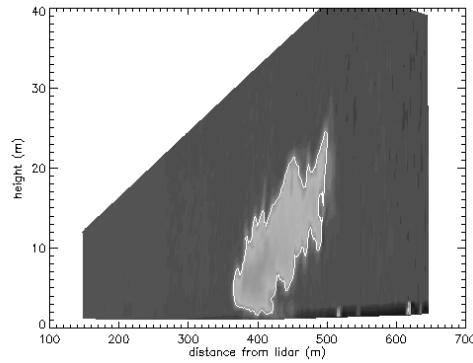


Figure 2: Representative cross sectional slice of aerial spray plume

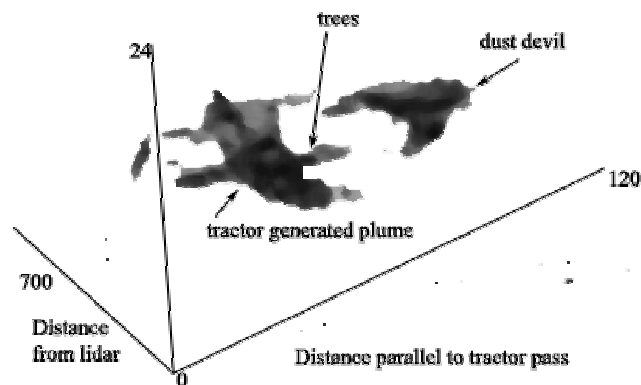


Figure 3: Lidar volume from one pass of the tractor. The reconstructed volume shows a line of trees behind the field, the dust generated from the tractor operation and a dust devil that occurred during the scan. Distance units are relative lidar units.

Determining Concentration

The technique to determine concentration consists of two steps. First the measured particle size distributions and emission rates of a test plume are used to calculate predicted plume concentration. Second, a conversion factor for lidar backscatter to concentration is found by finding the linear relationship between the predicted concentration and the total measured backscatter in the plume volume. This conversion factor is then used to convert the lidar measurements to concentrations across the plume. Figure 4 presents the relationship used to find the conversion factor for the airplane spray study. Figure 5 shows an example of the resulting spray plume concentration dynamics.

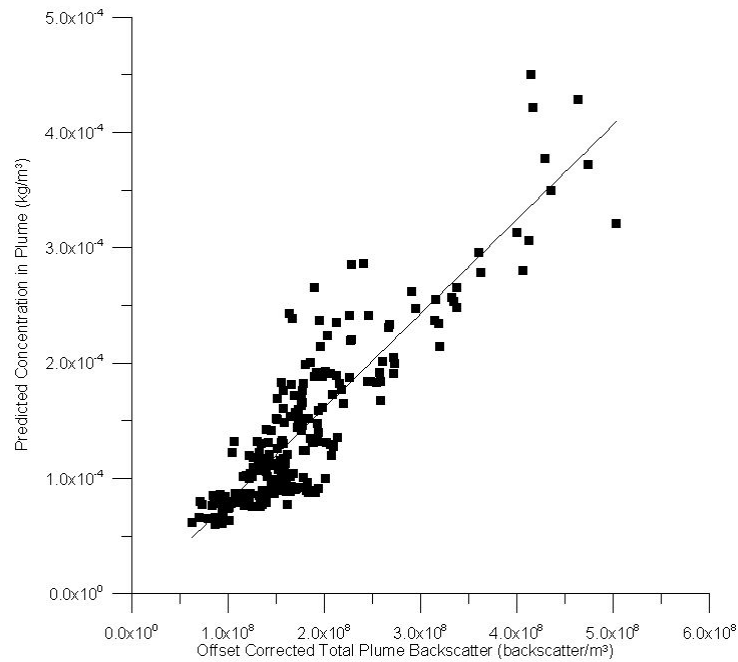


Figure 4: Predicted plume concentration vs. total lidar backscatter for three pass of the airplane. The linear relationship has a slope of 8.18e-13, which is used as a conversion factor between lidar backscatter and aerosol concentration.

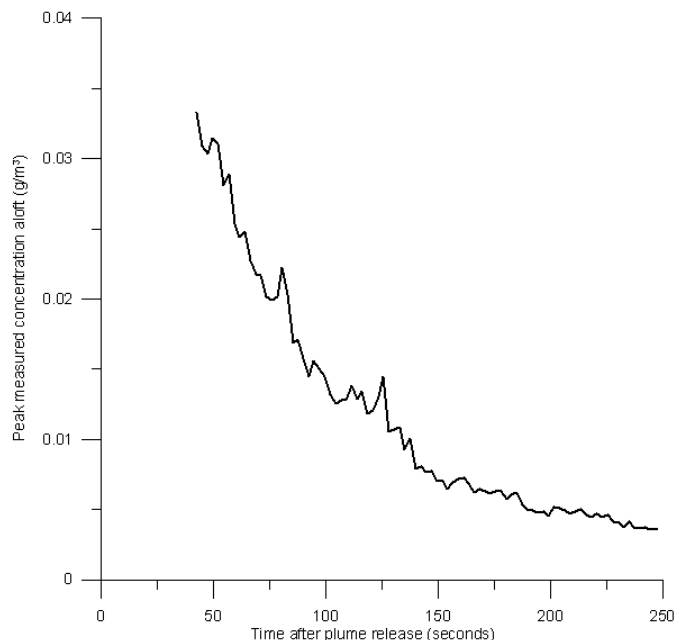


Figure 5: Remotely measured peak concentration in the plume after release for a single pass of the airplane.

Conclusions

It is possible to remotely measure the concentration of aerosol plumes in the air. Lidar offers a usable technique to perform such measurements and a viable technology for studying agricultural air quality.

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Biofiltration of the Critical Minimum Ventilation Exhaust Air

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Abstract

This research project investigated the gas and odor emission reduction potential from a deep-pit swine finisher using a strategy of partial biofiltration of a critical minimum amount of exhausted air (CMEA). The CMEA was defined as the amount of air exhausted to a stable hot-weather atmosphere, typical of summer night conditions. Ventilation air exhausted during the heat of summer days is exhausted into an atmosphere that is, for the vast majority of times, very unstable providing excellent natural mixing potential near the building source. In more stable evening atmospheres, biofiltration of the CMEA would reduce gas and odor emissions and the potential for odor plumes to travel long distances. The overall effect could be a more attractive and affordable biofiltration strategy that maximizes odor and gas reduction potential when it is needed most.

For this research project the CMEA was fixed at $75 \text{ m}^3\text{-h}^{-1}\text{-pig}^{-1}$. The preliminary results from this ongoing research effort indicate an ammonia reduction of the pit-exhaust air of 44 percent, a hydrogen sulfide reduction of 58 percent, and an odor reduction of 54 percent.

Introduction

Odor and gas dispersion from swine facilities is receiving much attention. Sources of odor and gas emissions include land application events, manure storage facilities, and building exhaust air. Ventilation air is typically exhausted into the ambient atmosphere without treatment. This exhaust air contains odorous gases and particulate matter that can represent a concentrated odor source.

Many researchers have examined odors from livestock facilities to determine the constituents that are most influential in olfactory perceptions. Hammond *et al.* (1979) found that the most important compounds were acids, phenols and carbonyls. However, results indicated that odors occurring at large distances from animal facilities were amplified by the presence of dust particles (Cai *et al.*, 2005). Hartung (1985) stated that filtering the dust from exhaust air can reduce the odor emission from animal houses up to 65 percent.

Biofiltration works well for treating odors and contaminated gases from livestock sources because an uncharacterized population of microorganisms can adapt to the profile of compounds to be treated. By-products are primarily water, CO_2 , mineral salts, and oxidizable inorganic compounds. Biofilters have been shown to remove in excess of 90 percent of the odor, 95 percent of the hydrogen, and 60-75 percent of the ammonia emissions from a livestock source. Thus, biofiltration is an effective technology to improve air quality adjacent to and downwind from livestock facilities. Biofilters are relatively economical and simple to install and maintain, but require a large land area and in most designs require higher capacity fans to move the ventilation air to be treated through the filter material.

Noren (1985) used peat and heather over wooden slats to form a biofilter for animal housing. It was found that odors were absorbed and converted by microorganisms to odorless substances after the biofilter was allowed to mature. Gases were decreased at an average rate of 50 percent with an 80 percent removal rate when the biofilter was kept at an optimal moisture content. Zeisiz and Munchen (1987) used several different materials including humus soil, compost, and peat. O'Neill and Stewart (1985) summarized the effectiveness of biofilters showing the odor removal efficiency ranged from 50 to 90 percent. Biofiltration adapted to U.S. animal production practices has been studied in detail with specific design guidelines given by Nicolai *et al.* (2002), Janni *et al.* (2001) and Nicolai and Janni (2001).

For this research project, the objective was to define and treat *via* biofiltration the least amount of ventilation air possible. This air, called the critical minimum exhaust air (CMEA), was defined as the amount of exhaust air that resulted in the most potential for downwind odor complaints by near-by residences. The objectives of this research project were to;

- 1). Determine and propose the CMEA for a typical deep-pit swine finishing facility,
- 2). Modify an existing deep-pit swine finisher to biofilter the CMEA, and,
- 3). Monitor concentrations of hydrogen sulfide, ammonia, and odor from a CMEA biofiltered barn compare with a conventional pit-ventilated deep-pit swine finisher.

Methods

Determining the Critical Minimum Exhaust Air (CMEA)

A six-state research project funded by the USDA-IFAFS program (APECAB) was conducted to investigate gas and particulate emissions from swine and poultry facilities. This project measured real-time emissions from fan ventilated animal and poultry housing facilities. A critical variable measured during this project was the real-time airflow rates required to assess gas and particulate emissions. In Iowa, a deep-pit swine finisher was monitored. The airflow data collected in Iowa showed that even in the heat of the summer, the evenings cooled enough forcing the barn to ventilate at a rate that was about 50 percent of the maximum mid-day requirements. A typical two-day result for August 2003 is given in Figure 1. The amount of ventilation air exhausted between day and night time periods is being delivered to two very distinct atmospheres. During hot summer days, the air is being exhausted to a predominantly unstable environment where very good natural vertical mixing of the exhausted air with fresh ambient air is being realized near the source. During the evening periods, the air is being exhausted into a relatively stable atmosphere, implying that natural vertical mixing is low and the potential for ground-level gas concentrations at further downwind distances is more probable than during the heat of the day. It is not uncommon for the ventilation rate to change from 35 fresh-air-changes per hour (ACH) during the night to a maximum of 100 ACH during the heat of the day. To biofilter all ventilation air (i.e. 100 ACH) would not be practical, and, based on atmospheric stability conditions is unwarranted. This change in ventilation requirements, as a function of atmospheric stability, can be used as an advantage with respect to ventilation air mitigation using biofilters or any other ventilation air mitigation strategy in general.

During the cooler and more stable evening hours, odors and gases exhausted from buildings have the potential to travel greater distances than those exhausted during the unstable daytime hours. Therefore, it makes practical sense to treat the night-time ventilation air to offset the lack of natural atmospheric mixing present during the heat of the day. This night-time exhausted air was defined for this research project as the CMEA. For this research project, the CMEA was defined as the amount of air associated with a rate equal to $75 \text{ m}^3\text{-h}^{-1}\text{-pig}^{-1}$. For the barn shown in Figure 1, with a capacity of 960 pigs, corresponds to a CMEA treatment level of $72,000 \text{ m}^3\text{-h}^{-1}$.

Barn Changes Made to Accommodate the CMEA

Two side-by-side 300-hd deep-pit swine finishing rooms were used at a cooperator's site in central Iowa (Figure 2). The control room was left unaffected and consisted of two 61-cm diameter variable speed fans drawing air from the head-space of the manure pit, exhausted directly to atmosphere. The balance of ventilation air was provided with side-wall curtains. The test room, originally identical to the control room, was modified in the following ways to accommodate the biofiltration of the CMEA:

- a. The two 61-cm diameter pit fans as shown in the control room were removed and replaced with four single-speed fans, two each in two separate exhaust plenums (Figure 2).
- b. A biofilter consisting of wood chips was installed at a depth of 27 cm and a surface area of 80 m^2 .

The installed biofilter fans, labeled as shown in Figure 2, were controlled in four stages with the characteristics given in Table 1.

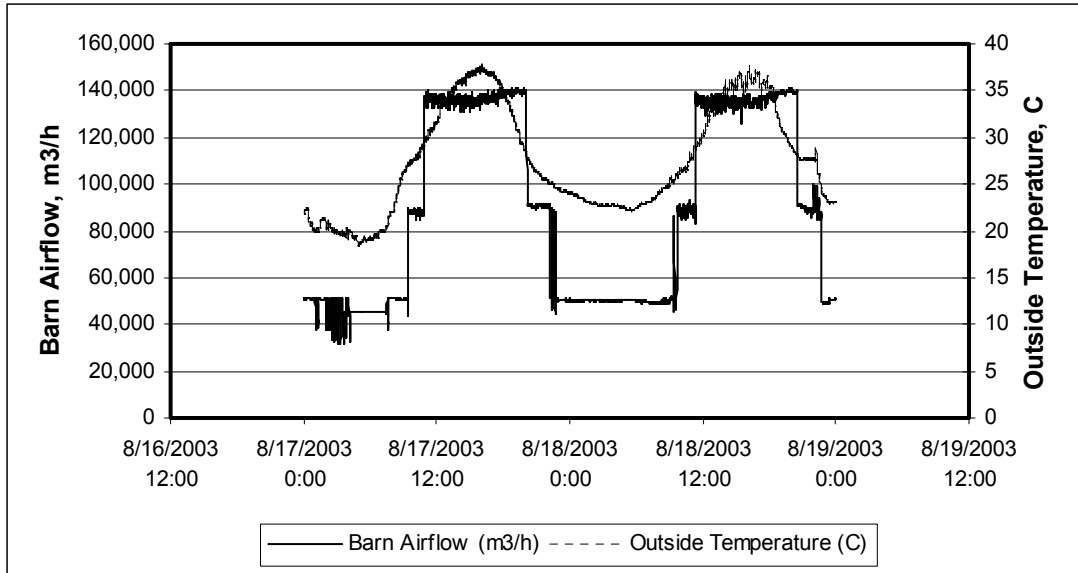


Figure 1. Building airflow changes with summer outside temperature.

Table 1. Fan staging to achieve the CMEA requirement .

Fan Stage	Fans Active*	Measured Operating Static Pressure (Pascals)	Estimated Airflow** (m ³ -h ⁻¹ -pig ⁻¹)
1	1	13	6
2	1 and 2	13	18
3	1 and 2 and 3	37	49
4	1 and 2 and 3 and 4	50	77

* Fan 1=30-cm diameter, Fan 2=41-cm, Fans 3 and 4=61-cm

** based on 85% of rated capacity at the operating pressures measured for each stage

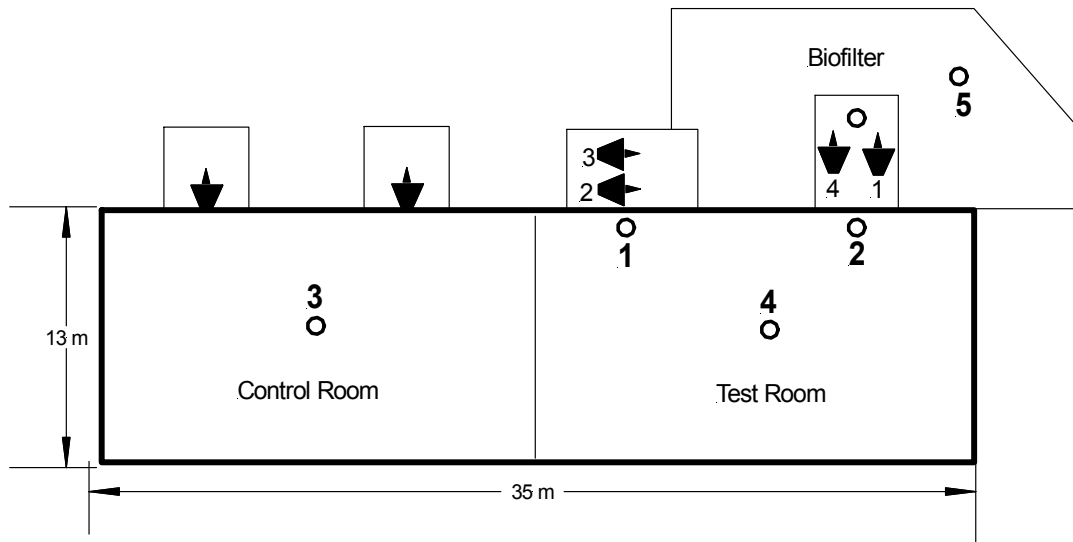


Figure 2. Building layout showing control and test room set-up with sampling locations (SL).

Gas Concentration Sampling

A Mobile Emissions Laboratory (MEL) was installed on-site that housed all monitoring equipment. Ammonia and hydrogen sulfide sampling was conducted semi-continuously with odor sampling conducted bi-weekly. Ammonia and hydrogen sulfide were monitored using a chemiluminescence-based analyzer (TEI, Inc; Model 17C) with hydrogen sulfide monitored using a pulsed-fluorescence-based analyzer (TEI, Inc; Model 45C). Gas samples (see Figure 2) were routed through 10-mm ID Teflon-lined sample lines to a common sampling system with switching solenoids to route samples at each location, in sequence, to the NH₃ and H₂S analyzers. All sample lines were heated at critical locations where condensation could occur and each sample line was fitted with a 0.45- μ m particulate filter at the intake. Samples from each sample location were presented to the analyzers for a total of 10-min with the final 4-min of each sampling sequence used for analysis. Odor samples were collected from each sampling location, collected in 10-L Tedlar sample bags and evaluated using dynamic dilution olfactometry (St. Croix Sensory, Inc; Model AC'SCENT).

Results and Discussion

Ammonia, Hydrogen Sulfide and Odor Concentration Results

Gas and odor concentrations were collected between July and October 2005 with the results shown in Table 2. Odor concentration entering the biofilter plenum from the head-space of the deep-pit (sample lines 1&2) averaged 1,169 \pm 788 OU/m³ with an average odor concentration leaving the biofilter (SL=5) of 540 \pm 316 OU/m³. These differences represented an average reduction in odor concentration of 54 percent ($p < 0.01$). The control barn room air (SL=3) odor concentration (603 \pm 484 OU/m³) compared with the test room air (SL=4; 577 \pm 546 OU/m³) was not significant ($p > 0.50$) and the biofilter exhaust air (SL=5) was not significantly different ($p > 0.50$) from the biofilter room air (SL=4). The ammonia concentration entering the biofilter plenum from the head-space of the deep-pit (SL=1) averaged 9.1 \pm 5.5 ppm with an average ammonia concentration leaving the biofilter (SL=5) of 5.1 \pm 3.4 ppm. These differences represented an average reduction in ammonia concentration of 44 percent ($p < 0.01$). The control barn room air (SL=3) ammonia concentration compared with the test room air (SL=4) was not significant ($p > 0.50$) and the biofilter exhaust air (SL=5) was not significantly different ($p > 0.10$) from the biofilter room air (SL=4). Similar results were found for hydrogen sulfide.

Table 2. Average (\pm SD) odor, ammonia and hydrogen sulfide concentrations measured between July and October 2005 for the sample locations shown in Figure 2.

Sample Location (SL)	Sample Descriptor	Odor Concentration (OU/m ³)	Ammonia Concentration (ppm)	Hydrogen Sulfide Concentration (ppb)
1 & 2	Pit Exhaust	1,169 \pm 788	9.1 \pm 5.5	253 \pm 263
3	Control Room	603 \pm 484	3.7 \pm 3.2	95 \pm 94
4	Test Room	577 \pm 546	3.6 \pm 3.1	90 \pm 113
5	Biofilter Exhaust	540 \pm 316	5.1 \pm 3.4	106 \pm 104

Assessment of Gas Emission Reduction Using Biofiltration of the CMEA

The odor, ammonia and hydrogen sulfide reductions reported here using the installed biofilter were modest. The biofilter installed was developed in such a way that fan pressure drop was a key variable used in an attempt to develop a biofilter that could be used with existing agricultural fans. For this design constraint to be realized, the decision was made to develop a biofilter that resulted in no more than a 60 Pascal pressure difference. The end result, after several iterations, was a biofilter consisting entirely of wood chips. Also, in order to achieve the reductions presented here, the biofilter media had to be frequently wetted to prevent drying of the material.

Conclusions

This on-going research on biofilter technology applied to fan ventilated animal housing facilities concentrated on the biofiltration of the critical minimum exhaust air (CMEA). This was defined as the amount of ventilation air required by swine finishing facilities during night-time summer periods. For this research project the CMEA was fixed at $75 \text{ m}^3\text{-h}^{-1}\text{-pig}^{-1}$. The preliminary results indicate an ammonia reduction of the pit-exhaust air of 44 percent, a hydrogen sulfide reduction of 58 percent, and an odor reduction of 54 percent. Further research will be conducted on biofilter additives to improve gas reduction efficiencies without increasing back-pressure on the exhaust fans. The authors would like to thank the Iowa Pork Producers Association and the USDA-Special Grants program for funding this on-going research.

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A Novel Rapid Method for the Characterization and Identification of Aerosols: Multi-Elemental Scanning Thermal Analysis (MESTA)

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Abstract

A rapid and sensitive method for the chemical analysis of aerosols is highly desirable for air-quality monitoring and research works. Here, I report the application of a newly developed multi-elemental scanning thermal analysis (MESTA) for chemical analysis of aerosols. We collected PM_{2.5} samples from the university campus (city) and from the Experimental Station (rural) and analyzed them with the MESTA. The results indicate that the PM_{2.5} campus and the that of the Experimental Station were different in the relatively low C/N/S ratio in the campus when compared to the rural Station. The PM_{2.5} collected near the pig pan of the Quincy Station, however, show similarity in chemical signature to that found in the campus. The weekend campus samples were similar to the weekday samples in chemical signature although the PM_{2.5} concentration was slightly higher during the weekends. In addition to the bulk chemical information, the MESTA reveals chemical signature of the PM_{2.5} components. Results of this study suggest that the MESTA technology could be a rapid and effective way to study aerosols in the air.

Introduction

A rapid and sensitively method for the chemical analysis of aerosols is highly desirable for air-quality monitoring and research works. Unfortunately, such basic requirement is still not met currently due to the limited separation techniques developed for solid samples. Traditional sequential extraction technique is not only tedious and incomplete, it may introduce contaminants and artifacts to an aerosol sample. Bulk elemental analysis can provide some chemical information but it is generally insufficient for identification and characterization purposes. Besides technical problem in separation, aerosol analysis requires high sensitivity and speed due to small size and possible post-sampling change of a sample. Various carbon scanning thermal analysis techniques have been developed to examine particulate air samples in the last two decades (Chow *et al.* 2001). In addition to the information of carbon content, carbon scanning thermal analysis also reveals about the temperature at which the carbon volatilizes (i.e., a carbon thermogram). Carbon scanning thermal has been quite efficient for differentiating different forms of carbon compounds in a solid sample. For example, organic, elemental and carbonate forms of carbon can be readily differentiated by their characteristic volatile temperatures in a carbon scanning thermal analysis (Conny *et al.* 2003). Carbon scanning thermal analysis reveals only the carbon information. Important information such as nitrogen and sulfur in aerosol is not available from carbon scanning thermal analysis. Here, I report the development of a multi-elemental scanning thermal analysis (MESTA) that can quickly and sensitively analyze aerosol samples and provide the information on C, N and S simultaneously. The simultaneous C, N and S thermograms of the MESTA provide chemical signature of organic and inorganic components in an aerosol sample. We demonstrate the application of MESTA to PM_{2.5} collected from the air of the campus and a rural Quincy Experimental Station. The simplicity, reproducibility and sensitivity of MESTA make the technology a good tool for air quality research.

Materials and Methods

The MESTA device

A prototype MESTA system has a quartz pyrotube that consists of a sample compartment and a combustion compartment. The combustion compartment is maintained at 1100 °C by a constant-temperature furnace. The sample compartment can be heated to 800 °C from ambient temperature at a given rate through a programmable furnace. The sample compartment has a gas inlet for carrier gas, a mixture of oxygen and helium. The mixing ratio of helium and oxygen in the carrier gas is adjustable by two flow meters. The

combustion compartment has an inlet for pure oxygen. The pyrotube is connected to detectors of C (a CO₂-IR analyzer), N (a NO₂ chemiluminescent detector) and S (a SO₂ chemiluminescent detector) in a serial manner. A PC based four-channel data logger (National Instrument) is used to record simultaneously the real-time temperature, C, N and S signals. When an analysis is performed, a weighed sample (0.1 – 20 mg) is loaded into a quartz boat and then inserted into the sample compartment. The sample compartment is then closed with a Teflon plug. After the air is purged out of the pyrotube by the carrier gas, the sample compartment is heated at a constant rate (50 °C/min) from ambient to 800 °C. As the temperature of the sample compartment increases, volatilized component of the sample is carried to the combustion compartment with carrier gas where C, N and S are oxidized to their respective oxides. The combusted gas stream is dehydrated and purified with filters and the oxidized C, N and S gases are detected by the respective detectors. The result of the analysis is displayed as C, N and S thermograms. Silver sulfide (Ag₂S) and elemental sulfur (S₈) are used as temperature markers to calibrate the thermograms as desired.

Ambient Air Sampling

Weekday and weekend air samples were collected from the Florida A&M University campus (Tallahassee, FL) on the rooftop of a 6-story building that was located approximately 600 meters from a main road intersection. Air samples were also collected from the Quincy Experimental Station of the Florida A&M University at two sites: one close (10 m) to the pig pan area and the other close to the office area, which is 300 m from the pig pan. PM_{2.5} samples were collected using a PM_{2.5} air sampling device that had been designated by EPA as Equivalent using the VSCC (Designation No. EQPM-0202-142; Model PQ200; BGI, Inc.) and with a pumping rate of 16.7 liters per minute. The PM_{2.5} was collected on a pre-weighed and pre-fired (900 °C, 2 h.) 47 mm quartz filter inside the sampler. Total sampling collection time ranged from 27 to 60 hrs. After the set sampling time, the filter was removed and weighed again and the PM_{2.5} concentration was calculated based on the filter weight difference and the total volume of air that passed through the air sampler. Each collected filter was put in a pre-fired stainless steel container and stored in a freezer until analysis. The weekday samples represent cumulative samples that were collected between 11 am and 6 pm each day for five days. The weekend air sample was collected for approximately 40 hours continuously during the weekends.

PM Sample Analysis

The quartz filter that contained the PM_{2.5} sample was weighed and cut into eight pie-shaped pieces. Each piece was weighed. The piece/total weight ratio was used to calculate the portion of the PM sample in that piece. A piece of the filter was loaded in the sample holder and the MESTA was carried out from 35 to 800 °C with 50 °C/min heating rate, 40 % oxygen 60 % helium carrier gas. The C, N and S thermograms of MESTA were recorded using a data logger (National Instrument). The C, N and S signals in the thermograms were calibrated to their relative atomic abundance with pure cystine standard.

Results and Discussion

The PM_{2.5} concentration and its chemical property of the air samples are listed in Table 1. The PM_{2.5} in the ambient air of the Tallahassee campus was significantly higher than that in the Quincy Experimental Station near the office area. The highest PM_{2.5} observed, however, was in the air close to the pig pan of the Quincy Station: about three times the PM_{2.5} concentration of the office area just 300 m away.

The PM_{2.5} in the campus during the weekends was slightly higher than that during the weekdays for some unknown reasons. The most noticeable is the higher nitrogen and sulfur contents relative to the carbon found in the impacted areas associated with higher PM_{2.5} concentration. For example, the campus PM_{2.5} during the weekdays had average C/N/S ratio of 5.9/1.7/1. The C/N/S ratio of the campus PM_{2.5} became 4.3/1.6/1 during the weekends when the PM_{2.5} was slightly higher. The PM_{2.5} concentration of the Quincy Station was lower than that in the campus. But the PM_{2.5} C/N/S ratio is much higher at 36.1/7.3/1. The impacted Quincy Experimental Station air near the pig pan area showed a distinct PM_{2.5} C/N/S ratio of 3.1/2.8/1. The above results indicate that the signature of an impacted air with higher PM_{2.5} is characterized by the higher N and S contents relative to the carbon. The black carbon (BC) content of the pig pan area was also significantly higher (11.2 % of the Total C) than the office area (2 % of the total C) and the campus (4-7.8 % of the total C).

Table 1. The PM_{2.5} concentration and its chemical property of the air sample collected in this study. (Units are in µg/m³)

Air sample	PM _{2.5}	Carbon	Nitrogen	Sulfur	BC	C/N/S
Campus weekdays	17.68-18.91	2.54-4.09	0.84-1.68	1.02-3.01	0.1-0.16	5.9/1.7/1
Campus weekends	26.54-32.44	4.33-5.31	1.87-2.82	2.37-4.19	0.34-0.41	4.3/1.6/1
Expt. Station near pig pan	31.68-43.47	6.37-7.59	4.15-7.61	3.89-6.06	0.71-0.85	3.1/2.8/1
Expt. Station near office	12.48	2.64	0.62	0.19	0.05	36.1/7.3/1

Apart from the bulk chemical information, the MESTA thermograms give the chemical signature pertaining to the PM_{2.5} components. Fig. 1 and 2 show the C, N and S thermograms of the PM_{2.5} samples collected in the city campus and the Quincy Station, respectively. The PM_{2.5} of the campus air is similar chemically between the weekday and weekend samples. Both PM_{2.5} samples have a low-temperature and a high-temperature components. Within the low-temperature component, there was a sulfur containing component and a non-sulfur containing component. The high-temperature component had low nitrogen content and virtually had no sulfur in it. The PM_{2.5} of the Quincy Station near the pig pan area had the similar chemical signature of the campus PM_{2.5}, except that the nitrogen content of the low-temperature component was significantly higher in the rural Quincy than in the city campus. The PM_{2.5} chemical signature of the PM_{2.5} in Quincy near the office area was quite different from the rest of the PM_{2.5} samples in that the nitrogen content was found only in the lowest temperature component and the sulfur does not synchronize with C or N. For comparison, we also analyzed the NIST SRM 1649a urban dust standard (Fig. 3). The NIST urban dust standard has also higher nitrogen content organic matter that is in the low-temperature component. In the NIST urban dust standard, there is a nitrogen component that does not synchronize with the carbon and the volatilization temperature is below 200 °C. Detail interpretation of the MESTA thermograms requires further work and analyses. Nonetheless, the MESTA provides new chemical information of the PM_{2.5} samples in a rapid and convenient manner, which is needed in air quality studies.

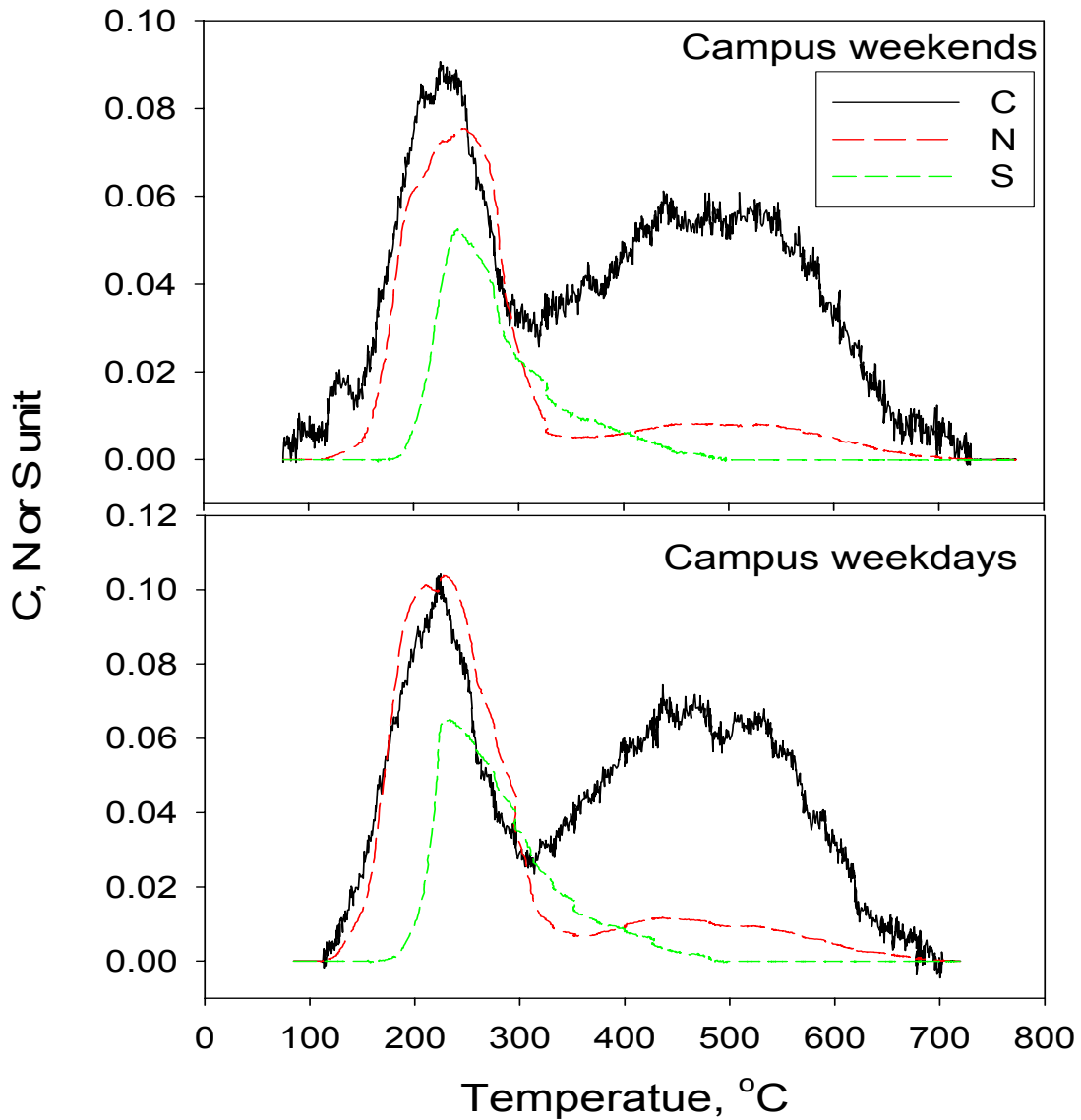


Figure. 1. The C, N and S thermograms of the air PM_{2.5} samples collected during weekdays and weekends, respectively in the Tallahassee Florida A&M University campus. The C, N and S units are expressed proportional to their relative atomic abundance.

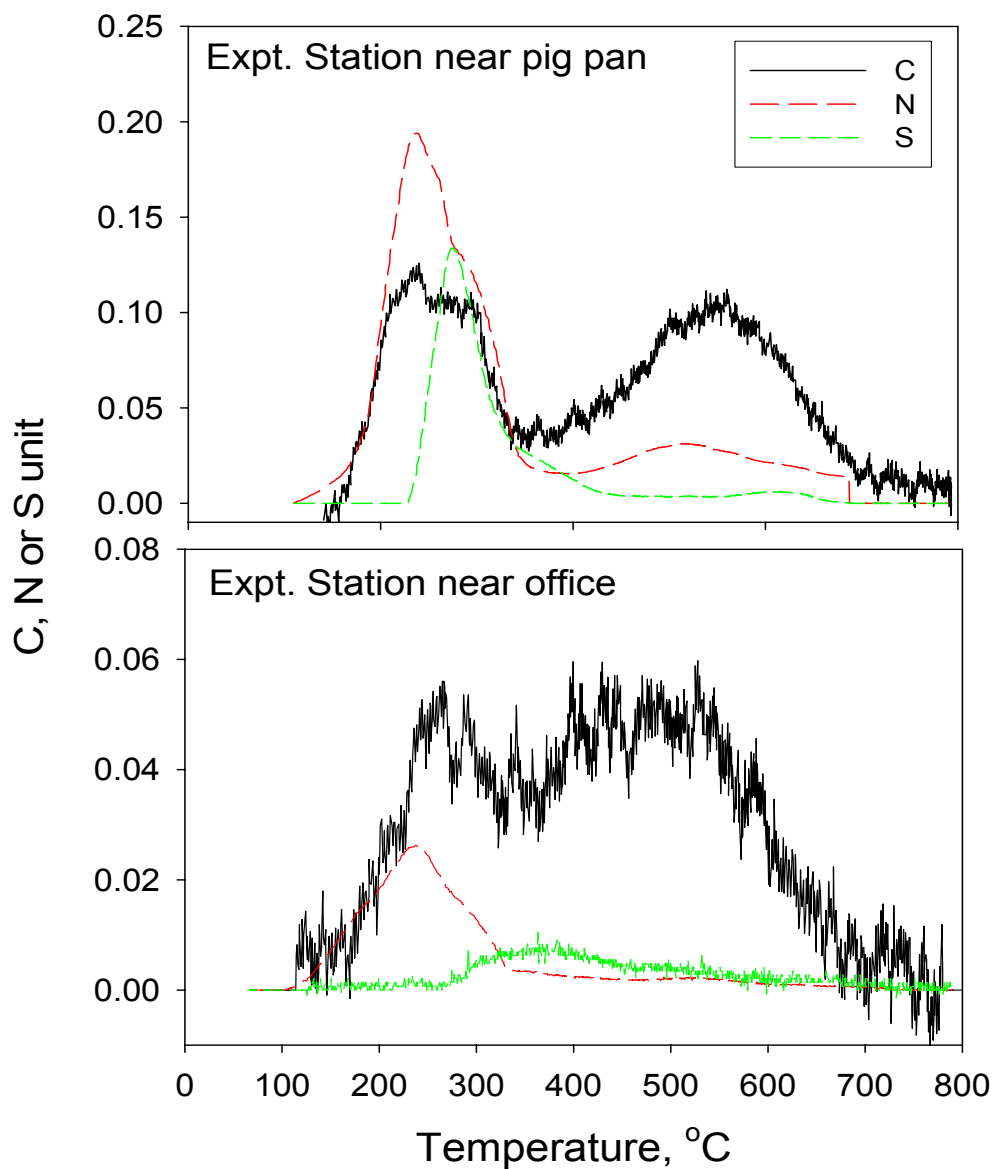


Figure. 2. The C, N and S thermograms of the air $PM_{2.5}$ samples collected from the pig pan area and the office area of the Quincy Experimental Station, respectively. The C, N and S units are expressed proportional to their relative atomic abundance.

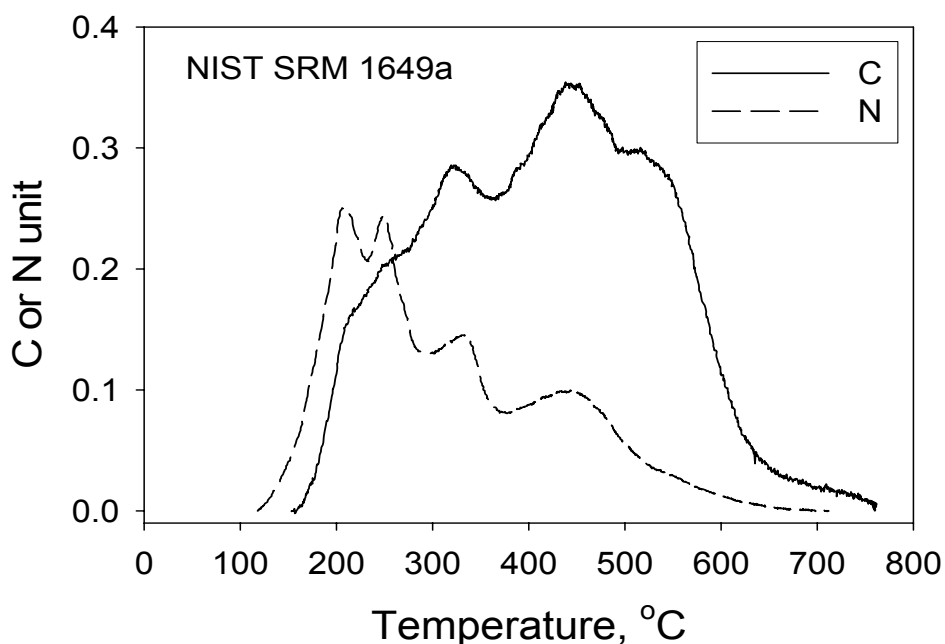


Figure 3. The MESTA thermograms of the NIST SRM 1649a urban dust standard. The C and N units are expressed proportional to their relative atomic abundance.

Conclusions

Analysis of PM_{2.5} samples collected from the Florida A&M University Tallahassee campus and the Quincy Experimental Station using the MESTA technology reveals that the chemical signatures of the Tallahassee campus and the Quincy Station were quite different in that the relative N and S contents to carbon is much higher in the former than the latter. The PM_{2.5} collected near the pig pan area in the Quincy Station, however, show similarity in chemical signature to that in the city. The weekend campus samples were similar to the weekday samples in chemical signature although the PM_{2.5} concentration was slightly higher during the weekends. The results of this study indicate that the newly developed MESTA technology is a rapid and effective way to study PM_{2.5} in the air not only in terms of the bulk chemical property but also in terms of the component chemical properties.

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Ammonia Emissions and their Implications on Fine Particulate Matter Formation in North Carolina

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Abstract

Ammonia (NH₃) is an important atmospheric pollutant that plays a key role in several air pollution problems. The accuracy of NH₃ emissions can have a large effect on air quality model (AQM) predictions of aerosol sulfate, nitrate, and ammonium concentrations. Large uncertainties exist in NH₃ emission inventories in both total emissions and temporal variations. In this work, sensitivity simulations are conducted to assess the impact of NH₃ emissions on the formation of PM_{2.5} and its composition in August and December 2002 in North Carolina.

Introduction

Ammonia in the atmosphere has a large effect on the formation of fine particles, such as aerosol ammonium sulfate and ammonium nitrate, due to its important role in the neutralization of acidic compounds (Seinfeld and Pandis, 1998). Major emission sources of NH₃ include animal waste, human waste, and biogenic soil emissions (Bouwman et al., 1997; Aneja et al., 1998; Goebes et al., 2003). Ammonia emission estimates for the state of North Carolina indicate that livestock and fertilizer together are responsible for about 80-90% of total NH₃ emissions (Wu et al., 2006). Accurate estimates of NH₃ emissions are needed for reliable AQM predictions, but current NH₃ emission inventories are uncertain both in annual estimates and in the monthly, daily, and diurnal variations. Since most ammonia emissions are from non-point sources such as livestock operations and fertilized fields, ammonia emissions from these sources are difficult to measure directly (Gilliland et al., 2003; Pinder et al., 2004 a, b, 2005).

To study the fate of NH₃ and the impact of the agriculture-livestock NH₃ emissions on ambient fine particulate matter (PM_{2.5}), two 1-month baseline AQM simulations have been recently conducted using the U.S. EPA Community Multiscale Air Quality (CMAQ) modeling system for August and December, 2002 in a Southeast U.S. domain that covers primarily the state of North Carolina at a 4-km horizontal grid spacing (Wu et al., 2005; 2006). The baseline simulation results are evaluated using the observational datasets from national and state-owned networks such as the Interagency Monitoring of Protected Visual Environments (IMPROVE), the EPA Speciation Trends Networks (STN), the Clean Air Status Trends Network (CASTNet), and the North Carolina Department of Environment and Natural Resources (NCDENR). The evaluation shows underpredictions for PM_{2.5} (with normalized mean bias (NMB) of -31.8% to -45.2%), NH₄⁺ (-18.0% to -35.2%), NO₃⁻ (-40.9% to -64.2%), SO₄²⁻ (-6.2% to -18.7%) in August; and overpredictions for PM_{2.5} (8.8% to 46.7%), NH₄⁺ (30.8% to 53.2%), NO₃⁻ (58.6% to 158.1%), and overprediction for SO₄²⁻ at the IMPROVE sites (9.1%) but underpredictions at the STN (-21.0%) and the CASTNet sites (-4.2%) in December. In addition to the inaccuracies in the simulated meteorological field and the uncertainties in the model treatment for aerosol dynamics and chemistry, the inaccuracies in the estimation of NH₃ emissions can have a large effect on the model performance on ammonium, nitrate, and PM_{2.5}. A comprehensive evaluation of both meteorological and chemical conditions along with a detailed process analysis for the baseline simulations in August and December 2002 are being performed (Krishnan et al., 2006; Wu et al., 2006). The relative importance of meteorological and chemical processes is being examined. The likely reasons for the discrepancies between the simulated and observed meteorological variables and chemical concentrations are being identified.

Summary of Sensitivity Simulations and Results

Differences exist in the domainwide total NH_3 emissions provided by different emission inventories due likely to different approaches used to create the emission inventories and different source categories, strengths, and emission factors included. For example, in the baseline simulations, the domainwide total NH_3 emissions are based on the emission inventory developed by the Visibility Improvement State and Tribal Association of the Southeast (VISTAS) (Morris and Koo, 2004). It gives a domainwide total of 568 tons per day in August and 334 tons per day in December. The Carnegie Mellon University (CMU) inventory (Goebes et al., 2003; Pinder et al., 2004b; Strader et al., 2005), on the other hand, gives a domainwide total of 735 tons per day in August and 226 tons per day in December. Using the CMU inventory as a benchmark, two methods have been applied to adjust the baseline NH_3 emissions that are based on the VISTAS emission inventory (referred to as Baseline hereafter). The first method is to use the total CMU NH_3 emissions but still keep the same diurnal variability as the baseline simulations (referred to as Sen_uniform hereafter), namely, multiplying the baseline VISTAS total NH_3 emissions by a domainwide uniform factor of 1.29 for August and by 0.68 for December to match the total CMU NH_3 emissions. The second method is to use the total CMU NH_3 emissions and the CMU diurnal variability (referred to as Sen_diurnal hereafter), namely, replacing the hourly NH_3 emission rates in the baseline simulations by those of the CMU inventory. The difference between Sen_uniform and Sen_diurnal lies in the diurnal variability profiles used, namely, Sen_diurnal gives higher daytime emission rates and lower nighttime emission rates than those of Sen_uniform in August, and has emission rates that are higher between 1-10 a.m., lower between 4 p.m.-11 p.m., and similar between 10 a.m.-4 p.m. in December. Sensitivity simulations with the two emission adjustment methods are performed to study the sensitivity of the concentrations of $\text{PM}_{2.5}$ and ammonium salts to the NH_3 emission adjustments in August and December, 2002. The sensitivity simulation results are evaluated in terms of spatial distributions, temporal variations, and the overall statistical performance.

The results of the sensitivity simulations show a large spatial and temporal variation of the impact of NH_3 emissions on PM ammonium and nitrate formation. In addition to emissions, other influential factors such as meteorological conditions and ambient chemical conditions are being analyzed along with the impacts of NH_3 emission adjustments. The statistical results show that the two emission adjustment methods improve the overall model performance in both August and December. For example, the NMBs of NH_4^+ change from -18.0% (Baseline) to -10.7% (Sen_uniform) and -11.8% (Sen_diurnal) in August and from 30.8% (Baseline) to 15.6% (Sen_uniform) and 16.8% (Sen_diurnal) in December at STN sites and from -35.2% (Baseline) to -28.6% (Sen_uniform) and -28.7% (Sen_diurnal) in August and from 53.2% (Baseline) to 41.9% (Sen_uniform) and 42.1% (Sen_diurnal) in December at IMPROVE sites. The correlation coefficients from the two sensitivity simulations remain the same as or similar to those from the baseline simulations for all species except NH_4^+ , whose correlation coefficient is slightly improved, changing from 0.81 to 0.86 and 0.85 for Sen_uniform and Sen_diurnal in August.

To identify the possible reasons for the similar performance of the two emission adjustment methods, we further analyze the predicted hourly $\text{PM}_{2.5}$ concentrations with the hourly observations that are available only at one NC DENR site, i.e., Grainger (GR) 371190041 site, in August. The statistics calculated for the GR 371190041 site show that Sen_uniform and Sen_diurnal give mean hourly $\text{PM}_{2.5}$ concentrations of 13.76 and 13.79 $\mu\text{g m}^{-3}$, respectively, and NMBs of -19.4% and -19.2%, respectively, during the daytime. During the nighttime, i.e., Sen_uniform and Sen_diurnal give mean hourly values of 13.88 and 13.79 $\mu\text{g m}^{-3}$, respectively, and NMBs of -19.6% and -20.1%, respectively. For comparison, the baseline simulation gives a mean hourly $\text{PM}_{2.5}$ concentration of 13.52 $\mu\text{g/m}^3$, with a NMB of -20.8% during the daytime and a mean hourly $\text{PM}_{2.5}$ concentration of 13.73 $\mu\text{g/m}^3$, with a NMB of -20.5% during the nighttime. The correlation coefficients for the two sensitivity simulations are the same as those of the baseline simulations. Two sensitivity simulations show similar predictions on hourly $\text{PM}_{2.5}$ concentrations to those of the baseline simulation, partially due to the fact that ammonium and nitrate account for only about 10% of the total $\text{PM}_{2.5}$ mass at this site, and the changes in the concentrations of ammonium and nitrate as a result of the emission adjustments in the two sensitivity simulations may be too small to change the predicted $\text{PM}_{2.5}$ concentrations appreciably. Another possible reason is that the site is located in ammonia-rich regime where there are sufficient amounts of ammonia to neutralize sulfate and nitrate ions. In such a regime, the increased amount of NH_3 will not result in a significant conversion to particulate NH_4^+ . The differences in the predicted concentrations of ammonium and nitrate between the two sensitivity simulations, therefore,

are overall not statistically significant at this site, resulting in similar PM_{2.5} concentrations by the two simulations. More detailed analyses are being conducted to fully understand all major reasons for the similar performance of the two emission adjustment methods.

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