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Research Paper

Spatial variability of mixing ratios of ammonia and tracer gases in a naturally ventilated dairy cow barn

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The use of the tracer gas ratio method to estimate emissions from naturally ventilated (NV) livestock barns excludes the need of monitoring ventilation rates. However, it requires accurate measurement of tracer release rate (Q_T) and a representative estimate of the mixing ratio between pollutant (P) and tracer (T) gases($\overline{[P]/[T]}$). While the quality of Q_T simply depends on using an accurate commercial mass flow controller, determination of a representative mixing ratio $\overline{[P]/[T]}$ is not trivial, since the NV livestock barn airspace presents complex movements that might be dependent on spatial vertical and cross horizontal dimensions. The goal was to assess the spatial variability of concentrations of the artificial tracer gas sulphur hexafluoride (SF_6), the metabolic carbon dioxide (CO_2) and the pollutant ammonia (NH_3), along with their mixing ratios ($[NH_3]/[CO_2]$, $[NH_3]/[SF_6]$, $[CO_2]/[SF_6]$), inside a NV dairy cow barn. The results indicated that the vertical variability of the calculated mixing ratios became more stable with increase in height, reaching approximately constant values above the animal occupied zone. Using both the metabolic CO_2 and the artificially injected SF_6 as tracer gases led to a homogeneous spread in behaviour of mixing ratios along V and HC directions. Finally, the possibility of finding a zone within the barn airspace where mixing ratios are considered to be representative for the whole barn, and the implications of applying artificial or metabolic tracers are discussed.

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Nomenclature

NV	Natural ventilation
T	Tracer gas of interest
[T]	Gaseous concentration of a tracer gas
Q_T	Release rate of a tracer gas T
AER	Air exchange rate
P	Pollutant gas of interest
[P]	Gaseous concentration of a pollutant gas
Q_P	Emission rate of a pollutant gas P
[P]/[T]	Mixing ratio between pollutant and tracer gases, both corrected for background
$\overline{[P]/[T]}$	Mean mixing ratio between pollutant and tracer gases, both corrected for background
$[NH_3]/[CO_2]$, $[NH_3]/[SF_6]$ and $[CO_2]/[SF_6]$	Mixing ratios of ammonia and carbon dioxide, ammonia and sulphur hexafluoride and carbon dioxide and sulphur hexafluoride, respectively. All concentrations corrected for background
L	Length dimension
W	Width dimension
H	Height dimension
V	Vertical direction within the barn
HC	Cross barn horizontal direction
PTFF	Polytetrafluoroethylene
ANOVA	Analysis of variance
GLM	General linear model
Y_{ij}	Measured pollutant or tracer gaseous concentration, or ratio between pollutant and tracer
Y	Gand mean concentration of pollutant or tracer gas, or ratio between pollutant and tracer throughout the experiment
V_i	Statistical effect of the vertical direction of the barn on concentrations of pollutant or tracer gases, or ratio between pollutant and tracer
HC_j	Statistical effect of the cross horizontal barn dimension on concentrations of pollutant or tracer gases, or ratio between pollutant and tracer
ε_{ij}	Independent normally distributed homogeneous random error
AOZ	Animal occupied zone

1. Introduction

Ammonia (NH_3) emissions from animal confinements have been the focus of research around the world for many years. The most important agricultural sources of pollutants are cattle housing systems (Bouwman et al., 1997; Dentener & Crutzen, 1994; Erisman, Bleeker, Galloway, Sutton, 2007; Ferm, 1998; Galloway & Cowling, 2002; Groot Koerkamp et al., 1998), where dairy cattle housing systems are mainly naturally ventilated (NV).

It is widely acknowledged that the quantification of emissions from NV buildings is a more complicated and challenging task than the quantification of emissions from mechanically ventilated buildings, given the difficulties that exist to accurately determine airflow rates (Scholtens, Dore, Jones, Lee, Philips, 2004). Considering the importance of NV buildings for cattle and other animal categories in many climate zones, a thorough understanding of their emission characteristics and potential mitigation options is highly relevant and requires a stronger methodological basis than is currently available (Ogink, Mosquera, Calvet, & Zhang, 2013).

According to Calvet et al. (2013), Ogink et al. (2013) and Takai et al. (2013), the tracer gas method has been considered a prominent candidate for the determination of flow rates and emissions from NV livestock buildings. Tracer gas studies are reported extensively in the contemporary literature dealing with residential, institutional (Eklund, 1999; Furtaw Jr., Pandian, Nelson, Behar, 1996; Han, Shin, Lee, & Kwon, 2011; Lim, Cho, & Kim, 2010; Santamouris et al., 2008; Xu, Luxbacher, Ragab, & Schafrik, 2013) and agricultural systems (Baptista, Bailey, Randall, & Meneses, 1999; Demmers et al., 1998; Demmers et al., 2001; Kaharabata, Schuepp, & Desjardins, 2000; Kiwan et al., 2013; Samer, Berg, et al., 2011; Samer, Loebsin, et al., 2011; Samer, Müller, Fiedler, Berg, & Brunsch, 2013; Schrade et al., 2012; Shen, Zhang, & Bjerg, 2012; Shen, Zhang, & Bjerg, 2013; Shen, Zhang, Wu, & Bjerg, 2013; Snell, Seipelt, Weghe, & Van Den, 2003; Van Buggenhout et al., 2009; Wu, Zhang, & Kai, 2012).

The theoretical foundation for tracer gas research is provided by the mixing-dilution first degree differential equation described by Barber and Ogilvie (1982), who demonstrated that in order to allow for the mass conservation of a given tracer (T) within the ventilated airspace, the constant injection rate (Q_T) of the tracer must equal the product between air exchange rate (AER, in per unit time) and the concentration of T corrected for background ([T]), in such a way that $Q_T = AER \cdot [T]$. Similarly, for a pollutant (P) being emitted to the same airspace, its emission rate Q_P can be calculated through $Q_P = AER \cdot [P]$. Hence, combining both relationships and solving for Q_P yields $Q_P = Q_T \cdot ([P]/[T])$. In other words, when using the tracer gas technique the complicated task of determining building ventilation rate in calculating gaseous emissions is suppressed/avoided. Instead, obtaining a representative mean mixing ratio $\overline{[P]/[T]}$ for the entire barn becomes essential.

The premise that both P and T present similar mixing behaviour in the region where the concentrations are to be measured might not be true when P and T have different physical properties, for instance, dissimilar molecular masses. Such discrepancies especially take effect when the mixing conditions are not ideal. Furthermore, an ideal T is the one that leads to mixing ratios that present constant values at least in some region of the barn ventilated airspace, indicating thorough homogeneous mixing. Also, it is of importance for representative mixing ratios that the release method of T sufficiently mimics the release of P. Several tracers have been used in past research studies, e.g. the carbon dioxide (CO_2) from the metabolism of animals and stored manure (Mosquera, Groenestein, Ogink, & Aarnink, 2012; Samer, Loebsin et al., 2011; Xin, Burns, Gates, Overhults, & Earnest, 2009); and artificially injected tracers such as krypton-85 (^{85}Kr) (Kiwan et al., 2013; Samer, Berg et al., 2011; Samer, Müller et al., 2011), trifluoromethyl sulphur pentafluoride (SF_5CF_3) (Schrade et al., 2012), and sulphur hexafluoride (SF_6) (Grainger et al., 2007; Kaharabata et al., 2000; Lassey, 2013; Schrade et al., 2012; Wu et al., 2012). However, a few studies have shown that not all chosen tracers, for example ^{85}Kr, behave the same when used for simultaneous determination of pollutant emissions from a ventilated airspace (Kiwan et al., 2013; Samer et al., 2012), and will lead to highly discrepant mixing ratio values due to the imperfect mixing and injection points of the tracer. The risk on discrepancies will increase with NV barns that have very open designs with large air exchange openings (Ogink et al., 2013).

Hence, the goal of this study was to assess the spatial variability of concentrations of pollutant and tracer gases and their ratios in a NV barn. The distribution of the artificial tracer gas SF_6, metabolic CO_2 as a tracer (produced by the animals through respiration and by their manure through bacteriological decomposition) and the pollutant NH_3, along with their mixing ratios ($[NH_3]/[CO_2]$, $[NH_3]/[SF_6]$, and $[CO_2]/[SF_6]$), were measured inside a naturally ventilated dairy cow barn. Specific objectives were (a) to delineate potential differences in distribution and mixing patters between the tracers SF_6 and CO_2 and (b) to determine whether it is possible to find a zone within the barn airspace where representative $\overline{[P]}/\overline{[T]}$ mixing ratios can be measured.

2. Methodology

A selected dairy cow barn was equipped with an injection system for the controlled release of SF_6. Two sampling poles were installed inside the dairy cow barn, with the goal of monitoring the patterns in dispersion of the pollutant NH_3 and the tracers CO_2 and SF_6 in the vertical (V) direction and in the horizontal cross (HC) barn direction. A more thorough description of barn, SF_6 injection system, experimental procedures and data analysis is provided below.

2.1. *Description of the dairy cow barn and animals management*

This study took place in a NV dairy cow barn (Fig. 1) with a design that is representative for modern dairy barns in Northern Europe. The barn was located in Bunschoten, in the middle of the Netherlands, was east-west oriented, had a roof with 37% slope, and dimensions of 64 Length × 38 Width × 4 Side wall Height m (L × W × H). The building envelope was composed of insulated roof and open side walls, the lateral openings on both sides are 2.75 m high, protected with stainless steel screens with openings of 50 × 50 mm and has manually operated curtains. The eastern part of the building features a small deep litter area of 6.5 L × 21 W m and a few cubicles with maximum housing capacity of 30 dry and pregnant cows. In the central part of the building, 3 double-rows of cubicles (paper chips bedding, 42 L × 21 W m) are located, with feeding alleys on both sides (north and south), and maximum housing capacity for 150 lactating cows. The last section of the barn is at the most western side, has an area of 13 L × 21 W m with similar cubicles and bedding system as for the lactating cows, where the heifers are kept

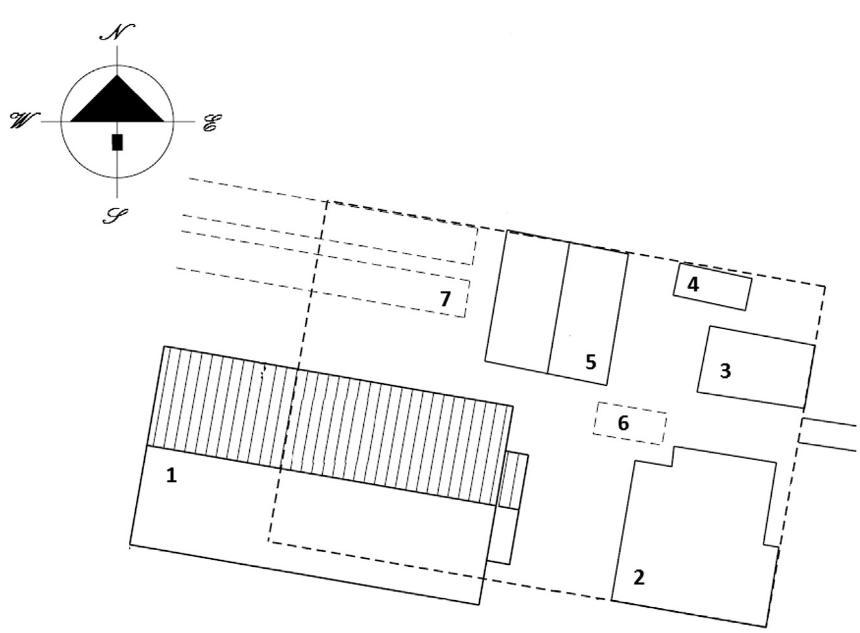

Fig. 1 – Farm site layout and description: 1 – dairy cow barn; 2 – young cattle barn; 3 – farmer's house; 4 – storage; 5 – machine shed; 6 and 7 – silos.

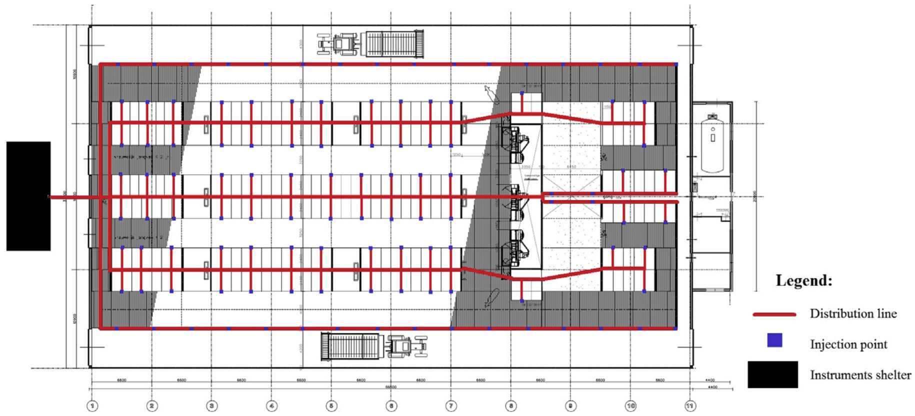

Fig. 2 – Drawing of the SF_6 distribution lines and injection points spread along the dairy cow barn.

(maximum capacity of 40 heifers). Barn cubicles area had concrete slatted walking alleys and a scraping robot that was automatically operated every 3 h. Manure was stored in a deep pit space of 65 L × 21 W × 2D m located under the slats and cubicles. The lactating cows had free access to 3 milking robot systems. All cows were kept inside all year long and were fed with roughage (grass and corn silage) and additional concentrates; bedding material consisted of shredded recycling paper which was replaced at every 4 months or as needed. Data collection tests in the barn were conducted during the summer season, in the months of June, July and August of 2012.

2.2. Description of the SF_6 injection system

The SF_6 injection system (Fig. 2) was designed to mimic the release of pollutant gases in the barn. Pure SF_6 tank and air compressor system were kept in a sheltered waggon placed outside near the western extremity of the barn. At a controlled mass flow rate of 0.027 l min^{-1} (GFM 57[1], Aalborg Instruments & Controls, Orangeburg, New York, USA), SF_6 (@ 99.9%) was mixed with compressed air at a flow rate of 10 l min^{-1} (GFM 57, Aalborg Instruments & Controls, Orangeburg, New York, USA), the air-SF_6 mixture was channelled into the barn through polyethylene tubing (6.3 mm inside diameter) where it was split into three branches (south, central and north). South and north branches were spread along both feeding fences. The central branch was spread along the cubicles area and beyond the milking parlour, with the injection points placed at approximately 0.5 m above the floor. A total of 116 injection points were distributed along the barn, including the lactating cows, dry cows and heifer's area. The injection points were equipped with capillary tubes to allow the passage of a specific amount of tracer gas. After installation, a few injection points in each branch of the injection system were checked for the presence of flow, by testing with soap bubbles.

[1] Mention of product or company names is for presentation clarity and does not imply endorsement by the authors or their affiliations, nor exclusion of other suitable products.

2.3. Vertical and horizontal cross barn variability of concentrations and mixing ratios

Two sampling poles were designed to measure concentrations of SF_6, CO_2 and NH_3 at 4 different vertical distances from the slats (V = 1, 2, 3 and 4 m from the floor) in two different locations (north and south cubicle alleys) along the width of the barn. The south location was 20 m west from the milking parlour, 11 m from the south side wall and the north location was also placed 20 m west from the milking parlour, but 11 m from the north wall (Fig. 3).

For each location, two sampling lines made of polytetrafluoroethylene tubing (PTFF, 6.3 mm inside diameter) were used to collect simultaneous air samples at every height (total of 8 sampling points per location); the sampling lines were supported by a 5 m (heigh) stainless steel pole (Fig. 4A). One of the sampling lines from each heigh was connected to a PTFF bag (maximum volume of 50 l), sampling air was aspirated to it by vacuum pump at a flow rate of 400 ml min^{-1}. The other sampling line coming to each height was attached to an impinger train. Each train consisted of three Greenburg-Smith® type impingers (WWR International©, Randor, Pensylvania, USA) connected in series with leak-free ground glass fittings and PTFF tubing. The first and second impingers contained each 125 ml of 0.1 N sulphuric acid (H_2SO_4) and the third was empty and collected excess acid solution that could potentially come from adjacent impingers. The impingers and the PTFF bags were connected to the vacuum pumps (1000 ml min^{-1} each) through capillary tubes that allowed for the passage of their specific flow rates, excess flow from the pump was discharged with a by-pass connection. Hence, each monitoring station consisted of four impinger trains, four PTFF bags and four vacuum pumps (Fig. 4B).

Measurements were carried out over the same period of the day (around noon), all pumps were turned on for 120 min. After the measuring period, both the PTFF bags and impingers were sent to the laboratory for analysis of gaseous concentrations of CO_2 and SF_6 through the gas chromatography method, while the impingers were analysed for ammonium-N content (mg l^{-1}) to assess NH_3 concentrations through chromatography.

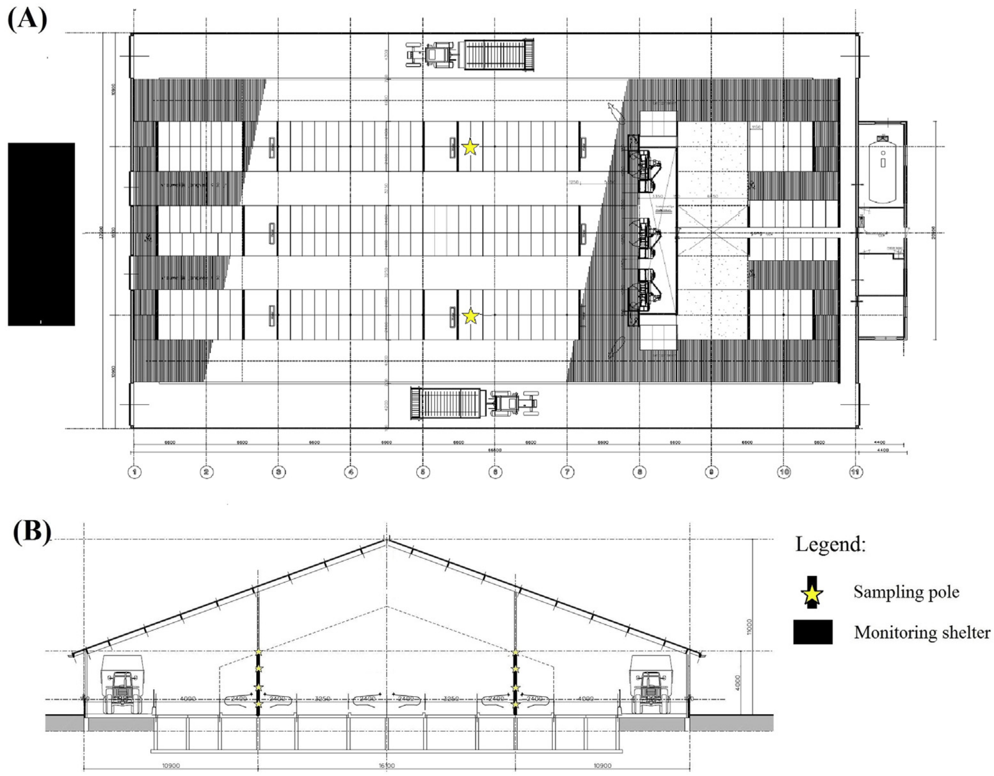

Fig. 3 – Plan (A) and cross-sectional (B) views of the investigated barn with the allocation of the sampling poles of carbon dioxide (CO_2), sulphur hexafluoride (SF_6) and ammonia (NH_3) of aerial concentrations (not to scale).

2.4. *Data processing and statistical analyses*

A total of 7 tests of 120 min duration each were performed in the summer of 2012. Prior to every monitoring session, barn screens were closed to 50% on both sides of the building, resulting in an effective opening area of 92 m^2 per side. The SF_6 injection system was turned on at least 30 min before measurements started, in order to allow full stabilisation of concentrations within the barn envelope.

Before data analysis procedures, data sets collected with the sampling poles from each side of the barn (north and south) were combined with wind direction, based on which

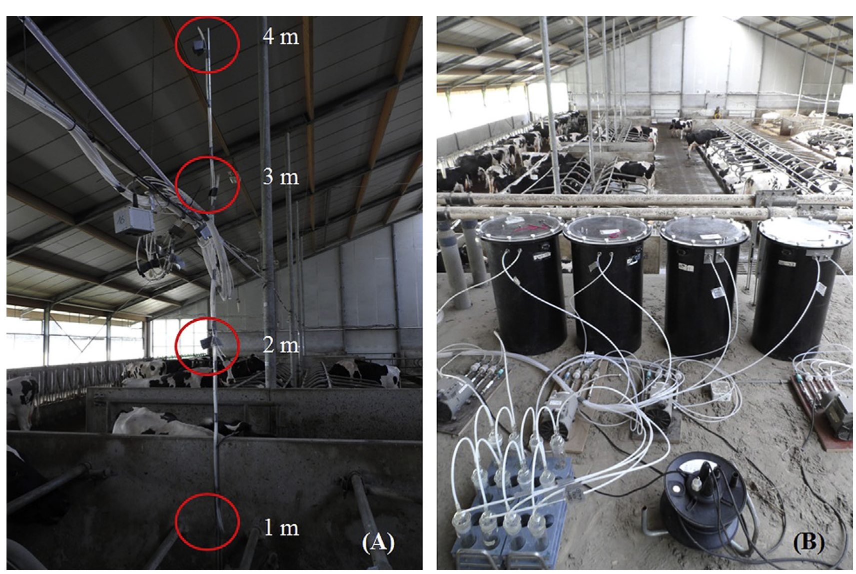

Fig. 4 – Diagram of a gaseous sampling pole (A), with highlights for the sampling ports at different heights; and a monitoring station (B), presenting the containers with PTFF bags, impingers' train and air pumps.

inlet and outlet sides were defined, thus, data sets collected from north and south poles were denominated "near inlet" or "near outlet" depending on the wind direction. Outside wind speed and direction data from the nearest meteorological station (KNMI, station De Bilt, at 20 m heigh and located 30 km from the measurement site) was used in this study.

A two-way analysis of variance (two-way ANOVA) was performed with the factors vertical component (V: 1, 2, 3 and 4 m from slats) and the cross horizontal component (HC, near inlet or outlet) with the response variable being gaseous concentration of pollutant and tracer gases and the mixing ratio between both. The analysis was done through the procedure general linear model (GLM) in SAS® (Version 9.4, Cary, North Carolina, USA) to test if the effects of V and HC on gaseous concentration can be explained by the fixed effects model described in Eq. (1). Pairwise test comparison between every level of V and HC was also implemented by adding the statement *pdiff* in the SAS® code, with *p* values adjusted with Tukey-Kramer corrections. All tests were performed on a significance level of 0.05.

$$Y_{ij} = Y + V_i + HC_j + \varepsilon_{ij} \tag{1}$$

where:

- Y_{ij} is the measured pollutant or tracer gaseous concentration, or ratio between pollutant and tracer;
- Y the grand mean concentration of pollutant or tracer gas, or ratio between pollutant and tracer present throughout the experiment;
- V_i the effect of the vertical dimension of the barn, or height from slats (1, 2, 3 or 4 m) on concentrations of pollutant or tracer gas, or ratio between pollutant and tracer;
- HC_j the effect of the cross horizontal barn dimension, or building width (near inlet or outlet) and
- ε_i the independent normally distributed homogeneous random error.

When the statistical analysis was performed with concentration data as response variable, the values were used "as-is". However, when analysis was done with mixing ratios of the monitored of P and T, their gaseous concentrations were corrected for the constant background values of 0.13 ppm_v, 417 ppm_v and 0 ppb_v, for NH_3, CO_2 and SF_6 respectively. These background concentration values were based on measurements from previous studies.

3. Results and discussion

A summary of numbers of cows and meteorological conditions, along with sidewall air inlet position data for each test are presented in Table 1. It can be seen that during most of the periods, the wind was coming mainly from the north, being the north sidewall considered the predominant air inlet.

3.1. *Spatial distribution analysis of the gaseous concentrations*

The V and HC distribution of gaseous concentrations of NH_3, CO_2 and SF_6 are plotted in Fig. 5. When looking at the vertical pattern of the measurements observed in Fig. 5, one notices that, in general, the concentrations tend to stabilise to a constant value as the distance from emitting source (animals) increases, especially where $V \geq 2$ m. The animal occupied zone (AOZ) in the barn, which extends to $V < 2$ m, is generally characterised by lower wind speeds and thus poorer mixing as compared to upper levels, and this might be the reason why some measurements in that zone were more unpredictable. This is presumably due to the presence of the animals and building structures such as fences, walls, cubicle metal frames, etc., which act as obstacles to the air flow thereby reducing wind speed. The complexity of the AOZ in terms of air and gaseous flow pattern predictability was acknowledged by Bjerg, Zhang, and Kai (2008) and Wu et al. (2012) when

Table 1 – Characteristics of the housed dairy cows and climate conditions during trials.

Variable	Test						
	1	2	3	4	5	6	7
Number of milking cows	140	135	136	134	135	142	144
Number of dry cows	22	25	24	29	28	21	22
Number of pregnant young cows	33	33	33	31	31	34	35
Milking cows liveweight (kg)[a]	664	663	663	661	660	662	658
Dry cows liveweight (kg)[b]	664	663	663	661	660	662	658
Pregnant young cows liveweight (kg)[c]	500	500	500	500	500	500	500
Milk production (kg/cow/day)	30.1	30.7	30.6	30.4	30.6	30.6	27.6
Inside air temperature (°C)	20.4 ± 0.2	18.3 ± 0.1	19.0 ± 0.2	25.6 ± 0.2	26.8 ± 0.3	19.9 ± 0.2	23.2 ± 0.1
Inside air relative humidity (%)	77 ± 1	89 ± 1	79 ± 2	63 ± 2	79 ± 2	95 ± 1	68 ± 1
Outside air temperature (°C)	20.4 ± 0.2	16.3 ± 0.1	17.3 ± 0.2	25.0 ± 0.4	25.4 ± 0.4	19.4 ± 0.3	22.3 ± 0.1
Outside air relative humidity (%)	68 ± 1	83 ± 1	67 ± 1	57 ± 2	72 ± 2	98 ± 1	63 ± 1
Wind speed ($m\ s^{-1}$)	5.67 ± 0.15	5.78 ± 0.15	2.93 ± 0.16	2.5 ± 0.13	1.91 ± 0.17	4.87 ± 0.40	4.33 ± 0.17
Wind direction (°)	212 ± 3	296 ± 2	338 ± 2	57 ± 2	341 ± 2	213 ± 4	224 ± 6
Side opening considered as inlet	South	North	North	North	North	South	South

[a] The liveweight of milking cows was measured on the day of the experiments at the milking parlour, with an electronic scale while they were being milked.
[b] The liveweight of dry cows was assumed to be the same as that of the milking cows.
[c] The liveweight of the young pregnant cows was estimated to be 500 kg.

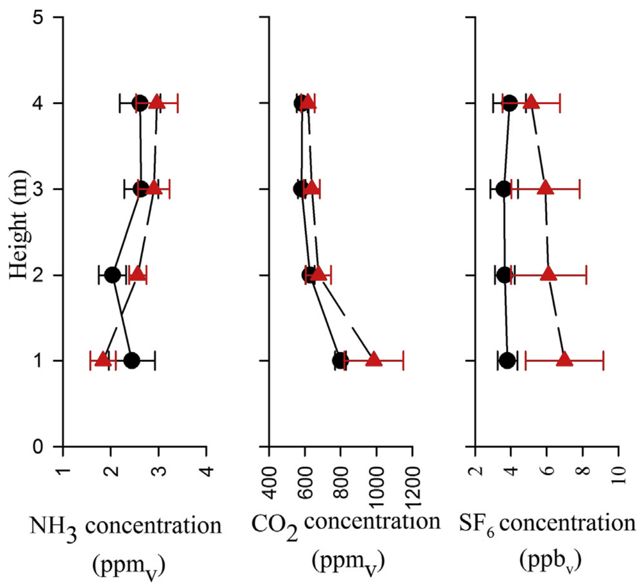

Fig. 5 – Vertical (height) and cross horizontal (near inlet or outlet) variability of concentrations of NH_3, CO_2 and SF_6 in the naturally ventilated dairy cow barn. Black dots and red triangles represent near inlet and near outlet concentrations, respectively. Error bars represent standard errors of the mean.

developing numerical models for air motion in NV dairy cow barns.

The results of the pairwise comparison analysis made for each monitored gas across heights is presented in Table 2, and show that the differences in concentrations between distinct levels was higher when the pairwise comparison was carried out in relation to V = 1 m, with a more pronounced effect for the gas CO_2. This outcome might be due to the close proximity of the sampling points to the CO_2 emitting source, i.e. the animals. The smaller differences in relation to the comparisons involving V = 1 m indicates that outside the AOZ, gaseous concentrations of NH_3, CO_2 and SF_6 were not sensitive to height, and were approximately constant.

Table 2 – Results of the pair-wise comparison test for heights. The numbers represent the absolute difference in concentration between two considered heights (HCs) and the values between brackets represent 95% confidence limits.

Absolute height comparison (m)	NH_3 (ppm_v)	CO_2 (ppm_v)	SF_6 (ppb_v)
1–2	0.16 (−0.81; 1.12)	223* (−50; 495)	0.270 (−3.6; 3.9)
1–3	0.63 (−0.33; 1.59)	262* (−11; 534.5)	0.125 (−3.5; 3.8)
1–4	0.65 (−0.31; 1.60)	267* (−5; 540)	0.272 (−3.3; 3.8)
2–3	0.47 (−0.49; 1.43)	39 (−234; 311.8)	0.145 (−3.5; 3.8)
2–4	0.49 (−0.47; 1.45)	45 (−288; 317)	0.002 (−3.6; 3.6)
3–4	0.02 (−0.94; 0.98)	6 (−267; 278)	0.146 (−3.7; 3.4)

*Mean difference is significantly different from zero at a significance level of 5%.

The HC distribution of the gases is represented by the concentrations that, at a certain height, were measured near barn inlet or outlet sides. It can be seen in Fig. 5 that for all gases (except NH_3 at H = 1 m), the concentrations measured near the outlet were slightly higher than those monitored near the inlet. This outcome evidences the existence of a cross air flow through the building, in which the fresher air entering at the inlet side opening purges the gases produced in the AOZ of the barn, leaving from the outlet side opening. This kind of flow is typical in NV barns and is mainly wind driven (Albright, 1990, chapter 11). However, the overlapping standard error bars at each level, indicate that the concentrations measured near inlet and outlet could not be distinguished in statistical terms. The ANOVA results showed that for all monitored gases under the conditions of this experiment and with a total number of 7 replicates, the barn width factor did not have a significant effect on concentration measurement (*p*-values ranging from 0.11 to 0.60).

Additionally, for a specific height, the concentration measured near outlet was divided by that measured near inlet; the results of this ratio are graphically presented in Fig. 6 for the gases NH_3, CO_2 and SF_6.

The average values for outlet/inlet ratios calculated between V = 3 and V = 4 m, where mixing show more stable patterns (Fig. 5), were (1.2 ± 0.1), (1.08 ± 0.03) and (1.5 ± 0.2) for the gases NH_3, CO_2 and SF_6, respectively. These ratios indicated that for NH_3 and CO_2, the increase in concentrations near the outlet was only 8–20% higher than that near the inlet, and that both NH_3 and CO_2 presented similar distribution patterns across barn width. However, for the gas SF_6, the concentration near outlet could be on average 50% higher than that at near the inlet. This means that the distribution of SF_6 across the barn width was not even, indicating a different behaviour in dispersion. A potential non-homogeneous

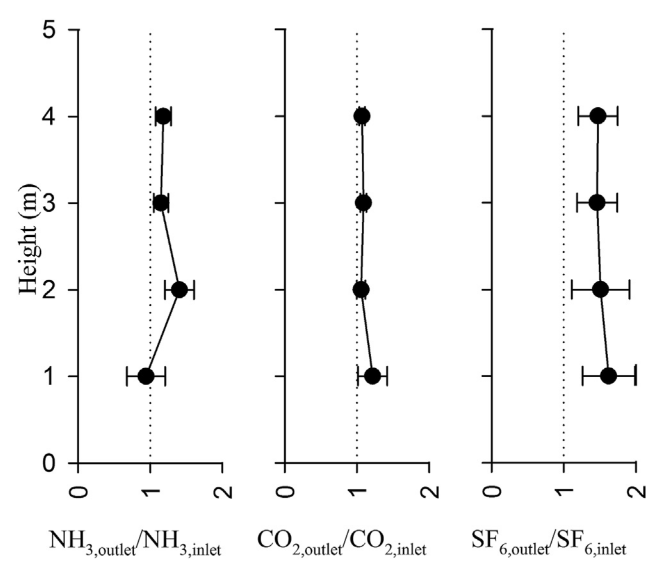

Fig. 6 – Vertical concentration ratios measured near inlet and outlet for the gases NH_3, CO_2 and SF_6, monitored in the naturally ventilated dairy cow barn, the error bars correspond to standard error of the mean.

dispersion of SF_6 across the barn may have occurred. From Fig. 6, a similarity in dispersion behaviour of NH_3 and CO_2 can be deduced. Possible reasons for the poorer distribution of SF_6 across the barn might be its relatively higher molecular weight (146.06 g mol^{-1}), as compared to CO_2 (44.01 g mol^{-1}) and NH_3 (17.03 g mol^{-1}), which might have caused it to present a different dispersion pattern, by remaining stagnated within the lower barn levels, while the other gases were able to rise and leave the barn through the outlets. The positioning of the SF_6 injection lines might also have played an important role on the spread of that gas, by defining its release and dispersion patterns. This outcome highlights the need of evaluating the performance efficiency of complex artificial tracer injection systems, such as the one used in this study, prior to the start of monitoring periods. This might be a motivation for further research.

3.2. *Analysis of spatial distribution of pollutant and tracer mixing ratios*

The spatial variability of the mixing ratios NH_3/CO_2, NH_3/SF_6 and CO_2/SF_6 are graphically represented in Fig. 7 and the results of the pair-wise comparison between ratios calculated for different heights are presented in Table 3. The ANOVA results indicated that the horizontal width dimension of the barn did not have a significant impact on the calculated mixing ratios (p-values ranging from 0.08 to 0.96).

The calculated mixing ratios involving NH_3 as pollutant, shown in Fig. 7, tended to present lower values within the AOZ and approximately constant values above it. Such results corroborated what was demonstrated before in this paper that the complex air flow patterns present in the AOZ makes it a risky place to determine gaseous concentrations and mixing ratios that are representative for the entire ventilated airspace, and should be avoided. The pair-wise comparison analysis output presented in Table 3 for the mixing ratios NH_3/CO_2 and NH_3/SF_6 supports the fact that in the AOZ, the ratios were significantly lower than all the mean ratios calculated above it. Based on this outcome, it is possible that if mixing ratios are calculated with concentration data monitored within the AOZ, the averaged barn mixing ratio will be biased. Preference should be given to sampling strategies set to monitor regions within the barn where mixing ratios present relatively constant values.

The presence of approximately constant mixing ratios between pollutant and tracer gases above the AOZ is an indication that both gases present similar mixing conditions, and hence, ratios measured at H > 2 m shall be used to estimate gaseous emissions of NH_3.

As for the comparison between mixing ratios near inlet vs. near outlet, it can be seen in Fig. 7 that above the AOZ, both mixing ratios NH_3/CO_2 and NH_3/SF_6 presented values near the inlet that were similar to those at the outlet. This result was corroborated by the pairwise test performed for the horizontal (near inlet and outlet) positions, resulting in ratios that were statistically the same. Such outcome was particularly true for the mixing ratio NH_3/SF_6, and indicates that even if SF_6 concentrations presented greater differences between near inlet and outlet positions than NH_3 concentrations, as discussed before in this paper, the horizontal variability of their mixing ratios were similar. This result demonstrates the robustness of the mixing ratio method for estimating gaseous emissions from NV livestock barns, and that P and T do not necessarily need to be homogeneously spread within the barn to yield homogeneous mixing ratios. The pitfall here, however, is that if there are problems with the artificial injection system that lead, for instance, to leakage of T, they cannot be easily detected.

The hypothesis of the similar mixing conditions between P and T tested in this study was proven to be valid outside the AOZ. On the other hand, the data presented in this paper indicated that when sampling of concentrations is done within the AOZ, the mixing ratios will present higher variability and may not be representative for the entire envelope of the barn.

Gaseous mixing may also be dependent on changing wind direction. This study was conducted in short trials (2 h) in which the wind conditions were approximately stable. Hence, longer tests with changing wind conditions and analysis of effects on the mixing of gases should be carried out in future research.

This research study can be seen as a first step to check vertical and horizontal variability of multiple P, T and their mixing ratios in NV livestock barns. A few questions that arise as a consequence of this study are: (a) what happens above H > 4 m? (b) Is the mixing ratio still representative if measured at the ridge? Moreover, it is known that a longitudinal variability also exists (different animals, presence of the litter, etc.), which was not evaluated in this study. Although these questions were not the aim of this present study, they may instigate further research. Lastly, since gaseous mixing might be also dependent on curtains opening, and since in this study

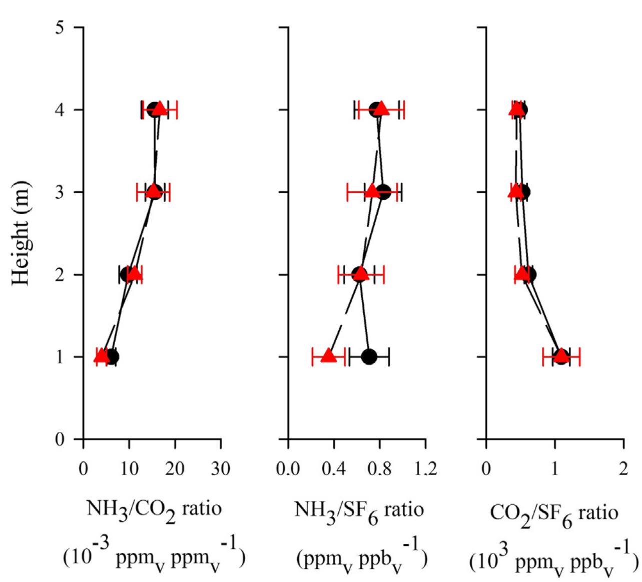

Fig. 7 – Vertical (height) and horizontal (near inlet or outlet) variability of the mixing ratios NH_3/CO_2, NH_3/SF_6 and CO_2/SF_6 in the naturally ventilated dairy cow barn. Black dots and red triangles represent near inlet and near outlet mixing ratios, respectively. Error bars represent standard errors of the mean.

Table 3 – Results of the pair-wise comparison test for heights. The numbers represent the absolute difference in mixing ratios between two considered heights (HCs) and the values between brackets represent 95% confidence limits.

Absolute height comparison (m)	NH_3/CO_2 (10^{-3}ppm$_v$ ppm$_v^{-1}$)	NH_3/SF_6 (ppm$_v$ ppb$_v^{-1}$)	CO_2/SF_6 (ppm$_v$ ppb$_v^{-1}$)
1–2	5.20 (−1.92; 12.35)	0.246 (−1.056; 0.565)	0.282 (−0.565; 1.128)
1–3	10.13* (3.00; 17.25)	0.543 (−0.267; 1.354)	0.203* (0.643; 1.128)
1–4	10.50* (3.38; 17.62)	0.444 (−1.238; 0.351)	0.353 (−0.477; 1.183)
2–3	4.93 (−2.20; 12.05)	0.298 (−0.513; 1.108)	0.078 (−0.768; 0.925)
2–4	5.93 (−1.83; 12.42)	0.198 (−0.597; 0.992)	0.071 (−0.902; 0.759)
3–4	0.37 (−7.50; 6.76)	0.099 (−0.700; 0.895)	0.150 (−0.980; 0.680)

*Mean difference is significantly different than zero at a significance level of 5%.

the curtains were kept constantly open at 50% on both sides, the similar mixing conditions may not be valid at very open situations. For this reason, it is recommended that the effect of different window openings, e.g. 75 and 100% both sides, on the spatial distribution of concentrations and mixing ratios be investigated.

4. Conclusions

A dairy cow barn was equipped with an injection system for controlled release of SF_6 and two sampling poles in order to monitor the patterns in vertical (V, 1–4 m above the slats) and cross horizontal barn (HC, near inlet and outlet) dispersion of the pollutant NH_3, the metabolically produced tracer CO_2, the artificially injected SF_6 and their mixing ratios. The following conclusions can be drawn:

1. The vertical variability of concentrations of NH_3, CO_2 and SF_6 and their mixing ratios became more stable with increase in height, reaching approximately constant values above the AOZ (V > 2 m);
2. The cross horizontal barn spread of the metabolically produced tracer CO_2 resembled more that of the pollutant NH_3, than the artificially injected tracer SF_6, as expressed by outlet/inlet concentration ratios.
3. The use of both, CO_2 and SF_6, as a tracer gas led to similar vertical and cross horizontal barn homogeneous spread in behaviour of mixing ratios.
4. In the conditions of this study, the most appropriate region in the barn to measure concentrations and determine the mixing ratios NH_3/CO_2 and NH_3/SF_6, is above the AOZ (V > 2 m).

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