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### DEVELOPMENT AND VALIDATION OF A SIMPLIFIED METHOD TO QUANTIFY GASEOUS EMISSIONS FROM CATTLE BUILDINGS

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**ABSTRACT** Obtaining representative gaseous emission factors from livestock production requires measurement methods adapted to a high number and a great diversity of livestock systems, including naturally ventilated buildings. The objective of this study is to propose such a method for cattle houses, based on livestock-related data (e.g., feeds, production, effluents) and intermittent measurements of gas concentrations (H<sub>2</sub>O, CO<sub>2</sub>, NH<sub>3</sub>, CH<sub>4</sub>, and N<sub>2</sub>O), temperature, and relative humidity. These data were used to estimate gas-concentration gradients and mass-balance deficits of C, N, and H<sub>2</sub>O). Emissions were calculated using the ratios of gas-concentration gradients. For quality control, the results were validated with water-budget observations and simulation model predictions of CH<sub>4</sub> and CO<sub>2</sub> emissions from cattle. During summer 2007, measurements were performed in seven types of barns commonly found in France. For buildings with slurry evacuated twice a day, good agreement was found between CO<sub>2</sub> and CH<sub>4</sub> emissions estimated with the simplified method and those predicted from models of cattle emission. For these buildings the observed emissions were homogeneous. For buildings with deep litter, observed emissions of CH<sub>4</sub> and CO<sub>2</sub> were higher than the predicted emissions. The difference indicates that a part of those gases was emitted by manure. Additional data and models should be used to improve this method for deep-litter systems. Data analysis continues to evaluate the method during winter conditions and in poultry or swine farms.

**Keywords:** emission, greenhouse gas, ammonia, dairy cattle, livestock building, method.

**INTRODUCTION** The 1997 Kyoto protocol commits its 143 ratifying countries, including France, to reduce their greenhouse-gas emissions over the 2008-2012 period by 5.2% compared to 1990 levels. France also signed the 1999 Gothenburg Protocol, which commits it to reduce its emissions of sulphur, NO<sub>x</sub>, volatile organic compounds, and ammonia.

To satisfy reduction objectives, however, it is essential to estimate the contribution of each economic activity, particularly agriculture, which is a major contributor of greenhouse gas and ammonia emissions. Agricultural activity is the origin of 98, 83, and 79% of ammonia (NH<sub>3</sub>), nitrous oxide (N<sub>2</sub>O), and methane (CH<sub>4</sub>) emissions, respectively, in France, the latter two accounting for 22% of national greenhouse-gas emissions (CITEPA, 2009a, 2009b). Cattle are the main contributors of ammonia (64%) and methane (48%) emissions (CITEPA, 2009b). These estimates, however, are based on global emission factors that do not consider specific characteristics of French livestock systems. Because of France's international commitments, methods to quantify gaseous emissions from a variety of livestock systems, particularly from their buildings and effluent-storage facilities are urgently needed. The size and cost of current methods for quantifying gaseous emissions from livestock buildings have limited their application.

The aim of this study was to develop and validate a simplified method to quantify gaseous emissions from cattle buildings. Ultimately, this new method can be used to estimate the efficiency of mitigation strategies.

**MATERIALS AND METHODS** We adapted the concentration-ratio method (CRM) of Paillat et al. (2005), developed to quantify gaseous emissions from compost heaps, to quantify gaseous emissions from cattle buildings. This adaptation required measurements under real conditions to evaluate the robustness of the method in a variety of buildings. For validation, measured emissions were compared to emissions predicted with the simulation model of Maxin et al. (2006).

**Concentration-ratio method** The CRM is based on the carbon mass-balance and measured gas-concentration ratios. When applying this method to cattle buildings, one can ignore the airflow rate, which is difficult to estimate in naturally ventilated buildings with large openings (e.g., most cattle buildings).

To apply this method to cattle buildings, the carbon mass balance was calculated as follows:

$$\begin{aligned} \text{C emissions} &= Q_{C, \text{feed}} + Q_{C, \text{litter}} - Q_{C, \text{milk}} - Q_{C, \text{gestation}} - Q_{C, \text{effluent}} \\ \text{C emissions} &= E_{\text{CO}_2, \text{C}} + E_{\text{CH}_4, \text{C}} \end{aligned}$$

where  $Q_{C, i}$  is the quantity of carbon in  $i$  ( $i = \text{feed, litter, milk, gestation, effluent}$ ) and  $E_{\text{CO}_2, \text{C}}$  and  $E_{\text{CH}_4, \text{C}}$  are C emissions in CO<sub>2</sub> and CH<sub>4</sub>, respectively.

Based on these equations, we expressed the emissions for each gas produced in the building:

$$E_{\text{CO}_2, \text{C}} = [Q_{C, \text{feed}} + Q_{C, \text{litter}} - Q_{C, \text{milk}} - Q_{C, \text{gestation}} - Q_{C, \text{effluent}}] / [1 + (\text{Gradient}_{\text{CH}_4, \text{C}} / \text{Gradient}_{\text{CO}_2, \text{C}})_{\text{mean}}]$$

$$E_{\text{CH}_4, \text{C}} = E_{\text{CO}_2, \text{C}} * (\text{Gradient}_{\text{CH}_4, \text{C}} / \text{Gradient}_{\text{CO}_2, \text{C}})_{\text{mean}}$$

$$E_{\text{NH}_3, \text{N}} = E_{\text{CO}_2, \text{C}} * (\text{Gradient}_{\text{NH}_3, \text{N}} / \text{Gradient}_{\text{CO}_2, \text{C}})_{\text{mean}}$$

$$E_{\text{N}_2\text{O}, \text{N}} = E_{\text{CO}_2, \text{C}} * (\text{Gradient}_{\text{N}_2\text{O}, \text{N}} / \text{Gradient}_{\text{CO}_2, \text{C}})_{\text{mean}}$$

where  $E_{\text{NH}_3, \text{N}}$  and  $E_{\text{N}_2\text{O}, \text{N}}$  are N emissions in NH<sub>3</sub> and N<sub>2</sub>O, respectively.

$\text{Gradient}_{\text{CH}_4, \text{C}}$  or  $\text{CO}_2, \text{C}$  or  $\text{NH}_3, \text{N}$  or  $\text{N}_2\text{O}, \text{N}$  are the differences between the indoor and outdoor gas concentrations. An average value was calculated for all gradient ratios using at least 10 gas concentrations measurements.

Concentration gradients for these equations were measured using the method presented below, and the carbon mass balance was estimated using the model of Maxin et al. (2006).

**Carbon mass balance estimate** The model of Maxin et al. (2006) predicts quantities of N, C, and minerals such as P and K emitted by cattle in urine, faeces, and gases (e.g., CH<sub>4</sub> and CO<sub>2</sub>), regardless of their age and physiological status. Farm surveys can be used to gather required input data, such as quantities of feed ingested, cow weight, quantity of milk produced, fat and protein content of milk, and cow gestation status.

Mineral composition of ingested feed was estimated with data from INRA (2007).

Estimating the mass balance of each element required data of the amount of effluent produced and its chemical composition when it was removed from buildings.

Effluents were sampled from one building of each of seven types, except for two of the types (deep litter houses and free stalls with concrete floors), for which effluents were sampled from three buildings. Analyses of the chemical composition of the dejections (i.e., N, P, K, C, organic matter, and dry matter) were performed using standard methods. No effluent sample was taken from free stalls with slatted floors due to the difficulty in obtaining a representative sample. For all samples, results were compared to those of Bodet et al. (2001) who give mean chemical compositions of manure for the different category of animal production. Quantities of effluent were estimated based on the effluent carbon content and the total excreted carbon estimated by the model of Maxin et al. (2006) and were weighted by the proportion of daily time spent indoors. These results were also compared with reference values (DEPSE, 2001). DEPSE (2001) is an official French document that gives reference tables with the quantities of effluent produced in function of the cattle breeding systems in order to estimate the needed storage volume.

**Building types** The most common cattle buildings in France were the following:

- Tie stall with straw bedding (TS)
- Free stalls with straw bedded floor (FSBF)
- Free stalls with concrete floor (FSLM)
- Free stalls with slatted floor (FSSF)
- Loose housing with concrete passageway (LHLM)
- Loose housing with bedded passageway (LHSM)
- Deep litter house (DLH)

These buildings can contain three effluent types: liquid manure (LM), compact solid manure (CSM), and very compact solid manure (VCSM) (Table 1). In some buildings, cattle presence was so low (i.e. during summer) that no straw was spread, leading to collection of only liquid manure. Measurements of gaseous emissions were performed in 21 buildings (three of each type) in five regions of France to evaluate the simplified method under a variety of building and climate conditions.

Dairy and suckler cows represented 42% of French cattle in 2006 (AGRESTE, 2007). Because development of the method required cow presence during measurements (for CH<sub>4</sub> emissions), we took gaseous measurements only from buildings holding dairy cows.

Table 1. Characteristics of the livestock buildings where measurements were performed

Building type	Livestock code	Area Region	Effluent <sup>2</sup>	Number of dairy cows	Other cattle <sup>1</sup>	Cow Indoor presence period
TS Tied stall with straw bedding	TS-1	Auvergne	CSM	30	1B	night
	TS-2	Auvergne	CSM	19		milking
	TS-3	Auvergne	CSM	29	1B	night
LHSM Loose housing with a bedded passageway	LHSM-1	Pays de la Loire	VCSM	28		milking
	LHSM-2	Bretagne	LM	32		milking
	LHSM-3	Bretagne	LM	51		milking
DLH Deep litter house	DLH-1	Picardie	VCSM	35	10H	24/24
	DLH-2	Picardie	VCSM	56	1H	24/24
	DLH-4	Nord Pas de Calais	VCSM	34		24/24
FSBF Free stalls with straw bedded floor	FSBF-1	Pays de la Loire	LM	69		milking
	FSBF-2	Pays de la Loire	LM	59		night
	FSBF-3	Pays de la Loire	LM	40		night
LHLM Loose housing with concrete passageway	LHLM-1	Pays de la Loire	VCSM+LM	70	35H	24/24
	LHLM-2	Pays de la Loire	LM	88		milking
	LHLM-3	Bretagne	LM	47	3H	milking
	LHLM-4	Basse-Normandie	LM	38	5H+2CT+1B	milking
FSCF	FSCF-1	Pays de la Loire	LM	30	35CT+35H	24/24
	FSCF-2	Bretagne	LM	100		night
	FSCF-3	Bretagne	LM	47	5CT	24/24
FSSF Free stalls with slatted floor	FSSF-1	Auvergne	LM	34	1B	milking
	FSSF-2	Auvergne	LM	31		milking

<sup>1</sup> B=Bulls; H = Heifers; CT = cull cow

<sup>2</sup>LM: liquid manure; CSM: compact solid manure; VCSM: very compact solid manure

**Emission measurements in buildings** Air samples were collected inside and outside each building with an inexpensive pump (ELITE®, with a flow of  $3.3 \times 10^{-6} \text{ m}^3/\text{s}$ ) connected to TEDLAR® (SKC®) storage bags. The volume of the bags was  $0.003 \text{ m}^3$  for indoor samples and  $0.008 \text{ m}^3$  for outdoor samples. After various tests to identify the best path in the building for sampling indoor air to obtain a representative sample, it was decided that the operator had to traverse the building along its length. The outdoor sample was collected from air on all sides of the building. Samples were analyzed using an infrared photoacoustic analyzer (INNOVA® 1312) to measure concentrations of CO<sub>2</sub>, CH<sub>4</sub>, NH<sub>3</sub>, N<sub>2</sub>O, and H<sub>2</sub>O. Indoor and outdoor temperature and moisture were monitored while collecting air samples using a TESTO® 400 thermohygrometer.

## RESULTS AND DISCUSSIONS

**Effluent composition** For the livestock with VCSM, large differences between dry matter (DM) and organic matter (OM) results and the data of Bodet et al. (2001) were observed (Figure 1). These differences can be explained by the strong variability in the quantity of straw bedding and indoor presence time of dairy cows. The largest differences

were observed for buildings LHSM-1, DLH-1, and DLH-4, which had the most straw bedding (11, 18, and 20 kg straw/cow/day, respectively).

In contrast, N, K<sub>2</sub>O, and P<sub>2</sub>O<sub>5</sub> contents of the VCSM agreed with Bodet et al. (2001), except for the N and P<sub>2</sub>O<sub>5</sub> content in LHSM-1, which may have been due to a sampling problem.

For systems with LM and CSM, chemical compositions of the samples agreed with the values observed by Bodet et al. (2001).

In conclusion, effluent sampling is not necessary when applying the CRM to systems with solid manure when straw supplies agree with standard practices. In contrast, since VCSM composition is strongly linked to straw supply, sampling and chemical analysis of effluents will have to be performed when straw supplies exceed 10 kg straw/dairy cow/day for loose housing systems and 14 kg straw/dairy cow/day for deep litter houses. Because sampling VCSM is difficult to perform, however, a sampling protocol needs to be developed.

**Effluent quantity estimates** Good agreement ( $R^2=0.73$ ) was found between quantities of effluents predicted with the model of Maxin et al. (2006) and reference values (DEPSE, 2001)(Figure 2). Quantities of effluents, however, predicted from total excreted carbon, are slightly lower than the reference data, particularly for buildings with a deep litter. This undervaluation can be explained by the quantity of carbon lost in the form of CO<sub>2</sub> and CH<sub>4</sub>, which is not considered when calculating effluent quantities.

These results validate the data collected from farms about feed, indoor presence time, and milk production that are used as input data in the model of Maxin et al. (2006) and the use of this model in the simplified method to estimate the carbon mass balance.

**Gaseous emissions** Emissions were calculated only when indoor and outdoor concentrations were significantly different and the carbon loss predicted with the model of Maxin et al. (2006) was positive.

For buildings with LM, there was good agreement between gaseous emissions predicted with the model of Maxin et al. (2006) and measured emissions (Figure 3). Estimates of CH<sub>4</sub> and CO<sub>2</sub> emission were similar in buildings with LM because these gases were produced mainly by the animals in the studied systems.

For the 5 buildings with VCSM (DLH-1,2,4 ; LHSM-1 ; LHLM1), predicted emissions are lower than measured emissions, probably because the model predicts mass balances at the animal scale, thus ignoring gaseous emissions from the manure.

Comparison of measured emissions between the different building types shows that CO<sub>2</sub> and CH<sub>4</sub> emissions are higher for systems with VCSM than for those with LM (Figure 4). But these observed differences are strong (four times higher for VCSM systems) and cannot be explained only by litter fermentation. Thus, further measurements in such systems will be necessary. For the systems with CSM, (TS-1,2,3), CO<sub>2</sub> and CH<sub>4</sub> emissions were lower than those with VCSM and equivalent to those with LM; this can be explained by the daily evacuation of CSM.

In thirteen buildings, the N<sub>2</sub>O emissions could not be calculated because no concentration gradient existed.

N<sub>2</sub>O emissions measured in buildings with VCSM had large variability. N<sub>2</sub>O is produced during nitrification and denitrification processes that can occur only under specific conditions in the litter (presence of anaerobic and aerobic areas in the VCSM). The highest N<sub>2</sub>O emission was observed for DLH-1, which also had the lowest NH<sub>3</sub> emission when compared to the other buildings with VCSM. The opposite trend was observed for DLH-4, which suggests that the VCSM in DLH-1 presented better conditions for the immobilization of nitrogen by microbial biomass than that in DLH-4, where NH<sub>3</sub> emissions were higher because of high moisture content in the litter. For systems with VCSM the results agree well with knowledge about biochemical processes occurring in this type of litter.

In contrast, N<sub>2</sub>O emission was higher and NH<sub>3</sub> emission was lower in buildings with slatted floors than those from other buildings. According to the literature, NH<sub>3</sub> is mainly emitted by liquid manure (Loyon et al., 2007; Hartung et al., 1997) and N<sub>2</sub>O by the solid manure. Further measurements are necessary for these buildings.

Nitrogen was lost mainly in the form of NH<sub>3</sub> in systems with LM, such as FSLM and FSBF with no straw during summer. For buildings without deep litter during summer and concrete or bedded passageways (LHSM-2 and 3, LHLM-2 and 3), the surface that received the effluents was limited because of an electric wire prohibiting access to deep litter zones. Because the heat-exchange surface of effluents with the air was decreased, NH<sub>3</sub> emissions decreased (Dollé, 1998).

Results obtained with the CRM among the 3 buildings of each type had too much variation to find correlations between gaseous emissions and building types. We noted, however, higher CO<sub>2</sub> and CH<sub>4</sub> emissions from buildings with VCSM and higher NH<sub>3</sub> emission from buildings where LM was excreted directly onto concrete (FSBF and FSLM).

**CONCLUSION** The measurements carried out in 21 buildings helped develop a simplified method to quantify gaseous emissions. A wide range of building types was sampled to test application of the method in different systems. This method relies on intermittent measurements of gas concentrations, temperature, and moisture and technical livestock data (e.g., feed intake, milk yield). Results showed that sampling effluents to assess their chemical composition and improve emission quantification only has to be performed in buildings with VCSM. Measured emissions agreed with current knowledge and with predictions of the model of Maxin et al. (2006) for CO<sub>2</sub> and CH<sub>4</sub> emissions. A measurement protocol has been constructed and gives recommendations concerning materials, the procedure for sampling air, farm data necessary and ideal conditions in which to perform measurements (e.g., no wind, presence of animals during measurement). To validate the simplified method, however, measurements also will have to be performed during winter as well.

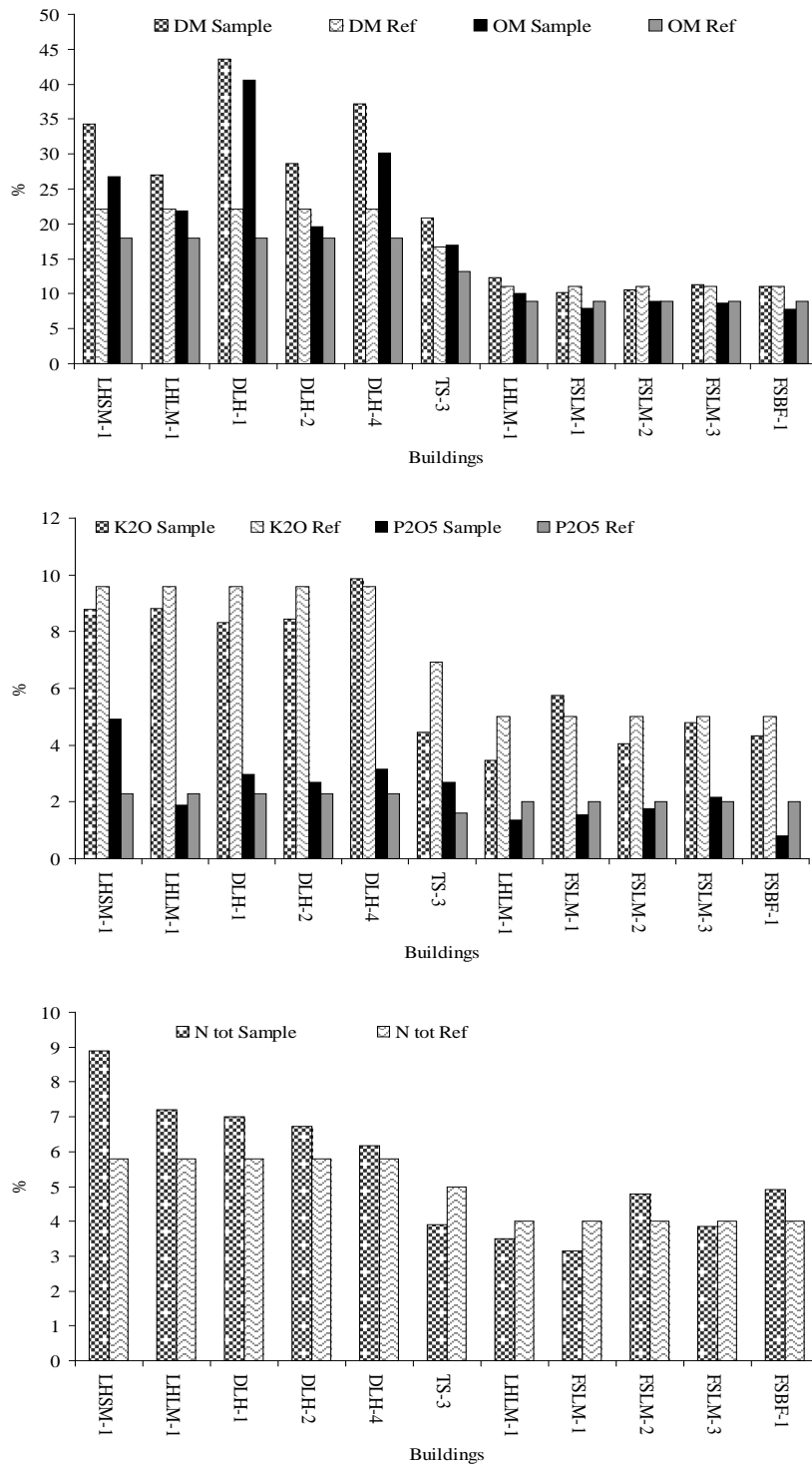


Figure 1. Comparison of the chemical composition (DM, OM, K<sub>2</sub>O, P<sub>2</sub>O<sub>5</sub>, N) of effluent samples collected in buildings in this study ("Sample") and the results observed by Bodet et al. (2001).

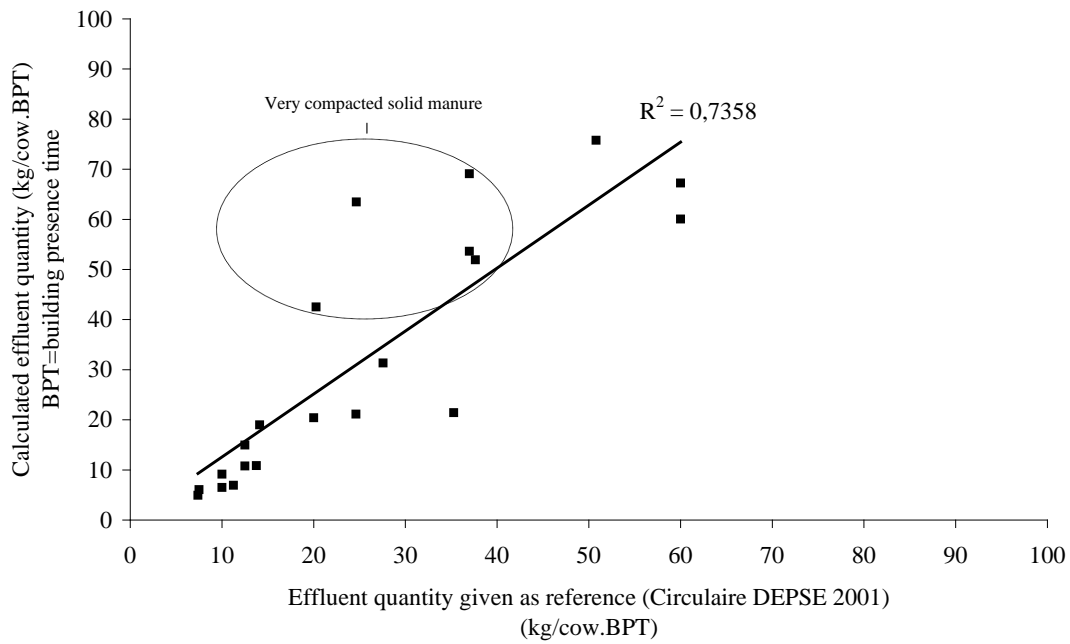


Figure 2. Correlation between the quantities of effluents predicted with the model of Maxin et al. (2006) and those provided DEPSE (2001)

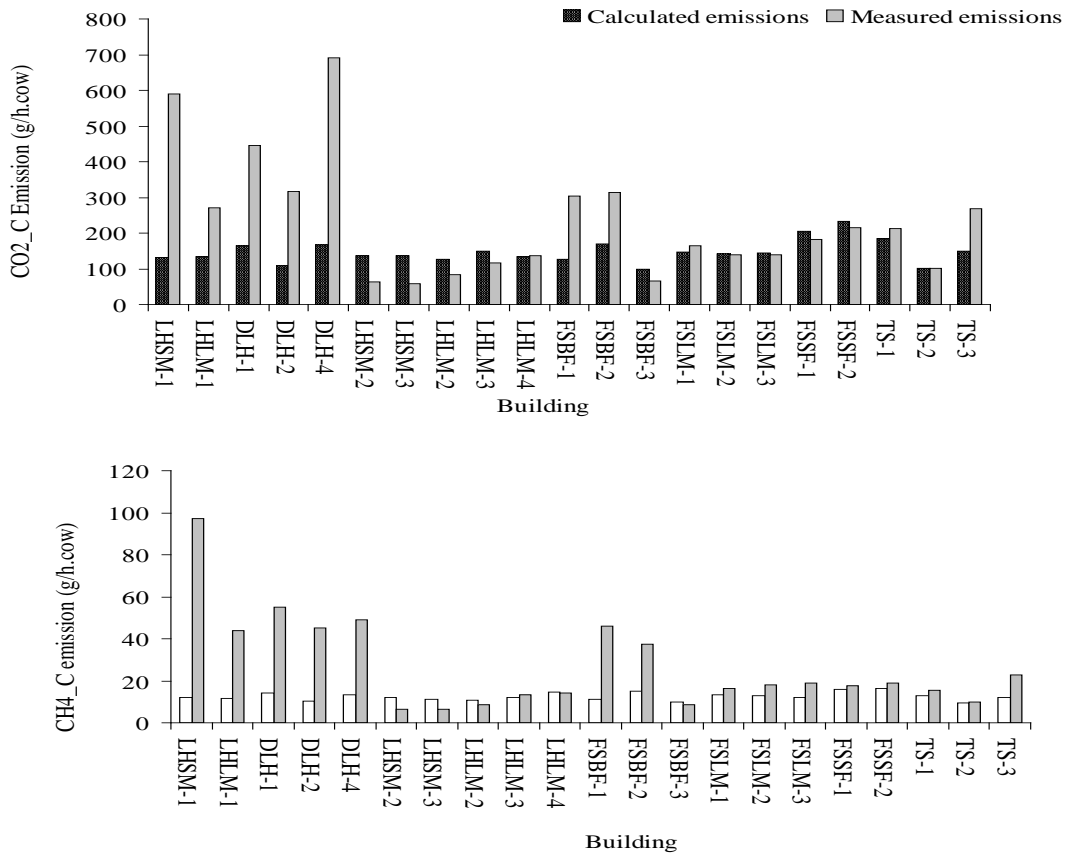


Figure 3. Comparison of predicted and measured CH<sub>4</sub>\_C and CO<sub>2</sub>\_C emissions (g/cow/h).



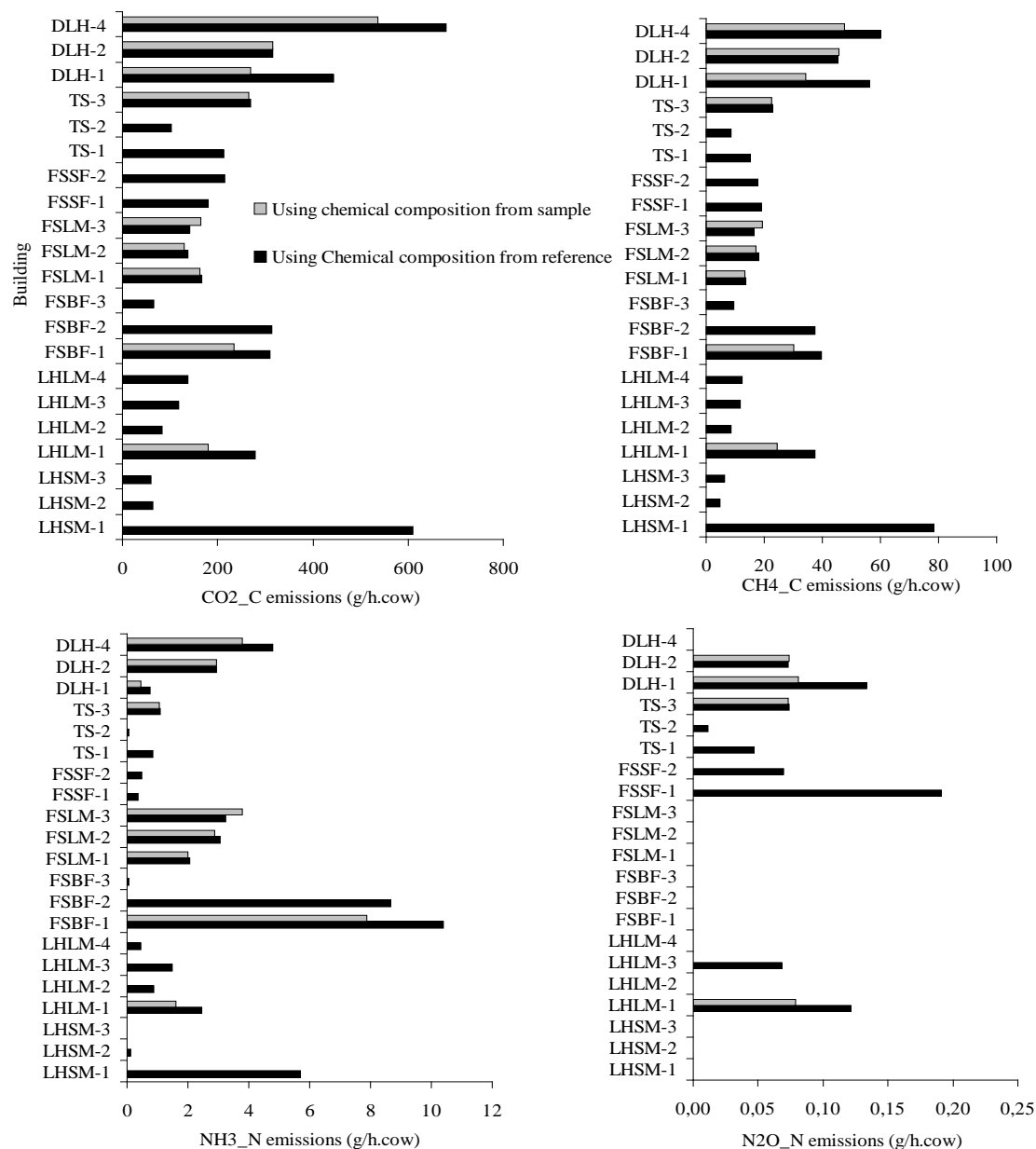


Figure 4. Hourly emissions estimated with the simplified method using effluent chemical compositions from the sample and from the reference. (g/h.cow)

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