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# Multi-location measurements of greenhouse gases and emission rates of methane and ammonia from a naturally-ventilated barn for dairy cows

## N.M. Ngwabie\*, K.-H. Jeppsson, S. Nimmermark, C. Swensson, G. Gustafsson

Department of Rural Buildings and Animal Husbandry, Swedish University of Agricultural Sciences, P.O. Box 86, S-230 53 Alnarp, Sweden

## article info

Article history: Received 21 August 2008 Received in revised form 21 January 2009 Accepted 7 February 2009 Published online 5 March 2009 Measurements of the gaseous emissions in livestock buildings are important as these pollutants may affect the health of farmers and the surrounding environment. Emission monitoring enables judgements on the effectiveness of mitigation strategies and controls on emission targets. The concentrations of CH<sub>4</sub>, N<sub>2</sub>O, NH<sub>3</sub> and CO<sub>2</sub> were measured in a naturally-ventilated dairy cattle building using a photoacoustic multi-gas analyser 1412 and a multiplexer 1309 (Lumasense Technologies SA, Ballerup, Denmark). The building had 164–195 Holstein dairy cows. The milk production was 31–33 kg cow $^{-1}\,\rm{d}^{-1}$ . Manure gutters beneath the partially slatted floor in the building were scraped twice a day. Measurements took place during the winter when the cows were permanently indoors and during 1 week in the spring when the cows were indoors at night and outside grazing during the daytime. The indoor concentrations were measured at nine evenly distributed locations, while outdoor concentrations were measured at two locations. The mean ventilation rate in winter was 250–265  $\mathrm{m^{3}LU^{-1}h^{-1}}$  and in spring, it was 401  $\mathrm{m^{3}LU^{-1}h^{-1}}$ . The emissions of NH $_{3}$ and CH<sub>4</sub> were in the range of 0.89–1.13 and 9–13  $\rm gLU^{-1}h^{-1}$ , respectively. A strong positive correlation was found between enhanced  $CO<sub>2</sub>$  and  $CH<sub>4</sub>$  concentrations.

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## 1. Introduction

Livestock management is a potential source of environmentally deleterious gaseous emissions that may affect the health of farm workers, animals and the surrounding communities (Zhang et al., 1998; Charavaryamath & Singh, 2006). Although the number of livestock animals may decrease regionally, as in Sweden during the years 2001–2006 (Statistics-Sweden, 2008), globally, the growing population and the increasing consumption of milk and meat in populous countries like China and India, will most certainly lead to an increase in emissions from livestock. Therefore, the problems associated with emissions from this sector are likely to persist. Emissions of NH<sub>3</sub> affect the ecosystem while CH<sub>4</sub> and N<sub>2</sub>O are greenhouse gases which influence the global climate.

About 94% of global anthropogenic emissions of  $NH<sub>3</sub>$  to the atmosphere originate from the agricultural sector of which close to 64% is associated with livestock management (FAO, 2006). Excessive levels of  $NH<sub>3</sub>$  emissions contribute to eutrophication and acidification (Schuurkes & Mosello, 1988). Extensive research has been carried out on this emission mechanism from livestock and the factors that affect it as well as suggestions on possible mitigation strategies (Hill & Barth, 1976; Chiba et al., 1987; Jeppsson,

Corresponding author.

E-mail addresses: ngwa.martin.ngwabie@ltj.slu.se (N.M. Ngwabie), knut-hakan.jeppsson@ltj.slu.se (K.-H. Jeppsson), sven.nimmermark@ltj.slu.se (S. Nimmermark), christian.swensson@ltj.slu.se (C. Swensson), gosta.gustafsson@jbt.slu.se (G. Gustafsson). 1537-5110/\$ - see front matter © 2009 IAgrE. Published by Elsevier Ltd. All rights reserved. doi:10.1016/j.biosystemseng.2009.02.004



1998; Hayes et al., 2004; Luo et al., 2004; Nimmermark & Gustafsson, 2005; Varel et al., 2007). A number of countries have set regional targets to decrease the emissions of  $NH<sub>3</sub>$ . In Sweden, the aim is that by 2010, emissions of  $NH<sub>3</sub>$  will have to be reduced by at least 15% compared to the levels in 1995 (Swedish-EPA, 2008).

Enteric fermentation and manure management account for 35–40% of the total anthropogenic  $CH<sub>4</sub>$  emissions and 80% of CH4 release from agriculture (FAO, 2006). Livestock activities contribute with 65% of the global anthropogenic  $N_2O$  emissions and account for 75–80% of the emission from agriculture (FAO, 2006). CH<sub>4</sub> and N<sub>2</sub>O are greenhouse gases with global warming potentials of 23 and 296 times that of  $CO<sub>2</sub>$ , respectively (IPCC, 2001). A number of investigations have been conducted on the emissions of these gases from livestock management (Jungbluthet al., 2001; Haeussermannet al., 2006; Monteny et al., 2006; Olesen et al., 2006; Sneath et al., 2006; Weiske et al., 2006). In Sweden, the target set for climate gases is that the average emission of greenhouse gases from 2008 to 2012 shall be at least 4% lower than in 1990 (Swedish-EPA, 2008).

Long-term mitigation strategies depend on an in-depth understanding of emission mechanisms and emission rates. An emission database can be a useful tool for improved emission models and for monitoring emission targets. It has been suggested that more research is needed to support existing publicationsonregionaland global emission factors(GrootKoerkamp et al., 1998; Amon et al., 2001; Jungbluth et al., 2001; Snell et al., 2003; Zhang et al., 2005; Starmans & Van der Hoek, 2007).

Generally, therelease rate of gases may vary from one region to another since emissions depend not only on the management systems, but also on the building types and the regional climates (Groot Koerkamp et al., 1998). While it is relatively easy to estimate emission rates from mechanically ventilated buildings, naturally-ventilated buildings are problematic because of difficulties with measuring air exchange rates. These types of buildings are commonly used for cattle since they are not especially susceptible to draughts and temperature changes and no extra heating is required. Air exchange rates in these buildings depend on the temperature gradient and the wind. In this case, the release rates of pollutants may also depend on external and uncontrollable parameters such as wind speed and the surrounding topography.

#### 1.1. Objectives

The present trend in milk production in Sweden is to change to systems with loose housing in naturally-ventilated buildings. Gaseous emissions may vary with building or manure systems applied. Emissions from dairy houses in Sweden may be different compared to emissions from other regions in Europe due to differences in climatic conditions. The hypothesis of this study was that emissions vary with time and season.

The purpose of the investigation was to:

- study the concentrations of  $CO<sub>2</sub>$ ,  $CH<sub>4</sub>$ , NH<sub>3</sub> and N<sub>2</sub>O during winter and spring in a naturally-ventilated dairy cattle building with a liquid manure system and frequent manure removal
- study gas concentration changes by both time and place inside the building
- $\bullet$  estimate the emissions of NH<sub>3</sub> and CH<sub>4</sub>
- $\bullet$  estimate the fraction of NH<sub>3</sub>–N loss
- analyse the correlations between indoor concentrations of  $CO<sub>2</sub>$ , CH<sub>4</sub> and NH<sub>3</sub>
- $\bullet$  investigate the influence of the ventilation rate (VR) on the indoor concentrations of  $NH<sub>3</sub>$  and CH<sub>4</sub>.

To achieve this, the concentrations of  $CO<sub>2</sub>$ ,  $CH<sub>4</sub>$ ,  $NH<sub>3</sub>$  and N2O were measured at nine locations inside, and at two locations outside a typical dairy cattle building.

#### 2. Materials and methods

#### 2.1. Experimental site

Measurements were carried out in a dairy cattle building with cubicles and a liquid manure system, in the south of Sweden. The building was 83.9 m long and 28.9 m wide. Measured from floor level, the height of the walls was 3.5 m and the height to the ridge was 8.7 m. The barn was naturally ventilated through automatically regulated ventilation flaps mounted on the sidewalls below the eaves and also at the ridge. Occasionally, the doors on the sidewalls were used to increase the VR. An integrated milking parlour protruded from one side of the building (Fig. 1). The building was located in open land with little shielding from the wind by trees or surrounding hills. It was divided into four sections: A–D, of which the first three had milking cows. Section D was further subdivided to hold pregnant cows on deep litter and calves that were kept for a week before being taken to another building. The floor had a raised platform with cubicles where the cows can lay down and a lower walkway made of concrete slats over





Fig. 1 – Layout of the dairy cattle building. The letters (A–D) represent sections in the building, and P1–P12 denote sampling locations for gas measurements.

manure gutters with scrapers. The manure in the gutters was scraped twice a day into manure storage tanks outside the building. Three open-surface concrete manure storage tanks were located about 15 m from the building.

During the measurement period, there were 164–195 dairy cows with an estimated average body weight of 600 kg in the barn. Daily milk production during the sampling period was within 31–33 kg cow $^{-1}\,\rm{d^{-1}}$ .

Daily activities in the building included a cycle which started with milking at 5 a.m. followed by feeding and ended with mechanical cleaning of the manure gutters at 8:50 a.m. A second cycle started at 4 p.m. with feeding and milking. It ended with cleaning of the manure gutters at 8:50 p.m. On weekdays from Mondays to Fridays, the cows were also fed at midday from 12 a.m. to 2 p.m. When the weather was warm (i.e. during May), the cows were outside grazing between 10 a.m. and 4 p.m.

#### 2.2. Animal diet

During periods without grazing, the average daily feed consumption was estimated by weighing the different feeds with an electronic scale. Daily feed consumption per cow was estimated at: 6.4 kg DM grass silage, 2.7 kg DM corn silage, 0.4 kg DM straw, 2.1 kg DM beet pulp, 4.2 kg DM wheat, and 4.3 kg DM protein premix (DM  $=$  dry matter). The cows were fed on totally mixed ration (TMR). The nitrogen content was analysed using the Kjeldahl method (carried out by Eurofins Laboratory, Kristianstad, Sweden). The average daily nitrogen intake was 0.5 kg N LU $^{-1}\,{\rm d}^{-1}$  according to the estimated feed consumption and analysis of the feed. The nitrogen content produced in the milk was 0.14 kg N LU $^{-1}\,{\rm d}^{-1}$  and that in the manure was 0.36 kg N LU $^{-1}$ d $^{-1}$ . During measurements with grazing in May, the cows were fed twice per day and got the rest of their feed from grazing.

## 2.3. Instrumental setup

The concentrations of  $CO<sub>2</sub>$ , N<sub>2</sub>O, NH<sub>3</sub>, CH<sub>4</sub> and water vapour in the air of the cattle building were measured using a photoacoustic multi-gas analyser 1412 and a multiplexer 1309 (Lumasense Technologies SA, Ballerup, Denmark). The detection thresholds of the gases were as follows: 0.2 ppm NH3, 0.03 ppm  $N_2O$ , 0.4 ppm CH<sub>4</sub> and 1.5 ppm CO<sub>2</sub>. The measuring accuracy for the multi-gas analyser according to data sheets from the manufacturer was  $\pm$ 2-3%. The default configurations of the multi-gas analyser were used with an automatic chamber flush time and a sample integration time of 5 s.

The instruments were placed on a 2.5 m high platform at the centre of the building. Air was sampled to the multiplexer through 3.2 mm (inner diameter) polytetrafluoroethylene (PTFE) tubes, which were equipped at the inlet with filters (Balston Filters, Parker Hannifin Corporation, Haverhill, MA, USA) for trapping large particles. The tubes from the sampling locations to the multiplexer were between 15 and 30 m long. Air was drawn into the multiplexer by an external booster pump connected to its exhaust. A flow rate of about 3.2  $\mathrm{l}\min^{-1}$ was measured at the exhaust of the booster pump. This pump ensured that air being analysed at any time was likely fresh from the sampling locations. A single booster pump at the exhaust of the multiplexer is better system than having 12 pumps in the supply tubes because it is cheaper and it ensures that the sampled air is not contaminated by the pumps before analysis. However, the length of the tubes meant that the air analysed at any time might have entered the tubes some time before. This potential time lag should not be a significant problem since continuous measurements were carried out and our interest was to measure mean values.

The multiplexer 1309 has 12 channels, 11 of which were used in this analysis. The first two channels had tubes connected to locations (P1, P2) outside the building for outdoor concentrations (Fig. 1). These locations were close to the outer walls at a height of 2 m from the ground. Nine channels had sample tubes evenly distributed inside the building with sampling locations (P3–P8 and P10–P12) above the cubicles and the feeding area at a height of 3 m from the floor.

Inaccuracies in the measurement of the concentrations of some gases such as  $NH<sub>3</sub>$  may arise from adsorption or desorption on the surface of the sampling tubes. This could happen when sampling high concentration differences or when changing sampling locations using the same tube. Tests have been conducted to assess  $NH<sub>3</sub>$  lose through adsorption as a function of sampling tube material, tube length, temperature and  $NH<sub>3</sub>$  concentrations (Mukhtar et al., 2003; Shah et al., 2006). In an experiment involving five types of tube materials: high-density polyethylene, polyvinyl chloride and three types of Teflon, Shah et al. (2006) reported  $NH<sub>3</sub>$ adsorption as being unaffected by the tube material. Mukhtar et al. (2003) recommended Teflon over low-density polyethylene (LDPE) for sampling NH<sub>3</sub>. Considering these reports, there might be some adsorption/desorption in the sampling tubes during concentrations spikes/decays or when changing from an indoor to an outdoor location. As a precaution, the tube between the multiplexer and the multi-gas monitor that conveyed air from all sampling locations was kept as short as possible (0.4 m).

Temperature was measured using Tiny-tags (Gemini Data Loggers, Chi Chester, UK) with an operating range of –40 to 85°C and an accuracy of  $\pm 1$ °C. Two Tiny-tags were placed inside the building close to the sampling locations P7 and P11 and two were placed outside, close to P1 and P2.

#### 2.4. Data acquisition

Data was collected during the cold weather period from December 2006 to March 2007 when the cows were confined inside the building. Measurements were also carried out for 1 week in May 2007 (22–29) when it was warmer outside. During this period, the cows were grazing daily between 10 a.m. and 4 p.m.

Gas concentrations were continuously recorded throughout the sampling period sequentially from positions P1 outside to P12 inside the building. The multi-gas analyser might not rapidly track changes in gas concentrations when switching between locations with high concentrations differences such as inside and outside animal buildings. The response time has been reported to be gas dependent with rapid tracking for  $CO<sub>2</sub>$  (Hinz & Linke, 1998) and delayed tracking for  $NH<sub>3</sub>$  (Rom & Zhang, 2008). In a comprehensive study, it was recommended to carry out a number of measurement cycles at each location before switching to a new location (Rom & Zhang, 2008). The measurements in this paper could be considered as close to this recommendation since concentrations were measured sequentially at several indoor locations before switching to outdoor locations.

Temperature readings were taken every 15 min throughout the entire measurement period. The relative humidity was calculated on a daily basis using the water vapour concentrations measured by the multi-gas analyser and the saturated vapour density at the corresponding surrounding temperature (Carl, 2006).

## 2.5. Ventilation and emission rate calculations

The air exchange rate in naturally-ventilated buildings cannot be directly measured. Indirect methods, such as the use of a tracer gas or the mass balance of  $CO<sub>2</sub>$  have been used for this purpose.

The VR in the building was calculated using  $CO<sub>2</sub>$  mass balance (CIGR, 2002) as presented in Eq. (1):

VR per HPU = 
$$
\frac{0.185}{(CO_2indoors - CO_2outdoors)10^{-6}}
$$
 (1)

where:

- $\bullet$  VR is the ventilation rate in  $m^3\,h^{-1}$  on a 24-h basis
- one heat producing unit (HPU) is 1000 W of the total heat produced by the animals at  $20^{\circ}$ C (CIGR, 2002)
- $\bullet$  0.185 is the CO<sub>2</sub> production in  $m^3\,h^{-1}\,HPU^{-1}$  and corresponds to a medium feeding level
- CO<sub>2</sub>indoors is the average indoor concentration in ppm from sampling locations P3–P8 and P10–P12 on a 24-h basis
- $\bullet$  CO<sub>2</sub>outdoors is the minimum average concentration in ppm from both outside locations on a 24-h basis. The minimum concentration at a specific time was chosen to represent the outdoor concentration in order to minimise any response time effect and problems with exhaust air increasing the CO<sub>2</sub> outdoor concentration.

The emission rates of  $NH<sub>3</sub>$  and CH<sub>4</sub> were calculated using the VRs from Eq. (1) and the enhanced concentration in the building as shown in Eq. (2):

$$
E = VR(C_{\rm in} - C_{\rm out})
$$
\n(2)

where:

- $\bullet\,$  E is the emission rate in mg  $\rm h^{-1}$
- $\bullet$  VR is the ventilation rate in  $m^3\,h^{-1}$  on a 24-h basis
- $\bullet$  C<sub>in</sub> and C<sub>out</sub> are the gas concentrations inside and outside the building, respectively, measured in mg  $m^{-3}$ .

Enhanced concentrations denote differences between indoor and outdoor concentrations.  $C_{\text{in}}$  was calculated as the mean of nine indoor sampling locations (P3–P8 and P10– P12). Outdoor concentrations might be higher on one side of the building than the other depending on the wind direction and the pattern of the exhausted air. To minimise this problem,  $C_{\text{out}}$  was represented by the minimum concentration at the two outdoor sampling locations. This also improved the concentrations of a gas like  $NH<sub>3</sub>$  with a high tracking time.

## 3. Results and discussion

The climatic conditions surrounding animal buildings are considered an extremely important factor concerning emissions. These conditions are probably essential for naturallyventilated buildings since the surrounding climate has a direct influence on the VR and most likely also on the emission rate. Table 1 shows the meteorological data during the

measurement period. The wind speed was obtained from a nearby weather station.

Analysis of the concentration profiles of  $CO<sub>2</sub>$ , CH<sub>4</sub> and NH<sub>3</sub> showed a variation by time and sampling locations during a short-term perspective (single days) while that of  $N_2O$ showed little variation by place. Fig. 2 shows an example of the variation of  $NH<sub>3</sub>$  concentration at some indoor sampling locations during a single day.

A one-way ANOVA was conducted on a linear model relating the natural logarithm of the indoor concentrations for a particular gas at all the sampling locations. It confirmed a significant difference ( $p < 0.001$ ) in the mean concentrations of  $CO<sub>2</sub>$ , CH<sub>4</sub>, NH<sub>3</sub> and N<sub>2</sub>O, respectively, at all indoor sampling locations. Gas concentrations for the sampling period in January are shown in Fig. 3. In comparison to the short-term perspective shown in Fig. 2 the differences between various sampling locations are much smaller. Fig. 2 shows the importance of multi-location sampling during short-term measurements in large animal buildings where there is hardly a uniform distribution of gases (Cnockaert & Sonck, 2007). However, when measurements are made frequently over longer periods (Fig. 3) a good choice of single sampling locations (for example P7 or P11) may give satisfactory results. The optimum number of sampling locations probably depends on factors like sampling interval, sampling time, animal distribution in the building, the building size and its orientation with respect to prevailing winds.

#### 3.1. Gas concentrations

Diurnal variations were observed for all gases with higher concentrations of  $CO<sub>2</sub>$ , CH<sub>4</sub> and NH<sub>3</sub> inside the barn than in the outdoor air (Tables 2 and 3). An exception was  $N<sub>2</sub>O$  where outdoor concentrations sometimes exceeded indoor concentrations.

The indoor concentrations of  $NH_3$ , CH<sub>4</sub> and CO<sub>2</sub> showed little variation during the winter months from December to March when the cows were confined in the building (Table 2). When the cows were in the building in May (5 p.m.–9 a.m.), the indoor concentrations of NH<sub>3</sub>, CH<sub>4</sub> and CO<sub>2</sub> amounted to 60%, 48% and 74% of the respective indoor winter values. The





Fig. 2 – Diurnal variation of  $NH<sub>3</sub>$  concentration at different sampling locations inside the dairy cattle building during a day in January, 2007. The solid line represents the mean of all nine indoor sampling locations (P3–P8 and P10–P12).

indoor  $N_2O$  concentrations were low, and were close to the outdoor concentrations (Tables 2 and 3). The  $N_2O$  indoor concentration was in the range of 0.29–0.36 ppm (December– March) and 0.34–0.39 ppm (May). Small concentrations and small concentration differences suggest that measurement methods with higher accuracy may give better estimates of N<sub>2</sub>O emissions. Livestock buildings with liquid manure systems and external manure tanks are not a major source of N<sub>2</sub>O compared to deep litter systems (Monteny et al., 2006). Difficulties in measuring low  $N_2O$  concentrations have been reported by (Jungbluth et al., 2001).  $N_2O$  indoor concentrations of 0.32–0.40 ppm in a dairy cattle building with a slatted floor (Berges & Crutzen, 1996) and of 0.56 ppm in a building with dairy cattle and heifers on a slatted floor have been measured (Jungbluth et al., 2001). Generally, the minimum and maximum indoor concentrations measured for the entire sampling period were as follows: 0.16-0.75 ppm  $N_2O$ , 1.70-17.93 ppm NH<sub>3</sub>, 9-283 ppm CH<sub>4</sub> and 644-3530 ppm CO<sub>2</sub>.

The outdoor concentrations of  $NH<sub>3</sub>$  and  $CH<sub>4</sub>$  were close to ambient levels found in other studies (Table 3). Outdoor concentrations denote gas concentrations measured just outside the building. Ambient air concentrations refer to the general atmospheric concentrations. On average, NH3 outdoor concentrations (0.9–1.1 ppm) amounted to about 14% of the indoor concentration (6.4–7.3 ppm) when the cows were confined in the building during the winter. An  $NH<sub>3</sub>$  outdoor concentration that amounted to 20% or more of the indoor concentration has been measured in other dairy cattle buildings (Groot Koerkamp et al., 1998). High outdoor concentrations of  $N_2O$  and  $CO_2$  were found compared to ambient air levels (EEA, 2008). The biggest difference was for  $CO<sub>2</sub>$  where the ambient air concentration amounted to 75–87% of the measured outdoor concentrations. The measured outdoor concentrations could have been influenced by emissions from the building, manure storage tanks, silos and grazing fields.



Fig. 3 – The concentrations of CO<sub>2</sub>, CH<sub>4</sub>, NH<sub>3</sub> and N<sub>2</sub>O at nine indoor sampling locations (P3–P8 and P10–P12) and two locations (P1, P2) outside the dairy cattle building in January, 2007. The box represents the inter-quartile range with the median indicated at the centre. The whiskers extend to the most extreme concentration which is not more than 1.5 times the inter-quartile range from both ends of the box.

#### 3.2. Ventilation and emission rates

#### 3.2.1. Ventilation rate

The mean monthly VRs were in the range of 250– 265  $\mathrm{m^{3}LU^{-1}h^{-1}}$  in winter and 401  $\mathrm{m^{3}LU^{-1}h^{-1}}$  in spring (Table 4). Mean VRs of 341  $\mathrm{m^{3} \, LU^{-1} \, h^{-1}}$  in winter and 404  $\mathrm{m^{3} \, LU^{-1} \, h^{-1}}$ in summer have been measured in Northern Europe (Seedorf



et al., 1998). The accuracy in estimating emissions is highly dependant on the accuracy in measuring VRs. In order to estimate possible errors in the current measurement method, the VR was also estimated when the outdoor  $CO<sub>2</sub>$  concentration was fixed to the ambient level of 381 ppm. The recalculated VRs were lower: 204–234  $\mathrm{m}^{3} \, \mathrm{L} \mathrm{U}^{-1} \, \mathrm{h}^{-1}$  in winter and 320  $\mathrm{m}^{3} \, \mathrm{L} \mathrm{U}^{-1} \, \mathrm{h}^{-1}$ in spring.





a 154 cows were lactating.

b Not considered in the total HPU since the calves were taken to another building after 1 week from delivery.

c One HPU is 1000 W of total heat at 20°C. Pregnancy contribution to total HPU was not considered.  $\tilde{y}$ : Mean. SD: standard deviation.

d  $1 LU = 500 kg$  animal weight.

The  $CO<sub>2</sub>$  mass balance method for estimating VR has been used successfully in situations where all the  $CO<sub>2</sub>$  in the building comes from animal respiration with negligible contribution from other sources (Van Ouwerkerk & Pedersen, 1994; Pedersen et al., 1998; CIGR, 2002). A good agreement between the VRs on  $CO<sub>2</sub>$  basis and measured using fans as a 24-h average has been found with a ratio between 1.02 and 1.17 (De Sousa & Pedersen, 2004). This method is meant to be applicable in buildings without deep litter where the  $CO<sub>2</sub>$ release from manure has been found to be below 4% of the total production (Aarnink et al., 1992; Van't Klooster & Heitlager, 1994; De Sousa & Pedersen, 2004). However, it may give erroneous results in deep litter buildings where  $CO<sub>2</sub>$  production from the manure can be almost at the same level as that from the animals (Van't Klooster & Heitlager, 1994; Jeppsson, 2000). However, a controversy still remains as  $CO<sub>2</sub>$  release from a liquid system (manure stored beneath the floor) has been reported to contribute about 37.5% to the total release in the building (Ni et al., 1999). In our study, manure was removed twice a day and considerable  $CO<sub>2</sub>$  release from the manure is probably less.

#### 3.2.2. Ammonia emission

The emission profile of  $NH<sub>3</sub>$  with significantly visible emission peaks is shown in Fig. 4. The peaks occurred in the mornings and in the evenings, and were probably related to mixing and removal of manure from the pit beneath the slatted floor to external storage tanks. The peaks could also be related to increased activity in the building when the cows were fed and milked. During this time, there is increased urination, defecation and mixing of the manure on the slatted floor. The mean monthly emission rates (measurement period values) are presented in Table 5. The lower NH $_3$  emission of 0.89 gLU $^{-1}\rm{h}^{-1}$ during the housing period in May could be due to lower amounts of manure in the building and less animal activity because the cows were grazing during part of the day. Measured  $NH<sub>3</sub>$  emissions (0.89–1.13 g LU $^{-1}\,\rm h^{-1}$ ) corresponded to an average nitrogen loss of about 0.02  $\rm kgNLU^{-1}d^{-1}$  from the manure. Based on an estimated 0.36 kg N LU $^{-1}\,\mathrm{d}^{-1}$  nitrogen content in the manure, the  $NH<sub>3</sub>$  emission corresponded to a nitrogen loss of 5.6%. In a tied stall dairy barn with sawdust on the floor, 4% of the nitrogen in the manure was found to be lost through  $NH<sub>3</sub>$ emission (Kaasik et al., 2002). In another study, in a barn with tied dairy cattle,  $NH<sub>3</sub>$  release corresponded to an average nitrogen loss of 4% (Gustafsson et al., 2005).

The measured NH $_3$  emission (0.89–1.13 g LU $^{-1}\rm{h}^{-1}$ ) was within the limits recorded in four European countries ranging from  $0.84$   $g$  LU<sup>-1</sup>  $h^{-1}$  in Denmark to  $1.77$   $g$  LU<sup>-1</sup>  $h^{-1}$  in the Netherlands (Groot Koerkamp et al., 1998). Sampling with a multi-gas analyser, and using the decay method for VR calculation, NH<sub>3</sub> emissions of  $1.62 \text{ gLU}^{-1} \text{h}^{-1}$  have been measured in a cattle building in Germany (Snell et al., 2003). In the United Kingdom, an NH<sub>3</sub> emission rate of 1.02  $\rm g \, LU^{-1} \, h^{-1}$ was measured in a slurry-based dairy cattle building (Demmers et al., 2001). In HPU, the NH<sub>3</sub> emission was in the range of 0.79– 0.98 g HPU<sup>-1</sup> h<sup>-1</sup> (Table 5). Emissions of 0.58-1.13 g HPU<sup>-1</sup> h<sup>-1</sup> have been reported in Denmark (Zhang et al., 2005).



Fig. 4 - Diurnal variations in  $NH<sub>3</sub>$  and CH<sub>4</sub> emission rates from the dairy cattle building during a period in February, 2007 (1 LU  $=$  500 kg animal weight).

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Table 5 – Mean monthly emissions of NH <sub>3</sub> and CH <sub>4</sub> from the dairy cattle building								
Month	$NH_3^a$ , g LU <sup>-1</sup> h <sup>-1</sup>		$NH3b$ , gHPU <sup>-1</sup> h <sup>-1</sup>		$CH_4$ , g LU $^{-1}$ h $^{-1}$		$CH_4$ , gHPU $^{-1}$ h $^{-1}$	
		<b>SD</b>		SD.	νĩ	<b>SD</b>		<b>SD</b>
December 2006 (20th-31st)	0.99	0.2	0.88	0.2	11.9	3	10.5	3
January 2007 (11th-31st)	1.03	0.3	0.89	0.2	13.0	6	11.2	
February 2007	1.03	0.3	0.90	0.3	11.6	4	10.1	3
March 2007	1.13	0.4	0.98	0.3	11.3		9.7	4
May 2007 (22nd-29th) (5 p.m.-9 a.m.)	0.89	0.3	0.79	0.3	9.0	3	8.0	3
<i><u><b><u>u Moon</u></b></u></i> CD: standard deviation								

ỹ: Mean. SD: standard deviation.

a  $1$  LU = 500 kg animal weight.

b One HPU is 1000 W of total heat at  $20^{\circ}$ C.

#### 3.2.3. Methane emission

 $CH<sub>4</sub>$  emission had a diurnal variation with distinct peaks, as shown in Fig. 4. The emission of  $CH<sub>4</sub>$  when the cows were indoors between December and March was 11.3–13 g LU $^{-1}\rm h^{-1}$ (Table 5). A lower emission of 9 gLU $^{-1}\rm{h}^{-1}$  was measured during the housing period in May (between 5 p.m. and 9 a.m.).  $CH<sub>4</sub>$  in a cow barn is mainly produced from enteric fermentation (Monteny et al., 2006). The measured  $CH_4$  emissions of 9–13 g LU $^{-1}\,\rm h^{-1}$  (Table 5) corresponded to 79–114 kg LU $^{-1}\,\rm yr^{-1}$ . An emission factor of 109 $\,\mathrm{kg\,head^{-1}\,yr^{-1}}$  from enteric fermentation has been reported for dairy cattle in Western Europe (IPCC, 2006). In a study with dairy cows in a respiratory chamber, CH<sub>4</sub> emission in the range of 5.4–12.1 g LU $^{-1}\rm h^{-1}$  has been reported (Jungbluth et al., 2001). Considerably higher CH<sub>4</sub> emissions of 16.23  $\rm g$ LU $^{-1}$ h $^{-1}$  have been measured from a dairy barn with cubicles and a combination of slatted and top floor systems (Snell et al., 2003). In the present study, the CH $_4$  emission calculated per HPU was 8.0–11.2 gHPU $^{-1}\rm{h}^{-1}$ (Table 5). In another study, emissions of 10.18– 14.13 g HPU $^{-1}\rm h^{-1}$  were found in a building with a slatted floor and scrapers in the manure gutter and 9.46–14.23 g HPU $^{-1}\rm h^{-1}$ was found in a building with back flushing (Zhang et al., 2005).



Fig. 5 – Correlation between the enhanced concentrations of  $CO<sub>2</sub>$  and  $CH<sub>4</sub>$  in December, 2006. The enhanced  $CH<sub>4</sub>$  and  $CO<sub>2</sub>$  concentrations denote the difference between indoor and outdoor concentrations.

Generally, emission rates differ between countries, seasons and building types. These variations, as well as differences concerning instruments and measurement methods, make it difficult to compare data from different studies.

## 3.3. Correlations

The relationships between the measured gas concentrations in December were analysed. A strong correlation was found between the enhanced  $CH_4$  and the enhanced  $CO_2$  indoor concentrations (Fig. 5).

Though less significant, positive correlations were also found for the enhanced concentrations of  $CH_4$  versus  $NH_3$  $(R^2 = 0.68$ , where  $R^2$  is the coefficient of determination) and for



Fig. 6 – Relationships between the VR and the daily indoor concentrations of  $NH<sub>3</sub>$  and  $CH<sub>4</sub>$  when the cows were confined in the building (December 2006–March 2007).

 $CO<sub>2</sub>$  versus NH<sub>3</sub> ( $R<sup>2</sup> = 0.61$ ). Although these gases have varying major emission sources, the positive correlations could be due to routine activities in the building, which resulted in simultaneous concentration spikes.

The relationship between  $NH<sub>3</sub>$  (or CH<sub>4</sub>) indoor concentration and the VR is shown in Fig. 6. An inverse linear relationship on the natural logarithmic scale was obtained with low concentrations corresponding to high VRs.

## 4. Conclusion

In this investigation, the concentrations of  $CO<sub>2</sub>$ ,  $CH<sub>4</sub>$ ,  $NH<sub>3</sub>$  and N2O were measured in a dairy barn with frequent removal of liquid manure. The VR was calculated based on  $CO<sub>2</sub>$  mass balance and the emissions of  $CH_4$  and  $NH_3$  were subsequently determined. The following conclusions could be drawn:

- $\bullet$  The concentrations of CO<sub>2</sub>, NH<sub>3</sub> and CH<sub>4</sub> varied considerably in time and place inside the naturally-ventilated barn.
- Multi-location sampling during short-term measurements increases the possibility for measuring representative gas concentrations and emissions.
- Single location sampling during long-term measurements may reveal representative values of concentrations and emissions if the sampling location is strategically chosen.
- $\bullet$  Low concentrations of N<sub>2</sub>O were measured, suggesting that cow barns with liquid manure systems and daily or frequent manure removal into external storage tanks do not constitute a major source of  $N_2O$ .
- $\bullet$  NH<sub>3</sub> emission from the building was in the range of 0.89-1.13 g LU $^{-1}\rm h^{-1}$  and corresponded to a mean nitrogen loss of about 0.02 kg $\rm NLU^{-1}d^{-1}$  from the manure. Considering an estimated 0.36 kg N LU $^{-1}$  d $^{-1}$  nitrogen content in the manure, the NH<sub>3</sub> emission corresponded to a nitrogen loss of 5.6%.
- A strong positive correlation was found between the enhanced  $CO<sub>2</sub>$  and  $CH<sub>4</sub>$  concentrations.

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