Time series measurements and process modelling of methane production and release in two Danish septic tanks

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Abstract-Investigation of two septic tanks in Northern Jutland, Denmark focusing on the release and transformation of carbon was performed over several days with timeseries measurements of CO_2 , H_2S and the pressure difference in the tank with the atmosphere. Estimated air exchange flow from the septic tank using the pressure difference was inaccurate due to the tanks not being sealed properly. H_2S concentrations in the gas phase was < 1 ppm during most of the experiment with peaks of 10-20 ppm which rapidly decreased afterwards indicating H_2S was released due to turbulence caused by the pumping of wastewater out of the septic tanks. CH_4 concentrations in the gas phase varied from 6300-8700 ppm and 900-5200 ppm in the two measured, locations which corresponded to previous measurements of the locations. Headspace CO₂ concentrations varied from 500-4000 ppm and 470-900 ppm in the two tanks respectively. The correlation of CO_2 to CH_4 is not directly proportional indicating that other processes than aceticlastic methanogenesis is occurring in the septic tanks or that fermentation and sulfate reduction is more prevalent in tank A.

I. INTRODUCTION

THE majority of Danish households are connected to a sewer system. However, some 312.000 properties are without connection to the public sewer network [1]. Treatment of the wastewater is therefore limited. Septic tanks are often used for these rural areas where solids settle and are retrieved for further treatment semi-annually. The long residence time combined with the high organic content of the wastewater result in anaerobic conditions. This in turn leads to production of hydrogen sulfide and CH_4 [2]. CH_4 is considered a harmful climate gas and a global warming potential over a 100 year time period in the atmosphere of 28-34 kg CO_2 equivalents per kg CH_4 [3]. Hydrogen sulfide on the other hand oxidizes rapidly, however, it can be a health hazard if one is exposed to high concentrations, corrosive to metals and concrete along with foul odours by the air vents of the septic tank [2]. However, with septic tanks being widely used and evidence for CH_4 leakages from these constructions, there are no time series measurements or long-duration analysis of the production of CH_4 in septic tanks in Denmark. A study in California found that the CH_4 production in septic tanks varied vastly between samples with an estimate of

11 g CH_4 d^{-1} PE^{-1} [4]. With Denmark being part of the UN IPCC program Denmark is required to report their environmental impact through inventory reports. The National Inventory Report 2020 of Danish emissions states the impact from septic tanks is estimated through a study done in 2018 on the capabilities of hybrid filters in septic tanks [5], [6]. The production of CH_4 in septic tanks was estimated to 0.695 $g CH_4 PE^{-1} d^{-1}$, but are also acknowledged as inaccurate and is therefore multiplied by a factor of 10 as to overestimate rather than underestimate the impact[6]. According to the IPCC report, septic tanks produce 1.47 kt of CH_4 out of the total of 2.04 kt of CH_4 from all wastewater treatment sources [6]. Almost 3/4 of the total CH_4 emissions from wastewater treatment are therefore assumed through a single paper with point measurements of two septic tanks in Northern Jutland. This paper will therefore estimate the emissions and processes from the same septic tanks used by Nielsen, A.H et al. 2018 [5] with time series measurements and based on the measurements, setup a conceptual model for estimating the carbon footprint of such systems.

II. SEPTIC TANK DESIGN AND USE

Septic tanks are generally only used as an option for areas where connection to a sewer is not available. Municipalities have the legal authority to demand residents to connect to a sewer, however, the cost of connecting some rural areas to the main sewer networks is not economically or environmentally feasible [7], [8]. Septic tanks' main purpose is to separate solids and liquids. The liquid is typically discharged into the soil whereas the solids will settle inside the tank's main chamber (see figure 3). Material that floats will also be separated by either dividing the tanks into several volumes with a connection below the water level (as illustrated on figure 3) or pumping from below the water level. There are many ways of constructing a septic tank, but for use in Denmark all tanks have to be VA-approved [1]. This separation and retention of solids, which are primarily organic matter, means a build up of sludge will happen over time which has to be pumped from the septic tanks. This sludge will be highly concentrated with a total COD content of upwards of 90000 $q m^{-3}$ [9]. This retention is however not a removal process as there is very little transformation of the organic matter whilst in the septic tanks due to the lack of oxygen. The sludge is instead regularly pumped out of the septic tanks and

handled elsewhere by a separate WWTP. Anaerobic processes is inherently less efficient than anoxic or aerobic processes due to the less favorable electron acceptor being carbon instead of oxygen or nitrogen.

III. PROCESSES IN SEPTIC TANKS

Septic tanks are in practice an accumulating reactor of sewage sludge with effluent water being regularly leaked or pumped out. There is little water movement in these systems causing the sludge to be permanently anaerobic [9].

A. Biological processes

In an environment full of organic matter, bacterial processes are bound to happen. The degradation in the sludge will primarily be anaerobic. The atmosphere in the septic tank will be oxygenated, however, the transfer of oxygen to the wastewater is low. The sludge is highly concentrated in organic matter and with limited oxygen transfer and wastewater in general being low in nitrate content, the sludge is permanently anaerobic [9]. Processes such as sulfate reduction, fermentation and methanogenesis will then occur in the sludge. The biofilm will also always have an anaerobic zone even when the wastewater is aerobic.

1) Sulfate reduction: Sulfate reduction occurs in the biofilm of the sewer walls. In septic tanks, this will also occur on the walls. Sulfate, being a common abundant and harmless ion in drinking water, will be present in wastewater. Both septic tanks are within the same area of catchment for drinking water and according to measurements the sulfate content is an average of $45 \ g \ m^{-3}$. A simplified pathway of sulfate reduction occurs through the usage of organic carbon.

$$SO_4^{2-} + 2CH_2O + 2H^+ \rightarrow 2H_2O + 2CO_2 + H_2S$$
 (1)

The organic matter is any easily biodegradeable organic matter (S_S) . Preferences for the type of S_S utilized for sulfate reduction exists, however for modelling purposes this is ignored.

2) Methanogenesis: CH_4 production is generally not seen in sewers due to sulfate reduction being more favorable and often fully penetrating the biofilm on the sewer wall [2]. Methanogenesis is assumed to only take place in the sludge. There are several pathways for methanogenesis to occur but this project assumes all CH_4 is produced from acetic acid:

$$CH_3COOH \to CH_4 + CO_2$$
 (2)

3) Fermentation: The acetic acid for methanogenesis is produced through fermentation of easily biodegradable organic matter. A variety of fermenting processes can take place in wastewater, however for modelling purposes all fermentation is assumed to be glucose, being the easily biodegradable organic matter and acetic acid being the product:

$$C_6H_{12}O_6 + 2H_{2O} \rightarrow 2CH_3COOH + 2CO_2 + 4H^+$$
 (3)

4) Hydrolysis: The larger organic compounds will be slowly degraded extracellular by enzymes from microorganisms in the wastewater. The large organic compounds cannot be consumed by the microorganisms before hydrolysis has taken place. Hydrolysis takes place in aerobic, anoxic and anaerobic conditions however the process rate is reduced during anaerobic conditions. The hydrolysis process is not selective so the hydrolysis process interacts with whatever is in the solution.

$$1 \ g \ COD \ X_{S2} + H_2O \xrightarrow{hydrolysis} 1 \ g \ COD \ S_F \quad (4)$$

Since wastewater consists of a multitude of different hydrocarbons the process is calculated through mass balance of the COD. The product of hydrolysis are readily fermentable biodegradeable substrate such as monosaccharides. The hydrolysis substrate can be divided into several subdivisions of degrees of hydrolyzability. This project will only include easily hydrolyzable (X_{S1}) and hydrolyzable substrate (X_{S1}) .

B. Monod kinetics

Modelling of these processes is done as monod kinetics. Monod kinetics describe the volumetric transformation rate of the substrate under the assumption that the only limiting factor for growth is substrate.

$$r_{V,X} = \mu_{max} \cdot \frac{S}{(S+K_s)} \cdot X_B \tag{5}$$

Where

- $r_{V,X}$ = the volumetric substrate consumption (*q* COD m⁻³ d⁻¹)
- μ_{max} = Maximum specific growth rate (d^{-1})
- S =Substrate concentration $(g \cdot m^{-3})$
- K_s = Half-saturation constant $(g \cdot m^{-3})$
- X_B = Biomass concentration (g COD $\cdot m^{-3}$)

Non-competitive inhibition will also affect the kinetics, e.g with nitrate in wastewater the anaerobic sulfate reduction will be reduced:

$$\mu'_{max} = \mu_{max} \cdot \frac{K_{S,I}}{K_{S,I} + C_I} \tag{6}$$

Where

- μ'_{max} = Maximum specific growth rate with inhibition(h^{-1})
- $K_{S,I}$ = Half-saturation constant of the inhibitor($g \cdot m^{-3}$)
- C_I = concentration of the inhibitor (g COD $\cdot m^{-3}$)

Following two-film theory, substrate will be limiting to some extent in the biofilm. Methanogenesis and fermentation are 1order kinetics in the sludge but the sulfate reduction is halforder in the biofilm. Order of kinetics is based on penetration. If penetration is full and the half-saturation constant is significantly lower than the substrate concentration at full penetration, then the substrate concentration will be irrelevant even at the deepest part of the biofilm. Thereby the process is only limited by maximum consumption rate and is a 1-order process [2].

C. Considerations

The residence time of the wastewater in the septic tanks depends on the amount of residents living on the property. A tank with a volume of 2 m^3 , which is assumed to be fit for 1-5 people the wastewater, would have a residence time between 18-3.7 days depending on the amount of people utilizing the tank [1], [10]. However, the Danish EPA recommends a residence time of the wastewater being less than 24 hours. With a 2 m^3 tank being used by two people producing 180 L of sludge + 60 L of float annually and the tank being emptied semi-annually, this is not possible [11]. Residence times in this paper are therefore significantly longer due to both septic tanks being 2.5 m^3 and being used by two people, even though these are recommended sizes for the households. The gas-residence time in these systems are also long enough for concentrations of CO_2 and CH_4 rising to many times the atmospheric concentrations. As the system is also with a constant sludge volume, the anaerobic methanogenesis is assumed to be approximately constant during the experiment. Due to equipment being incapable of logging CH_4 measurements, the CO_2 concentration is used as an indicator of the CH_4 concentration.

D. Physical processes

1) Diffusion: Gasses produced in the water phase will diffuse into the gas phase. However, for this model scenario, equilibrium is assumed to occur instantly. This is assumed to be a fair estimate due to the low exchange of gasses into the atmosphere with residence time being above 130 hours. The sludge and water phase is also assumed to be fully mixed in terms of the carbonate system and dissolved CH_4 . Even though the sludge is more active and producing more CO_2 the solubility of CO_2 is low, hence either the CO_2 will separate from the water phase or dissolve into the unsaturated water phase. The equilibrium of carbonate-carbon is also assumed to be instantaneous and pH being unchanged. pH measurements of the wastewater is extracted and measured in laboratory. Determining the solubility of molecules is done through Henry's law:

$$\rho_A = y_A \cdot \rho = H_A \cdot x_a \tag{7}$$

Where the concentration of gas in the water(x_a) and gas phase (y_A) is determined by a compound specific constant, H_A i.e the Henry's constant. In order to calculate the molar mass fraction of the water phase and gas phase, water is assumed to be pure water with a molar concentration of 55.56 $\frac{mol}{L}$ and the gas phase is atmospheric air with majority being nitrogen: 22.4 $\frac{mol}{L}$. When the system volume changes constantly, the sum of mass in the water phase and gas phase will vary. As the production of gasses is calculated from a mass perspective, the sum of the masses are divided into the volumes of water and gas corresponding to the concentration equilibrium:

$$C_{CH4,gas} = \frac{M_{CH4,total} \cdot F_C}{V_{gas}} \tag{8}$$

and water concentration:

$$C_{CH4,water} = \frac{M_{CH4,total} \cdot (1 - F_C)}{V_{water}}$$
(9)

where the F_C is the adjusted mass fraction in the gas phase during equilibrium:

$$F_{C} = \frac{\frac{C_{gas,eq}}{C_{gas,eq} + C_{water,eq}} \cdot V_{gas}}{\frac{C_{water,eq}}{C_{water,eq} + C_{gas,eq}} \cdot V_{water} + \frac{C_{gas,eq}}{C_{gas,eq} + C_{water,eq}} \cdot V_{gas}}$$
(10)

Where the equilibrium concentrations are at equilibrium in equal volumes of water to gas.

Henry's law is only valid during equilibrium which will be affected by the solution in which the gas is dissolved into. Wastewater has a lower solubility of e.g oxygen which is adjusted through a factor of 0.85-0.95 to the solubility concentration [2]. Transfer of oxygen is therefore reduced. Usually the overall oxygen transfer coefficient (K_La) is calculated through empirical equations for sewers [2]. However, in still-standing water in a septic tank these cannot be used. Instead another empirical correlation is used where the overall flux from air to water [12] :

$$J_{O2} = 9.03 \cdot 10^{-11} \ mol \ m^{-2} \ s^{-1} \ pa^{-1} \tag{11}$$

This is however only valid when the oxygen concentration is 0. For CO_2 the gas concentration is calculated in the same way as with CH_4 , however the water concentration is calculated as the fraction of CO_2 in equilibrium with the gas phase with the mol of HCO_3^- and CO_3^{2-} . The concentration of $HCO_3^$ and CO_3^{2-} is determined through acid dissociation constants. For HCO_3^- :

$$C_{F,HCO_3^-} = \frac{10^{-7.5} \cdot K_1}{10^{7.5^2} + K_1 \cdot 10^{-7.5} + K_1 \cdot K_2} = (12)$$

Where:

- + C_{F,HCO_3^-} is the fraction of HCO_3^- in a solution at pH 7.5 (-)
- K_1 is the dissociation constant for CO_2 to HCO_3^- (-)
- K_2 is the dissociation constant for HCO_3^- to CO_3^{2-} (-)

The same method is used for the two other parts of carbonate carbon CO_2 and CO_3^{2-} . H_2CO_3 is ignored as the fraction of carbonate carbon that exists at pH 7.5 is extremely low. In total, the carbonate carbon is calculated through a gas phase concentration of $CO_{2,g}$ to a water concentration of $CO_{2,w}$ multiplied by the amount of carbonate carbon that is in other forms than CO_2 .

IV. METHODS

A. Project site and septic tanks

The septic tanks examined in this project have according to the manufacturer three chambers with the three chambers being equal sizes. Due to the septic tanks being an older model, the documents for the dimensions are no longer obtainable online and the manufacturer was unwilling to describe the layout of the septic tanks. The septic tanks being used in this project are two 3-chambered septic tanks with effluent being pumped out. The two tanks will be referred to as tank A, which is the tank being measured on first, and tank B. Any details on the volumes are partly based on [5] and an email from the CEO of WaterCare claiming the tanks were of minimum water volume of 2 m^3 and a gas volume of 0.5 m^3 along with each compartment of the tank being equal sizes. Tank B has a longer neck which therefore results in a larger gas volume of an additional 0.15 m^3 . Both septic tanks are located in Northern Jutland close to the city of Brovst which is sparsely populated.



Fig. 1. Location of the city of Brovst where the septic tanks are located [13]

The only information given was a minimum constant water



Fig. 2. Conceptual illustration of the septic tank design with estimated volumes. The gas vent illustrated is taped off during the experiment

volume of 2 m^3 and an expected gas volume of 0.5 m^3 . This, along with the regulation of minimum pumped water volume per pumping event of 140-180 L, is then used to assume that the chambers are

$$V_{chamber} = \frac{2m^3 + 0.160m^3}{3 \ chambers} = 0.72m^3 \tag{13}$$

Where a pumping event will trigger when the 3rd chamber is filled up to the volume of $0.72 m^3$ where 160 L of wastewater will be pumped out of the tank restoring the minimum of 2 m^3 . The sludge volume is a part of the 2.16 m^3 and accumulation of sludge will reduce the water volume over time. The accumulation rate is assumed 180 L per year per capita [1].

$$V_{sludge}(t) = 180L \cdot PE^{-1} \cdot y^{-1} \cdot \frac{1}{365} \cdot t_e$$
(14)

where t_e is the amount of days since emptied to the start of the measurements. The two tanks were emptied within two

days of each other with the same amount of residents using the septic tanks. The sludge volume is therefore almost identical as the experiments were carried out continuously. Hence, when the measurements of the first tank were finished the setup was transported straight to the location of the second tank. Each



Fig. 3. Illustration of a septic tank [14]

compartment of the septic tank are assumed to be triangular prisms. This is not true, since the compartments are most likely half-circular, however, without knowing the curve of the upper walls and the bottom surface area, this would have to be estimated. Considering, the structural integrity of the tank would be worsened by a larger curve of the walls that makes a triangular prism most likely a fair estimate. The setup of the measuring equipment on the lid of the tank is illustrated on figure 4.



Fig. 4. Illustration of the setup in the septic tank. A is the CO_2 sensor sucking in air and pumping it to the flask with the CH_4 sensor (B). The H_2S sensor is placed underneath the lid as this needs direct contact with the gas phase in the tank and is hung underneath. The last sensor being the pressure sensor (D) is placed on its own connection measuring both the pressure inside the tank and the ambient pressure

B. Calibration of equipment

1) P-350-D-0.5inch calibration: To measure the pressure difference in air pressure between the septic tank gas and the ambient air a P350-D-0.1-inch Pressure Sensor (± 0.1 inches H_2O) from Pace Scientific is used with an XR5-SE Data Logger to record the measurements. Calibration of pressure sensor according to the flow of air through the smaller vent hole. This will be done in order to eliminate the error of the pressure sensor. Any irregularity in the zeroing of the

pressure sensor will not have an effect if measured flows to corresponding pressure differences. In order to measure the



Fig. 5. Measured pressure difference in pascal with changing flow through a plastic tube

flow of the air going in and out of the septic tank, the pressure sensor (P-350-D-0.5inch) is calibrated for the flow of air using a Brooks Instrument flow meter 0.1-0.5 LPM. The flow is then correlated to the pressure difference through the lid of the septic tank as illustrated on figure 5 The pressure during the experiment remains constant indicating that the flow meter is providing a constant flow. The pressure is averaged over the time of each event and plotted against the input flow as seen on figure 5. The linear correlation is very strong with a R^2



Fig. 6. Averaged pressure difference in pascal correlated with the flow to each event. The values on the graph are the corresponding flow in LPM by using a 0.1-0.5 LPM flow meter

of 0.9935. Slight variations are most likely due to the manual knob to set the flow.

2) LI-820 calibration: The CO_2 -sensor LI-820 is zeroed with a closed loop with a 2L BlueCap flask with 400 mL of water with 1.44 g of 97% NaOH being stirred. NaOH will cause CO_2 to precipitate into the water phase with the NaOH:

$$2NaOH_{(s)} + CO_{2_{(gas)}} \to 2Na^+CO_{3_{(aq)}}^{-2} + H_2O \qquad (15)$$

Excess of NaOH is used in order to ensure all CO_2 is removed from the airphase. Calibration of span was done with 10 & 20 mL of pure CO_2 in a 2 L BlueCap flask. The BlueCap flask was installed with a lid in with the pump sucking air from the flask through the LI-820 and pumping it back into the BlueCap in a closed loop. The inner volume was estimated by dry weight and then filling the flask with tap water. The difference in weight, assuming water is approximately 1 $g \ cm^{-3}$ in density, is therefore the volume. The volume of the entire flask is approximately 2271.7 cm^3 with tubing from the LI-820 back into the flask to be 18.26 cm^3 . 20 mL syringes were filled with pure CO_2 and inserted into the loop (see figure 7). The concentration of CO_2 measured before



Fig. 7. Measured initial concentration of CO_2 before and after addition. In all but one attempt the syinge was filled with 20 mL of CO_2 . In attempt 3 the syringe was only half full

should thereby change by:

$$C_{CO2,after} = \frac{\frac{V_{CO2,pure}}{\overline{V_{CO2,pure} + V_{total}}}}{10^6} + C_{before}$$
(16)

All concentrations are in ppm volume which is why the volume is divided by 10^6 . A volume of CO_2 inserted and measured equivalent volume is calculated and compared (see figure 8). The concentration change varies only slightly from

Inserted volume and measured change



Fig. 8. Volume of inserted CO_2 compared to the measured equivalent change in pure CO_2 volume

the predicted concentration. The minor variations is assumed to be due to the difficulty of extracting the CO_2 from the nozzle in the laboratory to the syringe and thereby diluting with atmospheric air into the syringe. All expected values were higher than measured resulting in a loss of CO_2 gas corresponding to the assumption of leakage.

3) ODA-logger calibration: An ODA-logger is used to measure the gas phase concentration of H_2S . Calibrated at 50 ppm H_2S (+- 5%, GasDetect SDS 102). The H_2S sensor is sensitive to long durations of high concentration

of H_2S and therefore the sensor was calibrated again before measurements of tank B began. A null concentration was also calibrated using pressurized air from the laboratory. The calibration corresponded to previous measurement calibration of the sensor which indicates that the calibration has not drifted during measurements of tank A even though these were repeated numerous times.

V. CARBON CYCLE

The initial carbon concentrations are estimated from an amount of accumulated sludge over time from when the tanks were last emptied and average concentrations of carbon in septic sludge from [9]. The water phase concentration of carbon is assumed through estimates of municipal wastewater concentrations of S_A , SF, XS1, XS2 and X_B [9]. The amount of water is calculated from an annual wastewater equivalent of water being 39400 L y^{-1} [11].

- $C_{XS2,in} = 290g \ COD \ m^{-3}$
- $C_{XB,in} = 120g \ COD \ m^{-3}$
- $C_{SA,in} = 90g \ COD \ m^{-3}$
- $C_{SF,in} = 180g \ COD \ m^{-3}$

All X_{S2} will be added to the sludge phase. Therefore the COD will be further fractioned by a part being COD in the water phase COD_s , and a part being COD in the sludge (COD_{pm}) . The processes consuming the carbon will be CH_4 production, sulfate reduction and growth. sulfate reduction will consume SS and sulfate to produce CO_2 . Fermentation will produce a subcategory of SS - readily biodegradeable acids, S_A , which is produced from the other subcategory of SS - the fermentable substrate S_F . The fermentation will also produce CO_2 and in this project is assumed to be consume on average 1 mole of CO_2 per mole of S_A produced.

VI. VOLUME CHANGES

The system is set up by three volumes. One being the wastewater (V_w) which is fully mixed and the buffer volume for which the wastewater is pumped out of the septic tank. The sludge volume (V_s) is the second volume where only anaerobic processes take place. The third volume being the gas phase (V_g) , which is in constant equilibrium with the water phase and exhausts gas when there is an inlet of wastewater and intake of atmospheric air when the pump initiates. As the system is open to the atmosphere, any changes in volume inside the tank will cause either a venting of gasses or an inlet of atmospheric air. If wastewater is pumped in:

$$V_{w,in} = V_{gas,out} \tag{17}$$

The volume of gas vented out is calculated through pressure measurements in the gas vent. The difference in pressure will correspond to a flow of air as calibrated in the laboratory:

$$Q_p = 0.0339 \cdot P_{dif}$$
 (18)

where Q_p is the flow through the vent hole in liters per minute (LPM) and P is the pressure in pascal. All pressure measurements equaling to a flow above +0.2 LPM are for modelling purposes assumed to be an influx event of wastewater from the residence and will be used as an increase of V_w and will subsequently result in a reduction of V_g and is therefore venting out gas. Each event of the LPM being above +0.2 will be multiplied with a factor of K_F . This factor is determined through the assumption of inlet wastewater being 39400 L per year per capita in a residence over the timeperiod of measuring.

$$V_{w,in} = \frac{Q_d \cdot t_{measured}}{N_{Q_{p,>+0.2}}} \tag{19}$$

where $N_{Q_{p,>+0.2}}$ is sum of events where Q_p is larger than 0.2 LPM and $t_{measured}$ is the time from start of measurement till the end of the experiment in days, Q_d is the amount of water inlet per day according to the Danish EPA per household capita [11]. Wastewater is pumped out of the tank when the wastewater level reaches $2.16m^3$ as previously calculated. This initiates the pumping of 160 L of wastewater out of the tank. Events of where wastewater is pumped out of the tank are compared to the events of where H_2S is measurable. H_2S is oxidized before reaching the lid of the septic tank where the H_2S -sensor was located. In both tanks this effect of H_2S concentration increase also correlated with a reduction of CO_2 concentration. This is assumed to be turbulence releasing H_2S causing an increase of H_2S and an afterwards dilution of the gas in the septic tank due to atmospheric air entering the septic tank.

A. Gas-phase concentrations

The gas in the tank is assumed to be fully mixed as with the wastewater. The exchange of CH_4 and CO_2 is assumed to be based on Henry's law using the Henry's constant of H_{CO_2} = 1640 atm and H_{CH_4} = 40200. The solubility of CH_4 is significantly lower than CO_2 .

B. Measurement setup

The measurements of the gas inside the septic tank was done through the lid of the two tanks. Both lids had four airtight push-in fittings installed. Two of the connections were used for in- and outlet of the gas measured with the LI-820 and the CH_4 sensor. The CO_2 sensor LI-820 has an integrated air-pump which the inlet was connected to a 500 mL Erlenmeyer flask in which the CH_4 sensor was connected to. CH_4 is measured using a RS PRO RS GD-36 combustible gas detector. The CH_4 measurements are monitored manually as the device can not log the data points itself. The exhausted gas is then pumped back into the tank in order to prevent removal of gas in the septic tank artificially. Another fitting was used for the pressure sensor in order to not be affected by the air pump. The fourth hole is then the only open vent of the lid which was left open as this was the hole the pressure sensor had been calibrated to the flow of.

VII. MODELLING OF THE SYSTEM

To be able to estimate the CH_4 production of the two septic tanks, a process model of transformation of organic components is set up. The process modelling will be done in respect to COD transformation and consumption which in turn produces CO_2 . These CO_2 producing processes will be used to correlate measurements of the system. This is due to the CH_4 sensor being unable to log data continuously. These point measurements of CH_4 are assumed to be fairly well correlated to the CO_2 and are extrapolated from these to a rate of CH_4 production. Several of the anaerobic processes use inverse correlations with oxygen and nitrate. These parts of the model have been removed as oxygen and nitrate is assumed to be 0 in the sludge and biofilm. A conceptual illustration of the model can be seen as Appendix I.

The rate of the fermentation will be modeled as such [2]:

$$R_{ferm} = q_{ferm} \frac{S_F}{K_{ferm} + S_F} \cdot \alpha^{T-20} \tag{20}$$

Where

- q_{ferm} is the fermentation rate constant (d^{-1})
- K_{ferm} is the saturation constant for the fermentation (g COD m^{-3})
- ϵ is the relative efficiency constant for hydrolysis in the biofilm biomass
- α is the temperature coefficient (-)
- T is the temperature of the wastewater

The sulfate reduction rate will be modelled as such:

$$R_{SO4,red} = a \cdot \sqrt{(S_F + S_A + X_{S_1})} \frac{S_{SO4}}{S_{SO4} + K_{SO_4}} \cdot A/V) \cdot \alpha^{T-2}$$
(21)

Where

- S_{SO4} is the concentration of sulfate
- K_{SO4} is the half saturation constant of sulfate

Transformation of the fractions X_{S1} and X_{S2} to S_F through hydrolysis will happen both in the water phase and the sludge. Anaerobic hydrolysis of COD is 14% of aerobic hydrolysis [15].

Hydrolysis of slow hydrolyzable substrate:

$$R_{hydr,XS2} = k_{XS2} \cdot \omega \cdot \frac{\frac{XS2}{X_B}}{K_{XS2} + \frac{XS2}{X_B}} \cdot X_B \cdot \alpha_W^{(T-20)}$$
(22)

Where

- k_{XS2} is the rate constant of hydrolysis for the hydrolyzable substrate (s^{-1})
- ω being the reduction factor (0.14)
- XS2 is the concentration of hydrolyzable substrate (g COD m^{-3})
- K_{XS2} is the half-saturation constant, fraction 2 (g COD m^{-3})

Hydrolysis of rapidly hydrolyzable substrate:

$$R_{hydr,XS1} = k_{h1} \cdot \omega \cdot \frac{\frac{X_{S1}}{X_B}}{K_{x1} + \frac{X_{S1}}{X_B}} \cdot X_B \cdot \alpha_W^{(T-20)}$$
(23)

Where

- k_{h1} is the rate constant for the easily hydrolyzable substrate (s^{-1})
- X_{S1} is the concentration of easily hydrolyzable substrate (g COD m^{-3})

• K_{x1} is the half-saturation constant, easily hydrolyzable substrate (g COD m^{-3})

Decay is assumed to be proportional to the maximum specific growth rate [2].

$$R_d = 0.1 \cdot \mu_{H,O2} \cdot X_B \tag{24}$$

Where

• $\mu_{H,O2}$ is the maximum specific growth rate

Methanogenesis:

$$R_{meth} = \mu_{max,CH4} \cdot \frac{S_A}{S_A + K_{CH4}} \cdot X_{B,meth}$$
(25)

TABLE I MASS BALANCES IN THE SLUDGE. S_A and S_F are the same for sludge and waterphase



TABLE II MASS BALANCES OF THE PROCESSES IN THE TANK. S_F and S_A are Assumed to be fully mixed in the tank.

Water phase				
X _B	$((X_{B_{t-1}} + Growth_{t-1} \cdot t - R_{decay,t-1} \cdot t)$			
	$V_{w,t-1} + \frac{V_{w,in}}{1000} \cdot X_{B,in} \cdot (V_{w,t-1} + \frac{V_{w,in}}{1000})^{-1}$			
S_F	-			
S_A	-			
X_{S1}	$X_{S1_{t-1}} - H_{fast_{t-1}} \cdot t$			
X_{S2}	$((X_{S2_{t-1}} - R_{hydr, XS2_{t-1}} \cdot t) \cdot V_{w,t-1} + \frac{V_{w,in}}{1000}$			
	$\cdot COD_{XS2,in}) \cdot (V_{w,t-1} + \frac{V_{w,in}}{1000})^{-1}$			
SO ₄	$((SO_{4_{t-1}} - SO_{4, redt-1} \cdot t) \cdot V_{w, t-1} + \frac{V_{w, in}}{1000} \cdot 45)$			
	$(V_{w,t-1} + \frac{V_{w,in}}{1000})^{-1}$			

The CO_2 producing steps are fermentation, methanogenesis and sulfate reduction. Growth is assumed to be zero and the biomass is constant. The biofilm is assumed to be in a steady-state and the sludge volume is constant in the model scenario. In order to determine the molar production of CO_2 the stochiometry from section III is used. A gram of H_2S will result in:

$$\frac{2 \ mol \ CO_2}{34 \ g \ mol^{-1} \ H_2 S} = \frac{0.05882 \ mol \ CO_2}{g \ H_2 S}$$
(26)

For 1 g COD of CH_4 through methanogenesis:

$$\frac{1 \ mol \ CO_2}{mol \ acetate \cdot 59 \ g \ mol^{-1} \cdot 1.08 \ g \ COD \ g \ acetate} = 0.01569 \ mol \ CO_2 \ g \ COD^{-1} \ acetate$$
(27)

And for fermentation:

$$\frac{2 \mod CO_2}{\mod glucose \cdot 180 \ g \mod^{-1} \cdot 1.0667 \ g \ COD \ g \ glucose} = 0.0104 \ mol \ CO_2 \ g \ COD^{-1} \ glucose$$
(28)

In summary the transport of organic matter is modelled as illustrated in figure 16.

VIII. RESULTS & DISCUSSION

Measurements of tank A is slightly shorter than the measurements of tank B. Several attempts at measuring in tank A failed due to a design flaw in which tubing would become loose. This wasn't identified and fixed until the measurements of tank B began. During three attempts of measuring with four day measuring periods on tank A, finally almost two consecutive days of data were intact. Measurements of the wastewater show a fair correlation between the H_2S presence in the atmosphere at sudden periods of time, see figure 9. Along with the spikes of H_2S , the concentration of CO_2 in the atmosphere can be seen spiking and dropping in the gas phase as well. This strongly suggests turbulent conditions within an interval that could be caused by pumping of the wastewater. The CO_2 changes in the tank are not as visible in tank A as in tank B. However the concentration of CO_2 in tank B is also vastly lower than in tank A. Since both tanks are operated by the same amount of people and being of the same design except having a longer neck, the gas phase might not be as well mixed as in tank A. Layering of the gas phase could be the cause for lower measured concentrations of CO_2 and CH_4 .

The concentration at the start of measuring in tank A is also



Fig. 9. Conceptual illustration of the septic tank design

vastly lower due to gas venting when installing the setup. For this reason model scenarios ignore the first measurements until the first CH_4 measurement the day after installation was logged. The amount of wastewater put into the system in both tank A and B with the experiment duration of 42 and 60 hours respectively and with the pumping volume assumed to be 160 L per event, a minimum of 3 events in tank B and twice in tank A should occur. As seen on figure 9, the hydrogen sulfide concentration spikes three times even in the measurements of tank A. This is however under the assumption that the tank was just emptied when started.

A. Airflow measurements

The septic tanks were however not sealed causing the calibration of the pressure sensor to flowrate to be highly inaccurate. Model scenarios with flowrates of gas equal to the calibration was unable to reduce concentration of CO_2 and CH_4 - even with zero production of the two - to any meaningful level. To counter this, a factor of 5 was used on the calibration of exchanged air volume in order to fit the CH_4 model concentration with the measurements. However this is very unlikely to be true and the model highly relies on the exchange of gasses from atmospheric air and the gas phase of the septic tanks. The parameters being calibrated in order to fit the model will therefore only be the $\mu_{max,ferm}$ and $\mu_{max,meth}$. The airflow during the night and morning hours is significantly lower than daytime values corresponding to water usage in residential households being less active during these hours [9].

As seen on figure 10, the majority of measurements are



Fig. 10. Measured pressure differences between the septic tank and the outside atmosphere calculated to flow volumes and multiplied with the model timestep into air-exchange volumes per timestep. The laboratory model of flow to pressure was edited to a factor of 5 times larger flow

positive exchanged air volumes. This indicates that there is a constant output of low amounts of gas from the lid. The amount of measurements with negative values indicate that the lid might be the outlet but not the inlet of gas and that the tank, which was not sealed properly, had a constant flow through at least the neck of the tank. This could be due to the power cord for the pump in tank A which went under the lid down to the pump and made it impossible to close the tank properly. In tank B, the neck had a metal pipe through it in order to keep the lid from being opened before this rod was removed. This was neither tight which caused a rather large gap in the neck. The hole and the open lid could have been a vent for the tank where the inlet of atmospheric air is entering and the purposefully drilled holes in the lid acted as a passage of air out.

1) Modelled CO_2 production compared to the measurements: As mentioned earlier, the model of CO_2 concentrations are very poorly correlated with the measurements. The exhaust of CO_2 instantly resets the concentration to equilibrium causing almost no changes in CO_2 . This is due to the larger fraction of carbonate-carbon being stored in the water phase which due to modelling constraints are at constant equilibrium. The reactions seen in the measurements will therefore never be present in the model. The interaction between the equilibrium of carbonate carbon and the diffusion into the gas phase affect the system too much to be able to do such a simplified model. The problem is also present in the model for tank B as



Fig. 11. CO_2 concentration in the gas phase measured compared to the modelled in tank A

well. The exchange of air needed in order for the CO_2 to be correlated with the measurements would be unrealistically high.

As seen on figure 11, there are two pumping events, however



Fig. 12. CO_2 concentration in the gas phase measured compared to the modelled in tank B

comparing to figure 9 the H_2S concentration peaks three times. This could be due to the assumption of the initial volume of the septic tank or the inlet volume in the modelling scenario not being the actual volume.

2) CO_2 correlation to CH_4 production: Correlating the production of CH_4 with the concentration of CO_2 is most likely inherently inaccurate without a rate of exchange between $CO_{2,g} \iff CO_{2,aq}$ and the accompanying carbonate system. In both tanks, the pH was measured after the experiment at pH 7.5 for both tanks which corresponds to the pH of the groundwater in the area [16]. When modelling the scenario of constant equilibrium the gas phase will be instantly stabilized resulting in almost no changes in the gas phase concentration of CO_2 . The model scenario is most likely overestimating the sum change of carbonate-carbon exhausted from the system but showing a higher concentration of $CO_{2,q}$ due to the instantaneous equilibrium with the larger mass of carbonatecarbon in the water phase - which most likely in the septic tank has not reached equilibrium throughout the atmosphere inside the tank. When trying to use the $CO_{2,q}$ to correlate the volume of exhausted gas in order to achieve a better fit the CH_4 is then completely exhausted due to the very low solubility of the gas compared to the sum of carbonate-carbon dissolved. With pH 7.5, the majority of carbonate carbon exists as the ion HCO_3^- which results in a much larger capacity of storage in the water phase of carbonate-carbon. The rate of equilibrium changes and gas to water phase interaction therefore needs a better model in order to be possible to correlate $CO_{2,q}$ concentrations to a CH_4 production in such a system. A better control of the exhaust volume is also needed in order to circumvent the modelling issue of calibrating the output of gasses.

B. CH₄ concentration measurements

 CH_4 concentrations of the gas phase was manually monitored with only a few point measurements. Even without having storage for logging, the sensor is able to save maximum measured concentration of 8700 ppm in tank A and 5200 ppm in tank B in between readings. As seen on figure 13, the concentrations of CH_4 are reduced during the measurement period for both experiments with only one measurement in tank B suddenly increasing to 4800 ppm. The first measurement is measured the day after the setup has been installed. This is due to the venting of the gas inside the tank when installing the setup as the lids of the septic tanks had to be replaced. Tank A measured around 5300 ppm right after the installation and tank B measured less than 1000 ppm. The installations effect on the CH_4 concentration is therefore assumed to be accounted for. The CH_4 concentrations in the gas phase are however higher than previous measurements of the tanks where the concentrations were approximated to 5000 ppm CH_4 [5].

The changes in the CH_4 concentration are fairly well correlated with the reduction of CO_2 , indicating the change is due to air exhaust and dilution. This can also be identified on figure 10 where the air exchange is significantly larger during the day of the 21st of April. Besides the fairly well correlated CO_2 concentration variations there is a larger difference in between the two tanks and their CO_2 to CH_4 concentrations. The only slightly above atmospheric CO_2 concentration in tank B shows that the correlation with CO_2 and CH_4 is not



Fig. 13. CH_4 concentrations measured during the two experiments in the two tanks with CO_2 concentrations in both tanks.

directly proportional even in septic tanks with same sludge age and load. This could be due to methanogenesis not being produced with acetic acid but with hydrogen:

$$CO_2 + 4H_2 \to CH_4 + 2H_2O \tag{29}$$

In municipal anaerobic systems, aceticlastic methanogenesis accounts for more than 2/3 of the methane production whereof the hydrogen process accounts for approximately 1/3 of the CH_4 production [17]. The difference in CO_2 to CH_4 ratio could also be due to fermentation and sulfate reduction being more prevalent in tank A than tank B.

C. Model of processes

Modelling of the processes in the septic tanks are based on a lot of assumptions of initial literature values or assumptions of the processes and components.

Initial values				
Time step	t	30	s	
Maximum specific growth rate	μ_{HO2}	$9.26 \cdot 10^{-5}$	s ⁻¹	
Temperature coefficient	α_W	1.07	-	
Rate contant, fraction 1	k_{h1}	$8.10 \cdot 10^{-6}$	s-1	
Rate contant, fraction 2	k_{h2}	$8.10 \cdot 10^{-6}$	s ⁻¹	
Half saturation constant, fraction 1	K_{x1}	1.5	$rac{g\ COD}{g\ COD}$	
Half saturation constant, fraction 2	K_{x2}	0.5	$\frac{g \ COD}{g \ COD}$	
Pumped water volume	V_{pumped}	160	L	
Maximum specific growth rate for methanogenesis	$\mu_{max_{methane}}$	$2.31 \cdot 10^{-6}$	s^{-1}	
Maximum specific growth rate for fermentation	$\mu_{max_{ferment}}$	$1.16 \cdot 10^{-5}$	s^{-1}	
Half saturation constant for fermentation	$KS_{ferment}$	0.03	$\frac{g \ COD}{g \ COD}$	
Half saturation constant for methanogenesis	KS_{meth}	0.1	$\frac{g \ COD}{g \ COD}$	
Heterotrophic biomass	X_b	50	$g \ COD \ m^{-3}$	
Easily degradable substrate	S_{f}	20	$g \ COD \ m^{-3}$	
Very easily degradable substrate	S_A	10	$g \ COD \ m^{-3}$	
Fast hydrolyzable substrate	X_{S1}	100	$g \ COD \ m^{-3}$	
Slowly hydrolyzable substrate	X_{S2}	60000	$g \ COD \ m^{-3}$	
Aceticlastic methanogenetic biomass	$X_{methanogenesis}$	10	$g \ COD \ m^{-3}$	
Input values				
Very easily degradable	$S_{A,in}$	90	$g \ \overline{COD} \ m^{-3}$	
Easily degradable	$S_{F,in}$	180	$g COD m^{-3}$	
Slowly degradable	$X_{S2,in}$	290	$g COD m^{-3}$	
Heterotrophic biomass	$X_{B,in}$	120	$g COD m^{-3}$	

Due to the exchange volume of the tank being unknown or at least not what was calibrated in the laboratory, the rate of processes is impossible to estimate. However, with the CO_2 concentration in the model being far above the measured air concentration, the air exchange either be enormously larger in which the CH_4 concentration will go to zero or the methane production is equally enormous in which the emission of CH_4 becomes unrealistically larger. The equilibrium of carbon and the rate of the exchange between air and water phase must be heavily affecting the $CO_{2,g}$ concentration in these systems. As seen on figure 14, the concentration of CH_4 correlates



Fig. 14. CH_4 concentrations measured along with the modelled concentration of CH_4 in the system and in the gas phase.

fairly well with the measurements of CH_4 . However, the sum molar concentration of CH_4 in the system remains fairly constant. The changes in gas concentration is mainly due to the pumping events where fresh air is sucked into the tank and wastewater is pumped out. The dilution effect of the gas along with the subsequent exhaust when filling the 0.16 m^3 void in the third compartment are the processes of reducing the concentration where the production of CH_4 in the sludge is acting oppositely. With the concentration during the measurement period being fairly constant, the production of CH_4 is approximately equal to the two counteracting forces. In tank B, see figure 15, the sum of CH_4 is vastly lower, however tank B also has a larger gas volume due to the larger neck of the tank. In tank B, the molar sum of CH_4 is fairly constant throughout the measurement period. With the longer neck of the tank this could also be due to a poorer mixing of the gas phase in the septic tank.

It is however also visible that the model cannot take all



Fig. 15. CH_4 concentrations measured during the two experiments in the two tanks with CO_2 concentrations in both tanks.

the variations into account in the system with the CH_4 concentration suddenly rising to 4500 ppm and afterwards dropping 900 ppm. Most likely, this is due to the gas phase not being fully mixed. A concentration gradient of CH_4 will almost certainly be present, however the magnitude of such gradient was not investigated. The sudden increases and subsequent drop in concentration indicate turbulence causing gas from near the water phase to move towards the lid where

the sensors are attached. However, the CO_2 measurements of the tank does not correlate to this happening. Considering that the H_2S measurements during peak events were similar in concentration between the two tanks, the gas phase must be equally mixed or proportionally poorer mixed and with a larger sulfide production (see figure 9).

The constant CH_4 concentration in the gas phase indicates that the methanogenesis process occurs constantly during the measurement period. The rate of methanogenesis in the model is limited due to the concentration of S_A . This is under the assumption of constant methanogenic biomass and with a $\mu_{max,meth} = 0.2 \ d^{-1}$. Assuming the exchange volume of gas is correct, the biomass could be lower but the $\mu_{max,meth}$ would then be proportionally smaller. The estimate for production however heavily relies on the exchanged volume of air. With the proposed correction of the calibration, the sum of produced CH_4 is 0.52 g and 0.17 g $d^{-1} PE^{-1}$. With the previous estimate in the IPCC report being a release of 0.695 g d^{-1} PE^{-1} there is further evidence that the CH_4 production in septic tanks is an emission factor, however not as large as previously estimated with reports of upwards of 11 g d^{-1} PE^{-1} [4]. The release of 0.695 g d^{-1} PE^{-1} is however also - a release. Not a production of CH_4 . Due to the system varying in gas/water volumes a release of CH_4 based on measurements of gas phase concentrations will be affected by pumping wastewater out and gas in due to the gas being diluted. Using the same method as done in [5] for a release during the measurement period the release of CH_4 is 1.44 g $d^{-1} PE^{-1}$ for tank A and 0.284 g $d^{-1} PE^{-1}$ for tank B. With the previous mentioned climate change effect of $28CO_{2,eq}$ the total environmental impact annually is 1.77-5.28 kg $CO_{2,eq}$ PE^{-1} y^{-1} or approximately 1-3 kt $CO_{2,eq}$ or 0.037-0.11 kt CH_4 if 10% of the Danish population uses a septic tank as according to the IPCC report [6]. The IPCC report estimates a total of 1.47 kt CH_4 for the year 2018. The estimate from IPCC is larger due to a multiplication factor of 10 to the emission of CH_4 due the assumption of the leakage from older tanks. This factor has not been factored into the estimate in this project. The variations can be caused by temperatures. The methanogenic bacteria are mesophilic and needs higher temperatures than what the Danish climate usually has [9]. The measurements were taken during days with air temperatures of 6-10 degrees Celsius and previous Danish study on the same tanks were done in January which usually has lower temperatures than what was measured in this study. The pH of the sludge is also affecting the methanogenic bacteria and are severely hindered at less than pH 5.5 [9]. The pH of the sludge is affected by fermentation due to acid production. However, the acids produced from fermentation is used by the methanogens to produce methane and thereby removing acid and increasing the pH. Undisturbed sediments in sewers have a higher pH than newly deposited sediments [2]. Sludge age has a positive correlation with the CH_4 production. The methanogenic biomass in wastewater is low and when the tanks are emptied, growth is necessary in order for production. With the yield of methanogenesis being low the growth is limited [9]. Emptying of septic tanks more frequently will reduce the emission of CH_4 due to a production reduction.

IX. CONCLUSIONS

• Gas phase concentrations of CH_4 and CO_2 are closely correlated, however with large variations among tanks with same sludge age and water volume. CH_4 concentrations varied from 6300-8700 ppm and 900-5200 ppm in the two measured locations.

The corresponding production rates of CH_4 equaled to 0.52 g and 0.17 g $d^{-1} PE^{-1}$ with 1.44 g $d^{-1} PE^{-1}$ and 0.284 g $d^{-1} PE^{-1}$ released into the atmosphere during the experiments.

- The environmental impact of the release equates to 1.77-5.28 kg $CO_{2,eq} PE^{-1} y^{-1}$ or a national emission production of 1-3 kt $CO_{2,eq}$ in septic tanks. CO_2 concentrations in the septic tanks varied from 500-4000 ppm and 470-900 ppm in the two tanks. However, the lower concentrations were mostly due to installation of measurement equipment causing the ventilation of the atmosphere inside the tank.
- H_2S concentrations in the gas phase is correlated with sudden increases of CO_2 indicating that the turbulence caused by pumping of wastewater has a large impact on the gas phase even of low soluble gasses such as H_2S .
- Modelling of the CO₂ concentration in the gas phase was unsuccessful presumably due to the carbonate system being slow to reach equilibrium with the gas phase.

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APPENDIX APPENDIX I



Fig. 16. Illustration of the processes in the septic tank and the components in the tank

APPENDIX APPENDIX II

Attached excel document