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Process Performance of the Anaerobic Digestion of Sewage Sludge for Increasing Operation Temperatures

Master's thesis in Environmental Engineering

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REPORT NO. BOMX02-17-91

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CHALMERS UNIVERSITY OF TECHNOLOGY
Göteborg, Sweden 2017

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Summary

This thesis was written to evaluate the potential and challenges of a thermophilic anaerobic digestion of wastewater treatment sludge from the Rya treatment plant in Göteborg, Sweden. Six pilot-scale reactors (8 L) were loaded on a daily basis with an average organic loading rate of 2.5 g_{VS}/L d. After 71 days at 35°C the temperature of three reactors (T1-T3) was increased to 55°C using different rates and loadings. The other three reactors served as reference and were operated at 35°C. The ratio of volatile fatty acids (VFA) and total alkalinity was used to decide on the loading and to evaluate the performance. Other performance parameters analysed were the amount of biogas produced and its methane (CH₄) content. Additionally, the effect of the temperature increase on the dewaterability of the digested sludge was evaluated using a pressure filtration test.

Depending on the conversion strategy high VFA concentrations could be observed that were however buffered well. Interrupting the loading of the reactor at 43°C kept the VFA concentrations at low levels but resulted in an unstable behaviour after the conversion. The operation of the thermophilic reactors proved to be more sensitive to changes in loading than that of the mesophilic ones.

The gas volume in the thermophilic reactors was lower than in the mesophilic ones independently of the conversion strategy and recovered only slowly. The CH₄ content of the biogas however could be restored after 2-3 weeks.

After the transition, the VFA/alkalinity ratio was 2-3 times higher in the thermophilic reactor than in the mesophilic reactors and the pH showed approximately 0.5 higher values.

The weight of the cake resulting from the pressure filtration test was identified as the most reliable parameter for a characterization of the dewaterability. It dropped during the transition phase but stabilized at a lower level with time. Using a polymer with a lower charge density could reduce the total suspended solids (TSS) found in the reject and increase the weight of the cake by 30% as compared to the standard polymer.

As best conversion strategy for the full-scale plant a fast temperature increase with loading rates monitored by the VFA/alkalinity was identified. As a critical factor, the loading frequency was determined whose effect on the performance should be assessed before further planning is done.

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Abbreviations

AD	Anaerobic digestion, anaerobic digester
COD	Chemical oxygen demand
CV	Coefficient of variation
EPA	Environmental protection agency
EPS	Extracellular polymeric substances
FAN	Free ammonia nitrogen
LOQ	Limit of quantification
MO	Microorganisms
NL	Normal litre
OLR	Organic loading rate
SRT	Solids retention time
TAN	Total ammonia nitrogen
TS	Total solids
TSS	Total suspended solids
VFA	Volatile fatty acid(s)
VS	Volatile solids
WW	Wastewater
WWT	Wastewater treatment
WWTP	Wastewater treatment plant

1 Introduction

The aim of wastewater treatment (WWT) is to remove substances that are harmful to the environment. This results in mainly two types of sludges:

1. Primary sludge which contains solids that are settled in the primary clarifier
2. Secondary sludge which consists mainly of the excess biomass and extracellular polymeric substances (EPS) from the biological treatment

Those sludges differ in several characteristics e.g., total solids (higher in primary sludge), volatile solids (lower in primary sludge), energy content (higher in primary sludge) or the organic acids (lower in primary sludge) (Coelho and Miguel 2012; Gebreeyessus and Jenicek 2016). The secondary sludge has a biogas potential of only 50-70% of that of the primary sludge (Nielsen and Petersen 2000).

If no treatment of the sludge is done it contains pathogens, is malodorous and consists to a large fraction of water. It is therefore difficult to handle, requires large storing and treatment facilities and can pose a risk to human health and the environment. Therefore, the Rya wastewater treatment plant (WWTP) treats the sludge like many other plants with the following steps:

1. Mixing of primary and secondary sludge in primary clarifier for co-settling
2. Thickening
3. Anaerobic digestion (AD)
4. Dewatering

All steps aim at a volume reduction. The 3rd step does this by converting parts of the organic matter into biogas. This results in a reduction of odours as well as conversion of complex nitrogen into compounds that allow for a better use as a fertilizer (Cartmell and Boyd 2003).

The share of the treatment and disposal costs of sludge on the total operational cost is high. At Gryaab, the sludge treatment accounts for almost 50% of the operational costs (I'Ons 2017). One option for the disposal of sewage sludge is spreading it on agricultural land which also helps to close the nutrient cycle especially for phosphorus. This is currently the case for 25% of the sludge in Sweden (Naturvårdsverkets 2013). Spreading WWT sludge on land that is used for food production can pose as risk to human health. The EU directive 86/278/EEC on the use of sewage sludge in agriculture (Directive 1986) was implemented by the Swedish Environmental Protection Agency (EPA) into the decree SNFS 1994: 2. Even though the latter regulates that the sludge has to be treated before the use on agricultural land, both regulations do not set limits concerning contagious pathogens. To fill in this gap in Sweden most sludge that is used in agriculture is nowadays certified voluntarily according to REVAQ (Revaq 2017). This certification system was initialized in 2008 by the wastewater industry and demands that the sludge is sanitized and controlled for salmonella in addition to stabilization. On a policy level, the Swedish government decided that at least 60% of the phosphorous in sewage sludge must be returned to productive land of which half must be arable land. In 2013 the EPA conducted a study where it identified an excessive risk for infection with the current regulation and proposed more stringent hygiene requirements (Naturvårdsverkets 2013). At that point in time the EPA announced new provision on hygiene treatment for the year 2019. To reach the recycling goals for phosphorous it can therefore be expected that in the next years a change in legislation will regulate the sanitation. Gryaab wants to increase the share of sludge that is used in agriculture from currently 45% to 80% (I'Ons 2017) and therefore wants to be prepared for changes in legislation.

One method to sanitize the sludge is the thermophilic AD that was identified by Gryaab as the most easily implemented method since the sludge is already treated anaerobically at mesophilic temperatures. According to the EPA the sludge has to be treated in batches with a minimum of 55°C for at least 8 hours to be considered as sanitized (Naturvårdsverkets 2013).

A study conducted by the EU concluded that 4 hours at that temperature are sufficient to get nearly pathogen-free sludge (Carrington 2001). To ensure a complete sanitation the reactor must be operated in a batch-mode.

While all studies conducted on the effect of a temperature increase in AD proved a better pathogen removal, the effect on the reactor performance and operation is less clear. While some studies showed a higher biogas production (up to +108%) (Boušková et al. 2005; Iranpour et al. 2002; Cavinato et al. 2013; Zabranska et al. 2002; Hidaka et al. 2016; Ghasimi et al. 2015; Nielsen and Petersen 2000; Gebreeyessus and Jenicek 2016) others did not find a difference for this parameter compared to mesophilic operation (Palatsi et al. 2009; Tezel et al. 2014; de la Rubia et al. 2005). There are also contradictory results on the methane content of the biogas (higher than in mesophilic: Ziemińska-Buczyńska (2014) and Tezel et al. (2014), similar: Cavinato et al. (2013), de la Rubia et al. (2005) and Zabranska et al. (2002)). The recovery times that are reported after a temperature increase range from 14 days (Iranpour et al. 2002) to 70 days (Boušková et al. 2005) and are often not clearly reported.

Seeding the mesophilic reactor with thermophilic sludge is no option for Gyaab due to the size of the plant and the lack of thermophilic digesters nearby. The rate of stabilization and the performance after the temperature increase will therefore depend on the community of anaerobic microorganisms (MO) present in the mesophilic sludge and other sludge characteristics. A study using this sludge is therefore necessary since only very little is known on key organisms and many studies do not clearly state the operational conditions and feed characteristics. The dewatering behaviour is a key factor at Gyaab and is assessed only in two of the studies mentioned above which come to different conclusions (better than in mesophilic: Nielsen and Petersen (2000), worse: Tezel et al (2014)). For the planning of the temperature conversion it is also essential for Gyaab to know what behaviour of the reactors is to be expected during the temperature conversion and if the faster kinetics at thermophilic temperatures mentioned by Tezel et al. (2014) result in a bigger treatment capacity at the same retention time. All this lead to the decision to conduct a pilot scale experiment in whose context this thesis is placed.

2 Background

2.1 Anaerobic Digestion of Wastewater Sludges

Anaerobic digestion has the aim to “reduce pathogens, reduce biomass quantity [...], and produce usable gas as a by-product” (Vesilind 2003). It is a complex process that involves many microorganisms (MO) and can be divided into the following steps whereby each step uses the products of the previous step (Mc Graw-Hill 2010; Baier 2015; Vesilind 2003, 1974; Cartmell and Boyd 2003):

1. Hydrolysis: break-down of complex organics polymers (e.g., carbohydrates) into soluble monomers (e.g., glucose, amino acids) that are available for other MO
2. Acidogenesis/Fermentation: break-down to volatile fatty acids (VFA), H_2 and CO_2
3. Acetogenesis: conversion of higher organic acids to acetic acid
4. Methanogenesis: conversion to CH_4 and CO_2 by two pathways:
 - a. From CO_2 and H_2 (30%)
 - b. Directly from acetic acid (70%)

The rate limiting step for complex substrates as they can be found in WWTP is the hydrolysis (Vesilind 2003; Baier 2015; Ghasimi et al. 2015; Gebreeyessus and Jenicek 2016; Mc Graw-Hill 2010).

While the acidogens and acetogens are not very sensitive to changes in the environmental conditions the methanogens are highly disturbed by variations in pH, alkalinity, ammonia or temperature (Gebreeyessus and Jenicek 2016, 2016; Vesilind 1974). Since methanogens also have a doubling time of 5-16 days (Baier 2015) there is a danger of wash-out if the reactors

are not operated carefully (Ziemińska-Buczyńska et al. (2014) found ca. 3.5 d of doubling time).

The optimum pH for the acidogens is between 5.5 and 6.5 while for methanogens it is between 6.5 and 8.2 (Mao et al. 2015). Since the methanogens are the more sensitive organisms and the acidogenesis is fast the pH should stay between 6.6 and 7.4 to support the methanogenesis (Lahav and Morgan 2004; Vesilind 2003). In step two of the AD buffering capacity is consumed but is recovered in step four such that the pH stays constant if the steps are well balanced.

The temperature has an influence on the digestion rate especially for hydrolysis and methanogenesis and should therefore be kept as stable as possible (Vesilind 2003). Reactors are usually either operated at mesophilic (around 35°C) or thermophilic (around 55°C) temperature (Metcalf & Eddy 2014). The operation temperature determines which MO are present since most of the MO have an optimum growth rate in one of the two ranges. However, some MO are insensitive to temperatures between 35 and 55°C (Ziemińska-Buczyńska et al. 2014). Therefore, at 55°C real thermophilic as well as thermotolerant MO can be found (Zabranska et al. 2002).

2.2 Start-up Strategies for Thermophilic Anaerobic Digestion

When the temperature increase is done without seeding the reactor with thermophilic sludge the thermotolerant MO need time to adapt their enzymes and other cell components to the higher temperatures (Zabranska et al. 2002). The thermophilic organisms that are present in small numbers in the mesophilic sludge must outcompete the mesophilic ones by their higher growth rate at thermophilic temperatures. This results in a transition phase during which the reactor performance decreases. Attention must be paid to the conversion strategy since it determines the final performance of the reactor and must ensure that the reactor will not break down irreversibly.

In literature, the following two factors are identified as critical:

- Rate of temperature increase:
 - Directly from 35°C to 55°C
 - Stepwise increase
 - Slow continuous increase
- Loading during the transition phase:
 - Continuous
 - Interrupted/reduced

The rate of temperature increase determines which MO are selected depending on their growth rates. The loading is critical because the acidogenesis is not very temperature sensitive while the methanogenesis is disturbed by temperature changes. This can result in an accumulation of VFA and a drop of pH to values outside the range at which AD can take place (Labatut and Gooch 2012). Different authors (e.g., (Palatsi et al. 2009; de la Rubia et al. 2005; Boušková et al. 2005) identified 43°C as the critical temperature at which the performance of the reactor decreases dramatically. Tab. 14 (appendix) gives an overview on the strategies used in some studies that look at the temperature conversion. Additional information on the operational conditions is provided for comparison with the experiments of this thesis. The different authors chose various combinations of temperature increase and loading.

2.3 Dewatering of Stabilized Wastewater Sludges

The digested sludge still contains more than 95% of water which can be partly removed by dewatering. During dewatering a filtration (cake formation) and consolidation (cake compression) takes place (Christensen et al. 2015). The resulting cake behaves like a semi-solid or solid (Mc Graw-Hill 2010). There are different methods for sludge dewatering. At Gyaab, screw presses are used since 2017.

According to Vesilind (1974) the water contained in the sludge can be classified into free, floc, capillary and particle water. The removal of this water becomes increasingly difficult in this order. Colin and Gazbar (1995) therefore divide the water into free water, bound water removable by moderate mechanical strain, bound water removable by maximal strain and bound water that is not removable mechanically. When a certain pressing pressure is reached, increasing the pressure only results in a compression of the cake but will not remove more water (Sveegaard, Keiding, and Christensen 2012). The dewatering behaviour can however be enhanced by conditioning. At Gryaab this is done by using a cationic polyelectrolyte (polymer). This polymer neutralizes the negatively charged sludge particles and allows for a coagulation of the solids so that the absorbed water is released. Cationic polymers are available with different charge densities (defined as percent of units along the polymer chain that are charged). The polymer dose has to be carefully chosen such that an overdosing and an excess of positively charged particles are avoided and large flocs can be formed (Metcalf & Eddy 2014).

There are many sludge properties that influence how well the conditioning works and what polymer dose is required (Mc Graw-Hill 2010; Christensen et al. 2015; Vesilind 2003):

- Particle size: the smaller the worse dewaterability
- Suspended solids concentration: the higher the worse dewaterability
- Sludge source: the more biologically digested and stored the worse dewaterability (more hydration for long storing times)
- Microorganisms: the more extracellular polymeric substances (EPS) formed the worse dewaterability
- pH and alkalinity: the higher the worse dewaterability
- Polyvalent ions (e.g., Ca^{2+}): the lower the worse dewaterability
- Viscosity: the higher the worse dewaterability
- Handling/transport: the more shear stresses the worse dewaterability

2.4 The Rya Wastewater Treatment Plant

The Rya Wastewater Treatment Plant (WWTP) is operated by Gryaab AB and treats the wastewater (WW) of the Gothenburg region in the west of Sweden. Almost 800 000 PE (based on BOD_7) are connected and on average 3.9 m³/s of WW are treated (Gryaab 2016). There is no strong industrial influence on WW quality. The WW undergoes a conventional highly loaded activated sludge treatment and post-nitrification and -denitrification.

The sludge is treated by gravity belt thickening and then digested anaerobically. The digestion takes place in three reactors that are operated in series whereby the last reactor is not heated. The produced biogas (2016: 73 GWh) is upgraded and used as vehicle fuel. The sludge is further dewatered after polymer conditioning (170 t_{polymer}/a). A REVAQ certification is done for 45% the rest is co-composted to produce soil-products (I'Ons 2017). The sanitation of the REVAQ sludge is done by long term storage (min. 6 month) outside the WWTP. 3% of the biogas comes from co-digestion of grease and food waste. Some operational key parameters can be found in Tab. 2.

2.5 Preliminary Work on Dewatering at Gryaab

The dewaterability of the digested sludge is an important parameter that Gryaab wants to assess during and after the temperature increase. Yulang (2017) therefore performed experiments to identify a reliable method to measure this parameter. No repeatable results could be achieved by the capillary suction time (CST) that is a commonly used method. Hence, Yulang (2017) developed a pressure filtration test. He evaluated the effect of different polymer doses, pressing times and pressures, and defined standard values for these factors that yield

good results. All dewatering experiments performed in the context of this thesis are based on this method.

3 Scope and Objective

The aim of this thesis is to broaden the foundations for an objective decision as to whether and how an increase of the operation temperature of the AD to thermophilic conditions is feasible for the specific sludge that can be found at the Rya plant.

The reactor performance is therefore evaluated

- during the temperature conversion
- after reactor stabilization

The performance is characterized based on the:

- transition time until stabilization is reached
- gas production and methane content during transition and after stabilization
- dewaterability of the digested sludge during transition and after stabilization
- process stability to changes in loading after stabilization

Different conversion strategies based on various rates of temperature increase and loadings during the transition are evaluated to identify the best way of implementing the temperature change.

The thesis also identifies gaps in knowledge that must be closed before the planning can be done.

This shall give Gryaab an indication on the

- extent to which the operation of the AD can be continued during the transition phase (storage capacity for sludge needed, usability of biogas etc.)
- long term capacity needs

and provide rough fundamentals for a more detailed planning.

4 Material and Methods

4.1 Experimental Setup and Feed Characteristics

Six pilot-scale reactors (Belach Biotechnik AB) were available for the experiments and operated for 71 days at 35°C before the start of the thermophilic experiments. Three served as reference and remained at 35°C (M1-M3). The other three (T1-T3) were converted to 55°C according to the strategies displayed in Tab. 1 which use different combinations of the key factors identified in chapter 2.2.

Tab. 1: Strategies for the conversion of reactors from mesophilic to thermophilic operation temperatures.

	T1	T2	T3
Temp. increase	Immediately	1°C/d	1°C/d
Loading	Interrupted immediately	Interrupted at 55°C	Interrupted at 43°C

To determine when the loading can be re-started the ratio of VFA/alkalinity was chosen. Therefore, samples were titrated in the morning and the loading of the corresponding day was chosen based on these results. When the ratio dropped below 0.3 the loading was increased carefully such that the ratio remained below 0.3 (details see Tab. 18, appendix). The scheme couldn't be followed to 100% because during the weekend measurements were not possible. If no loading took place the volume taken out to measure VFA and alkalinity (15 mL) was replaced by digested sludge from M1.

Tab. 2 characterized the pilot-scale reactors in comparison to the full-scale plant. The loading took place each day around 10 o'clock except on Sundays. The volume to be loaded was first withdrawn from the reactors and used for analysis. Then, the corresponding amount of raw sludge was loaded to the reactor. Two loading schemes were applied and specified in Tab. 3. In both schemes the same volume is loaded per week but in different portions over the week. It should be noticed that the loading was based on volume and not volatile solids (VS). This results in fluctuations of the organic loading rate (OLR) between days that have the same volumetric loading (see Fig. 18, appendix, for an example) and in loadings of up to 5.1 g_{VS}/L d. A comparison of different points in time is therefore only possible to a limited extent but different reactors can be compared for a given point in time.

Fig. 19 (appendix) shows the variation of the feed composition over time whereby some of the fluctuations are probably due to inhomogeneous nature of the raw sludge that is difficult to measure. The feed consists of 46% primary and 54% secondary sludge and the average feed composition is the following: TS: 7%, VS: 78%, COD: 68 g/L, COD_{soluble}: 8 g/L, NH₄⁺: 0.57 g_N/L, TKN: 2.8 g_N/L, P: 20 g/L, Acetic acid: 720 mg/L, Propionic acid: 450 mg/L, Butyric acid: 150 mg/L, total VFA: 1400 mg/L. The three VFA mentioned here account for ca. 95% of the total VFA.

Tab. 2: Average characteristics of the pilot-scale reactor during the mesophilic phase and the full-scale reactor in 2016.

	Pilot-scale	Full-scale
Volume	8 L (active volume)	11380 m ³
SRT [d]	21.1	23.0
OLR [gVS/L d]	2.5	2.7
Feeding scheme	1/d, 6/week	Continuous
Operation temperature [°C]	34.6-35.0	35.4

Tab. 3: Loading volumes with the loading scheme “normal” which took place before the start of the temperature increase and until day 75 and the loading scheme “new” which started on day 76.

	Mon	Tue	Wed	Thu	Fri	Sat	Sun
Normal loading [mL]	380	380	380	380	570	570	0
New loading [mL]	445	445	445	445	445	445	0

4.2 Parameters Measured

A wide range of parameters was measured and is shown in Tab. 4. This allows to

- monitor the reactor and intervene to prevent potential reactor failure
- assess the reactor performance to show differences in time and between the reactors.

The data of the continuous measurements done in the reactors by sensors are available in high temporal resolution and for this thesis minute values were chosen. Some measurements of the sludge and the gas quality were conducted in an external lab (see Tab. 4). The sludge sample analysed externally on a weekly basis always consisted of a mix of two successive days (see Tab. 15, appendix, for details).

Tab. 4: Overview on the parameters analysed. Heavy metals = Pb, Cd, Cu, Cr, Hg, Ni, Zn. Gas quality = CH₄, CO₂, O₂, N₂, H₂, CO, H₂O. Medium: s = sludge, g = gas, r = reject. Place: e = external laboratory, i = own analysis, r = reactor. Freq. = Frequency of measurements. Cont. = continuous measurement by online sensors. Precision as coefficient of variation (CV). Precision of heavy metals and gas depending on substance (1% for CH₄ and CO₂).

Parameter	Medium	Place	Freq.	Method	Precision
Temperature	s	r	Cont.	Sensor	unknown
Gas flow	g	r	Cont.	Level sensor (see below)	unknown
Gas quality	g	e		GC (SS-ISO 6974)	± 1-20%
pH	s	r	Cont.	Sensor	unknown
Conductivity	s	i	1/d	Sensor	unknown
Alkalinity	s	i		Titration with H ₂ SO ₄ (see below)	< ± 5% (n = 4)
VFA	s	i		Titration with H ₂ SO ₄ (see below)	< ± 10% (n = 4)
		e	1/w	GC (Journal of Chrom. A 963 (2002))	± 15 % (± 20 for acetic acid)
TS		i	2/w	Drying	± 3% (n = 6)
		e	3/w	Drying (SS-EN 12880:2000)	± 10%
VS	s	e	3/w	Drying (SS-EN 12879:2000)	± 10%
COD & COD _{filtr.}	s	e	1/w	ISO 15705:2002(E))	± 10%
TKN	s	e	1/m	SS-EN 13342	± 10%
Heavy metals	s	e	1/m	Different	± 15-25%
Phosphorus	s	e	1/m	SS 028150-2 / ICP-AES	± 15%
Dewaterability	s	i	2/w	Pressing device	± 1% for TS of cake
NH ₄ ⁺	r	i	2/w	Ion chromatography	unknown
	s	e	1/w	STANDARD METHODS 1998, 4500	± 20%
TSS	r	i	2/w	Filtration	± 5% (n = 3)

4.2.1 Gas Flow and Quality

The volumetric gas flow was measured with a device that uses two level sensors as shown in Fig. 20 (appendix). The gas that leaves the reactor first passes through a condenser where it is cooled using tap water. Then the gas flows through a tree-way valve into the measurement device where it increasingly pushes back the water column. When the surface of the water column reaches the second level sensor the valve opens towards the atmosphere, the gas is released and the signal recorded. After the water column is settled, the filling starts again. The results depend on the temperature of the tap water and the flow rate of the gas because this determines how much the gas is cooled and therefore its volume. If a cooling to the temperature of the tap water takes place reactors of different operation temperatures can be compared. Comparing the results of the pilot-scale reactors on different days to each other and to the full-scale plant is only approximative since the tap water temperature fluctuates and the results of the full-scale plant are given in normal litre (NL). A recalculation to NL was not done since the differences are small (for a tap water temperature of 7°C the pilot-scale results would need to be multiplied by 0.9653 to quantify them in NL) and the exact tap water temperatures are not known.

To measure the gas composition the biogas that leaves the reactor was collected in gas bags. It was then analysed in an external laboratory using gas chromatography equipped with a thermal conductivity detector (TCD). The water content was determined using an optical feedback cavity enhanced absorption spectrometer (OFCEAS).

4.2.2 Volatile Fatty Acids and Total Alkalinity

As described in chapter 2.1 volatile fatty acids (VFA) are an intermediate in the AD. VFA is a generic term for the fatty acids consisting of two to seven carbon atoms. The following VFA exist: acetic acid (HAc), propionic acid (HPr), butyric acid (HBu), iso-butyric acid (iso-HBu), pentanoic acid (HPe), iso-pentanoic acid (iso-HPe), n-hexanoic acid, n-iso-hexanoic acid and heptanoic acid. Changes in VFA concentrations are usually buffered and the alkalinity expresses the amount of acid that the buffers can neutralize before the pH starts dropping. If only the pH in an AD is monitored a high VFA concentration has to be present before any change can be observed. At that time it is usually too late to prevent reactor failure (Lahav and Morgan 2004). Since the loading in the experiments of this thesis are additionally based on the VFA/alkalinity ratio VFA and alkalinity have to be measured frequently and accurate results are needed. Hence, a 4-point potentiometric titration method to predefined pH values was developed. This method is adapted to the sample volumes and equipment available at Gryaab and uses the following standard procedures as reference methods:

- Alkalinity: Standard Method 2320 (Standard Methods 1999)
- Total VFA: Method of Kapp which was developed for sludge from mesophilic AD (Buchauer 1998)

Fig. 21 (appendix) shows the experimental setup used. Sulfuric acid (H_2SO_4) was titrated with the help of glass burettes. The acid was prepared with two dilution steps in a volumetric flask and it was assumed that this procedure is precise enough so that the standardization against sodium carbonate as described in the Standard Method 2320 is not needed. The pH was measured using a VWR pH110 pH meter that was calibrated with buffer solution of pH 7 and 4 every day. The initial pH and the acid consumed until pH 5, 4.3 and 4 is reached was recorded. Thus, for the measurement of alkalinity an endpoint of pH 4.3 was used at which all bicarbonate is converted into carbonic acid. The samples were centrifuged two times for 15 minutes (the second time taking the supernatant). The sample was then placed in a 50-mL glass beaker and stirred with a magnetic stirrer at a velocity of 300 rpm. This allowed for a fast mixing but didn't create a vortex and therefore reduced the gas exchange with the atmosphere to minimum. Only small sample volumes could be taken due to the limited volume of the pilot

scale reactors. Therefore, the samples were diluted with 20 mL of type 1 water (Elga purelab flex, 18.2 MΩ cm) so that the tip of the pH meter was immersed into the sample.

Tab. 5 shows the different steps of the method development. Both reference methods use 0.1 N sulfuric acid, corresponding to a molar concentration of 0.05 M. In the first step of the method development only this concentration was used in a 25-mL burette (step size: 0.1 mL).

The titration from a relatively high initial pH in the range of 7.7-8.2 to pH 5 worked well. However, for the titration to 4.3 and 4 the concentration of the acid was too high with respect to the sample volume and resulted in big pH jumps and unprecise results. In a second step 0.01 M acid in a 10-mL burette (step size: 0.05 mL) was used additionally. For the titration to pH 5 the 0.05 M acid was kept using to avoid a further dilution of the sample due to high titrated volumes and long titration times with higher rates of gas exchange with the atmosphere and stripping.

In the first two steps, the samples were centrifuged two time with a velocity of $4.4 \cdot 10^3$ rpm (Eppendorf 5702). This didn't result in a clear sample and since the Standard Method 2320 mentions a potential influence of suspended solids on the results it was decided in a third step to take the supernatant of the slower first centrifugation and centrifuge it at a velocity of $14.65 \cdot 10^3$ rpm (Eppendorf 5424). Since only 2 mL tubes could be used on this centrifuge the supernatant of the first centrifugation had to be split up into three tubes. From each of this tubes 1.67 mL were take (5 mL total sample volume).

After immersing the pH probe into the sample, the measured pH started drifting very fast. In the first three steps of the method development the initial pH was read out after a waiting period of 5 min. After evaluating the effect of this adaption time on the alkalinity and VFA results it was decided to read out the initial value directly after the probe was placed.

Tab. 5: Steps of method development. 4 = final method. Centrifugation for 15 min each. Dilution with 20 mL of water -> different dilution factors.

Step	Volume sample [mL]	Dilution sample	Centrifugation speed [10^3 rpm]	Waiting before start [min]	H ₂ SO ₄ concentration [M]
1	8	1:3.5	2x 4.4	X	0.05
2	8	1:3.5	2x 4.4	X	0.05 & 0.01
3	5	1:5	1x 4.4, 1x 14.65	X	0.05 & 0.01
4	5	1:5	1x 4.4, 1x 14.65		0.05 & 0.01

The following equations adapted to a titration with two different acid concentrations were used to calculate the total alkalinity and VFA:

$$\text{Alk} = \frac{\left(A_1 + \frac{A_2 * C_2}{C_1}\right) * N_1}{V} * 1000 * 50 \quad (1)$$

$$\text{VFA} = \frac{131340 * \frac{B * C_2}{C_1} * N_1}{V} - 3.08 * \text{Alk} - 25 \quad (2)$$

In final method:

Alk = Alkalinity [mg CaCO₃/L]

VFA = Volatile fatty acids [mg/L]

C₁ = Acid concentration 1 [mol/L]

C₂ = Acid concentration 2 [mol/L]

0.05

0.01

A_1 = Volume of acid 1 titrated to reach pH 5 [mL]
 A_2 = Volume of acid 2 titrated between pH 5 and 4.3 [mL]
 B = Volume of acid 2 titrated between pH 5 and 4 [mL]
 N_1 = Normality of acid 1 [eq/L] 0.1
 V = Volume sample 5

The method of Kapp overestimates the VFA concentration due to the effects of ammonium, hydrogencarbonate, sulphide or phosphate (Buchauer 1998; Lahav and Morgan 2004; Lützhøft et al. 2014). It was therefore decided to add known amounts of 1M HAc (the most important VFA found in the sludge of AD) and determine the recovery by comparing the measured concentration with the expected concentration:

$$\text{VFA}_{\text{expect}} = \frac{V * \text{VFA}_0 + V_{\text{HAc}} * M_{\text{HAc}} * C_{\text{HAc}}}{V + V_{\text{HAc}} * 10^{-3}} \quad (3)$$

Values used

$\text{VFA}_{\text{expect}}$ = VFA concentration expected after addition of know

amount of HAc [mg/L]

V = sample volume [mL]

VFA_0 = VFA concentration before the addition of HAc (mean value) [mg/L]

V_{HAc} = Volume of HAc added to sample [μL]

M_{HAc} = Molar weight of HAc [g/mol] 60.04

C_{HAc} Molar concentration of HAc [mol/L] 1

In the original method of Kapp the sample is filtered with a pore size of 0.45 μm . For one sample, the effect of this preparation was tested using disk filters and a syringe.

Since the pKa values of the different VFA are close together (Buchauer 1998) the titration method can give a good estimation of the total VFA. To get an overview on the fractions of the different VFA gas chromatographic measurements were done once per week as specified in Tab. 4. For this method, the recovery was also validated.

The development of the titration method took place in parallel with the experiments. For this reason, some samples from reactor T1 (details Tab. 16, appendix) were measured as described in step 1 in Tab. 5 and there was not enough volume left to repeat the analysis with the final method. Therefore additional samples were measured according to the first and last step of the method development to be able to convert the results of the first step and allow for comparison.

Samples from the other reactors were measured with the final method and were stored in tubes without headspace at 6°C with a storage time of up to 35 days. Also, the external lab stored the sample up to 21 days before analysis was done so that the effect of the storing was evaluated. Therefore, sludge from a mesophilic and thermophilic reactor (M1 and T1) was stored in completely filled 15 mL tubes for up to 30 days. Analysis was done after 5, 9, 13, 16, 20, 23 and 30 days.

4.2.3 Dewaterability and Characterization of Reject

The dewaterability of the sludge was measured by the pressure filtration test described in chapter 2.4 (details can be found in (Yulang Guo 2017)). 100 g of sludge were mixed with 11.5 g polymer/kg TS. It was then pressed for 40 min at 3.1 bar in the device that is depicted in Fig. 22 (appendix). The dose is a compromise between clean reject water (requires high dose) and high TS values of the resulting cake (requires low dose). The weight and TS of the resulting cake were determined and the volume of the reject was measured. Two polymers from BASF were used:

- Zetag 8180 (Z8180) for mesophilic and thermophilic sludge (also used in the full-scale plant)
- Organopol 6007 (O6007) for thermophilic sludge

Both polymers are cationic but the first has a charge density of 80%, the latter of 10%. O6007 also consists of beads and therefore has a narrower molecular weight distribution.

The reject quality was characterized by measuring the following two parameters:

- Ammonium nitrogen: by ion chromatography
- TSS: directly and after a centrifugation of 1 min at 0.5 rpm to remove particles that can easily be settled. Munktell Ahlstrom grade MGA filter paper was prepared according to the Standard Method 2540 D (Standard Methods 1999)

4.3 Data Processing

For the analysis, big amounts of data from different sources and in different formats has to be handled. The processing was done using the open source programming language R. All available data was read in and stored in one dataframe for each pilot scale reactor. More details on the structure of the codes can be found in Tab. 19, appendix. Only the most important codes and measurements are shown to limit the page number but a complete digital dataset and code is provided with this thesis.

4.3.1 Calculation of Gas Volume per Cycle

After each loading of the reactor a new cycle starts in which biogas is produced. The amount of gas produced per cycle must be calculated from the gas sensor data. In a first step, the outliers of the gas flow that are generated every time the reactor is opened for loading were removed. This was done by calculating the moving average over 60 data points (60 min.). If a data point was bigger than 1.15 or small than 0.8 times the moving average it was defined as outlier and replaced by a mean of the point before and after the outlier. Additionally, all data points above 25 mL/min were defined as outlier to make sure that the algorithm also works for longer periods of open reactors.

In a second step, the gas volume after each loading (= cycle) was then calculated from the area under the gas flow curve. Therefore, the start of each cycle had to be determined since the loading was not done at the exact same time each day. This was done by searching for days with loading and identifying the point where the change of values over time was the biggest since the gas production increases rapidly in the beginning of the cycle. The starting points of the cycles were then used to calculate the total gas volume produced in the time between two starting points. This was done by splitting the data of each cycle into slices of 5 minutes length and using the average gas flow per slice to calculate the area and adding up all areas per cycle. Only cycles with normal length (approximately 1 day from Monday to Friday and 2 days on Saturday) were used to avoid falsification of the results by integration over too long time periods.

4.3.2 The Time Aspect in Data Processing

As shown in Tab. 4 the analysis of the different parameters was done in various frequencies. Moreover, the external analysis of the raw and digested sludge was done for a mix of sludge from two days. If an analysis involves parameters that were measured with different frequencies not all parameters are available at a given point in time. Therefore, either the data with low frequency must be extrapolated which increases the uncertainty or the more frequent data has to be averaged which reduces the points available.

When doing mass balances, the time period between a change in the input characteristics and a result in the output characteristics is unknown. To evaluate this effect a model was developed. It was assumed that the further away in time the measurement of the output is from the input the lower is the influence of the input on it. For a given input the output was calculated by summing up weighted outputs over a period that can be specified. The weighing factors were estimated based on a linear model as shown in Fig. 1 (more realistic would probably be an exponential decrease) with equation (4). With the total area set to 1 the different areas represent the weighing factors that become smaller the further away the output is from the input. By choosing the divisor D the period of influence can be adjusted.

$$A_i = \frac{1 + (i - 1) * 2}{D^2} \quad (4)$$

$i = 1, 2, \dots D$

A_i = Weighing factor
 D = Divisor

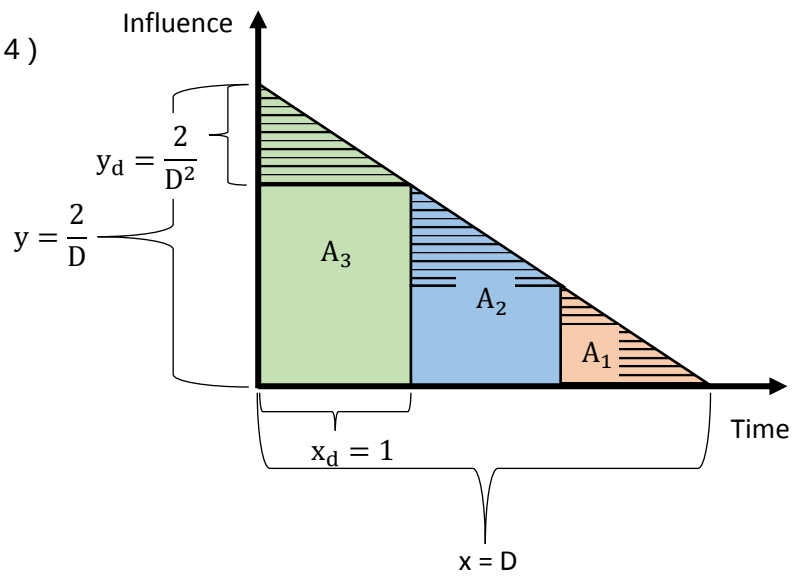


Fig. 1: Visualization of equation (4). The different areas represent the factor that is used to multiply the data at different points in time. Here $D = 3$.

5 Results and Discussion

5.1 Method for the Determination of Alkalinity and Volatile Fatty Acids

As described in chapter 4.2.2 several influencing factors were considered in the method development.

Even though a higher velocity of the second centrifugation removes many suspended solids and the liquid becomes clear it does not have a big influence on the alkalinity or VFA results (see Fig. 23, appendix). Since this effect was tested on sludge from the full-scale plant which is less fine than that of the thermophilic AD it was still decided to centrifuge with a higher speed in the final method.

No effect can be seen concerning the adaption time before the start of the measurements either (see Fig. 24, appendix). In the final method, the titration was therefore done directly after immersion the pH probe to save time.

The filtering of the sample shows no effect on the results of the alkalinity measurements but results in a considerable decrease of VFA (see Fig. 2). Even though the sample had a light-yellow colour the filter clogged after about 1 mL of sample filtered.

Therefore, it was

necessary to use big pressing forces and change the filter several times to get the desired amount. The drop in VFA could be explained by the handling of the sample. Some air is sucked up together with the sample and then filtered with high pressure. This could lead to a stripping of VFA. Since the comparison of the different centrifugation velocities does not show an influence of even bigger particles and the filtering as described above is not feasible on an everyday basis this was not further investigated.

As can be seen in Fig. 25 (appendix) the storing does not have an influence on the results of both alkalinity and VFA. The storing time should be limited as much as possible but if samples cannot be measured directly they can be stored at 6°C without concerns for up to 30 days.

When comparing the measurements of given samples using a titration according to the first and last step of the method development the following equations for conversion can be found (also see Fig. 26, appendix):

$$\text{Alk}_{\text{final}} = -237.37 + 1.0356 * \text{Alk}_{\text{first}} \quad R^2 = 0.9934 \quad (5)$$

$$\text{VFA}_{\text{final}} = -63.047 + 0.9779 * \text{VFA}_{\text{first}} \quad R^2 = 0.9984 \quad (6)$$

$\text{Alk}_{\text{final}}$ and $\text{VFA}_{\text{final}}$ = Alkalinity and VFA using the final method

(step 4) of the method development

$\text{Alk}_{\text{first}}$ and $\text{VFA}_{\text{first}}$ = Alkalinity and VFA using the first method
(step 1) of the method development

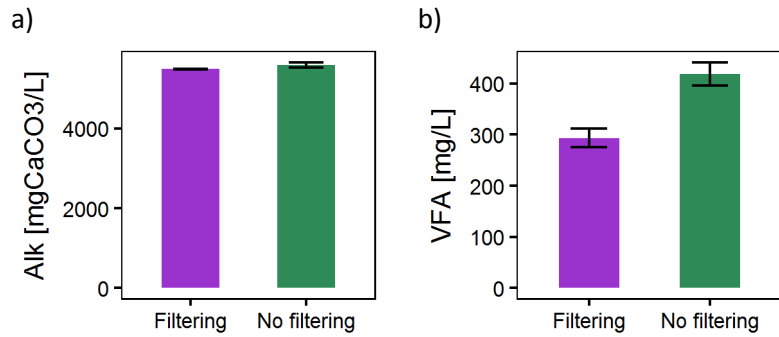


Fig. 2: Alkalinity (a) and VFA (b) measurements with and without filtration of the centrifuged sample with a 0.45 µm disk filter. Filtered samples measured as duplicates, non-filtered ones as triplicates. Error bar shows 1 standard deviation.

When comparing the expected and measured concentrations of samples spiked with HAc a clear trend showing an overestimation becomes visible (see Fig. 3 a)) and an equation for the correction of all titration results based on recovery can be determined:

$$\text{VFA}_{\text{corr}} = 173.27 + 0.8028 * \text{VFA}_{\text{titration}} \quad R^2 = 0.9887 \quad (7)$$

$\text{VFA}_{\text{real}} = \text{VFA}$ corrected based on recovery

$\text{VFA}_{\text{titration}} = \text{VFA}$ measured by titration according to Kapp

This correction is based on measurements of HAc only but should give a good estimation since HAc is the VFA with the highest concentration.

As can be seen in Fig. 3 b) no systematic trend can be found for the recovery of VFA using the GC. However, the number of VFA present in the sample influences the total VFA measured (the more present the higher the total VFA). On one hand, this is due to the limit of quantification (LOQ). VFA other than HAc and HPr are only present in low concentrations. For the calculation of the total VFA a concentration of zero was assumed if the result was below the LOQ. This probably results in a small underestimation of the concentration and is also the reason for the higher scattering of this data points in Fig. 3. On the other hand, it is possible that the peak picking works less well with many VFA present. Since the effect of the two factors mentioned is not entirely clear the results of the GC were used without correction.

The titration method could be improved if higher sample volumes were available. This would allow for measurements without dilutions and the use of an Erlenmeyer flask instead of a beaker to limit the gas exchange with the atmosphere. It should also be considered to use an automatic titration so that a titration curve can be obtained. This would allow to get the exact location of the characteristic points.

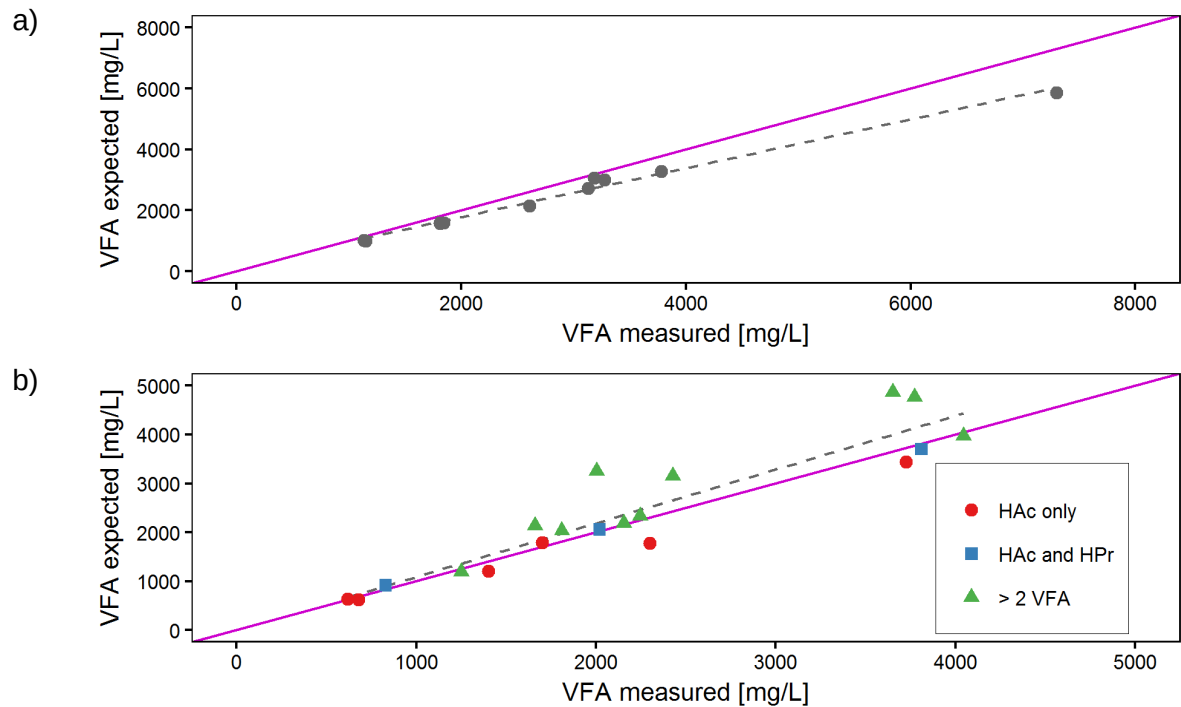


Fig. 3: Comparison of the VFA concentration measured by titration (a) and GC (b) with the expected concentration based on different amounts of acetic acid added. HAc = acetic acid, HPr = propionic acid. Titration results show the average of duplicates or triplicates (mean CV: 4.1 %).

5.2 Data Processing

Fig. 4 shows exemplary for reactor T3 how the algorithm for cleaning the gas flow data removes the outliers. Since the cleaned data is plotted on top of the raw values the latter are only visible if they were identified as outliers. In Fig. 27 (appendix) the results for the complete dataset are shown.

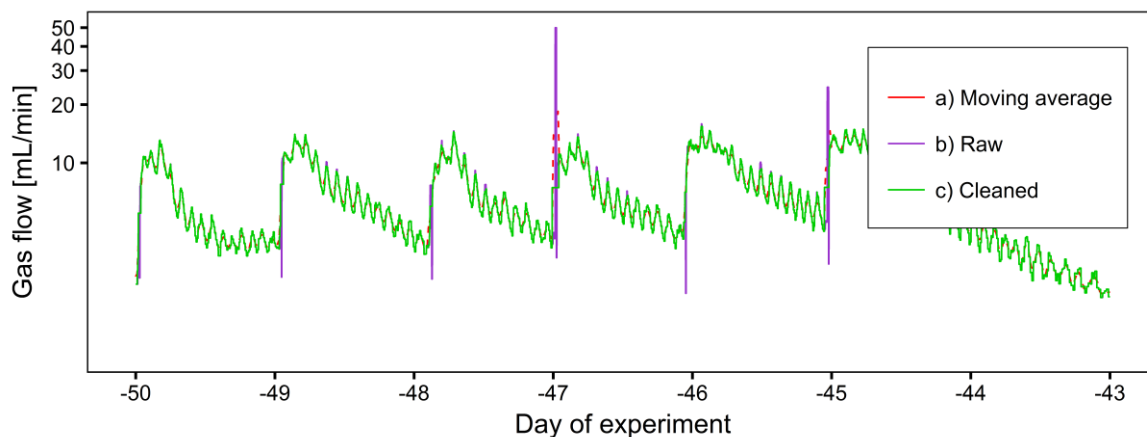


Fig. 4: Raw (purple) and corrected (green) gas flow data over one week in reactor T3. The red line shows the moving average that was used to identify outliers. Y-axis in logarithmic scale.

The calculation of the total gas volume for each loading is visualized in Fig. 5 for reactor T3 over one month (plot over the whole experimental period: see Fig. 28, appendix). It can be seen how the algorithm identifies the starting points of each cycle. The gas volume (assigned to the middle of the cycle) is shown together with the area used for its calculation. The gas volume during Saturdays is higher than for the rest of the week since here the cycle lasts for approximately two days. As mentioned in chapter 4.3.1 only normal cycle lengths were used for the calculation of the gas volume. This results in gaps during the time of reduced feeding or when the normal feeding scheme could not be followed due to public holidays (see Fig. 28 , appendix).

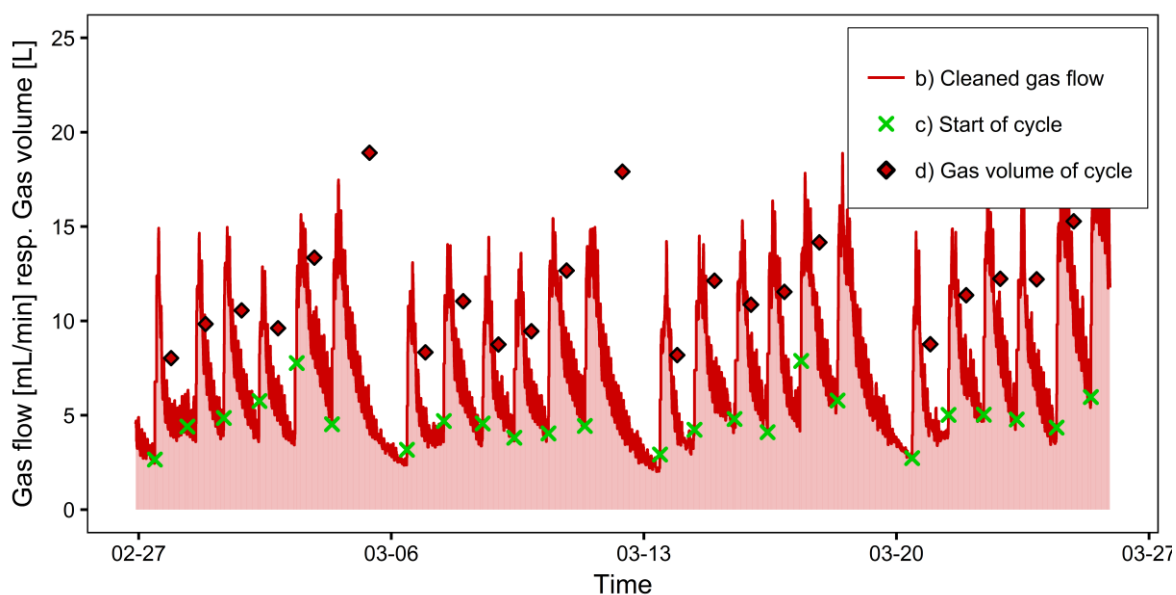


Fig. 5: Results of the calculation of the gas volume in reactor T3 for 1 month. The gas flow in mL/min is shown as solid line. The gas volume for each cycle in L is denoted as diamonds together with the shaded area under the gas flow curve that was used for the calculation. The green crosses denote the start of each cycle.

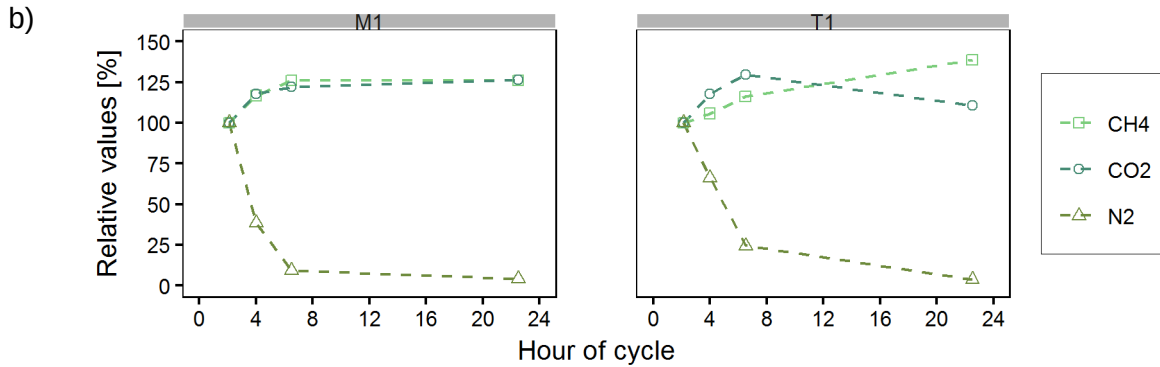


Fig. 7: Variation of measured parameters in sludge and gas over the course of one cycle. a) 2017-06-15 and 16 during normal feeding scheme; b) 2017-07-25 and 26 during new feeding scheme.

Fig. 8 shows for the example of the VS removal the effect of different time shifts between the measurement of ingoing and outgoing sludge. If values of the same day are used (no shift) there is no pattern visible since changes in the characteristic of the incoming sludge cannot be reflected in the characteristics of the outgoing sludge measured at the same time. When a shifting is included a weekly pattern becomes visible with a lower VS removal in the beginning of the week. This is in accordance with what can be observed in Fig. 6. Which time shift matches the real characteristics of the reactor could best be determined by interrupting the feeding for a longer period and determine the drop of gas production.

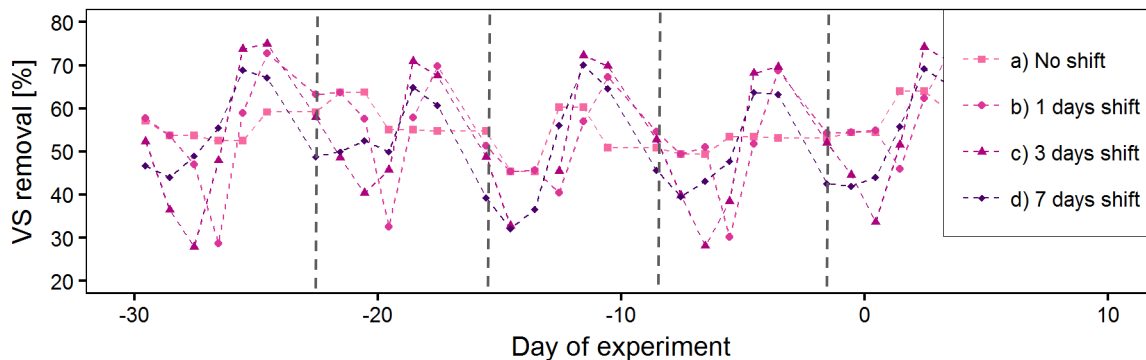


Fig. 8: VS removal over time in reactor T2 for different time shifts between the value of the VS input and output used to calculate the removal. Calculation bases on equation (4). The vertical lines mark the beginning of a week.

5.4 Influence of the Temperature Increase on the Reactor Performance

In the following the most important parameters characterizing the reactor performance are analysed over time to evaluate the best strategy for temperature conversion. Additionally, mass balances are done to compare the performance before and after the conversion. Data points are considered as being part of the phase after conversion if they are measured after the normal loading scheme as described in Tab. 3 is reached again. On day 59 the cake that formed on the stirrer in the reactors was removed which had an influence on the performance. For a better visibility only one mesophilic reactor (M1) is shown in the plots. The performance of M2 and M3 is comparable and shown in Fig. 30 (appendix). If data is split according to time phases a different number of points is assigned to the different phases depending on the time the reactors needed to reach the normal loading scheme again. Important points in time are marked in the graphs according to Tab. 7.

Tab. 7: Points in time that are marked in graphs.

Label in plots	Day of experiment	Description
1	0	Start of the temperature increase. Direct increase to 55°C in T1
2	7	Critical temperature of 43°C reached in T2 and T3. Loading in T3 interrupted
3	19	55°C reached in T2 and T3. Loading in T2 interrupted
4, 5, 6	27, 41, 55	T1, T2 and T3 back to normal loading scheme. Start of phase “after” for the corresponding reactors
7	76	New loading scheme established

5.4.1 Reactor Transition and Stabilization

Fig. 9 shows the most important parameters of reactor performance. The highest VFA concentration develops in reactor T2. In this reactor, the peak concentration of HAc is approximately 150 times higher than before the temperature rise (up to 3900 mg/L, not shown in Fig. 9 b) for better visibility but included Fig. 31, appendix) and that of HPr 80 times (up to 820 mg/L). The concentrations of the other VFA show a simultaneous peak (see Fig. 31, appendix). The VFA/alkalinity ratio also shows a peak for this reactor. However, here the values are only approximately 15 times higher than before the temperature rise which shows that the buffering of the reactor works well. Reactor T1 also shows an increase in the VFA/alkalinity ratio and the different VFA but the peaks especially for the HAc are much smaller than in reactor T2. Interrupting the loading when the temperature of 43°C is reached like it was done in reactor T3 keeps the VFA concentrations lower than in the other reactors (see Tab. 8). However, after interrupting the loading of T2 at 55°C this reactor stabilizes quickly while the concentration of HPr continues rising in reactor T3 even though almost no loading took place. The stabilization of reactor T3 was difficult because an increase of the loading resulted in an immediate increase of the VFA/alkalinity ratio so that the loading had to be reduced again. Only after the HPr concentration decreased, the loading could be increased to the normal amount. Tab. 8 shows that T1 was the reactor where the normal loading scheme could be re-established the fastest (after 4 weeks) while T3 could be loaded normally only 2 months after the start of the temperature increase and had the highest number of days without any loading. The number of days without loading is comparable for T1 and T2 but the overall transition period of T2 lasts longer since the increase of the temperature takes almost 3 weeks during which the reactor can only partially stabilize.

In all reactors, the HAc concentration rises first before the HPr also increases. This is consistent with what can be found in literature (Iranpour et al. 2002; Boušková et al. 2005). After stabilization, the thermophilic reactors show approximately two to three times higher VFA/alkalinity values than the mesophilic ones and values fluctuate between ca. 0.15 and 0.3 (as compared to ca. 0.1 in M1-M3. In literature, different statement on the VFA levels after stabilization are made. While Zabranska et al. (2002) found lower levels, Iranpour et al. (2002) and Cavinato et al. (2013) report similar levels and de la Rubia et al. (2005), Nielsen and Petersen (2007) and Tezel et al. (2014) obtained higher concentrations. From literature, it cannot be concluded which rate of temperature increase or loading favours low VFA/alkalinity ratios. For inoculated sludge Ghasimi et al. (2015) found that in the first 24 hours after loading the HAc concentration was nine times higher in thermophilic reactors in comparison to mesophilic ones. If a similar behaviour can be found in the reactors used for this thesis this could lead to an accumulation of VFA and explain the higher VFA values in the thermophilic reactors.

After temperature increase the HPr concentrations are high in all reactors (almost as high as the concentrations of HAc). HPr was identified by Van Lier et al. (1990) as the VFA that reacts most sensitive to sudden temperature changes. According to Nielsen, Uellendahl and Ahning

(2007) it is the most suitable parameter for process optimization because it shows a reactor upset 12-18 days before the CH_4 production decreases. Gebreeyessus and Jenicek (2016) reported that the oxidation of HPr is sensitive to high H_2 levels. Since the concentration of this gas is zero during the whole experimental period it can be suspected that step 3 of the AD (see chapter 2.1) does not work with a sufficient rate. Even after stabilization iso-HBu and iso-HPe can be found in T1-T3 (see Fig. 32, appendix). Lin and Hu (1993) reported that HAc inhibits the degradation of iso-HBu more than that of HBu. That iso-HBu persists in the thermophilic reactors could therefore indicate a slow rate in step 3 of the AD as well. When comparing continuously fed reactors with direct and stepwise temperature increase Palatsi et al. (2009) found a similar behaviour of HAc during the transition but higher peaks for HPr in the reactor with direct increase. This is the contrary to what is found in the experiments of this thesis.

The variations of the concentrations of the different VFA are probably caused by the variations in the feed (see Fig. 19, appendix) and can also be observed in M1-M3 but on a lower level (see Fig. 31, appendix). It should also be noticed that due to inhomogeneities in the sludge the uncertainties of the measurements are high.

Tab. 8: Overall days without any loading and time until the normal loading scheme was reached again and peak VFA values.

	T1	T2	T3
Days without loading	9	10	16
Length transition phase [weeks]	4	6	8
Rounded peak VFA concentration	2900	6600	2700

The recovery of the gas production takes long (see Fig. 9 d)) and the thermophilic reactors cannot reach the levels of the mesophilic ones. It must be noticed that in Fig. 9 d) there are gaps in the gas volume data during the transition phase due to the way the total gas volume is calculated (see chapter 4.3.1). Fig. 28 (appendix) shows that the gas production drops to zero in reactor T1, while in reactor T2 it is never below approximately 2.5 mL/min and there is a slow de- and increase in reactor T3 with a minimum around day 14. The lower gas production in the thermophilic reactors becomes visible in Fig. 33 (appendix) which shows the cumulative gas volume after the normal loading scheme was re-established and in Fig. 10 which compares the average gas volume of the mesophilic reactors (M1-M3 are in the same range, see Fig. 30, appendix) with that of T1-T3. Tab. 9 shows the results of a t-test on the hypothesis H_0 that the gas volume of the mesophilic reactors is not higher than in the thermophilic ones. According to this test, before the temperature increase the performance of all reactors is in the same range except for T1. After the conversion, all thermophilic reactors produce significantly less biogas. Both plots and the t-test include all date points after the normal loading scheme was reached (while the gas production was still recovering) but even before the new loading scheme starts the gas production is not as high as in the mesophilic reactors.

Contrary to the volume of biogas its CH_4 content recovers fast (see Fig. 9 e)). In reactor T1 the period of reduced CH_4 content lasts approximately two weeks, in reactor T2 and T3 three weeks. There is a difference between the reactors concerning the composition of the biogas during the time of reduced CH_4 content. As can be seen in Fig. 34 (appendix) in reactor T1 and T2 the drop in the CH_4 share is due to an increase of CO_2 while in reactor T3 it is caused by an increase of N_2 . In literature, the recovery of the CH_4 or biogas production (often not specified exactly) is reported to last between 20 (Cavinato et al. 2013) and 70 (Boušková et al. 2005) days. Boušková et al. (2005) found that a direct temperature increase leads to faster recovery than a stepwise (30 vs. 70 d) while Palatsi et al. (2009) made the opposite observation (35 vs. 20 d) and Palatsi et al. report a similar speed but a more stable behaviour in the stepwise increase.

43°C proved to be the critical temperature of the continuous temperature conversion. At lower temperatures, there is no deterioration in any of the performance parameters of reactor T2 and T3 shown in Fig. 9.

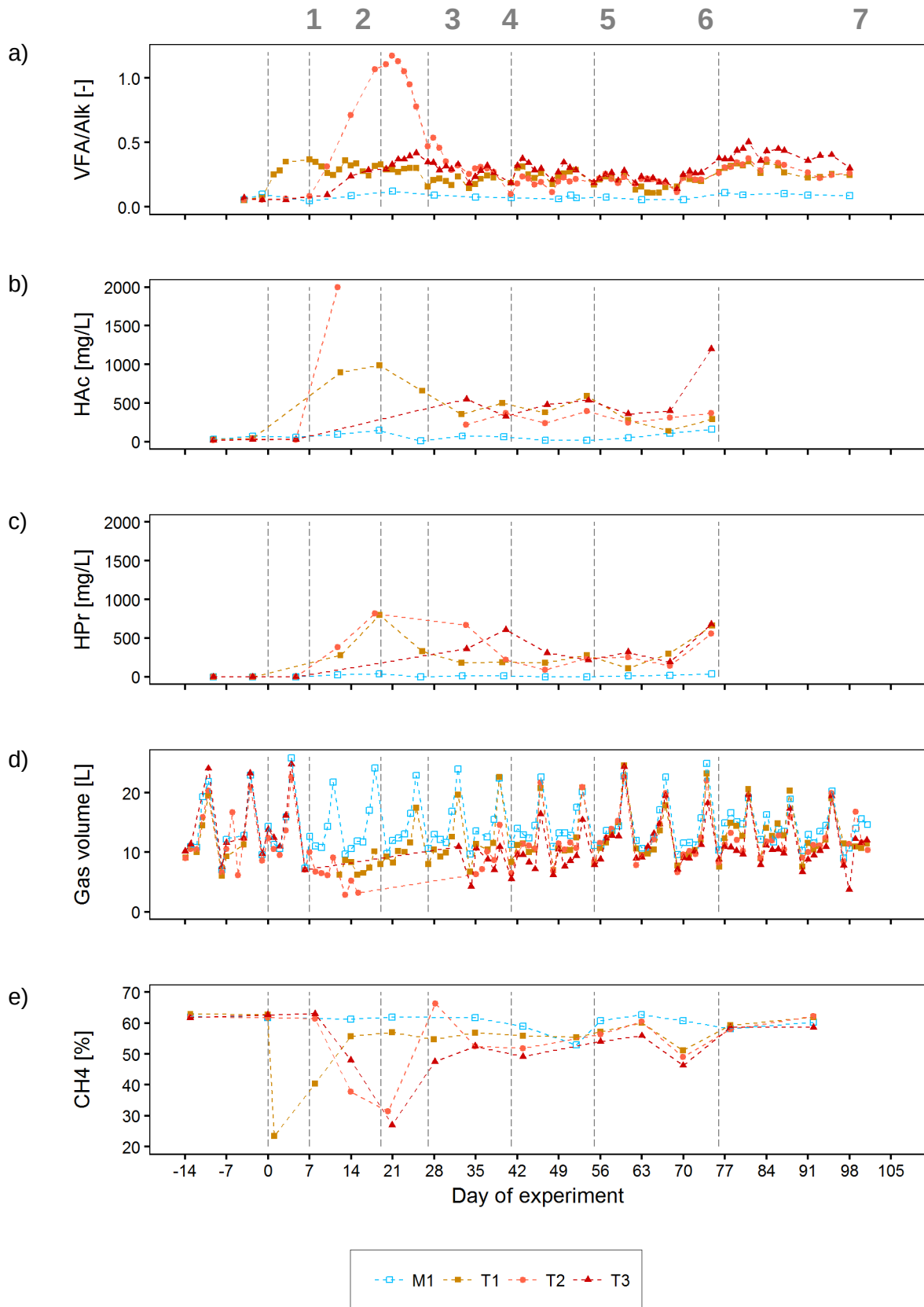


Fig. 9: Parameters of reactor performance over time. Time phases: 1: Start of temperature increase; T1: 55°C; 2: T2 & T3: 43°, loading of T3 stopped; 3: T2&T3: 55°C, loading of T2 stopped; 4: T1: loading normal; 5: T2: loading normal; 6: T3: loading normal; 7: new loading scheme.

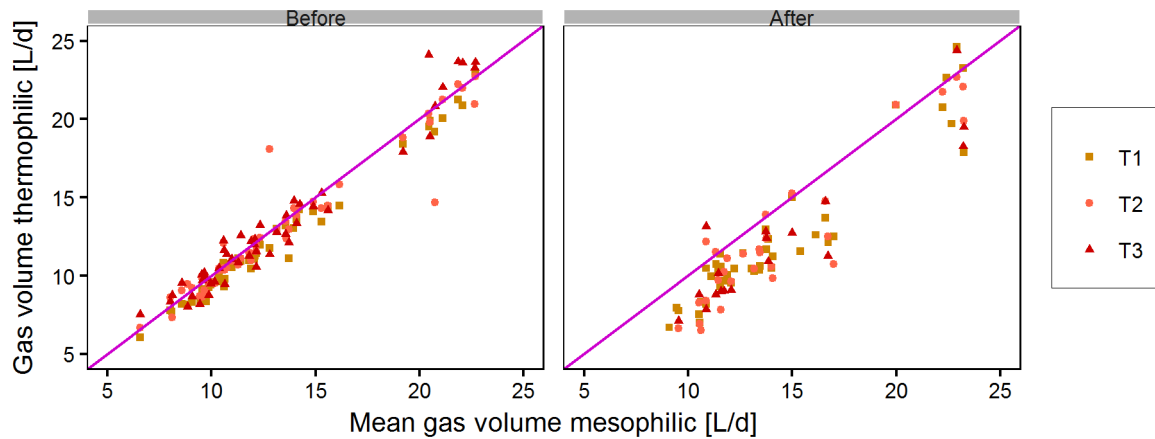


Fig. 10: Average gas volume per cycle in reactor M1-M3 (x-axis) vs. gas volume per cycle in reactor T1-T3 (y-axis) before and after the temperature transition.

Tab. 9: Comparison of the mean difference between the average gas volume in reactor M1-M3 and the gas volume in reactors T1-T3 before and after temperature transition. p-values for t-test on H_0 : thermophilic not smaller mesophilic gas volume. Values < 0.05 are highlighted.

	T1		T2		T3	
	Before	After	Before	After	Before	After
Gas volume mesophilic minus thermophilic reactors	0.6	1.9	0.2	1.8	0.05	1.4
p-values	5.8*10 ⁻⁷	1.7*10 ⁻⁸	0.09	1.1*10 ⁻⁴	0.6	0.02

Fig. 11 and Tab. 10 show the results of a COD balance. For the calculation of the COD leaving the reactor in form of CH_4 a value of $0.363 \text{ L}_{\text{CH}_4}/\text{g}_{\text{COD,removed}}$ was used which assumes a gas temperature of 7°C (estimation of the tap water temperature). The different amounts of COD fed to T1-T3 in the period “after” are due to the different starting points of this period and a variation in the feed (see Fig. 35, appendix). The reduced gas volume described above leads to lower COD values in the biogas after the transition but also the COD values in the digested sludge are higher than before the transition. This balance highly overestimates the COD leaving the reactor (see Tab. 10) both before and after the transition. One reason for this could be the variation of the CH_4 content during the cycle (see Fig. 7) so that the gas composition depends on the sampling period. The average ratio of COD/VS is around 0.89 in the feed and around 0.63 in the digested sludge. This means that the reduction of COD is higher than that of the VS. The COD/VS ratio of the raw sludge is low so that the biogas potential is also low and the sludge probably contains little amounts of substances with a high COD like proteins or lipids (Cavinato et al. 2013; Vesilind 2003). While the total COD that leaves the reactor with the digested sludge stays approximately the same over time for all reactors, the proportion of the soluble COD on the total COD that leaves the reactors increases in T1-T3 but seems to decrease again after stabilization but there are big fluctuations (see Fig. 35, appendix). Even though the TS and VS content of the feed is approximately constant (see Fig. 19, appendix) there is a slight increase of the TS and decrease of the VS in all reactors starting around day 28 (see Fig. 36, appendix). This could be caused by the build-up of the cake on the stirrer as described above since with the removal of it on day 59 this trend is stopped after some disturbances.

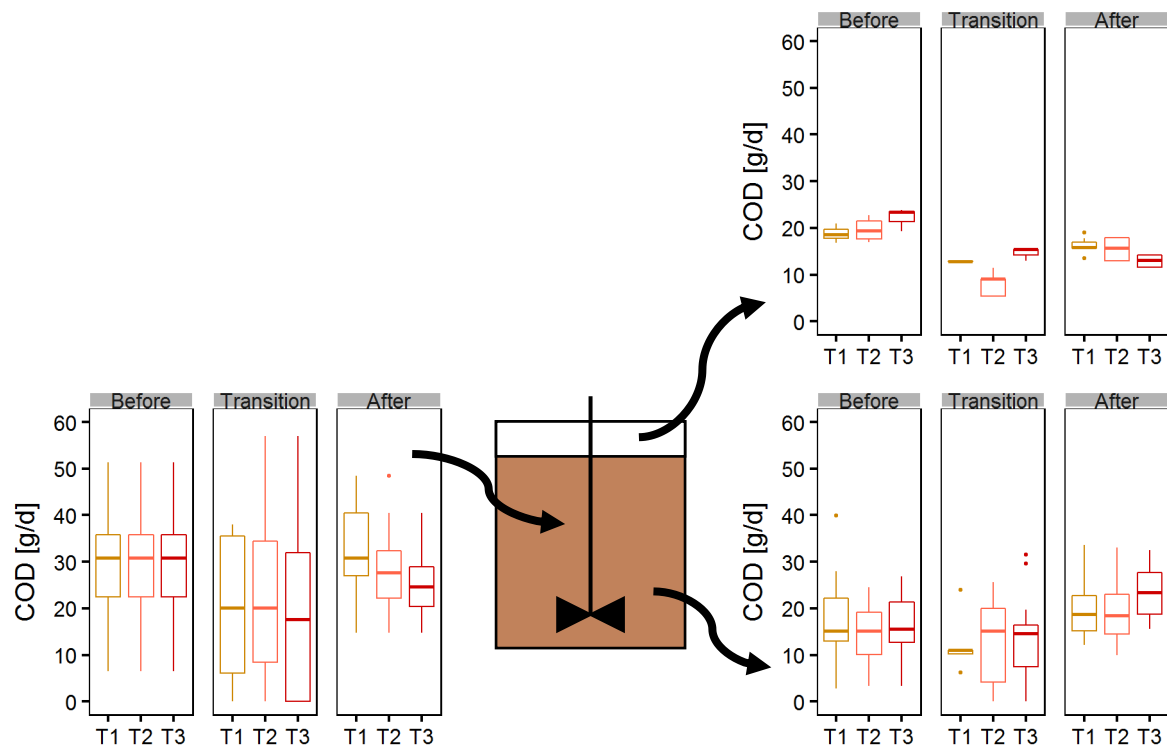


Fig. 11: COD balance considering the raw and digested sludge and the methane in the biogas produced.

Tab. 10: Comparison of COD out (sludge and biogas) and COD in for the different reactors and experimental phases.

	T1	T2	T3
Before [% of COD in]	119	115	126
After [% of COD in]	112	121	142

Measuring the pH in the reactors proved to be difficult. The calibration was not possible for some of the online sensors and the sensors failed successively. According to the manufacturer this can only be solved by expensive software changes. Starting from around day 40 the pH was therefore measured manually once per day in the sludge taken out from the reactors at the end of each cycle. Like in the titration experiments the measured value drifts quickly after the immersion of the probe and therefore is only an estimation. The thermophilic reactors display a pH that is approximately 0.5 higher than in the mesophilic reactors. In the mesophilic reactors, the pH is in the range of 7.3 to 7.4 while in the thermophilic ones values between 7.7 and 7.9 are measured. The higher pH in the thermophilic reactors is also reported by Ziemińska-Buczyńska et al. (2014).

As expected, the change in temperature does not show any effect on the heavy metal content of the sludge. The phosphorous levels in the digested sludge are not influenced by the temperature either. It should however be noticed that those values were only assessed in a mix of sludge over one month so that short time variations are not depicted.

Fig. 12 shows the mass balance for the nitrogen (considering TKN and NH_4^+) over the reactor and the NH_4^+ that can be found in the reject after dewatering. The nitrogen that leaves the reactor in form of N_2 in the biogas is not considered here since the values depend on the time of measurement during the cycle (see Fig. 7). The TKN values are only measured as average over one month and depending on the length of the depicted period are not available or only in form of one value. 50-60% of the TKN is converted into NH_4^+ in the reactor like it is reported

by Vesilind (1974). Contrary to what was found by Nielsen and Petersen (2000) and Tezel et al. (2014) this is independent of the temperature.

It is important to notice that there is an equilibrium between ammonium (NH_4^+) and free ammonia (NH_3 = FAN) which together are referred to as total ammonia nitrogen (TAN). Especially the FAN can inhibit the methanogenesis but the bacterial community can adapt partially to high FAN levels (Yenigün and Demirel 2013). The review done by Yenigün and Demirel (2013) however only covers mesophilic conditions. The equilibrium between NH_4^+ and NH_3 depends on the temperature and pH (Gujer 2007) which are both different between the thermo- and mesophilic reactors. Thereby, especially the influence of the pH is high (see Tab. 11). The NH_4^+ and TKN values in the feed are comparable and the maximum NH_4^+ concentration in all reactors does not exceed 2200 $\text{mg}_\text{N}/\text{L}$. These values are however measured at the end of each cycle in sludge that has the same temperature and due to the higher pH and temperature in the thermophilic reactors high FAN concentrations can be expected in T1-T3. It is likely that these concentration lead to an inhibition of the methane production. Fig. 7 and Tab. 17 (appendix) show that the gas composition in the beginning of each cycle is similar in M1 and T1. However, in T1 the CH_4 concentration only rises after the N_2 concentrations dropped. This could indicate that first parts of the FAN must leave the reactor in form of N_2 before higher methanogenesis rates are possible.

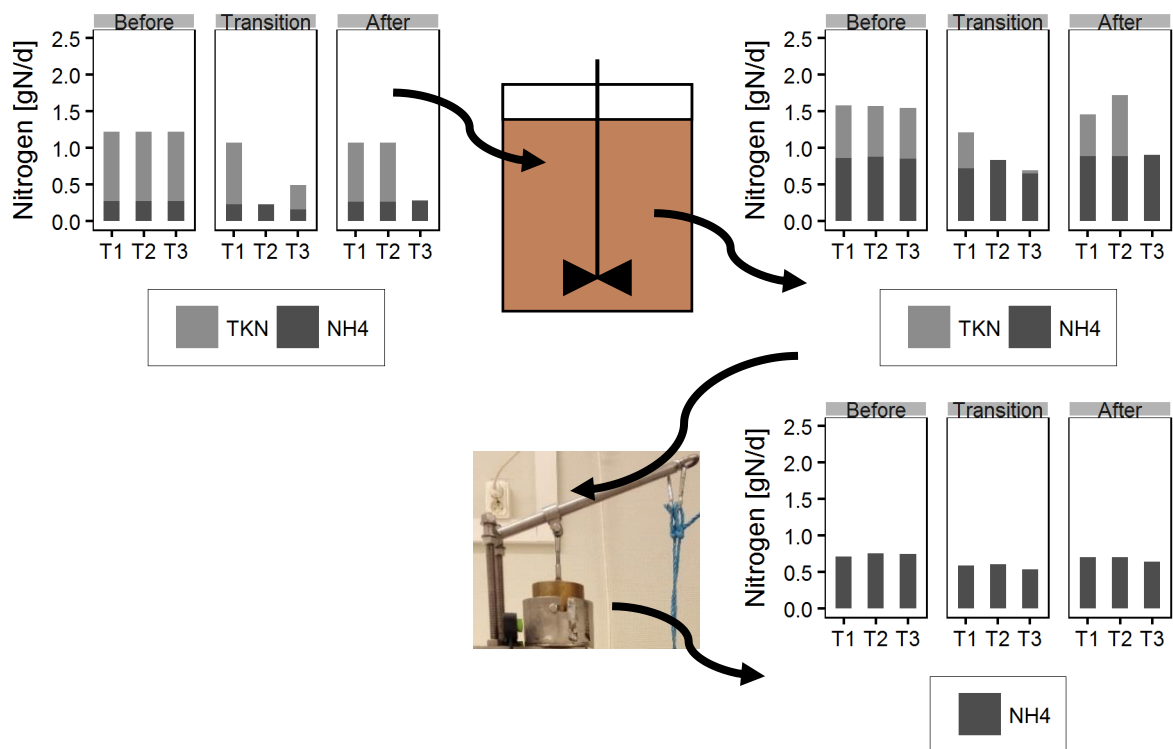


Fig. 12: Nitrogen balance considering the TKN and NH_4^+ of the raw and digested sludge and the NH_4^+ of the reject after dewatering.

Tab. 11: Overview on fraction of TAN that exists as FAN for different temperatures and pH values.

	pH 7	pH 7.5	pH 8
35°C	1.6%	5%	
55°C	4%	12%	29%

5.4.2 Dewaterability of the Digested Sludge

In the following the different aspect related to this key parameter are analysed. Fig. 13 shows how the dewatering behaviour of the sludge is influenced by the change in operation temperature. The outliers visible on day 63 are due to the removal of the cake on the stirrer mentioned above.

The weight of the cake was identified as the most reliable parameter characterizing the dewatering behaviour. This parameter shows a decrease after the temperature change took place but stabilizes on a lower level after the normal loading scheme is reached and the gas production increases. Using the polymer that is adapted to the mesophilic sludge (Z8180) leads to a dark reject with higher TSS contents. This was also reported by Nielsen and Petersen (2000). There is no big difference between the TSS that is measured directly or after a short centrifugation which means that the particles in the reject are small. The NH_4^+ content of the reject increases slightly during the transition period but there are big fluctuations also in M1. The influence of the temperature on the reject NH_4^+ content that is reported Gebreyesus and Jenicek (2016) is not clearly visible and it was therefore decided to not further analyze this parameter. Fig. 12 shows that approximately 70-90% of the NH_4^+ found in the digested sludge is transferred into the reject because of its high solubility independently of the polymer used.

The weight of the cake in the reference reactor M1 increases slightly over time in parallel with the increasing TS values in the sludge (see Fig. 36, appendix). This is not expected since the polymer is dosed based on the TS values of the sludge. The method for the TS determination for dewatering however underestimates the TS content so that the polymer dose that is actually added is smaller than intended. According to Yulang (2017) this results in a lower TS and higher weight of the cake than expected. The underestimation is due to the method that uses a syringe to handle the unhomogenized sludge so that clots in the sludge are not considered in the measurements. The underestimation is the higher the TS content is (see Fig. 37, appendix, which shows a comparison to the external measurements). In parallel to the increase of the cake TS in M1 the directly measured TSS in the reject also increases. There are however mainly big particles formed that can be removed by centrifugation (see results of TSS after centrifugation with stable values for M1).

Fig. 13 also evaluates the effect of the polymer used for the dewatering. It can be seen that adapting the kind of polymer to the changing sludge characteristics (use of polymer O6007 for thermophilic sludge) can increase the weight of the cake by approximately 30% so that around 90% of the weight found for reactor M1 can be reached (see also Tab. 12). This also reduces the TSS in the reject and increases the size of the particles that pass the grid (visible difference between TSS direct and after centrifugation). The TSS in the reject can be reduced to levels found in M1 or below with polymer O6007 but the values after centrifugation are slightly higher indicating that the particles are finer in the thermophilic sludge. Reactor T1 generally shows a higher weight of the cake with both polymers compared to the other thermophilic reactors.

Tab. 12: Average weight of the cake after dewatering for the time period 2017-06-05 to 2017-07-09 (phase where both polymers used).

	T1	T2	T3	Mean T1-T3	M1
O6007	11.9	11.2	11.2	11.4	
Z8180	9.3	8.2	8.8	8.8	12.4

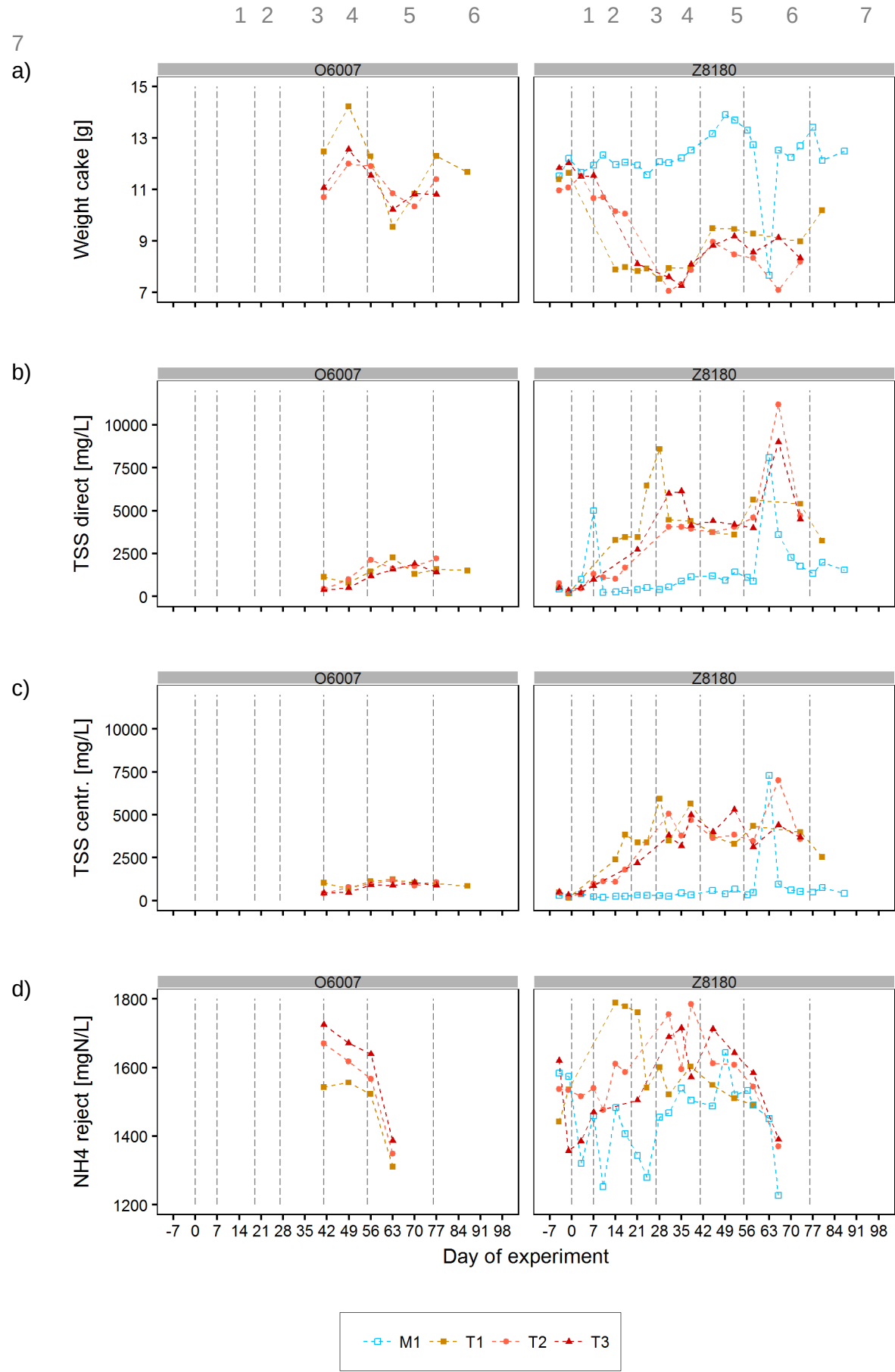


Fig. 13: Results of the dewatering experiments over time for the two polymers used. Time phases line are defined in Fig. 9.

During the dewatering experiments parts of the total mass and the TS are lost in the pressing device. How much is lost depends on the operation temperature and the kind of polymer used and is shown in Tab. 13 and Fig. 14. A comparison between reactors and polymers is therefore only an approximation. The reject dominates the balance of the total weight. Therefore, the difference for the different polymers used on the thermophilic sludge is small (see Tab. 13). However, when using the polymer Z8180 on the thermophilic sludge lots of solids pass the grid but get stuck in the device and are therefore not found in the reject. This influences the TS balance which is dominated by the fraction found in the cake. Parts of the solids that pass through also stick to the bottom of the grid and are therefore included in the measurement of the cake weight even though they still contain lots of water. This leads to an overestimation of the weight of the cake. Since only the part of the cake on top of the grid is used to determine its TS there is an error introduced. The TS mass balance shown in Fig. 14 also only ignores the soluble parts of the reject since only the TSS is measured.

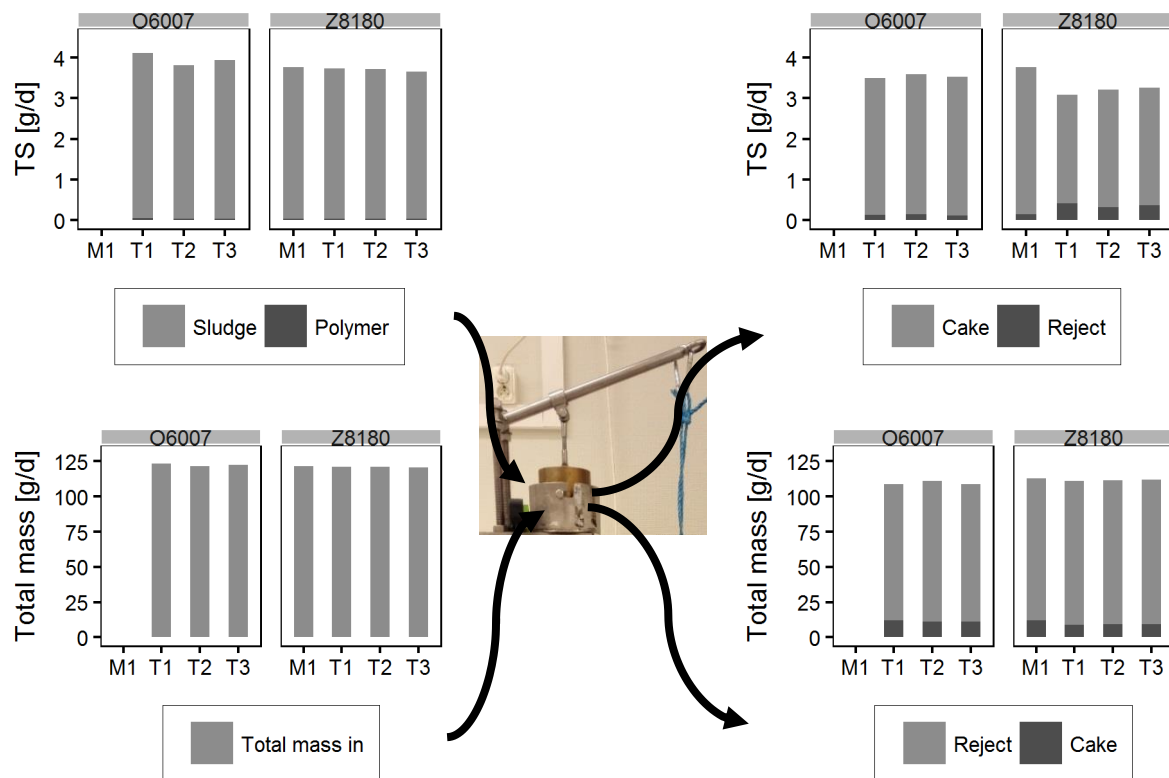


Fig. 14: TS and total mass balance of the dewatering process considering the TS of the sludge and polymer before the pressing as well as the TS of the cake and the TSS of the reject after the pressing. Considered time period: day 34 (stabilization of weight cake) to 77 (change of loading).

Tab. 13: Total mass and TS lost in pressing device for the different polymers for the time period between day 34 and 77 (where both polymers used).

	M1	T1	T2	T3
Weight loss O6007 [%]		11.9	8.9	11.1
Weight loss Z8180 [%]	7.7	9.4	9.4	8.8
TS loss O6007 [%]		15.2	5.8	10.4
TS loss Z8180 [%]	0.7	21.8	19.3	15.3

5.5 Influence of the Loading Scheme and Adaptability to Changes

It can be seen in Fig. 6 that the reactor needs some time to adapt to changes in loading. The gas volume produced on Monday after the reactor is not loaded on Sunday is approximately 75% of that of the other days with the same loading and even though the loading rate is increased to 150% on Friday compared to Thursday the gas production only rises to approximately 130%. Additionally, Vesilind (1974) emphasises the importance of uniform loading rates for a successful digester operation and recommends at least two loadings per day. It was therefore decided to start a new loading scheme on day 76 where the same amount is fed per week but in equally sized doses each day (see Tab. 3). This allows to test if a more stable operation is possible with less frequent changes in loading and how the reactors react to changes in loading schemes.

The change of the loading scheme results in a drop of the gas production in all reactors (see Fig. 38, appendix). Fig. 15 however clearly shows that reactor T1 recovers the fastest while the gas production of T3 is disturbed significantly. T3 also shows a comparable high rise in the VFA/alkalinity ratio while there is no effect visible for the reference reactor M1 (see Fig. 9 a)). After around 3 weeks the VFA/alkalinity ratio stabilizes again in T1-T3 but remains at a higher level than before due to both higher VFA and lower alkalinity values. This shows that the thermophilic reactors are much more sensitive to changes in the loading than the mesophilic ones. The more regular loading also influences the gas composition. Fig. 7 and Fig. 34 (appendix) show that the new loading scheme results in a bigger drop of the N_2 fraction to almost zero in the end of the cycle.

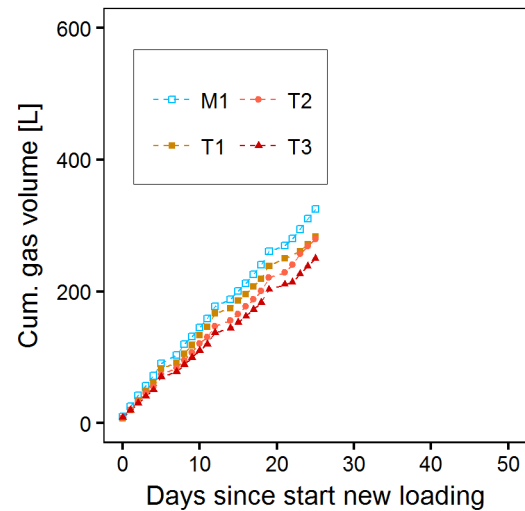


Fig. 15: Cumulated gas volume starting from 2017-07-10 when the new loading scheme was established.

5.6 Transferability of Pilot-Scale Results to the Full-Scale Plant

Fig. 18 compares the weekly VS removal in the mesophilic pilot-scale reactors to that of the full-scale plant at the same points in time. The continuous loading that is done in the full-scale plant seems to result in higher gas production rates (see also Fig. 6 that shows that also the mesophilic reactors need time to adapt to changes in loading). The CH_4 content in the full-scale biogas shows stable values between 61 and 63%. In M1 the average value is 59.5% but depends on when the collection of the gas is done while the values of the full-scale plant are measured online. The gas volumes cannot be compared directly since the results of the full-scale plant are given in NL while the values of the pilot-scale reactors are not recalculated (see chapter 4.2.1). The alkalinity of the full-scale plant is in the same range as in the pilot-scale

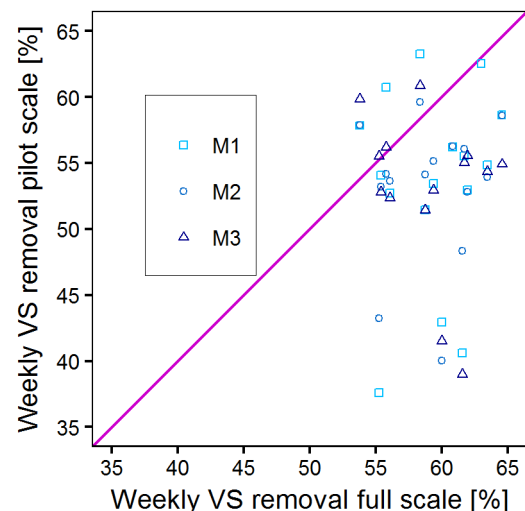


Fig. 16: VS removal averaged over a week in the full- and pilot-scale reactors at mesophilic temperature.

reactors. The buffering capacity for VFA increases during temperature changes should therefore be comparable. The NH_4^+ values in the reject are slightly higher in the pilot-scale reactors. Since the NH_4^+ values do not change with the temperature increase no big increase in the loading of the full-scale plant with NH_4^+ must be expected. Compared to other studies the OLR is high in both the full- and pilot-scale reactors so that the results of other authors can only be used with care.

There is a significant difference in the way the gas flow and gas composition changes over the cycle in the thermo- and mesophilic reactors. Apart from the lower overall gas production in the thermophilic reactors Fig. 17 also shows that M1 a significant peak after the loading while the thermophilic reactors show a more even gas flow over time. When the REVAQ requirements of 8 h digestion at 55°C have to be met two batch feedings per day of 12 hours length are realistic (since there must be time for loading and heating). This would however mean, that some sludge is already taken out before it could release most of its biogas potential. The CH_4 yield can also be expected to be lower for a thermophilic operation because as Fig. 7 clearly shows the CH_4 share still increases after the first 12 hours in T1 as opposed to M1. Zabranska (2002) and (2000) observed a higher process stability for higher OLR. However, Ghasimi et al. (2015) associated a faster increase to the peak with an operation below full conversion capacity. According to this, the OLR of the thermophilic reactors could not be increased since they do not show clear peaks and therefore operate at their limit.

Kim and Speece (2002a) and (2002b) compared the performance of reactors that were fed once per day or continuously at mesophilic and thermophilic temperatures. The reactor fed daily needed longer for stabilization to a given OLR and showed fluctuations in pH. The VS removal was comparable but the continuously fed reactor produced less biogas. However, the reactor failure occurred at more than twice the OLR in the continuously fed reactor (continuous feeding: 20 g/L d at mesophilic and thermophilic, daily feeding: 10 g/L d at mesophilic and 7.4 g/L d at thermophilic). Golkowska, Sibisi-Beierlein, and Greger (2012) also showed that the capability of the reactor to adapt to changes and the degradation rates increase with a higher loading frequency. With a continuous feeding scheme like in the full-scale reactor it can therefore be expected for thermophilic temperatures that higher OLR are possible and reactors are less sensitive to changes in loading than in the pilot-scale reactors.

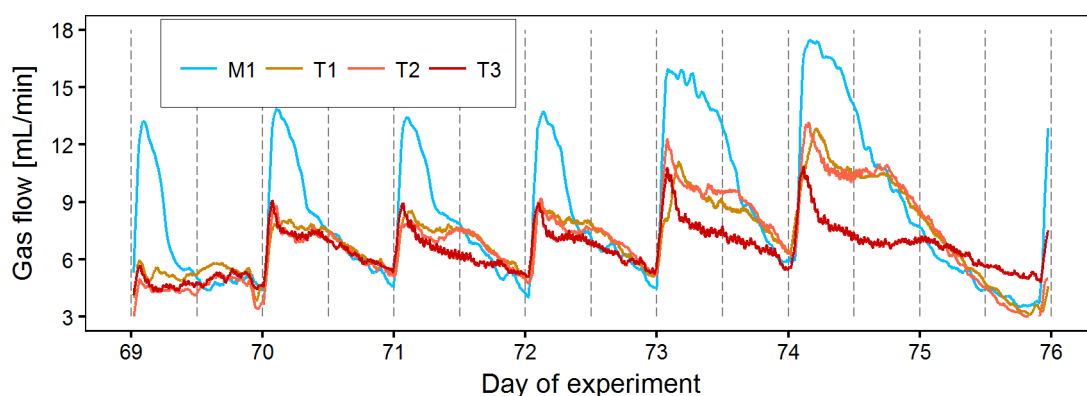


Fig. 17: Gas flow over the week before the loading scheme was changed. Data smoothed by moving average over 60 minutes for a better visibility. Original data: see Fig. 39, appendix. Vertical lines denote 12 h periods.

6 Conclusion and Outlook

Overall, it can be concluded that the temperature increase in the full-scale plant should be done as fast as possible based on the heating capacity while continuing with the loading. This corresponds to a mix between scenario T1 and T2. A slow and careful increase of the temperature seems to favour unstable behaviour and high VFA levels. During the transition phase, the VFA/alkalinity ratio should be analysed daily and used to decide on reduction of the loading after the critical temperature of 43°C is reached. HPr should be monitored closely during the transition phase since it is the most sensitive parameter indicating reactor upset. The buffering capacity of the reactor is sufficient to take up high VFA concentrations during the transition phase (as observed in T2). It is therefore probably sufficient to only reduce but not interrupt the loading of the digesters. This implicates that no big storing capacities for the sludge are necessary. If the personnel situation allows for it the time of the year with reduced sludge production (2016: July and August) should be chosen to increase the temperature. The OLR should be changed carefully and should be kept as constant as possible after stabilization since the thermophilic MO seem to react more sensitive to changes in loading and the bacterial community changes with the loading (Mao et al. 2015).

After the temperature increase a polymer with less charge density must be used. The right charge density and dose must be determined after the process stabilized and the loading is fixed since this can influence the dose. A higher loading could also improve the dewaterability TS values of the cake in the range of mesophilic sludge are realistic but the thermophilic sludge contains more fines that could be recycled with the reject. A much higher loading of the plant with NH_4^+ will probably not occur.

A lower amount of biogas must be expected. The CH_4 content of the gas will probably drop only for a limited period (2-3 weeks) and it should be burnable throughout the transition.

The pilot-scale experiments indicate a lower gas production at thermophilic temperatures. Each of the four steps of the AD could be the cause for this since they build on one another. The reason for the lower gas production could either be an inhibition or the absence of the MO adapted to the temperatures and capable of performing at a high rate:

- The high levels of HPr and iso-HBu could indicate that the acetogenesis (step 3) is inhibited by the high levels of HAc.
- That the CH_4 content of the biogas increases slowly and only after N_2 levels drop could indicate an inhibition of the methanogenesis (step 4) by FAN
- The rise of CO_2 before more CH_4 is produced could indicate that methanogens are present that can only perform at a low rate so that their substrate (CO_2) accumulates
- The concentrations of soluble COD that are high compared to the total COD could indicate that the steps after the hydrolysis work at low rates only

Ziemińska-Buczyńska et al. (2014) found that the biodiversity of the MO community decreased with the temperature increase and that the temperature shift had a stronger influence on the methanogens than on the rest of the MO. If the right MO are therefore not present in the mesophilic sludge or washed out this can have a big impact on the performance. To evaluate if the necessary MO are present it is essential to analyse the samples collected during the experiments for the composition of the microbial community.

For further analysis before conversion of the full-scale plant the following is suggested:

- Operation of the reactors with a loading based on VS that takes place at least two times per day on all days of the week to evaluate the effect of a more stable operation (closer to the continuous loading of the full-scale plant) on the gas production and VFA concentration
- Sludge biodegradability test to assess the degradability of the feed at different temperatures (refer to Tezel et al. (2014) and Palatsi et al. (2009))

- Methanogenic activity test to assess the maximal specific biogas production rate for the given MO community and the ability to handle extra load. Therefore add known amounts of VFA and analyse response of the reactors (refer to Zabranska et al. (2002) and Ahring et al. (2002))
- Calculate the net energy balance to analyse if a conversion reasonable from an energetic point

7 References

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Appendix

Additional Data and Plots

Tab. 14: Studies on strategies for converting mesophilic AD to thermophilic temperatures. Loading during transition: c = continuous, i = interrupted; Scale: p = pilot-scale, f = full-scale; Feed: p = primary sludge, s = secondary sludge. Empty cells if parameter not provided in study.

Study	Temp. change	Loading during transition	Scale	OLR [$\text{kg}_{\text{VS}}/\text{m}^3 \text{ d}$]	Feed	Feeding scheme	SRT [d]
(Boušková et al. 2005)	- Stepwise: 37, 42, 47, 51, 55 - Direct	c	p (2.5 L)	1.38	p (40%) & s (60%)	6/d	20
(Cavinato et al. 2013)	- Direct (pilot-scale) - 1.2°C/d (full-scale)	i	- p (380 L) - f (2000 m^3)	- p: 1.6 (start-up), 2.2 (final) - f: 1.28	s & biowaste		22
(Iranpour et al. 2002)	3°C/d	i	f (4500 m^3)		p (30%) & s (70%)	different	12 after stabilization
(Palatsi et al. 2009)	- Stepwise: 35, 43, 50, 55 - Direct	c	p (5 L)	1.29-1.73	p (50%) & s (50%)		22 d
(de la Rubia et al. 2005)	0.38°C/d until 45°C, 50, 52, 0.13°C/d until 55°C	i	p (150 L)	1.26 (mesophilic), 1.48 (thermophilic)	p & s	3/d	27
(Tezel et al. 2014)	3°C/d	c	p (3 L)	1.1	p (20%) & s (80%)	1/d	30
(Zabranska et al. 2002)	Stepwise with increments < 4°C		f (2x 4800 m^3 in series)				5.1 (first stage), 9 (both reactors)
(Ziemińska-Buczyńska et al. 2014)	3°C/d		P (200 L)		P (66%) & excess sludge from SBR (33%)		33

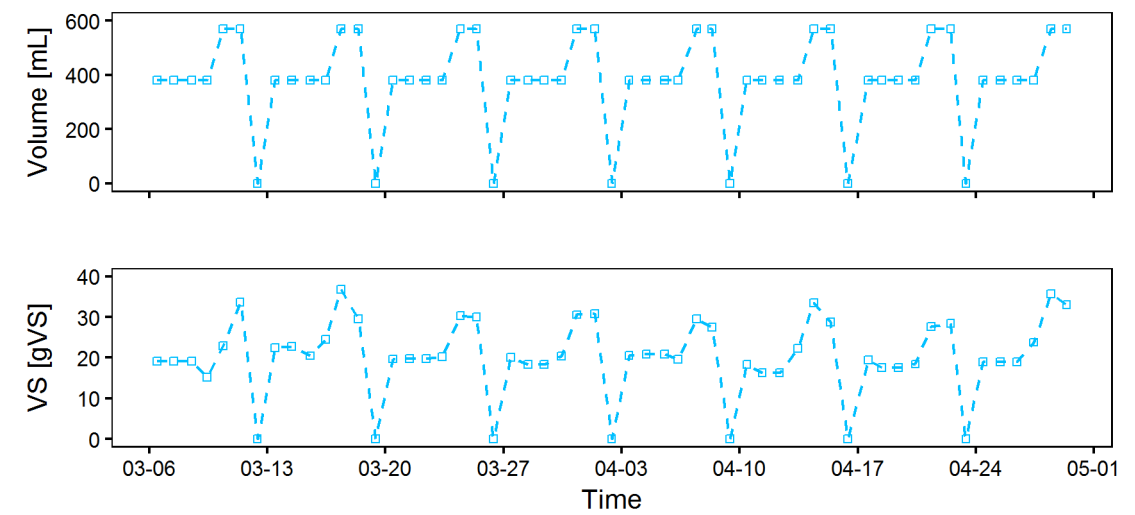


Fig. 18: Loading rates of reactor M1 based on volume and VS (OLR) during the mesophilic phase.

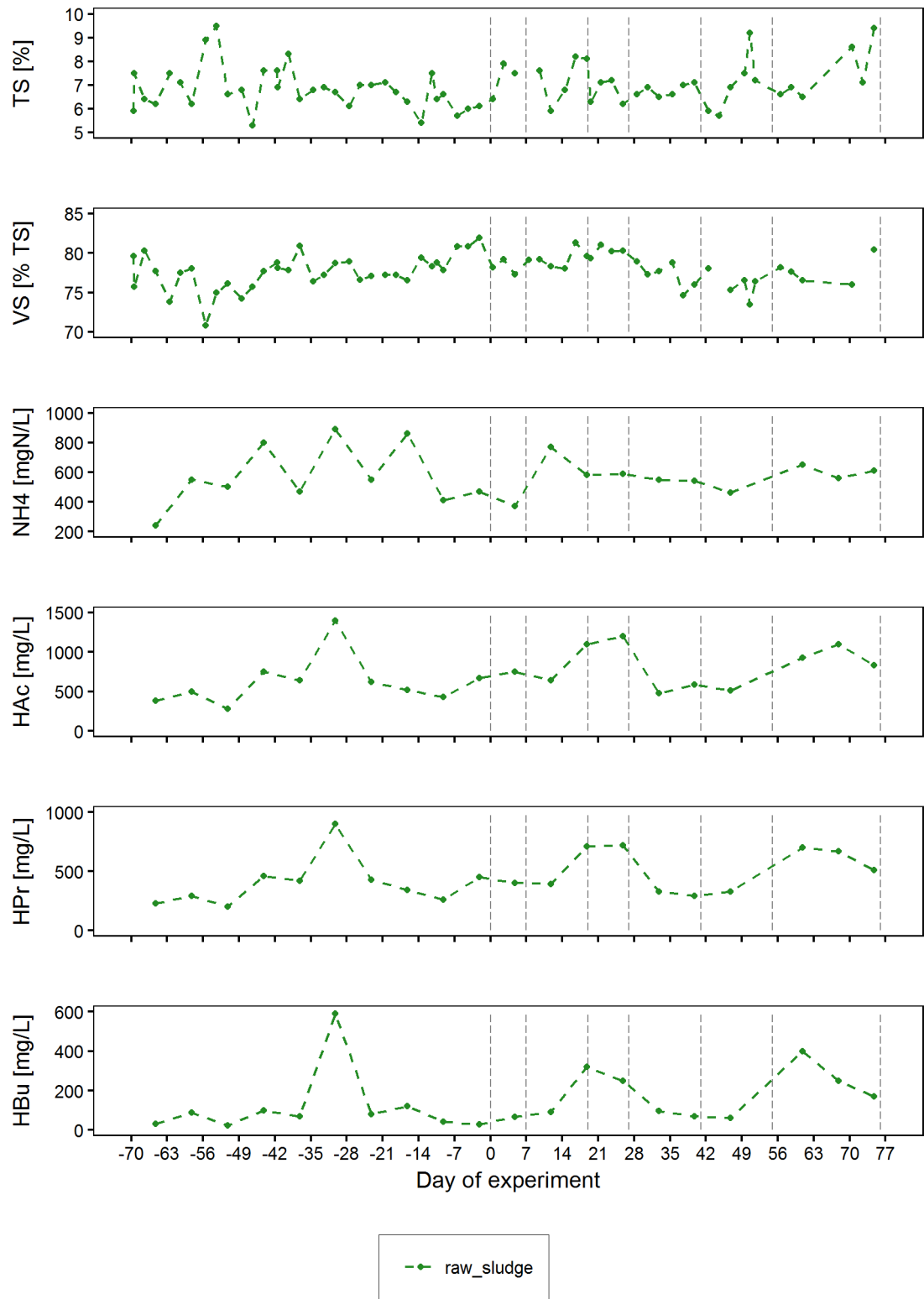


Fig. 19: Feed composition over the course of the experiments. HAc = acetic acid, HPr = propionic acid, HBu = butyric acid.

Tab. 15: Overview on the sampling scheme for the external analysis of sludge quality.

	Tue	Wed	Thu	Fri	Sat	Mon
Parameters analysed externally	TS, VS		TS, VS		TS VS, COD, COD _{filtr.} , NH ₄ ⁺ , VFA	

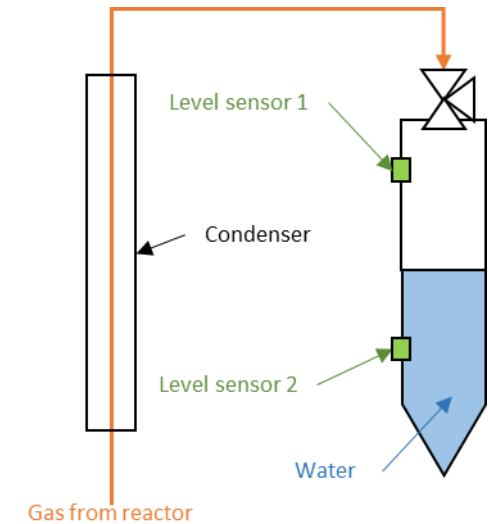


Fig. 20: Device for the measurement of gas flow according to (Belach Biotechnik AB 2011).

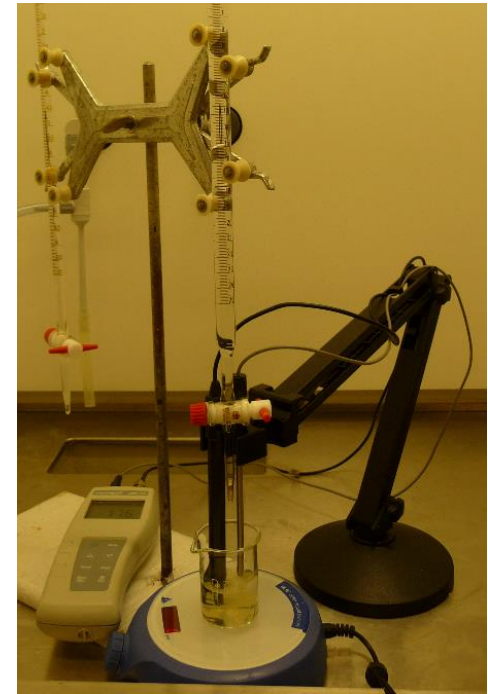


Fig. 21: Experimental setup for the determination of total alkalinity and VFA.



Fig. 22: Experimental setup for the determination of dewaterability (Yulang Guo 2017).

Tab. 16: Titration methods used for the different samples. 1 = according to first step of method development. 4 = according to final step of method development. Day = Day of experiment.

Date	Day	T1	T2	T3	M1	M2	M3
2017-04-21	-4	1	4	4	1	4	4
2017-04-24	-1	1	4	1	4	4	4
2017-04-26	1	1					
2017-04-27	2	1					
2017-04-28	3	1	4	4			
2017-04-29	4	1					
2017-05-01	6	1					
2017-05-02	7	1	4	4	3	3	3
2017-05-03	8	1					
2017-05-04	9	1					
2017-05-05	10	1	4	1			
2017-05-06	11	4					
2017-05-07	12	3					
2017-05-08	13	3					
2017-05-09	14	3	4	4	4	4	4
2017-05-10	15	4					
2017-05-11	16	4					
2017-05-12	17	4		4			
2017-05-13	18	4	4				
2017-05-14	19	4					
2017-05-15	20	4	4	4			
2017-05-16	21	4	4	4	4	4	4
2017-05-17	22	4	4	4			
2017-05-18	23	4	4	4			
2017-05-19	24	4	4	4			
2017-05-20	25	4	4	4			
2017-05-22	27	4	4	4			
2017-05-23	28	4	4	4	4	4	4
2017-05-24	29	4	4	4			
2017-05-25	30	4	4	4			
2017-05-26	31	4	4	4			
2017-05-27	32	4	4	4			
2017-05-29	34	4	4	4			
2017-05-30	35	4	4	4	4	4	4
2017-05-31	36	4	4	4			
2017-06-01	37	4	4	4			
2017-06-02	38	4	4	4			
2017-06-05	41	4	4	4	4	4	4
2017-06-06	42	4	4	4			
2017-06-07	43	4	4	4			
2017-06-08	44	4	4	4			
2017-06-09	45	4	4	4			
2017-06-10	46	4	4	4			
2017-06-12	48	4	4	4			
2017-06-13	49	4	4	4	4	4	4
2017-06-14	50	4	4	4			
2017-06-15	51	4	4	4	4		
2017-06-16	52	4	4	4	4		
2017-06-19	55	4	4	4			
2017-06-20	56	4	4	4			
2017-06-21	57	4	4	4	4	4	4
2017-06-22	58	4	4	4			

2017-06-23	59	4	4	4			
2017-06-24	60	4	4	4			
2017-06-26	62	4	4	4			
2017-06-27	63	4	4	4	4	4	4
2017-06-28	64	4	4	4			
2017-06-29	65	4	4	4			
2017-06-30	66	4	4	4			
2017-07-01	67	4	4	4			
2017-07-03	69	4	4	4			
2017-07-04	70	4	4	4	4		4
2017-07-05	71	4	4	4		4	
2017-07-06	72	4	4	4			
2017-07-07	73	4	4	4			
2017-07-10	76	4	4	4			
2017-07-11	77	4	4	4	4	4	4
2017-07-12	78	4	4	4			
2017-07-13	79	4	4	4			
2017-07-14	80	4	4	4	4		
2017-07-15	81	4	4	4			
2017-07-17	83	4	4	4			
2017-07-18	84	4	4	4		4	4
2017-07-20	86	4	4	4			
2017-07-21	87	4	4	4	4		
2017-07-25	91	4	4	4	4	4	4
2017-07-27	93	4	4	4			
2017-07-29	95	4	4	4			
2017-08-01	98	4	4	4	4		

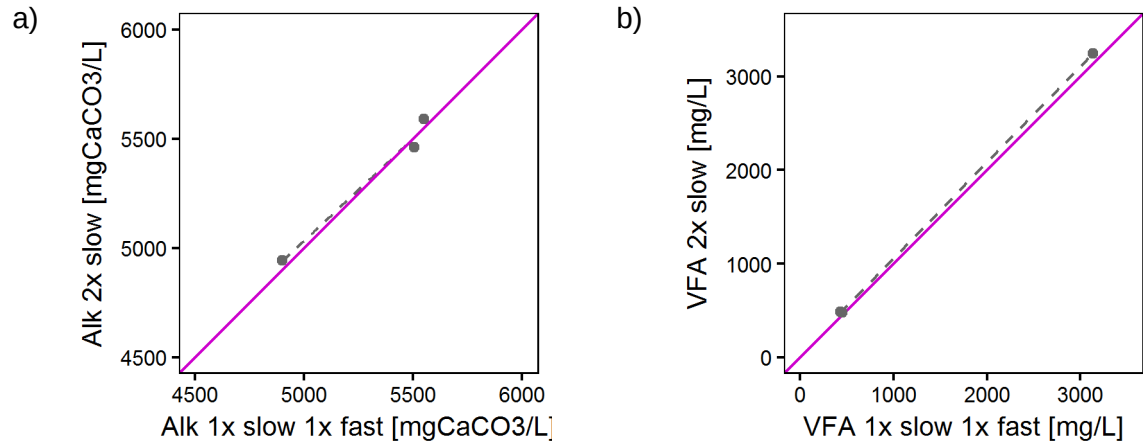


Fig. 23: Influence of the **centrifugation speed** on the alkalinity (a) and VFA (b) measurements by titration. Slow centrifugation = $4.4 \cdot 10^3$ rpm. fast centrifugation = $14.65 \cdot 10^3$ rpm. Each measurement as average of triplicates. Average CV: a) Values on x-axis: 0.8%. values on y-axis: 0.4%; b) Values on x-axis: 3.8%. values on y-axis: 3.8%.

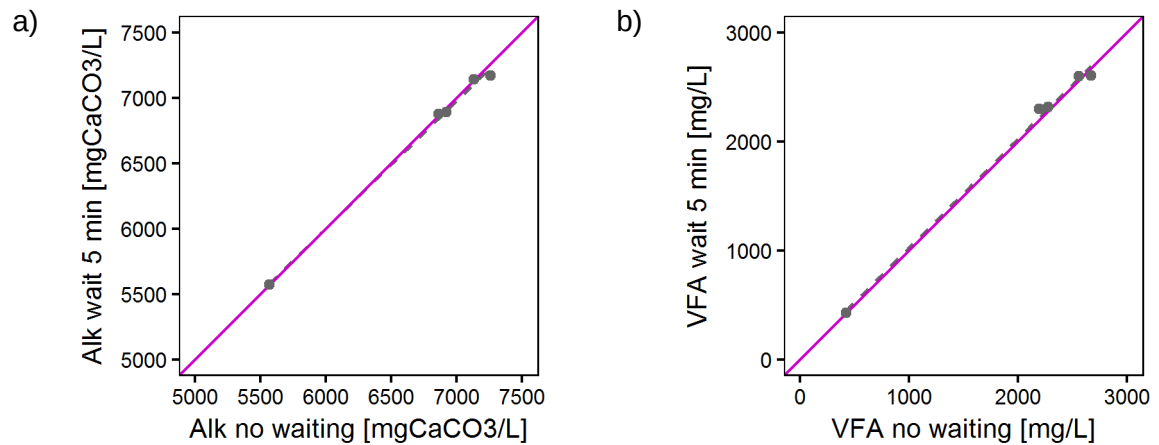


Fig. 24: Influence of the **adaption time of the pH probe before the start of measurements** on the alkalinity (a) and VFA (b) measurements by titration. Each measurement as average of duplicates or triplicates. Average CV: a) Values on x-axis: 0.7%. values on y-axis: 0.4%; b) Values on x-axis: 2.2%. values on y-axis: 2.0%.

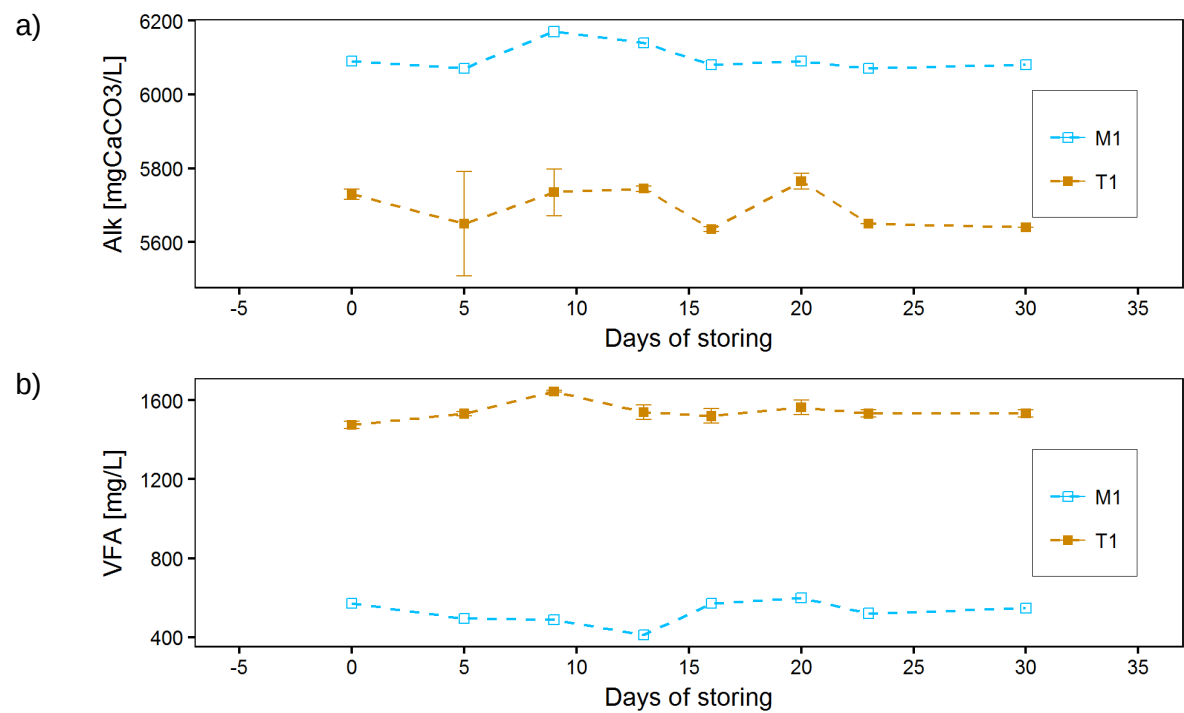


Fig. 25: Influence of **sample storing** on the alkalinity (a) and VFA (b) results obtained by titration. T1: each measurement as duplicate.

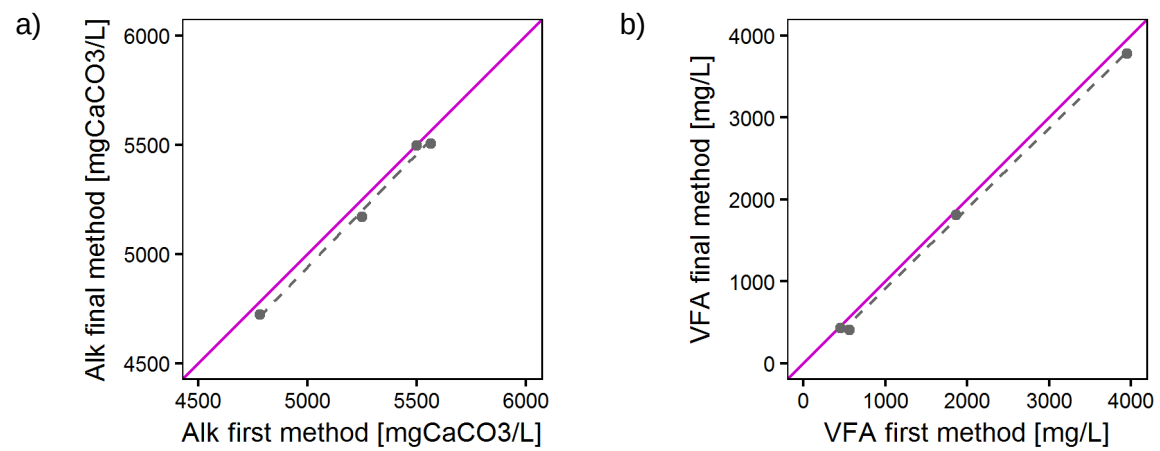


Fig. 26: Results of the measurement of the alkalinity (a) and VFA (b) in different samples with the parameters of the **first and last step of the method development**. Each measurement as average of duplicates or triplicates. Average CV: a) Values on x-axis: 0.2%. values on y-axis: 1.1%; b) Values on x-axis: 5.7%. values on y-axis: 4.9%.

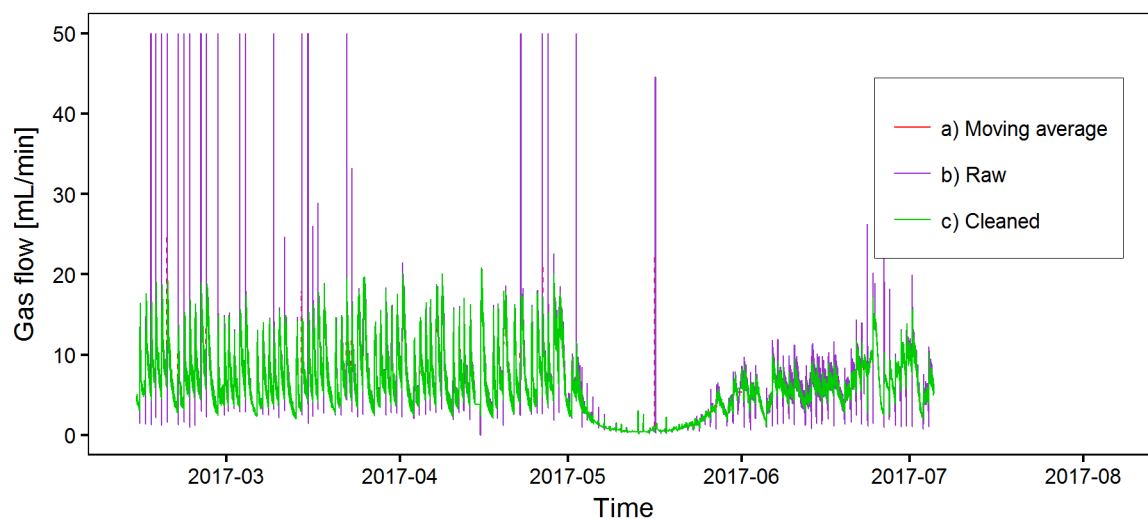
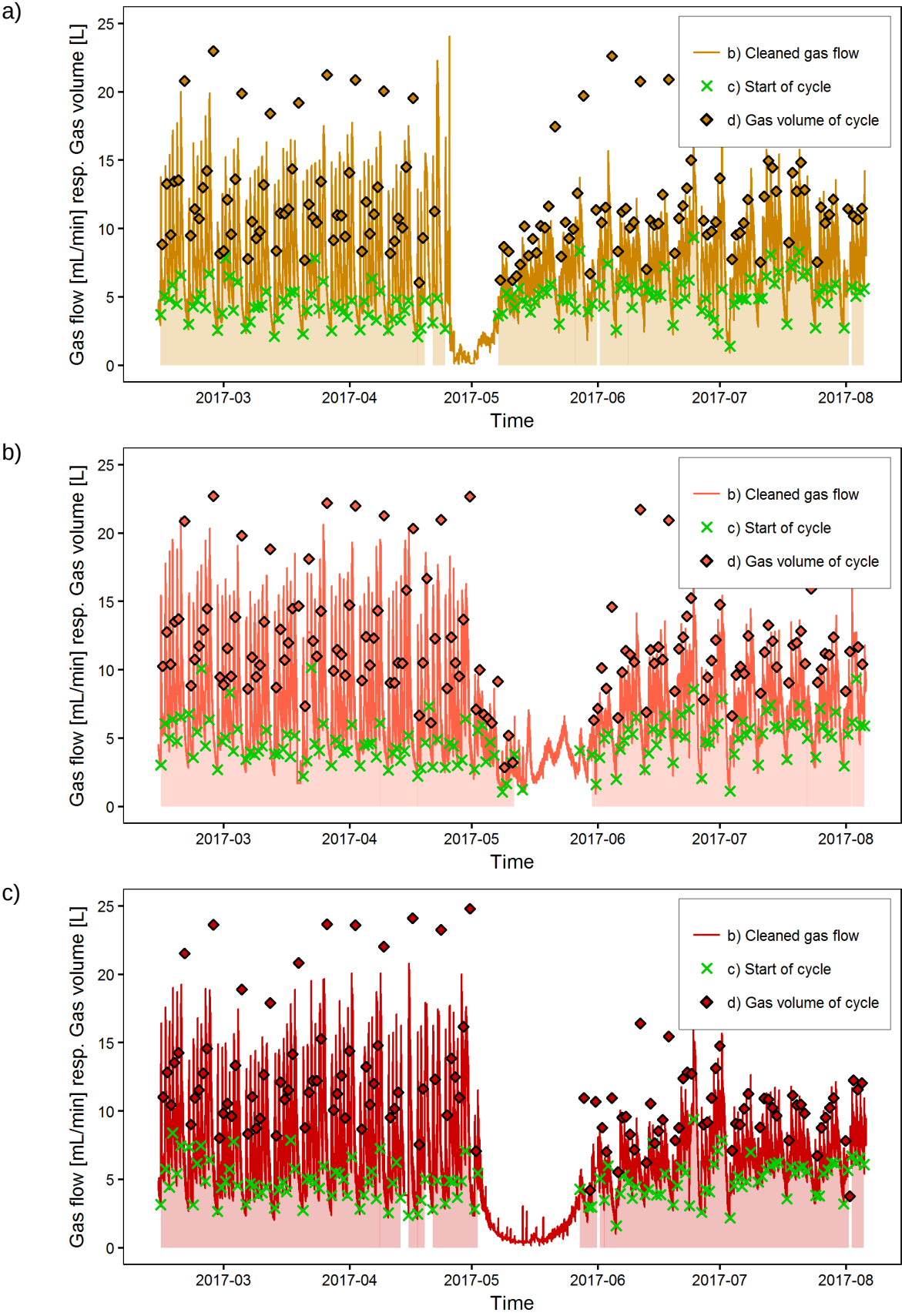
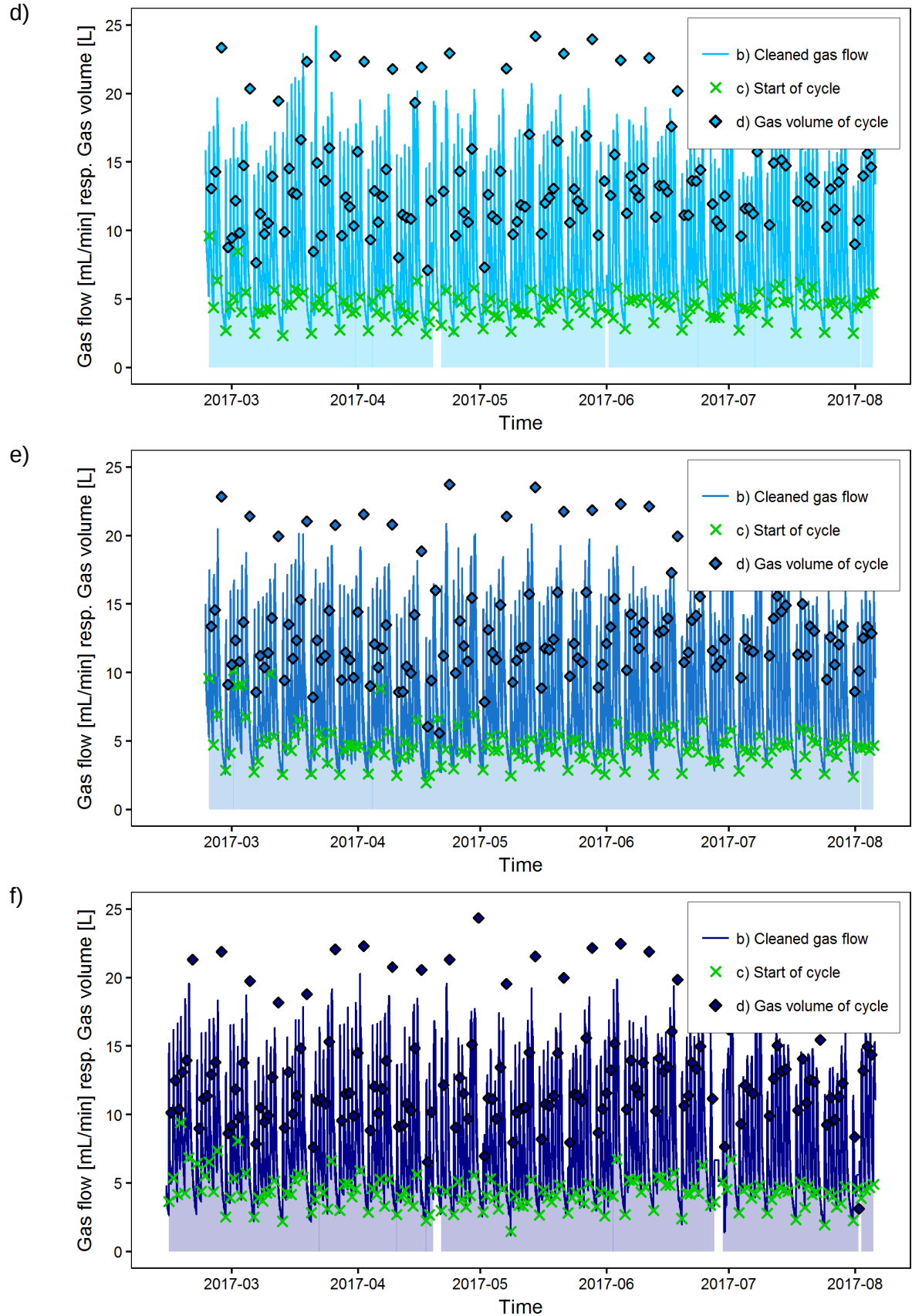


Fig. 27: Raw (purple) and corrected (green) gas flow data in the reactor T3.





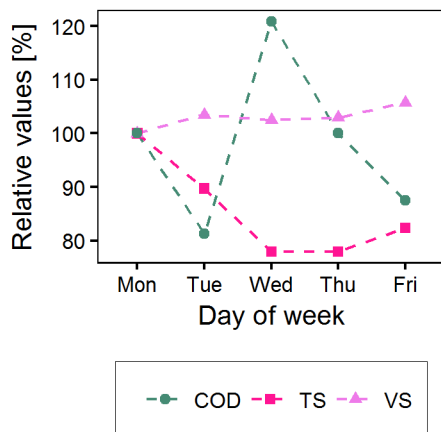


Fig. 29. Variation of measurements of sludge parameters over the course of a week for reactor T1.

Tab. 17: Values at the start of the cycle that are set to 100% in Fig. 7.

		Alk [mg _{CaCO3} /L]	CH ₄ [%]	CO ₂ [%]	N ₂ [%]	TS [%]	VFA [mg/L]
2017-06-15	M1	6030	46	33	16	4.07	720
	T1	6330	44	32	20	3.98	1633
2017-07-25	M1		48	30	17		
	T1		44	33	18		

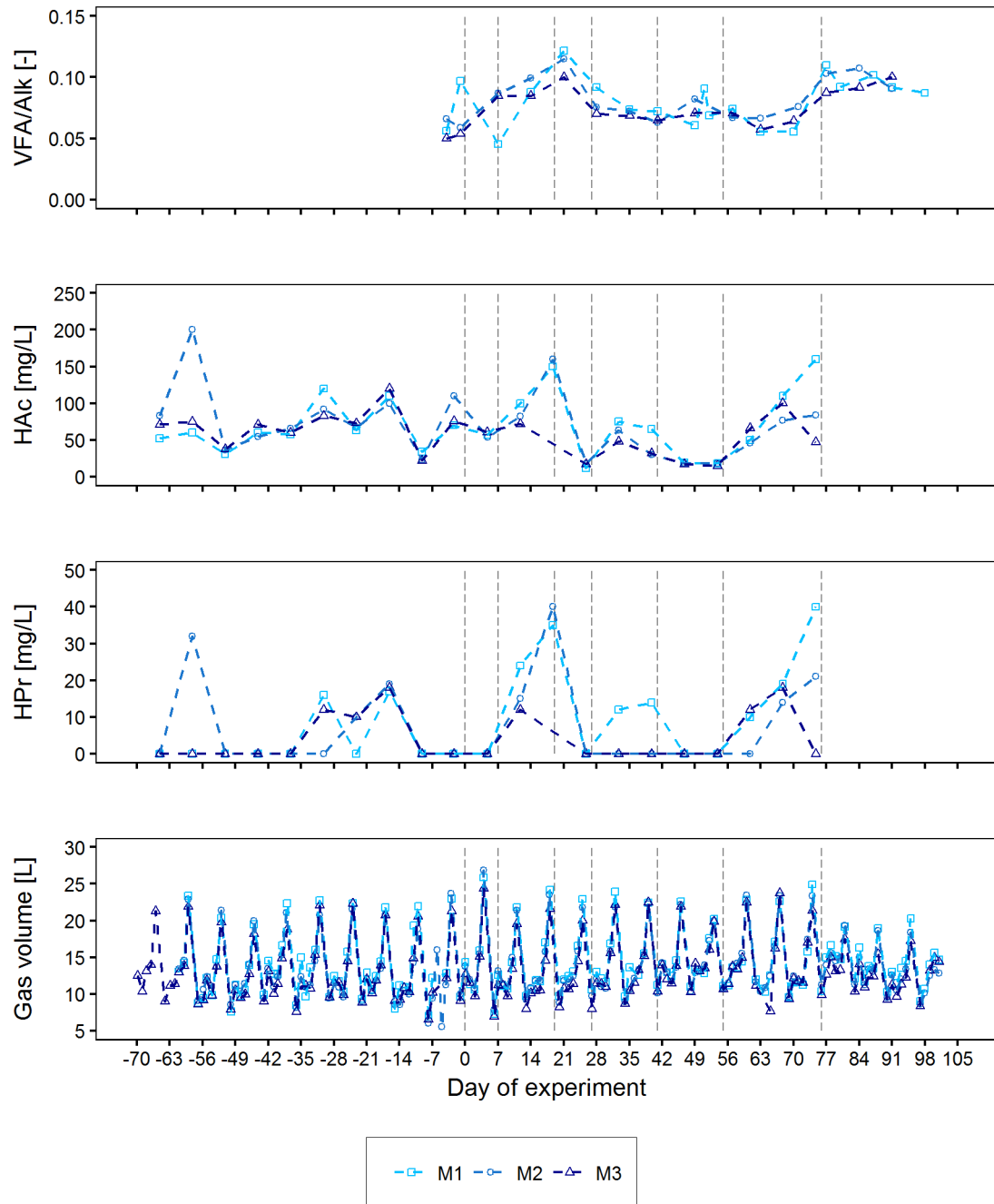


Fig. 30: Performance parameters in all mesophilic reactors over time.

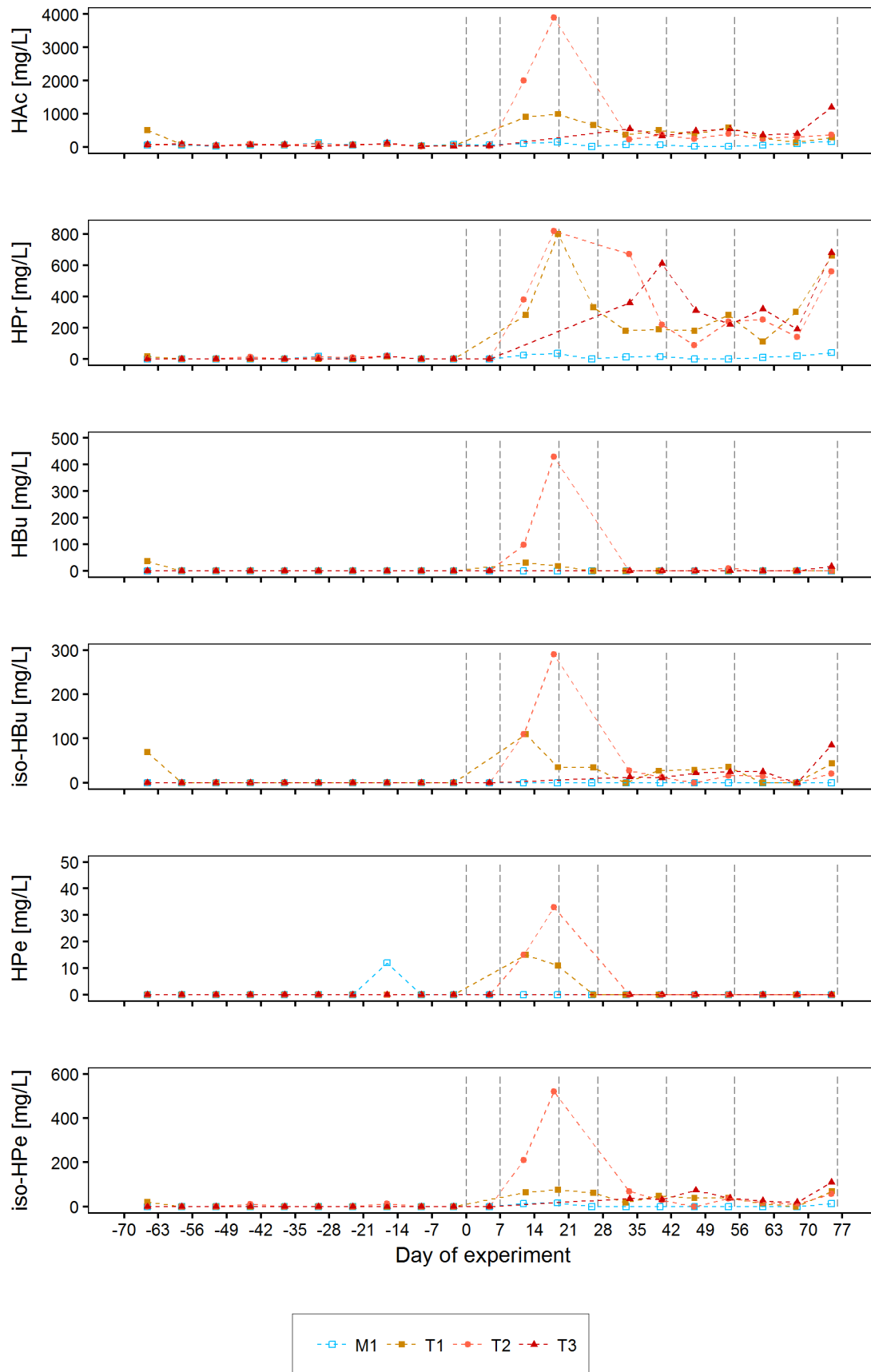


Fig. 31: Variation of the different VFA over the total length of experiments. HAc = acetic acid, HPr = propanoic acid, HBu = butyric acid, HPe = pentanoic acid. Hexanoic and heptanoic acid not shown because always < 10 mg/L.

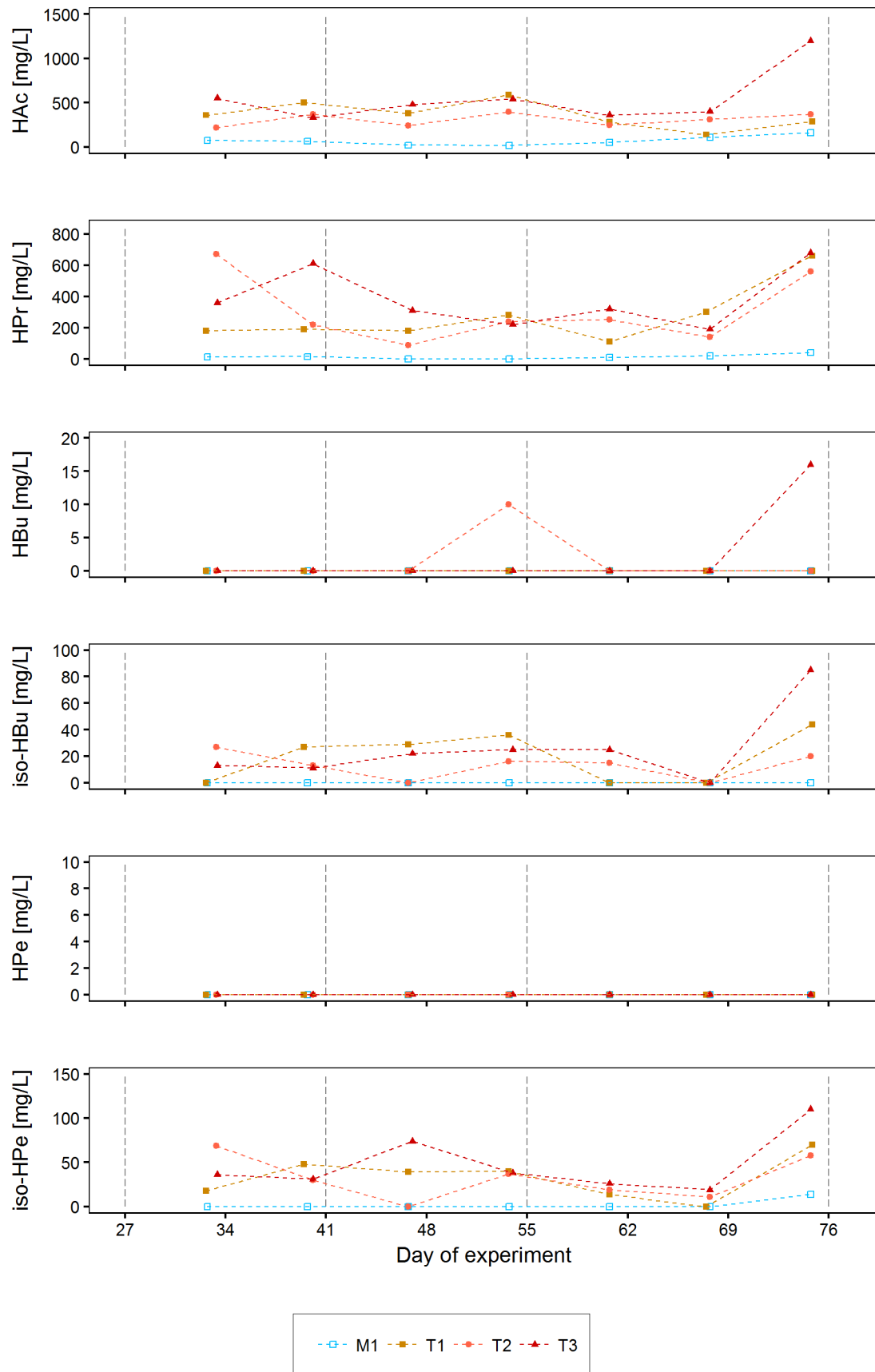


Fig. 32: Variation of the different VFA starting from day 27 of the experiments. HAc = acetic acid, HPr = propanoic acid, HBu = butyric acid, HPe = pentanoic acid. Hexanoic and heptanoic acid not shown because always < 10 mg/L.

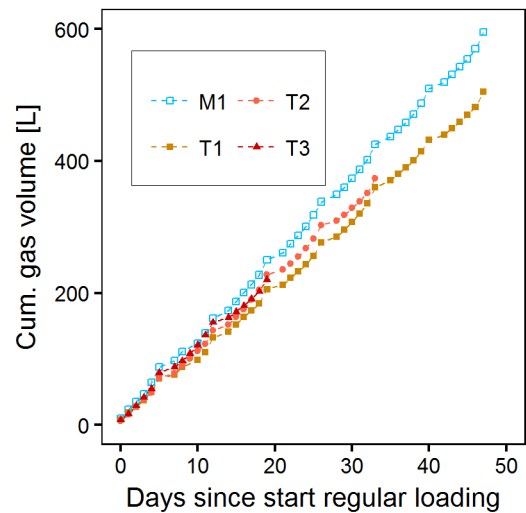


Fig. 33. Cumulated gas volume after reactor is loaded according to the regular loading scheme again. For M1 day 0 chosen as the same as for T1.

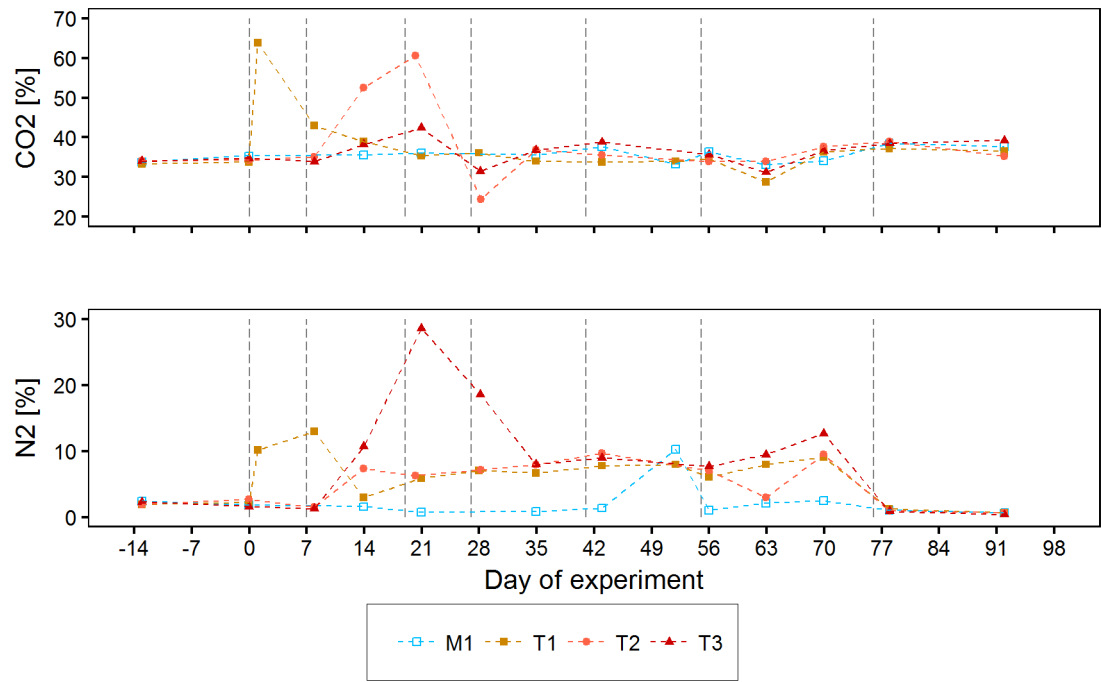


Fig. 34: CO₂ and N₂ in the biogas during the course of the experiments. Time phases like in Fig. 9.

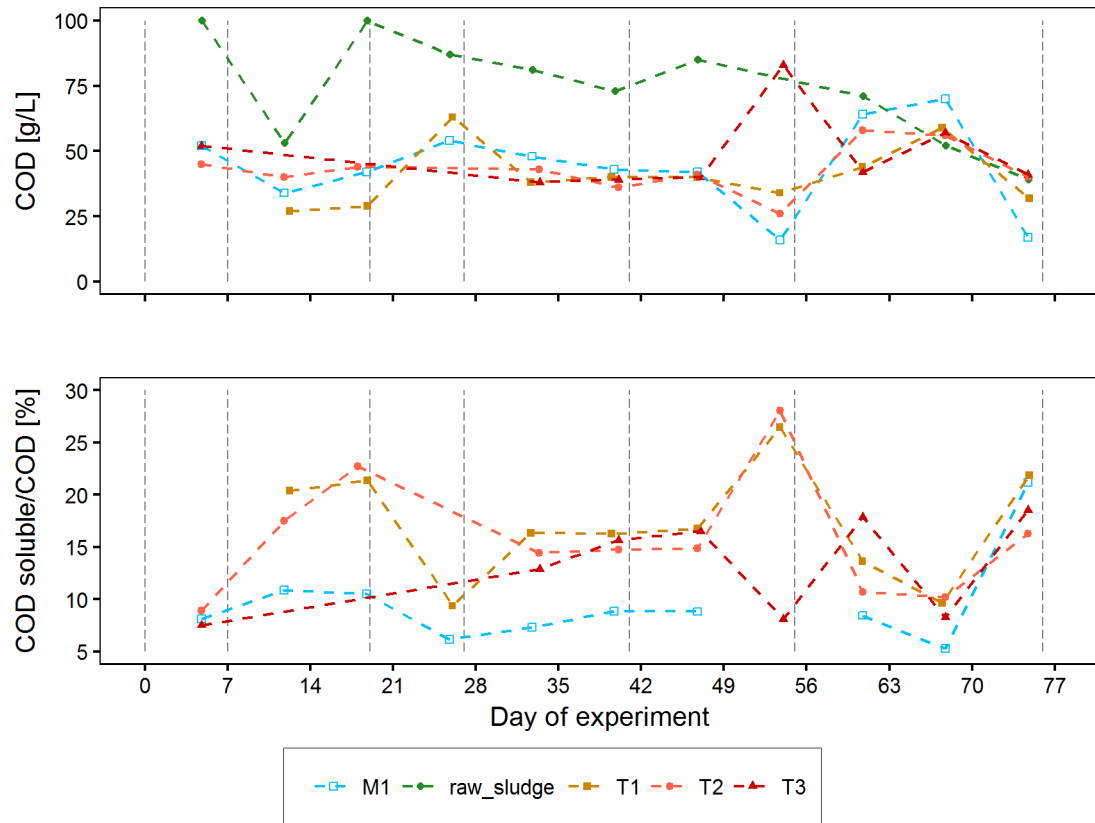


Fig. 35: COD in the raw digested sludge and the share of soluble COD on the total COD in the digested sludge over time.

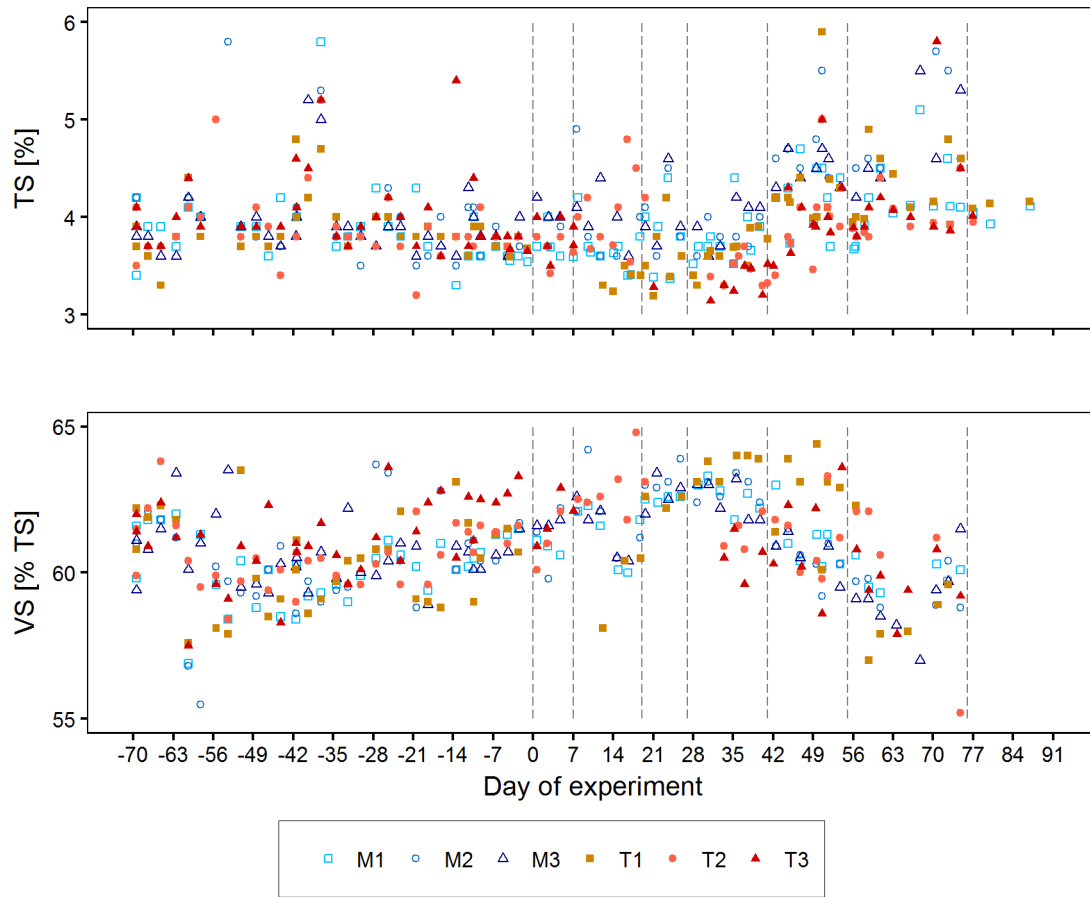


Fig. 36: TS and VS values over the time of the experiments. TS values above 6% cut off for better visibility since there is a low possibility that they are realistic. Around day 65 has outliers in TS measurements due to removal of cake formed on stirrer (values up to 8.3% which are cut off).

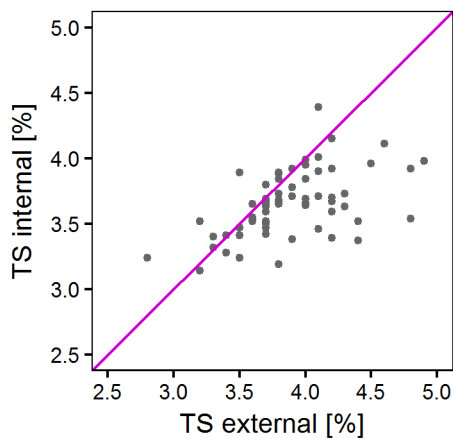


Fig. 37: TS measurements in the external lab and own measurements use to determine the polymer dose for dewatering.

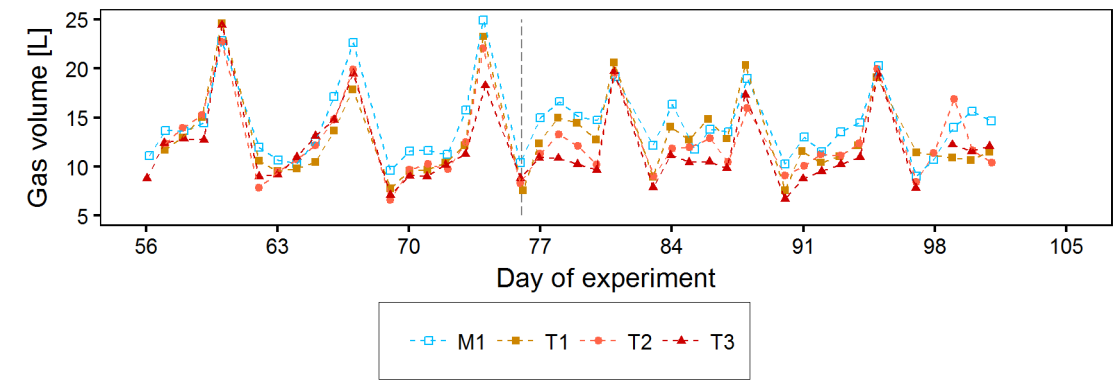


Fig. 38: Gas volume before and after the change of the feeding scheme.

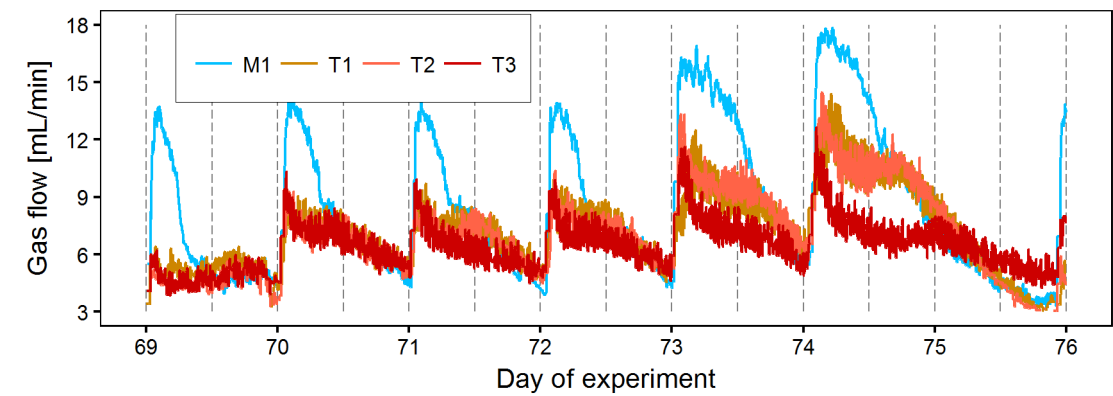


Fig. 39: Gas flow over one week. Data not smoothed.

Tab. 18: Amount of sludge taken out and loaded and sludge sent to external lab. Day = Day of experiment.

Day	Date	Volume in and out [mL]						Volume for external analysis [mL]					
		T1	T2	T3	M1	M2	M3	T1	T2	T3	M1	M2	M3
-4	21.04.2017	570	570	570	570	570	570	349	349	349	349	459	459
-3	22.04.2017	570	570	570	570	570	570	569	569	569	569	569	569
-2	23.04.2017												
-1	24.04.2017	380	380	380	380	380	380	155	155	155	155	265	265
0	25.04.2017	15	380	380	380	380	380	0	155	155	155	155	140
1	26.04.2017	15	380	380	380	380	380	0	265	265	265	265	250
2	27.04.2017	15	380	380	380	380	380	0	265	265	265	265	250
3	28.04.2017	15	570	570	570	570	570	0	235	235	345	455	440
4	29.04.2017	15	570	570	570	570	570	0	455	455	455	455	440
5	30.04.2017												
6	01.05.2017	15	380	380	380	380	380	0	265	265	265	265	250
7	02.05.2017	15	380	380	380	380	380	0	45	45	45	155	140
8	03.05.2017	15	380	0	380	380	380	0	265	0	265	265	250
9	04.05.2017	15	380	0	380	380	380	0	265	0	265	265	250
10	05.05.2017	115	570	15	570	570	570	0	235	0	345	455	440
11	06.05.2017	230	570	0	570	570	570	100	455	0	455	455	455
12	07.05.2017												
13	08.05.2017	380	380	0	380	380	380	155	265	0	265	265	265
14	09.05.2017	230	380	15	380	380	380	0	45	0	45	155	140
15	10.05.2017	230	380	0	380	380	380	0	265	0	265	265	265
16	11.05.2017	230	380	0	380	380	380	5	265	0	265	265	265
17	12.05.2017	380	570	15	570	570	570	0	235	0	345	455	440
18	13.05.2017	380	570	0	570	570	570	215	405	0	455	455	455
19	14.05.2017												
20	15.05.2017	380	15	115	380	380	380	140	0	0	265	265	250
21	16.05.2017	380	15	115	380	380	380	125	0	0	45	155	130
22	17.05.2017	380	15	15	380	380	380	235	0	0	265	265	235
23	18.05.2017	380	15	15	380	380	380	250	0	0	265	265	235
24	19.05.2017	380	15	15	570	570	570	125	0	0	345	455	425
25	20.05.2017	380	15	15	570	570	570	250	0	0	455	455	425
26	21.05.2017												
27	22.05.2017	380	15	15	380	380	380	250	0	0	265	265	235
28	23.05.2017	380	15	15	380	380	380	140	0	0	45	155	125
29	24.05.2017	380	15	115	380	380	380	250	0	0	265	265	250
30	25.05.2017	380	15	115	380	380	380	250	0	0	265	265	250
31	26.05.2017	570	115	115	570	570	570	330	0	0	345	455	455
32	27.05.2017	570	230	230	570	570	570	440	100	100	455	455	455
33	28.05.2017												
34	29.05.2017	380	380	380	380	380	380	210	210	250	265	265	265
35	30.05.2017	380	230	380	380	380	380	140	0	140	140	250	250
36	31.05.2017	380	230	380	380	380	380	250	100	30	45	265	265
37	01.06.2017	380	230	230	380	380	380	250	100	100	265	265	265
38	02.06.2017	570	230	230	570	570	570	330	0	0	345	455	455
39	03.06.2017	570	230	230	570	570	570	455	115	115	455	455	455
40	04.06.2017												
41	05.06.2017	380	380	380	380	380	380	140	140	140	250	250	250
42	06.06.2017	380	380	380	380	380	380	250	250	250	265	265	265

43	07.06.2017	380	380	230	380	380	380	140	140	0	265	265	265
44	08.06.2017	380	380	230	380	380	380	250	250	100	265	265	265
45	09.06.2017	570	570	380	570	570	570	330	330	140	345	455	455
46	10.06.2017	570	570	380	570	570	570	440	440	250	455	455	455
47	11.06.2017												
48	12.06.2017	380	380	380	380	380	380	250	250	250	265	265	265
49	13.06.2017	380	380	380	380	380	380	140	140	140	140	250	250
50	14.06.2017	380	380	230	380	380	380	250	250	100	265	265	265
51	15.06.2017	380	380	230	380	380	380	235	250	100	250	265	265
52	16.06.2017	570	570	380	570	570	570	240	330	140	255	455	455
53	17.06.2017	570	570	380	570	570	570	440	440	250	455	455	455
54	18.06.2017												
55	19.06.2017	380	380	380	380	380	380	250	250	250	265	265	265
56	20.06.2017	380	380	380	380	380	380	140	140	140	155	265	265
57	21.06.2017	380	380	380	380	380	380	130	250	250	115	250	250
58	22.06.2017	380	380	380	380	380	380	140	140	140	155	265	265
59	23.06.2017	570	570	570	570	570	570	440	440	440	455	455	455
60	24.06.2017	570	570	570	570	570	570	440	440	440	455	455	455
61	25.06.2017												
62	26.06.2017	380	380	380	380	380	380	250	250	250	265	265	265
63	27.06.2017	380	380	380	380	380	380	140	140	140	140	250	250
64	28.06.2017	380	380	380	380	380	380	250	250	250	265	265	265
65	29.06.2017	380	380	380	380	380	380	250	250	250	265	265	265
66	30.06.2017	570	570	570	570	570	570	330	330	330	345	455	455
67	01.07.2017	570	570	570	570	570	570	440	440	440	455	455	455
68	02.07.2017												
69	03.07.2017	380	380	380	380	380	380	140	250	250	265	265	265
70	04.07.2017	380	380	380	380	380	380	30	140	140	140	250	250
71	05.07.2017	380	380	380	380	380	380	140	250	250	265	250	265
72	06.07.2017	380	380	380	380	380	380	140	250	250	265	265	265
73	07.07.2017	570	570	570	570	570	570	220	330	330	345	455	455
74	08.07.2017	570	570	570	570	570	570	330	440	440	455	455	455
75	09.07.2017												
76	10.07.2017	380	380	380	380	380	380	250	250	250	265	265	265
77	11.07.2017	445	445	445	445	445	445	205	205	205	205	315	315
78	12.07.2017	500	500	500	500	500	500	370	370	370	385	385	385
79	13.07.2017	445	445	445	445	445	445	315	315	315	330	330	330
80	14.07.2017	445	445	445	445	445	445	205	205	315	95	330	330
81	15.07.2017	445	445	445	445	445	445	315	315	315	330	330	330
82	16.07.2017												
83	17.07.2017	445	445	445	445	445	445	315	315	315	330	330	330
84	18.07.2017	445	445	445	445	445	445	205	205	205	205	315	315
85	19.07.2017	445	445	445	445	445	445	330	330	330	330	330	330
86	20.07.2017	445	445	445	445	445	445	315	315	315	330	330	330
87	21.07.2017	445	445	445	445	445	445	205	205	315	220	330	330
88	22.07.2017	445	445	445	445	445	445	330	330	330	330	330	330
89	23.07.2017												
90	24.07.2017	445	445	445	445	445	445	330	330	330	330	330	330
91	25.07.2017	445	445	445	445	445	445	315	315	315	315	315	315
92	26.07.2017	445	445	445	445	445	445	330	330	330	330	330	330

93	27.07.2017	445	445	445	445	445	445	315	315	315	330	330	330
94	28.07.2017	445	445	445	445	445	445	330	330	330	330	330	330
95	29.07.2017	445	445	445	445	445	445	315	315	315	330	330	330
96	30.07.2017												
97	31.07.2017	445	445	445	445	445	445	330	330	330	330	330	330
98	01.08.2017	445	445	445	445	445	445	205	315	315	205	315	315
99	02.08.2017	445	445	445	445	445	445	315	315	315	330	330	330
100	03.08.2017	445	445	445	445	445	445	315	315	315	330	330	330
101	04.08.2017	445	445	445	445	445	445	315	315	315	330	330	330
102	05.08.2017	445	445	445	445	445	445	315	315	315	330	330	330

R Codes

Tab. 19: Overview on the scripts available for data handling and plotting.

Script	Functions	Description
eval.R		Control code to: • set working directory and load packages and theme (always) • read and clean raw data (for new data only) • load data of specified time period (if data read in already) • set standard colours and symbols for the different reactors (always)
read-and_clean_pilot_scale.R	read_and_clean_pilot	Code for reading in all data of the pilot-scale reactors, cleaning of the gas sensor data and calculation of the gas volume. Generates 1 data.frame for each reactor and the raw sludge (feed) containing all measured variables. Generates 1 data.frame that contains units for all reactors. Output saved in R-files: • data_pilot_scale_raw.R • data_pilot_scale_cleaned.R Plots generated: • Cleaning of the gas flow data • Calculation of the gas volume
read_full_scale.R	read_full	Code for reading in data from the full-scale plant Output saved in R-file: data_full_scale.R
load.R	load_data	Code for extracting a data period specified by user from the complete data set so that resulting data.frames only contain specified dates
plot_overview.R	plot_same_x_axis	General code to generate plots with time on the x-axis. Can do facetting (splitting data into categories that are plotted separately), set number of plots and the reactors per plot. User can customize appearance of plot by many variables (for details see description in code header).
plot_using_plot_overview.R (not shown)		Code that calls the function plot_same_x_axis with different parameters

plot_mass_balances.R (not shown)		Code for reading out all data needed for mass balances and save it in data_pilot_scale_mass_balance.R. Generates the plots of the mass balances
plot_several_points_in_cycle_and_week.R (not shown)		Generates plots that show how parameters vary over the week and cycle and compares the gas production on different weekdays
plot_titration.R		Code for generating the plots that evaluate the influencing factors of the titration method development.
plot_others.R (not shown)		Code for doing plots that are not covered by the other scripts: • Gas volume mesophilic vs. thermophilic • Gas volume for each weekday • Variation over week • Variation over cycle • Cumulative gas volume
theme.R	theme_own	Contains the default settings that are used for all plots (e.g., colour of background)

read_and_clean_pilot_scale.R

Created: Tineke Bittlingmayer (bit), May 2017

Last update: bit, 2017-08-08

=====

Code for reading in all data of the pilot-scale reactors,

cleaning of the gas sensor data and calculation of the gas volume

=====

=====

read_and_clean_pilot = function(factor_lower, factor_upper, width_integration){

 colour_general <- c(M1 = "deepskyblue1", M2 = "dodgerblue3", M3 = "blue4",

 T1 = "orange3", T2 = "tomato", T3 = "red3",

 raw_sludge = "forestgreen") # in plots picks right name

=====

files_to_read <- c("raw_sludge", "T1", "T2", "T3", "M1", "M2", "M3")

for (i in 1:length(files_to_read)){

 # =====

 # Read out data of type 1 (all reactors saved in 1 file)

 # =====

 # -----

 # Read out feeding

 # -----

if(! files_to_read[[i]] == "raw_sludge"){

 sourcename <- paste(path, "/Data/Data_Pilot_scale/Lab_analysis_external/Feeding.csv", sep = "")

 temp_data <- read.csv(sourcename, header = T, sep = ";", fill = T, stringsAsFactors = F)

 # assign 10:00 to all dates

 temp_data\$Time <- as.POSIXct(temp_data\$Time, format = "%d.%m.%Y", tz = "Etc/GMT+1")+ (60*60*10)

 feeding_position <- vector()

 # to calculate the TS and VS of the feeding using measurements in raw sludge

for (j in 1:nrow(temp_data)){

 time_diff <- abs(temp_data\$Time[j]-raw_sludge\$Time)

 time_min <- min(time_diff, na.rm = T)

 feeding_position[j] <- which(time_diff == time_min)

 }

temp_data_feeding_volume <- data.frame(temp_data\$Time,

```

      temp_data[files_to_read[i]])
temp_data_feeding_TS <- data.frame(temp_data$Time,
      temp_data[files_to_read[i]]*raw_sludge$TS[feeding_position]/100)
temp_data_feeding_VS <- data.frame(temp_data$Time,
      temp_data[files_to_read[i]]*raw_sludge$TS[feeding_position]/100*
      raw_sludge$VS[feeding_position]/100)

colnames(temp_data_feeding_volume) <- c("Time", "Feeding_volume")
colnames(temp_data_feeding_TS) <- c("Time", "Feeding_TS")
colnames(temp_data_feeding_VS) <- c("Time", "Feeding_VS")

temp_data_feeding_volume$Cat <- "Feeding"
temp_data_feeding_volume <- temp_data_feeding_volume[, c(1, ncol(temp_data_feeding_volume),
      3:ncol(temp_data_feeding_volume)-1)]
temp_data_feeding_TS$Cat <- "Feeding"
temp_data_feeding_TS <- temp_data_feeding_TS[, c(1, ncol(temp_data_feeding_TS),
      3:ncol(temp_data_feeding_TS)-1)]
temp_data_feeding_VS$Cat <- "Feeding"
temp_data_feeding_VS <- temp_data_feeding_VS[, c(1, ncol(temp_data_feeding_VS),
      3:ncol(temp_data_feeding_VS)-1)]

temp_units_feeding_volume <- data.frame(V1 = "mL")
colnames(temp_units_feeding_volume) <- "Feeding_volume"

temp_units_feeding_TS <- data.frame(V1 = "gTS")
colnames(temp_units_feeding_TS) <- "Feeding_TS"

temp_units_feeding_VS <- data.frame(V1 = "gVS")
colnames(temp_units_feeding_VS) <- "Feeding_VS"
}

# -----
# Read out weight factors for results of external lab analysis
# -----
sourcename <- paste(path, "/Data/Data_Pilot_scale/Lab_analysis_external/Weights.csv", sep = "")
weight <- read.csv(sourcename, header = T, sep = ";", fill = T, stringsAsFactors = F)
weight$Time <- as.POSIXct(weight$Time, format = "%d.%m.%Y", tz = "Etc/GMT+1")

```

```

# =====
# Read out data of type 2 (1 file for each reactor)
# =====
name_read <- c("Titration", "Gas", "Dewatering")
name_assign <- c("titration", "gas", "dewatering")

if(! files_to_read[[i]] == "raw_sludge"){
  for (j in 1:length(name_read)){
    if(name_read[j] == "Gas"){
      sourcename <- paste(path, "/Data/Data_Pilot_scale/Lab_analysis_external/",
                          name_read[j], ".csv", sep = "")
    } else {
      sourcename <- paste(path, "/Data/Data_Pilot_scale/Lab_analysis_own/",
                          name_read[j], ".csv", sep = "")
    }

    temp_header <- read.csv(sourcename, nrow = 1, header = F, sep = ";", dec = ",",
                           fill = T, stringsAsFactors = F)

    temp_data <- read.csv(sourcename, skip = 2, header = F, sep = ";", dec = ",",
                         fill = T, stringsAsFactors = F)

    colnames(temp_data) <- temp_header

    if (name_read[j] == "Titration"){
      # sometimes measured before feeding -> then keep time, else replace with 10:00
      temp_data$Hour[which(temp_data$Hour == "")] <- "10:00:00"
      temp_data <- unite(temp_data, "Time", c(Time, Hour), sep = " ")
      temp_data$Time <- as.POSIXct(temp_data$Time, format = "%d.%m.%Y %H:%M", tz = "Etc/GMT+1")

      temp_data$"VFA_Alkalinity" <- temp_data$VFA/temp_data$Alkalinity
    } else {
      if (name_read[j] == "Gas"){
        time_start <- unite(temp_data, "Time_start", c(Time, Time_start), sep = " ")
        time_start <- time_start[which(colnames(time_start) == "Time_start")]
        time_start$Time_start <- as.POSIXct(time_start$Time_start, format = "%d.%m.%Y %H:%M", tz = "Etc/GMT+1")

        time_end <- unite(temp_data, "Time_end", c(Time, Time_end), sep = " ")

```



```

time_end <- time_end[which(colnames(time_end) == "Time_end")]
time_end$Time_end <- as.POSIXct(time_end$Time_end, format = "%d.%m.%Y %H:%M", tz = "Etc/GMT+1")

time_average <- (time_end$Time_end - time_start$Time_start)/2
temp_data$Time <- as.POSIXct(temp_data$Time, format = "%d.%m.%Y", tz = "Etc/GMT+1")
temp_data$Time <- time_start$Time_start + time_average
temp_data <- temp_data[-which(colnames(temp_data) %in% c("Time_start", "Time_end"))]

} else {
# to pick right format for conversion to POSIX:
if (grepl("2017-", temp_data$Time[1])){
temp_data$Time <- as.POSIXct(temp_data$Time, format = "%Y-%m-%d", tz = "Etc/GMT+1")+(60*60*10)
} else {
temp_data$Time <- as.POSIXct(temp_data$Time, format = "%d.%m.%Y", tz = "Etc/GMT+1")+(60*60*10)
}
}
}

# Convert results of IC from mM to mgN/L
if (name_read[j] == "Dewatering"){
temp_data$NH4 <- temp_data$NH4*1400.1
}

# pick only measurements of reactor which currently reads in
temp_data <- temp_data[which(temp_data$Reactor == files_to_read[[i]]), c(-2)]

if (nrow(temp_data) > 0){ # because of M2 and M3
if (!name_read[j] == "Dewatering"){
temp_data$Cat <- name_read[j]
temp_data <- temp_data[, c(1, ncol(temp_data), 3:ncol(temp_data)-1)]
} else {
# has to assign polymer type to cat and remove this column
temp_data$Cat <- paste(name_read[j], "_", temp_data$Polymer, sep = "")
temp_data <- temp_data[, c(1, ncol(temp_data), 4:ncol(temp_data)-2)]
}
}

} else { # for M2 and M3 and dewatering and reject which should have same format to avoid problems in the following code
if (name_read[j] == "Dewatering"){

```

```

    temp_data <- temp_data[, -which(colnames(temp_data) == "Polymer")]
  }

  temp_data$Cat <- character(0)
}

temp_units <- read.table(sourcename, nrow = 1, skip = 1, header = F,
                        sep = ";", fill = T, stringsAsFactors=F)
colnames(temp_units) <- temp_header

if (name_read[j] == "Titration"){
  temp_units$"VFA_Alkalinity" <- "-"
}

if (name_read[j] == "Dewatering"){
  temp_units$NH4 <- "mgN/L"
}

temp_units <- temp_units[, -which(colnames(temp_units)
                                %in% c("Time", "Cat", "Polymer", "Reactor",
                                      "Hour", "Time_start", "Time_end"))]

assign(paste("temp_data_", name_assign[j], sep = ""), temp_data)
assign(paste("temp_units_", name_assign[j], sep = ""), temp_units)

}
}

# =====
# Read out sensor data
# =====
if (!files_to_read[[i]] == "raw_sludge"){
  names_var_system_sensor <- c("Time", "TEMPERATURE", "PH", "GASFLOW")
  names_var_assigned_sensor <- c("Time", "Temperature", "pH", "Gasflow")

  temp_data_cleaned <- list()

  sourcename <- paste (path, "/Data/Data_Pilot_scale/Sensor_analysis/", files_to_read[i], ".csv", sep = "")
  temp_header <- read.table(sourcename, nrow = 1, header = F,

```

```

sep = ",", dec = ".", fill = T, stringsAsFactors=F)

temp_data <- read.table(sourcename, skip = 1, header = F, colClasses = "character",
  sep = ",", dec = ".", fill = T, stringsAsFactors=F)

temp_data <- unite(temp_data, "Time", c(V1, V2), sep = " ")
temp_header <- unite(temp_header, "Time", c(V1, V2), sep = " ")
temp_header$Time <- "Time"
temp_data$Time <- as.POSIXct(temp_data$Time,format = "%m/%d/%y %H:%M:%S", tz = "Etc/GMT+1")

for (j in 1:length(names_var_system_sensor)){
  # make sure that assigns data to the right variables
  # independently of the order in the raw data
  temp_data_cleaned[j] <- temp_data[which(grepl(names_var_system_sensor[j], temp_header))]
}

temp_data_cleaned <- as.data.frame(temp_data_cleaned)

# make numbers numeric
temp_data_cleaned[,c(2:ncol(temp_data_cleaned))] <- lapply(temp_data_cleaned[,c(2:ncol(temp_data_cleaned))], function (x) as.numeric(as.character(x)))

colnames(temp_data_cleaned) <- names_var_assigned_sensor
temp_data_cleaned$Cat <- "Sensor"

# when problem with computer system (e.g., power cut) has gaps in sensor data
# -> has to remove to avoid problems with the following code pos_na <- list()
pos_na <- list()
for (j in 1:(length(names_var_assigned_sensor)-1)){
  temp_data_cleaned_variable <- temp_data_cleaned[[names_var_assigned_sensor[j+1]]]
  pos_na[[j]] <- which(is.na(temp_data_cleaned_variable))
}

# at some points all sensors have NA values, at some not
pos_na <- unlist(pos_na)
if (length(pos_na) > 0){
  pos_na <- pos_na[!duplicated(pos_na)]

  temp_data_cleaned <- temp_data_cleaned[-pos_na, ]
}

```

```

# order columns
temp_data_cleaned <- temp_data_cleaned[, c(1, ncol(temp_data_cleaned), 3:ncol(temp_data_cleaned)-1)]

temp_units <- data.frame("-", "°C", "-", "mL/min")
colnames(temp_units) <- names_var_assigned_sensor
temp_units <- data.frame(lapply(temp_units, as.character), stringsAsFactors=FALSE)
temp_units <- temp_units[,-which(colnames(temp_units) == "Time")]

assign("temp_data_sensor", temp_data_cleaned)
assign("temp_units_sensor", temp_units)
}

# =====
# Read out results of external lab
# =====
sourcename <- paste (path, "/Data/Data_Pilot_scale/Lab_analysis_external/Results_lab_analysis_",
                    files_to_read[i], ".csv", sep = "")

temp_header <- read.table(sourcename, nrow = 1, header = F,
                        sep = ";", dec = ",", fill = T, stringsAsFactors=F)

temp_data <- read.csv(sourcename, skip = 2, header = F,
                    sep = ";", dec = ",", fill = T, stringsAsFactors=F)
colnames(temp_data) <- temp_header

# to pick right format for conversion to POSIX:
if (grepl("2017-", temp_data$Time[1])){
  temp_data$Time <- as.POSIXct(temp_data$Time, format = "%Y-%m-%d", tz = "Etc/GMT+1")
} else {
  temp_data$Time <- as.POSIXct(temp_data$Time, format = "%d.%m.%Y", tz = "Etc/GMT+1")
}

temp_time <- list()

for (j in 1:nrow(temp_data)){
  if(temp_data$Cat[[j]]==1){
    time_start <- temp_data$Time[[j]]-(60*60*24*6)
    time_end <- time_start+(60*60*24*1)
  }
}

```

```

}

if(temp_data$Cat[[j]]==2){
  time_start <- temp_data$Time[[j]]-(60*60*24*4)
  time_end <- time_start+(60*60*24*1)
}

if(temp_data$Cat[[j]]==3){
  time_start <- temp_data$Time[[j]]-(60*60*24*2)
  time_end <- time_start+(60*60*24*2)
}

if(temp_data$Cat[[j]] == "month"){
  m <- month(temp_data$Time[[j]])
  y <- year(temp_data$Time[[j]])
  temp_data$Time[[j]] <- as.POSIXct(paste("15.", m-1, ".", y, sep = ""),
    format = "%d.%m.%Y", tz = "Etc/GMT+1")
} else {
  position_start <- which(weight$Time == time_start)
  position_end <- which(weight$Time == time_end)
  position_reactor <- which(colnames(weight) == files_to_read[i])

  time_start <- time_start + (60*60*10)
  time_end <- time_end + (60*60*10)
  time_diff <- time_end - time_start

  temp_weight <- weight[[position_end, position_reactor]]/
    (weight[[position_start, position_reactor]] + weight[[position_end, position_reactor]])

  temp_data$Time[[j]] <- time_start + temp_weight*time_diff
}
}

temp_data$VFA <- temp_data$Acetic_acid + temp_data$Propionic_acid+
temp_data$Butyric_acid + temp_data$Iso_Butyric_acid +
temp_data$Pentanoic_acid + temp_data$Iso_Pentanoic_acid +
temp_data$n_Hexanoic_acid + temp_data$n_iso_Hexanoic_acid +
temp_data$Heptanoic_acid

```

```

temp_data$Cat <- "Sludge"

temp_units <- read.table(sourcename, nrow = 1, skip = 1, header = F,
                        sep = ";", dec = ",", fill = T, stringsAsFactors=F)
colnames(temp_units) <- temp_header
temp_units$VFA <- "mg/L"
temp_units$NH4 <- "mgN/L" # to have same unit as in reject (assuming density = 1)
temp_units <- temp_units[,-which(colnames(temp_units) %in% c("Time", "Cat"))]

assign("temp_data_sludge", temp_data)
assign("temp_units_sludge", temp_units)

# =====
# Assemble data
# =====
if (!files_to_read[[i]] == "raw_sludge"){
  temp_names_data <- list(temp_data_sensor, temp_data_sludge, temp_data_feeding_volume,
                        temp_data_feeding_TS, temp_data_feeding_VS,
                        temp_data_dewatering, temp_data_titration, temp_data_gas)

  for (j in 1:length(temp_names_data)){
    if (j == 1){
      col_names <- colnames(temp_names_data[[j]])
    } else {
      col_names <- c(col_names, colnames(temp_names_data[[j]]))
    }
  }
}

joining_names <- unique(col_names[duplicated(col_names)])

for (j in 1){
  joining_names_exist_1 <- joining_names %in% colnames(temp_names_data[[j]])
  joining_names_exist_2 <- joining_names %in% colnames(temp_names_data[[j+1]])
  joining_names_exist <- joining_names[which(joining_names_exist_1 == joining_names_exist_2)]
  temp_data_joined <- full_join(temp_names_data[[j]], temp_names_data[[j+1]],
                              by = joining_names_exist)
}

for (j in 3:length(temp_names_data)-1){

```

```

joining_names_exist_1 <- joining_names %in% colnames(temp_data_joined)
joining_names_exist_2 <- joining_names %in% colnames(temp_names_data[[j+1]])
joining_names_exist <- joining_names[which(joining_names_exist_1 == joining_names_exist_2)]
temp_data_joined <- full_join(temp_data_joined, temp_names_data[[j+1]],
                             by = joining_names_exist)
}

temp_data_joined <- temp_data_joined[order(temp_data_joined$Time),]

if (TRUE %in% is.na(temp_data_joined$Time)){
  temp_data_joined <- temp_data_joined[-c(which(is.na(temp_data_joined$Time))),]
}

assign(files_to_read[[i]], temp_data_joined, env = .GlobalEnv)

temp_names_units <- list(temp_units_sensor, temp_units_sludge, temp_units_feeding_volume,
                        temp_units_feeding_TS, temp_units_feeding_VS,
                        temp_units_dewatering, temp_units_titration, temp_units_gas)
for (j in 1:length(temp_names_units)-1){
  if (j == 1){
    temp_units <- data.frame(temp_names_units[j], temp_names_units[j+1])

  } else {
    temp_units_2 <- data.frame(temp_names_units[j+1])
    same_name <- which(colnames(temp_units_2) %in% colnames(temp_units))

    if (length(same_name) > 0){
      temp_units <- data.frame(temp_units, temp_units_2[-c(same_name)])
    } else {
      temp_units <- data.frame(temp_units, temp_units_2)
    }
  }
}

temp_units <- data.frame(temp_units, Time = "-", Cat = "-")

for (j in 1:length(colnames(temp_data_joined))){
  position_name <- which(colnames(temp_units) == colnames(temp_data_joined[j]))

```

```

    if (j == 1){
      units_pilot_scale <- temp_units[position_name]
    } else {
      units_pilot_scale <- data.frame(units_pilot_scale, temp_units[position_name])
    }
  }
  assign("units_pilot_scale", units_pilot_scale, env = .GlobalEnv)

} else {
  temp_data_sludge <- temp_data_sludge[order(temp_data_sludge$Time),]

  assign(files_to_read[[i]], temp_data_sludge, env = .GlobalEnv)

  assign("units_pilot_scale", temp_units_sludge, env = .GlobalEnv)
}

write.table(eval(as.name(files_to_read[[i]])), file = paste(path, "/Data/Data_from_R/", files_to_read[[i]], ".csv", sep = ""),
  row.names = FALSE, sep = ";", dec = ",")
write.table(units_pilot_scale, file = paste(path, "/Data/Data_from_R/units_pilot_scale.csv", sep = ""),
  row.names = FALSE, sep = ";")

}

save(raw_sludge, T1, T2, T3, M1, M2, M3, units_pilot_scale, file = "data_pilot_scale_raw.R")

# =====
# Clean the gas sensor data and calculate gas volume
# =====
files_to_clean <- c("T1", "T2", "T3", "M1", "M2", "M3", "raw_sludge")
for (i in 1:length(files_to_clean)) {
  data_complete <- eval(as.name(files_to_clean[[i]]))

  if (!files_to_clean[i] == "raw_sludge"){
    data_sensor <- data_complete[which(data_complete$Cat == "Sensor"), ]
    data_gasflow <- data_sensor$Gasflow

    width_ma <- 60
    ma <- rollapply(data_gasflow, width = width_ma, mean, na.pad = TRUE, na.rm = TRUE)
  }
}

```



```

limit_lower <- factor_lower*ma
limit_upper <- factor_upper*ma

lower <- data_gasflow < limit_lower
upper <- data_gasflow > limit_upper

lower[is.na(lower)] <- FALSE
upper[is.na(upper)] <- FALSE

outlier <- vector()
for (j in 1:length(lower)){
  if (lower[j] == FALSE && upper[j] == FALSE){
    outlier[j] <- FALSE
  } else {
    outlier[j] <- TRUE
  }
  # additional criteria
  if(data_gasflow[j] > 25 && !is.na(data_gasflow[j]) && outlier[j] == FALSE){
    outlier[j] <- TRUE
  }
}

not_outlier <- which(outlier == FALSE)
outlier <- which(outlier == TRUE)

for(j in 1:length(outlier)){
  diff_start <- outlier[j]-not_outlier
  diff_min_start <- min(diff_start[! diff_start < 0])
  pos_start <- outlier[j]-diff_min_start

  diff_end <- not_outlier-outlier[j]
  diff_min_end <- min(diff_end[! diff_end < 0])
  pos_end <- outlier[j]+diff_min_end

  data_gasflow[pos_start:pos_end] <- mean(c(data_gasflow[pos_start],
                                           data_gasflow[pos_end]))
}

p1 <- ggplot()+

```

```

geom_line(aes(data_sensor$Time, ma, colour = "a) Moving average"),
  na.rm = T, size = 0.3, linetype = "dashed")+
geom_line(aes(data_sensor$Time, data_sensor$Gasflow,
  colour = "b) Raw"), na.rm = T, size = 0.3)+
geom_line(aes(data_sensor$Time, data_gasflow,
  colour= "c) Cleaned"),na.rm = T, size = 0.3)+
labs(x = "Time", y = "Gas flow [mL/min]")+
scale_y_continuous(limits = c(0,50))+
scale_x_datetime(date_breaks = "1 month", date_labels = "%Y-%m",
  limits = as.POSIXct(c("2017-02-13", "2017-08-06")))+
scale_colour_manual(values = c("red", "darkorchid", "green3"))+
theme_own()+
theme(legend.title = element_blank(), legend.position = c(0.85, 0.65))

print(p1)

ggsave(filename = paste("Correct_gasflow_", files_to_clean[i],
  ".png", sep = ""),
  plot = p1, path = paste(path_save_plots, "/Cleaning", sep = ""),
  height = 7, width = 15.5, units = "cm")

# overwrite original gasflow with corrected values
data_sensor$Gasflow <- data_gasflow
data_complete[which(data_complete$Cat == "Sensor"),
  which(colnames(data_complete) == "Gasflow")]<-data_gasflow

# Pick start of cycle (start of day and of cycle shifted)
# looking at gasproduction of cycle only makes sense if there was feeding on same day
dates_all <- date(data_sensor$Time)
data_feeding <- data_complete[data_complete$Cat == "Feeding", ]
data_feeding <- data_feeding[which(data_feeding$Feeding_volume > 0), ]
dates_feeding <- date(data_feeding$Time)

pos_start <- vector()

for (j in 1:length(dates_feeding)){
  # check for every day that has sensor data if there was feeding
  # find start of cycle on the day that had feeding
  pos_date <- which(dates_feeding[j] == dates_all)

```

```

data_gasflow_date <- data_gasflow[pos_date]

diff <- vector()
for (k in 1:length(data_gasflow_date-1)){

  # locate area in plot with biggest changes
  diff[k] <- data_gasflow_date[k+1]-data_gasflow_date[k]

}
# pick position of minimum gasflow in this area
pos_change <- which(diff > 0.8)

# for better picking: choose in right time area (after 7)
pos_change_7 <- which(hour(data_sensor$Time[pos_date[1]+pos_change]))>7)
pos_start[j] <- pos_change[pos_change_7[1]]+pos_date[1]-1

pos_start <- pos_start[!is.na(pos_start)]

}

# Gasflow and time of the slices used for integration in each cycle
gasflow_integration <- list()
time_integration <- list()

volume_gas <- list()
time_volume_gas <- list()
time_volume_gas_plot <- list()

for (j in 1:(length(pos_start)-1)){
  gasflow_integration_k <- vector()
  # take only full cycles (last j not used)
  pos_start_integration <- vector()
  pos_end_integration <- vector()

  pos_start_integration[1] <- pos_start[j]
  pos_end_integration[1] <- pos_start[j] + width_integration
  k = 2

  # take only samples that follow regular feeding pattern to avoid falsification of results (e.g. 19.4.)

```

```

if(pos_start[j+1]-pos_start[j] < 1.5*1440 &&
  !weekdays(data_sensor$Time[pos_start[j]]) %in% c("lördag", "Samstag",
    "Saturday"))||
  pos_start[j+1]-pos_start[j] < 2.5*1440 &&
  weekdays(data_sensor$Time[pos_start[j]]) %in% c("lördag", "Samstag",
    "Saturday")){

  while(k < (pos_start[j+1]-pos_start[j])/width_integration){
    pos_start_integration[k] <- pos_start_integration[k-1] + width_integration
    pos_end_integration[k] <- pos_end_integration[k-1] + width_integration
    k = k+1
  }

  # to take data of remaining increment
  pos_start_integration[k] <- pos_end_integration[k-1]

  # can only take start of next cycle as end point if no gap between cycles
  pos_end_integration[k] <- pos_start[j+1]

  for (k in 1:length(pos_start_integration)){
    gasflow_integration_k[k] <- width_integration * mean(data_gasflow[pos_start_integration[k]:pos_end_integration[k]])
  }

  gasflow_integration[[j]] <- gasflow_integration_k
  time_integration[[j]] <- seq(from = data_sensor$Time[pos_start[j]],
    length.out = length(gasflow_integration[[j]]),
    by = paste(width_integration, " min", sep = ""))

  # Total gas volume per cycle
  volume_gas[[j]] <- sum(gasflow_integration[[j]])/1000
  time_volume_gas[[j]] <- time_integration[[j]][1]

  diff_time <- (data_sensor$Time[pos_start[j+1]]-
    data_sensor$Time[pos_start[j]])/2
  time_volume_gas_plot[[j]] <- as.POSIXct(data_sensor$Time[pos_start[j]]+diff_time)

} else {
  gasflow_integration[[j]] <- NA
  time_integration[[j]] <- NA

```

```

    volume_gas[[i]] <- NA
    time_volume_gas[[i]] <- NA
    time_volume_gas_plot[[i]] <- NA
    #mean_time[i] <- NA
  }
}

# check for cycles with no data
not_is_na <- which(!is.na(volume_gas))

gasflow_integration <- gasflow_integration[not_is_na]
time_integration <- time_integration[not_is_na]
volume_gas <- volume_gas[not_is_na]
time_volume_gas <- time_volume_gas[not_is_na]
time_volume_gas_plot <- time_volume_gas_plot[not_is_na]

# unlist times, gasflows and -volumes during cycles
gasflow_integration <- unlist(gasflow_integration)
time_integration <- as.POSIXct(unlist(time_integration),
                              origin = "1970-01-01", tz = "Etc/GMT+1")
volume_gas <- unlist(volume_gas)
time_volume_gas <- as.POSIXct(unlist(time_volume_gas),
                              origin = "1970-01-01", tz = "Etc/GMT+1")
time_volume_gas_plot <- as.POSIXct(unlist(time_volume_gas_plot),
                                   origin = "1970-01-01", tz = "Etc/GMT+1")

volume_gas_df <- data.frame(volume_gas)
time_volume_gas_df <- data.frame(time_volume_gas)

data_volume_gas <- as.data.frame(c(time_volume_gas_df, volume_gas_df))
colnames(data_volume_gas) <- c("Time", "Volume_gas")

names_colour_general <- names(colour_general)
colour_plot <- colour_general[[which(names_colour_general == files_to_clean[i])]]
p2 <- ggplot()+
  geom_rect(aes(xmin = time_integration,
                xmax = as.POSIXct(as.numeric(time_integration)+width_integration*60,
                                   origin = "1970-01-01", tz = "Etc/GMT+1"),
                ymin = rep(0, length(time_integration)),

```

```

      ymax = gasflow_integration/width_integration), # to get same height),
      fill = colour_plot, alpha = 0.25, na.rm = T)+
geom_line(aes(x = data_sensor$Time, y = data_sensor$Gasflow,
      colour = "b) Cleaned gas flow"), na.rm = T)+
geom_point(aes(x = data_sensor$Time[pos_start], y = data_sensor$Gasflow[pos_start],
      colour = "c) Start of cycle"), size = 1.5, shape = 4, stroke = 1, na.rm = T)+
geom_point(aes(x = time_volume_gas_plot, y = volume_gas,
      colour = "d) Gas volume of cycle"),
      size = 1.5, stroke = 0.7, shape = 23, fill = colour_plot, na.rm = T)+
scale_y_continuous(limits = c(0,25))+
scale_x_datetime(date_breaks = "1 month", date_labels = "%Y-%m",
      limits = as.POSIXct(c("2017-02-13", "2017-08-06")))+
labs(x = "Time", y = "Gas flow [mL/min] resp. Gas volume [L]")+
scale_colour_manual(values = c(colour_plot, "green3", "black"))+

guides(colour = guide_legend(override.aes = list(linetype = c(1,0,0),
      shape = c(NA, 4, 23))),
      fill = FALSE, alpha = FALSE)+
theme_own()+
theme(legend.title = element_blank(), legend.position = c(0.85, 0.8))

print(p2)

ggsave(filename = paste("Volume_gas_", files_to_clean[i],
      ".png", sep = ""),
      plot = p2, path = paste(path_save_plots, "/Cleaning", sep = ""),
      height = 8, width = 15.5, units = "cm")

# Replace times (not for sludge)
pos_variables <- which(data_complete$Cat %in%
      c("Feeding", "Dewatering_Z8180", "Dewatering_O6007"))

pos_titration <- which(data_complete$Cat == "Titration")
data_titration <- data_complete[pos_titration, ]
pos_titration_replace <- which(as.character(times(format(data_titration$Time, "%H:%M:%S")))
      == "10:00:00")
pos_variables <- c(pos_variables, pos_titration_replace)
# still contains also times of sensor measurements that doesn't want to replace

```

```

pos_date <- which(date(data_complete$Time) %in% date(data_volume_gas$Time))
pos_variables_replace <- which(pos_variables %in% pos_date)
pos_variables_replace <- pos_variables[pos_variables_replace]

# pick replacement values
time_replace <- list()
for (j in 1:length(pos_variables_replace)){
  time_replace[[j]] <- data_volume_gas$Time[which(date(data_complete$Time[pos_variables_replace[j]]) ==
                                                    date(data_volume_gas$Time))]
}
time_replace <- as.POSIXct(unlist(time_replace),
                           origin = "1970-01-01", tz = "Etc/GMT+1")

data_complete$Time[pos_variables_replace] <- time_replace

# only after replacement so that times and cat put together when joining
data_volume_gas$Cat <- "Feeding"
data_complete <- full_join(data_complete, data_volume_gas, by = c("Time", "Cat"))
data_complete <- data_complete[order(data_complete$Time), ]
}

data_complete$Weekday <- weekdays(data_complete$Time)
data_complete$Weekday <- as.factor(data_complete$Weekday)

if ("Dienstag" %in% data_complete$Weekday){
  data_complete$Weekday <- revalue(data_complete$Weekday,
                                   c("Montag" = "Mon", "Dienstag" = "Tue", "Mittwoch" = "Wed",
                                     "Donnerstag" = "Thu", "Freitag" = "Fri",
                                     "Samstag" = "Sat", "Sonntag" = "Sun"))
}

if ("Tuesday" %in% data_complete$Weekday){
  data_complete$Weekday <- revalue(data_complete$Weekday,
                                   c("Monday" = "Mon", "Tuesday" = "Tue", "Wednesday" = "Wed",
                                     "Thursday" = "Thu", "Friday" = "Fri",
                                     "Saturday" = "Sat", "Sunday" = "Sun"))
}

if ("tisdag" %in% data_complete$Weekday){

```

```

data_complete$Weekday <- revalue(data_complete$Weekday,
                                c("måndag" = "Mon", "tisdag" = "Tue", "onsdag" = "Wed",
                                  "torsdag" = "Thu", "fredag" = "Fri",
                                  "lördag" = "Sat", "söndag" = "Sun"))
}

data_complete$Weekday <- factor(data_complete$Weekday,
                                levels = c("Mon", "Tue", "Wed", "Thu",
                                             "Fri", "Sat", "Sun"))

# different time formats (for whatever reason)
if(files_to_clean[i] == "raw_sludge"){
  data_complete$Day_of_experiment <- as.numeric(round((data_complete$Time -
                                                       as.POSIXct("2017-04-25 10:00:00", tz = "Etc/GMT+1"))/
                                                       24, digits = 5))
} else {
  data_complete$Day_of_experiment <- as.numeric(round((data_complete$Time -
                                                       as.POSIXct("2017-04-25 10:00:00", tz = "Etc/GMT+1"))/
                                                       (24*60*60), digits = 5))
}

data_complete <- data_complete[, c(1, ncol(data_complete),
                                   ncol(data_complete)-1, 4:ncol(data_complete)-2)]

if (i == 1){ # needs to only read in once
  units_pilot_scale <- data.frame(units_pilot_scale[, 1], "Day_of_experiment" = "-",
                                  "Weekday" = "-",
                                  units_pilot_scale[, 2:ncol(units_pilot_scale)],
                                  "Volume_gas" = "L")
  colnames(units_pilot_scale)[1] <- "Time"
}
assign(files_to_clean[[i]], data_complete)
}

save(raw_sludge, T1, T2, T3, M1, M2, M3, units_pilot_scale, file = "data_pilot_scale_cleaned.R")
}

```


read_full_scale.R

```

# Created: Tineke Bittlingmayer (bit), May 2017
# Last update: bit, 2017-08-08
# =====
# Code for reading in data of the full-scale plant
# =====
# =====
read_full = function(){
  sourcename <- paste (path, "/Data/Data_Full_scale/data_fullscale.csv", sep = "")

  temp_header <- read.table(sourcename, nrow = 1, header = F,
                           sep = ";", dec = ".", fill = T, stringsAsFactors=F)

  temp_data <- read.csv(sourcename, skip = 8, header = F,
                      sep = ";", dec = ".", fill = T, stringsAsFactors=F)
  colnames(temp_data) <- temp_header

  temp_data$Time <- as.POSIXct(temp_data$Time, format = "%d.%m.%Y %H:%M", tz = "Etc/GMT+1")

  # wants to have data in same units as for pilot scale:
  temp_data$Alkalinity_R1 <- temp_data$Alkalinity_R1*1000
  temp_data$Alkalinity_R2 <- temp_data$Alkalinity_R2*1000

  temp_units <- read.table(sourcename, nrow = 1, skip = 3, header = F,
                          sep = ";", dec = ".", fill = T, stringsAsFactors=F)
  colnames(temp_units) <- temp_header
  temp_units[is.na(temp_units)] <- "-"
  temp_units[colnames(temp_units) %in% c("Alkalinity_R1", "Alkalinity_R2")] <- "mg CaCO3/L"
  temp_units[colnames(temp_units) %in% c("TS_in", "TS_out")] <- "%"
  temp_units[colnames(temp_units) %in% c("VS_in", "VS_out")] <- "% TS"

  assign("full_scale", temp_data, env = .GlobalEnv)
  assign("units_full_scale", temp_units, env = .GlobalEnv)

  save(full_scale, units_full_scale, file = "data_full_scale.R")
}

```

load.R

```

# Created: Tineke Bittlingmayer (bit), May 2017
# Last update: bit, 2017-08-08
# =====
# Extracts the data period specified by user from the complete data set
# =====
# =====
load_data = function(date_start, date_end, data){
  for (i in 1:length(data)){
    load(data[i])
  }

  # find substrings to account for different names after manipulation of data of pilot scale
  if (TRUE %in% grepl("pilot", data)){
    units_pilot_scale <- units_pilot_scale
  }

  if (TRUE %in% grepl("full", data)){
    units_full_scale <- units_full_scale
  }

  if (length(data) == 1){
    if (TRUE %in% grepl("pilot", data)){
      files_to_read <- c("raw_sludge", "T1", "T2", "T3", "M1", "M2", "M3")
    }

    if (TRUE %in% grepl("full", data)){
      files_to_read <- "full_scale"
    }

  } else {
    files_to_read <- c("raw_sludge", "T1", "T2", "T3", "M1", "M2", "M3", "full_scale")
  }

  for (i in 1:length(files_to_read)){
    temp_data <- eval(as.name(files_to_read[[i]]))

    time_diff_start <- temp_data$Time -
      as.POSIXct(date_start, format = "%Y-%m-%d %H:%M:%S", tz = "Etc/GMT+1")
  }
}

```

```
pos_time_start <- which(time_diff_start == min(time_diff_start[which(!time_diff_start<0)]))

if (length(pos_time_start) > 1){ # possible if data of different categories
  pos_time_start <- pos_time_start[1]
}

time_diff_end <- as.POSIXct(date_end, format = "%Y-%m-%d %H:%M:%S", tz = "Etc/GMT+1") - temp_data$Time

pos_time_end <- which(time_diff_end == min(time_diff_end[which(!time_diff_end<0)]))

if (length(pos_time_end) > 1){ # possible if data of different categories
  pos_time_end <- pos_time_end[2]
}

temp_data <- temp_data[pos_time_start:pos_time_end, ]

temp_data <- temp_data[! duplicated(temp_data), ]

assign(files_to_read[[i]], temp_data, env = .GlobalEnv)

if (temp_data$Time[1] > as.POSIXct(date_end, format = "%Y-%m-%d %H:%M:%S", tz = "Etc/GMT+1")){
  rm(list = files_to_read[[i]])
}
}
```

theme.R

```

# Created: Tineke Bittlingmayer (bit), May 2017
# Last update: bit, 2017-08-08
# =====
# Code containing the default setting for all plots
# =====
# =====
theme_own <- function (base_size = 10) {
  theme_bw(base_size = base_size) %+replace%
  theme(
    plot.title = element_text(size = rel(1), face = "bold", margin = margin (b = 10, unit = "pt")),
    plot.background = element_rect(fill = "white", colour = NA),

    panel.background = element_rect (fill = "transparent", colour = NA),
    panel.border = element_rect(fill = NA, colour = "black", size = 0.5),
    panel.grid.major = element_line(colour = "transparent", size = 0.2),
    panel.grid.minor = element_line(colour = "transparent", size = 0.2),
    panel.spacing.x = unit(30, "pt"),
    panel.spacing.y = unit(30, "pt"),

    strip.background = element_rect(fill = "grey70", colour = "white"),
    strip.text.x = element_text(size = rel(1)),

    axis.title = element_text(size = rel(1), colour = "black"),
    axis.text = element_text(size = rel(0.8), colour = "black"),
    axis.line = element_blank(),
    axis.ticks = element_line(colour = "black"),

    legend.title = element_text(size = rel(1)),
    legend.background = element_rect(fill = "white", colour = "black", size = 0.2),
    legend.key = element_rect(fill = "white", colour = "white"),
    legend.text = element_text (size = rel(0.8))
  )
}

```

plot_overview.R

```

# Created: Tineke Bittlingmayer (bit), May 2017
# Last update: bit, 2017-08-08
# =====
# General plot that can be adapted by the user via input parameters
# =====
# =====

# Parameters to adapt:
# variable_x_axis: "Time" or "Day_of_experiment"
# variable_y_axis: combination of variables that wants to plot. 1 plot for each.
# names_variables: if wants to have different labels on y-axis
# reactors_to_plot: list which contains for each variable_y_axis the reactors that wants to plot
# x_axis_ticks:
# - if variable_x_axis = "Time": specify as min, hour, day, week, month
# - if variable_x_axis = "Day_of_experiment": specify as number
# format_x_axis_ticks:
# - if variable_x_axis = "Time": specify format e.g., %Y-%m
# - if variable_x_axis = "Day_of_experiment": put "none"
# x_axis_limits:
# - if variable_x_axis = "Time": put "none"
# - if variable_x_axis = "Day_of_experiment": set lower and upper limit
# y_axis_ticks: list which contains for each variable_y_axis the ticks
#   in form of lower, upper, increment
# y_axis_limits: list which contains for each variable_y_axis the lower and upper limit
# facetting: variable to use to split data into different plots
# seq_facet: put cut points if facetting done on continuous variable (e.g. pH)
#   and wants fixed cut points. else: put "none"
# cut_facetting: list containing the min, max and increment for cuts.
#   If puts "none" for min and max: no data points are cut off at the ends
# labels_facetting: labels for the different facets
# graph_points: put "yes" or "no" for each variable_y_axis and each reactors_to_plot
# point_size: size of points in graph
# graph_lines: analogue to graph_points
# line_size: size of line in plot
# vertical_line: lines to divide plot into periods. "none" or x-values
# legend_position:
# - if "none": 1 legend at bottom.
# - if specified: legend for each plot in specified position

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# save_name: name to use for saving
# height_single_plot <- height of each plot showing variable_y_axis

plot_same_x_axis = function(colour_general, variable_x_axis, variable_y_axis,
                             names_variables, reactors_to_plot,
                             x_axis_ticks, format_x_axis_ticks, x_axis_limits,
                             y_axis_ticks, y_axis_limits, facetting,
                             seq_facet, cut_facetting, labels_facetting,
                             graph_points, point_size, graph_lines,
                             line_size, vertical_line, legend_position, save_name, height_single_plot){

plots <- list()
y_ticks <- list()

for (i in 1:length(variable_y_axis)){

  # Take out data to plot and stack
  for (j in 1:length(reactors_to_plot[[i]])){
    data_reactor <- eval(as.name(reactors_to_plot[[i]][j]))

    pos_x_variable <- which(colnames(data_reactor) == variable_x_axis)
    pos_y_variable <- which(colnames(data_reactor) == variable_y_axis[i])

    if (!facetting == "none"){
      pos_facet <- which(colnames(data_reactor) == facetting)
    }

    if (! facetting == "none"){
      data_full <- data_reactor[, c(pos_x_variable, pos_y_variable, pos_facet)]
    } else {
      data_full <- data_reactor[, c(pos_x_variable, pos_y_variable)]
    }

    pos_not_na <- which(!is.na(data_full[[variable_y_axis[[i]]]]))
    data_full <- data_full[pos_not_na, ]

    if (j == 1){
      data_variable <- data_full[, 1:2] # pick x and y values
      if (!facetting == "none"){

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    data_facet <- data_full[,c(1,3)]
  }

  } else {
    data_variable <- full_join(data_variable, data_full[,1:2],
                               by = variable_x_axis)
    if(!facetting == "none"){
      data_facet <- full_join(data_facet, data_full[, c(1,3)],
                              by = variable_x_axis)
    }
  }
}

colnames(data_variable) <- c(variable_x_axis, reactors_to_plot[[i]])

if(!facetting == "none"){
  colnames(data_facet) <- c(variable_x_axis, reactors_to_plot[[i]])
}

y_data_stacked <- stack(data_variable[2:ncol(data_variable)])
x_data_stacked <- rep(data_variable[, variable_x_axis], length(reactors_to_plot[[i]]))

if (! facetting == "none"){
  facet_stacked <- stack(data_facet[2:ncol(data_facet)])
  facet_stacked <- facet_stacked$values
}

if (! facetting == "none" && seq_facet == "none"){
  if (is.numeric(facet_stacked)){ # because can do splitting only if not string
    if (cut_facetting[[1]] == "none"){
      # wants that all data is included in some category and not excluded due to rounding
      diff_facet_start <- min(facet_stacked, na.rm = T) -
        round(min(facet_stacked, na.rm = T), digits = 0)
      if(diff_facet_start < 0){
        facet_start <- round(min(facet_stacked, na.rm = T), digits = 0) - 1
      } else {
        facet_start <- round(min(facet_stacked, na.rm = T), digits = 0)
      }
    } else {

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    facet_start <- cut_facetting[[1]]
  }

  if (cut_facetting[[2]] == "none"){
    diff_facet_end <- round(max(facet_stacked, na.rm = T), digits = 0)-
      max(facet_stacked, na.rm = T)

    if(diff_facet_end < 0){
      facet_end <- round(max(facet_stacked, na.rm = T), digits = 0) + 1
    } else {
      facet_end <- round(max(facet_stacked, na.rm = T), digits = 0)
    }

  } else {
    facet_end <- cut_facetting[[2]]
  }

  seq_facet <- seq(facet_start, facet_end, cut_facetting[[3]])
  # to avoid NA
  if (seq_facet[length(seq_facet)] < facet_end){
    seq_facet <- c(seq_facet, seq_facet[length(seq_facet)]+cut_facetting[[3]])
  }
}

pos_not_na <- which(!is.na(y_data_stacked$values))

x_data_stacked <- x_data_stacked[pos_not_na]
y_data_stacked <- y_data_stacked[pos_not_na, ]

if (! facetting == "none"){
  facet_stacked <- facet_stacked[pos_not_na]
}

if (! facetting == "none"){
  if (is.numeric(facet_stacked)){
    facet_stacked <- cut(facet_stacked, seq_facet)
    #data_facet <- cut(data_variable$facet,
    #seq(facet_start, facet_end, cut_facetting[[3]]))
  }
}

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}

facet_stacked <- as.factor(facet_stacked)

if (!labels_facetting[1] == "none"){
  levels(facet_stacked) <- labels_facetting
}

df <- data.frame(x = x_data_stacked, y = y_data_stacked$values,
  Reactor = y_data_stacked$ind, facet = facet_stacked)
} else {
  df <- data.frame(x = x_data_stacked, y = y_data_stacked$values,
    Reactor = y_data_stacked$ind)
}

if (!vertical_line[1] == "none"){
  if (!facetting == "none"){
    length_facet <- length(levels(facet_stacked))
    facet_vertical <- list()
    for (k in 1:length(levels(facet_stacked))){
      facet_vertical[[k]] <- rep(levels(facet_stacked)[k], length(vertical_line))
    }
    facet_vertical <- unlist(facet_vertical)

    df_vertical <- data.frame(x = rep(vertical_line, length_facet),
      xend_v = rep(vertical_line, length_facet),
      y_v = rep(y_axis_limits[[i]][1], length(vertical_line)*length_facet),
      yend_v = rep(y_axis_limits[[i]][2], length(vertical_line*length_facet)),
      facet = facet_vertical)
    df <- full_join(df, df_vertical, by = c("x", "facet"))
  } else {
    df_vertical <- data.frame(x = vertical_line, xend_v = vertical_line,
      y_v = rep(y_axis_limits[[i]][1], length(vertical_line),
      yend_v = rep(y_axis_limits[[i]][2], length(vertical_line)))
    df <- full_join(df, df_vertical, by = "x")
  }
}

plots[[i]] <- ggplot()

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if(!vertical_line[1] == "none"){
  plots[[i]] <- plots[[i]]+
    geom_segment(data = df, aes(x = x, xend = xend_v,
      y = y_v, yend = yend_v),
      colour = "grey50", linetype = "longdash", size = 0.3, na.rm = T)
}

if (graph_points[[i]] == "yes"){
  plots[[i]] <- plots[[i]]+
    geom_point(data = df[!is.na(df$y), ],
      aes(x, y, colour = Reactor, shape = Reactor),
      na.rm = T, size = point_size)
}

if (graph_lines[[i]] == "yes"){
  plots[[i]] <- plots[[i]]+
    geom_line(data = df[!is.na(df$y), ],
      aes(x, y, colour = Reactor), na.rm = T, size = line_size,
      linetype = "dashed")
}

y_ticks[[i]] <- seq(y_axis_limits[[i]][1], y_axis_limits[[i]][2], y_axis_ticks[[i]][3])

plots[[i]] <- plots[[i]]+
  scale_colour_manual(values = colour_general)+
  scale_shape_manual(values = symbols_general)+
  scale_y_continuous(breaks = y_ticks[[i],
    limits = y_axis_limits[[i]])+
  labs(y = paste(names_variables[i], " [",
    as.character(units_pilot_scale[[variable_y_axis[[i]]]]), "]", sep = ""))+
  theme_own()+
  theme(legend.position = legend_position, legend.title = element_blank(),
    plot.margin = unit(rep(.2, 4), "lines"))+
  guides(col = guide_legend(nrow = 1))

if (variable_x_axis == "Time"){

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plots[[i]] <- plots[[i]] + scale_x_datetime(date_breaks = x_axis_ticks,
                                             date_labels = format_x_axis_ticks)+
  labs(x = "Time")
}

if (variable_x_axis == "Day_of_experiment"){
  x_ticks <- seq(x_axis_limits[1], x_axis_limits[2], x_axis_ticks)

  plots[[i]] <- plots[[i]] +
    scale_x_continuous(breaks = x_ticks, limits = x_axis_limits)+
    labs(x = "Day of experiment")
}

if (!variable_x_axis == "Time" && !variable_x_axis == "Day_of_experiment"){
  x_ticks <- seq(x_axis_limits[1], x_axis_limits[2], x_axis_ticks)

  plots[[i]] <- plots[[i]] +
    scale_x_continuous(breaks = x_ticks, limits = x_axis_limits)+
    labs(x = paste(variable_x_axis, " [",
                  as.character(units_pilot_scale[[variable_x_axis]]), "]", sep = ""))
}

if (length(variable_y_axis) > 1){
  if (!i == 1){
    plots[[i]] <- plots[[i]] + theme(legend.title = element_text(colour = "white"))
  }

  if (i < length(variable_y_axis)){
    plots[[i]] <- plots[[i]] + theme(axis.title.x = element_text(colour = "white"),
                                     axis.text.x = element_text(colour = "white"))
  }
}

if(!facetting == "none"){
  plots[[i]] <- plots[[i]] + facet_wrap(~ facet)+
    theme(panel.spacing.x = unit(0.5, "lines"))
}

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    }

    if (i == 1){
      g <- ggplotGrob(plots[[i]])
    } else {
      g <- rbind(g, ggplotGrob(plots[[i]]))
    }
  }

  if (legend_position[1] == "none"){
    prow <- plot_grid(g, align = 'vh', hjust = -1, nrow = 1)
    legend_b <- get_legend(plots[[1]] + theme(legend.position="bottom"))

    p <- plot_grid(prow, legend_b, ncol = 1, rel_heights = c(1, .1))

  } else {
    prow <- plot_grid(g, align = 'vh', hjust = -1, nrow = 1)

    p <- plot_grid(prow, ncol = 1, rel_heights = c(1, .1))
  }

  height_plot = height_single_plot*length(variable_y_axis)

  ggsave(filename = paste(save_name, ".png", sep = ""),
        plot = p, path = paste(path_save_plots, "/Overview", sep = ""),
        height = height_plot, width = 14.8, units = "cm")
}

```