Quantification of methane emissions from cattle farms, using the tracer gas dispersion method

Citation for the original published paper (version of record):
Dos Reis Vechi, N., Mellqvist, J., Scheutz, C. (2022). Quantification of methane emissions from cattle farms, using the tracer gas dispersion method. Agriculture, Ecosystems and Environment, 330.
http://dx.doi.org/10.1016/j.agee.2022.107885

Agriculture, Ecosystems and Environment 330 (2022) 107885

Contents lists available at ScienceDirect

Agriculture, Ecosystems and Environment

journal homepage: www.elsevier.com/locate/agee

Quantification of methane emissions from cattle farms, using the tracer gas dispersion method

Nathalia T. Vechi [a,b,*], Johan Mellqvist [b], Charlotte Scheutz [a]

[a] *Department of Environmental Engineering, Technical University of Denmark, Kongens Lyngby, Denmark*
[b] *Department of Space, Earth and Environment, Chalmers University, Göteborg, Sweden*

ARTICLE INFO

Keywords:
Dairy cows
Beef cattle
GHG inventory models
Enteric fermentation
Manure management

ABSTRACT

In Denmark, agriculture is the largest source of anthropogenic methane emissions (81%), mainly from cattle (dairy and beef) farms. Whole-farm methane emissions were quantified at nine Danish cattle farms, using the tracer gas dispersion method. Five to six measurement campaigns were carried out at each farm, covering a full year. Of the nine cattle farms, seven were home to dairy cows and two to beef cattle. The farms represented typical breeds, housing and management systems used in Denmark. Whole-farm methane emission rates ranged from 0.7 to 28 kg h^{-1}, with the highest measurements seen at locations with the highest number of animals. Emissions tended to be higher from August to October, due to elevated temperatures and high amounts of stored manure during this period of the year. The average emission factor (EF) for dairy cow farms was 26 ± 8.5 g Livestock Unit $(LU)^{-1}$ h^{-1}, whereas it was 16 ± 4.1 LU^{-1} h^{-1} for beef cattle farms, i.e. 38% lower for the latter. The use of deep litter house management explained some of the differences found in the EFs for dairy cows. Methane emission rates estimated using IPCC models and national guidelines tended, on average for all farms and measurements, to be underestimated by 35% in comparison with the measured methane emissions, for all models and farms. The results suggest that future improvements to inventory models should focus on enteric methane emissions from beef cattle and manure methane emissions for both dairy cows and beef cattle, especially from deep litter management.

1. Introduction

Many countries have strengthened their greenhouse gas (GHG) reduction targets and are putting efforts into reducing associated emissions with the goal of minimising climate change impacts. Globally, methane is the second most important emitted GHG and constitute 19% of the combined effective radiative forcing (relative to 1750) of the well-mixed GHGs (Forster et al., 2021). Atmospheric methane concentrations are increasing and reached 1.89 ppm in 2020, corresponding to an increase of 16% since 1985 (NOAA, 2021). Due to the relatively short atmospheric lifetime of methane (IPCC, 2013), reducing its emissions will lead to cost-effective mitigation of climate change impacts in the short term (Johansson et al., 2008). In Denmark, 81% of all anthropogenic methane emissions come from agriculture, produced by both enteric fermentation and manure management (Nielsen et al., 2021). Of the methane emitted from cattle livestock, about 76% is due to enteric emissions from the ruminant digestive system and about 24% comes from degradation of animal manure, stored either under the animals' house or at outside tank storages (Nielsen et al., 2021). Approximately 1.5 million cattle (dairy and non-dairy), accounting for about 20% of total agricultural exports (Danish Agriculture and Food Council, 2018), contribute 59% of all national methane emissions, which is a reflection of the large dairy industry in the country.

At the Danish national level, methane emissions from livestock production are estimated by using the empirical models provided in the International Panel on Climate Change (IPCC) 2006 guidelines for national inventory GHG emission reporting (IPCC, 2006). However, these models lack spatial resolution, and many of their parameters are based on limited or outdated research (Hristov et al., 2018). In order to improve them, the IPCC recently published a refined version of their guidelines, updating and supplementing some of the calculations and factors and using more actual research (IPCC, 2019). Where available, nations are encouraged to apply national models and emission factors, with the justification that they will better reflect their animal management policies.

Few studies have compared estimated methane emissions using these

* Corresponding author at: Department of Environmental Engineering, Technical University of Denmark, Denmark.
E-mail address: navei@env.dtu.dk (N.T. Vechi).

https://doi.org/10.1016/j.agee.2022.107885
Received 18 August 2021; Received in revised form 19 January 2022; Accepted 23 January 2022
Available online 10 February 2022

IPCC models with emissions based on whole-farm scale measurements representing real management conditions. This form of study is important for evaluating model accuracy and consequently improving emission predictions. Additionally, for models to achieve reliable predictions, a large dataset with a wide range of feed compositions and management systems across different regions remains to be built (Hristov et al., 2018).

Methods for quantifying enteric emissions from a single cow have mostly used respiratory chambers (Hammond et al., 2015), SF_6 tracer flux (Grainger et al., 2007) and, head-chamber systems (Sorg et al., 2018), albeit these are difficult to apply in real farm-scale measurements. Similarly, studies on methane emissions from farm-scale manure tanks are scarce, but they are still needed in order to obtain emission rates that reflect real conditions, preferably using non-intrusive methods (Kupper et al., 2020). Flux chambers (Amon et al., 2006), inversion dispersion models such as backward Lagrangian Stochastic (bLS) dispersion modelling (Baldé et al., 2016) and micrometeorological mass balance methods (Wagner-Riddle et al., 2006) are commonly used to measure emissions from manure tanks. Quantifying whole-farm CH_4 emissions is useful in understanding mitigation efforts. In addition, inventory models have been reported to underestimate livestock emissions (Owen and Silver, 2015; Wolf et al., 2017), therefore farm measurements representing real management conditions can help to improve model accuracy for the whole-farm system; however, they are few in number and mostly focused in North America (Arndt et al., 2018; Hensen et al., 2006; McGinn and Beauchemin, 2012; VanderZaag et al., 2014). The mobile tracer gas dispersion method (sometimes also referred to as the tracer correlation method or tracer flux ratio method) is a ground-based remote method that has been used to quantify emissions from many different area sources, such as landfills (Börjesson et al., 2009; Scheutz et al., 2011; Mønster et al., 2015), wastewater treatment plants (Delre et al., 2017; Samuelsson et al., 2018), biogas plants (Scheutz and Fredenslund, 2019) and livestock production facilities in North America (Arndt et al., 2018; Daube et al., 2019). This method has been described elsewhere (Galle et al., 2001; Roscioli et al., 2015; Scheutz and Kjeldsen, 2019) and can be used to quantify whole-farm emissions and, in some cases, emissions from physically well-separated farm operations such as manure tanks and cattle barns (Arndt et al., 2018). The tracer gas dispersion method uses a tracer gas to mimic the source methane release, which it is a principle also used in other techniques such as the SF_6 tracer flux method where it has been used to measure enteric methane emissions from individual cows (Grainger et al., 2007), and from manure tanks (Kaharabata and Schuepp, 1998) by performance of stationary plume sampling. The mobile tracer gas dispersion method applies an easier approach using a mobile analytical platform and measures multiple complete plume transects at a greater distance to the emission source securing sufficient mixing and source simulation thus reducing measurement uncertainty.

The objective of this study was to quantify whole-farm methane emissions from cattle farms, using a mobile tracer gas dispersion method, and to compare measured emissions with estimated methane emissions, using IPCC guidelines and national inventory models. We directly measured methane emissions consistently over one year for nine farms, which were selected to represent the different housing systems, manure management systems and breeds currently used in Danish agriculture. This study targets the lack of studies assessing whole-farm direct CH_4 emissions from cattle livestock and the need for IPCC model validation.

2. Materials and methods

2.1. Investigated sites

Nine cattle farms (C1-C9) were chosen for this study (Table 1). Seven of them were dairy farms, while the other two focused on beef production. Their selection was based on wishing to represent typical breeds, housing systems and manure management methods used in Denmark, where three dairy cow breeds predominate: the Danish Holstein (70%), Danish Jersey (12%) and Red Danish (5%) (Danish agriculture and Food Council, 2020a). Among beef cattle, the Limousine is the most common breed, accounting for 18% of all beef animals (SEGES, 2021). Considering these statistics, we selected at least one farm for each breed. Regarding manure management, 28% of the manure produced by cattle (dairy and non-dairy) is treated by anaerobic digestion, while 8% of the farms use deep litter and 60% have loose-holding housing systems for dairy cows (Nielsen et al., 2021). Therefore, three farms with loose-holding and different floor types (drained or slatted floor) (C1, C2 and C4), two with deep litter with a long eating space (C3 and C6), three with a mix of both systems (C5, C7, and C9) and one with deep litter only (C8) were selected. For the farms applying deep litter with a long eating space (C3 and C6), 60% of the excreted manure is deposited in straw beds, forming the deep litter, while 40% is collected as liquid/slurry. The anaerobic digestion of manure at the studied farms is performed in centralised biogas plants, i.e. raw manure is collected from pits under

Table 1
Overview of the investigated farms. For more detailed information, refer to Table A1 in Appendix A.

Farm Name	Type of animal	Numbers of animals			Livestock unit (LU)[a]	Animal breed	House	Manure handling
		Dairy	**Heifer/young bulls**	**Calves**				
C1	Dairy cow - Organic	600 – 745	289–765	0–643	970–1250	Jersey	Loose-holding drained floor	Liquid/slurry manure
C2	Dairy cow	250	110	100	330	Jersey	Loose-holding slatted floor	Biogas
C3	Dairy cow	420	180	150	540	Jersey	Deep litter with long eating space	Biogas
C4	Dairy cow - Organic	240	180	70	230	Holstein	Loose-holding slatted floor	Biogas
C5	Dairy cow	526	405	212	1055	Holstein	Loose-holding slatted floor (65%)/deep litter with long eating space (35%)	Biogas
C6	Dairy cow	160	110	40	305	Holstein	Deep litter with long eating space	Biogas
C7	Dairy cow	190	103	44	320	Red Danish	Loose-holding slatted floor (50%)/deep litter with long eating space (50%)	Liquid/slurry manure (3/20 – 9/20) Biogas (9/20–1/21)
C8	Beef cattle	30	40	20[b]	130	Limousine	Deep litter	Solid piles
C9	Beef cattle		560	143	545	Holstein	Loose-holding slatted floor (80%)/deep litter with long eating space (20%)	Biogas

[a] LU = 500 kg of body weight. Jersey dairy = 0.89 LU; Holstein and Red Danish dairy = 1.2 LU; Jersey heifer 0.65 LU; Holstein and Red Danish heifer 0.78 LU; Jersey bull = 0.68 LU; Holstein bull = 0.85 LU, Jersey calves = 0.16; Holstein and Red Danish calves = 0.21 LU; Limousine cows = 1.6 LU; Limousine heifer or bulls = 0.8–1 LU; Limousine calves 0.3 = LU.

[b] On average, farm C8 had 26 heifers and 13 bulls.

the barn one or more times a week and then taken to a biogas plant, which in turn returns degassed biomass to the farm's manure tanks. The amount of degassed manure received is around 90% of the initial volume; however, the composition largely differs, since biogas plants employ a mix of manure, deep litter, organic industrial waste and energy crops. Six farms send their manure for anaerobic digestion, while C7 only started sending it after September. For the other two farms, the manure was not treated and was instead either stored in liquid/slurry form (C1) or as a solid in deep litter piles (C8). Organic farms account for 14% of Danish dairies (Danish agriculture and Food Council, 2020b); hence, two organically managed farms were selected (C1, and C4). Among other things, milk cows spend part of their day grazing during the summer (~7 h over 196 days) and are fed with a high percentage of organic fodder. Enteric methane emissions vary according to the animal feed intake and the energy used, therefore different breed with differences in weight will produce distinctive emissions. As for manure emissions, the amount of manure stored in the house, the use of straw in the house or any other treatment the manure is subjected to can affect the strength of emissions.

The farms had a constant number of animals during the year. One exception was C1, which has seasonal calving in late spring, as all cows are inseminated at the same time, thereby affecting milk production and especially the amount of heifers and calves during the year. Methane emission factors (EFs) were calculated by normalising measured emission rates to the body weight base unit, whereby one livestock unit (LU) corresponded to 500 kg of body weight (Table 1). The animal weights used and other information on the farm's management were obtained by interviewing the farmers (Table A1 in Appendix A). Animal numbers used to model annual farm emissions were obtained from the animal central database (Centrale Husdyrbrugsregister (CHR)), to ensure consistency among the farms and to account for monthly variations. At the dairy farms, in addition to dairy cows, the numbers of heifers and calves were also included in the assessment of whole-farm emissions. These animals are sometimes managed at individual housing systems apart from the dairy cows' housing, which was taken into consideration when calculating inventory emissions. The only exception was farm C4, where measurements included only dairy cows, while heifers and calves were located at a building further away from the main farm, in which case they were not included in neither the measurements nor the modelling.

2.2. The tracer dispersion method

Methane emissions were quantified using the mobile tracer gas dispersion method, which has been employed previously to quantify methane emissions from dairy livestock in the USA (Arndt et al., 2018; Daube et al., 2019). The method involved the controlled release of tracer gas and simultaneous measurements of methane and tracer gas concentrations downwind of the farms, using high-precision instruments installed on a mobile platform (Galle et al., 2001; Mønster et al., 2014; Scheutz et al., 2011). Although the method has limited temporal resolution, it has a short setup time and can cover more than one facility in a single day; additionally, it is independent of weather conditions and atmospheric modelling. The method is based on the assumption that the source and tracer gas disperse similarly (Mønster et al., 2014), and therefore the tracer gas can be used to simulate target gas (methane) emissions, which can then be calculated by considering the ratio between the target and the tracer gas, and the known constant emission rate of the tracer gas (Eq. 1)

$$E_{tg} = Q_{tr}\frac{\int_{x1}^{x2}\left(C_{tg}\text{- } C_{tg\text{-}bg}\right)dx}{\int_{x1}^{x2}\left(C_{tr}\text{- } C_{tr\text{-}bg}\right)dx}\cdot\frac{MW_{tg}}{MW_{tr}} \tag{1}$$

where E_{tg} (kg h^{-1}) is the target gas emission, Q_{tr} (kg h^{-1}) is the known tracer gas flux, C_{tg} and $C_{tg\text{-}bg}$ (ppb as mass mixing ratio) are target gas concentrations measured inside the plume and in the background, respectively, similar to C_{tr} (ppb) and $C_{tr\text{-}bg}$ (ppb) for the tracer gas. MW_{tg} (g/mol) and MW_{tr} (g mol^{-1}) are the molecular weights for the target and the tracer gas, respectively. X2 (m) and X1 (m) represent the end and the beginning of the plume, respectively. The ratio is estimated by integrating the plume concentration of each gas, because this has been demonstrated as the best approach for minimising minor tracer gas misplacement (Fredenslund et al., 2019; Mønster et al., 2014). A number of transects provided average emissions, and in order to be considered a measurement, a minimum of 10 transects should be performed (Fredenslund et al., 2019). Acetylene (C_2H_2) was selected as the tracer gas, due to its negligible atmospheric background concentrations and long atmospheric lifetime. Recent studies, using the tracer gas dispersion method by performing controlled releases, have shown that method uncertainty is no higher than ± 20% in a 95% confidence interval (Fredenslund et al., 2019; Mønster et al., 2014).

The present study used a mobile analytical platform equipped with fast-response and sensitive gas analysers and a global positioning system (GPS) connected to the van. Air was sampled from the car's roof (two meters above ground) with the help of an external pump, and measured concentrations were shown in real time. Three different gas analysers (based on cavity ring-down spectroscopy) were used, based on instrument availability. The instrument used most during the measurement campaigns was a methane and acetylene analyser (G2203, Picarro Inc., CA), with a measurement frequency of two seconds and a precision (3σ) of 2.14 and 0.34 ppb for methane and acetylene, respectively. In addition, two other instruments were used in combination during the campaigns carried out from January 2020 to June 2020. One instrument measured acetylene (S/N JADS 2001, Picarro Inc., CA), with a response on average every three seconds and a precision (3σ) of 2.5 ppb, while the other instrument measured methane (G1301, Picarro Inc., CA), with a response time of four seconds and a precision (3σ) of 3.4 ppb. Acetylene (≥ 99.5%) was released from gas cylinders (at one to two locations usually close to the animal house and manure tanks), using constant flow rates set with calibrated high-precision flowmeters. In addition, the cylinders were weighed before and after each measurement campaign, in order to determine the precise mass of the released tracer. The tracer release rates varied from 0.6 to 2 kg h^{-1}.

Prior to the measurements, a desktop study was performed in order to evaluate the best weather conditions for optimal measurement performance, especially with regards to interfering methane sources and road availability. During the field campaign, the measurements can be described in three phases – as defined earlier (Scheutz and Kjeldsen, 2019). First, an on-site screening of the farm was performed to identify the main methane emission sources, which were mainly the animals' barns and manure tanks. Second, the farm's surroundings were screened by driving along available roads, in order to identify any interfering sources located in the area. Lastly, tracer gas was released and plume measurements performed. If the tracer and target gas plumes did not correlate well, meaning that they did not start and finish at the same time, the tracer gas bottles were repositioned. Crossing the whole plume downwind is important, to define baselines for the integrated plume calculations and assuring that the whole emission from the target source is measured.

2.3. Measurement campaigns

Table 2 provides an overview of the performed measurement campaigns. In total, 60 quantitative emission measurement campaigns were taken, all fulfilling the requirement of at least 10 plume transects carried out over 1–2 h. Most quantifications were performed on roads more than 1 km away from the target source, to ensure sufficient plume mixing.

Whole-farm methane emission rates were measured every second month, covering all seasons over one year. However, C1 was measured over a period of two years. Since the measurements were distributed equally around the year, the simple average of all measurements was considered as the annual average emissions. One exemption was beef farm C8, on which the animals grazed during the summer, and so

Table 2
Summary of the quantitative emission measurements.

Farm	Date	Time interval	Wind speed (m s^{-1}) and direction[a]	T (°C)	Road distance (km)	Number of transects	Methane emission ± SD (kg h^{-1})	Emission factor ± SD (g LU^{-1} h^{-1})
C1	07–02–19	11:00–13:20	4.5, SSW	3	1.1 (10) 2.1 (14)	24	15.3 ± 4.3	15.9 ± 4.5
	11–04–19	17:57–19:10	4, ENE	7	1.5	11	18.8 ± 1.4	20.4 ± 1.5
	28–06–19	22:09–23:15	2, WSW	18	1.5	20	13.0 ± 1.6	13.5 ± 1.7
	25–09–19	16:00–18:00	6, ENE	15	1.5	12	25.2 ± 4.1	25.9 ± 4.1
	14–11–19	17:00–18:40	1.5, ESE	4	2.4	11	28.5 ± 4.0	30.5 ± 4.3
	22–01–20	20:00–22:00	6, W	6	1.4	12	25.1 ± 4.2	26.8 ± 4.7
	04–03–20	16:00–18:00	2.5, WSW	7	1.4	20	24.1 ± 3.3	20.3 ± 2.9
	23–06–20	18:00–19:00	2, WSW	20	1.4	13	16.6 ± 4.2	13.5 ± 3.5
	13–07–20	17:30–19:00	2.5, W	18	1.4	20	15.9 ± 1.8	13.9 ± 1.6
	02–10–20	14:45–16:00	2.5, ESE	16	0.95	18	27.6 ± 4.2	24.9 ± 3.8
	05–11–20	14:45–15:45	5, W	11	1.4	17	22.1 ± 2.2	20.2 ± 2.0
	12–12–20	14:00–15:30	2, E	3	0.95	22	21.6 ± 1.7	20.0 ± 1.6
C2	05–03–20	19:00–21:00	1.5, ENE	3	0.7 (14) 1.0 (4)	18	9.2 ± 1.9	28.5 ± 5.9
	12–05–20	20:00–22:00	3.5, NW	5	0.4	17	5.9 ± 0.5	17.8 ± 1.5
	08–07–20	16:00–18:00	4, NW	14	0.4	25	7.7 ± 1.1	24.1 ± 3.4
	09–09–20	07:00–08:30	3, SW	17	1.0	20	7.2 ± 1	23.0 ± 3.2
	10–11–20	11:00–13:00	2.5, SE	7	1.1	22	6.4 ± 0.6	19.9 ± 1.9
	04–01–21	13:00 – 14:15	5, ESE	2	0.6 (22) 1.0 (3)	25	6.5 ± 1.0	20.2 ± 3.1
C3	06–03–20	15:30–17:30	4, NE	5	1.4	14	23.7 ± 4.1	40.6 ± 7.5
	08–05–20	17:30–19:00	3, W	16	1.2	17	22.3 ± 5.5	43.5 ± 11.3
	07–07–20	21:00–22:30	1.5, W	14	1.2	30	16.6 ± 1.1	32.3 ± 2.1
	08–09–20	10:00–12:00	3.5, W	18	1.2	27	18.7 ± 3.6	35.6 ± 6.8
	12–11–20	13:00–15:00	2.5, SSE	9	1 (9) 2.2 (14)	23	20.1 ± 2.8	38.0 ± 5.3
	05–01–21	09:00 – 11:00	1, NE	3	2	17	19.9 ± 2.4	37.4 ± 4.5
C4	17–03–20	13:30–15:30	5, SW	9	1.6	19	7.7 ± 1.3	26.4 ± 4.5
	14–05–20	16:00–18:00	1.5, NW	13	0.8	24	5.7 ± 1.0	19.5 ± 3.4
	07–07–20	15:00–16:30	4.5, WSW	17	1.6	18	5.6 ± 1.2	19.5 ± 4.2
	07–09–20	17:30–18:45	2, WSW	15	1.6	24	4.8 ± 0.6	16.9 ± 2.1
	10–11–20	14:30–18:00	1.5, SSE	6	0.9	21	7.8 ± 1.5	27.2 ± 5.2
	04–01–21	16:00 – 18:00	4.5, NE	1	1.7	21	6.6 ± 0.8	21.3 ± 2.6
C5	05–03–20	16:00–18:00	2.5, ENE	5	1.4	19	18.1 ± 2.9	18.2 ± 3.1
	12–05–20	12:00–16:00	3.5, W	7	1.8 (10) 0.4 (3)	13	17 ± 3.3	17.2 ± 3.2
	07–07–20	17:20–18:45	2.5, W	14	1.8	20	19.6 ± 1.8	20.1 ± 1.8
	07–09–20	16:00–15:30	4.5, SW	17	1.8	21	23.9 ± 3.6	24.1 ± 3.6
	11–11–20	17:00–18:30	1.5, ESE	7	1.2 (19) 0.6 (2)	21	21.3 ± 2.7	21.3 ± 2.7
	05–01–21	16:30 – 18:30	2, ENE	1	1.1	19	23.4 ± 1.6	23.3 ± 1.6
C6	29–03–19	20:15–22:30	4, SW	10	1.4	29	7.5 ± 1.2	26.0 ± 4.2
	20–08–19	21:00–23:00	2, SW	13	1.4	32	8.9 ± 1.0	30.0 ± 3.4
	13–02–20	10:00–13:00	2.5, S	3	1.5	11	7.8 ± 1.9	27.9 ± 6.8
	18–05–20	16:30–18:30	1.5, WSW	11	1.4	20	7.3 ± 1.2	24.7 ± 4.1
	27–06–20	21:30–23:30	1, S	20	1.5	16	8.0 ± 1.5	27.1 ± 5.1
	10–10–20	15:00–17:00	1.5, WSW	10	1.5	20	8.4 ± 1.1	29.0 ± 3.8
	18–12–20	10:45: 12:15	3.5, SSW	8	1.8	19	5.4 ± 0.8	18.3 ± 2.7
C7	16–03–20	11:00–13:00	3.5, W	8	1.0	19	13 ± 1.8	39.6 ± 5.5
	07–05–20	19:45–22:15	3.5, W	14	1.0	18	8.7 ± 0.9	27.0 ± 2.8
	06–07–20	20:30–22:00	4.5, W	13	1.0	30	12.1 ± 1.3	37.2 ± 4.0
	09–09–20	15:30–17:30	3.5, W	18	1.0	26	16.9 ± 1.7	54.4 ± 5.5
	12–11–20	14:00–15:30	2.5, ESE	7	1.8	20	9.3 ± 1.1	30.2 ± 4.8
	12–01–21	10:00 – 12:00	3, WSW	1	1.0	23	7.6 ± 0.8	24.2 ± 2.5
C8 - House	16–03–20	18:00–19:30	1.5, WSW	7	0.7	20	1.2 ± 0.2	11.6 ± 1.9
	07–05–20	21:30–23:45	1, WSW	8	0.7	13	1.9 ± 0.5	16.9 ± 4.5
	11–11–20	07:30–09:00	1, SE	7	0.7	17	1.1 ± 0.3	17.9 ± 4.9
C8 - Grazing	11–11–20	10:30–12:00	1, SE	7	0.5	19	0.4 ± 0.1	23.5 ± 5.9
C8 - House	06–01–21	08:00 – 10:00	4, NE	1	1.0	16	2.7 ± 0.6	22.3 ± 3.6
C9	17–03–20	17:30–19:30	4.5, WSW	7	0.8	16	6.2 ± 1.1	11.5 ± 2.0
	08–05–20	20:00–22:00	2.5, W	11	0.8	18	5.9 ± 0.8	10.6 ± 1.4
	08–07–20	20:00–21:30	1, WNW	14	1.5	29	7.4 ± 0.7	18.0 ± 1.7
	08–09–20	13:30–14:45	0.5, WNW	21	1.2	21	8.6 ± 1.3	18.4 ± 2.8
	12–11–20	17:00–18:15	1, S	8	0.6	24	6.1 ± 0.7	11.8 ± 1.4
	05–01–21	13:00 – 15:00	2.5, NE	2	1.5	21	6.6 ± 0.7	11.9 ± 1.3

[a] Wind speed measured by a vane anemometer at a height of 1.5 m.

measurements were only taken during the winter season (from November to May). However, in the November measurement campaign, about two-thirds of the animals were housed inside while the others grazed on a nearby field. Emissions from both groups of cows were therefore measured in this campaign. For the dairy farms and beef farm C9, measurements were only taken when animals were inside, although cows did graze outside for part of the day (~7 h) at the organic farms (C1, C4).

2.4. Methane emission estimation, following the IPCC and Danish inventories

Measured methane emission rates were compared to modelled emission rates, following the IPCC's inventory guidelines and the Danish national guideline. The IPCC guidelines are divided into Tier 1, 2, and 3, differing on the level of information used in the calculation (IPCC, 2006); for this study, Tier 2 was adopted. Additionally, the IPCC recently published a refined version of their models, which included improvements to the estimations of methane emissions from cattle production (IPCC, 2019); therefore, for comparison, both the 2006 IPCC model (I06) and its 2019 refined version (I19) were used here. The Danish GHG inventory uses a similar approach to the IPCC (2006), applying extra information and models, which should reflect better the management systems used at Danish farms (Nielsen et al., 2021). Both the IPCC guidelines (I06 and I19) and the Danish guideline (DK) calculate EFs for enteric fermentation and manure management, and each uses similar equations (Eq. A1 and A2 in Appendix A).

Farm-specific information obtained from the farmers was used in the modelling, such as type of housing system, manure treatment, frequency of manure removal (when applicable in IPCC, 2019), milk production and animal body weight (Table A1). The calculations resulted in estimated EFs per animal head (Table A5), which were then converted to annual emissions by multiplying the EFs by the number of animals in that month (the same month as the measurement was done) and then averaged over all months to get an average annual emission rate. The estimated annual emissions were compared to the measured annual emissions, which was the average of all measurements.

As values for animal feed intake were not available or unknown for some of the farms, enteric emissions were calculated for consistency by following the indicated approach for each of the models. Gross energy (GE) is the main parameter used to estimate enteric emissions. The IPCC calculates GE based on animal used net energy (NE) and digestible energy (DE) (IPCC, 2006) (Fig. 1), while the Danish guideline uses GE per feed intake for dairy cows, or feeding units (FU) for other cattle (Nielsen et al., 2021). Standardised GE values for the different livestock and breeds in the country (Table A2) (Børsting et al., 2020) are provided based on national feeding plans for 15–18% of Danish dairy production (Nielsen et al., 2021). For three of the farms (C1, C2 and C3), feeding plans were known; however, the difference between real feeding and standardised values were minimal and did not affect the models' results.

Manure emissions are estimated by the models using information on volatile solid contents (VS), the methane conversion factor (MCF) (Table A3) and maximum methane-producing capacity (B_0) (for more information, see Appendix A). The IPCC calculates the quantity of VS excreted based on energy intake, while the Danish guideline provides standard values for excreted manure based on information on typical animal characteristics (Børsting et al., 2020) and according to the types of housing systems most frequently used in the country (Table A4) (Fig. 1). The estimation of VS by the Danish inventory approach results in higher values than the VS based on GE, mainly due to the inclusion of bedding material in the first option (Nielsen et al., 2021). The IPCC 2019 refinement contains updated model parameters and improvements to the way the methane conversion factor (MCF) for liquid manure is estimated. The newest version applies a more detailed sub-model, using temperature-dependent degradation functions, and considers manure storage time. A similar sub-model is also applied in the Danish national

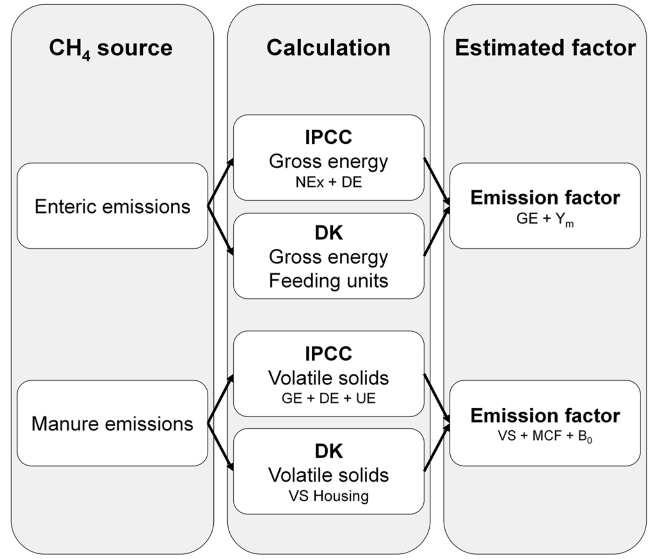

Fig. 1. The IPCC and Danish inventory models estimate enteric and manure methane emissions, using similar equations (Appendix A). NEx = Net energy for each type of activity (Maintenance, growth, activity, pregnancy and lactation). DE = Digestible energy; GE = Gross energy, UE = urinary energy fraction. VS = Volatile solids. MCF = Methane conversion factor. Ym = Methane yield. The most important parameters for enteric fermentation are GE and Y_m, while manure emissions are based on VS, MCF and B_0. Both methods differ in the calculation of.

guidelines, following the most common practices and data available for the country, resulting in recommended values for annual MCF for cattle manure handled as liquid slurry or treated biogas (Table A3) (Nielsen et al., 2021). For deep litter, the Danish model adopts the MCF provided in the IPCC 2006 model. Additionally, the Danish model considers different temperatures for the storage of manure in barns and for external manure tanks in order to calculate MCF factors (Nielsen et al., 2021). According to the IPCC guidelines and the Danish national inventory the uncertainty on the EF estimates using Tier 2 is on the order of 20%, which is a reflection of the level of information available.

3. Results and discussion

3.1. Methane emission rates

Whole-farm methane emissions were quantified at nine farms, and during all quantitative measurement campaigns, well-confined methane and tracer gas plumes were obtained, thus avoiding any influence of methane plumes from neighbouring sources. Fig. 2 shows a representative example of an on-site and off-site methane screening campaign as well as plume concentration transects. The average measurement time was ~ 90–120 min, and within this time interval no temporal emission variations were observed, not even for longer measurement periods (> 4 h). Whole-farm emissions rates ranged from 0.7 kg h^{-1} to 28 kg h^{-1} (Table 2). The highest emission rates were measured at the three dairy farms C1, C3 and C5, which were also home to the highest number of animals. The lowest emission rates were recorded at beef cattle farm C8, which had only approximately 100 animals (including animals at different life stages). During one of the campaigns at C8, methane emissions of 0.4 kg h^{-1} from 16 cattle grazing in the field were measured.

Methane emissions fluctuated throughout the year, but these oscillations were small, with emissions varying on average between − 16% and + 13% of the mean annual emission measured at the individual farm (Fig. 3). The measured emission rates contained both enteric and manure methane emissions. Enteric emissions were expected to be

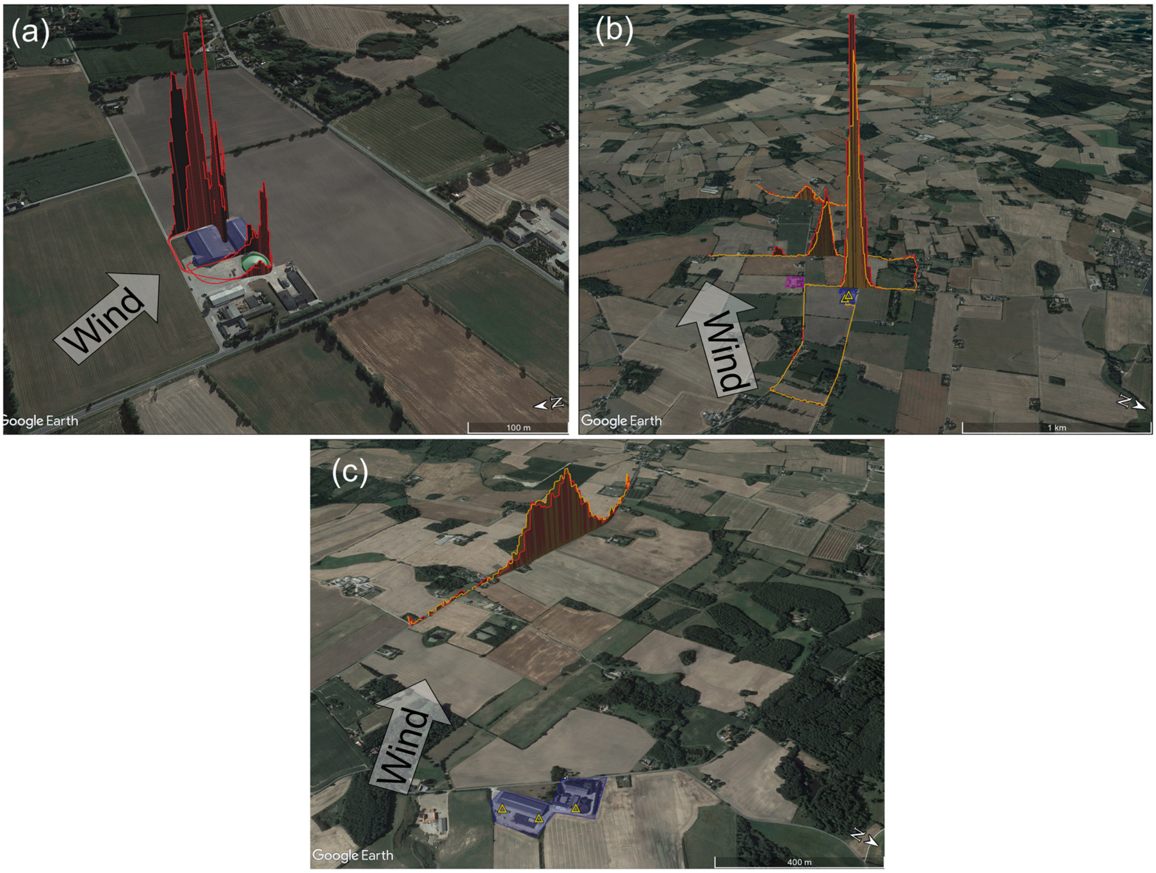

Fig. 2. (a) Example of on-site screening performed at farm C2, showing methane (red) concentrations (above background). The blue area indicates the location of the animal housings and the green area the location of the manure tank. (b) Example of off-site screening and plumes at different distances away from the farm (C2) (blue area) (160 m, 600 m and 1400 m). Two tracer gas bottles (yellow triangles) were positioned close to the animal barn, and the wind was blowing from the east-north-eastern direction. For the second plume (600 m), the methane (red) peak concentration was 108 ppb above background and 11 ppb for acetylene (yellow), while for the third plume the values were 30 ppb and 3.4 ppb, respectively. The small methane emission plume on the left of the target farm's plume came from a small horse farm, indicated by the purple area, which was clearly distinguishable from the farm's emissions, due to the lack of tracer gas. (c) Example of a plume transect at farm C9. Three gas bottles were positioned close to the animal barn and manure tanks, the wind blew in the north-easterly direction and the methane peak (red) was 17 ppb above background, with 4 ppb for acetylene (yellow). (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)

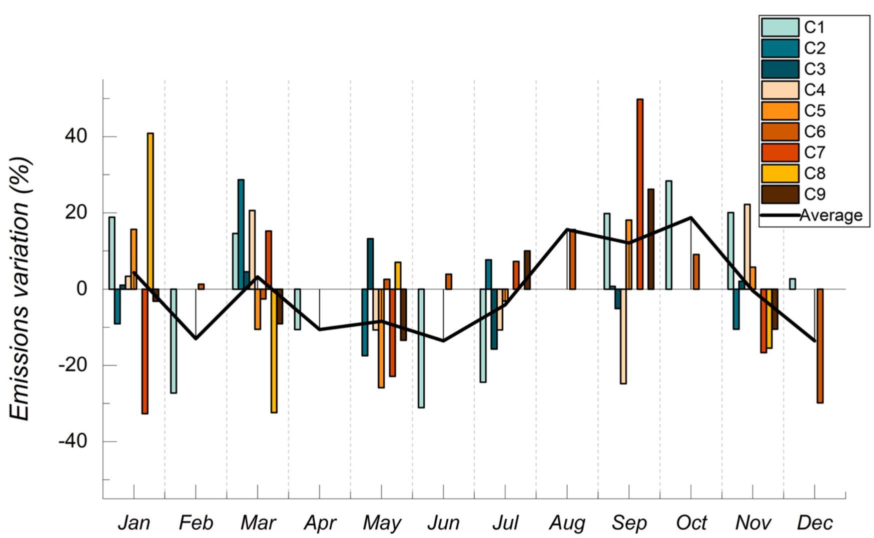

Fig. 3. Methane emission variations during the year. For each farm the variation is the measured emission in a specific month minus the mean annual emission of the farm and divided by the mean annual emission of the farm. The black line represents average variations for each month. Note that in some months, only one or two measurements were available.

constant in most of the farms, since animal numbers or feeding patterns did not vary much throughout the year. Therefore, monthly variations are most likely an effect of fluctuations in manure emissions, which in turn is expected due to changes in atmospheric temperature and the amount of manure stored during the year. In addition, most of the farms stored anaerobically digested (degassed) manure, so the expected contribution of this source was reduced, consequently causing small variations in the total farm emissions – as indicated by the results. The highest average emissions were seen in late summer/early autumn (August to October), which was expected, because at this time of year, there is a combination of higher temperatures and higher amounts of stored manure, thereby increasing emissions (Fig. 3) (Kariyapperuma et al., 2018). At some farms (C1, C2, C3, C4 and C7), slightly higher emissions were recorded in March, which was due to the high amount of manure stored at the time. The manure tanks are emptied in early spring (April/May), thus explaining the reduction in emissions at this point in time.

The maximum and minimum values observed were + 49% and − 32% of the mean annual emissions, respectively. They were found on farm C7, most likely because, during the first measured months (March to September), raw manure was being stored in on-site manure tanks before being sent for anaerobic digestion, following which the farm stored degassed manure. Farm C1 had a lower number of heifers in combination with its dairy cows producing less milk in spring due to pregnancy, thereby decreasing enteric emissions, which is in agreement with the observed data showing lower emissions in April and June in comparison to September and October (Fig. 3). C8 also had a few more suckling cattle in January than in March, possibly explaining the higher emissions in January. Some of the outliers can be explained; for example, C4 had lower emissions in September, in complete opposition the overall trend, because the manure tanks remained empty due to the manure constantly being applied to the fields from March to November.

Finally, other factors might explain some of the observed emission variation, such as time of the measurement in relation to feeding and general activity of the cattle, type of fodder, amount of manure accumulated under the housing, measurement uncertainty and others. Diurnal variation of methane emissions caused by periods of feeding has being observed in some studies (Ngwabie et al., 2011; VanderZaag et al., 2014) although, others have not observed such significant emission variation (Arndt et al., 2018; Bühler et al., 2021). At the farms investigated in this study, fodder was available for the animals during the whole day, therefore the animals would have alternate periods of feeding and resting, which would level out any emission variation to due feed intake. However, the data set did not allow for an in depth analysis of the influence of these factors on the measured emissions.

3.2. Methane emission factors

Converting the measured emission rates to EFs resulted in EFs ranging from 11 to 54 g LU^{-1} h^{-1}, with an average EF of 23 ± 9 g LU^{-1} h^{-1} and a median of 22 g LU^{-1} h^{-1} (Table 2 and Fig. 4). For dairy cows, normalising the measured emissions according to milk production resulted in EFs ranging from 21 to 67 g L_{milk}^{-1}, with an average EF of 39 g L_{milk}^{-1} or 35 g $head^{-1}$ h^{-1} when normalising milk per head of cow (Table A1).

For comparison, Table 3 compiles methane EFs from dairy cows, including only studies where whole-farm methane emissions were measured. The values ranged from 7.1 to 60.2 g LU^{-1} h^{-1}, which is comparable to the values found in this paper (11–54 g LU^{-1} h^{-1}); however, a direct comparison must be made with caution, due to differences in management systems, seasons and measurement techniques. First of all, there are relatively few studies, and most of them were performed in the USA or Canada, which might not reflect Danish conditions in terms of either climatic conditions or manure management practices (Arndt et al., 2018; Bjorneberg et al., 2009; Leytem et al., 2011). An important difference in manure management between DK and North America dairy farms (especially in USA) is the use of open anaerobic lagoons by the latter, while in DK external manure is stored in concrete tanks. Only three European studies on whole-farm methane emissions were found, and each reported very different EFs, i.e. relatively low 11.2 – 15.0 g LU^{-1} h^{-1} in Austria (Amon et al., 2001) and 11 – 14 g LU^{-1} h^{-1} in Switzerland (Bühler et al., 2021) and relatively high 28.7 – 50.5 g LU^{-1} h^{-1} in Netherlands (Hensen et al., 2006) in comparison to the EFs in our study. In addition, seasonal variations can play a role in emissions, and most of these studies did not systematically measure emissions across the whole year. Therefore, it is difficult to conclude whether the EFs found in our study were elevated or not when compared to the studies compiled in Table 3, due to the lack of comparability between them.

In the following sections different factors are discussed, which can explain some of the variations in EFs found across the farms investigated in this study (Fig. 4).

3.2.1. Housing type

Of the Jersey farms (C1, C2 and C3), farm C3 had an EF significantly higher than the other two Jersey farms ($p < 0.05$, Tables A6 and A7). Of the Holsteins dairy farms (C4, C5 and C6), C6 had a significantly higher EF than C5 and C4 ($p < 0.05$, Table A6), while the C4 and C5 farms were significantly similar ($p > 0.05$, Table A6). Farms C3 and C6 both used deep litter with two months of retention time as their main house management system (60% of the manure produced is deposited in deep litter and mixed with straw) (Table A1), which might explain the higher methane emissions (Fig. A1b in Appendix A). In the deep litter house management system, faeces, urine and straw are compressed into mats, thereby limiting oxygen diffusing into the material, and as a result anaerobic conditions and methane formation develop in the bottom and centre of the material, potentially leading to higher emissions than other housing systems (Nicks et al., 2003; Webb et al., 2012). Farms C5 and C7 also used deep litter, albeit to a lesser extent (between 13% and 30% of the manure is handled as deep litter). Additionally, C5 also applied a

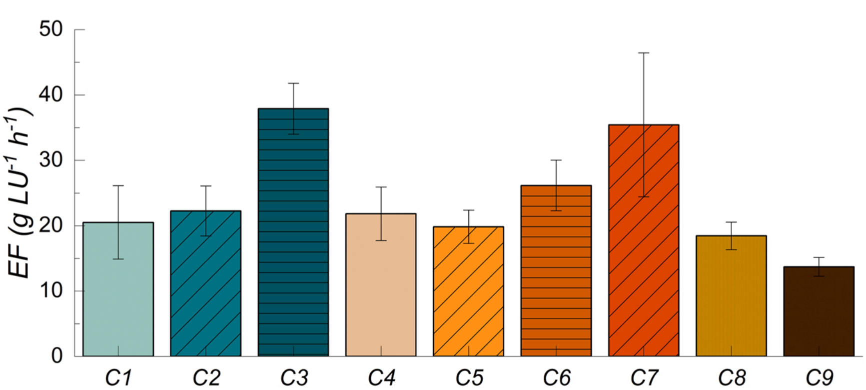

Fig. 4. Average yearly methane emission factors (EFs) (g LU^{-1} h^{-1}). Error bars represent the standard deviations of six or more measured EFs. C1- Organic dairy, Jersey, no manure treatment; C2 – Traditional dairy, Jersey, biogas; C3 – Traditional dairy, Jersey, deep litter and biogas; C4 – Organic dairy, Holstein, biogas, C5 – Traditional dairy, Holstein, deep litter (35%) and biogas; C6 – Traditional dairy, Holstein, deep litter and biogas; C7 – Traditional dairy, Red Danish cow, deep litter (50%) and biogas; C8 – Traditional Beef cattle, Limosine, deep litter and grazing; C9 – Traditional beef cattle, Holstein, deep litter (20%) and biogas.

Table 3
Overview of whole-farm methane emission factors (EFs) measured at dairy farms.

Ref	Country	Period	Farm management	Measurement technique	EFs (g LU^{-1} h^{-1})
Present study	Denmark	Yearly	Dairy and beef cows with manure tank	TDM	23.6
(Arndt et al., 2018)	USA-California	Summer Winter Summer Winter	Dairy cows 1 (Jersey) - free stalls and anaerobic lagoon Dairy cows 2 (Jersey) - free stalls and anaerobic Lagoon	Open-path spectrometer and inverse dispersion modelling, TDM and aircraft close-path	60.2 28.5 46.8 18.9
(Leytem et al., 2011)	USA- Idaho	Spring, summer, winter and fall	Dairy cows CAFO - anaerobic lagoons	Open-path spectrometer and inverse dispersion modelling	57.9
(Bjorneberg et al., 2009)	USA- Idaho	Spring, summer, winter and fall	Dairy cows - anaerobic lagoons	Open-path spectrometer and inverse dispersion modelling	9.7
(McGinn and Beauchemin, 2012)	Canada	Fall	Dairy cows 1 - open lagoon Dairy cows 2 - open lagoon Dairy cows 3 - open lagoon	Open-path spectrometer and inverse dispersion modelling	7.5 7.1 8.1
(VanderZaag et al., 2014)	Canada	Spring Fall Spring Fall	Dairy cows 1 - earthen storage Dairy cows 2 - earthen storage and concrete tank	Open-path spectrometer and inverse dispersion modelling	7.3 19.6 10.4 21.8
(Amon et al., 2001)	Austria	Yearly	Tied stall and solid manure (heap - aerobic conditions) Tied stall and solid manure (heap - anaerobic conditions)	FTIR - flux conversions using exhaust air flow or open dynamic chambers	11.2 15.0
(Hensen et al., 2006)	Netherlands	Spring and summer	Dairy cows - manure tanks - slurry based system Dairy cows - manure tanks - straw based systems	TDLAS - Gaussian plume method and fast box measurement Technique	28.7 50.5
(Bühler et al., 2021)	Switzerland	Fall (Sep-Oct) Fall (Nov-Dec)	Dairy cows - loose- holding, slurry pit	Open-path spectrometer and inverse dispersion modelling	14.2 11.2

comparatively short manure retention time (6 weeks), thus reducing emissions caused by deep litter accumulation (IPCC, 2019).

3.2.2. Animal breed

Emission factors for Jersey cow farms (C1 – C3) were 21 ± 6, 22 ± 4 and 38 ± 4 g LU^{-1} h^{-1} for farms C1, C2 and C3, respectively (Fig. 4). For the Holstein dairy cows (C4 – C6), the average EFs were 22 ± 4.1, 21 ± 2.7 and 26 ± 3.9 g LU^{-1} h^{-1}, for C4, C5 and C6, respectively, while for the Red Danish milk breed (RDM) farm C7 the averaged emission factor obtained was 35 ± 11 g LU^{-1} h^{-1}. Comparing the two groups of dairy farms (Jersey versus heavy species (Holstein and RDM)), EFs did not differ as a result of breed differences. The same was the case when emission rates were normalised by the number of cows instead of body weight (Table A1).

3.2.3. Production target

The largest difference in EFs was recorded between the dairy and beef farms (Fig. A1a). Emission factors for the C8 and C9 beef cattle farms were between 11 and 24 g LU^{-1} h^{-1} (Fig. 4) with an average EF of 16 ± 4.1 g LU^{-1} h^{-1}, which is approximately 38% lower than the average EF for dairy cows 26 ± 8.5 g LU^{-1} h^{-1} (Fig. 4). The higher EF for dairy cows is mainly caused by differences in enteric emissions, due to the higher feed intake (caused by milk production) – as described in the Danish guideline (Børsting et al., 2020). Dry matter intake is known to be correlated with enteric emissions (Hristov et al., 2018).

3.2.4. Other factors

Farms C1 and C2 had similar average EFs, even though farm emissions were significantly different according to the Wilcoxon test ($p < 0.05$, Table A6, Fig. 4). Farms C1 and C2 had rather different management systems; C1 is an organic farm where cows spend part of their time grazing, and they do not treat the manure but instead apply it to the fields more frequently. Farm C2 is a conventional farm and treats its manure at a centralised biogas plant. Stored digestate in the tanks is only removed in spring, therefore it is difficult to draw a conclusion from their comparison. C7 had the highest EF among the heavy race farms, two factors might have caused this elevated EF. First, 50% of the dairy cows were managed in deep litter with a long eating space and an extended retention time (4 months), and the second, there was a lack of liquid/slurry manure treatment (only implemented in September) (Fig. 4). This notion is also supported by the large variability in emissions seen on this farm, which was potentially caused by variations in manure emissions, since enteric emissions is expected to have been constant during the year as no changes in animal numbers occurred (Arndt et al., 2018; VanderZaag et al., 2014). Generally, for dairy cows, a significant difference between EFs was observed for farms using deep litter management and other treatments (Fig. A1b).

The two beef farms had similar average EFs in spite of different housing systems, with C8 having 18 ± 2.1 g LU^{-1} h^{-1} and C9 having 14 ± 1.4 g LU^{-1} h^{-1} ($p > 0.05$ for a t-test, but $p < 0.05$ for a Wilcoxon test, Tables A6 and A7). At farm C8, methane emissions were not measured during the summer months, because the cows grazed outside (24/7) from May to November. However, in November, emissions were measured for two groups of cows, namely those grazing in the field and those in a barn, resulting in an EF of 24 ± 6 g LU^{-1} h^{-1} for the first cohort, higher than second group in the barn at 18 ± 5 g LU^{-1} h^{-1}. These results are most likely due to the high feed intake in pasture conditions, during measurements or issues in the adopted normalization (calculation of livestock units), since the animals here were a mix of calves, heifers and suckling cows. Nevertheless, more data are required, in order to support a more substantive conclusion.

Housing type and production target was the two factors, which caused a larger impact on the EFs. Other factors like the use of anaerobic digestion for manure treatment did not show a large impact because among the dairy cows only one farm did not treat their manure and their management was not comparable with the other farms.

3.3. Comparison of measured methane emissions with international and national inventory estimates

The measured emissions were compared to methane emissions predictions made by IPCC models and the Danish national model. Both IPCC models showed a similar average underestimation of emissions (−35% for the 2019 refinement, and −33% for the IPCC 2006 model (Fig. 5),

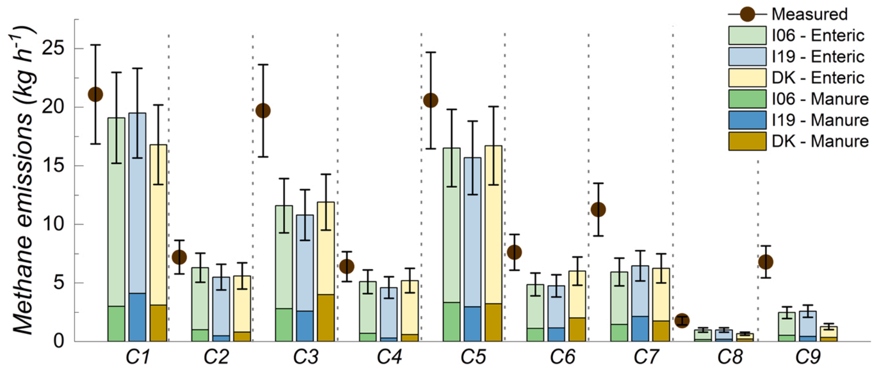

Fig. 5. Comparison between inventories and measured methane emission rates. The bars represent the emission estimated by the models, where the light colours correspond to enteric emissions and the dark colour shows manure emissions. The brown dot shows measured emissions. The error bar for the models corresponds to their respective uncertainties (± 20%), and for the measured emissions it corresponds to method uncertainty (± 20%) based on Fredenslund et al. (2019).

while the national Danish inventory resulted in the highest underestimation (−37%). Although the measured annual averaged emissions were higher than estimated by the inventory models for all farms (Fig. 5), the differences were within uncertainty limits when considering the models and measurements' uncertainty (as indicated by the error bars in Fig. 5). Exceptions were C3, C7, C8 and C9, for which none of the models managed to estimate emissions within the uncertainty limit. C3 and C7 utilised a deep litter house with high retention times (> 1 month). This type of management resulted in the largest differences between the measured and modelled emissions. Similar observations were noted at farm C6, which also used deep litter (Fig. 5). For this type of house management (with deep litter), the Danish model considers both the manure produced and the straw used for the beds in the VS estimation (Table A4), resulting in a higher manure methane emission than the IPCC models, although it is still lower than the measured emissions for C3 and C7 (Fig. 5). Fig. 6 shows the difference between international and national VS estimations, whereby the farms using deep litter as part of their dairy cow management (C3, C5, C6 and C7) all have higher VSs in the Danish model, in comparison to the IPCC models.

Considering the beef farms (C8 and C9), the national inventory performed worse (higher underestimation in comparison to measured emissions) than the IPCC models (Fig. 5). IPCC models estimate enteric emissions based on the weight of and energy used by the animals, whereas the Danish national guidelines use predefined feed intake according to the breed and the animal's life stage. As a result, the IPCC models calculated a significantly higher gross energy intake of 146 MJ head^{-1} day^{-1} in comparison to the Danish model, resulting in a value of 63 MJ head^{-1} day^{-1} (Fig. 6 farms C8 and C9), which might suggest the need to revise the Danish model's feed intake values used for bulls for slaughtering. The low gross energy used in the Danish models resulted in lower enteric emissions for this type of animal in this model (Table A5) and consequently in lower emission estimations. However, in comparison, the IPCC models also underestimated the emissions on these farms (C8 and C9), which might point to a lack of knowledge on emissions from beef cattle production, since it is unclear whether the source of error is enteric or manure emissions estimations. For C8, deep litter with a high retention time might have played a role in increasing model underestimation, as observed in the dairy cows' inventory comparison.

The differences between the 2006 and 2019 IPCC models were, on average, small, because the models are very similar in structure and to a large extent use analogous equations and approaches to estimate the variables, and in some cases they even use the same input parameters (such as for the net energy calculations) (IPCC, 2019, 2006). This is especially the case for modelling enteric fermentation emissions, which accounted for most of the estimated emissions (77% in average) (Fig. 5). Therefore, the differences between the two IPCC models (2006 and 2019 refinement) are mostly caused by differences in the modelling of emissions from manure management. For the anaerobic digestion of manure, IPCC 2019 uses a lower methane conversion factor (MCF) (3.5%) than IPCC 2006 (7.5%, based on the national model), resulting in a lower emission estimation by the 2019 methodology (farms C2, C3, C4, C5, C7 and C9). For on-site storage of liquid slurry (as was the case for farms C1 and C7), MCF values for IPCC 2019 were higher (14% and 22% for C1 and C7, respectively) in comparison to the MCF values used in IPCC 2006 (10% and 17% for C1 and C7, respectively), which made the manure emissions slightly higher in the IPCC 2019 predictions for those farms. For the Danish inventory, the annual MCF factors were estimated according to typical management conditions in the country, for cattle manure without (12.5%) and with biogas treatment (7.5%), while the MCF value for deep litter (17%) was adopted from the IPCC 2006 model.

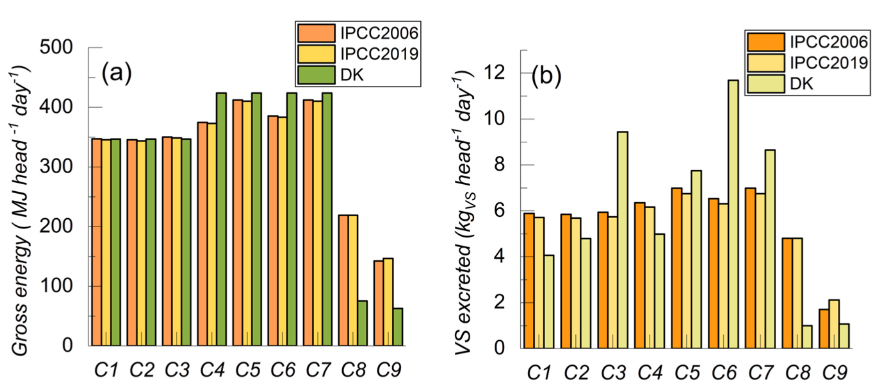

Fig. 6. Parameters used in the EF model calculations, estimated according to each model's methodology (for more information, please refer to Fig. 2. or Appendix A). (a) Gross energy intake (MJ head^{-1} day^{-1}) for dairy cows (C1 to C7) and bulls (C8 and C9). (b) Volatile solids excreted (kg_{VS} head^{-1} day^{-1}) for dairy cows (C1 to C7) and bulls (C8 and C9).

It is difficult to assess the impact of the MCF on emissions calculations in the Danish model in comparison to the IPCC models, because other parameters also play an important role, as discussed previously for VS.

A European study comparing top-down (tower measurements and inverse dispersion modelling) and bottom-up approaches (UNFCC/IPCC approaches) generated similar results to ours, with emissions being lower when using bottom-up inventories in comparison to top-down measurement approaches, albeit they were still within the uncertainty limits set for the modelled and measured emissions (Bergamaschi et al., 2015). In addition, a Danish study estimated methane emissions from manure stored under animal housing by calculating methane emission rates produced by incubating the collected manure at ambient temperatures (Petersen et al., 2016). They found that only the contribution made by manure emissions from animal housing was close to the total modelled manure emissions (accounting housing and external storage sources), which might indicate underestimations of total manure emissions by the national inventory (Petersen et al., 2016).

4. Conclusion

Annual whole-farm methane emissions were measured at nine cattle farms in Denmark, using the tracer gas dispersion method. Of the nine farms, seven were dairy and two beef cattle, and they were representative of common Danish breeds, housing systems and manure management practices. The seasonality of the emissions was addressed by measuring emissions every second month throughout a whole year. Methane emissions varied from 0.7 to 28 kg h^{-1}, while normalised measured emission factors (EFs) ranged between 14 and 54 g LU^{-1} h^{-1} for dairy and 11–24 g LU^{-1} h^{-1} for beef.

On average, the EF for dairy cows was 26 g LU^{-1} h^{-1} and for beef cattle 16 g LU^{-1} h^{-1}, the latter being approximately 38% lower than for dairy cow farms. Methane emissions tended to be higher in late summer/autumn (August to October), but annual variations in measured methane emissions were in general relatively low, varying between − 16% and + 13% of the annual mean emission for all farms. Among the dairy farms, housing systems using deep litter with high retention times seemed to result in higher emissions in comparison to farms using slatted or drained floors. Measurements of more farms are necessary to strengthen the conclusion that higher emissions are caused by deep litter house management and the mitigation of emissions using anaerobic digestion, which was not possible to evaluate herein.

A comparison of the measured emissions with modelled emissions showed an underestimation by all models: − 35%, − 33% and − 37% for IPCC 2019 and 2006 and the Danish national inventory, respectively. The underestimation fell within uncertainty limits for the modelled and measured emissions for most of the farms while for the beef farms this difference was large. The national model largely underestimated the measured emissions therefore a revision of national values in terms of feed intake for bulls for slaughter might be needed. Additionally, in order to improve model estimations of dairy cow methane emissions, the focus should fall on the estimation of manure emissions, with particular emphasis on deep litter management.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was funded in part by the Kvægafgiftsfoden: Måling af udledning af metan fra kvægbrug (27-2020), Denmark, and SEGES, Denmark. The authors wish to thank the farmers for their willingness to participate in the study and their assistance in collecting information from their facilities.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.agee.2022.107885.

References

Amon, A., Kryvoruchko, V., Amon, T., Zechmeister-Boltenstern, S., 2006. Methane, nitrous oxide and ammonia emissions during storage and after application of dairy cattle slurry and influence of slurry treatment. Agro Food Ind. Hi. Tech. 28, 2700–2704. https://doi.org/10.1016/j.agee.2005.08.030.

Amon, B., Amon, T., Boxberger, J., Alt, C., 2001. Emissions of NH3, N2O and CH4 from dairy cows housed in a farmyard manure tying stall (housing, manure storage, manure spreading). Nutr. Cycl. Agroecosyst. 60, 103–113. https://doi.org/10.1023/A:1012649028772.

Arndt, C., Leytem, A.B., Hristov, A.N., Zavala-Araiza, D., Cativiela, J.P., Conley, S., Daube, C., Faloona, I., Herndon, S.C., 2018. Short-term methane emissions from 2 dairy farms in California estimated by different measurement techniques and US Environmental Protection Agency inventory methodology: a case study. J. Dairy Sci. 101, 11461–11479. https://doi.org/10.3168/jds.2017-13881.

Baldé, H., VanderZaag, A.C., Burtt, S.D., Wagner-Riddle, C., Crolla, A., Desjardins, R.L., MacDonald, D.J., 2016. Methane emissions from digestate at an agricultural biogas plant. Bioresour. Technol. 216, 914–922. https://doi.org/10.1016/j.biortech.2016.06.031.

Bergamaschi, P., Corazza, M., Karstens, U., Athanassiadou, M., Thompson, R.L., Pison, I., Manning, A.J., Bousquet, P., Segers, A., Vermeulen, A.T., Janssens-Maenhout, G., Schmidt, M., Ramonet, M., Meinhardt, F., Aalto, T., Haszpra, L., Moncrieff, J., Popa, M.E., Lowry, D., Steinbacher, M., Jordan, A., O'Doherty, S., Piacentino, S., Dlugokencky, E., 2015. Top-down estimates of European CH4 and N2O emissions based on four different inverse models. Atmos. Chem. Phys. 15, 715–736. https://doi.org/10.5194/acp-15-715-2015.

Bjorneberg, D.L., Leytem, A.B., Westermann, D.T., Griffiths, P.R., Shao, L., Pollard, M.J., 2009. Measurement of atmospheric ammonia, methane, and nitrous oxide at a concentrated dairy production facility in southern Idaho using open-path FTIR spctrometry. Trans. Am. Soc. Agric. Biol. Eng. 52, 1749–1756. https://doi.org/10.13031/2013.29137.

Börjesson, G., Samuelsson, J., Chanton, J., Adolfsson, R., Galle, B., Svensson, B.H., 2009. A national landfill methane budget for Sweden based on field measurements, and an evaluation of IPCC models. Tellus Ser. B Chem. Phys. Meteorol. 61 B, 424–435. https://doi.org/10.1111/j.1600-0889.2008.00409.x.

Børsting, C.F., Hellwing, A.L.F., Lund, P., 2020. Normative Figures 2020. DCA - Danish Centre for Food and Agriculture, Aarhus University accessed 3.15.21. http://anis.au.dk/forskning/sektioner/husdyrernaering-og-fysi-ologi/normtal/.

Bühler, M., Häni, C., Ammann, C., Mohn, J., Neftel, A., Schrade, S., Zähner, M., Zeyer, K., Brönnimann, S., Kupper, T., 2021. Assessment of the inverse dispersion method for the determination of methane emissions from a dairy housing. Agric. . Meteor. 307 https://doi.org/10.1016/j.agrformet.2021.108501.

Danish agriculture & Food Council, 2020a. Statistics 2019 - Beef. URL ⟨https://agricultureandfood.dk/prices-and-statistics/annual-statistics⟩ (accessed 3.14.21).

Danish agriculture & Food Council, 2020b. Statistics 2019 - Dairy. URL ⟨https://agricultureandfood.dk/prices-and-statistics/annual-statistics⟩ (accessed 3.14.21).

Danish Agriculture & Food Council, 2018. Danish Dairy Industry [WWW Document]. Danish Agric. Food Counc, accessed 3.14.21. http://www.agricultureandfood.dk/danish-agriculture-and-food/danish-dairy-industry.

Daube, C., Conley, S., Faloona, I.C., Arndt, C., Yacovitch, T.I., Roscioli, J.R., Herndon, S. C., 2019. Using the tracer flux ratio method with flight measurements to estimate dairy farm CH4 emissions in central California. Atmos. Meas. Tech. 12, 2085–2095. https://doi.org/10.5194/amt-12-2085-2019.

Delre, A., Mønster, J., Scheutz, C., 2017. Greenhouse gas emission quantification from wastewater treatment plants, using a tracer gas dispersion method. Sci. Total Environ. 605–606, 258–268. https://doi.org/10.1016/j.scitotenv.2017.06.177.

Forster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J.L., Frame, D., Lunt, D.J., Mauritsen, T., Palmer, M.D., Watanabe, M., Wild, M., Zhang, H., 2021. The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity. In: Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S.L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M.I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J.B.R., Maycock, T.K., Waterfield, T., Yelekçi, O., Yu, R., Zhou, B. (Eds.), Climate Change 2021: the physical science basis. contribution of working group i to the sixth assessment report of the intergovernmental panel on climate change. Cambridge University Press.

Fredenslund, A.M., Rees-White, T.C., Beaven, R.P., Delre, A., Finlayson, A., Helmore, J., Allen, G., Scheutz, C., 2019. Validation and error assessment of the mobile tracer gas dispersion method for measurement of fugitive emissions from area sources. Waste Manag 83, 68–78. https://doi.org/10.1016/j.wasman.2018.10.036.

Galle, B.O., Samuelsson, J., Svensson, B.H., Borjesson, G., 2001. Measurements of methane emissions from landfills using a time correlation tracer method based on FTIR absorption spectroscopy. Environ. Sci. Technol. 35, 21–25. https://doi.org/10.1021/es0011008.

Grainger, C., Clarke, T., McGinn, S.M., Auldist, M.J., Beauchemin, K.A., Hannah, M.C., Waghorn, G.C., Clark, H., Eckard, R.J., 2007. Methane emissions from dairy cows measured using the sulfur hexafluoride (SF6) tracer and chamber techniques. J. Dairy Sci. 90, 2755–2766. https://doi.org/10.3168/jds.2006-697.

Hammond, K.J., Humphries, D.J., Crompton, L.A., Green, C., Reynolds, C.K., 2015. Methane emissions from cattle: estimates from short-term measurements using a

GreenFeed system compared with measurements obtained using respiration chambers or sulphur hexafluoride tracer. Anim. Feed Sci. Technol. 203, 41–52. https://doi.org/10.1016/j.anifeedsci.2015.02.008.

Hensen, A., Groot, T.T., Van Den Bulk, W.C.M., Vermeulen, A.T., Olesen, J.E., Schelde, K., 2006. Dairy farm CH4 and N2O emissions, from one square metre to the full farm scale. Agric. Ecosyst. Environ. 112, 146–152. https://doi.org/10.1016/j.agee.2005.08.014.

Hristov, A.N., Kebreab, E., Niu, M., Oh, J., Bannink, A., Bayat, A.R., Boland, T.M., Brito, A.F., Casper, D.P., Crompton, L.A., Dijkstra, J., Eugène, M., Garnsworthy, P.C., Haque, N., Hellwing, A.L.F., Huhtanen, P., Kreuzer, M., Kuhla, B., Lund, P., Madsen, J., Martin, C., Moate, P.J., Muetzel, S., Muñoz, C., Peiren, N., Powell, J.M., Reynolds, C.K., Schwarm, A., Shingfield, K.J., Storlien, T.M., Weisbjerg, M.R., Yáñez-Ruiz, D.R., Yu, Z., 2018. Symposium review: Uncertainties in enteric methane inventories, measurement techniques, and prediction models. J. Dairy Sci. 6655–6674. https://doi.org/10.3168/jds.2017-13536.

IPCC, 2019. 2019 Refinement to the 2006 IPCC Guidlines for National Greenhouse Gas Inventories. Vol. 4 Agric. For. other L. use 4, 209. URL ⟨https://www.ipcc-nggip.iges.or.jp/public/2019rf/index.html⟩ (accessed 3.14.21).

IPCC, 2013. Climate change 2013: the physical science basis. Contribution of Working Group I to the Fifth assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press,, Cambridge, United Kingdom and New York, NY, USA. https://doi.org/10.1017/CBO9781107415324.

IPCC, 2006. IPCC Guidelines for National Greenhouse Gas Inventories, Agriculture, forestry and other land use, Prepared by the National Greenhouse Gas Inventories Programme. CIGES,, Japan. Accessed 3.14.21. http://www.ipcc-nggip.iges.or.jp/public/2006gl/index.html.

Johansson, D.J.A., Persson, U.M., Azar, C., 2008. Uncertainty and learning: implications for the trade-off between short-lived and long-lived greenhouse gases. Clim. Change 88, 293–308. https://doi.org/10.1007/s10584-007-9381-x.

Kaharabata, S.K., Schuepp, P.H., 1998. Methane emissions from aboveground open manure slurry tanks oxygen demand z 218. Glob. Biogeochem. Cycles 12 (3), 545–554. https://doi.org/10.1029/98GB01866.

Kariyapperuma, K.A., Johannesson, G., Maldaner, L., VanderZaag, A., Gordon, R., Wagner-Riddle, C., 2018. Year-round methane emissions from liquid dairy manure in a cold climate reveal hysteretic pattern. Agric. . Meteorol. 258, 56–65. https://doi.org/10.1016/j.agrformet.2017.12.185.

Kupper, T., Häni, C., Neftel, A., Kincaid, C., Bühler, M., Amon, B., VanderZaag, A., 2020. Ammonia and greenhouse gas emissions from slurry storage - a review. Agric. Ecosyst. Environ. 300 https://doi.org/10.1016/j.agee.2020.106963.

Leytem, A.B., Dungan, R.S., Bjorneberg, D.L., Koehn, A.C., 2011. Emissions of Ammonia, methane, carbon dioxide, and nitrous oxide from dairy cattle housing and manure management systems. J. Environ. Qual. 40, 1383. https://doi.org/10.2134/jeq2009.0515.

McGinn, S.M., Beauchemin, K.A., 2012. Dairy farm methane emissions using a dispersion model. J. Environ. Qual. 41, 73. https://doi.org/10.2134/jeq2011.0266.

Mønster, J., Samuelsson, J., Kjeldsen, P., Rella, C., Scheutz, C., 2014. Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements - A sensitivity analysis based on multiple field surveys. Waste Manag 34, 1416–1428. https://doi.org/10.1016/j.wasman.2014.03.025.

Mønster, J., Samuelsson, J., Kjeldsen, P., Scheutz, C., 2015. Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method. Waste Manag. 35, 177–186. https://doi.org/10.1016/j.wasman.2014.09.006.

Ngwabie, N.M., Jeppsson, K.H., Gustafsson, G., Nimmermark, S., 2011. Effects of animal activity and air temperature on methane and ammonia emissions from a naturally ventilated building for dairy cows. Atmos. Environ. 45, 6760–6768. https://doi.org/10.1016/j.atmosenv.2011.08.027.

Nicks, B., Laitat, M., Vandenheede, M., Désiron, A., Verhaeghe, C., Canart, B., 2003. Emissions of ammonis, nitrous oxide, methane, carbon dioxide and water vapor in the raising of weaned pigs on straw-based and sawdust-based deep litters. Anim. Res. 299–308. https://doi.org/10.1051/animres.

Nielsen, O.-K., Plejdrup, M.S., Winther, M., Nielsen, M., Gyldenkærne, S., Mikkelsen, M. H., Albrektsen, R., Thomsen, M., Hjelgaard, K., Fauser, P., Bruun, H.G., Johanssen, V., Nord-Larsen, T., Vesterdal, L., Stupak, I., Scott-Bentsen, N., Rasmussen, E., Petersen, S.B., Olsen, T.M., Hansen, M.G., 2021. Denmark's national inventory report 2021. Emission Inventories 1990–2019 - submitted for publication under the United Nations Framework Convention on Climate Change and the Kyoto Protocol. URL ⟨http://dce2.au.dk/pub/SR437.pdf⟩ (accessed 3.14.21).

NOAA, 2021. NOAA/GML. ⟨https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/⟩ (Accessed 3.14.21).

Owen, J.J., Silver, W.L., 2015. Greenhouse gas emissions from dairy manure management: a review of field-based studies. Glob. Chang. Biol. 21, 550–565. https://doi.org/10.1111/gcb.12687.

Petersen, S.O., Olsen, A.B., Elsgaard, L., Triolo, J.M., Sommer, S.G., 2016. Estimation of methane emissions from slurry pits below pig and cattle confinements. PLoS One 11, 1–16. https://doi.org/10.1371/journal.pone.0160968.

Roscioli, J.R., Yacovitch, T.I., Floerchinger, C., Mitchell, A.L., Tkacik, D.S., Subramanian, R., Martinez, D.M., Vaughn, T.L., Williams, L., Zimmerle, D., Robinson, A.L., Herndon, S.C., Marchese, A.J., 2015. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement results. Atmos. Meas. Tech. 8, 2017–2035. https://doi.org/10.5194/amt-8-2017-2015.

Samuelsson, J., Delre, A., Tumlin, S., Hadi, S., Offerle, B., Scheutz, C., 2018. Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. Water Res. 131, 299–309. https://doi.org/10.1016/j.watres.2017.12.018.

Scheutz, C., Fredenslund, A.M., 2019. Total methane emission rates and losses from 23 biogas plants. Waste Manag. 97, 38–46. https://doi.org/10.1016/j.wasman.2019.07.029.

Scheutz, C., Kjeldsen, P., 2019. Guidelines for landfill gas emission monitoring using the tracer gas dispersion method. Waste Manag. 85, 351–360. https://doi.org/10.1016/j.wasman.2018.12.048.

Scheutz, C., Samuelsson, J., Fredenslund, A.M., Kjeldsen, P., 2011. Quantification of multiple methane emission sources at landfills using a double tracer technique. Waste Manag. 31, 1009–1017. https://doi.org/10.1016/j.wasman.2011.01.015.

SEGES, 2021. Årsstatistik 2020 - Dansk kødkvæg 1–12. URL ⟨http://www.danskkoedkvaeg.dk/media/3541/aarsstatistik-2020.pdf⟩ (accessed 3.14.21).

Sorg, D., Difford, G.F., Mühlbach, S., Kuhla, B., Swalve, H.H., Lassen, J., Strabel, T., Pszczola, M., 2018. Comparison of a laser methane detector with the GreenFeed and two breath analysers for on-farm measurements of methane emissions from dairy cows. Comput. Electron. Agric. 153, 285–294. https://doi.org/10.1016/j.compag.2018.08.024.

VanderZaag, A.C., Flesch, T.K., Desjardins, R.L., Baldé, H., Wright, T., 2014. Measuring methane emissions from two dairy farms: seasonal and manure-management effects. Agric. . Meteorol. 194, 259–267. https://doi.org/10.1016/j.agrformet.2014.02.003.

Wagner-Riddle, C., Park, K.H., Thurtell, G.W., 2006. A micrometeorological mass balance approach for greenhouse gas flux measurements from stored animal manure. Agric. . Meteorol. 136, 175–187. https://doi.org/10.1016/j.agrformet.2004.11.014.

Webb, J., Sommer, S.G., Kupper, T., Groenestein, K., Hutchings, N.J., Eurich-Menden, B., Rodhe, L., Misselbrook, T.H., Amon, B., 2012. Emissions of ammonia, nitrous oxide and methane during the management of solids manures. Agroecol. Strateg. Clim. Change Sustain. Agric. Rev. Volume 8. ed.

Wolf, J., Asrar, G.R., West, T.O., 2017. Revised methane emissions factors and spatially distributed annual carbon fluxes for global livestock. Carbon Bal. Manag. https://doi.org/10.1186/s13021-017-0084-y.