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Research article

In-vitro method and model to estimate methane emissions from liquid manure management on pig and dairy farms in four countries

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ABSTRACT

Methane (CH_4) emissions from manure management on livestock farms are a key source of greenhouse gas emissions in some regions and for some production systems, and the opportunities for mitigation may be significant if emissions can be adequately documented. We investigated a method for estimating CH_4 emissions from liquid manure (slurry) that is based on anaerobic incubation of slurry collected from commercial farms. Methane production rates were used to derive a parameter of the Arrhenius temperature response function, *lnA*', representing the CH_4 production potential of the slurry at the time of sampling. Results were used for parameterization of an empirical model to estimate annual emissions with daily time steps, where CH_4 emissions from individual sources (barns, outside storage tanks) can be calculated separately. A monitoring program was conducted in four countries, i.e., Denmark, Sweden, Germany and the Netherlands, during a 12-month period where slurry was sampled to represent barn and outside storage on finishing pig and dairy farms. Across the four countries, *lnA*' was higher in pig slurry compared to cattle slurry ($p < 0.01$), and higher in slurry from barns compared to outside storage ($p < 0.01$). In a separate evaluation of the incubation method, *in-vitro* CH_4 production rates were comparable with *in-situ* emissions. The results indicate that *lnA*' in barns increases with slurry age, probably due to growth or adaptation of the methanogenic microbial community. Using *lnA*' values determined experimentally, empirical models with daily time steps were constructed for finishing pig and dairy farms and used for scenario analyses. Annual emissions from pig slurry were predicted to be 2.5 times higher than those from cattle slurry. Changing the frequency of slurry export from the barn on the model pig farm from 40 to 7 d intervals reduced total annual CH_4 emissions by 46 %; this effect would be much less on cattle farms with natural ventilation. In a scenario with cattle slurry, the empirical model was compared with the current IPCC methodology. The seasonal dynamics were less pronounced, and annual CH_4 emissions were lower than with the current methodology, which calls for further investigations. Country-specific models for individual animal categories and point sources could be a tool for assessing CH_4 emissions and mitigation potentials at farm level.

1. Introduction

Methane (CH_4) is the second-most abundant anthropogenic greenhouse gas (Myhre et al., 2013), and livestock production is responsible for around a third of anthropogenic CH_4 emissions (UNEP, 2021). Globally, enteric CH_4 dominates emissions from livestock farms

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at around 90 % (Reisinger et al., 2021), and with a contribution from manure management of only around 10 % the need for mitigation of this source is often neglected (e.g., Herrero et al., 2016; Chang et al., 2019). However, there are regional differences, and the livestock sector of both the US (EPA, 2022) and the EU (Leip et al., 2010) have a share of CH_4 emissions from manure management of around 25 %, indicating that manure is a key source in some production systems. For example, on dairy farms in California with a Mediterranean climate, the contribution to CH_4 emissions from livestock production associated with manure management may exceed 50 % (Owen and Silver, 2015). Further, with intensive pig production the manure accounts for 87 % of CH_4 emissions according to the Global Livestock Environmental Assessment Model (FAO, 2017). Hayek and Miller (2021) reported that CH_4 emissions from intensive livestock production could be substantially underestimated, and while the causes are still unknown, the authors draw attention to "manure fluxes influenced by climate and geography".

Reducing enteric CH_4 emissions via feeding, additives or breeding requires meticulous investigations of animal health and productivity, and implementation has been slow (Jayasundara et al., 2016; Melgar et al., 2021). Therefore, in a 2030 or 2050 perspective, the technical potential for mitigation of CH_4 emissions from manure through treatment or changes in management may be equally high as that of enteric fermentation (Petersen, 2018).

Considering the regional distribution of manure management systems (Gerber et al., 2011), it can be estimated that liquid manure (slurry) accounts for >90 % of CH_4 emissions from manure management in both Europe and North America. Furthermore, liquid manure management is likely to increase in many parts of the world due to intensification of livestock production (FAO, 2010; Hayek and Miller, 2021), emphasizing the importance of mitigation strategies for liquid manure. For example, the biodegradation of slurry volatile solids (VS) during storage may be extensive (Møller et al., 2004a), and since these may increase with storage temperature (Patni and Jui, 1987; Popovic and Jensen, 2012), a more frequent removal from barns to cooler outside storage or for treatment, can reduce CH_4 emissions (Groenestein et al., 2012; Ma et al., 2023).

Following accepted inventory guidelines (IPCC, 2006; IPCC, 2019), annual CH_4 emissions from manure are currently estimated from CH_4 production potentials (B_0) of fresh manure and methane conversion factors, MCFs, that are determined by, e.g., manure management system, duration of storage, and temperature of the storage environment (IPCC, 2019). No distinction is made between different stages of manure management, yet manure in pits under slatted floors in barns, pumping pits, and outside storage facilities (tanks or ponds) are point sources that may contribute differently to farm emissions and have different mitigation potentials. Consequently, the effects of management such as frequency of export from barns, or the net effect of manure treatment prior to outside storage, are difficult to represent with a single MCF. The lack of a method to verify CH_4 emissions from individual sources on commercial farms may thus be a key barrier towards the development and adoption of effective mitigation technologies.

Quantifying CH_4 emissions from manure management is challenged by the complexity of farm environments. A recent survey of existing measurement data presented results from laboratory-, pilot- and commercial-scale studies that were based on tracer-ratio measurements, micrometeorological methods, or enclosure-based methods with a duration of at least six days (Hassouna et al., 2022). On-farm CH_4 measurements are inherently sensitive to enteric emissions and disturbance of wind profiles, while off-farm measurements may not be able to reproduce *in-situ* conditions over time. The present study evaluates an alternative method for quantifying CH_4 emissions which is based on short-term (24 h) anaerobic incubation of fresh manure at the *in-situ* temperature, i.e., off-farm but realistic with respect to time-lag and environmental conditions. This assay provides point measurements in time, not estimates of cumulative CH_4 emissions. Instead, the measurements are used to derive a key parameter for an empirical model with daily time steps (see next section).

In this study, liquid manure was sampled on intensive finishing pig and dairy farms in each of the four countries Denmark, Sweden, Germany and the Netherlands for determination of CH_4 production rates. The aim was to examine country- and system-specific differences in CH_4 production potential, and to calculate daily and annual CH_4 emissions using an empirical model. We hypothesized that estimates of CH_4 emissions would be higher for pig slurry compared to cattle slurry, and higher for slurry in barns compared to outside storage, especially for pig slurry. This is because the availability of degradable VS is expected to be higher in pig slurry, and because of the higher average storage temperature in barns compared to outside storage.

2. Empirical model

Liquid manure has a high biological oxygen (O_2) demand and a low oxygen exchange rate, and so is likely to be a predominantly anaerobic environment. The methodology investigated here assumes that the CH_4 production rate in livestock slurry stored on the farm (*in situ*) will be the same if the freshly collected slurry is incubated anaerobically (*in vitro*) at the temperature of the slurry at the time of collection. If this assumption is valid, as suggested by a pilot study (Petersen et al., 2016a), then sampling and incubation of slurry from commercial farms could provide a database of CH_4 production rate measurements representing a given livestock category or point source on the farm. Methane production rates will vary as a function of, e.g., the amount and composition of residual VS, storage temperature, and changes in the microbiological potential for CH_4 production (Dalby et al., 2021). However, it is neither feasible nor cost-effective to measure CH_4 emissions repeatedly on multiple farms; a model is therefore needed for estimating the daily CH_4 emission based on farm-specific activity data.

A simple model based on the Arrhenius equation was proposed by Sommer et al. (2004) to estimate CH_4 emissions from liquid manure based on residual VS and a temperature response function for methanogenesis:

$$F_t = (VS_d + 0.01VS_{nd})_t \, e^{\left(lnA - \frac{E_a}{RT_t}\right)}, \tag{1}$$

where F_t is CH_4 production rate (g CH_4 kg^{-1} VS h^{-1}) at time *t*, VS_d and VS_{nd} (kg kg^{-1} VS) are the residual fast degradable and slowly degradable ("nondegradable") fractions, respectively, of volatile solids (VS) in the slurry, E_a (J mol^{-1}) and *lnA* (g CH_4 kg^{-1} VS h^{-1}) are Arrhenius parameters, *R* is the universal gas constant (J K^{-1} mol^{-1}), and *T* is manure storage temperature (K). The parameter E_a represents the apparent activation energy of methanogenesis, and the average value 81 kJ mol^{-1} (95 % C.I. 74.9–87.1 kJ mol^{-1}) reported by Elsgaard et al. (2016) for methanogenesis in pig and cattle slurry, and digestates, represents the current best estimate of this property for methanogens in livestock manure (Baral et al., 2018; IPCC, 2019). In graphical depictions of the Arrhenius equation, process rate is plotted as a function of 1/T (e.g., Fig. 2 in Elsgaard et al., 2016). Here, *lnA* is the y intercept for $1/T \approx 0$ and represents a theoretical CH_4 production potential as determined by substrate availability and methanogenic potential of microorganisms in the chemical environment of the manure. If E_a is known, then it is possible to calculate *lnA* from measurements of CH_4 production rate, VS composition, and temperature by rearrangement of Eq. (1) (Petersen et al., 2016a):

$$lnA = ln\left[\frac{F_t}{(VS_d + 0.01VS_{nd})_t}\right] + \frac{E_a}{RT_t} \tag{2}$$

It has previously been shown that estimates of CH_4 emission with an Arrhenius response function are extremely sensitive to the value of *lnA*, and much less sensitive to VS degradability (Chianese et al., 2009; Petersen et al., 2016a). Since VS degradability is not easily measured, it was recently proposed to simplify Eq. (2) by expressing *lnA* with reference to total VS (Møller et al., 2022). Since lignin is not biodegradable in anaerobic environments, a better approach could be to use $dVS = total\ VS - lignin$ (Appuhamy et al., 2018) as the reference for derivation of *lnA*:

$$ln\ A' = ln\left[\frac{F_t}{dVS_t}\right] + \frac{E_a}{RT_t} \quad (3)$$

where *dVS* is slurry VS corrected for lignin (kg kg^{-1}), and the apostrophe indicates that $ln\ A'$ was calculated with reference to *dVS* and not VS_d+ $0.01VS_{nd}$ as originally proposed. The fraction of lignin may be estimated from measurements, or from diet composition and the characteristics of feed ingredients based on standard values. Hence, in the present study the empirical model used to calculate daily CH_4 production rates was:

$$F_t = dVS_t\ e^{\left(lnA' - \frac{E_a}{RT_t}\right)} \quad (4)$$

Finally, the CH_4:CO_2 ratio must be defined to estimate daily VS loss during slurry storage, since there is a need to account for the contribution of processes other than methanogenesis to VS loss, including aerobic degradation at the slurry-air interface (Møller et al., 2004b). Currently, this ratio is a simple average of 1:3 based on previous studies where emissions of both gases were quantified (e.g., Grant et al., 2015; Sommer et al., 2007). Ideally both CH_4 and CO_2 production rates should be determined experimentally, and for different stages of management, but this is complicated by the diversity of housing designs, storage systems and management practices. Møller et al. (2022) presented a sensitivity analysis for the empirical model and found that changing the CH_4:CO_2 ratio between 1:1 and 1:7 changed cumulative CH_4 emissions from untreated cattle and pig slurry by a maximum of 21 %, indicating that the error associated with this variable is modest when calculating CH_4 emissions with reference to total VS corrected for lignin.

Equation (4) is consistent with the IPCC methodology in that both calculations estimate CH_4 production from VS in the slurry, a temperature response function, and a methanogenic potential; in Eq. (4) the latter is represented by *lnA*', and in the IPCC methodology by the maximum methane producing capacity (B_0) of fresh excreta and a MCF for the corresponding livestock and manure management category. A key feature of the method investigated here is that it can represent the actual storage environment as modified by partial VS degradation, temperature and microbiology at each stage of manure management. With Eq. (3) and a database of measured CH_4 production rates (F_t) representing a specific source, it may thus be possible to determine region- and system-specific estimates of lnA' and parameterize Eq. (4) with a temperature response function that is based on observations, and to use this model for estimation of daily CH_4 emissions from information about slurry VS and its retention time at individual stages of manure management.

3. Materials and methods

The experimental part of the study was planned to acquire data to support model development. First, a database of *in-vitro* CH_4 production rates was obtained through monitoring in all four countries. In parallel, continuous measurements of slurry temperature and slurry level were recorded in selected countries to support a separate slurry temperature model. And finally, two method tests were carried out, the first test to compare *in-vitro* CH_4 production rates with actual emission measurements and the second test to examine effects of slurry age on the CH_4 production potential. In the following sections, these activities are described in more detail.

3.1. In-vitro assay

In preparation for monitoring, all partners adopted the *in-vitro* assay first described by Elsgaard et al. (2016) in their own laboratories. Briefly, 3-g samples of sieved (<2 mm) slurry ($n = 8$) are incubated anaerobically in crimp-seal test tubes at a constant temperature (here: within ±3 °C of the temperature of the slurry at the time of collection to support methanogens that were active in the original slurry environment). Anaerobic conditions should be ensured during handling and transfers of slurry subsamples. Two test tubes are terminated immediately to determine the amount of dissolved CH_4, while the other six test tubes are incubated over-night. The amount of VS in fresh and sieved samples is determined separately. Additional details are given by Elsgaard et al. (2016).

All monitoring programs followed the protocol, with incubation of subsamples within 24 h of sampling as described by Petersen et al., 2016a, but with some local modifications which are summarized in Table S2.

3.2. Monitoring on livestock farms

Representative finishing pig and dairy farms were selected in each country (Denmark, Sweden, Germany and the Netherlands). The distribution of the farms is shown in Fig. 1 and selected characteristics in Table 1; the results of a survey based on interviews are given in Table S1. Slurry samples representing barn or outside storage tanks were collected 3 to 5 times during a year (Table S1). In Denmark, three pig farms delivered freshly exported slurry to local biogas plants, and hence on these farms the outside storage tank contained digestate produced from a mixture of pig and cattle slurry, and other biomasses. Depth-integrated slurry samples were collected from pits in the barn, or from outside pumping pits on days of export, and the slurry temperature was simultaneously recorded. In the Netherlands, livestock slurry is collected in deep pits under slatted floors, and slurry was therefore only sampled in barns. In the other three countries, depth-integrated samples were also collected from the outside storage tanks.

3.3. Activity data

Slurry temperature is a key parameter, but difficult to document. Pig houses are heated with forced ventilation, and the temperature may be regulated from 20 down to 16 °C over the course of the production cycle (Petersen et al., 2016b), but in warmer periods or climates this may not be possible. Unpublished measurements of slurry temperature in Danish pig barns covering different periods of the year showed an average temperature of 19.7 °C ($n = 8$), and the average temperature of individual periods varied between 17.5 and 21.1 °C (H.B. Møller, personal communication). In the Netherlands, the slurry temperatures recorded in deep pits of pig houses were on average 21.6 °C. For model exercises, a constant in-house pig slurry temperature of 21.6 °C was assumed for the Netherlands, and 19.7 °C for Denmark and Germany (data from Sweden were not part of model calculations). Cattle barns are naturally ventilated, but with a higher average temperature in indoor slurry pits than in outside storage tanks (Groenestein et al., 2012). Here, it was assumed that slurry temperature in barns was always 5 °C higher than slurry temperature in the outside storage tank. These assumptions about in-house slurry temperature are consistent with the National Inventory Report for Denmark (Mikkelsen et al., 2016). In Sweden, there was no indoor storage, and instead the slurry was transferred daily to an outside pumping pit before further distribution to long-term storage.

Slurry temperature in outside storage tanks was recorded every 3 h on all farms in the Swedish monitoring program using Tinytag TG-4100 Temperature Data Loggers from Intab (Stenkullen, Sweden), which were attached to a chain hanging from a buoy at depths of 0.5, 1.5 and 2.5 m below the surface. When slurry was field applied in spring, the lower temperature loggers would record from the bottom position until the slurry level increased again. Additional data were obtained in Denmark and Germany using the same procedure. The temperature data were used to develop a model for predicting slurry temperature in outside storage tanks (Hafner and Mjöfors, 2023).

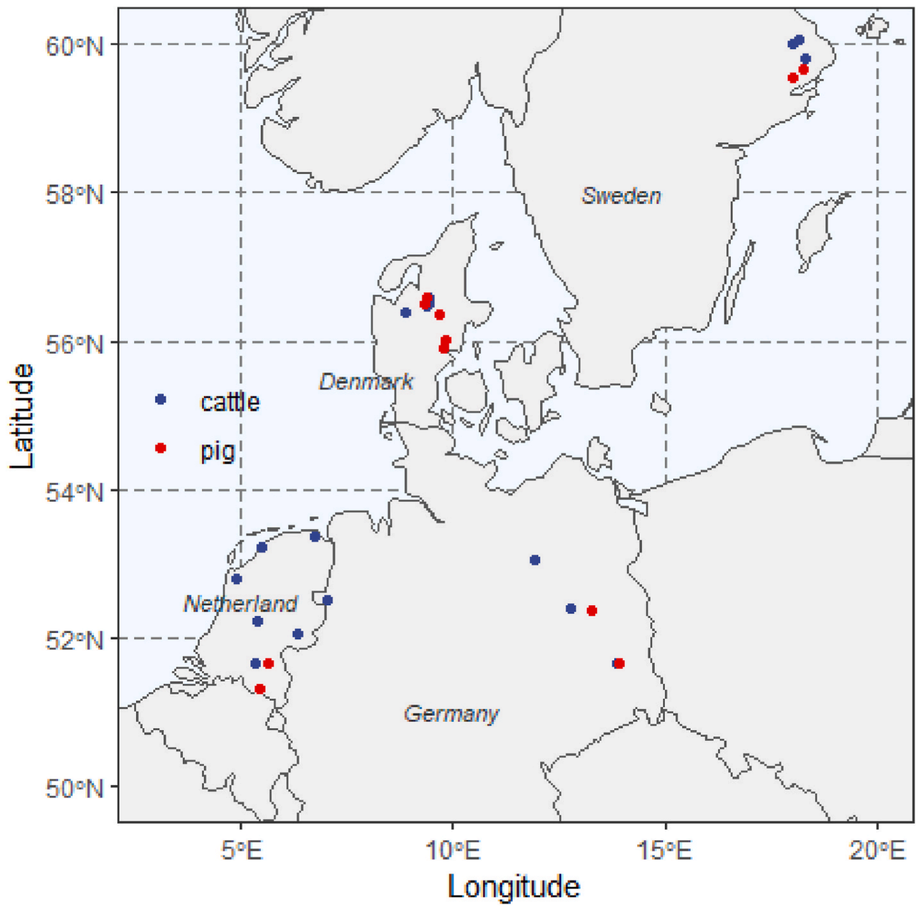

Fig. 1. Sampling sites of dairy and finishing pig farms included in the monitoring programs in Denmark, Sweden, Germany and the Netherlands. Selected information about farms is shown in Table 1, and a detailed account of farm characteristics and management practices in Table S1.

Slurry levels in storage tanks were recorded at the time of sampling only. However, continuous data on slurry level are needed for model calculations, and actual measurements could provide activity data as input for monitoring, reporting and verification (MRV). In two selected storage tanks from the Danish monitoring program with, respectively, pig slurry and cattle slurry, the level was continuously monitored during a full year with hourly measurements. Microflex D ultrasonic level transmitters (Interautomatika; Vilnius, Lithuania) were mounted on a custom-made platform fixed to the upper rim of the storage tank (Fig. S1). Distance to the surface was recorded hourly by a PMS-90R datalogger (Aplisens; Warszawa, Poland).

Total VS in sieved and unsieved slurry samples were determined by first drying subsamples at 105 °C for 24 h, followed by 3 h at 550 °C. The VS removed by sieving was assumed to have the same specific CH_4 production rate as the VS retained in sieved samples (Witarsa and Lansing, 2015). The lignin content of fresh excreta from dairy cattle and finishing pigs were 127 g kg^{-1} VS and 49 g kg^{-1} VS, respectively (Møller et al., 2004b), and the lignin content of digestates from centralised biogas plants processing a mixture of cattle and pig slurry and fiber-rich biomasses averaged 142 g kg^{-1} VS (Møller et al., 2020).

3.4. *In-vitro method evaluation*

To compare *in-vitro* CH_4 production rates with actual emissions, slurry was collected during an on-going pilot-scale study to determine emissions of CH_4 and other gases from pig slurry that was exported from barn to outside storage at 2–3 or 7 d intervals (Ma et al., 2023). The 6.5 m^3 storage tanks were ventilated at around 100 m^3 h^{-1} to simulate open storage, and ventilation air was subsampled hourly and collected in gas sampling bags for determination of weekly average CH_4 emissions based on concentrations measured by gas chromatography (Petersen et al., 2012) and ventilation rates. Samples for lab incubations were collected on 15, 18 and 25 June, and July 2, 2020, whereas composite ventilation air samples were retrieved on 18 and 25 June, and on 2 and July 7, 2020.

The effect of slurry age on CH_4 production rate was evaluated in a separate sampling campaign in December 2022 where one Danish pig farm was re-visited. Depth-integrated slurry samples were collected with a hand-operated pump below slatted floors in five different sections, four of which had 40 cm deep pits and 75 % slatted floor and contained either 40 kg or 60 kg body weight finishing pigs; here, slurry pits had been emptied either 9 or 15 d before sampling. The last section had a 60 cm deep pit and 100 % slatted floor, and contained 50 kg body weight pigs; in this section slurry had been collected during 29 d. In both tests, CH_4 production rates were determined using the *in-vitro* assay as described by Elsgaard et al. (2016).

3.5. *Data analyses*

To compare CH_4 production rates and lnA' of slurry from different sampling positions or livestock categories, a repeated-measures mixed-effects analysis of variance was carried out with livestock category and manure source as fixed effects. The analytical unit was the average values recorded for the individual farm, sampling position and day. Farm was nested within country as a random effect, and sampling date as a separate random intercept term. Methane production rates were log-transformed to improve the model fit. The 'lme4' package (v1.1-31) (Bates et al., 2015) in R (v4.1.1) was used to run the mixed-effects models, and the 'lmerTest' package (v3.1-3) was used to obtain F and

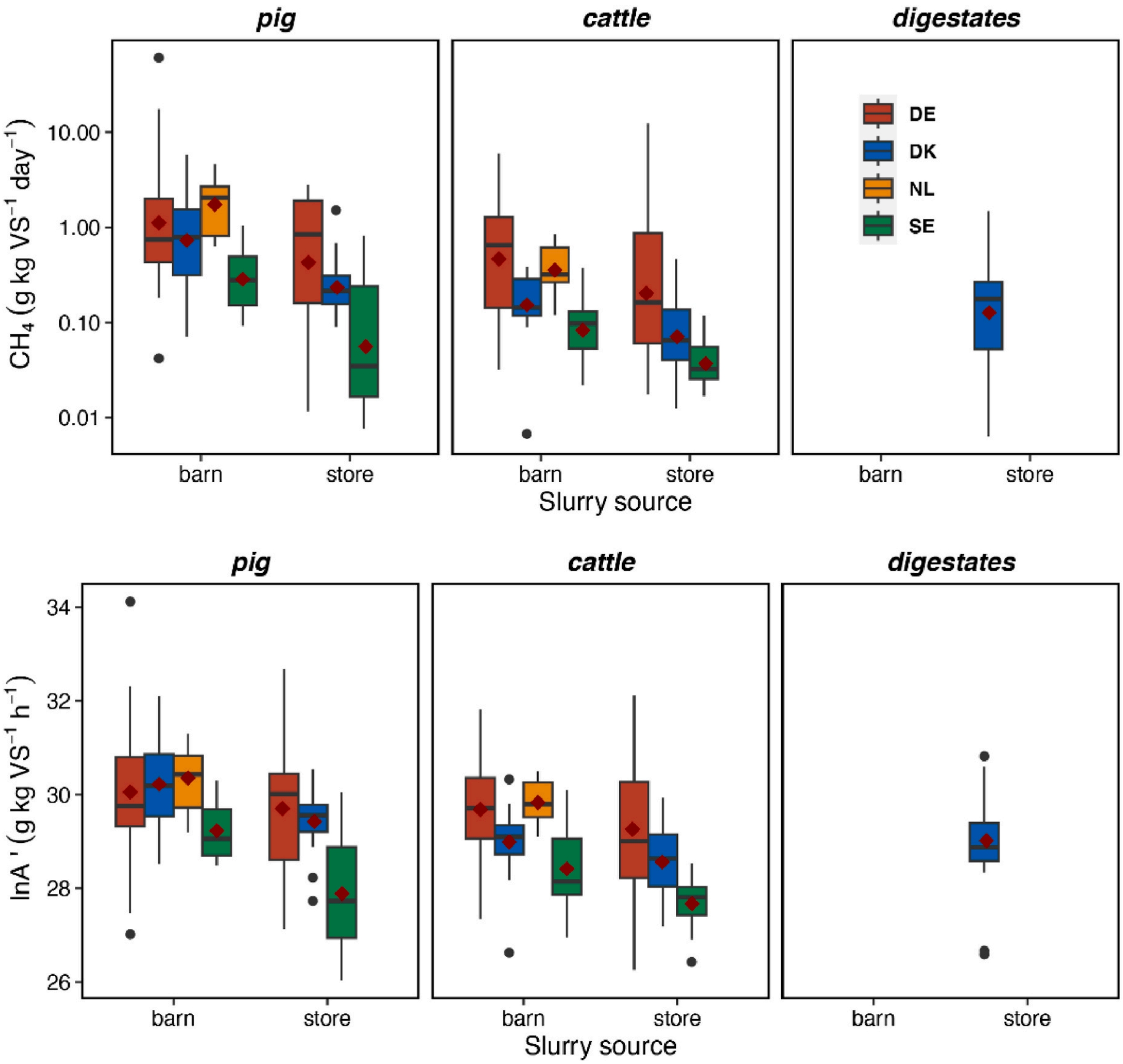

Fig. 2. Methane production rates (upper panel; note log scale) and the *lnA'* values (lower panel) of slurry from dairy farms and finishing pig farms as observed in 3–5 campaigns during a 12-month period in each of the countries Denmark, Sweden, Germany and the Netherlands. Box boundaries indicate the 25th and 75th percentiles. The horisontal lines indicate medians, and red dots the means. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

P values with Kenward-Roger approximation for degrees of freedom. In the formula notation of the lme4 R package, the models had the structure of log (CH_4 production rate) ~ livestock × source + (1|country/farm) + (1|date) and lnÃ livestock × source (1|country/farm) + (1|date).

The relationship between *in-situ* and *in-vitro* CH_4 production rates was analyzed by Spearman correlation analysis. Differences between slurry with different lengths of storage were evaluated with the Kruskal-Wallis test. All statistical analyses were run using the R statistical software (version 4.1.1) (R Core Team, 2023).

3.6. Scenarios

Based on Eq. (4), empirical models with daily time steps were defined to estimate daily and annual CH_4 emissions from slurry in barns and outside storage. The farms that were part of the monitoring program (Table S1) represented a wide range of housing and storage conditions, and management, and this limited dataset was insufficient to define models at country level. Instead, exemplary models were defined as described below.

Scenarios to investigate effects of export frequency and climate were based on livestock numbers and excretal returns in finishing pig and dairy farms previously defined for the farm model FarmAC (https://www.farmac.dk/), which was developed as part of the European project AnimalChange (https://animalchange.eaap.org/). The dairy farm was assumed to have 168 cows and 168 heifers in a barn with ring channels and outside manure storage capacity of 5087 m^3. Calculations assumed an excretion of 1200 kg VS LU^{-1} yr^{-1}. The finishing pig farm was assumed to produce 3395 pigs yr^{-1} in barns with shallow pits having a pull-plug system for export. Calculations were based on the excretion of 158 kg VS (pig place)$^{-1}$ yr^{-1}. The outside storage tank in this case had a capacity of 2707 m^3.

In the scenarios investigated, the driving variables were the daily excretion of *VS*, the export frequency and time of field application, and the daily air temperature. The excretion of VS, and conversion of VS to slurry volumes, were based on standard values (Børsting and Hellwing, 2022) and assuming 80 % VS in excreted dry matter. On dairy farms the typical slurry residence time in the barn has been estimated at 20.1 d (Mikkelsen et al., 2016), corresponding to a 40 d interval between exports, although with ring channels only a minor part of the slurry is

Table 1
Selected characteristics of the farms in Fig. 1 where manure was sampled 2–5 times during a one-year period. A more detailed account of farm characteristics and management practices is available in Table S1.

Country	Annual mean air temperature	Manure export frequency, pig/cattle	Manure management, pig/cattle
	°C	Interval, avg. days [a]	
Sweden (SE)	6.9	1/0.38	Storage for field application/Storage for field application
Denmark (DK)	9.0	17/7	Storage for field application or biogas treatment/Storage for field application
Germany (DE)	10.8	52/2.75	Storage for field application/Storage for field application
Netherlands (NL)	11.1	49/120	Storage for field application or biogas treatment/Storage for field application or biogas treatment

[a] Wide ranges in some cases, see Table S1. In cattle barns with ring channels, only part of the slurry is exported.

exported on each occasion (cf. Fig. S1A). Similarly, the average residence time of slurry in barns with finishing pigs has been determined at 19.0 d in Denmark (Mikkelsen et al., 2016). Based on this information, a reference situation with 40 d between exports from barn to an outside storage was selected. It was assumed that 95 % of the accumulated cattle slurry in outside storage tanks was removed for field application in April and again in July (Table S1), while for pig slurry a 95 % removal for field application was assumed to occur in April, which is a simplification of actual practice (Table S1).

Slurry temperature in the outside storage tank was calculated with the STM model (Hafner and Mjöfors, 2023) using daily air temperature (Denmark) for the period 2019–2021. Slurry temperature was calculated at daily resolution using v1.0 of the heat transfer model STM (https://github.com/AU-BCE-EE/STM/releases/tag/v1.0) with default input parameters (pars.txt file v1.0) together with slurry tank dimensions and other settings specified separately for each tank in a user-defined parameter file (see Table S4 for an example). Other inputs included daily mean air temperature and global radiation, which were taken from nearby weather stations, while slurry level was defined as described below.

The parameter values for lnA' used to calculate CH_4 emissions from barn and outside storage on finishing pig and dairy farms, respectively, were means of the values obtained in monitoring programs in Denmark, Germany and the Netherlands (barn only), with Sweden excluded for reasons discussed below. For each daily time-step, *VS* degradation was calculated and subtracted as input for the next time-step. The carbon (C) content of VS was set to 0.45 kg kg^{-1} (Petersen et al., 2016a), and it was assumed that C was lost as CH_4 and CO_2 emissions at a molar ratio of 1:3 (Nielsen et al., 2021).

The first model exercise examined the effect of changing the export frequency from 40 to 7 d intervals. In a second scenario using the model dairy farm, daily CH_4 emissions from barn and outside storage were calculated, and total monthly emissions were compared with monthly CH_4 emissions as calculated by the spreadsheet provided in the 2019 refinement of guidelines for national inventories (IPCC, 2019). In both scenarios, daily CH_4 emissions were calculated for a three-year period to stabilize VS and slurry volumes in the storage tank (IPCC, 2019). When referring to annual emissions, this represents the 3rd year of model runs.

4. Results

4.1. On-farm monitoring of CH_4 production potentials

Using an *in-vitro* assay, CH_4 production rates were determined in slurry samples from barns and outside storage tanks collected during a one-year period in each of four countries (Fig. 2, upper panel; in the Netherlands from barns only). The CH_4 production rates showed high variability, and the distributions are depicted as box plots on a logarithmic scale.

Using Eq. (3), average values of lnA' were calculated by country, livestock category and source as a measure of CH_4 production potential of the samples (Fig. 2, lower panel). For barn as well as outside storage, lnA' values of pig slurry were significantly higher than those of cattle slurry ($p < 0.01$), and the lnA' values of slurry in barns were higher than those of slurry in outside storage tanks ($p < 0.01$; Table S3). The lnA' of digestates from centralised biogas plants, which represented a mixture of pig and cattle slurry co-digested with other organic wastes, were not significantly different from lnA' estimates of untreated slurry ($p > 0.05$, post-hoc Tukey's test).

As mentioned above, in Germany and the Netherlands, slurry samples representing barn storage were collected directly from pits below slatted floors, whereas in Denmark and Sweden the slurry was collected from a pumping pit on days of slurry export to outside storage tanks. Germany, the Netherlands and Denmark showed similar lnA' values for barns (Fig. 2), whereas in Sweden with daily export the $ln\,A'$ values of slurry from barns as well as outside storages were significantly lower compared to the three other countries ($p < 0.01$, post-hoc Tukey's test). Seasonal trends were absent across the four countries, and therefore the uncertainty ranges in Fig. 2 represent between-farm as well as seasonal variation.

Short-term changes in $ln\,A'$ were seen in the separate campaign in Denmark where pig slurry was sampled from pits underneath sections with finishing pigs at 9, 15 or 29 d after the previous removal. The two shorter collection periods (9 and 15 d) showed similar rates at a level that was five-fold lower than the rate observed after a 29 d collection period (Fig. 3A). This difference was reflected in the derived $ln\,A'$ values (Fig. 3B).

4.2. Slurry temperature and volume

Documenting slurry export from barn to outside storage could become an important part of future MRV schemes for greenhouse gas mitigation, and a method to monitor slurry level in storage tanks throughout the year was investigated, in which distance to the liquid surface was recorded with ultrasound level transmitters installed on the edge of a storage tank with pig slurry having a tent cover (Fig. S1A), and an open storage tank with cattle slurry (Fig. S1B). While the seasonal dynamics and times of import and export were visible in the recorded slurry levels, even on a sub-weekly basis, there were also some transient peaks, marked in dark green in Fig. S1, which could not be explained. Due to this challenge, and to the fact that documentation of daily slurry level in outside storage tanks was not available for modeling of CH_4 emissions on most farms in the monitoring program, slurry volumes were instead calculated from standard values for manure production and VS content (Børsting and Hellwing, 2022).

4.3. Scenarios with daily and annual CH_4 emissions

Management effects on CH_4 emissions from slurry were calculated for typical dairy and finishing pig farms using the means of $ln\,A'$ values for barn and outside storage observed in the monitoring programs conducted in Denmark, Germany and the Netherlands (barn only), and climatic conditions for Denmark. In the last year of a three-year model run, the total emission from cattle slurry constituted 12.6 g CH_4 kg^{-1} VS yr^{-1}, and the emission from pig slurry 43.2 g CH_4 kg^{-1} VS yr^{-1}. Both of

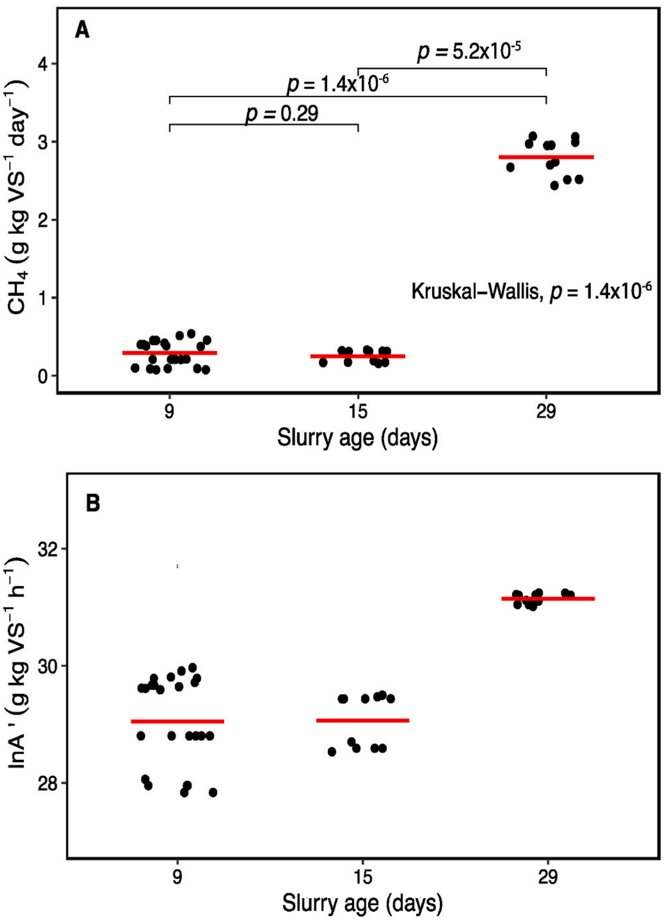

Fig. 3. Methane production rates (A) and corresponding $ln\,A'$ values (B) of finishing pig slurry collected from pits below slats in sections where the slurry had been collected for 9, 15 or 29 d, see text for further details. Overall p-values represent results from a Kruskal–Wallis test among slurry age groups, and pairwise p-values were obtained by the Wilcoxon test.

these scenarios assumed a 40 d interval between exports from barn to outside storage.

The emissions of CH_4 from pig slurry in barn and outside storage were calculated to be 21.1 and 22.1 g CH_4 kg^{-1} VS yr^{-1} (Table 2), and the corresponding daily emissions are shown in Fig. 4A. Changing the export interval to 7 d (Fig. 4B) greatly reduced emissions from slurry in the barn to 4.47 g CH_4 kg^{-1} VS yr^{-1}, whereas emissions from the outside storage increased to 25.1 g CH_4 kg^{-1} VS yr^{-1} (total: 29.6 g CH_4 kg^{-1} VS yr^{-1}). Overall, a 46 % reduction was predicted by this management change.

For cattle slurry, predicted daily CH_4 emission rates from barn and outside storage are shown in Fig. 5A for a three-year period. The specific CH_4 emission per kg VS was consistently higher for the barn compared to outside storage, but due to the longer residence time in the outside storage tank, the cumulative CH_4 emission was higher from this source (Table 2). The monthly CH_4 emissions calculated by the empirical model were compared with those calculated using the recently updated IPCC methodology (Fig. 5B). The seasonal dynamics were greater with the IPCC method, and cumulative emissions were much higher, i.e., 30.3 *vs.* 12.6 g CH_4 kg^{-1} VS yr^{-1} (Table 2); a difference, albeit less, was also observed with pig slurry. Possible reasons for these deviations are discussed below.

Table 2
Annual average CH_4 emission per kg VS excreted on dairy and finishing pig farms with liquid manure management as calculated with an empirical model using daily time steps, and with separate accounting of emissions from barn and outside storage (40 d export interval). The $ln\,A'$ values used for model parameterization were the mean of measurement results in Denmark, Germany and the Netherlands (excluding Sweden which had daily export), and climate corresponded to Denmark; see section 3.5 for additional details. Annual CH_4 emissions per kg VS excreted were also calculated with the IPCC methodology (IPCC, 2019) using the spreadsheet provided to account for mean monthly temperature (available at: https://www.ipcc-nggip.iges.or.jp/public/2019rf/vol4.html).

		Empirical model			IPCC
		barn	storage	total	total
		g kg^{-1} VS yr^{-1}			g kg^{-1} VS yr^{-1}
Cattle	1st year	5.3	4.7	10.0	28.6
	2 nd year	5.2	7.1	12.3	29.3
	3rd year	5.2	7.4	12.6	30.3
Pig	1st year	21.0	15.2	36.2	46.2
	2 nd year	21.1	21.4	42.5	74.7
	3rd year	21.1	22.1	43.2	74.7

4.4. *In-vitro vs. in-situ measurements*

The assumption that a laboratory assay can represent *in-situ* CH_4 emissions was evaluated by *in-vitro* measurements of pig slurry sampled four times at weekly intervals (June–July 2020) during an 8-week storage experiment where CH_4 production rates could be compared with actual CH_4 emissions. The two independent estimates of CH_4 emissions were comparable ($p = 0.164$), but with higher rates from the 7 d treatment ($p < 0.001$) and no interaction between measurement method and treatment, and thus both methods indicated lower emissions for 2–3 d compared to 7 d export intervals (Fig. 6).

5. Discussion

In support of nutrient circularity, livestock manure is increasingly stored for extended periods to ensure efficient use of manure nutrients

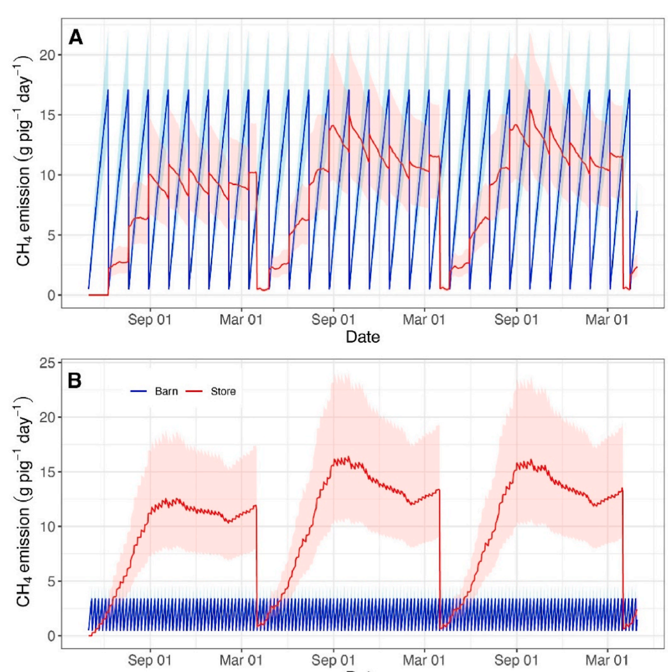

Fig. 4. Daily CH_4 emissions (g pig^{-1} d^{-1}) from a typical finishing pig production system during a three-year period as calculated using Eq. (4) in an empirical model with 40 d (A) or 7 d (B) interval between exports from barn to the outside storage. In both scenarios, 95 % of the manure in the outside storage was assumed to be field-applied on 1 April. The shaded areas represent the uncertainty of 95 % confidence ranges for $ln\,A'$.

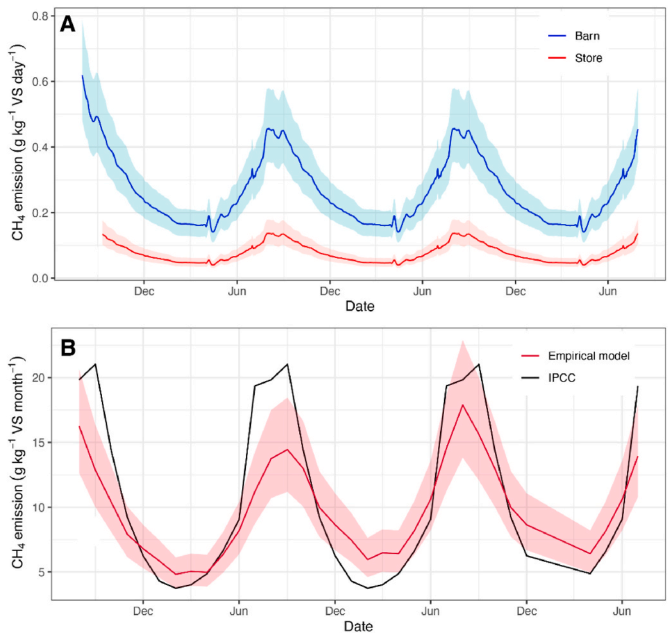

Fig. 5. The upper panel (A) shows specific CH_4 emission rates (g CH_4 kg^{-1} VS d^{-1}) in barn and outside storage during a three-year period for a typical dairy production system (see text for management). The lower panel (B) shows specific total CH_4 emission rates (g CH_4 kg^{-1} VS month^{-1}), as calculated using Eq. (4), where shaded areas represent the uncertainty of 95 % confidence ranges for $ln\,A'$. The lower panel also shows (black line) the corresponding rates calculated according to IPCC, 2019.

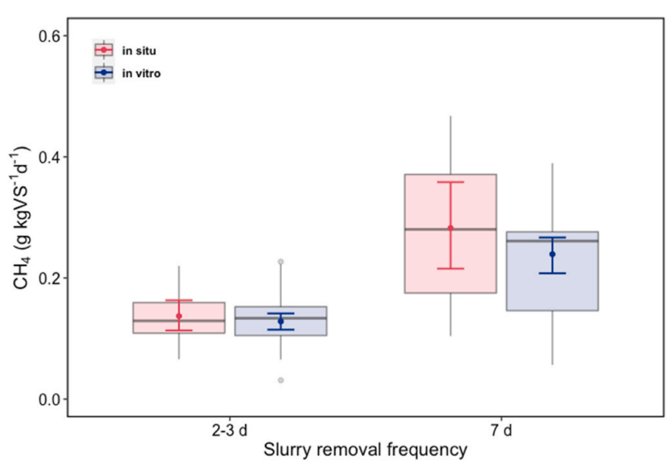

Fig. 6. The CH_4 production rates determined with the *in-vitro* assay were compared with actual emissions in connection with an on-going pilot-scale storage experiment, where pig slurry was exported from the barn at either 2–3 d intervals (three times per week) or weekly. The CH_4 emission rate measurements were based on time-integrated sampling of ventilation air in gas sampling bags for weekly analysis by gas chromatography (Ma et al., 2023).

for crop production by timely application in the field (Oenema et al., 2011; van der Wiel et al., 2020). With liquid manure management, the CH_4 emission during storage may dominate the carbon footprint of a manure management chain that includes field application (Baral et al., 2018; Meng et al., 2023), and this study was motivated by the need for methods to monitor CH_4 emissions, and effects of mitigation measures.

5.1. Barn vs. outside storage

There was a significant difference between $ln\,A'$ values of slurry from barns and outside storage tanks. A main reason for lower CH_4 production potentials in outside storage tanks is probably depletion of substrates to sustain the methanogenic community (Dalby et al., 2021). Even if slurry from the barn is introduced at regular intervals, the average age and degree of degradation of manure VS in the outside storage will always be greater than in the barn. Management may also influence CH_4 production potentials, and in Sweden with daily export the $ln\,A'$ values of slurry were significantly lower compared to those in Denmark, Germany, and the Netherlands where the retention time of slurry in barns was several weeks or even months (Table S1). The average air temperature was lower in Sweden compared to the other three countries (Fig. 1), but this should not have influenced the CH_4 production potential of slurry from climate-controlled pig barns. Previous studies have also concluded that frequent export will reduce CH_4 emission rates from animal houses (Sommer et al., 2007; Dalby et al., 2022; Ma et al., 2023), especially when the pits are cleaned between collection periods (Haeussermann et al., 2006).

A lower $ln\,A'$ value was observed in Sweden also during outside storage, although the retention time here was at least as long in Sweden as in Denmark and Germany. Methanogens in fresh excreta are adapted to the digestive system of the animals and probably not active in slurry pits with ambient temperature and a different chemical environment (Demirel and Scherer, 2008), and thus activity and growth of other methanogens adapted to the slurry environment may be a precondition for substantial CH_4 emissions. Fotidis et al. (2013) reported that changes in the availability of acetate and ammonium induced changes in the methanogen community composition, and Habtewold et al. (2018) observed that increasing CH_4 emissions during a 100-day storage of cattle slurry were accompanied by an increasing proportion of methanogens belonging to the acetotrophic (but metabolically diverse) genus *Methanosarcina*. Low numbers of adapted methanogens in daily exported slurry, in combination with lower outside storage temperature, could therefore help explain the lower CH_4 production potential developing in Sweden in this study (Fig. 2).

Lower CH_4 production rates and $ln\,A'$ values were observed in 1–2 week old compared to 4 week old pig slurry in shallow pits in the barn (Fig. 3), and lower CH_4 production rates during outside storage were observed with 2–3 d as opposed to 7 d export interval (Fig. 6). In accordance with this, Ma et al. (2023) reported lower cumulative CH_4 emissions from 2–3 compared to 7 d export interval in the barn, as well as during outside storage. Together these observations suggest that a shorter interval between excretion and export to an outside storage with lower temperature can reduce CH_4 emissions from liquid manure, not only because of the temperature difference but also because the growth of methanogens adapted to the slurry environment is suppressed. In the context of the empirical model, this would translate into a lower CH_4 production potential as defined by $ln\,A'$.

The present study included housing systems with slurry export intervals ranging from <1 d (Sweden) to 180 d (the Netherlands), and this probably contributed to the variation in $ln\,A'$. However, only Sweden with daily export deviated significantly from other countries. The anaerobic degradation of organic matter consists of four steps, i.e., hydrolysis, acidogenesis, acetogenesis, and methanogenesis (Dalby et al., 2022), and possibly in the early stages of degradation the microbial potential for methanogenesis is a limiting factor unless a well-adapted inoculum is present in residual slurry, whereas at later stages the hydrolysis of residual VS becomes more important.

The scenarios with 40 d (Fig. 4a) *vs.* 7 d (Fig. 4b) export intervals for pig slurry indicated a nearly ten-fold reduction of CH_4 emissions from manure in the barn, and a reduction in total annual emissions of 46 %. In a separate study presenting experimental results for the same two export intervals (Ma et al., 2023), reductions of CH_4 emissions from the barn with 7 d export interval were 55 and 61 %, and reductions in total CH_4 emissions 17 and 52 %, under summer and winter storage conditions, respectively, which is in reasonable agreement with model estimates.

Hence qualitatively, the model gave a realistic response to this management change.

5.2. Livestock category

Methane production potentials, as represented by $ln\,A'$, were higher for pig slurry than for cattle slurry, in accordance with the default values for biodegradability of VS in fresh excreta from pigs and cattle assumed in the IPCC methodology of 0.45 and 0.24 m^3 CH_4 kg^{-1} VS, respectively (IPCC, 2006). Hence the difference in theoretical potential for CH_4 production was still visible during storage despite partial degradation and likely changes in the chemical composition of slurry VS. The higher $ln\,A'$ would, for a given storage temperature, translate into higher CH_4 emissions from pig slurry compared to cattle slurry.

The empirical model predicted that CH_4 emissions from pig slurry would be 2.5-fold higher compared to cattle slurry, and this was also the case when estimated with the updated IPCC methodology (IPCC, 2019), cf. Table 2. Studies on commercial farms have reported CH_4 emission rates from pig slurry that are twice as high as from cattle slurry (Kupper et al., 2020), a difference that was reasonably well predicted by both calculation methods. However, the emissions of CH_4 estimated with the empirical model were only around 50 % of those estimated with the IPCC methodology. The estimates of the empirical model were, especially for cattle slurry, in the lower end of the range presented by Hassouna et al. (2022) in a review of results compiled in a global database, DATAMAN, with a predominance of experimental results from liquid manure management in wet temperate climate, as in the present study. Potential biases must therefore be considered, including those of model parameters, activity data, and the sampling and analytical procedures.

5.3. Accuracy and precision of Arrhenius parameters

The parameter E_a of the temperature response function was adopted from Elsgaard et al. (2016), and this is also the case in the revised IPCC methodology (IPCC, 2019). Hence, any error in this parameter would not explain the difference between the empirical model and the IPCC-based estimates. However, the average value for slurry and digestates of 81 kJ mol^{-1} reported by Elsgaard et al. (2016) had a wide confidence range of 74.9–87.1 kJ mol^{-1}, and hence there could be a general bias associated with this parameter.

We interpret $ln\,A'$ as a measure of CH_4 production potential as determined by availability of degradable substrate and the microbiological capacity for conversion of degradable VS to CH_4 as modified by the chemical environment. It is an empirical value as enzymatic processes have a defined optimum and temperature range (Peleg et al., 2012), but Elsgaard et al. (2016) found a linear relationship with 1/T for typical storage temperatures (5–35 °C) which allows estimation of $ln\,A'$. During monitoring, the *in-vitro* assay was, for logistical reasons, initiated the day after sampling, and this could have reduced the amount of degradable VS remaining following overnight storage at near-ambient temperature. Therefore, the pig slurries sampled from pits after 9, 15 or 29 d collection periods were used to compare CH_4 production rates incubated on the day of sampling and 24 h later (data not shown), but the difference was not significant (pig slurry, $p = 0.12$; $n = 5$).

The ranges of $ln\,A'$ observed in the national monitoring programs (Fig. 2) may be compared with previous studies. Elsgaard et al. (2016) reported *lnA* values for a selected cattle and pig slurry of 33.3 and 31.1 g CH_4 kg^{-1} VS d^{-1}, respectively, which is equivalent to 30.1 and 27.9 g CH_4 kg^{-1} VS h^{-1}, i.e., $ln\,A_h = \ln(exp(lnA_d)/24)$ where A_d and A_h represent the pre-exponential factor per day and per hour, respectively. This was higher and lower, respectively, than the levels observed for outside storages in Denmark and Germany. For pig slurry, this can be explained by the fact that slurry was sampled from a storage tank which had not received fresh material for at least six months. In contrast, the cattle slurry storage sampled by Elsgaard et al. (2016) had received fresh material from the barn and was mixed on the day of sampling, and therefore the average degradability may have been relatively high. Møller et al. (2022) estimated $ln\,A'$ from selected storage experiments in a desk study where average values for cattle and pig slurry were 29.2 and 30.3 g CH_4 kg^{-1} VS h^{-1}, respectively, which was in reasonable agreement with the present study.

Sieving was part of the standard procedure to improve reproducibility, and this was not expected to change short-term rates substantially (Witarsa and Lansing, 2015), but could be an issue with prolonged incubation (Rico et al., 2007). Some fiber-rich cattle slurries were difficult to sieve, and prolonged exposure to oxygen could have stressed the methanogens. A procedure with larger sample volumes could enable incubation without sieving without loss of precision, and as part of the project behind this study an instrument for the determination of CH_4 production rates in up to 2-L samples at ambient temperatures is currently being developed by BPC Instruments (https://bpcinstruments.com/). Increasing the sample volume would also increase gas production and potentially reduce analytical error and the variability among observations, which was high (Fig. 2), although it should be noted that this variability also represented the diversity of farms and management practices (Table S1). Prolonging the anaerobic incubation to 7 or more days was also examined as a strategy to increase the sensitivity of the *in-vitro* assay. Results (not shown) were promising, but different from the observations reported by Elsgaard et al. (2016), and therefore incubation conditions must be investigated further.

The estimates of $ln\,A'$ from individual campaigns were pooled because of the limited number of sampling days that were possible with the available resources. Yet, $ln\,A'$ may vary during the year, and since the response of methanogenesis to temperature is nonlinear (Elsgaard et al., 2016) a simple average could underestimate annual emissions. A higher storage temperature may promote the growth of methanogens (van den Berg, 1977), although Guo et al. (2020) found that an observed increase in CH_4 emissions with increasing temperature was not associated with community changes, but rather with increased activity of existing methanogens as revealed by transcription of *mcrA*, a gene encoding a key enzyme in methanogenesis. The storage temperature could influence CH_4 production potential indirectly via the hydrolysis of complex carbohydrates to substrates for fermentation and methanogenesis (Vavilin et al., 2008). The fact that the empirical model was used with annual average $ln\,A'$ values rather than, e.g., monthly values may partly explain the deviations in CH_4 emission from the IPCC methodology across the year (Fig. 5), and the sensitivity of $ln\,A'$ to storage temperature should be explored further.

5.4. Accuracy and precision of other variables

The empirical model originally proposed for the estimation of CH_4 emission from liquid manure (Eq. (1)) defined two pools of VS, i.e., fast degradable VS and "the rest" (Sommer et al., 2004). This approach was later used in other studies (e.g., Rotz et al., 2010; Petersen et al., 2016a; Baral et al., 2018). The initial degradability of VS can be estimated from the biochemical composition of fresh excreta, but a reliable method for determining degradable VS in a slurry sample of unknown age and composition is currently not available. Therefore, in the present study *dVS*, i.e., non-lignin organic dry matter, was proposed as basis for estimation of residual degradable VS. Methods to determine lignin in livestock manure are well-established, and this approach may therefore be more robust for characterisation of bioavailable VS during manure storage. Experimental data on lignin content in slurry from barns and outside storage tanks were recently reported by Hilgert et al. (2023) that were consistent with the lignin contents of Møller et al. (2004b) for fresh excreta used in the present study.

There is limited knowledge about the proportion of C in VS that is emitted as CH_4 when degraded. The relationship between organic C and VS, as determined by loss-on-ignition, is subject to analytical error but appears robust with R^2 values of 0.95 or higher in most cases (Pribyl, 2010). The VS in excreta contains a high proportion of carbohydrates, and the C content of 0.45 kg kg^{-1} assumed in the present study is within the range 0.40–0.45 kg kg^{-1} characteristic of carbohydrates (Pribyl, 2010).

The relationship between VS degradation and CH_4 emission calculated depends on the CH_4:CO_2 ratio assumed, and this was set to a molar ratio of 1:3 (0.33) for untreated slurry in the present study. During on-farm storage of pig slurry in lagoons, Viguria et al. (2015) observed average ratios of 0.21–0.27, Laguë et al. (2005) found a CH_4:CO_2 ratio of 0.65 during summer storage in a slurry tank, and Leytem et al. (2011) reported ratios increasing from 0.09 to 0.74 between spring and fall when measuring emissions from a dairy wastewater pond. In view of this dynamic picture, there is a need for a cost-effective technique to determine CH_4 and CO_2 emissions at high spatial and temporal resolution. The instrument from BPC Instruments mentioned above has been developed to measure emissions of both gases under both aerobic and anaerobic conditions, but was not available for testing as part of the present study.

Grant et al. (2015) reported a positive relationship between CH_4:CO_2 ratio and temperature in slurry that indicates a shift in organic matter degradation pathways and methanogenic activity and growth with increasing storage temperature and, possibly, storage time. Aerobic processes at the manure-air interface will contribute to production of CO_2 (Møller et al., 2004a), and hence storage conditions (surface-to-volume ratio) and presence of a crust (Laguë et al., 2005) may also influence this ratio. The predictions of daily VS loss may improve with a better understanding of the dependence of CH_4:CO_2 ratios on storage conditions and changes in manure composition. On the other hand, sensitivity indices were <0.2 for the CH_4:CO_2 ratio and was not considered a major source of error in the estimation of CH_4 emissions despite the uncertainty discussed above.

Among activity data, slurry temperature is probably more important than VS composition (Chianese et al., 2009; Petersen et al., 2016a). For outside storage, a model was developed and parameterized based on continuous measurements on multiple tanks during a year (Hafner and Mjöfors, 2022), whereas the temperature in slurry pits could not be investigated in this study. Both Chianese et al. (2009) and Petersen et al., 2016a calculated a sensitivity index for selected parameters and found that the sensitivity of CH_4 production rate to lnA was more than ten-fold greater than the sensitivity to slurry temperature. However, if temperature can affect estimates of lnA, as discussed above, then accurate representation of *in-situ* slurry storage temperature may still be critical. In view of the sensitivity of model results to lnA, the analytical procedure for determination of CH_4 production rates used to derive this parameter could be a key source of error, and it should be the subject of further investigation and accurate estimation.

5.5. Application for inventories and mitigation

National inventories of CH_4 emissions from manure management are currently based on annual emission factors, but in a recent refinement of guidelines for emission inventories the monthly mean air temperature was introduced into the calculations (IPCC, 2019). However, for adoption of mitigation measures at farm level a realistic representation of manure management practices is needed. In particular it is important to distinguish between barns and outside storage considering the differences in CH_4 production potential demonstrated in the present study, as well as any difference in storage temperature. With the method described in this paper, $ln\,A'$ for individual sources and livestock categories could be expressed as a mean with confidence limits. The option to statistically analyse effects of management changes or manure treatment on emissions could remove a barrier towards verification of mitigation strategies.

Besides export frequency, a variety of other factors may influence CH_4 emissions from manure management, such as housing design, animal feed rations and the use of bedding material. The *in-vitro* assay provides an opportunity to determine CH_4 production rates in liquid manure at farm level, and with supporting information about manure management etc. it may be possible to identify low- or high-emission production systems.

The experimental results suggested that frequent export of slurry from the barn is a CH_4 mitigation strategy for temperate and cool climates with lower average outside slurry storage temperature. This was also predicted by the model in scenarios with 7 *vs.* 40 d export interval. More frequent export will increase the average degradability of VS in outside storage tanks which could result in higher CH_4 emissions from this source and partly offset the mitigation effect. On the other hand, with daily export the limited growth and adaptation of methanogens may lead to lower emissions also in the outside storage tank, as observed in Sweden; this will likely depend on outside storage temperature. There are treatment technologies such as anaerobic digestion (Maldaner et al., 2018) and slurry acidification (Ma et al., 2022; Lemes et al., 2022) which have been shown to be effective for CH_4 mitigation, but if slurry treatment takes place before or during outside storage, then only CH_4 emissions from this source will be affected, and scenarios for such treatments require a model that can estimate CH_4 emissions from slurry in barns and outside storage facilities separately.

6. Conclusions

A new methodology for estimation of CH_4 emissions from liquid manure management was investigated, in which a key parameter representing the CH_4 production potential of the liquid manure material, $ln\,A'$, was determined experimentally. Significant effects of livestock category, and of manure management stage, indicated that $ln\,A'$ was an important slurry characteristic. Daily export was associated with lower $ln\,A'$ on both cattle and pig farms, and separate tests suggested this may be due to the suppression of growth or adaptation of methanogens during the first several weeks of storage. It implies that, besides degradable VS and temperature, the methanogenic microbial community of manure environments represents an important control of CH_4 emissions, and a key mitigation target. In scenarios with empirical models using experimentally determined $ln\,A'$ values, the predicted differences in CH_4 emission between pig and cattle slurry, and the effects of more frequent export, were consistent with experimental observations, even if both were lower than predicted by the current IPCC methodology. Additional work is needed to validate analytical procedures, but these first results from a transnational monitoring study suggest that a simple empirical model with experimentally determined parameters has potential for assessing CH_4 emissions from individual manure management stages as modified by treatment and management, and climate. Given the diversity of farm operations, such an approach may be needed for accurate accounting of CH_4 emissions and effects of mitigation measures.

CRediT authorship contribution statement

Søren O. Petersen: Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Chun Ma:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Julio E. Hilgert:** Writing – review & editing, Investigation, Formal analysis, Data curation, Conceptualization. **Kristina Mjöfors:** Writing – review & editing, Investigation, Formal analysis, Data curation, Conceptualization. **Paria Sefeedpari:** Writing – review & editing, Investigation, Formal analysis, Data curation, Conceptualization. **Barbara Amon:** Writing – review & editing, Supervision, Conceptualization. **André Aarnink:** Writing – review & editing, Supervision, Conceptualization. **Balázs Francó:** Writing – review & editing, Methodology, Investigation, Conceptualization.

Federico Dragoni: Writing – review & editing, Investigation, Formal analysis, Data curation, Conceptualization. **Karin Groenestein:** Writing – review & editing, Supervision, Investigation, Formal analysis, Conceptualization. **Steen Gyldenkærne:** Writing – review & editing, Methodology, Formal analysis, Conceptualization. **Christiane Herrmann:** Writing – review & editing, Investigation, Formal analysis, Data curation, Conceptualization. **Nicholas J. Hutchings:** Writing – review & editing, Investigation, Formal analysis, Conceptualization. **Ib S. Kristensen:** Writing – review & editing, Investigation, Formal analysis, Conceptualization. **Jing Liu:** Writing – review & editing, Investigation, Data curation, Conceptualization. **Jørgen E. Olesen:** Writing – review & editing, Methodology, Formal analysis, Conceptualization. **Lena Rodhe:** Writing – review & editing, Supervision, Investigation, Formal analysis, Conceptualization.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2024.120233.

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