



Greenhouse gas emissions during storage of manure and digestates: Key role of methane for prediction and mitigation

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ABSTRACT

Treatment of liquid manure and other wastes by anaerobic digestion (AD) adds to renewable energy targets, and it is thus a favorable strategy for greenhouse gas (GHG) mitigation. Both untreated manure and digestates are typically stored for a period in order to recycle nutrients for crop production, and emissions of methane (CH₄), nitrous oxide (N₂O) and ammonia (NH₃) during storage contribute to the overall GHG balance. We determined emissions of all three gases during summer and autumn storage of digestates and untreated manure in pilot-scale experiments. Using these and other data, GHG balances were calculated for treatment, post-treatment storage, and field application. The GHG mitigation potential of AD was demonstrated, but CH₄ emissions during storage dominated the overall GHG balance irrespective of treatment; hence for GHG inventories and mitigation efforts, the correct estimation of this source is critical. Current inventory guidelines from the Intergovernmental Panel on Climate Change (IPCC) estimate CH₄ emissions from manure management based on a simple classification of livestock production systems, volatile solids (VS) excreted, and annual average temperature, and the effects of treatment and management at farm level are therefore not accounted for in any detail. Two empirical models were evaluated, which instead calculate VS degradation and storage temperature with daily time steps; both models were based on concepts presented by Sommer et al. (2004). Parameters for the Arrhenius temperature relationship of CH₄ production, i.e., apparent activation energy, E_a , and pre-exponential factor, A , could be selected, for which cumulative CH₄ emissions calculated with the two models approached observed emissions. However, the magnitude of emissions during a warm period was not well reproduced, and the parameters identified for the two models differed. Sensitivity analyses showed that deviations from observations could not be explained by errors in manure storage temperature. The results thus suggest that CH₄ emissions cannot be predicted from VS and temperature alone, i.e., that the methanogenic potential changes during storage. Determination of parameters for estimation of CH₄ emissions from manure management is discussed with reference to recent literature.

1. Introduction

Animal agriculture is responsible for around 20% of non-CO₂ greenhouse gas (GHG) emissions such as nitrous oxide (N₂O), methane (CH₄) and fluorinated compounds (Owen and Silver, 2015). Globally, manure management contributes about 10% of agricultural CH₄ emissions (Yusuf et al., 2012), but in confined livestock production systems (e.g. dairies and piggeries) with liquid manure management this proportion can exceed 50% depending on climate (Owen and Silver, 2015). Furthermore, Davidson (2009) estimated that management and field

application of livestock manure overall accounts for 44% of total anthropogenic N₂O emissions. These figures emphasize the need to improve estimates of GHG emissions from manure management.

Country-specific practices for manure management significantly influence GHG emissions (Sommer et al., 2009). For example, in Denmark around 80% of livestock manure is managed as slurry (Foged, 2012), most of which is stored until spring for land application as fertiliser due to regulatory provisions to enhance nitrogen use efficiency (Danish Environmental Protection Agency, 2012). Methane emissions during liquid manure storage can thus be significant due to its high

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content of degradable organic matter (volatile solids, VS) and predominantly anaerobic conditions (Umetsu et al., 2005; Sommer et al., 2007).

Several manure treatment technologies have the potential to modify CH₄ emissions (Hou et al., 2015). An increasing proportion of liquid manure is treated by anaerobic digestion (AD) in an effort to capture the CH₄ produced during VS degradation for bioenergy purposes (Nkoa, 2014; Dhamodharan et al., 2015), and this may reduce CH₄ emissions from the manure during post-treatment storage (Amon et al., 2006; Nkoa, 2014). Predicting CH₄ emissions from digestates, however, is complicated by the fact that manure is often co-digested with other wastes, and how this affects emissions during storage depends on the substrate composition (Rodhe et al., 2015), as well as process parameters (Triolo et al., 2011) and storage conditions (Clemens et al., 2006; Wood et al., 2012). The degradation of VS during AD also leads to net nitrogen (N) mineralization (Koirala et al., 2013), which increases the potential for emissions of N₂O and ammonia (NH₃), an indirect source of N₂O, during storage (Misselbrook et al., 2005; Nielsen et al., 2010). Determining the net GHG balance of contrasting treatment and management practices thus requires that emissions of N₂O and NH₃ are also quantified.

National GHG inventories for agriculture are based on guidelines proposed by the Intergovernmental Panel on Climate Change (IPCC, 2006), in which annual CH₄ emissions from manure management are estimated from bioavailable VS in excretal returns and CH₄ conversion factors for a limited number of livestock production systems, and average annual temperature. This approach assumes that a methanogenic potential is present and not a limiting factor. Annual emissions of N₂O and NH₃ are similarly estimated using emission factors (EFs) and total manure N. However, annual EFs are not suited to describe the dynamic changes resulting from treatment and variable retentions time in barns and outside storage, and therefore a prediction model with higher temporal resolution is needed.

In the present study, GHG balances were determined, and two empirical models for prediction of CH₄ emissions evaluated, using data from pilot-scale storage experiments with four different liquid manure materials as case. Two management practices involving AD were represented, as well as untreated cattle and pig slurry for reference. We hypothesized i) that AD treatment would reduce CH₄ emissions during storage compared to untreated slurries; ii) that the overall GHG balance of scenarios with AD would be lower than those of untreated manure; and iii) that CH₄ emissions during storage of liquid manure materials could be predicted from VS composition and storage temperature.

2. Model applications

Sommer et al. (2004) proposed an empirical model to estimate daily CH₄ emissions based on concentrations of total and degradable volatile solids (VS, kg) excreted, storage temperature, and duration of storage. The model distinguished two pools of VS, i.e., an easily degradable fraction (VS_d, kg kg⁻¹ VS) and a “non-degradable” fraction (VS_{nd}, kg kg⁻¹ VS) with a much slower decomposition rate set to 1% of the rate for VS_d:

$$F_{CH_4} = (VS_d + 0.01VS_{nd}) e^{\left[\frac{\ln(A) - E_a}{RT}\right]} \times 24 \quad (1)$$

where F_{CH_4} is CH₄ emission rate (g CH₄ kg⁻¹ VS day⁻¹), A is the pre-exponential factor (g CH₄ kg⁻¹ VS h⁻¹) and E_a the apparent activation energy (J mol⁻¹) of an Arrhenius equation describing the temperature response of CH₄ production, R the gas constant (8.314 J mol⁻¹ K⁻¹), and T is temperature (K). Two modified versions of this model were applied here to evaluate the feasibility of this approach against observations, though only for cattle slurry (treatment CS, cf. Section 3), since parameters for both models were not available with the other materials.

2.1. Integrated farm system model (IFSM)

The sub-model of the Integrated Farm Systems Model (IFSM) for CH₄ emissions from manure management in beef and dairy production systems (Chianese et al., 2008; USDA-ARS, 2009) shares several features with the model of Sommer et al. (2004). The IFSM has been used in several studies to assess the impact of GHG reduction strategies for dairy and beef farms (e.g., Rotz and Hafner, 2011; Dutreuil et al., 2014). The model performance in predicting CH₄ emissions during manure storage has not been extensively verified, but Chianese et al. (2009) did predict annual CH₄ emissions using 25 years of weather data and found that the results were in good agreement with an on-farm monitoring study (Husted, 1994), in which observed daily CH₄ emissions from a cattle slurry storage tank varied between 5 and 35 g CH₄ m⁻³ d⁻¹ during a 12-month monitoring period.

The IFSM model adopts Eq. (1) to calculate CH₄ emissions and uses the Arrhenius parameters proposed by Sommer et al. (2004), i.e., an E_a value of 112.7 and $\ln(A)$ value of 43.33. A modification to the original model has been introduced to calculate the fraction of degradable VS (VS_d) as a function of the VS contained in the manure storage on a given day using Eq. (2):

$$VS_d = \left(VS_{in} \cdot \left(\frac{B_0}{E_{CH_4, pot}} \right) - VS_{loss} \right) / VS_T \quad (2)$$

where VS_T is the total volatile solids contained in the storage tank on a given day (kg), VS_d the degradable VS fraction in the manure (kg kg⁻¹ VS⁻¹), VS_{in} the VS loaded into the storage tank up to the given day (kg), B_0 the achievable emission of CH₄ during anaerobic digestion (0.2 kg CH₄ kg⁻¹ VS, cf. Sommer et al., 2004), $E_{CH_4, pot}$ the potential CH₄ yield of the manure (0.48 kg CH₄ kg⁻¹ VS, cf. Sommer et al., 2004), VS_{loss} the VS lost from the storage up to the given day (kg), which is predicted in IFSM as three times the CH₄ loss from the stored manure. Secondly, VS_T is calculated as a function of the total solids (TS) content in the manure (Eq. (3)):

$$VS_T = M_{manure} \cdot P_{TS} \cdot P_{VS} - VS_{loss} \quad (3)$$

where M_{manure} is the accumulated mass of manure entering the storage tank (kg), P_{TS} the total solids (TS) content in the manure (kg TS kg⁻¹ manure), and P_{VS} the fraction of VS in TS (0.726 kg VS kg⁻¹ TS, cf. Chianese et al., 2008).

Storage temperature is an important driver of CH₄ emissions from liquid manure in both models. The IFSM model uses a 10-day moving average of daily air temperature (Chianese et al., 2009), and this approach was maintained here.

2.2. Modified Sommer model

An alternative adaptation of the model of Sommer et al. (2004) was recently proposed (Petersen et al., 2016a), which introduced new parameters for the Arrhenius temperature response function. An E_a value of 81 kJ mol⁻¹ determined by Elsgaard et al. (2016) was used, and a $\ln(A)$ value of 31.2 g CH₄ kg VS⁻¹ h⁻¹ which was derived from CH₄ production rates in slurry collected from several cattle barns (Petersen et al., 2016a).

In the original model, VS_d was estimated from the composition of fresh excreta, but VS degradation would have already taken place during pre-storage prior to the pilot-scale experiment, and therefore the model was modified to determine VS_d experimentally (cf. section 3.2). It was further assumed that CH₄ and CO₂ were the only significant carbon (C) compounds produced during VS degradation (Spellman and Whiting, 2007), and the CH₄-C/(CH₄-C + CO₂-C) ratio observed in treatment CS, i.e., 0.55, was therefore used to calculate daily VS loss. This was done using Eq. (1), since daily CH₄-C emission could then be converted to daily total C loss by dividing with 0.55; C loss in turn was converted to VS loss assuming a C content in VS of 0.43 kg kg⁻¹

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