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USE OF GAS CONCENTRATION DATA FOR ESTIMATION OF METHANE AND AMMONIA EMISSION FROM NATURALLY VENTILATED LIVESTOCK BUILDINGS

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ABSTRACT Determination of emission of contaminant gases as ammonia and methane from natural ventilated livestock buildings with large opening is a challenge due to the large variety in gas concentration and air velocity in the openings. The close relation between calculated animal heat production and the carbon dioxide production from the animals have in several cases been utilized for estimation of the emission of ammonia and other gasses. Using this method the problem of the complicated air velocity and concentration distribution in the openings is avoided, but still there is considerable doubt about (1) the precision of the estimations (2) the requirement for the length of measuring periods, and (3) the required measuring point number and location. The purpose of this work was to investigate how estimated average gas emission and the precision of the estimation is influenced by different calculation procedures, different measuring period length, different measure point locations, different measure point numbers and different criteria for excluding measuring data. The analyses is based on existing data from a 6 days measuring period in a naturally ventilated, 150 milking cow building, and it shows that the methane emission can be determined with much higher precision than ammonia emission, and, for methane, relatively precise estimations can be based on measure periods as short as 3 hours. This result makes it feasible to investigate the influence of feed composition on methane emission in a relative large number of operating cattle buildings and consequently it can support a development towards reduced greenhouse gas emission from cattle production. For ammonia the analyses did not reveal any shortcuts to precise and simple estimation of emission from naturally ventilated buildings.

Keywords: Methane, ammonia, carbon dioxide, emission, naturally ventilated buildings.

INTRODUCTION Determination of emission of contaminant gases as ammonia and methane from natural ventilated livestock buildings with large opening is a challenge due to the large variety in gas concentration and air velocity in the openings. The close relation between calculated animal heat production (CIGR. 2002) and the carbon dioxide production from the animals (Pedersen et al., 2008) have, in several cases, been utilized in estimation of the emission of ammonia and other gasses (Zhang et al., 2005; Ngwabie et al., 2009). The calculations in published articles have used the average concentration of carbon dioxide in the livestock building over a specific time span, but the accuracy may potentially be increased considerably by using the method proposed by Madsen et al. (2010), where the relation between the concentration of the gas in question, - ex. methane and carbon dioxide is calculated on the individual air samples instead of using mean values for carbon dioxide concentrations. Using this method the problem of the complicated air velocity and concentration distribution in the openings is avoided, but still there is considerable doubt about (1) the precision of the estimations (2) the requirement for the length of measuring periods, and (3) the required measuring point number and location.

The purpose of the work is to investigate how estimated average gas emission and the precision of the estimation is influenced by different calculation procedures, different measuring period length, different measure point locations, different measure point numbers and different criteria for excluding observations.

METHODS: The analyses in this work is based on gas concentration data recorded for six days in a naturally ventilated milk production building (see figure 1), which were included in a larger investigation by Zhang et al., 2005. Details on the measurements is given by Zhang et al., 2005, where the building included in this work is named building 5.

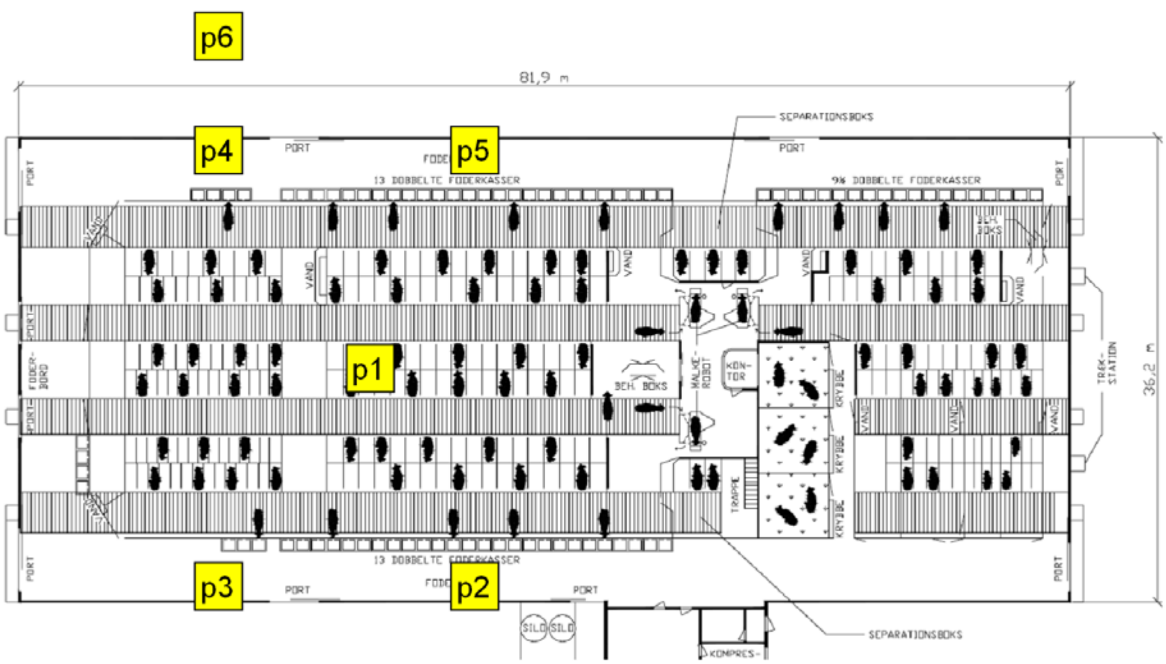

Figure. 1. Measurement section and sensor positions

The investigated data contains measured carbon dioxide, ammonia and methane concentration at 5 locations inside the building and one location outside the building. Figure 1 shows that the measure point inside the building were located close to the openings and distributed in a 20 m long section of the building. Four of the five measurement points were located in corners of the measurement section about 0.25m from the side wall openings and 0.25m below the eaves. Measure point 1 was located in the middle of the measurement section about 1m below the ridge opening.

The data contains measured concentration at each point with a frequency of 9 – 10 measurement per hour. Figure 2 shows the relation between measured values for carbon dioxide and ammonia or methane concentrations for all measurements in all points.

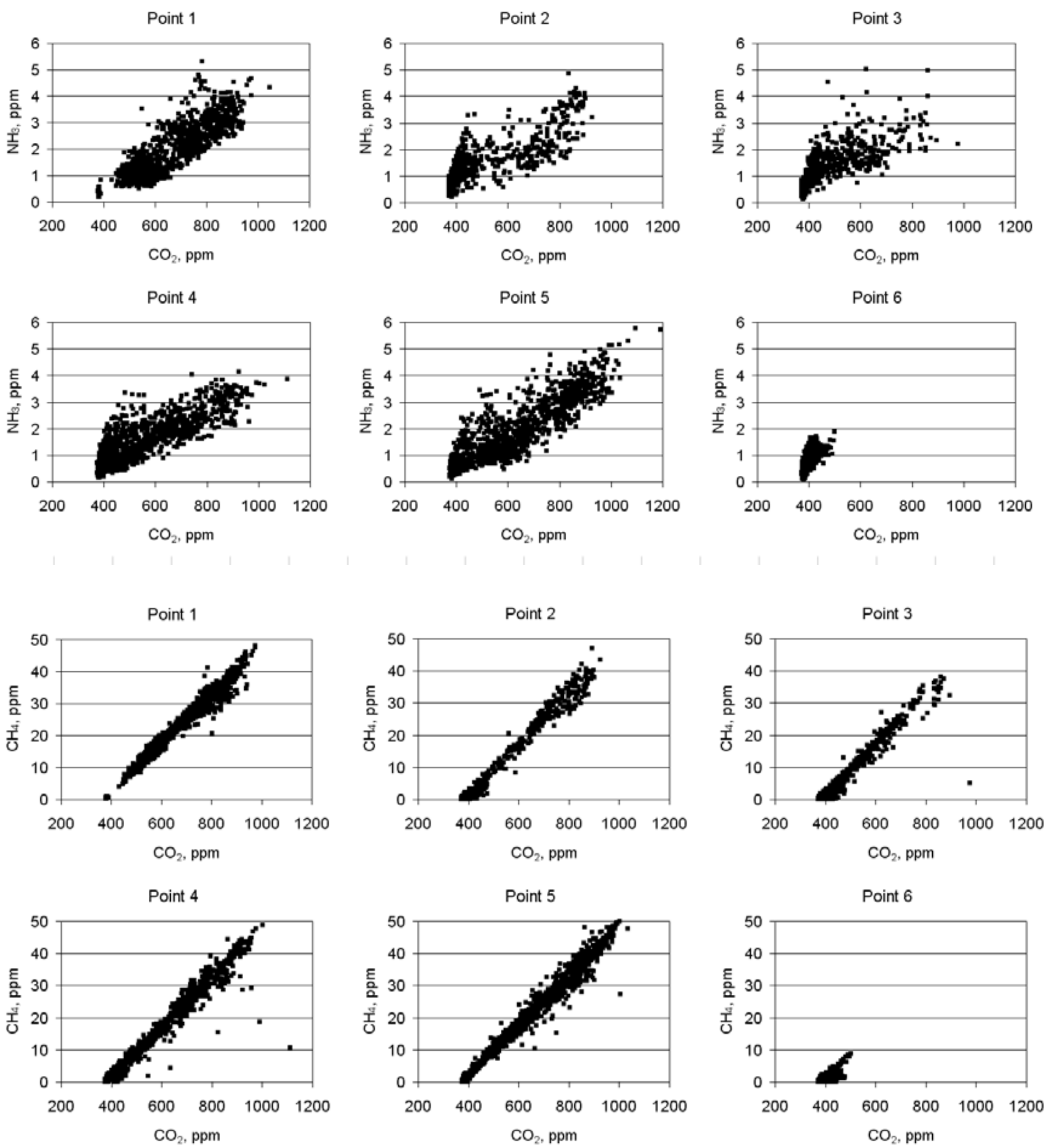

Figure 2. Ammonia (above) or methane (beneath) concentrations as function of carbon dioxide concentration in all measure points.

Determination of ammonia and methane emission can be based on an assumed strong correlation between carbon dioxide production and calculated animal heat production (Pedersen et al 2008). A discussion of the exact value of the expected carbon dioxide production is not an objective of this work and consequently the value of 0.185 m^3HPU^-

$^1h^{-1}$, stated by CIGR 2002, is used in all calculation in this work. Using this value air change per HPU (Heat Production Unit) can be calculated as:

$$Q_{HPU} = \frac{0.185}{\left(c_{CO_2, outlet} - c_{CO_2, inlet}\right) \cdot 10^{-6}} \qquad [1]$$

Where Q_{HPU} is air change, $m^3h^{-1}HPU^{-1}$

c_{CO_2} concentrations of CO_2, ppm

Production of ammonia or methane can be calculated as:

$$P_{gas} = \frac{0.185\left(c_{gas, outlet} - c_{gas, inlet}\right)}{\left(c_{CO_2, outlet} - c_{CO_2, inlet}\right)} \qquad [2]$$

Where P_{gas} Production of ammonia or methane, m^3h^{-1} HPU^{-1}

c_{CO_2} Concentrations of CO_2, ppm

Zhang et al., (2005) assumed that the average of the five indoor located measure points were representative for concentration in outlet. In this work it is investigated how estimated emission is influenced by using one measure point only, and by the different location of measure point.

Zhang et al., (2005) used daily average values to calculate standard deviations of each data set. In this work it is investigated how time averaging of concentrations influences the accuracy of the estimation, expressed as interval of confidence.

The accuracy of carbon dioxide measurements has a large influence on estimated gas production if the difference in carbon dioxide concentration in outlet and inlet is small. In this work observation is generally excluded if this difference is less than 6 ppm, corresponding to specified accuracy of the instrument, and it is investigated how larger thresholds influence the estimated emission.

Zhang et al. (2005) assumed that the outdoor measure point were representative for concentrations in inlets. To simplify measuring procedure it could be considered to use fixed values for outdoor concentration based on measurement from a period before or after the indoor measurement. This simplification makes it possible to conduct measurements with one single channel measuring device only. To simulate that possibility the average concentrations in the outdoor measurements at day 1 and 6 where used as fixed values for inlet concentration (404 ppm CO_2, 0.91 ppm NH_3 and 0.96 ppm CH_3), and the used indoor observation were restricted to measurement at point 1 in the four days period in between.

A further simplification of the measuring procedure would be to use indoor measurement only. This possibility was simulated by assuming no ammonia or methane in the inlet air and by using linear regression to calculate the carbon dioxide concentration where that assumption was fulfilled. For the entire measure period the estimated carbon dioxide concentration corresponding to zero ammonia concentration were 422 ppm ($R^2 = 0.67$), and the corresponding value for zero methane concentration were 371 ppm ($R^2 = 0.97$). Subsequently, the ammonia and methane production was calculated as:

$$P_{gas} = \frac{0.185 c_{gas}}{\left(c_{CO_2} - c_{CO_2,0}\right)} \qquad [3]$$

Where P_{gas} Production of ammonia or methane, m^3h^{-1} HPU^{-1}

$c_{CO_2,0}$ Concentrations of CO_2 where $c_{gas} = 0$, ppm.

Zhang et al., (2005) measured in 6 days periods. The influence of reducing the measuring period to 24, 3 or 1 hour is investigated in this work.

RESULTS AND DISCUSSION

Influence of averaging period Table 1 show that the interval of confidence for gas emission measurements for the entire 6 day period can be reduced for both methane and ammonia by using one hour averaging periods, and further reduced by using each single measurement in the calculation, and thereby considering the composition of each individual sample as a certain portion of air from the cows and a certain portion of outside air (Madsen et al., 2010). Consequently, the following calculation is based on each single measurement.

Table 1. Average value and interval of confidence for estimated ammonia and methane emission using different averaging periods.

Length of averaging period, h	n	NH_3 emission gday^{-1} HPU^{-1}	CH_4 emission, gday^{-1} HPU^{-1}
24	6	14.4 ± 2.31	243 ± 4.41
1	144	14.4 ± 0.89	243 ± 2.54
No averaging	1269	14.4 ± 0.41	243± 1.33

Influence of number and location of measure points used for estimation of outlet concentration. Table 2 shows that using point 1, as the only point for assumed outlet condition, results in similar methane emission, and a about 18 percent lower ammonia emission, than using average value from point 1 to 5. For the wall located points (2-5) a larger number of observations were excluded due to low carbon dioxide concentration difference. Consequently point 1 is used as assumption for outlet condition in the following calculations.

Table 2 also shows that the relative deviation in estimated emission between the single measure point was about 10 times higher for ammonia than for methane. It can be anticipated that the low number of included observation at point 2 and 3 was a result of the dominating wind direction which caused carbon dioxide concentrations differences below the threshold for longer periods. It is calculated that exclusion of those two points would reduce the coefficient of variation between different measure points for methane emission to less than 0.01.

Table 2. Average value and interval of confidence for estimated ammonia and methane emission using different points for assuming outlet conditions.

Measure points used as outlet conditions	n	NH_3 emission gday^{-1} HPU^{-1}	CH_4 emission, gday^{-1} HPU^{-1}
Average of 1-5	1269	14.4 ± 0.41	243 ± 1.33
1	1288	12.3 ± 0.27	241 ± 0.94
2	369	30.4 ± 4.64	260 ± 10.1
3	319	30.9 ± 5.24	263 ± 9.37
4	998	23.1 ± 3.87	248 ± 3.87
5	1034	19.4 ± 1.44	250 ± 3.39
Coefficient of variation between points (n=5)		0.34	0.036

Influence of different criteria for excluding observations Generally in this work observation were excluded if the difference between carbon dioxide concentration in outlet and inlet were less than 6 ppm. This is done to make sure that there is a high concentration of air from the cows and thereby minimizing the influence of the composition of the outside air. Table 3 shows that increasing that limit to 100 ppm has no influence on calculated emissions. If the limit is increased to 400 ppm 85 percent of the observation is excluded but the estimated methane emission is still at the same level.

Table 3. Average value and interval of confidence for estimated ammonia and methane emission using different limits for excluding observations.

Observation excluded if: $c_{CO_2,outlet} - c_{CO_2,inlet}$ <, ppm	n	NH_3 emission gday^{-1} HPU^{-1}	CH_4 emission, gday^{-1} HPU^{-1}
6	1288	12.3 ± 0.27	241 ± 0.94
50	1283	12.3 ± 0.28	241 ± 0.94
100	1227	12.3 ± 0.28	240 ± 0.94
200	726	13.2 ± 0.36	241 ± 1.14
400	195	13.6 ± 0.40	240 ± 1.77

Influence of using assumptions of fixed values for inlet condition Table 4 shows that using the indoor measurement (point 1) for determining inlet condition results in a large deviation in estimated ammonia emission. This is mainly caused by the poor correlation between ammonia concentration and carbon dioxide concentration, which results in assuming a to high carbon dioxide concentration in the inlet. For methane the correlation with carbon dioxide concentration was much better and, consequently, this method resulted in a reduction of estimated methane emission of 7 percent, only. For both gases the deviations were smaller if the fixed values for inlet conditions where estimated from the outdoor measurement (point 6) at day 1 and 6, and the used observation were restricted to the 4 days in between.

Table 4. Average value and interval of confidence for estimated ammonia and methane emission using different assumption for inlet conditions.

Assumed inlet condition	n	NH_3 emission $gday^{-1}$ HPU^{-1}	CH_4 emission, $gday^{-1}$ HPU^{-1}
Point 6	1288	12.3 ± 0.27	241 ± 0.94
Fixed values calculated from point 1*:	1316	27.9 ± 0.73	225 ± 1.17
Fixed values calculated from point 6*:	698	11.8± 0.60	244 ± 1.76

* see methods secton

Influence of using data from reduced measuring periods Table 5 compares estimated emission for each day with the estimation for the entire period.

Table 5. Average value and interval of confidence for estimated ammonia and methane emission at different days.

Day	n	NH_3 emission $gday^{-1}$ HPU^{-1}	CH_4 emission, $gday^{-1}$ HPU^{-1}
1-6	1288	12.3 ± 0.27	241 ± 0.94
1	193	10.5 ± 0.50	242 ± 1.83
2	220	10.6 ±0.75	242 ± 3.17
3	219	10.3 ± 0.50	238 ± 2.25
4	218	13.6 ± 0.60	236 ± 1.91
5	219	15.7 ± 0.82	241 ± 2.07
6	219	13.0 ± 0.47	246 ± 1.98
Coefficient of variation between days (n=6)		0.17	0.013

It can be seen that the relatively deviation in emission estimations between different days is more than 10 times larger for ammonia than for methane. The small difference between daily estimations of methane emission makes it relevant to investigate even shorter measuring periods for methane. Table 6 shows estimated methane emission based on 3 hour periods beginning at 9 am on each day, and 1 hour periods beginning at 2 pm.

Comparison of results from table 5 and 6 shows, that reducing the measuring periods to three hour had a very limited influence on estimated methane emission, and on the deviation between different days. Table 6 shows that a further decrease of the measure period - to one hour, only - results in a significant larger, but still relatively low, variation between days.

Table 6. Average value and interval of confidence for estimated methane emission at different days at a 3 hour period beginning at 9 am on each day, and a 1 hour period beginning at 2 pm.

Day	3 hour periods beginning at 9 am n	$gday^{-1}$ HPU^{-1}	1 hour periods beginning at 2 pm n	$gday^{-1}$ HPU^{-1}
1-6	139	243 ± 3.60	55	243 ± 5.40
1	3	238 ± 32.7	10	244 ± 20.3
2	28	242 ±8.88	9	266 ± 12.1
3	27	246 ± 1.42	9	232 ± 16.1
4	27	242 ± 4.97	9	229 ± 9.87
5	27	247 ± 9.35	9	246 ± 9.05
6	27	236 ± 10.7	9	241 ± 7.29
Coefficient of variation between days (n=6)		0.017		0.054

Influence of estimating inlet concentration from indoor measurements for three hour periods For the 3 hour measuring periods table 7 shows estimated methane emission based on the assumption that inlet condition can be calculated from included measurement by using data from point 1 and linear regression to calculated the carbon dioxide concentration that corresponds to no methane concentration.

Table 7. Average value and interval of confidence for estimated methane emission at different days at a 3 hour period beginning at 9 am, using data from point 1 to calculate inlet concentrations.

Day	CO_2 concentration where CH_4 concentration is zero, ppm	R^2	n	CH_4 emission, $gday^{-1}$ HPU^{-1}
1-6	375	0.97	158	237 ± 5.40
1	375	0.99	27	236 ± 29.7
2	410	0.75	27	307 ± 12.1
3	376	0.87	27	243 ± 5.6
4	392	0.98	27	255 ± 4.01
5	640	0.25	27	678 ± 49.4
6	496	0.78	27	327 ± 6.8
Coefficient of variation between days (n=6)				0.49

For the entire period and on day 1, 3 and 4 there were a high correlation between methane and carbon dioxide concentration and, consequently, the estimated methane emission was at the same level as if other methods where used. The days with poor correlation between methane and carbon dioxide concentration where characterized with small differences in carbon dioxide concentration during the measure period, which might contribute to the poor correlation between methane and carbon dioxide concentration.

Influence of using fixed value assumption based on outdoor measurements for 3 hour periods. For three hour measuring periods table 8 shows the influence of using fixed values for inlet condition based on outdoor measurement in point 6 from 9 am to 10 am. Due to the one hour spent on outdoor measurement the utilized indoor observations were restricted to measurement in point 1 in the two hour period from 10 am to 12 am.

Comparison of table 6 and 8 shows that the relative variation between estimated daily methane emissions increase nearly 3 times, if the first hour of a three hour measuring periods is used for determining inlet concentration, compared with simultaneous indoor and outdoor measurements. But, nevertheless, the level of coefficient of variation between days is still relatively low.

Table 8. Average value and interval of confidence for estimated methane emission at different days using a 3 hour periods beginning at 9 am on each day, and using the first hour to determine fixed values for inlet conditions.

Day	n	$gday^{-1}$ HPU^{-1}
1-6	97	256 ± 5.51
1	19	244 ± 8.77
2	18	253 ± 6.06
3	18	246 ± 4.11
4	18	273 ± 13.7
5	18	233 ± 12.2
6	6	275 ± 17.3
Coefficient of variation between days (n=6)		0.066

CONCLUSIONS Analyses based on data from a 6 days measuring period in a naturally ventilated, building for milking cows, shows that methane emission can be determined with much higher precision than ammonia emission.

Estimated daily emission was predicted with significant higher accuracy if the individual air sample were used, instead of average values for one or 24 hour measuring periods.

Estimated daily methane emission was not significantly influenced:

- By reducing the number of measure point for determination of outlet concentrations from 5 to 1.
- By the location of measure point for determination of outlet concentration, if locations close to inlet was avoided.
- By excluding observation where the difference between carbon dioxide concentration in outlet and inlet were below different thresholds in the interval between 6 and 400 ppm
- By reducing the measure period from six days to three hours.

Measuring procedures can be simplified by using fixed values for assumed inlet concentration. If suitable values are used the influence on estimated gas emission is limited, and this way it becomes possible to conduct measurement with a single - one channel - measuring system. Fix values for inlet concentrations can be based on outdoor or indoor measurement. If indoor measurement is used for determination of inlet conditions, the quality of the estimation is highly dependent on the correlation between the concentration of carbon dioxide and the concerned gas.

The relatively moderate measuring requirements for methane emission estimations makes it feasible to investigate the influence of feed composition in a relative large number of operating cattle buildings and consequently it can support a development towards reduced greenhouse gas emission from cattle production.

For ammonia the analyses did not reveal any shortcuts to precise and simple determination of emission from naturally ventilated buildings.

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