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## Biogas plant management decision support – A temperature and time-dependent dynamic methane emission model for digestate storages

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## ABSTRACT

The aim was to develop a temperature and time-dependent model that can calculate the methane production in an anaerobic digester and its subsequent digestate storage tank under realistic and variable conditions. With a daily resolution, the model was applied to a Swedish dairy farm under two different climatic conditions. The most influential parameters were hydraulic retention time and the substrate specific first order reaction rates in the digester, which have a big influence on the residual biogas potential, and hence the potential methane production in the digestate storage. The management of the storage can have a large impact on the emissions from the storage due to its temperature dependence. The model can be used to support plant design and operation of anaerobic digesters and storages, but further research is needed to determine first-order reaction rates and the relationship between the ambient and digestate temperatures at different times of the year.

### 1. Introduction

Anaerobic digestion is a way of increasing the value of organic residues, such as animal manure and crop residues, by converting the organic C in the residues into biogas. The methane (CH<sub>4</sub>) in the biogas acts as an energy carrier, which can be used to substitute fossil fuels. Digestate is also generated as a byproduct from the anaerobic digestion process. The digestate is an organic fertilizer with readily plant-available nutrients, which is often easier to handle and apply to the field than raw manure (Montes et al., 2013; Sajeev et al., 2018).

Biogas production can however have negative impacts on the climate, mainly due to unwanted production of CH<sub>4</sub> during the storage of digestate (Rodhe et al., 2015). Fresh digestate is pumped from the digester to the digestate storage tank to match the influent of substrate to the digester, thus maintaining an even volume in the digester. The digestate storage tank is necessary to temporarily store the digestate before it can be applied to an agricultural field when the soil is not frozen. In central Sweden this can translate to a storage capacity requirement of over 6 months (SJVFS, 2010). The result is a variable residence time for digestate entering the storage throughout the year.

The production of CH<sub>4</sub> in the storage tank depends on the residence time, the residual CH<sub>4</sub> production potential and the temperature of the digestate in the storage tank (Muha et al., 2015). The CH<sub>4</sub> production rate in a storage tank is highly temperature dependent (Baldé et al.,

2016; Maldaner et al., 2018). Studies have shown that CH<sub>4</sub> production in the storage is low when the temperature of the digestate is below 10 °C to 15 °C (Hansen et al., 2006; Maldaner et al., 2018). It can therefore be expected that the timing of digestate removal from the storage tank will impact the total amount of CH<sub>4</sub> lost to the atmosphere as a result of undesired production in the storage tank. This has been confirmed in modelling studies using the anaerobic digestion model no.1 (e.g. Baral et al., 2018; Vergote et al., 2019).

To minimize the generation of unwanted CH<sub>4</sub> emissions it is however necessary to be able to predict which actions will have a significant impact on the production of CH<sub>4</sub> in the storage tank (Baral et al., 2018). The main parameters under the control of the operator of a biogas plant are the management of the digestate, i.e. when to empty the storage tank and spread the digestate, and to some extent the substrate mixture properties and flow rates into the digester. The designer can influence the size and configuration of the biogas plant components based on the expected operating and ambient conditions, effectively determining the upper limits of the hydraulic retention time (HRT) in the digester, which indirectly impacts on the CH<sub>4</sub> production potential of the digestate entering the storage tank (Vergote et al., 2019).

An alternative to a complex dynamic model such as anaerobic digestion model no.1 is to use a simplified approach based on first-order kinetics (Linke, 2006; Linke et al., 2013; Muha et al., 2015). The advantage of this modelling approach is that it can be more practically

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useful under normal operating conditions where all the data needed for complex models are not easily accessible.

The aim of this study was to develop a model based on first-order kinetics, that can readily be incorporated in a tool to estimate the potential CH<sub>4</sub> production in the digestate storage tank. This is related to the CH<sub>4</sub> produced in the anaerobic digester and can be used to identify the most influential parameters that determine the magnitude of the unwanted CH<sub>4</sub> production in the storage tank.

## 2. Material and methods

The model developed in this study simulates a biogas plant consisting of a continuously operated anaerobic digester which empties into a digestate storage tank. The storage tank temporarily holds the digestate, from when it leaves the anaerobic digester up until the time at which it is withdrawn from the storage tank to be applied on a field.

The model is able to handle much of the variability directly under the control of the designer or operator of a biogas plant, both in the anaerobic digester and the digestate storage tank. This includes variability in the temperature of stored digestate as a result of ambient temperatures, timing and size of the volumetric flows to and from the digester and storage tank, as well as variability in the characteristics of the substrate entering the digester. These factors all affect the production of both useful CH<sub>4</sub> in the digester and potential CH<sub>4</sub> emissions in the digestate storage tank.

The model was applied to a generic dairy cattle farm in order to demonstrate its functionality and to perform a sensitivity analysis, using liquid cattle manure as substrate. The sensitivity analysis was carried out to identify the parameters that exert the biggest influence on the generation of CH<sub>4</sub> in the digestate storage predicted by the model under varying conditions.

### 2.1. Theoretical background

The parting point for choosing modelling approach was to make the model depend only on data input that can be readily collected, rather than data that is available at the time of writing. Two previously published functions (Linke, 2006; Linke et al., 2013) that describe the cumulative CH<sub>4</sub> yield ( $y^{CH_4}$ ) in the digester (superscript D, Eq. (1)) and digestate storage tank (superscript S, Eq. (2)) form the basis for the calculations in this study. Both equations are of a single step first order kinetic type (Brulé et al., 2014) and use the volatile substance of the substrate mixture entering the digester ( $VS_{in}^D$ ) as reference value. This enables a straightforward comparison between the cumulative CH<sub>4</sub> yields in the digester and the digestate storage tank ( $y^{D,CH_4}$ ,  $y^{S,CH_4}$ ). Furthermore, it eliminates the need to calculate the VS reduction in the digester.

$$y_{VS_{in}^D}^{D,CH_4}(HRT) = \frac{HRT \cdot k^D \cdot y_{VS_{in}^D}^{max,CH_4}}{HRT \cdot k^D + 1} \quad [LCH_4/kg \ VS_{in}^D] \quad (1)$$

The  $y^{D,CH_4}$  from a single input depend on the maximum potential CH<sub>4</sub> yield ( $y^{max,CH_4}$ ) and first-order reaction rate in the digester ( $k^D$ ) of the VS in the substrate mixture entering the digester as well as its HRT (Eq. (1)). Eq. (1) was first parametrised for thermophilic conditions (Linke, 2006), but later applied to mesophilic conditions with a range of different substrates (Linke et al., 2013). The model functions the same way under both temperature regimes, but the  $k^D$ -values are temperature dependent, and thus significantly lower under mesophilic conditions.

$$y_{VS_{in}^D}^{S,CH_4}(T, t) = y_{VS_{in}^D}^{res,CH_4} \cdot (1 - \exp(-k^S(T) \cdot t)) \quad [LCH_4/kg \ VS_{in}^D] \quad (2)$$

The  $y^{S,CH_4}$  from a single input depend on the residual CH<sub>4</sub> potential ( $y^{res,CH_4}$ ) of the original VS in the substrate mixture entering the digester, as well as the residence time of the digestate in the storage ( $t$ ) and the first order reaction rate in the storage ( $k^S$ ) (Eq. (2)).

The  $y^{res,CH_4}$  represents the fraction of the  $y^{max,CH_4}$  that has not been

converted into CH<sub>4</sub> when leaving the digester. It can be calculated as  $y^{max,CH_4} - y^{D,CH_4}$ . The  $k^S$  depends on the temperature of the digestate in the storage tank ( $T$ ), analogously to the  $k^D$ -value of the digester. The main difference between the digester and the storage tank is that the temperature in the digester is stable, while the temperature in the storage tank varies with the outside temperature, both on a daily and seasonal basis. This makes the calculation of a daily  $k^S$ -value essential to estimate daily contributions to  $y^{S,CH_4}$ . In Linke et al. (2013), the temperature dependence of  $k^S$  was modelled using a single Arrhenius relationship (Randall et al., 1982) for the temperature range 12 °C to 37 °C (Eqs. (B.1) and (B.2)).

In this study all yields and potentials refer to CH<sub>4</sub> and is relative to the  $VS_{in}^D$ . For this reason, and to increase readability, the superscript CH<sub>4</sub> and subscript  $VS_{in}^D$  will be omitted, using  $y^D$ ,  $y^S$ ,  $y^{max}$  and  $y^{res}$  in place of  $y_{VS_{in}^D}^{D,CH_4}$ ,  $y_{VS_{in}^D}^{S,CH_4}$ ,  $y_{VS_{in}^D}^{max,CH_4}$  and  $y_{VS_{in}^D}^{res,CH_4}$ , respectively.

### 2.2. Relative emission potential

An indicator was formulated to compare the potential CH<sub>4</sub> emissions with the benefits from biogas production, as well as to serve as a benchmark when comparing scenarios and different emission sources in the biogas value chain.

In this study it is called the relative emission potential (REP), representing the percentage of CH<sub>4</sub> yield in the digestate storage during one year ( $y^S$ ) relative to the yield in the digester ( $y^D$ ), after correcting for fugitive emissions from the biogas plant itself ( $em^{fug}$ ) ( $y^S / (y^D - em^{fug}) \cdot 100$ ). The  $em^{fug}$  were assumed to be 3.5% of  $y^D$ , based on a number of studies on biogas plant emissions (Flesch et al., 2011; Hrad et al., 2015; Holmgren et al., 2015; Reinelt et al., 2016).

Fugitive emissions taking place after the collection of the biogas produced in the anaerobic digester were not included in  $em^{fug}$  since these may differ depending on the type of end use. Such emissions might occur when upgrading the biogas to vehicle fuel quality (Kvist and Aryal, 2019) or when combusting the biogas in a gas engine (Holmgren et al., 2015).

### 2.3. Development of the time-dependent model

The model developed in this study uses a daily time step in order to accommodate the introduction of information about the substrate and operation of the biogas plant by the user in a spreadsheet tool on a daily basis. Data supplied by the user determines the daily volumes entering the digester ( $V_i^D$ ).

The daily volume leaving the digester and entering the storage tank ( $V_j^S$ ) is given by the volumetric capacity of the anaerobic digester or the user. The digester is assumed to always be operating at full volumetric capacity if  $V_j^S$  is not given by the user.

The user also has to supply the daily volumes leaving the storage tank ( $V_m^F$ ), which are equal to the volumes removed when applying digestate to a field. Subscripts  $i$ ,  $j$  and  $m$  denote the day of the study period ( $t$ ) in which a volume enters the digester or the storage tank, as well as when it is removed from the storage tank, respectively.

Different designations are used to represent time in this model, depending on their reference point. Time relative to the start of the study period are designated by  $t$ , while time relative to the day of introduction of individual daily input into the digester and storage tank ( $V_i^D$  and  $V_j^S$ , respectively) are designated by  $n$ . The relationship between  $n$  and  $t$  can be expressed as  $0 \leq n \leq t - \{i, j\}$ .

Eqs. (1) and (2) are directly applicable to batch digestion experiments where the HRT is identical for the entire input mixture. In a continuous stirred tank reactor (CSRT) a fraction of  $V_i^D$  ( $v_i^D[n]$ ) is however removed with  $V_j^S$  every day. To calculate  $y^D(t)$  under these conditions it is necessary to take into account the size and residence times ( $n$ ) of all remaining  $V_i^D$  fractions, having entered the reactor prior to day  $t$ .

Analogously to the liquid in the digester, the digestate in the storage

tank will be made up of a mixture of all  $V_j^S$  having entered the storage up to  $t$ . Each  $V_j^S$  will have a different  $y_j^{res}$  and residence time in the storage at time  $t$ , affecting their contribution to  $y^S(t)$ .

### 2.3.1. Calculating the volumetric flows

Assuming a completely mixed liquid volume, the size of  $v_i^D[n]$  is proportional to the share of the initial  $V_i^D$  remaining in the digester, relative to the total liquid volume in the digester ( $V^D$ ) (Eq. (3)) at time  $n - 1$ , multiplied by  $V_j^S$  at time  $n$ .

$$v_i^D[n] = V_j^S[n] \cdot \frac{V_i^D[n-1]}{V^D[n-1]} \quad [\text{m}^3/\text{day}] \quad (3)$$

The liquid volume in the digester at any given day,  $t$ , is equal to the balance between the previous day incoming and outgoing volumetric flows ( $V^D[t] = V^D[t-1] + V_i^D[t] - V_j^S[t]$ ). At the same time  $V_j^S$  is the sum of all volumetric fractions leaving the digester each day ( $V_j^S = \sum_{i=0}^{j-1} v_i^D[n]$ ).

The liquid volume in the digestate storage tank ( $V^S$ ) at any given day is calculated the same way as that of the digester, substituting  $V_j^S$  for  $V_i^D$  and  $V_m^F$  for  $V_j^S$  ( $V^S[t] = V^S[t-1] + V_j^S[t] - V_m^F[t]$ ).

In the present model the composition of  $V_m^F$  is not calculated since the fate of the VS remaining after storage is not included within the system boundaries. To calculate the remaining fractions of all  $V_j^S$  after removing digestate, each  $V_j^S$  is reduced proportionally to its volumetric share of the total volume in the storage tank at the time of removing  $V_m^F$ . This assumes that the stored digestate is completely mixed when removed. The implications of this assumption will be addressed in the results and discussion section.

### 2.3.2. Calculating the methane yields

The daily  $VS_{in}^D$  is calculated from information supplied by the user. In the model, information is entered in one of two ways: either as total wet weight (WW) of individual substrates in the input mixture, accompanied by information on their respective dry matter content (DM) and density; or as total volumetric flow of the input mixture ( $V_i^D$ ) together with information on the DM content and density of the entire mixture, as well as information on the size of the DM fraction of each individual substrate in the total mixture. An arbitrary number of up to  $z$  substrates ( $x$ ) can be used in the model.

The user also has to supply the VS share of the DM,  $k^D$ -value and  $y^{max}$  of each individual substrate. In this study the digester is assumed to be operated at 37 °C (mesophilic). The  $k^D$ -values used have to be determined under the same conditions as those in the digester being studied. From the given information the fraction of individual VS inputs, relative to the total substrate mixture VS is calculated ( $p_{x,i}^{VS}$ ). This is used to calculate individual  $y_i^{max}$  (Eq. (4)) and  $k_i^D$ -values (Eq. (5)) for each  $V_i^D$ , as well as the sizes of  $VS_{in,i}^D$  and  $V_i^D$ .

$$y_i^{max} = \sum_{x=1}^z y_x^{max} p_{x,i}^{VS} \quad [\text{LCH}_4/\text{kgVS}_i^D] \quad (4)$$

$$k_i^D = \sum_{x=1}^z k_x p_{x,i}^{VS} \quad [1/\text{day}] \quad (5)$$

#### 2.3.2.1. Methane yield in the digester.

A basic assumption for the calculation of  $y^D(t)$  is that both the liquid and the VS is completely mixed in the digester. This leads to proportional mass and volumetric flows out of the digester ( $m_j \propto V_j^S$ ).

No corrections are made in the model for mass losses from the digester. Thus, the  $V_j^S$  is assumed to be equal to  $V_i^D$  under normal operation. This assumption is feasible when working with dilute substrates, such as liquid cattle manure. In case the VS concentration of the substrate mixture is higher it might be necessary to correct the mass and volumetric flows for mass losses in order to adjust the cumulative  $\text{CH}_4$  yield calculations in the digestate storage ( $y^S(t)$ ).

The cumulative yield from a unit of the daily input mixture ( $y_i^D(n)$ )

is calculated based on Eq. (1), replacing  $n$  for HRT and using the  $y_i^{max}$  and  $k_i^D$ -values corresponding to the  $VS_{in,i}^D$  of day  $i$ .

Due to the proportionality between mass and volumetric flows, the total contribution to the daily yield of  $\text{CH}_4$  in the digester from each daily input mixture ( $y_{i,t}^D$ ) is equal to the contribution from a unit  $VS_{in,i}^D$  multiplied by the amount of  $VS_{in,i}^D$  present in the digester at each time step ( $VS_{in,i}^D(n)$ , Eq. (6)).

$$y_{i,t}^D = (y_i^D(n) - y_i^D(n-1)) \cdot VS_{in,i}^D(n) \quad [\text{LCH}_4/\text{day}] \quad (6)$$

Total cumulative yield in the digester,  $y^D(t)$ , is then calculated as the sum of the total daily yields ( $y_i^D$ ) (Eq. (8)), which in turn is the sum of the contribution from all daily inputs up until day  $t$  (Eq. (7)).

$$y_i^D = \sum_{i=0}^t y_{i,t}^D \quad [\text{LCH}_4/\text{day}] \quad (7)$$

$$y^D(t) = \sum_{m=0}^t y_m^D \quad [\text{LCH}_4] \quad (8)$$

#### 2.3.2.2. Methane yield in the storage tank.

In order to calculate  $y^S(t)$  under realistic conditions it is necessary to take into account the daily variability of the temperature in the storage ( $T_t^S$ ), since it affects both the first order reaction rate  $k^S(T^S)$  in the storage tank and the effective residence time ( $t_j^{eff}$ ) experienced by each  $V_j^S$ . This is done based on the cumulative yield from a unit of the daily digestate input ( $y_j^S$ ) using Eq. (9), which is a modified version of Eq. (2).

$$y_j^S(T_t^S, n) = y_j^{res} \cdot (1 - \exp(-k^S(T_t^S) \cdot t_j^{eff}(n))) \quad [\text{LCH}_4/\text{kgVS}_{in}^D] \quad (9)$$

The  $k^S(T^S)$  in Eq. (9) depends only on the storage temperature and affects all  $V_j^S$  alike. In this study, the same Arrhenius relationship was used as in Linke et al. (2013) (Eqs. (B.1) and (B.2)). The parametrization may affect the result, but this was not further investigated in this study. A challenge when modelling  $y^S$  lies in accurately determining the dependence between  $T_t^S$  and the ambient temperature. The approach used here is further described in Sections 2.4.1 and 2.5.

Calculating  $y_j^S(T_t^S, n)$  requires an iterative approach since  $t_j^{eff}$  is not the actual residence time of a single days input in the storage tank, but an apparent time which depends both on  $k^S(T^S)$  and the fraction of  $y_j^{res}$  remaining at the beginning of each time step. It can be calculated using Eq. (10), which was derived from Eq. (9).

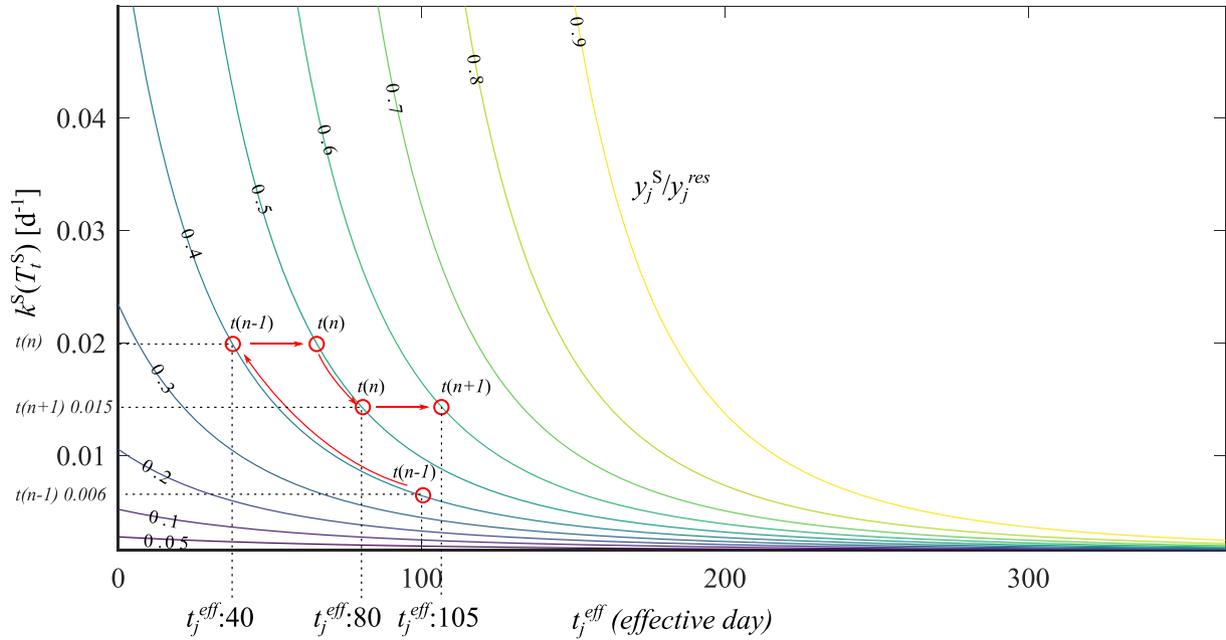
$$t_j^{eff}(n) = 1 - \frac{\ln(1 - y_j^S(n)/y_j^{res})}{-k^S(T_t^S)} \quad (10)$$

The first step to calculate the  $t_j^{eff}$  to use at  $t = n$  in Eq. (9) is to determine the cumulative yield up until the beginning of the time-step ( $y_j^S(n-1)$ ). This is inserted into Eq. (10) together with the  $k^S(T^S)$  of time-step  $t = n$  (Fig. 1). These yields the  $t_j^{eff}(n-1)$  and  $y_j^S(n-1)$ , but under the first-order reaction rate of  $t = n$ . After adding one day to the  $t_j^{eff}(n-1)$  and inserting it into Eq. (9) together with the  $k^S(T^S)$  of time-step  $t$ ,  $y_j^S(n)$  can be calculated. Since  $y_j^S(0) = 0$ ,  $t_j^{eff}(1)$  will always be 1, which will give the  $y_j^S$  of the first time-step.

It is necessary to calculate  $t_j^{eff}(n)$  separately for all  $V_j^S$  since they will have different  $y_j^S(n-1)/y_j^{res}$  at time  $t$ , which affects the shift in  $t_j^{eff}$  between consecutive days (Fig. 1).

The model also includes a function to set the  $y_j^S(T_t^S, n)$  to 0 below a certain temperature threshold. This cut-off temperature ( $T_{cut-off}$ ) was set to 12 °C in all scenarios, based on the range given for the Arrhenius relationship in Linke et al. (2013).

Knowing  $y_j^S(T_t^S, n)$ , the total contribution to the daily yield of  $\text{CH}_4$  in the storage tank from  $V_j^S$  at time  $t$ ,  $y_{j,t}^S$ , the total yield from a single day,  $y_j^S$ , and the total cumulative yield,  $y^S(t)$  (Eqs. (11) to (13)) can be calculated analogously to Eqs. (6) to (8).



**Fig. 1.** The  $t_j^{eff}$  each consecutive day is determined by adding 1 to the  $t_j^{eff}$  calculated from  $y_j^S(n-1)/y_j^{res}$  at the beginning of the day and  $k^S(T_t^S)$  of time-step  $t = n$ . This can be found by following the isoline for  $y_j^S(n-1)/y_j^{res}(t_j^{eff}: 100; k^S(T_t^S): 0.006)$  to the intersection with  $k^S(T_t^S)$  at day  $t$  ( $t_j^{eff}: 40; k^S(T_t^S): 0.02$ ). After adding one day ( $t_j^{eff}(n) = 41$ ) the  $y_j^S(n)/y_j^{res}$  can be calculated. Additional  $CH_4$  produced during day  $t$  will move the  $y_j^S/y_j^{res}$  to the right, perpendicular to the y-axis. This exercise is repeated for each consecutive day of the study period.

$$y_{j,t}^S = (y_j^S(T_t^S, n) - y_j^S(T_{t-1}^S, n-1)) \cdot VS_{in,j}^S(n) \quad [LCH_4/day] \quad (11)$$

$$y_t^S = \sum_{j=0}^t y_{j,t}^S \quad [LCH_4/day] \quad (12)$$

$$y^S(t) = \sum_{m=0}^t y_t^S \quad [LCH_4] \quad (13)$$

#### 2.4. Application to generic farm

The model was applied to a generic dairy farm with two different animal keeping regimes: *grazing* and *indoor*. The same farm was investigated using two different digestate management regimes: *multiple* and *single* application to the field. This resulted in four different scenarios. All four scenarios were tested under two different climates in order to illustrate how the model behaves and improve the understanding of what parameters are most important under different conditions.

The farm was assumed to hold 100 milk cows, 50 heifers and 50 calves. Under the *grazing* regime, all animals were assumed to be kept indoors between the 30th of September and the first of May, while they were allowed to graze for the rest of the year. The milk cows were assumed to be kept outdoors 12 h a day during the grazing period while the heifers were kept outdoors and the calves were kept indoors 24 h a day. Under the *indoor* regime, all animals were assumed to be kept indoors 24 h a day throughout the entire year.

The collection of manure was assumed to be proportional to the production from different animal types, the number of individuals and their length of stay indoors each day of the year (Appendix A).

In the *multiple* application digestate management regimes the digestate was assumed to be removed from the storage tank three times between April and August (12/4, 9/7 and 11/8), leaving approximately 200 m<sup>3</sup> of digestate in the storage tank. In the *single* digestate application management regime the digestate was only removed from the storage tank once a year to make room for new digestate, letting the digestate accumulate in between the single annual applications on the

30th of September.

All four scenarios were run using temperature data from two different climates to calculate the daily temperatures in the digestate storage,  $T_t^S$ . Bollerup in Southern Sweden was used to represent a warm climate, while Piteå in Northern Sweden was used to represent a cold climate. The objective of this was to investigate the influence of the ambient temperature ( $T^{amb}$ ) on the model outcome under otherwise identical conditions, not to quantify  $CH_4$  emissions from a typical dairy farm in the two regions from which the time-series were taken. In a real scenario the length of the growing season would have influenced the grazing periods and timing of application of digestate significantly. That would in turn affect the amount of manure collected and  $CH_4$  produced, both in the digester and storage tank.

In all scenarios the digester was assumed to be operated at total volumetric capacity throughout the year. In the *grazing* scenarios this lead to a variable average HRT in the digester as the amount of collected manure varied throughout the year. In the *indoor* scenarios the HRT was constant since the amount of manure did not vary. The volume of the digester was set so that the average HRT became 30 days at the time of maximum manure production.

All manure was assumed to have a  $y^{max}$  value of 270 L/kgVS<sub>in</sub><sup>D</sup> (Linke et al., 2013) and a  $k^D$ -value of 0.1 d<sup>-1</sup>. The  $k^D$ -value was set in between the 0.02 d<sup>-1</sup> and 0.2 d<sup>-1</sup> that was used in Muha et al. (2015) and Linke et al. (2013), respectively. This gives a  $y^{res}$  of 25% relative to  $y^{max}$  with a HRT of 30 days, calculated using Eq. (1).

In order to have normal operating conditions at the start of the study period, the model included a startup period of 60 days. During this period the digester received influent according to the last 60 days of the study period. This assured that the digester was full and that the composition of the mixture in the digester was representative for a continuously operating biogas plant at  $t = 0$ .

##### 2.4.1. Temperature time-series

An annual daily mean temperature time series was calculated from hourly data for the warm and the cold climate using the Ekholm-Modén formula (Ma and Guttorp, 2012). A period average daily mean temperature time series was calculated from the annual daily means in

2001–2011 to reduce the influence from individual weather events.

The warm climate (Bollerup) time-series (SMHI, 2020a) represented a temperate oceanic climate according to the Köppen-Geiger climate classification (Kottek et al., 2006). It had a period average annual mean temperature of 8.6 °C for 2001–2011. The maximum and minimum period average daily mean during the same period was 19.8 °C and –2.6 °C, respectively.

The cold climate (Piteå) time-series (SMHI, 2020b) represented a subarctic climate and had a period average annual mean temperature of 2.9 °C for 2001–2011. The maximum and minimum period average daily means for the same time period were 17.7 °C and –14.0 °C, respectively.

## 2.5. Digestate temperature calculation

The period average daily mean temperature time series was transformed to a five-day running mean ( $T_t^{\text{amb}}$ ) of the five days directly preceding  $t$  to take into account that the temperature in the digestate does not change instantaneously with the air temperature, but normally exhibits a lag time (Rodhe et al., 2015). This may be due to the heat capacity of the digestate and response time in the biological activity.

The digestate storage tank temperature time series ( $T_t^S$ ) was calculated using the relationship  $T_t^S = 0.75 \cdot T_t^{\text{amb}} + 6$  [°C], based on studies of the dependency between digestate storage and ambient temperatures in similar sized tanks in Denmark (Hansen et al., 2006). It was also assumed that the digestate storage never freeze. Days for which the above formula yielded results below freezing the digestate temperature was set to 0 °C. Since this was below  $T_{\text{cut-off}}$  the assumption did not affect model results.

## 2.6. Sensitivity analysis

The model sensitivity to the cut-off temperature for CH<sub>4</sub> production in the digestate storage tank, different HRTs and  $k^D$ -values in the anaerobic digester was investigated. The aim of the sensitivity analysis was to increase understanding of how the model behaves under different conditions and identify which variables have the biggest influence on the modelling outcome under these conditions. To achieve this two lower cut-off temperatures were used, as well as a range of HRT and  $k^D$ -values.

The  $y^{\text{max}}$  is also an important factor determining the absolute production of biogas and CH<sub>4</sub> emissions from the digestate storage tank. However, increasing or reducing the  $y^{\text{max}}$  without changing the HRT or the  $k^D$ -value will not affect the relative size of the VS reduction in the digester predicted by the model. This assumes that the change in organic loading rate does not have a negative effect on the biological activity in the reactor. The  $y^{\text{res}}$  of the digestate, and hence, CH<sub>4</sub> production will therefore scale linearly and proportionally with the change in  $y^{\text{max}}$ . As a consequence, relative emission calculations are not affected by a change in  $y^{\text{max}}$ . It was therefore not included in the sensitivity analysis.

### 2.6.1. Temperature cut-off

The Arrhenius relationship used in this study was calibrated in Linke et al. (2013) for digestate storage temperatures under limited psychrophilic-mesophilic conditions (12 °C to 37 °C). It is possible that the first-order reaction rate is not accurately described by a single Arrhenius relationship throughout the entire psychrophilic-mesophilic range due to changes in dominant microbial species and limiting biochemical reaction rates (Randall et al., 1982). For this reason the temperature cut-off was set to 12 °C in the base case, leading to no CH<sub>4</sub> generation in the model under this temperature.

The volume of stored digestate is however normally largest early spring, before spreading the digestate to growing crops. This coincides with the time of year when ambient temperatures are rapidly increasing. Whether the spreading takes place before or after the

temperature in the digestate storage tank has surpassed 12 °C can therefore be suspected to have a large impact on the total predicted CH<sub>4</sub> production in the digestate storage tank. Furthermore, Randall et al. (1982) concluded that the substrate degradation rate in highly diverse microbial cultures can be accurately predicted using the Arrhenius relationship over the entire 5 °C to 40 °C temperature range.

To test the influence of the cut-off temperature it was therefore decreased to 8 °C and 4 °C, despite it being outside the calibration range of the Arrhenius relationship used. This was done using both the warm and cold climate temperature profiles.

### 2.6.2. First-order reaction rate in digester and hydraulic retention time

The first order reaction rate of the substrate mixture entering the digester,  $k^D$ , was varied between 0.02 d<sup>-1</sup> and 0.2 d<sup>-1</sup> (Linke et al., 2013; Muha et al., 2015). With a 30 day HRT this resulted in a variation in  $y^{\text{res}}$  between 73% and 14% of  $y^{\text{max}}$ , respectively.

The HRT has an obvious influence on the CH<sub>4</sub> production potential in the storage tank since it directly affects the  $y^{\text{res}}$  of the digestate leaving the anaerobic digester.

Swedish farm-scale biogas plants are on average operated at mesophilic conditions and a HRT of 30 days, where 95% of the substrate is manure (Ahlberg-Eliasson et al., 2017). This can be compared with Germany where the average HRT was 101 days and the substrate mixture consisted of 37% manure (Weiland et al., 2009). However, according to Witt et al. (2011, pp.92), the HRT at biogas plants in Germany mainly operated on manure was in the range of 50–60 days.

The average HRT at the time of maximum manure production was varied between 20 d and 120 d. These two HRTs corresponded to  $y^{\text{res}}$  values of 33% and 8% of  $y^{\text{max}}$ , respectively, using a  $k^D$ -value of 0.1 d<sup>-1</sup>.

## 3. Results and Discussion

### 3.1. Temperatures in digestate storage tank

The  $T_t^S$  calculated from  $T_t^{\text{amb}}$  was significantly lower in the cold climate from the beginning of October to the end of June. This reflected the differences in the 10 year period average daily mean temperature (Fig. 2c and d). The  $T_t^S$  in the warm climate never went below 4.8 °C while it remained below 0 °C in the cold climate during 62 days of the year. The  $T_t^S$  remained above the 12 °C cut-off value for production of CH<sub>4</sub> in the digestate storage tank during 181 and 131 days in the warm and cold climate, respectively. The dates at which  $T_t^S$  reached 12 °C in autumn were fairly close to each other in both climates. In spring, they were however reached the second week of April in the warm climate, which was almost six weeks earlier than in the cold climate.

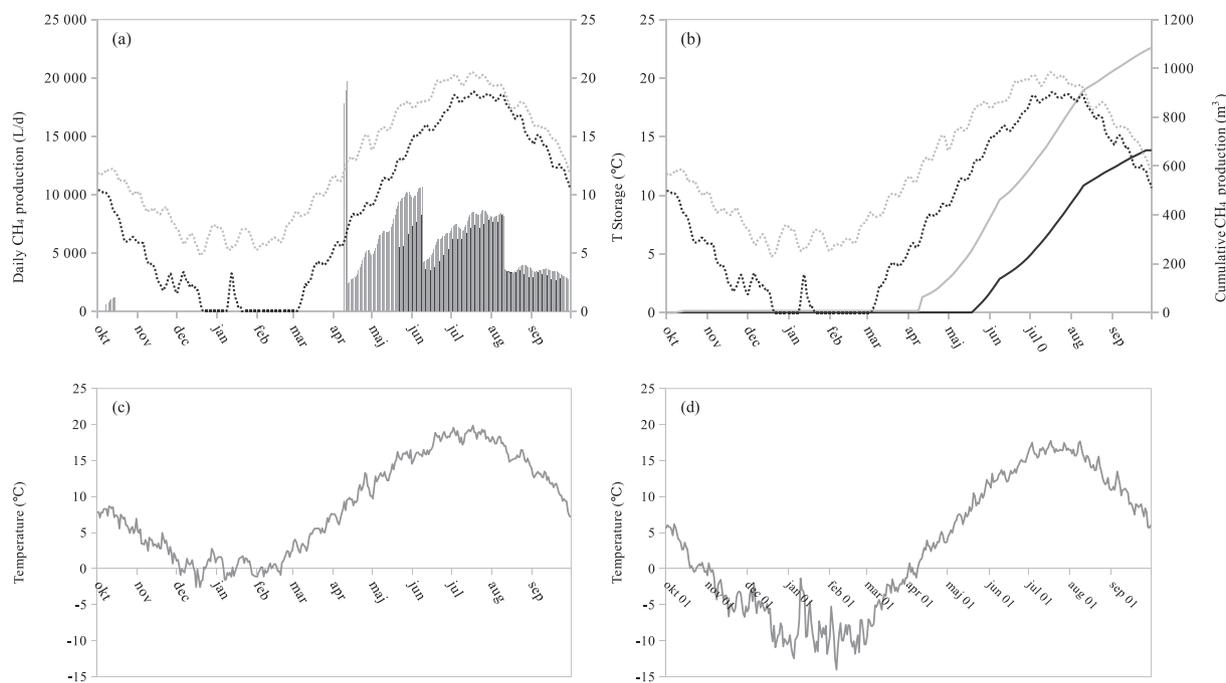
### 3.2. Digester input volumes and residual methane potentials

The daily volumetric flow of substrate into the digester,  $V_i^D$ , was 8.3 m<sup>3</sup>/day with all animals kept indoors 24 h a day. In the grazing scenarios  $V_i^D$  decreased to 3.9 m<sup>3</sup>/day between the first of May and the 30th of September. This increased the average HRT from 30 to 64 days during this period. Since the digester was operated at full capacity throughout the year the flow of substrate from the digester to the digestate storage tank,  $V_j^S$ , was equal to  $V_i^D$ .

The  $y^{\text{res}}$  decreased from 97 to 67 L/kgVS<sub>in</sub><sup>D</sup> during the grazing period (36% and 25% of  $y^{\text{max}}$ ) as a direct consequence of the increased average HRT. These values are similar to those calculated from substrates and digestates on Swedish dairy farms having similar HRTs and substrate composition (Rodhe et al., 2015, 2018).

### 3.3. Methane production and emissions

The multiple-grazing and multiple-indoor scenarios had an REP of 3.6% and 4.8% in the warm climate and 2.2% and 3.2% in the cold climate, respectively. The multiple-grazing scenario in the warm climate



**Fig. 2.** Daily (a, bars) and cumulative (b, solid lines) CH<sub>4</sub> production in the digestate storage of the *multiple-grazing* scenarios, depending on the  $T_i^S$  (dashed lines) in the warm (light grey) and cold (dark grey) climate, which was calculated from the 10-year average mean temperature for the warm (c) and cold climate (d).

could be considered a realistic case of a well managed dairy farm with biogas production in southern Sweden, generating relatively low emissions.

The REP was highly dependent on the digestate management regime. In the *single* application scenarios the model estimated emissions of 21% and 23%, and 15% and 17%, respectively for the *grazing* and *indoor* scenarios in the warm and cold climate, respectively. This was an effect of the storage tank being relatively full during the warmer summer months when daily CH<sub>4</sub> production rates are high (Fig. 3b). This is however not a normal management regime. In case the storage tank would only be emptied once a year it would make more sense to do so in spring (Sommer et al., 2009) when crops can make better use of the nutrient content in the digestate.

The REP was also significantly affected by  $T_i^S$ . It decreased by 39% and 35% in the *multiple* application *grazing* and *indoor* scenarios, respectively, and 28% and 27% in the *single* application scenarios when switching from the warm to the cold climate. This was mainly due to the shorter period of  $T_i^S$  above the 12 °C cut-off value in the cold climate (Fig. 2). Absolute emission reduction was however larger in the *single* application scenarios due to the large amount of CH<sub>4</sub> generated during summer storage in these (Fig. 3).

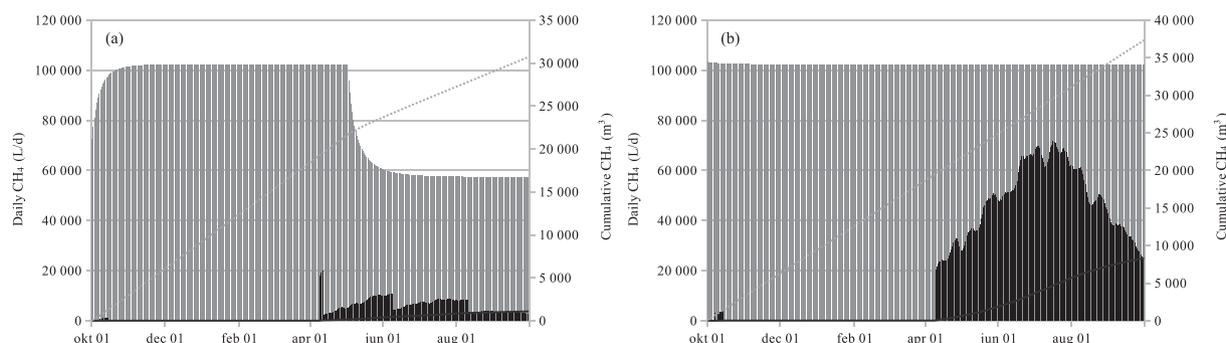
There was a difference between the *grazing* and *indoor* scenarios, but

it was not as important as the digestate management regimes or the digestate temperatures. This can be explained by the fact that an increase in the manure collection rate lead to an almost proportional increase in the production of undesired CH<sub>4</sub> in the digestate storage and useful CH<sub>4</sub> in the digester.

### 3.3.1. Storage emissions in relation to other emission sources

Digestate storage emissions calculated using the present model can be compared to other sources of CH<sub>4</sub> previously reported in the literature, such as biogas plant infrastructure, pressure release valves (PRV), digestate storage, upgrading to vehicle fuel quality and stationary gas engine stack emissions (Fig. 4).

It is not always clear what the reference value of the reported emissions are. The term produced biogas is frequently used, but can be interpreted either as the amount of biogas generated in the digester (including fugitive emissions at the biogas plant), the amount of useful biogas produced at the biogas plant or the amount of biogas after upgrading. In this comparison, values that have a clear reference value other than useful biogas produced at the biogas plant have been recalculated to be comparable to the REP-values, otherwise they have been used as originally reported. A weighted average REP-value was calculated for each type of emission source, taking into account the



**Fig. 3.** The daily ( $y_i^S$ , bars) and cumulative ( $y^S(t)$ , lines) CH<sub>4</sub> production in the digester (light grey, dashed) and digestate storage tank (dark grey, solid) of the *repeat-grazing* (a) and *single-indoor* (b) warm climate scenarios.

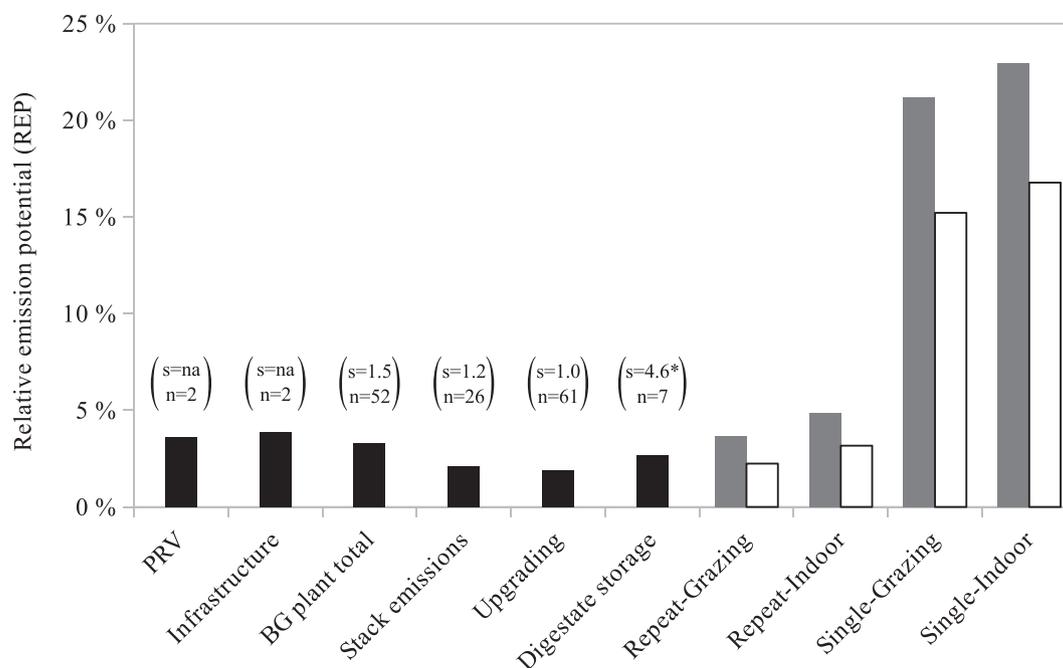


Fig. 4. The REP-values calculated for the four different scenarios in this study, using the warm (grey) and cold (white) climate temperature profiles, compared to other biogas related emission sources (black) reported in the literature (PRV: pressure release valves; BG: Biogas; s: standard deviation; n: number of biogas plants; \*a single plant measurement of 12% had a large influence on s due to the limited number of measurements).

number of biogas plants included in each study, but not the number of individual observations at each plant.

Biogas plant total emissions have been measured and reported in several studies (Flesch et al., 2011; Hrad et al., 2015; Holmgren et al., 2015). It is difficult to distinguish digestate storage emissions from other emission sources with techniques measuring total plant emissions. The studies included here have however attempted to quantify and remove storage emissions from total emissions. Total average CH<sub>4</sub> emissions in the three studies were 3.3%, with the highest and lowest reported value being 5.5% and 1.6%, respectively.

Emissions of 3.6% have been reported from pressure release valves (Reinelt et al., 2016). These make up a part of the total biogas plant emissions. In the same study total biogas plant emissions were reported to be 3.9%.

Emissions of uncombusted CH<sub>4</sub> emissions from stationary gas engines using raw biogas as fuel have been measured in Germany and France, varying between 0.7% and 3.3%, with an average of 2.1% (Holmgren et al., 2015).

Upgrading of biogas can also contribute to significant emissions, although this often takes place downstream from the biogas plant. Values varying between 0.05% and 5.3%, with a weighted average of 1.9% have been reported (Holmgren et al., 2015; Kvist and Aryal, 2019; Ravina et al., 2019). The magnitude of these are related to the type of upgrading technology used (Kvist and Aryal, 2019).

Finally, several studies have reported emissions from different types of digestate storages (Poeschl et al., 2012; Hrad et al., 2015; Holmgren et al., 2015; Baldé et al., 2016). The reported values are highly variable, depending on the type and management of the storage as well as measurement technique applied. Emission values were on average 2.7%, with a maximum and minimum value of 12% and 0.2%. The REP-values from the present model compares fairly well to these values, taking into account that the *single* application scenarios (> 20%) include an extremely unfavourable digestate storage management regime.

It can be seen from the results of this study that the CH<sub>4</sub> emissions from digestate storages can potentially be of similar magnitude or larger than those from the rest of the biogas value chain (Fig. 4). This

emphasizes the importance of being aware of the factors that affect the emissions from digestate storages in order to be able to prevent them.

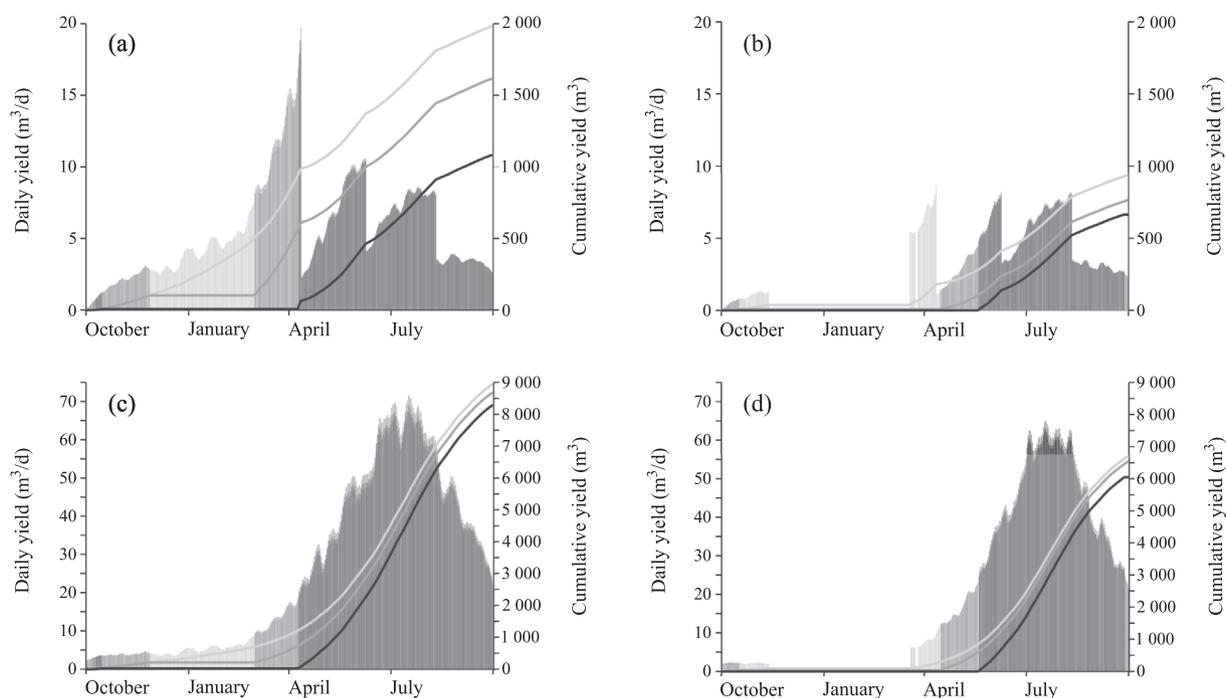
### 3.4. Sensitivity analysis

#### 3.4.1. Temperature cut-off

The REP-value is considerably affected by the cut-off temperature chosen when calculating  $k^S$ , used in Eq. (9). The importance of the cut-off temperature increases along with the number of days that  $T_i^S$  stays between the original cut-off value (12 °C) and the new cut-off value (8 °C or 4 °C). This could be clearly seen in the daily production rates off CH<sub>4</sub> in the digestate storage of the *multiple-grazing* scenarios (Fig. 5). In the warm climate  $T_i^S$  stayed between 12 °C to 8 °C and 8 °C to 4 °C 90 and 94 days, respectively, leading to a 50% and 83% increase in the REP-values. In the cold climate it stayed between the same temperatures 54 and 52 days, leading to a smaller increase of 15% and 41%. The higher relative increase when setting the cut-off temperature to 4 °C instead of 8 °C was in this case explained by the higher amount of digestate present in the storage tank under the 8 °C to 4 °C temperature range compared to 12 °C to 8 °C.

The sensitivity of a specific scenario to the cut-off temperature also depends on the digestate storage management regime. In the *single-indoor* scenario relative emissions increased with 5% and 8% in the warm climate, and 8% and 11% in the cold climate when setting the cut-off temperatures to 8 °C and 4 °C, respectively (Fig. 5). The lower relative increase in the REP-values compared to the *multiple* application scenarios was an effect of a larger share of the total CH<sub>4</sub> production taking place at temperatures above 12 °C. It is important to note that the lower sensitivity to the temperature cut-off in the *single* application scenarios was an indication of higher negative impacts, which in practice can be easily avoided by not having a storage tank full of digestate during the warm period of the year.

Several empirical studies have reported low CH<sub>4</sub> emission rates at temperatures below 10 °C (Baldé et al., 2016; Rodhe et al., 2015; Maldaner et al., 2018). In the study by Baldé et al. (2016), CH<sub>4</sub> emissions were measured continuously during a three-year period in Ontario, Canada. They observed that 92% of the emissions took place at



**Fig. 5.** The effect on daily ( $y_t^S$ , bars) and cumulative ( $y^S(t)$ , lines)  $\text{CH}_4$  production in the storage tank when setting the temperature cut-off level in the model to either 4 °C, 8 °C or 12 °C (light grey, medium grey and dark grey, respectively) in the warm (a and c) and cold (b and d) *multiple-grazing* (a and b) or *single-indoor* (c and d) scenarios, respectively.

temperatures above 10 °C (May–October) and the temperature did not go below 8 °C. The later observation significantly differed from the  $T_t^S$  calculated with the presented model, considering the climatic conditions were similar. The measurements were however done on a storage three times the size of the one modelled in this study, and the storage was never emptied below 3500 m<sup>3</sup>. Both of these factors likely affects the heat balance in the digestate storage. This emphasizes the need to further investigate the dependence between digestate storage and ambient temperatures at different conditions in order to accurately estimate REP-values.

### 3.4.2. Hydraulic retention time and first-order reaction rate

In order to understand how the HRT in the digester or the  $k^D$ -value affects the relative production of  $\text{CH}_4$  in the digestate storage tank it is important to understand how the  $y^{\text{res}}$  of the digestate leaving the digester is affected by the interdependence of these parameters. In the model presented here, the  $y^{\text{res}}$  is a fraction of the  $y^{\text{max}}$ , dependent only on the HRT and  $k^D$ -value of the substrates entering the digester (Eq. (1)). Any factor that increases the VS reduction in the digester reduces the relative impact of  $\text{CH}_4$  in the digestate storage tank through the simultaneous absolute increase of useful  $\text{CH}_4$  production in the digester, and absolute reduction of  $\text{CH}_4$  production in the digestate storage tank.

In a scenario with a given digester temperature, the  $y^{\text{max}}$  and  $k^D$ -values are given by the substrates fed into the digester (Linke, 2006). The HRT is then the sole variable in the model deciding the VS reduction of each substrate in the digester, and consequently the  $y^{\text{res}}$  of the digestate leaving the digester. A higher HRT leads to less VS in the digestate and thus a lower  $y^{\text{res}}$ . This makes the model more sensitive to small increments in HRT at lower HRTs. The effect is clearly illustrated when plotting the change in the REP against different HRTs (Fig. 6). Varying the HRT with  $k^D$  set to 0.02 d<sup>-1</sup> and 0.2 d<sup>-1</sup> was also tested but had a negligible impact on the relative change in REP.

A change in the  $k^D$ -value of the substrate fed to the digester has a larger effect on the REP-values from the model for substrates that require longer time to be degraded, since a lower first-order reaction rate translates into a lower VS reduction in the digester under the same

HRT.

First-order reaction rates are rarely reported in the literature, but can be derived from long term batch experiments (Linke, 2006) or continuously operating digesters as long as information about the HRT,  $y^D$  and  $y^{\text{max}}$  per unit  $\text{VS}_m^D$  are all reported. Unfortunately, this is rarely the case. Furthermore, the specific  $k^D$ -value to use depends on the type of equation used to describe the anaerobic digestion process. The same one used to model the process Eq. (1) has to be applied when deriving the  $k^D$ -values for the substrates.

Ahlberg-Eliasson et al. (2017) collected data from 27 digesters, four of which were operating under similar conditions to the one modelled in this study. From their data the  $y^D$  and HRT could be derived, but  $y^{\text{max}}$  was not reported. Assuming the same  $y^{\text{max}}$  as in this study (270) would lead to  $k^D$ -values in those reactors of 0.04 d<sup>-1</sup> to 0.06 d<sup>-1</sup>. Assuming the same  $k^D$ -value as in this study (0.1 d<sup>-1</sup>) would lead to  $y^{\text{max}}$ -values of 191 L  $\text{kg}_{\text{VS}}^{-1}$  to 235 L  $\text{kg}_{\text{VS}}^{-1}$ . All of these  $y^{\text{max}}$  values are within the range reported by Labatut et al. (2011), indicating that 0.04 d<sup>-1</sup> to 0.1 d<sup>-1</sup> is a feasible range for the  $k^D$ -values of dairy manure to use in this model. Applying a  $k^D$ -value of 0.2 d<sup>-1</sup>, as in Linke et al. (2013) would lead to  $y^{\text{max}}$  of 169 L  $\text{kg}_{\text{VS}}^{-1}$  to 209 L  $\text{kg}_{\text{VS}}^{-1}$ . That is in the lower range, but not unfeasible values for liquid slurry dairy manure.

Given the large impact on the REP-value that a change in  $k^D$  from 0.1 d<sup>-1</sup> to 0.04 d<sup>-1</sup> would have (Fig. 6) more investigation into the reaction rates of different substrates is recommended. This is a general recommendation, not only restricted to this model.

### 3.5. Limitations of the model, uncertainties and future development

The model presented here can include several different substrates and the relation between the substrates can be changed for modelling co-digestion. However, the substrate reaction rates are independent of each other in the model, with no synergistic or antagonistic effects due to variations in the substrate mixture. Furthermore, the model is of a single step first-order kinetic type that does not take into account any rate limiting reactions in the biochemistry throughout the different steps in the anaerobic digestion process (hydrolysis-acidogenesis-

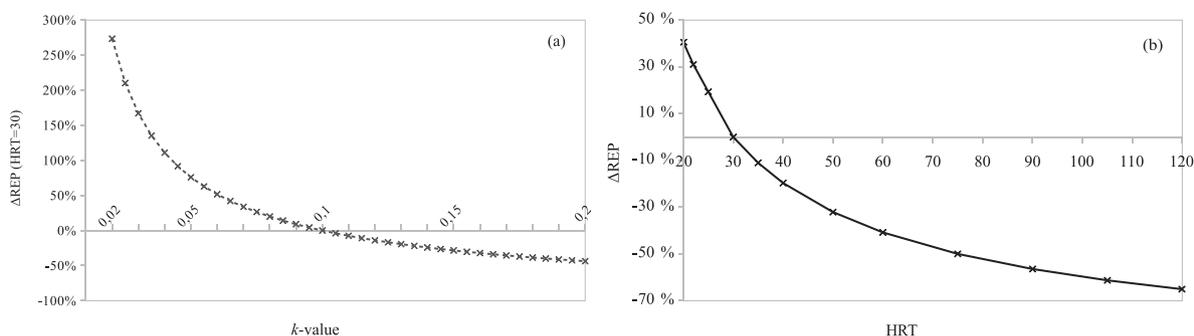


Fig. 6. The change in REP, relative to a  $k^D$ -value of  $0.1 \text{ d}^{-1}$  (a) and an HRT of 30 days (b) when varying the  $k^D$ -value and HRT in the digester between  $0.02 \text{ d}^{-1}$  to  $0.2 \text{ d}^{-1}$  and 20 d to 120 d, respectively.

acetogenesis-methanogenesis) (Brulé et al., 2014). The user should be aware of these limitations, and not use the model to draw conclusions under conditions where such effects may occur.

In the present model  $y_i^S$  co-vary linearly with  $T_i^S$  based on a single Arrhenius relationship. It has been observed in several studies that this assumption holds fairly well in spring, when temperatures are increasing, but that  $y_i^S$  is higher in autumn under the same  $T_i^S$  (Baldé et al., 2016; Maldaner et al., 2018). This hysteresis-like relationship between  $y_i^S$  and  $T_i^S$  is something that requires more research (Kariyapperuma et al., 2018). A likely explanation for this phenomenon is an enrichment of methanogens in the digestate storage tank throughout the year, which leads to a higher  $\text{CH}_4$  production activity during the autumn as compared to the spring at the same temperature. Improving the description of the temperature dependence of  $y_i^S$  under different conditions could improve the accuracy and usefulness of this and similar models to predict and avoid unnecessary  $\text{CH}_4$  emissions.

One limitation of the presented model is the treatment of the digestate storage as completely mixed. In real storage tanks the digestate is rarely stirred and will form a sludge layer at the bottom which is hard to extract by pumping. This is the reason why so much digestate was left in the storage after emptying the storage in the study by Baldé et al. (2016). Stirring the digestate may in fact lead to increased  $\text{CH}_4$  emissions. The shape of the storage tank and removal technique will restrict the amount of digestate that can be removed. This can be controlled by setting the volumes removed to the appropriate values in the model. The sludge layer will however consist of organic material that has considerably longer HRT, and thus a lower  $\text{CH}_4$  forming potential than the rest of the volume in the digestate storage tank. This is not considered in the model when treating the digestate as completely stirred. By removing a proportional value of all digestate in the tank the remaining fraction will have a lower  $y^S/y^{\text{res}}$  than what would be the case if the HRT of the organic material in the sludge and suspended layer was considered separately. That will lead to an overestimation of  $y^S(t)$  from old digestate after repeated emptying events. The size of this

## Appendix A. Input data

Manure production from milk cows (8 t (ECM milk)/yr), heifers ( $\geq 1$  yr) and calves ( $< 1$  yr) was calculated from Table 4 in Börling et al. (2018). A 10% dilution with rainwater was assumed and extracted from the total production. The three animal groups produced  $0.072 \text{ m}^3_{\text{slurry}} \text{ d}^{-1}$ ,  $0.026 \text{ m}^3_{\text{slurry}} \text{ d}^{-1}$  and  $0.015 \text{ m}^3_{\text{slurry}} \text{ d}^{-1}$  having a DM content of  $0.09 \text{ g g}_{\text{ww}}^{-1}$ ,  $0.10 \text{ g g}_{\text{ww}}^{-1}$  and  $0.10 \text{ g g}_{\text{ww}}^{-1}$ , respectively.

## Appendix B. Arrhenius equation

The  $k^S(T_i^S)$  value is the same for all  $V_j^S$  at each time step and can be calculated for an arbitrary temperature between  $12 \text{ }^\circ\text{C}$  to  $37 \text{ }^\circ\text{C}$  using an Arrhenius equation (Eqs. (B.1) and (B.2)), based on Linke et al. (2013).

$$f_T = \left( \frac{k^S(T_2)}{k^S(T_1)} \right)^{\frac{1}{T_2 - T_1}} \quad (\text{B.1})$$

$$k^S(T_i^S) = k^S(T_1) \cdot f_T^{(T_i^S - T_1)} \quad [\text{d}^{-1}] \quad (\text{B.2})$$

overestimation has not been quantified, but should be kept in mind when interpreting the results from the model.

## 4. Conclusions

The model developed and presented in this study can be used by both plant designers and operators to get an integrated understanding of the effects of the design, management and operation of the plant on potential  $\text{CH}_4$  emissions from the digestate storage.

It is important to consider the range of plausible  $y^{\text{max}}$  and  $k^D$ -values of the substrates used during the design of an anaerobic digester as well as the effect that a change in substrates and substrate properties may have on the  $y^{\text{res}}$  from a given reactor in order to reduce  $\text{CH}_4$  emissions from the digestate storage.

## CRedit authorship contribution statement

**Niclas Ericsson:** Conceptualization, Methodology, Investigation, Visualization, Writing - original draft. **Åke Nordberg:** Conceptualization, Funding acquisition, Writing - review & editing. **Maria Berglund:** Writing - review & editing.

## 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.

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In this study the same parameters as in Linke et al. (2013) was used to calculate  $k^S(T_i^S)$ :  $k^S[22] = 0.0063$ ,  $k^S[37] = 0.050$ .

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