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Characterizing reduced sulfur compounds emissions from a swine concentrated animal feeding operation

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HIGHLIGHTS

- Intensification of livestock production methods has affected air quality.
- There is limited information on RSCs emissions from swine CAFOs.
- This article provides a comprehensive analysis of RSCs emissions from a swine CAFO.

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ABSTRACT

Reduced sulfur compounds (RSCs) emissions from concentrated animal feeding operations (CAFOs) have become a potential environmental and human health concern, as a result of changes in livestock production methods. RSC emissions were determined from a swine CAFO in North Carolina. RSC measurements were made over a period of ≈1 week from both the barn and lagoon during each of the four seasonal periods from June 2007 to April 2008. During sampling, meteorological and other environmental parameters were measured continuously. Seasonal hydrogen sulfide (H_2S) barn concentrations ranged from 72 to 631 ppb. Seasonal dimethyl sulfide (DMS; CH_3SCH_3) and dimethyl disulfide (DMDS; $CH_3S_2CH_3$) concentrations were 2–3 orders of magnitude lower, ranging from 0.18 to 0.89 ppb and 0.47 to 1.02 ppb, respectively. The overall average barn emission rate was 3.3 g day^{-1} AU^{-1} (AU (animal unit) = 500 kg of live animal weight) for H_2S, which was approximately two orders of magnitude higher than the DMS and DMDS overall average emissions rates, determined as 0.017 g day^{-1} AU^{-1} and 0.036 g day^{-1} AU^{-1}, respectively. The overall average lagoon flux was 1.33 μg m^{-2} min^{-1} for H_2S, which was approximately an order of magnitude higher than the overall average DMS (0.12 μg m^{-2} min^{-1}) and DMDS (0.09 μg m^{-2} min^{-1}) lagoon fluxes. The overall average lagoon emission for H_2S (0.038 g day^{-1} AU^{-1}) was also approximately an order of magnitude higher than the overall average DMS (0.0034 g day^{-1} AU^{-1}) and DMDS (0.0028 g day^{-1} AU^{-1}) emissions. H_2S, DMS and DMDS have offensive odors and low odor thresholds. Over all four sampling seasons, 77% of 15 min averaged H_2S barn concentrations were an order of magnitude above the average odor threshold. During these sampling periods, however, DMS and DMDS concentrations did not exceed their odor thresholds. The overall average barn and lagoon emissions from this study were used to help estimate barn, lagoon and total (barn + lagoon) RSC emissions from swine CAFOs in North Carolina. Total (barn + lagoon) H_2S emissions from swine CAFOs in North Carolina were estimated to be $1.22*10^6$ kg yr^{-1}. The barns had significantly higher H_2S emissions than the lagoons, contributing ≈98% of total North Carolina H_2S swine CAFO emissions. Total (barn + lagoon) emissions for DMS and DMDS were 1–2 orders of magnitude lower, with barns contributing ≈86% and ≈93% of total emissions, respectively. H_2S swine CAFO emissions were estimated to contribute ≈18% of North Carolina H_2S emissions.

1. Introduction

Reduced sulfur compounds (RSCs) emissions from concentrated animal feeding operations (CAFOs) can have a wide range of environmental impacts. On the local scale, the primary environmental

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effect is odor. RSCs are generally odorous, and are therefore key contributors to odorous emissions from CAFOs. Emissions of odorous compounds are important as they can cause health symptoms and additionally health effects on people in nearby areas (Schiffman and Williams, 2005), as well as affecting their quality of life (Wing et al., 2008; Wing and Wolf, 2000).

RSCs can also have regional environmental impacts as a result of the oxidation of RSCs, which leads to the formation of sulfur dioxide (SO_2). SO_2 can in turn further react to form aerosols such as ammonium sulfate and ammonium bi-sulfate, which through inhalation can affect human health (Coffin and Knelson, 1976) and decrease visibility (Malm et al., 1996).

Swine farming is one of North Carolina's largest animal agricultural industries, with a swine population of ≈10 million (United States Department of Agriculture (USDA, 2013)). The majority of the swine reside in the southeastern coastal plain of North Carolina. Therefore, RSC emissions from waste at swine CAFOs in North Carolina are an issue of potential environmental concern. Of the RSCs emitted from swine CAFOs, hydrogen sulfide (H_2S) is the most extensively studied. Emissions of H_2S from swine CAFO barns have been reported in previous studies (i.e. Zhu et al., 2000; Kim et al., 2008; Blunden et al., 2008; Ni et al., 2002; Rahman and Newman, 2012). Because H_2S emissions can vary due to differences in production, management and environmental conditions, there is a need for comprehensive regional measurement of swine CAFO emissions. Presently, a study by Blunden et al. (2008) is the only known study that has determined H_2S emissions from a swine CAFO barn in North Carolina. In comparison to H_2S, there are fewer measurement studies that have determined other RSC concentrations and emissions from swine CAFOs (Clanton and Schmidt, 2000; Trabue et al., 2008; Blunden et al., 2005; Kim et al., 2007; Feilberg et al., 2010; Hansen et al., 2012; Yao et al., 2011; Schiffman et al., 2001) with the majority of these studies only reporting concentrations. Furthermore, there is limited analysis of emissions with respect to seasonal variations.

With respect to H_2S, this study builds upon the Blunden et al. (2008) study by making an additional four seasons of H_2S barn emission measurements at the same commercial swine farm in eastern North Carolina. Measured H_2S emissions are evaluated with respect to seasonal and diurnal variations and environmental factors. A brief summary of H_2S lagoon fluxes from the sampling campaign is provided in this manuscript. For an in-depth analysis and discussion of H_2S lagoon fluxes with respect to diurnal and seasonal variations as well as environmental factors, the reader is referred to Rumsey and Aneja (2014). In addition, the emissions of other RSCs from the barn and lagoon are also determined over four seasonal sampling periods. These RSC emissions are also evaluated with respect to seasonal variations and environmental parameters. The potential local environment impact of RSCs emissions from swine CAFOs is assessed by comparing concentrations to their odor threshold. Additionally, measured emissions are used to estimate the total (barn + lagoon) North Carolina RSCs emissions from swine CAFOs. This analysis also allows an estimation of the relative contribution of barn and lagoon RSCs emissions to swine CAFO emissions in North Carolina.

2. Method and materials

2.1. Sampling site and scheme

The swine CAFO sampling site is an operational commercial swine finishing CAFO located in the eastern coastal plain of North Carolina. The swine CAFO consists of eight mechanically ventilated barns and an anaerobic lagoon. At the beginning of each rotation, 800–1100 swine are placed in each barn with an initial weight of 17–25 kg. The swine farm handles its waste using a method known as 'Lagoon and Spray Technology' (LST). Swine waste accumulates in a shallow manure collection pit under the barn, which has an average depth of ≈0.7 m. The waste is flushed weekly from the shallow manure collection pit into the anaerobic treatment lagoon. The lagoon surface area was measured during each sampling period and had an average surface area of 18,145 m^2. The lagoon waste is used to flush the shallow manure collection pits and is also sprayed onto crops for nutrient enrichment when needed.

Both barn and lagoon measurements were made over an approximate one week period in four different seasons (summer: June–August; fall: September–November; winter; December–February; spring: March–May) in 2007–2008, the summer season from June 8th–June 28th, 2007; the fall season from the October 20th–November 12th, 2007; the winter season from February 8th–February 29th, 2008; and the spring season from April 11th–April 28th, 2008.

2.2. Field sampling technique and instrumentation

2.2.1. H_2S measurements

Continuous H_2S measurements were made with a TEI model 450C pulsed fluorescence H_2S/SO_2 analyzer (Thermo Environmental Corporation, Mountain View, CA) with a range of 0–1000 ppb. Prior to each sampling season the analyzer was calibrated. During the sampling periods and also just after each sampling period, zero and span checks were conducted on the H_2S/SO_2 analyzer.

2.2.2. Canister field sampling and analysis

The field sampling of other RSCs was conducted by collecting whole air samples using both 6-L (liter) SUMMA treated stainless steel (SUMMA) canisters and fused-silica lined stainless steel (FSL) canisters. RSCs were measured using a gas chromatography–flame ionization detection (GC-FID) system at the National Exposure and Research Laboratory of the US Environmental Protection Agency (EPA) in Research Triangle Park, NC. Information on the GC-FID analytical system is provided in Rumsey et al. (2012) and Blunden et al. (2005). Calibration of the GC-FID system was performed using 0.25 ppm ± 1.2% propane in air (National Institute of Standards and Technology Standard Reference Material). From the slope of the multi-point calibration curve, a response factor is determined based on parts per billion carbon (ppbC). The FID has a uniform carbon response for all hydrocarbon type compounds. Consequently, a single response factor can be used to represent all these type compounds (Blades, 1976; Sternberg et al., 1962). Substituted hydrocarbons require the adjustment of the FID response using an effective carbon number (ECN) to correctly report compound concentration (Scalon and Willis, 1985; Kallai and Balla, 2002; Jorgensen et al., 1990). The limited ECN information available suggests that the sulfur atom in the form of alkyl sulfide has no effect on FID response (Jorgensen et al., 1990). Consequently, the RSCs of interest dimethyl sulfide (DMS; CH_3SCH_3), dimethyl disulfide (DMDS; $CH_3S_2CH_3$) and methyl mercaptan (CH_3SH) are reported with no correction to the observed response. Column retention times for the RSCs were determined using known mixtures of these compounds. Identification of the reported RSCs was confirmed by a gas chromatography–mass spectrometry (GC–MS) system. However, in this study, the GC–MS was not used to quantify compounds. For further information on the GC–MS, the reader is referred to Blunden et al. (2005).

The suitability of using SUMMA and FSL canisters to collect whole air samples for DMS, DMDS and CH_3SH analysis was assessed by conducting canister stability tests using prepared mixtures of these compounds in humidified zero air. The details of these tests

are provided in the Supplementary material. In summary, the results of these tests indicated that DMS and DMDS are stable in humidified SUMMA and FSL canisters for a period of at least three weeks. In contrast, the test results indicated that CH_3SH is extremely unstable in humidified SUMMA canisters, and therefore will not be measured in these canisters. In comparison to humidified SUMMA canisters, the recovery of CH_3SH in humidified FSL canisters was improved, indicating that CH_3SH may be measured at reduced concentrations in humidified FSL canisters.

Nine to eleven canister samples were taken from both the barn and lagoon in each measurement period over a minimum of four different days. Barn canister samples were collected from the barn fan exhaust (see Section 2.3.1. for additional details). Lagoon samples were collected just above the lagoon surface using a dynamic flow-through chamber system (see Section 2.3.2. for additional details). Barn and lagoon canister samples were collected over a period of ≈5 min at different times of the day (between 8:00 and 18:00 EST). A mixture of 6-L SUMMA and FSL canisters were used for sampling. Prior to sampling, the canisters were cleaned by a XonTech Model 960 canister cleaning system. The automated system performs a cycle of cleaning, where canisters are evacuated, filled with humidified air and then baked at 120 °C. The canisters were cleaned using 2 cycles. After the cleaning, the system evacuates the canisters to <0.05 mm Hg using a vacuum pump.

2.3. Barn, lagoon and environmental parameter measurements

2.3.1. Barn measurements

Barn measurements were made at one of the eight swine barns at the sampling site. The barn contained five fans, which turned on in a set sequence as the temperature increases inside the swine barn. Two of the fans were direct driven and three were belt driven. Barn emissions were determined as follows:

$$J = C^* \sum f \quad (1)$$

where J is the compound emission, C is the barn outlet concentration and $\sum f$ is the sum of the flow rates of each individual fan.

Barn concentration measurements were made by placing a sample line made of Teflon tubing (0.64 cm outer diameter, 0.4 cm inner diameter) directly in front of the first fan to turn on. During the continuous sampling of H_2S, background samples were collected upwind of the barns using FSL canisters and were instantly drawn in the H_2S/SO_2 analyzer. Concentrations were negligible in comparison to corresponding H_2S concentrations measured from the ventilation fan and therefore they were not considered during emission calculations. For other RSCs, background canister samples were taken upwind of the barn at simultaneous times to the barn sample collection. RSCs were not observed in background samples.

The flow rate for each individual fan was calculated as follows:

$$FFR_c = FFR_m \times \frac{RPM_{meas}}{RPM_{spec}} \quad (2)$$

where FFR_c is the calculated fan flow rate, FFR_m is the manufacturers fan flow rate, RPM_{meas} is the measured revolutions per minute and RPM_{spec} is the specified revolutions per minute. The specified revolutions per minute were based on manufactures specifications. Measured revolutions per minute (rpm) were calculated by attaching Mabuchi VDC motors (Santa Clara, CA) to the fans. Measurements of the static pressure difference between the inside and outside of the barn were used to adjust the manufacturers fan flow rate. For additional information on the use of the motors to determine fan rpm and quality assurance/quality control (QA/QC) associated with this methodology the reader is referred to Rumsey et al. (2012). Rumsey et al. (2012) also provides further details on the static pressure difference measurements.

Table 1
Seasonal barn concentrations, ventilation rates, emissions, and corresponding environmental parameters for H_2S.

Season	Concentration (ppb)	Ventilation rate (m^3 min^{-1})	Emissions (g day^{-1})	Barn temperature (°C)	Ambient temperature (°C)
Summer	72,[a] 73[b] (43)[c] N[d] = 518	1763 (691) N = 518	189 (42) N = 518	27.9 (2.7) N = 518	26.0 (4.1) N = 518
Fall	327, 307 (158) N = 741	327 (180) N = 741	206 (89) N = 741	19.9 (2.4) N = 740	8.4 (5.2) N = 741
Winter	164, 150 (63) N = 507	262 (174) N = 507	80 (54) N = 507	18.4 (3.8) N = 507	11.3 (6.2) N = 507
Spring	631,[e] 645 (240) N = 649	601 (321) N = 649	647 (219) N = 649	26.5 (1.5) N = 630	19.0 (4.2) N = 632

[a] Mean value.
[b] Average daily mean value.
[c] ±1 standard deviation.
[d] N represents the number of 15 min averaged data points.
[e] 173 (27%) of the 15 min averaged data points had at least one minute average above the limit of detection of the analyzer.

2.3.2. Lagoon measurements

A dynamic flow-through chamber system was used to determine anaerobic lagoon fluxes (Rumsey and Aneja, 2014; Rumsey et al., 2012; Blunden and Aneja, 2008). A description of the H_2S lagoon flux measurement methodology is provided in Rumsey and Aneja (2014). The same methodology was applied to the collection of lagoon canister samples, with the exception that the flow rate for the samples ranged from 4 to 6 L min^{-1}. Further information on the lagoon canister collection is provided in Rumsey et al. (2012).

2.3.3. Environmental parameter measurements

During both lagoon and barn sampling, ambient relative humidity, air temperature and solar radiation measurements were made at a height of 2 m. Ambient Wind speed and wind direction measurements were made at a height of 10 m. During lagoon flux measurements, lagoon temperature and pH were monitored at a depth of ≈7 cm. During barn sampling, temperature was measured at the fan outlet. Additional information on the instruments used to make the environmental parameter measurements is provided in Rumsey et al. (2012).

3. Results and discussion

3.1. RSCs emissions

The RSCs, DMS and DMDS were identified in almost every barn and lagoon sample. Therefore, DMS and DMDS concentrations and emissions were further analyzed in addition to H_2S. CH_3SH was not identified in any lagoon or barn samples, despite analyzing FSL canisters within an appropriate time period. Therefore, it is hypothesized that a combination of low concentrations and instability, resulted in CH_3SH not being detected. The RSC, dimethyl trisulfide ($CH_3S_3CH_3$) was occasionally observed in lagoon and barn samples. However, the compound concentration was near the detection limit of the GC-FID system and was therefore not selected for further analysis. The GC–MS additionally identified carbon disulfide (CS_2) in some canister samples; however, as mentioned,

quantitative measurement could not be determined with the GC–MS system.

3.1.1. Barn concentrations and emissions

3.1.1.1. Seasonal H_2S concentrations and emissions. The seasonal H_2S barn concentrations and emissions, as well as ventilation rates, and the environmental parameters, barn temperature and ambient temperature are presented in Table 1. The highest seasonal concentration of 631 ppb occurred in the spring sampling season. However, in this season, 173 of the 649 (27%) 15 min averaged H_2S concentrations had concentrations above the maximum range of the H_2S analyzer (1000 ppb). These H_2S concentrations were set to the value of 1000 ppb, therefore the actual average concentration is higher than the reported value. For the remainder of the manuscript, the reader should take this into account when spring concentration and emission values are discussed. The spring seasonal concentration was almost twice as high as the next highest seasonal concentration, which was 327 ppb for the fall season. The two lowest seasonal concentrations were in the winter (164 ppb) and summer (72 ppb). H_2S concentration is expected to be influenced by ventilation rate. This relationship was investigated by using the coefficient of determination (r^2). Log H_2S concentration was found to have a fairly strong negative relationship with ventilation rate ($r^2 = 0.45$, $p < 0.0001$).

H_2S emissions (Table 1) ranged from 80 g day^{-1} (winter) to 647 g day^{-1} (spring). Total animal weight is considered to be one of the largest factors influencing emissions from a barn. Emissions were therefore normalized by 500 kg of live animal weight (LAW), also known as 1 AU (animal unit). The calculated LAW, the corresponding pig production data and the normalized H_2S emissions are shown in Table 2. After taking into account the LAW, there is still considerable variance in the emission rate. The highest seasonal H_2S normalized emission rate occurs in the spring with an emission of 7.3 g day^{-1} AU^{-1}. The next highest is the fall season with an emission of 3.0 g day^{-1} AU^{-1}, followed by the summer season with an emission of 2.2 g day^{-1} AU^{-1}. The lowest normalized emission rate was in the winter with an emission of 0.7 g day^{-1} AU^{-1}. The overall average normalized emission rate (average of seasonal normalized emission rates) is 3.3 g day^{-1} AU^{-1}. Analysis of the diurnal trends of H_2S emissions as well as H_2S concentrations and ventilation rates are provided in the Supplementary material.

In comparison to the Blunden et al. (2008) study, which determined barn emissions at the same sampling site as this study, the magnitude of the emissions are fairly similar. In the Blunden et al. (2008) study, reported emissions ranged from 1.2 g day^{-1} AU^{-1} in the summer to 4.2 g day^{-1} AU^{-1} in the winter with an overall average normalized emission rate of 2.6 g day^{-1} AU^{-1}. The variance in concentrations and emissions in this study and in the Blunden et al. (2008) study are the result of the influence of a number of different factors. Manure properties such as manure temperature and manure pH can influence emissions as well as the speed of the air movement across the manure surface, which is controlled by the ventilation rate. In this study, barn temperature which is likely related to manure surface temperature was measured. However, it was beyond the scope of this study to measure manure temperature as well as manure pH and the speed of the air movement across the manure surface. Analysis of the diurnal trends of H_2S emissions, H_2S concentrations and ventilation rates (provided in the Supplementary material) suggests that barn temperature could be a significant factor in influencing emissions. Statistical analysis of the influence of barn temperature on H_2S emissions as well as on DMS and DMDS emissions is provided in Section 3.1.1.3. As mentioned, ventilation rate influences the speed of air movement across the manure surface. It is hypothesized that the low winter emissions (0.7 g day^{-1} AU^{-1}) observed in this study were influenced by a combination of ambient temperature and air flow over the manure. During the winter sampling season, the barn temperature was low, which resulted in one fan only working intermittently for 54% of the sampling period. This results in less air movement over the manure surface, thus resulting in lower manure emissions.

Table 3 presents the concentrations and emissions of H_2S from other swine finishing CAFO studies. Studies have been selected that made H_2S emission measurements continuously or semi-continuously and reported emissions normalized for LAW. The exception was the Zhu et al. (2000) study, which did not report emissions normalized for LAW, however the numbers of pigs and their average weight during the sampling period was provided in the manuscript, which allowed the LAW to be calculated. Of these previous swine CAFO finishing studies, two were conducted in the Midwest of U.S.A (Zhu et al., 2000; Ni et al., 2002), one in South Korea (Kim et al., 2008) and one in North Carolina at the same sampling site as this study (Blunden et al., 2008). The swine CAFO average daily mean (ADM) concentrations and emissions in this study are of a similar magnitude to other swine CAFO studies, which range from 47 to 632 ppb and 1.2 to 8.5 g H_2S day^{-1} AU^{-1}, respectively (Table 3). The variance of emissions observed between studies in different locations (Table 3) occurs as a result of different management and environmental conditions.

3.1.1.2. Seasonal DMS and DMDS concentrations and emissions. Seasonal DMS and DMDS concentrations, ventilation rates, emissions, normalized emissions (emissions normalized for LAW using the information provided in Table 2) and corresponding environmental parameters are presented in Table 4. DMS and DMDS concentrations ranged from 0.18 to 0.89 ppb and 0.47 to 1.02 ppb, respectively. The highest seasonal concentration occurred in the fall season for both compounds. The lowest seasonal concentration for both DMS and DMDS occurred in the summer season. In comparison, DMS seasonal concentrations were higher than DMDS seasonal concentrations in the winter and spring, whereas DMDS seasonal concentrations were higher than DMS seasonal concentrations in the summer and fall. The highest DMS sample concentration was 2.09 ppb, which occurred in the fall season. The highest DMDS sample concentration occurred in the spring season with a value of 1.69 ppb. In comparison to H_2S concentrations, DMS and DMDS concentrations were 2–3 orders of magnitude lower.

Table 2
Seasonal pig production information and the calculated normalized H_2S emission rate.

Sampling Season	Number of pigs	Number of weeks in rotation	Average weight (kg)	Live animal weight (LAW) (kg)	Normalized H_2S emission rate (g day^{-1} AU^{-1})
Summer	884.5	7–8	48.7	43,049	2.2 (0.5)[a]
Fall	994.5	4–5	34.6	34,428	3.0 (1.3)
Winter	476[b]	20–21	116.6	55,513	0.7 (0.5)
Spring	874.5	8–9	50.6	44,262	7.3 (2.5)

[a] ±1 standard deviation.
[b] Sampling occurred at end of rotation, when some pigs had been sold.

Table 3
H_2S barn concentrations and emissions from this study and previous swine finishing CAFO studies.

Reference	Location of study	Ventilation type[d]	Manure collection system	Month	ADM Conc (ppb)[e]	Live animal weight (kg)	Emission rate (g day^{-1} AU^{-1})
Ni et al. (2002)	IL[a]	MV	Deep pit	Jun–Sep	173	48,783	8.3
Zhu et al. (2000)	Midwest[b]	MV	Deep pit	Sep	414	44,990	2.0[g]
Zhu et al. (2000)	Midwest	NV	Deep pit	Sep	271	43,640	3.3
Kim et al. (2008)	S. Korea	NV	Deep pit	May–Jun & Sep–Oct	296[f]	–	6.7[h]
Kim et al. (2008)	S. Korea	MV	Deep pit	May–Jun & Sep–Oct	613	–	8.5
Kim et al. (2008)	S. Korea	NV	Scraper removal	May–Jun & Sep–Oct	115	–	5.8
Kim et al. (2008)	S. Korea	MV	Scraper removal	May–Jun & Sep–Oct	270	–	6.3
Kim et al. (2008)	S. Korea	NV	Deep bedded	May–Jun & Sep–Oct	138	–	3.0
Blunden et al. (2008)	NC[c]	MV	Shallow pit	Feb	632	48,963	4.2
Blunden et al. (2008)	NC	MV	Shallow pit	Apr	441	73,895	3.3
Blunden et al. (2008)	NC	MV	Shallow pit	Jun	47	33,952	1.2
Blunden et al. (2008)	NC	MV	Shallow pit	Oct	304	38,390	1.7
This study	NC	MV	Shallow pit	Jun	73	43,049	2.2
This study	NC	MV	Shallow pit	Nov	307	34,428	3.0
This study	NC	MV	Shallow pit	Feb	150	55,513	0.7
This study	NC	MV	Shallow pit	Apr	645	44,262	7.3

[a] IL = Illinois, U.S.A.
[b] Location is assumed to be Midwest of U.S.A. Location is not specified in paper.
[c] NC = North Carolina, U.S.A.
[d] MV = mechanically ventilated, NV = naturally ventilated.
[e] ADM Conc = Average daily mean concentration.
[f] Concentrations presented from Kim et al. (2008) are average concentrations.
[g] Emissions reported in the Zhu et al. (2000) study were not normalized by live animal weight. However, the Zhu et al. (2000) study reported the number of pigs and the average body weight during the sampling period, therefore the reported emissions for the Zhu et al. (2000) study were normalized for live animal weight using these values.
[h] Emission units from Kim et al. (2008) were converted from 75 kg of live animal weight.

DMS and DMDS concentrations reported in this study are similar to concentrations reported for barn ventilation exhaust air and barn room air by previous swine CAFO studies, which range from ≈0 (i.e. not detected) to 13.8 ppb for DMS and ≈0 (i.e. not detected) to 4.7 ppb for DMDS (Clanton and Schmidt, 2000; Trabue et al., 2008; Blunden et al., 2005; Kim et al., 2007; Feilberg et al., 2010; Hansen et al., 2012; Yao et al., 2011; Schiffman et al., 2001). Overall, taking into account the variations in production, management and environmental conditions, the measured concentrations in this study compare well to the previous swine CAFO studies.

Observed DMS and DMDS concentrations are expected to be influenced by ventilation rate. The relationship between these RSCs and ventilation rate was investigated by using the coefficient of determination (r^2). Both DMS ($r^2 = 0.47$, $p < 0.001$) and DMDS ($r^2 = 0.22$, $p = 0.003$) were found to have significant negative relationships with ventilation rate.

The seasonal DMDS emission rates were higher for all four seasons in comparison with DMS (Table 4). DMDS highest emission rate was in the summer (4.25 g day^{-1}). The lowest DMDS emission rate was in the winter with a value of 1.41 g day^{-1}. DMS seasonal emissions ranged from 0.90 g day^{-1} (summer) to 2.19 g day^{-1} (fall). Seasonal barn emission rates for DMS and DMDS were two to three orders of magnitude lower than H_2S barn emission rates.

Similar to H_2S emissions, DMS and DMDS seasonal emissions were normalized by 500 kg of LAW (see Table 2 for LAW calculation). Normalized seasonal emissions for DMS and DMDS ranged from 0.010 to 0.032 and 0.013 to 0.061 g day^{-1} AU^{-1}, respectively (Table 4). Both compounds had their highest normalized seasonal emissions in the fall and their lowest in the winter. The overall average normalized barn emission (average of seasonal normalized emission rates) for DMDS (0.036 g day^{-1} AU^{-1}) is approximately twice as high as the emission for DMS (0.017 g day^{-1} AU^{-1}). DMS and DMDS normalized seasonal emissions are significantly lower

Table 4
Seasonal barn concentrations, ventilation rates, emissions and corresponding environmental parameters for DMS and DMDS.

Season	Concentration (ppb)		Ventilation rate (m^3 min^{-1})	Emissions (g day^{-1}/g day^{-1} AU^{-1})		Barn temperature (°C)	Ambient temperature (°C)
	DMS	DMDS		DMS	DMDS		
Summer	0.18[a]	0.47	2040	0.90[d] (0.90)	4.25 (2.38)	30.27	31.15
	(0.22)[b]	(0.39)	(589)	0.010[e] (0.01)	0.050 (0.03)	(3.18)	(2.48)
	n[c] = 10	$n = 10$	$n = 10$	$n = 10$	$n = 10$	$n = 10$	$n = 10$
Fall	0.89	1.02	725	2.19 (1.28)	4.19 (2.03)	16.42	23.70
	(0.61)	(0.34)	(289)	0.032 (0.02)	0.061 (0.03)	(3.67)	(1.81)
	$n = 10$	$n = 10$	$n = 10$	$n = 10$	$n = 10$	$n = 10$	$n = 10$
Winter	0.84	0.66	435	1.10 (0.50)	1.41 (0.98)	14.05	19.78
	(0.41)	(0.32)	(296)	0.010 (0.01)	0.013 (0.01)	(4.15)	(2.78)
	$n = 9$	$n = 9$	$n = 9$	$n = 9$	$n = 9$	$n = 9$	$n = 9$
Spring	0.57	0.50	850	1.56 (0.51)	1.90 (1.31)	22.42	27.61
	(0.25)	(0.51)	(366)	0.018 (0.01)	0.021 (0.02)	(3.47)	(1.62)
	$n = 9$	$n = 9$	$n = 9$	$n = 9$	$n = 9$	$n = 9$	$n = 9$

[a] Mean value.
[b] ±1 standard deviation.
[c] n is the number of canister samples.
[d] Emissions in units of g day^{-1}.
[e] Normalized emissions in units of g day^{-1} AU^{-1}.

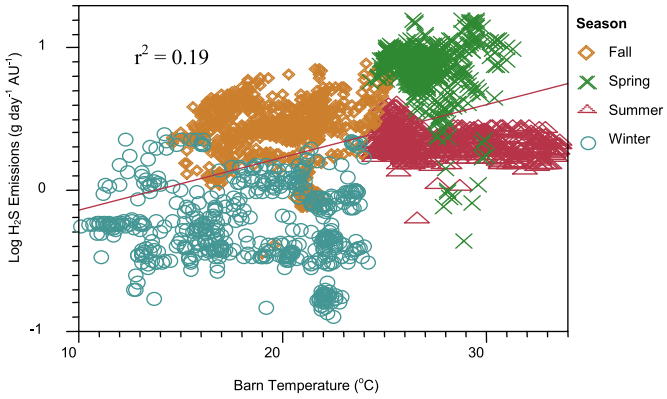

Fig. 1. The relationship between log H_2S emissions (g day^{-1} AU^{-1}) and barn temperature (°C). Data points (n = 2222) represent 15 min averages.

than those for H_2S, with the overall average DMS and DMDS emissions approximately two orders of magnitude lower than the overall average H_2S emission (3.3 g day^{-1} AU^{-1}).

Seasonal normalized DMS and DMDS emissions in this study are an order of magnitude lower than a previous swine CAFO study that also reported emissions normalized for animal weight (Kim et al., 2007). The Kim et al. (2007) study was conducted in South Korea and determined normalized emissions of DMS and DMDS for five different types of pig production stages, gestation, farrow, nursery, grow and finish. Normalized emissions in the Kim et al. (2007) study ranged from 0.22 to 0.94 g day^{-1} AU^{-1} for DMS and 0.12 to 0.53 g day^{-1} AU^{-1} for DMDS over the five different pig production stages. It is expected that the difference in emissions between the studies is caused by the different production, management and environmental conditions.

3.1.1.3. The influence of barn temperature on barn emissions. As mentioned in Section 3.1.1.1, analysis of the diurnal trends of H_2S emissions, H_2S concentrations and ventilation rates (see Supplementary material) suggest that barn temperature could be a significant factor in influencing emissions, with increases in barn temperature resulting in increased emissions. The explanation for this phenomenon is that increasing barn temperatures will increase the manure temperature, which in turn can increase the mass transfer of H_2S across the manure surface due to the effect of increasing temperature on H_2S diffusivity and solubility (Arogo et al., 1999).

The influence of barn temperature on RSC barn emissions was investigated using linear regression. H_2S barn emissions were found to have a significant, but weak positive correlation with barn temperature ($r^2 = 0.19$, $p < 0.0001$) (Fig. 1). There was no significant relationship between DMS and DMDS emissions and barn temperature. The r^2 values were 0.05 ($p = 0.1607$) and 0.05 ($p = 0.1538$), for DMS and DMDS, respectively. The relationships might be stronger if the emissions were correlated with manure temperature. However, it was beyond the scope of the study to measure this environmental parameter.

A major reason that there is a weak relationship between barn temperature and RSCs emissions is due to the dynamic barn environment. The ventilation rate of a barn controls the amount of gas emissions exiting the barn. Therefore the varying ventilation rate will in combination with the waste emission rate influence the barn exhaust emissions, thus making it difficult to determine the influence of barn temperature and other environmental factors on barn emissions.

3.1.2. Lagoon fluxes

H_2S, DMS and DMDS seasonal lagoon fluxes and emissions as well as the environmental parameters during the canister sampling are presented in Table 5. As mentioned, only a brief summary of H_2S lagoon fluxes from the sampling campaign is provided in this manuscript. However, for an in-depth analysis and discussion of H_2S lagoon fluxes with respect to diurnal and seasonal variations as well as environmental factors, the reader is referred to Rumsey and Aneja (2014). Rumsey and Aneja (2014) also compare measured H_2S lagoon fluxes to other previous swine CAFO lagoon studies.

Seasonal H_2S lagoon fluxes are higher than DMS and DMDS seasonal fluxes in all seasons, particularly in the summer season, where they are over an order of magnitude higher. The overall average (average of seasonal fluxes) H_2S flux of 1.33 μg m^{-2} min^{-1} is

Table 5
Seasonal H_2S, DMS and DMDS lagoon fluxes and emissions, and the environmental parameters during canister sampling.

Season	Flux (μg m^{-2} min^{-1})/Emission (g day^{-1} AU^{-1})			Lagoon temperature (°C)	Lagoon pH	Wind speed (m s^{-1})	Air temperature (°C)
	H_2S[a]	DMS	DMDS				
Summer	3.81[b]	0.26	0.22	27.0	7.33	2.23	26.7
	(3.24)[c]	(0.08)	(0.04)	(3.34)	(0.16)	(0.87)	(3.32)
	N = 705[d]	n = 10[f]	n = 8	n = 10	n = 5	n = 10	n = 10
	0.11[e]	0.0073	0.0063				
Fall	1.17	0.11	0.04	23.3	7.63	3.07	25.5
	(1.62)	(0.08)	(0.01)	(1.67)	(0.16)	(2.03)	(3.01)
	N = 646	n = 10	n = 10	n = 10	n = 10	n = 10	n = 10
	0.033	0.0031	0.0011				
Winter	0.08	0.05	0.02	12.3	8.08	2.38	8.59
	(0.09)	(0.04)	(0.02)	(2.50)	(0.11)	(0.77)	(1.23)
	N = 631	n = 11	n = 11	n = 11	n = 11	n = 11	n = 11
	0.0023	0.0014	0.00071				
Spring	0.27	0.06	0.11	19.6	8.03	4.37	18.3
	(1.71)	(0.03)	(0.03)	(1.77)	(0.07)	(1.80)	(6.24)
	N = 478	n = 10	n = 10	n = 10	n = 10	n = 10	n = 10
	0.0081	0.0017	0.0033				

[a] H_2S flux data from Rumsey and Aneja (2014).
[b] Mean flux value.
[c] ±1 standard deviation.
[d] N represents the number of 15 min averaged data points.
[e] Emission value.
[f] n is the number of canister samples.

over an order of magnitude higher than the fluxes for DMS (0.12 μg m^{-2} min^{-1}) and DMDS (0.09 μg m^{-2} min^{-1}). In comparison to DMDS, DMS fluxes were slightly higher in all seasons apart from spring. DMS and DMDS seasonal fluxes are both highest in the summer sampling season with values of 0.26 μg m^{-2} min^{-1} and 0.22 μg m^{-2} min^{-1}, respectively. DMS seasonal flux is 2nd highest in the fall and 3rd highest in the spring. This trend is reversed for DMDS seasonal fluxes. DMS and DMDS have their lowest fluxes in winter with values of 0.05 and 0.02 μg m^{-2} min^{-1}, respectively. Lagoon flux values were converted into lagoon emission values in units of g day^{-1} AU^{-1} based on lagoon area and the average of the total live animal weight of all eight barns in each sampling period. H_2S seasonal emissions ranged from 0.0023 g day^{-1} AU^{-1} to 0.11 g day^{-1} AU^{-1} with an overall average of 0.038 g day^{-1} AU^{-1}. DMS and DMDS emissions ranged from 0.0014 g day^{-1} AU^{-1} to 0.0073 g day^{-1} AU^{-1} and 0.00071 g day^{-1} AU^{-1} to 0.0063 g day^{-1} AU^{-1}, respectively. The overall average DMS emission was 0.0034 g day^{-1} AU^{-1}, which was slightly higher than the DMDS emission value of 0.0028 g day^{-1} AU^{-1}.

A previous swine CAFO lagoon study by Schiffman et al. (2001) identified DMS and DMDS in three lagoon waste samples from different swine CAFOs in North Carolina. However, the Schiffman et al. (2001) study did not determine lagoon emissions.

3.1.2.1. The influence of environmental parameters on RSC lagoon fluxes. The influence of lagoon temperature and lagoon pH on DMS and DMDS fluxes was investigated using linear regression. For lagoon temperature, there were significant (p-value <0.01) positive relationships for both compounds (Fig. 2a and b), indicating that as lagoon temperature increases, DMS and DMDS fluxes increase. Lagoon temperature also had a significant positive relationship with H_2S flux (Rumsey and Aneja, 2014). The explanation for the relationship between lagoon temperature and H_2S lagoon fluxes is the same as for the influence of barn manure temperature on H_2S barn emissions (Section 3.1.1.3). It is hypothesized that increasing lagoon temperature also increases DMS and DMDS emissions across the lagoon–air interface in a similar way.

For lagoon pH, there were significant (p-value <0.01) negative relationships for DMS and DMDS (Fig. 3a and b), indicating that as pH increases, DMS and DMDS fluxes decrease. Lagoon pH also had a significant negative relationship with H_2S flux (Rumsey and Aneja, 2014). There are no known studies discussing the aqueous dissociation of DMS and DMDS. The influence of pH on H_2S is supported by the aqueous dissociation of H_2S described in literature (Rumsey and Aneja, 2014).

3.2. Odorous emissions

RSCs typically have unpleasant odors and low odor thresholds, therefore their emissions are of concern locally. The potential local environmental impact of RSC emissions was assessed by comparing measured barn concentrations to their odor threshold. A range of odor thresholds for H_2S have been determined, 4.5 ppb (American Industrial Hygiene Association, 1989), 8.1 ppb (Devos et al., 1990), and 17.8 ppb (Amoore and Hautala, 1983). For this study the average of these three H_2S odor thresholds was used (10.13 ppb). H_2S has an odor characteristic described as rotten eggs (Schiffman et al., 2001). DMS has an odor threshold of 2.24 ppb (Devos et al., 1990) with an odor characteristic described as stench (Schiffman et al., 2001). DMDS has an odor threshold of 12.3 ppb (Devos et al., 1990) with an

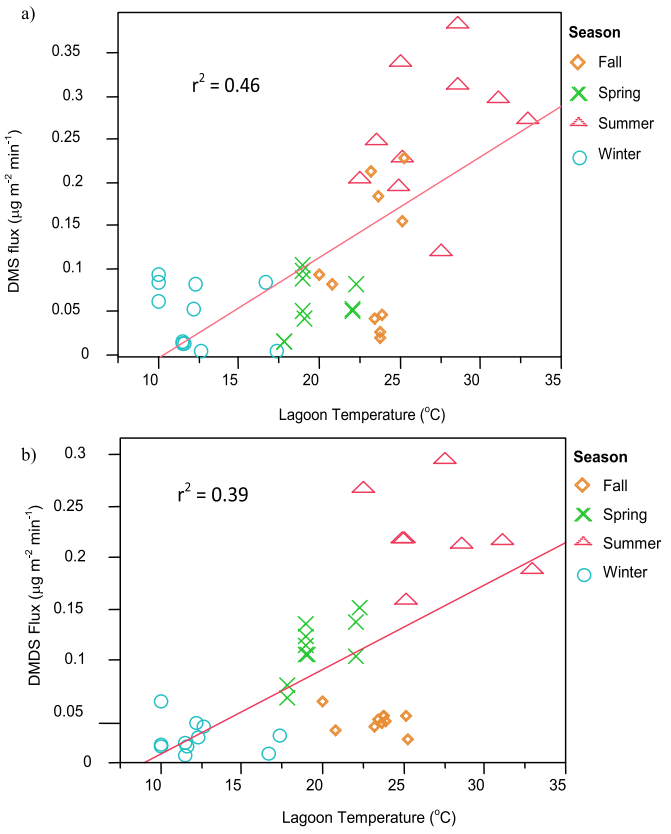

Fig. 2. Influence of lagoon temperature (°C) on a) DMS flux (μg m^{-2} min^{-1}), b) DMS flux (μg m^{-2} min^{-1}).

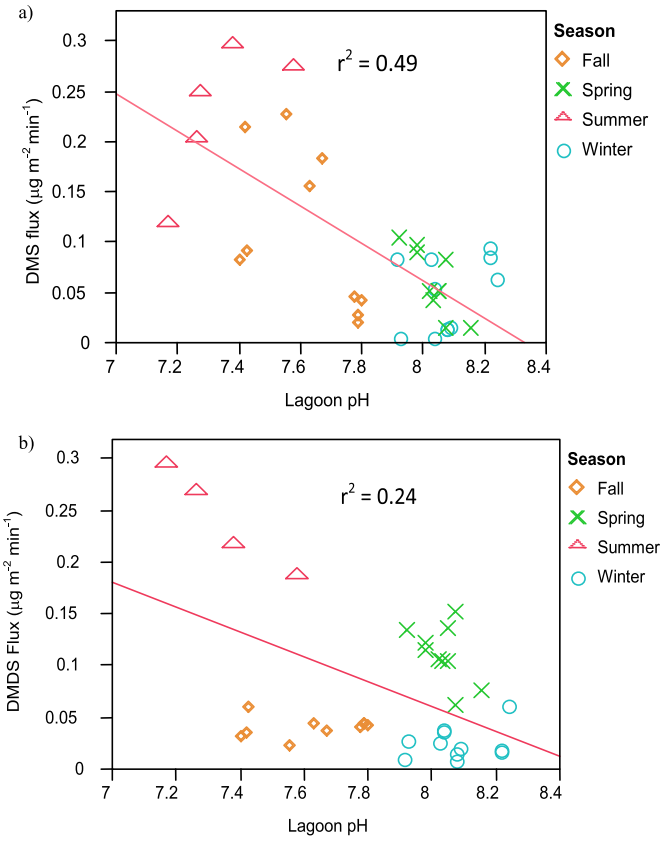

Fig. 3. Influence of lagoon pH on a) DMS flux (μg m^{-2} min^{-1}), b) DMS flux (μg m^{-2} min^{-1}).

odor characteristic described as putrid garlic (Schiffman et al., 2001).

All 15 min average H_2S barn concentrations were higher than the average odor threshold of 10.13 ppb. An examination of the seasonal barn concentrations and their standard deviation (Table 1) gives an approximate assessment of how high observed concentrations were in comparison to their odor threshold. A further assessment of the extent to which H_2S concentrations were greater than their odor threshold was conducted by comparing 15 min average concentrations to a value (101.3 ppb) which was an order of magnitude higher than the average of the three odor thresholds. In spring, 97% of 15 min average concentrations were above 101.3 ppb. Fall and winter were next highest with 93% and 79% of 15 min average concentrations above 101.3 ppb. The season with the lowest percentage of 15 min average concentrations above 101.3 ppb was summer with 27%. Over all four seasons, 77% of 15 min average concentrations were above 101.3 ppb.

DMS barn concentrations did not exceed their odor threshold (2.24 ppb), with the highest sample concentration (2.09 ppb) just below the odor threshold. Similarly, DMDS barn concentrations did not exceed their odor threshold of 12.3 ppb. The highest DMDS sample concentration was 1.69 ppb.

The lagoon is also a source of H_2S, DMS and DMDS and therefore contributes to odorous emissions. H_2S, DMS and DMDS lagoon fluxes are provided in Section 3.1.2 in this manuscript.

3.3. North Carolina RSC emissions

An estimate of RSCs swine CAFO emissions for North Carolina was calculated by estimating North Carolina barn and lagoon emissions. North Carolina barn and lagoon emissions were calculated by applying measured emission values to an estimation of the number and weight of swine in North Carolina. The number and weight of swine in North Carolina was estimated using the methodology described in Rumsey et al. (2012). A summary of the swine numbers and weight provided by this methodology is presented in a table in the Supplemental material. The overall average H_2S emissions determined in this study for the barn (3.3 g day^{-1} AU^{-1}) and lagoon (0.038 g day^{-1} AU^{-1}) were applied to all swine weight classes apart from breeding. To estimate H_2S emissions for the swine population classified as breeding, emission values were determined from previous barn and lagoon emission measurement studies, which measured H_2S barn emissions (Rahman and Newman, 2012) and lagoon emissions (Grant et al., 2013) from swine breeding operations. Emission values were determined as 1.56 g day^{-1} AU^{-1} and 0.10 g day^{-1} AU^{-1} for the barn and lagoon, respectively. A description of the methodology for the determination of these emission values is provided in the Supplemental material. There are no known studies that have reported DMS and DMDS animal weight normalized emissions from a swine breeding operation in the Unites States. Therefore, North Carolina swine CAFO DMS and DMDS emissions were estimated using two different approaches. Firstly, by calculating emissions from all swine except those associated with breeding operations and secondly by applying the emission values to all swine including those associated with breeding operations.

North Carolina barns were estimated to emit 1.20*10^6 kg yr^{-1} of H_2S. North Carolina lagoon emissions were over an order of magnitude lower with an emission of 23,692 kg yr^{-1}. Total (barn + lagoon) emissions from North Carolina swine CAFOs were 1.22*10^6 kg yr^{-1} for H_2S. North Carolina H_2S barn emissions contributed ≈98% of total (lagoon + barn) North Carolina emissions. Estimating North Carolina H_2S swine CAFO emissions using the previous studies by Blunden et al. (2008) and Blunden and Aneja (2008), which made measurements of barn and lagoon emissions at the same sampling site as this study produced similar North Carolina emission values to this study (see Supplemental material). Statewide estimates of DMS and DMDS emissions were significantly lower. For all swine except the breeding operations, barn emissions were 5169 kg yr^{-1} and 10,705 kg yr^{-1} for DMS and DMDS, respectively. For all swine, barn emissions were 7617 kg yr^{-1} and 15,775 kg yr^{-1} for DMS and DMDS, respectively. RSC lagoon emissions for all swine except the breeding operations were estimated as 1002 kg yr^{-1} for DMS and 845 kg yr^{-1} for DMDS. For all swine, lagoon emissions were 1477 kg yr^{-1} for DMS and 1245 kg yr^{-1} for DMDS. DMS total (barn + lagoon) emissions from North Carolina were 6171 kg yr^{-1} for all swine except the breeding operations and 9093 kg yr^{-1} for all swine. DMDS total (barn + lagoon) emissions from North Carolina were 11,550 kg yr^{-1} for all swine except the breeding operations and 17,020 kg yr^{-1} for all swine. For DMS and DMDS, North Carolina barn emissions were not as dominant as for H_2S, with lagoon emissions contributing ≈14% and ≈7% of total emissions, respectively.

The North Carolina Division of Air Quality (NCDAQ) released an North Carolina H_2S emission inventory for 2002 stating total emissions of 5.40*10^6 kg yr^{-1} (NCDAQ, 2003). However, this inventory did not include emissions from animal operations. By adding the contribution of the swine CAFO emissions (using the H_2S emissions determined from measurements in this study) to the inventory, it is estimated that H_2S emissions from swine CAFOs in North Carolina comprise ≈18% of statewide H_2S emissions.

4. Conclusions

Measurements of RSCs emissions were made over four seasonal sampling periods from a barn and anaerobic lagoon at a swine CAFO in North Carolina. These emissions were evaluated with respect to seasonal variations and environmental factors. Overall average barn emissions for H_2S were 3.3 g day^{-1} AU^{-1}, which was approximately two orders of magnitude higher than DMS and DMDS barn emissions, which were 0.017 g day^{-1} AU^{-1} and 0.036 g day^{-1} AU^{-1}, respectively. Overall average H_2S lagoon emissions were over an order of magnitude higher than DMS and DMDS lagoon emissions, 0.038 g day^{-1} AU^{-1} compared to 0.0034 g day^{-1} AU^{-1} and 0.0028 g day^{-1} AU^{-1}, respectively.

The potential local environment impact of RSC swine CAFO emissions was evaluated by comparing RSC barn exhaust concentrations to their odor threshold. 77% of measured H_2S barn exhaust concentrations were an order of magnitude above their average odor threshold (101.3 ppb). To further assess the potential effects of the H_2S concentrations, the dispersion of H_2S from the swine CAFO should be modeled to predict fence-line/property boundary concentrations.

Barn and lagoon emission values from this study as well as from previous studies that had made emission measurements at swine breeding operations (Rahman and Newman, 2012; Grant et al., 2013) were used to estimate RSCs swine CAFO emissions for North Carolina. The total (barn + lagoon) emissions were 1.22*10^6 kg yr^{-1} for H_2S, which is estimated to be ≈18% of statewide H_2S emissions. It is estimated that North Carolina H_2S barn emissions contribute ≈98% of the total (barn + lagoon) swine CAFO emissions. In comparison to H_2S, total (barn + lagoon) North Carolina swine CAFO emissions for DMS and DMDS were 1–2 orders of magnitude lower.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.05.041.

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