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Methods for measuring gas emissions from naturally ventilated livestock buildings: developments over the last decade and perspectives for improvement

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Abstract

The objectives of this paper are: 1) to give an overview of the development of methods for measuring emission rates from naturally ventilated livestock buildings over the last decade, 2) to identify and evaluate strengths and weaknesses, 3) to summarize and conclude the current state-of- art of available measurement concepts and their perspectives for improvement. The methods reviewed include determination of concentration and air exchange rate separately, tracer gas ratio, passive flux samplers, flux chambers, and combined downwind measurement and dispersion modelling. It is concluded that passive flux samplers, flux chambers and combined measurement and dispersion modeling, are useful, but for limited fields of application only and require further development and validation against reference methods. The most robust method to investigate emission rates available at this stage is the tracer gas ratio method, but improvements are required. They include more detailed estimates of CO_2 release rates (when using CO_2 as a tracer) and research into optimising dosing performance of tracer gas release systems. The reliability of tracer gas ratio methods applied in buildings with large ventilation openings needs to be improved by a more profound understanding of tracer-pollutant ratios and their spatial variability, and the development of improved sampling methods for concentration ratios. There is a need for a field reference method against which other methods can be evaluated. None of the discussed measurement methods can be marked as a solid reference for all conditions; tracer gas ratio methods are the most likely candidate but need further improvement.

Keywords: livestock housing; gas emissions; measurement methods; natural ventilation

1. Introduction

Gaseous emissions from livestock buildings and their environmental effects have been studied for a number of decades over the world to provide accurate information on their magnitude and relevance and, where needed, to develop options for mitigation. The majority of methods for measuring gaseous emission from buildings that are nowadays available have been mainly developed in the eighties and nineties of the last century. From its early beginnings it has been widely acknowledged that the quantification of emissions from naturally ventilated (NV) buildings is a more complicated and challenging task compared to mechanically ventilated barns, due to the difficulties to accurately determine airflow rates. Airflow patterns in NV buildings are a function of many parameters including wind speed and direction, temperature, animal heat production, air inlet and outlet constructions and it results in gas concentrations with a high and incongruous variability in space and time. So far no clear and undisputed reference field method for emission measurement can be identified in the variety of available methodological approaches. As a result information on emissions from NV buildings for the main animal categories is scarce and subject to discussion on measurement uncertainty. A typical example is the large range in reported ammonia emissions of dairy cattle in basically similar cubicle loose housings (Schrade, 2009; Pereira, Misselbrook, Chadwick, Coutinho, Trindade, 2010; Samer et al., 2011), where it is unclear whether the reported variations in annual emission factors, differing up to a factor two, can be attributed to effects of management, housing lay-out and climate or different measurement concepts. Given 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, requiring a stronger methodological basis than currently available.

Phillips, Scholtens, Lee, Garland, and Sneath (2001) provided an extensive review of measurement methods for ammonia emission rates from NV livestock buildings that included a comprehensive overview of research on methods developed up to 2000. Over the last decade, new insights on existing methods and some new methodological concepts have been developed and presented in studies at laboratory scale and field scale, comparative field studies with different methods, and research into emission factors of NV buildings. The objectives of this paper are:

(a) To give an overview of the development of new and existing methods for measuring emission rates from NV buildings over the last decade;

(b) To identify and evaluate theoretical and practical strengths and weaknesses of these concepts when applied in NV buildings, and considerations for further research and developments;

(c) To summarize and conclude the current state-of- art of available measurement concepts taking into account the relationships with different objectives for establishing emission research, discuss major conceptual and operational problems and their perspectives for improvement.

The research studies reported on this subject have been mainly focused on NV buildings for cattle and on ammonia emissions. However the scope of this paper is not exclusively restricted to cattle and ammonia, but attempts where possible to present a wider perspective to other animal categories and emissions. The focus in the discussion of measurement methods will be on their concept in relation to the specific characteristics of NV buildings. Where appropriate, more detailed information on measurement principles and performances of analysers and other devices will be given.

The review is organized in categories of measurement methods along the lines of the main measurement principles.

2. Developments in measurement methods for NV livestock buildings

Methodological research into the quantification of emissions from NV livestock buildings can be aimed at developing appropriate methods for the following applications:

a) scientific purposes: achieving knowledge on processes involved in the formation and emission of pollutants;

b) mitigation assessment: evaluating the effects of mitigation options against a reference;

c) quantification of emission factors: measuring emissions of buildings and housing systems for regulatory purposes.

These applications require methods that may differ in required time resolution, sampling strategy, overall accuracy and precision, and affordable costs. Moreover choice of methods and sampling strategies may also depend on the possibility to measure emissions of different pollutants simultaneously. Earlier extended reviews of the different existing methods for measuring the concentration, ventilation rate and emissions of different pollutants from livestock buildings can be found in the literature (Arogo, Westerman, Heber, Robarge, Classen, 2001; Hofschreuder, Mosquera, Hol, Ogink, 2003; Mosquera et al., 2002; Mosquera, Monteny, Erisman, 2005; Ni & Heber, 2001; Phillips, Scholtens, Lee, Garland, Sneath, 2001; Scholtens, 1993; Van ‘t Klooster, Heitlager, Van Gastel, 1992). Our overview is organized based on the main measurement concepts for emissions from NV livestock buildings. The following main categories can be distinguished:

1) emission calculated as a product of measured differential concentration and ventilation rate
2) emission calculated using a tracer gas ratio method
3) emission directly measured using passive flux samplers
4) flux chamber methods
5) combination of downwind measurement and dispersion modelling

2.1. Emission calculated as a product of differential concentration and ventilation rate

In this section we restrict our overview to the measurement of ventilation rates in NV buildings, being the most challenging component when applying this method. A number of techniques have been used to measure the ventilation rate from NV buildings, including: pressure difference, hot wire or ultrasonic anemometers and tracer gas methods.

2.1.1. Pressure difference

The pressure difference method is a local measurement method which relies on using the pressure difference between the air inside and outside the building across the ventilation openings to determine the ventilation rate. The main drawbacks of this approach are the non-uniform distribution of the pressure differences and the velocity profile across the ventilation openings and through time, in particular at low wind speed conditions. With very large openings in a modern cattle building, uncertainty of the method is increased. This method is not considered reliable to estimate the ventilation rate in NV buildings (Demmers et al., 2001; Samer et al., 2011; Ozcan, Vranken, Berckmans, 2007).

2.1.2. Anemometers

Another approach is to measure the air velocities at ventilation openings and to integrate the velocities and the opening areas into a ventilation rate. Hot wire anemometers can be used to measure the air velocity in a ventilation opening based on the heat loss of the hot wire element by force convection when air passes through it. However, it has to be a three dimensional sensor, since the direction of airflow passing the opening is varied depending on outdoor wind conditions. It gives a local measurement of the air velocity and consequently of the ventilation rate and, therefore, the main drawback is the representativeness of the measurement for the entire ventilation openings of a building (Ozcan, Vranken, Berckmans, 2007). It is noted that a three dimensional hot wire anemometer is expensive and not robust for long time field measurements.

Three dimensional ultrasonic wind anemometers can be also used to measure the air velocities at the ventilation openings. By multiplying the measured wind velocity with the represented opening area, the local ventilation rate can be estimated. Comparing to hotwire anemometer, an ultrasonic sensor is inexpensive and robust, and suitable for field measurements. However, it also gives only local measurements and has as main drawback for dealing with the high variation in wind velocity at different positions of the ventilation openings. Increasing the numbers of the sensors may overcome the drawback. According to literature, however, few scientific reports on the subject can be found in field measurements.

2.1.3. Tracer gas

Tracer gas techniques have become widely used to indirectly measure the ventilation rates in buildings. The basic principle of the method is the conservation of mass (of tracer gas) in the ventilation process: by monitoring the injection and concentration of the tracer, the exchange of air in the building can be determined.

A number of factors should be taken into account when applying the tracer gas method, including 1) approach/method, 2) type of tracer gas, 3) release and sampling, and 4) validation against a reference method

2.1.3.1. Tracer gas methods defined by injection and monitoring.

The tracer gas method can be applied in following three different ways.

Tracer gas decay method. In this method, a tracer gas is initially injected into the building and mixed with the indoor air. After mixing of the tracer gas to a constant concentration level, the injection is stopped and the concentration is monitored during the decay period without further injection to calculate the air exchange rate. The rate of decay is the least disruptive tracer method and requires the minimum of equipment to perform a test. However, this method does not give a continuous indication of the ventilation rate and is not suitable for long-term airflow measurements.

Constant concentration method. In this method a tracer gas is distributed in the air of a building to a certain concentration level. The amount of tracer gas required to maintain a constant concentration in the building is then a direct function of the ventilation rate. In this method it is necessary to continuously monitor both the injection rate and the tracer gas concentration in the room space. To implement the constant concentration concept in practice, it is necessary to accurately measure the rate of tracer gas injection to maintain a given level of concentration throughout the whole enclosure. This makes this method not suitable for research in buildings with unstable (continuously changing) ventilation rates, as is the case for NV livestock buildings.

Constant injection rate. In this method the building is initially charged with tracer gas and then the injection rate is set to a constant value that produces an easily measurable concentration, within the detection range of the measuring equipment. The constant injection method is the most commonly used technique to estimate the ventilation rate in NV buildings, because it allows semi-continuous measurement of ventilation rates (Scholtens, Van der Heiden-de Vos, Huis in ‘t Veld, 1996).

2.1.3.2. Choosing tracer gas.

The tracer gas should fulfil the following requirements:

- Low, constant background concentration in ambient air
- Non-toxic to humans or animals at the concentrations injected in the building
- Safe regarding fire hazard and explosion
- Possible to measure at low concentrations with existing equipment
- Chemically inert
- Cheap and commercially available

- Good mixing properties with ambient air (e.g. by mixing the tracer gas with air before being injected in the livestock building)

Tracer gases used in the literature include nitrous oxide (N_2O; Demmers, Burgess, Phillips, Clark, Wathes, 2000), carbon monoxide (CO; Demmers et al., 2001), carbon dioxide (CO_2; Müller, Möller, Gläser, Cespiva, 2007; Müller et al., 2007; Ngwabie, Jeppsson, Nimmermark, Swensson, Gustafsson, 2009; Ngwabie, Jeppsson, Gustafsson, Nimmermark, 2011; Zhang et al., 2005, Samer et al., 2011), tetrahydrothiophene (C_4H_8S; Lung, Müller, Gläser, Möller, 2002), perfluorocarbon (Okuyama, Onishi, Tanabe, Kashihara, 2009), sulphur hexafluoride (SF_6; Pinares-Patino & Clarck, 2008; Scholtens, Dore, Jones, Lee, Phillips, 2004; Schrade et al., 2007; Schrade, Keck, Hartung, 2008; Müller, Möller, Gläser, Cespiva, 2007; Van Duinkerken, André, Smits, Monteny, Sebek, 2005; Van Duinkerken, Smits, André, Sebek, Dijkstra, 2011; Zhang et al., 2005; Snell, Seipelt, Van den Weghe, 2003), trifluormethyl sulphur pentafluoride (SF_5CF_3; Schrade et al., 2007; Schrade, Keck, Hartung, 2008), and krypton (^{85}Kr; Müller, Möller, Gläser, Cespiva, 2007; Müller et al., 2007; Samer et al., 2011; Lung, Müller, Gläser, Möller, 2002).

The CO_2 mass balance method is a particular form of tracer gas method. In this approach, the tracer gas (CO_2) is not artificially injected in the building, but naturally released by the animals (and the manure) in the building. To determine the ventilation rate from the building by this method the calculation rules of the International Commission of Agricultural and Biosystems Engineering may be used (CIGR, 2002; Pedersen et al., 2008), which provide prediction equations for the CO_2 production of the animals. By using the metabolically produced CO_2 as tracer, this method benefits from the very homogeneous distribution of CO_2 by animal sources throughout the building, providing a better mixing of the tracer (CO_2) and air than normally at reasonable costs can be achieved by injection systems. The accuracy of this method depends on the estimation of the CO_2 production of the animals, which varies with animal breed, weight, activity, productivity and pregnancy. In addition, due to the generally small difference in CO_2 concentration between inlet and outlet air, CO_2 concentrations should be measured accurately. In deep-litter buildings the CO_2 production from the manure can be almost at the same level as that from the animals (Van ‘t Klooster & Heitlager, 1994; Jeppsson, 2000), and cannot be ignored, yet its CO_2 release is not easily determined. In buildings without deep-litter where the CO_2 release from manure is less than 5% of the total CO_2 production (Aarnink, Van Ouwerkerk, Verstegen, 1992; Van ‘t Klooster & Heitlager, 1994; Schneider, 1988; De Sousa & Pedersen, 2004; Van Ouwerkerk &

Pedersen, 1994; Pedersen et al., 1998; CIGR, 2002), the CO_2 production from manure may be neglected. The CO_2 mass balance method has been applied to determine the ventilation rate from both mechanically (Blanes & Pedersen, 2005; Calvet, Estellés, Cambra-López, Torres, Van den Weghe, 2011; Li et al., 2004, 2005; Pedersen et al., 1998; Xin et al., 2006) and from NV (Van ‘t Klooster & Heitlager, 1994; Zhang et al., 2005; Mosquera et al., 2010) livestock buildings.

2.1.3.3. Release and sampling.

The tracer gas should be injected in the animal house in a way that mimics the emission sources being measured. Additionally, the tracer gas and the gas being measured should disperse in a similar way. Finally, the concentration measured at the sampling point(s) should be representative for the average concentration in the building. Van Buggenhout et al. (2009) tested the tracer gas rate of decay method in a mechanically ventilated scale model where a standardized orifice served as reference method for ventilation rate. For single sampling points distributed throughout the compartment they showed large (up to 86%) measurement deviations when calculated on basis of decay curves. The lowest errors (3 – 10% deviation at varying ventilation rates) were measured at the outlet position. Because perfect mixing is seldom the case in a NV livestock building, selection of the sample location(s) to measure a representative average building concentration is critical when using this method. The outlets of the building can normally be considered as the most representative sampling location. However, defining a building opening as an inlet or as an outlet may be challenging in situations with rapidly changing wind direction conditions. In this case, mixing of ventilated air and ingoing air may lead to lower tracer gas concentration at the sample location, resulting in an overestimation of the ventilation rate.

2.1.3.4. Validation against a reference method.

The tracer gas method has been evaluated against different methods. Comparison with mechanical ventilation is preferred because of the lower errors of the reference method (calibrated fan) used. The results presented in Table 1 show in general a good agreement between the tracer gas method and the reference method for buildings with mechanical ventilation. In NV buildings, no reference method is available to evaluate the accuracy of the tracer gas method. Table 1 shows a review of studies focusing on the comparison between the tracer gas method and the CO_2 mass balance method, also commonly applied to estimate the ventilation rate in NV livestock buildings. Differences among these two methods are substantial, with the tracer gas method resulting in higher ventilation rate estimates compared to the CO_2 mass balance method. These differences may be partly explained by a possible overestimation of the ventilation rate when using the tracer gas method (see 2.1.3.3), and a possible underestimation of the ventilation rate when using the CO_2 mass balance method (e.g. by not fully accounting for the CO_2 production from the manure).

Table 1: Validation of tracer gas methods in livestock buildings

Method tested	Reference method	Comparison	Literature
Constant injection	Calibrated fan	-6 to -12%	Demmers (2007)
Constant injection^1	Calibrated fan	< ±4%	Mosquera , Hol, and Huis in ‘t Veld (2008)
Constant injection^2	Calibrated fan	> +50%	Mosquera , Hol, and Huis in ‘t Veld (2008)
CO_2 balance	Calibrated fan	+2 to +17%	De Sousa and Pedersen (2004)
CO_2 balance	Calibrated fan	-3% to +8%	Xin et al. (2009)
Constant injection	CO_2 balance	+75%	Mosquera, Hol, and Groenestein (2010)
Decay method	CO_2 balance	+100%	Samer et al. (2011)

1) sampling tracer gas at ventilation exhaust 2) sampling in middle section of the compartment

2.2 Emission calculated using a tracer gas ratio method

The tracer gas ratio method has been widely used to measure emissions from NV livestock buildings (Huis in ‘t Veld & Groot Koerkamp, 2001; Huis in ‘t Veld & Monteny, 2003; Huis in ‘t Veld, Monteny, Scholtens, 2001; Monteny, Smits, Van Duinkerken, Mollenhorst, De Boer, 2002; Monteny, Hol, Mosquera, 2005; Mosquera & Ogink, 2004; Mosquera, Hol, Huis in ‘t Veld, 2005; Sonneveld et al., 2008; Snell, Seipelt, Van den Weghe, 2003; Stout & Richard, 2003; Zhang et al, 2005; Smits & Huis in ‘t Veld, 2007; Schrade et al., 2007). In the tracer gas ratio method, the ratio of the emission rate of the pollutant (EP) and the tracer gas (ET) is assumed to be equal to the ratio of the pollutant and tracer gas concentration C_p and C_t, both corrected for background concentrations, C_{bp} and C_{bt} respectively. In the ideal case of continuous homogenous mixing throughout the building this relation applies for any particular location in the building:

$$EP = ET\ (C_p - C_{bp})/(C_t - C_{bt}) \qquad \text{(Eq. 1)}$$

In less ideal and more practical situations the concentration ratio in the outlet passage can be assumed to be equal to the emission rate ratio of pollutant and tracer. The ratio method can be considered as a special case of the constant injection method, with the single purpose to have an accurate estimate for the pollutant emission only and not for ventilation rate *per se*. The ratio approach requires (Scholtens, Dore, Jones, Lee, Phillips, 2004):

- Similar atmospheric transport and dispersion properties of the tracer and pollutant over the distance and time scales under consideration
- The loss of either compound through chemical removal or deposition must be similar over the distance and time scales under consideration
- The source pattern of the two species must be similar from the perspective of the measurement location

The tracer gas ratio method can be applied in two different ways, either by measuring inside the building (internal tracer gas ratio method) or downwind the building (external tracer gas ratio method). The internal tracer gas ratio method has the advantage of being less dependent on changes to the wind direction than the external version, and is capable of measuring individual barn sources within a farm, which is not possible in case of the external tracer gas ratio method. However, the internal tracer gas ratio method demands a much better similarity in source patterns of tracer and the pollutant of interest than is necessary in the external version where mixing can take place downwind. This means that more care should be taken in designing and installing a tracer gas release system. When the external tracer gas ratio method is applied to estimate the pollutant emission from a group of sources in a farm and the tracer gas is released in only one source of the group, the position at which the concentrations are measured needs to be sufficiently downwind to allow the group of sources to be considered as a point source. In practice this means measuring downwind at a distance of at least 10 times the overall width of the group of sources.

The main advantage of the tracer gas ratio method is that it can be used to estimate the pollutant emission directly without the need to calculate the ventilation rate. Complete mixing of the air within the building is therefore not a requirement as long as the tracer and pollutant themselves are properly mixed (e.g. by mixing the tracer gas with air before being injected into the livestock building). Similar to the use of tracers for determination of ventilation rate, the most representative sampling location will be situated near to the outlet passage. However, as stated in section 2.1.3.3., in some cases (e.g. by rapidly changing wind direction conditions), it may not be easy to define whether a building opening is behaving as an inlet or as an outlet. In addition, the measurement locations for background concentrations are also important and should be chosen representatively (e.g. not too close to the building, to avoid building effects; continuous measurements, in particular by rapidly changing wind direction conditions, to filter out values from air coming from the building).

Scholtens, Dore, Jones, Lee, and Phillips (2004) validated both the internal and external methods using SF_6 as a tracer in a NV test building with a known high and low release rate of ammonia from an artificial source. The internal approach was based on sampling tracer and pollutant near the three building openings (both sidewall passages and ridge). During a 1-week test the recovery rate, averaged over all sampling points, amounted 102% for the high release rate. For the low release rate only 78% was recovered, but this figure was considered uncertain as a result of measurement problems in the release rate. The external tracer gas approach showed, during a 1-day measurement period, a bias between -25% and +43% compared to the weighed ammonia release from the source. This bias could not be explained. The low tracer and pollutant concentrations at the external sampling point might make this method more sensitive to instrumental errors than the internal tracer approach. Dore et al. (2004) found significantly higher ammonia emissions from a NV cow house when using the external tracer gas ratio method compared to the internal tracer gas ratio method. They attributed this difference in emissions to incomplete mixing of the tracer (SF_6) and NH_3 within the livestock building.

Demmers et al. (2001) tested the internal tracer gas ratio method (using CH_4 as tracer) against a reference method based on the known release of CO in an empty NV building at a fixed rate. Injection of the tracer by a point and a line source were both studied. They used leeward outlet concentrations of the tracer for determination of emission rates, and found in general a good correlation between the measured and the actual release rates of the reference gas CO. A mean CO recovery rate of 108% for the point source and 102% for a line source was observed, both based on four observations.

Demmers (2007) used heater banks and lay flat tubings that released ammonia at a known rate to simulate animals and their emissions in a NV building. Measured at the building perimeter the emission could be determined within 10% overestimation. Average samplings from the cross section however resulted in large deviations from the actually released ammonia of +/- 30%.

2.3 Emission directly measured using passive flux samplers

The measuring principle of this method and its performance to measure NH_3 emissions is described in detail in Scholtens, Hol, and Phillips (2003a, 2003b). In brief, passive flux samplers rely on capturing NH_3 (using tubes with acid coatings) at a rate proportional to the NH_3 concentration and the wind velocity of the air flowing through it, without the need of a pump or other instrument requiring power supply. They are easy to construct, transport and handle, and have little laboratory requisites, although preparation of samplers may be laborious. In order to apply the samplers in a proper way, the flow around the sampler should be oriented in the direction of the sampler. In addition, existing obstacles should not change the pressure difference along the sampler. The sampler should be first calibrated in a wind tunnel, by measuring the pressure difference through the sampler for different wind speeds. In addition, passive flux samplers are based on the principle of diffusion to a reaction surface and, therefore, need longer sampling periods. This increases the risk of changing wind directions during the measurement period, therefore affecting the applicability of the method. Also, they give and average value over the measurement period and no information on variation in emission patterns through time. Passive flux samplers have been applied to measure NH_3 emissions from both mechanically ventilated (Mosquera, Scholtens, Ogink, 2003) and NV livestock buildings (Dore et al. 2004; Burton, Misselbrook, Welch, Hampson, 2007; Pereira, Misselbrook, Chadwick, Coutinho, Trindade, 2010). A comparison between passive flux samplers and a reference method for mechanically ventilated buildings (Mosquera, Scholtens, Ogink, 2003) showed no significant differences between NH_3 emission estimates of passive flux samplers and the reference method, although passive samplers seemed to underestimate (on average) the NH_3 emission from the building.

Scholtens, Dore, Jones, Lee, and Phillips (2004) validated the use of passive flux samplers in a simulated animal building with a known release of ammonia from an artificial source. In five separate tests they mounted twelve re-curved flux samplers across the space-boarded area on both sides of the building, and eight re-curved flux samplers evenly spaced along the open roof ridge, applying a minimal exposure time of four hours. On average they found a very constant recovery rate of 66% with a standard deviation between tests of 3%. Given this constant recovery rate they consider this technique feasible for use in practice, if a correction rate for partial recovery is applied, but note that further research is needed to identify the causes of partial recovery.

2.4 Emission calculated using flux chamber methods

Two different types of flux chambers can be defined. In non-steady-state (also known as static or closed) chambers, air inside the chamber is usually re-circulated to allow for a good mixing of all gases in the headspace of the chamber, but no air replacement occurs. The rate of increase in concentration over time inside the chamber is then used to calculate the emission from the surface covered by the chamber. By steady-state (also known as open or dynamic) chambers, the headspace of the chamber is flushed with clean air at a known flow rate (Reichman & Rolston, 2002) from one side of the chamber (inlet) to the other side of the chamber (outlet) passing above the emission surface. A flow rate measurement device is often equipped to monitor the airflow rate and consequently the average air speed through the chamber cross section. The concentration difference between the outlet and the inlet is monitored and used to determine the emission strength of the pollutant from the covered chamber area.

Flux chambers are generally low cost in investment and easy to use. However, they can interfere and influence the emission and exchange processes between the emitting surface and the air in the building. By injecting a tracer gas into the chamber and applying the internal tracer gas approach to calculate the emission (Hensen et al., 2004), measurements can be performed over a short period of time minimizing the (possible) influence of the chamber on the emission processes. Mosquera, Van Dooren, Aarnink, and Ogink (2010) performed NH_3 emission measurements with a dynamic flux chamber at three different (floor) places in a dairy barn and at four airflow levels. They found a positive, non-linear relation between the emission rate and the airflow in the flux chamber. Similar results were found by Wheeler et al. (2010) when comparing NH_3 emission measurements in a mechanically ventilated room by using both a static and a dynamic flux chamber, with the results of direct measurements of room ventilation rate and gas concentration at the exhaust. They found the best results by the non-steady-state flux chamber arriving at 90% to 119% of the emissions measured with the reference method. This chamber overestimated the emissions at higher internal recirculation speeds. Since the test was performed at airflow rates lower than those expected in a dairy cattle barn, measurements at “normal” airflow rates could therefore result in an even higher overestimation of the emissions. The steady-state flux chamber in their study seemed to underestimate the emissions by 14%-50% comparing with the reference.

Spatial variability is also a major limitation for the accurate quantification of emission using flux chambers, unless a large number of measurements is performed in different places across the building. Mosquera, Van Dooren, Aarnink, and Ogink (2010) found a significant difference of the measured emission rates among the different locations in a barn. Measurements also showed a significant variation of ammonia emissions in different measurement days, but not within a measurement day. They concluded that for a good estimation of the ammonia emissions, measurements at different locations in a barn for a number of days, if possible under different weather conditions, are advised. The number of measurements performed was not sufficient to perform a statistical analysis of variability, to determine the proper number of sites to measure.

Flux chambers are suitable to compare emissions from different floor and slurry pit systems based on a case-control approach, as shown by Van Dooren and Mosquera (2010), and by Adviento-Borbe et al (2010). Adviento-Borbe et al (2010) used a non-steady-state flux chamber to evaluate the effect of different diets on the emissions of NH_3, CO_2, CH_4 and N_2O from a NV dairy barn. Van Dooren and Mosquera (2010) measured the emission of three ammonia emission reduction systems (based on floor measures) at three dairy cattle barns in the Netherlands where also a reference floor (concrete slatted floor with slurry pits) was present. Measurements were performed by using a steady-state flux chamber. They concluded that results from flux chamber measurements cannot be directly compared to the absolute emission of the whole building, or for the estimation of the variation between farms.

To use the flux chamber method to estimate a total emission from a NV building, the airflow characteristics at floor level should be investigated and the chamber design as well as measurement protocol should reflect the air speed and turbulent level at floor regions (Mosquera, Van Dooren, Aarnink, Ogink, 2010; Parker et al., 2012). In published literature, however, very few investigations on these issues can be found. The chamber sizes were found to affect emission rates (Frechen, Frey, Wett, Loser, 2004; Hudson et al., 2009). Smaller chamber height could enhance emission rate due to the larger air velocity gradient at the release surface. Saha, Wu, Zhang, and Bjerg (2011) reported a comprehensive investigation of five flux chambers in different dimension using CFD (Computational Fluid Dynamics) method and support by laboratory experimental results. They concluded that the emission mass transfer in the chambers varied with the chamber heights and air velocity above the emission surface.

2.5. Emission calculated by a combination of downwind measurements and dispersion modeling

Pollutant emissions from NV livestock buildings may also be estimated by combining concentration measurements downwind of the building with dispersion models. The Gaussian plume dispersion model is the most common air pollution dispersion model. This model assumes a Gaussian concentration distribution in the horizontal and vertical directions downwind from a source. This model is easy to use, but is not designed for situations where buildings may have a dominant influence on the dispersion process, which is the case close to buildings (distance less than 100 m).

Another alternative to estimate pollutant emissions downwind the livestock building is the use of a backward Lagrangian stochastic model (Flesch, Wilson, Harper, Crenna, Sharpe, 2004; Flesch, Wilson, Harper, Crenna, 2005; Sommer, McGinn, Hao, Larney, 2004) together with one-point or line measurements of wind velocity and concentrations. This model calculates trajectories of air parcels (or particles) upwind from the receptor point and backwards to the emitting source. This technique looks promising as a non-interference measurement technique to measure the whole-farm emissions, but has not been proven yet for complex farm situations. These approaches require measurements to be carried out downwind. Therefore wind conditions determine sampling locations to be chosen for each measurement session, resulting in practical limitations where sampling locations are not accessible.

3. Discussion

The main part of research over the last decade into emissions from NV livestock buildings has been focused on tracer gas related methods as dealt with in 2.1 and 2.2. Other approaches such as the passive flux sampler, flux chamber and combined measurement and dispersion modeling have been further developed but, at this stage, cannot be seen as fully equivalent substitutes because of their restricted field of application and limited validation against reference methods. Passive flux samplers seem to be particularly suited for research into emission factors, but need further study to explain and control their partial recovery rates that so far are not fully understood (Scholtens, Dore, Jones, Lee, Phillips, 2004). They are not suitable for application to measure multiple sources close to each other, or when rapidly changing wind directions may be expected. Flux chambers can be useful in comparing the effects of different floor surface lay-outs on emissions but scaling chamber data to full barn emissions (Wheeler et al., 2010) is difficult due to the large variation of climatic conditions in barns and NV barn designs. Dispersion model based approaches especially have practical limitations with regard to their dependency on suitable wind directions and accessible space around farm complexes, and cannot distinguish between different farm sources. Furthermore they require validation against other methods.

No significant progress could be reported in the development of direct methods for the determination of ventilation rates of NV buildings. Investigated methods all depend on local measurements in the ventilation openings and face problems in dealing with incongruous wind velocity patterns that cause a high spatial and temporal variability, a phenomenon to be considered typical for NV livestock buildings. Here, improvements may possibly be reached by combining local measurements with modeling of airflow patterns in ventilation openings, research on scale models of barns and investigations using CFD method. The latter may require air velocity data generated in field measurements.

Indirect determination of ventilation rates by tracer gas methods make use of the mixing process of tracers and pollutants in the barn that ideally would lead to a completely homogenous distribution of both gases and no spatial variability in gas concentrations, thus allowing a single local measurement point to be sufficient. Van Buggenhout et al. (2009) showed high spatial variations in tracer gas concentrations in a lab setting that would lead to huge errors if estimates would be based on randomly chosen single measurement points. Even in case of using the intensively distributed metabolic CO_2 as tracer, researches in NV cattle barns showed considerable spatial variability of tracer gas concentrations (Bjerg, Zhang, Madsen, Rom, 2010; Ngwabie, Jeppsson, Nimmermark, Swensson, Gustafsson, 2009). Samer et al. (2011) attempted to improve mixing of the tracer gas (krypton) by applying large ceiling fans, but concluded that still incomplete mixing of krypton must have led to a strong bias in their results. The alternative approach is to assume only the gas concentrations in the outlets as representative for the tracer. Validation studies of tracer applications in mechanically ventilated buildings show consistent similar results between anemometer based ventilation rates and tracer estimates based on concentrations at ventilation exhaust (Demmers, 2007; Mosquera, Hol, Groenestein, 2010). However, frequently changing outlet and inlet positions between different ventilation openings of NV livestock buildings are not easily identified (Demmers et al., 2001). Unobserved mixing of ventilated air and ingoing air still may cause large errors in estimated ventilation rates.

Facing the gas mixing limitations we conclude that the most robust method to investigate emission rates available at this stage is the tracer gas ratio method that does not distinguish between ventilation rate and concentrations but yields an emission rate directly. This approach is less demanding with regard to mixing conditions and establishing average gas concentrations in the barn as long as the dispersion pattern of both the tracer gas and the pollutant of interest are similar, both gases are allowed to mix before measuring their concentrations inside the livestock building, and the overall ratio between concentrations of tracer and pollutant can be accurately determined. Similarly to the determination of ventilation rates best representative sampling conditions may be expected in the outlet openings. However, still many practical problems and pitfalls may be encountered in its applications. The performance of the method depends on three basic elements: the accuracy of determining the tracer gas source strength, adequate dosing of the tracer near the full pollutant surface, and representative sampling of gas ratios. With regard to the tracer source strength no problems are to be expected for external tracers where their release can be controlled by orifices and mass flow controllers. The accuracy of determining the source strength of metabolic CO_2 however depends on models as provided by CIGR (2002). Setbacks here are that these models generally predict 24-hours values. Diurnal variations may be monitored but depend on additional recording of animal activity (Pedersen et al., 2008; Ngwabie, Jeppsson, Gustafsson, Nimmermark, 2011). Small additional CO_2 sources, like stored manure, may easily lead to biases of a few percent when a fixed proportion of manure contribution is applied (Pedersen et al., 2008), and more elaborated procedures need to be developed. Mosquera, Hol, and Groenestein (2010) tested the CIGR prediction models for different monogastric animal categories in mechanically ventilated buildings, and found generally high correlations and no systematic deviations between CO_2 derived ventilation rates and values from calibrated fans.

A seemingly clear advantage of metabolic CO_2 over external tracer gases is their evenly spread release in barns with as many release points as animals. Yet it is not clear whether this can be matched or improved by adequately designed injection installations releasing external tracers. Analysing the results of Zhang et al. (2005), Bjerg, Zhang, Madsen, and Rom (2010) noted high correlations between CO_2 and CH_4 at different sampling points across the measuring compartment of a NV cattle building, however correlations with NH_3 were much weaker. Both CO_2 and CH_4 are emitted here from identical release points, i.e. the exhaled air of animals. An ideal tracer release system for NH_3 would have approached these high correlations, indicating that even compared to CO_2 distribution further improvement might be possible. Despite their big potential impact on the overall measurement performance, in many studies the layout of injection systems is not or only superficially specified. We found no research where the performances of differently positioned tracer gas release systems and the frequency and distribution of injection points have been systematically investigated. Considerable improvements can be made here.

Next to adequately distributed and located dosing of tracers, representative sampling of gas concentration ratios remains an important issue. Especially the combination of insufficiently spread injection points of tracers, large sidewall openings and strong wind will lead to risks on biased emission rates. In such conditions, tracers may be systematically missed in sampling lines leading to underestimated tracer concentrations and overestimated emission rates. These or part of these factors may explain the systematically higher ammonia emission rates based on external tracers compared to simultaneously measured emission rates based on metabolic CO_2 in NV cattle barns, as reported by Mosquera, Hol, and Groenestein (2010) and Samer et al. (2011). Windtunnel experiments on small scale models (Ikeguchy & Moriyama, 2010) demonstrate how tracer concentration can be underestimated. Especially barn designs that facilitate cross ventilation may be subject to inadequate sampling. There is a need for criteria that help to identify when inappropriate sampling conditions occur and that indicate how sampling strategies should be modified accordingly. Such criteria can only be developed if a more profound understanding of spatial variability of tracer-pollutant ratios in outlet openings is developed, and the relations between this spatial variability on one hand and airflow patterns and dosing efficiency of the tracer on the other are investigated. With some exemptions (Ikeguchi & Moriyama, 2010) these issues have been hardly researched. The reliability of tracer gas ratio methods would be much improved if such studies would be undertaken extensively.

A basic and common requirement for all methods is that they should be validated in terms of accuracy and precision against an accepted reference method. The reference should be rooted in the SI-system, and in representative conditions, i.e. a NV compartment. Both Demmers et al. (2001) and Scholtens, Dore, Jones, Lee and Phillips (2004) performed such validations by testing measurement methods of ventilation rates against the known release of an artificial source of either carbon monoxide or ammonia gas in an empty NV building. The actual release can be reliably verified by following the decrease in mass of the artificial source. It is noted that this procedure applies to the validation of measuring emission rates (emitted mass per time unit) and not to air volumes and gas concentrations as independent quantities. Conditions for such a validation setup are that emission patterns and airflow fluctuations are representative for a specified type of NV livestock buildings, and that the essential features of investigated methods are diligently described. The validations by Demmers et al. (2001) and Scholtens, Dore, Jones, Lee and Phillips (2004) used the same type NV section with space boarding openings. Estimation of ventilation rates in such a building using tracer gas method will result in less error than in a NV building with much larger side-wall openings. Therefore, in generalization of validation results building types and climate conditions have to be carefully judged case by case. It underlines the need for the development of a more practical field method that can be used as a reference against which other methods can be evaluated. At this stage none of the discussed field methods can be marked as a solid reference for all conditions. Improvements in tracer gas ratio methods, as discussed before, might lead to a widely applicable reference method.

Depending on its field of application, measurement methods may have very different requirements with regard to time resolution and precision of their smallest observational units. For research into emission factors, observations based on daily means as smallest unit can be sufficient, whereas investigation of physical processes in emissions from NV buildings may require time resolutions at hourly basis or even shorter. Similarly, required precision of observational units for research into emission factors of housing systems can be less demanding because sampling strategies are generally the dominant element in the overall precision of calculated factors (Ogink, Mosquera, Melse, 2008). Practical considerations and costs may also lead to different approaches in the mentioned fields of application. Research setups based on short field observation at different farm locations require flexible mobile equipment and personal expertise that is not suited for long term in depth research in experimental settings. All these elements should be taken into account in the development and assessment of measurement methods.

4. Conclusions

In summary we arrive at the following conclusions and perspectives for improvement of determining emission from NV livestock buildings:

- Methods based on passive flux samplers, flux chambers and combined measurement and dispersion modeling, are useful for limited fields of application, but require further development and validation against reference methods;
- It is difficult in practice to apply direct ventilation rate methods to deal with high spatial and temporal variations in ventilation openings. It may be improved by combining local air velocity measurements with modeling of air flow patterns, scale model research and investigation using CFD or other techniques with necessary air velocity data in NV buildings with large openings;
- The most robust method to investigate emission rates available at this stage is the tracer gas ratio method, but methodological improvements are required;
- The accuracy of estimating the release rate of CO_2 can be improved by developing procedures that better estimate metabolic CO_2 related to animal activity and CO_2 contribution from additional manure sources;
- Effective dosing of tracers near emitting surfaces is essential for overall accuracy; research into the dosing performance of tracer gas release system is lacking, and may lead to considerable improvement in measurement accuracy;
- The reliability of tracer gas ratio methods applied in buildings with large ventilation openings needs to be improved; a more profound understanding of tracer-pollutant ratios and their spatial variability is required to develop criteria for representative sampling methods of concentration ratios.
- There is a need for a field reference method for assessment of other methods; at this stage none of the discussed measurement methods can be marked as a solid reference for all climatic conditions and NV building designs; tracer gas ratio methods are the most likely candidate but need further improvement to be applicable under all conditions.

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