Co-digestion of food and garden waste with mixed sludge from wastewater treatment in continuously stirred tank reactors

Fitamo, Temesgen Mathewos; Boldrin, Alessio; Boe, Kanokwan; Angelidaki, Irini; Scheutz, Charlotte

Published in:
Bioresource Technology

Link to article, DOI:
10.1016/j.biortech.2016.01.085

Publication date:
2016

Document Version
Peer reviewed version

Link back to DTU Orbit

Citation (APA):
Co-digestion of food and garden waste with mixed sludge from wastewater treatment in continuously stirred tank reactors

Fitamo, T., Boldrin, A., Boe, K., Angelidaki, I., Scheutz, C.

Department of Environmental Engineering
Technical University of Denmark
Kgs. Lyngby, Denmark

"NOTE: this is the author’s version of a work that was accepted for publication in Bioresource Technology journal. Changes resulting from the publishing process, such as peer review, editing, corrections, structural formatting, and other quality control mechanisms may not be reflected in this document. Minor changes may have been made to this manuscript since it was accepted for publication. A definitive version is published in Bioresource Technology, vol 206, pp 245-254, doi: dx.doi.org/10.1016/j.biortech.2016.01.085"
Abstract

Co-digestions of urban organic waste were conducted to investigate the effect of the mixing ratio between sludge, food waste, grass clippings and green waste at different hydraulic retention times (HRTs). Compared to the digestion of 100% sludge, the methane yield increased by 48% and 35%, when co-digesting sludge with food waste, grass clippings and garden waste with a corresponding VS% of 10:67.5:15.75:6.75 (R1) and 10:45:31.5:13.5 (R2), respectively. The methane yield remained constant at around 425 and 385 NmL CH₄/g VS in R1 and R2, respectively, when the reactors were operated at HRTs of 15, 20 and 30 days. However, the methane yield dropped significantly to 356 (R1) and 315 (R2) NmL CH₄/g VS when reducing the HRT to 10 days, indicating that the process was stressed. Since the methane production rate improved significantly with decreasing HRT, the trade-off between yield and productivity was obtained at 15 days HRT.

Keywords

Anaerobic digestion, biogas, urban organic waste, sewage sludge, hydraulic retention time, biochemical methane potential
1. Introduction

As a consequence of increasing demand for renewable energy, biogas production is expected to increase in many countries and to play an important role in future energy systems. To catch up with the increasing demand, due to political targets set by many countries, additional types of biomass will have to be exploited synergistically, including organic waste originating from urban areas. For example, the European Union has adopted renewable energy directives to increase the share of energy from biofuels by at least 10% in the transport sector across the member states (European Union, 2009). Similarly, the People’s Republic of China has planned to construct several rural household biogas digesters and biogas service system (Peidong et al., 2009), while the USA, among different initiatives, established the energy independence and security act of 2007 to increase energy production from advanced biofuels (U.S. Congress, 2007).

The Danish government has an ambitious energy strategy to become 100% independent of fossil fuels by 2050 (Energinet.dk, 2010; The Danish Government, 2011). The major source of renewable energy is predicted as being wind-sourced, but biogas production is also expected to provide a significant contribution in the near future. Danish centralised and farm-scale biogas plants currently deliver about 3.8 PJ of renewable energy by processing only 7% of the manure produced in the country (Energistyrelsen, 2014). Recent policies have focused on progressively introducing the mandatory separate collection of biowaste by 2020 so that increasing amounts of organic wastes from households, restaurants, businesses and industries can be used as feedstock for anaerobic digestion (AD) plants in the coming years, whilst also decreasing the amount of organic waste sent to incineration (The Danish Government, 2013). While the establishment of new AD capacity will be needed, the achievement of targets will also require a general optimisation of biogas production by synergistically exploiting different flows of available organic materials (Boldrin et al., 2015).

AD reactors fed with manure as feedstock are commonplace, but they are typically located in rural areas, and transporting organic waste from urban areas can be costly. Conversely, AD facilities are often found in wastewater treatment plants (WWTPs), which are typically located near urbanised areas. WWTPs generate substantial amounts of primary and secondary sludge that is treated/stabilised in AD reactors to produce biogas. In many countries, sludge AD is often integrated within a WWTP, and the energy produced from biogas is mostly used to cover on-site demand, albeit in some cases surplus energy is generated and delivered to the electricity network and in other cases to the district heating system. AD of municipal wastewater provides significant benefits such as savings related to sludge disposal and odour reduction, environmental benefit as well as rescind pathogens (Appels et al., 2008). While the cost of AD in a WWTP is high and energy production is sub-optimised (Weiland, 2010), many facilities currently also have spare AD capacity, owing to a general decrease in wastewater generation in Denmark. In this context, treating urban organic waste anaerobically in a WWTP could be a way to exploit this surplus capacity while ensuring the management of waste at a local level and possibly improving WWTP economy. In fact, processing sludge and urban waste co-substrates could have some synergetic benefits. On the one hand, the addition of co-substrates into existing sewage sludge-based AD reactors has the potential to significantly increase biogas production that will in turn lead to the increased net energy generation of WWTP. On the other hand, sludge would provide both dilution and buffering effects, making the AD of urban waste more of a feasible undertaking. Moreover, the performance of co-digestion can be enhanced with AD of food and co-substrates due to buffer capacity and nutrient balance (Zhang et al., 2014).

In AD, organic matter available in the waste is converted into biogas that can be used to generate power, while the remaining digestate can be used as a soil conditioner in agriculture. AD technologies comprise a rather wide variety of options, the suitability of which may depend on the type of feedstock. In general, compared to a mesophilic process, thermophilic (i.e. 55°C) AD provides improved gas yield and organic matter reduction as well as better biological and chemical reactions, reduced disposal costs for the sludge and better hygienisation (Angelidaki et al., 2006). Thermophilic AD using sludge and food waste as single substrates has been studied rather extensively in the literature, generally showing the technical feasibility of the process for sludge (Astals et al., 2012; Ferrer et al., 2008; Gavala et al., 2003) and for food
waste (Climenhaga and Banks, 2008; Forster-Carneiro et al., 2008) while identifying some challenges for regular operation. For example, Banks et al. (2011) showed that, because of its high protein content, using food waste as a single substrate could result in the accumulation of VFA due to ammonia inhibition. Therefore, proper feedstock composition is of great importance, and optimal composition can be achieved through appropriate co-substrate mixing. Previous studies have examined the effects of mixing different substrates. For example, Bolzonella et al. (2006) and Cecchi et al. (1988) showed that co-digesting activated sludge with food waste resulted in improved process performance compared to the single substrate, in both pilot and full-scale tests. Co-digestion of food and green waste was also investigated in a few cases based on batch experiments, showing that biogas yield increased when increasing the fraction of food waste compared to green waste (Chen et al., 2014), or that thermophilic conditions provide higher biogas yield compared to mesophilic settings (Liu et al., 2009).

In general terms, even though studies have been conducted on the co-digestion of sewage sludge as a main substrate, there is clearly a lack of information regarding the co-digestion of food and green waste as a main substrate with sludge (Mata-Alvarez et al., 2014). In particular, the steady operation of a continuous process in thermophilic conditions, using sludge and food/green waste as the substrate, has not been reported previously in the literature.

The objective of this study was to explore the possibility of utilising urban organic waste, such as garden and food waste, for biogas production in co-digestion with mixed sewage sludge. This was done by: 1) co-digesting food waste and garden waste with WWTP sludge in continuously stirred tank reactors (CSTRs) operated at 55 °C, ii) investigating the effect of the co-substrate mixing ratio on biogas yield and iii) identifying the optimal hydraulic retention time (HRT) for the process. Moreover, the biochemical methane potential (BMP) was set up to analyse the biogas potential of individual substrates.

2. Materials and Methods

2.1. Waste sampling, sample preparation and characterization

Food waste was obtained from the main canteen at the Technical University of Denmark, grass and garden waste was sampled from private gardens and the Borgervænget recycling station and sludge (collected separately as primary and secondary sludge but later mixed at a volume ratio of 1:1) was sampled from the Avedøre wastewater treatment plant (WWTP) in Denmark. A series of mechanical pretreatment steps were employed to obtain representative samples with a particle size suitable for the analyses and the lab-scale reactor experiments. Food waste, grass clippings and garden waste were mechanically mixed and shredded with a shear-shredder (ARP SC 2000) to particle sizes of 16 mm. Woody materials were separated from the garden waste by manual sorting, whereas soil was separated from the grass clippings by mesh sieving. Representative samples were attained via laying the sample lots in elongated 1-D multilayer piles and dividing the sample lot into equal portions that were either accepted or discarded until the required sample amount was achieved. Further size reduction was conducted with a comminutor (Fitzmill model D,Daso-6) and a cutter knife mill (Wiencken 19225), in order to obtain 4 mm particle size, which could be pumped through the lab scale pipelines. The pre-treated samples were then kept in small containers and stored at -20°C. Inoculum for continuous and batch reactor experiments was obtained from Avedøre WWTP, Denmark, and Va Syd WWTP, Malmö (Sjölunda), Sweden, respectively.

The waste samples were characterised by determining total solids (TS), volatile solids (VS), pH, total ammonia concentration and total Kjeldahl nitrogen (TKN), according to APHA standard methods for the examination of water and wastewater (APHA, 2005). The content of fat, total nitrogen and total carbon was analysed with DHF 42, ISO 13878 and DS/EN 13137, respectively, at a commercial laboratory (Eurofins, DK). The analyses of volatile fatty acids (VFAs) and alcohol were performed with gas chromatography (Shimadzu GC-2010AF, Kyoto, Japan) fitted with a flame ionisation detector (FID).

2.2. Biochemical methane potential (BMP)

The experimental biochemical methane potential (BMP) of food waste, grass clippings, garden waste and sludge was conducted in triplicate in a mesophilic incubator in a 1 L batch reactor, with a working
volume of 0.3 L. The organic loading rate (OLR) of substrates was 2.7 g VS/L, whereas the VS-based inoculum-to-substrate ratio (ISR) was maintained at 2 (Hansen et al., 2004). Inoculum collected from the Va Syd Sjölunda WWTP (Malmö, SE) was transferred into all assay batch reactors in the required amounts. Avicel (Fluka, Denmark) was used as the standard substrate in the control experiments, while blanks were used to quantify the contribution of gas production from the inoculum. The batch reactors were purged with 100% N₂ in order to achieve anaerobic conditions. Methane production from inoculum was subtracted from the substrates’ assays, in order to remove any background methane production (Angelidaki et al., 2009). The theoretical BMPs (T-BMPs) for the input feedstock were determined based on the organic chemical composition (proteins, carbohydrates and lipids) of each input waste material (Møller et al., 2004; Triolo et al., 2011). Ultimate substrate methane yields were calculated by taking the chemical components of the organic waste fraction into consideration as well as the corresponding maximum methane yield of the chemical components.

2.3. Continuously stirred tank reactor set-up and monitoring

The experiment was carried out in two continuously stirred tank reactors (CSTRs), R1 and R2, each with a working volume of 7.5 L and an operating temperature of 55°C. The temperature was kept constant by circulating hot water in the outer glass chambers of the reactors. Reactor feeding was conducted by an automated system connecting the feed tank via a pump leading into the reactor (pictures of the sample preparation and system set-up are provided in the supplementary information). Reactor mixing during the operation was performed by an automated stirring system running at 150 rpm with 2-minute on/off interval throughout the experiment. Each reactor was equipped with a liquid sampling port, a gas sampling port and an automatic stirring control unit. The volume of gas production was measured using a liquid displacement gas metering system, as described by Angelidaki et al. (1992). Reactor digestate and gas samples were taken twice a week to measure VFA, ammonia, pH and biogas composition.

The reactors were operated for a period of 230 days and divided into five phases in order to study the effect of changes in the co-substrate’s VS ratio at various HRTs on biogas production. Table 1 shows an overview of the process parameters during the operation of the reactors. The TS and VS of the co-substrates were analysed in duplicate, whereas the OLR and HRT were average values in steady state conditions. During the first phase, only sludge prepared by mixing primary and secondary sludge, 1:1 (v/v %), was fed into both reactors as the substrate, in order to achieve a baseline biogas generation rate without co-digestion. During phases 2 through to 5, the two reactors were fed with different sewage sludge, food waste, grass clipping and garden waste mixing ratios.

Feedstock composition was chosen based on current application in wastewater treatment plants, where sewage sludge is digested alone, compared to the co-digestion of sludge and urban waste with a view to maximising biogas production. Accordingly, the fraction of mixed sludge was fixed at 10% VS of the total VS in the feedstock for both reactors, which was enough to ease pumping of the reactor feedstock. Similarly, the green waste contained a fixed ratio of 70% VS grass clippings and 30% VS garden waste, in order to avoid highly woody material, branches and soil in the garden waste. The % VS ratios of food waste: green waste was set as 75:25 and 50:50 for R1 and R2, respectively. Based on the criteria above, a mixture of sludge, food waste, grass clippings and garden waste was fed with corresponding VS ratios of 10:67.5:15.75:6.75 for R1 and 10:45:31.5:13.5 for R2, respectively. The feedstock ratio remained constant during the reactor experiments, whereas the HRT of the reactors was changed. During phase 1+2, phase 3, phase 4 and phase 5, the HRTs were 30, 20, 15 and 10, respectively. Due to pipe size limitations in relation to the lab-scale reactors and constraints in feedstock pumping, the TS content of co-substrates was diluted by a dilution factor of 1.25 in R1 and 1.29 in R2. The difference in the dilution factor was due to the high amount of green waste in R2.

As the amount of particulate matter and soils in the feed was relatively high, and the mixing of the reactors was relatively weak, we anticipated the accumulation of some solids inside the reactor. Therefore, sediment build-up inside the reactors was quantified. At the end of the experimental period, 12% of the working volume was found to be filled up with sediments, containing soil and lignocellulosic materials
partially composing the substrates (especially grass clippings and garden waste). For this reason, correction factors for the HRT, based on actual working volume and the average daily feed rate, were taken into consideration by assuming a linear build-up of sediments when analysing the results (see Table 1). The OLR rate was also corrected based on measured TS/VS of feedstock prepared each time and actual daily feed pumping. Reactor process parameters, such as the ammonia, VFA, pH and methane content of the biogas produced during the reactor operation, were measured twice a week throughout the experiment.

Table 1 – Co-digestion of mixed sludge with food and green waste: overview of process parameters, VS mixing ratios of substrates, characteristics of mixed sludge and co-substrates. The TS and VS co-substrate analyses were conducted in duplicate, whereas the organic loading rate (OLR) and hydraulic retention time (HRT) data were determined in steady-state conditions. Numbers in brackets give the standard deviation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Phase 1 (day 0-74)</th>
<th>Phase 2 (day 75-130)</th>
<th>Phase 3 (day 131-164)</th>
<th>Phase 4 (day 165-204)</th>
<th>Phase 5 (day 205-230)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>R1</td>
<td>R2</td>
<td>R1</td>
<td>R2</td>
<td>R1</td>
</tr>
<tr>
<td>HRT</td>
<td>Days</td>
<td>30</td>
<td>30</td>
<td>20</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>55</td>
<td>55</td>
<td>55</td>
<td>55</td>
<td>55</td>
</tr>
<tr>
<td>Corrected working</td>
<td>L</td>
<td>7.0</td>
<td>7.4</td>
<td>7.0</td>
<td>7.4</td>
<td>7.0</td>
</tr>
<tr>
<td>working volume</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Feed</td>
<td>L d⁻¹</td>
<td>~0.250</td>
<td>~0.250</td>
<td>0.262</td>
<td>0.381</td>
<td>0.508</td>
</tr>
<tr>
<td>HRT¹</td>
<td>D</td>
<td>~28</td>
<td>~29.60</td>
<td>26.72</td>
<td>18.37</td>
<td>13.78</td>
</tr>
<tr>
<td>OLR²</td>
<td>g VS L⁻¹d⁻¹</td>
<td>0.65 (0.03)</td>
<td>0.62 (0.04)</td>
<td>2.55 (0.21)</td>
<td>2.25 (0.15)</td>
<td>3.91 (0.08)</td>
</tr>
<tr>
<td>TS</td>
<td>% ww</td>
<td>2.85 (0.1)</td>
<td>2.85 (0.1)</td>
<td>7.97 (0.1)</td>
<td>8.19 (0.3)</td>
<td>8.33 (0.1)</td>
</tr>
<tr>
<td>VS</td>
<td>% ww</td>
<td>2.02 (0.1)</td>
<td>2.02 (0.1)</td>
<td>7.06 (0.1)</td>
<td>6.92 (0.2)</td>
<td>7.3  (0.1)</td>
</tr>
<tr>
<td>VS/TS</td>
<td>% TS</td>
<td>70.93 (4)</td>
<td>70.93 (4)</td>
<td>88.50 (2)</td>
<td>84.25 (4)</td>
<td>87.19 (2)</td>
</tr>
<tr>
<td>Mixed sludge</td>
<td>% VS</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
</tr>
<tr>
<td>Food waste</td>
<td>% VS</td>
<td>0</td>
<td>0</td>
<td>67.50</td>
<td>45.00</td>
<td>45.00</td>
</tr>
<tr>
<td>Grass clippings</td>
<td>% VS</td>
<td>0</td>
<td>0</td>
<td>15.75</td>
<td>31.50</td>
<td>15.75</td>
</tr>
<tr>
<td>Garden waste</td>
<td>% VS</td>
<td>0</td>
<td>0</td>
<td>6.75</td>
<td>13.50</td>
<td>6.75</td>
</tr>
</tbody>
</table>

The volume of sediments accumulated in the CSTR in the final phase of the reactor operation was found to be 0.9 L.

²: values based on corrected working volume due to accumulation of soil, sand and sediments in a steady state.

2.4. Gas chromatographic analysis

Methane generation during the BMP assay was analysed by taking gas samples over time, followed by manual injection on a thermos-scientific trace gas chromatograph (TRACE 1310 GC) equipped with a thermal flame ionisation detector (FID). The oven temperature was set at 160°C and the instrument method used was SOP SSL FID, while the data treatment method was carried out via methane Q-plot 8m.prometh.

Biogas composition (content of CH₄ and CO₂) during CSTR operation was determined by thermal conductivity detector gas chromatography (TCD-GC) (GC82 MikroLab, Aarhus A/S, DK) equipped with a packed column for compound separation (main column: 1.1m x 1/16” Molsive 137 + 0.7m x ¼” Lithiumsorb
The injector, detector and oven temperatures were set at 50°C. The carrier gas, hydrogen, flowing over the column was at 40 ml/min (10 ml/15 sec on a flow meter).

3. Results and discussion

3.1. Substrate characterisation

The physicochemical characteristics of substrates used as feedstock in the CSTR co-digestion process are shown in Table 2. The VS content (expressed as a percentage of TS) of food waste and grass clippings was higher than for the garden waste and sludge, due partly to the high content of inorganic material in the garden waste and recalcitrant (cell biomass in secondary sludge) matter in the sludge. The high C/N ratio in food waste compared to mixed sludge made it possible to adjust the C: N ratio when using them as co-substrates in AD. The VFA content of the input substrates was less than 5% of wet weight (ww), and ammonia concentration was insignificant (Table 2). The highest amounts of fats and proteins were found in the food waste samples, possibly originating from fatty materials such as various kinds of meat, oils and cheese.

Table 2 – Physicochemical characteristics of the substrates used in the CSTR experiments. TS, VS, total carbon, total nitrogen and lipids were analysed in triplicate, whereas the rest of the parameters were performed in duplicate. Numbers in brackets give the standard deviation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Food waste</th>
<th>Grass clippings</th>
<th>Garden waste</th>
<th>Primary sludge</th>
<th>Secondary sludge</th>
</tr>
</thead>
<tbody>
<tr>
<td>DM</td>
<td>g/kg ww</td>
<td>160 (1.1)</td>
<td>211 (1.84)</td>
<td>369 (1.12)</td>
<td>38 (0.51)</td>
<td>17 (0.28)</td>
</tr>
<tr>
<td>VS</td>
<td>g/kg ww</td>
<td>149 (0.93)</td>
<td>184 (1.66)</td>
<td>249 (1.60)</td>
<td>28 (0.45)</td>
<td>12 (0.27)</td>
</tr>
<tr>
<td>VS % DM</td>
<td></td>
<td>93.41 (0.13)</td>
<td>87.28 (0.11)</td>
<td>67.62 (0.37)</td>
<td>72.94 (0.22)</td>
<td>69.08 (0.49)</td>
</tr>
<tr>
<td>Carbon (total) % DM</td>
<td></td>
<td>50 (10)</td>
<td>46 (9.2)</td>
<td>35 (7)</td>
<td>39 (7.8)</td>
<td>34 (6.8)</td>
</tr>
<tr>
<td>Nitrogen (total) % DM</td>
<td></td>
<td>3.5 (0.7)</td>
<td>3.9 (0.78)</td>
<td>1.6 (0.32)</td>
<td>2.2 (0.44)</td>
<td>6 (1.2)</td>
</tr>
<tr>
<td>Lipids g/kg ww</td>
<td></td>
<td>30.56 (2.14)</td>
<td>12.48 (0.87)</td>
<td>9.05 (0.63)</td>
<td>2.26 (0.16)</td>
<td>0.75 (0.05)</td>
</tr>
<tr>
<td>VFA (total) g/kg ww</td>
<td></td>
<td>2.89 (0.002)</td>
<td>4.03 (0.97)</td>
<td>0.83 (0.04)</td>
<td>1.35 (0.03)</td>
<td>0.07 (0.004)</td>
</tr>
<tr>
<td>Alcohol g/kg ww</td>
<td></td>
<td>5.3 (0.06)</td>
<td>1.07 (0.001)</td>
<td>0.02 (0.002)</td>
<td>0.024 (0.001)</td>
<td>0.002 (0.001)</td>
</tr>
<tr>
<td>TKN g/kg ww</td>
<td></td>
<td>5.20 (0.38)</td>
<td>6.85 (0.31)</td>
<td>5.38 (0.23)</td>
<td>0.99 (0.05)</td>
<td>1.07 (0.17)</td>
</tr>
<tr>
<td>Protein g/kg ww</td>
<td></td>
<td>44.80 (1.72)</td>
<td>35.20 (1.72)</td>
<td>23.60 (1.16)</td>
<td>4.93 (0.25)</td>
<td>8.14 (0.41)</td>
</tr>
<tr>
<td>NH$_3$-N g/kg ww</td>
<td></td>
<td>0.56 (0.16)</td>
<td>0.54 (0.17)</td>
<td>0.43 (0.03)</td>
<td>0.13 (0.02)</td>
<td>0.20 (0.03)</td>
</tr>
</tbody>
</table>

3.2. Biochemical methane potential (BMP) assay

The experimental and theoretical BMPS (NmL CH$_4$/g VS) of individual substrates and co-substrates are shown in Figure 1. While an incubation period of 28 days was used, the cumulative methane yield reached a plateau (stable condition) at day 15, when about 80% of the gas was produced. The theoretical methane potential was calculated based on the assumption of the full degradation of the organic substrate components shown in Table 2. TS, VS, total carbon, total nitrogen and lipids were analysed in triplicate, whereas the rest of the parameters were performed in duplicate. Biodegradability is defined as the ratio of actual BMP to T-BMP multiplied by 100%. The maximum methane yield in the batch tests was obtained for
food waste (579 NmL CH$_4$/g VS), while garden waste presented low degradability (160 NmL CH$_4$/g VS). The measured feedstocks’ BMPs were comparable with values reported in the literature for similar materials, namely at 500-700 NmL CH$_4$/g VS for food waste, 160 -390 NmL CH$_4$/g VS for grass and garden waste and up to 590 NmL CH$_4$/g VS for the primary sludge (Chynoweth et al., 1993; Nallathambi Gunaseelan, 1997; Zhang et al., 2007).

Source-segregated food waste collected from a canteen had a BMP of between 467 and 529 mL/g VS (Browne and Murphy, 2013). Maximum BMP value and better degradability was obtained for food waste possibly owing to the abundance of fats and proteins in the waste compared to the other substrates.

BMP values measured for secondary sludge, grass clippings and garden waste were significantly lower than the respective theoretical ones, most likely due to lignocellulosic components in plant materials and recalcitrant cell biomass in the secondary sludge. The BMP results also demonstrate how mixing different substrates can potentially influence the performance of the co-digestion process, as seen when comparing results for individual substrates with three different mixing ratios (70:30, 50:50, 30:70 on VS basis) of grass and garden, which provided methane yields of 283 NmL CH$_4$/g VS, 249 NmL CH$_4$/g VS and 201 NmL CH$_4$/g VS, respectively. In particular, it is evident that the higher the amount of garden waste, the lower the obtained BMP because of the increased content of lignocellulosic material in garden waste compared to grass. Finally, the specific methane yield of R1 feedstock is higher than the BMP for R2 because of the greater content of food waste.

**Figure 1** – Experimental and theoretical biochemical methane potential (BMP) of the substrates and co-substrates used in our CSRT experiments. Error bars report standard deviation calculated based on three replicates.

### 3.3 Co-digestion process in CSTR

Reactor performance parameters, methane productivity and specific methane yield are shown in Figure 2, in which we note that methane productivity increased considerably when HRT decreased. For both the R1 and R2 mixing ratios, the addition of urban organic waste as a co-substrate in the feedstock improved methane productivity considerably, as seen when passing from phase 1 (mixed sludge only) to phase 2 (co-substrates). The R1 mixing ratio showed considerably higher (~21%) methane productivity compared to R2 at 30 days HRT, owing to the larger content of food waste, which had the highest BMP (see Figure 1). Overall, R1 showed 10-20% higher productivity compared to R2. Similarly, R1’s mixed substrates
provided a substantially higher methane yield compared to R2 at all HRT; the difference was estimated to be 9, 13, 14 and 14% for 30, 20, 15, 10 days HRT, respectively. The specific methane yield obtained in phase 1 with sludge-based feed was 287 NmL/g VS; the yield increased considerably to 424 NmL/g VS in R1 and 391 NmL/g VS in R2 during phase 2. Studies have shown that 50:50 of food waste and activated sludge provided a yield of 321 ml CH₄/g VS (Heo et al., 2004) at HRT of 13 days and an OLR of 2.43 g VS/(L. day).

Figure 2 – Co-digestion of mixed sludge with food and green waste: development of the organic loading rate (OLR), yield and productivity in R1 and R2 during phase 1 (0-75 day), phase 2 (76-130 days), phase 3 (131-164), phase 4 (165-203) and phase 5 (204-230) days.
When increasing the feed rate, i.e. lower HRT from 30 to 20 and 15 days, respectively, the methane yield remained relatively constant at around 430 NmL/g VS and 376 NmL/g VS in R1 and R2, respectively. A significant decrease in methane yield was observed in both reactors when the HRT was decreased to 10 days, owing to the overloading and washing-out of the microbial community. At 10 days HRT, the specific methane yield dropped by 17% compared to 15 days of HRT, i.e. declining from 430 to 359 NmL/g VS in R1, and from 375 to 315 NmL/g VS in R2. A summary of the reactor performance results and operational parameters in steady-state conditions is provided in Table 3.

Table 3 – Summary of reactor performance parameters for all experimental phases performed in R1 and R2 in steady-state conditions. The given parameters were average values in steady-state conditions. Numbers in brackets give the standard deviation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Phase 1</th>
<th>Phase 2</th>
<th>Phase 3</th>
<th>Phase 4</th>
<th>Phase 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>55</td>
<td>55</td>
<td>55</td>
<td>55</td>
<td>55</td>
</tr>
<tr>
<td>HRT</td>
<td>Days</td>
<td>30</td>
<td>30</td>
<td>20</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>Feedstock</td>
<td></td>
<td>Mixed sludge</td>
<td>Co-substrate</td>
<td>Co-substrate</td>
<td>Co-substrate</td>
<td>Co-substrate</td>
</tr>
<tr>
<td>Steady state</td>
<td>Days</td>
<td>37-62</td>
<td>112-130</td>
<td>149-163</td>
<td>193-204</td>
<td>220-230</td>
</tr>
<tr>
<td>Reactor</td>
<td></td>
<td>R1</td>
<td>R2</td>
<td>R1</td>
<td>R2</td>
<td>R1</td>
</tr>
<tr>
<td>Productivity</td>
<td>mL CH₄/(L day)</td>
<td>186 (12)</td>
<td>178 (16)</td>
<td>1078 (61)</td>
<td>895 (65)</td>
<td>1695 (56)</td>
</tr>
<tr>
<td>Methane yield</td>
<td>mL CH₄/(g VS)</td>
<td>287 (19)</td>
<td>291 (31)</td>
<td>424 (20)</td>
<td>391 (15)</td>
<td>433 (13)</td>
</tr>
<tr>
<td>Methane content</td>
<td>CH₄%</td>
<td>69 (1)</td>
<td>68 (1)</td>
<td>60 (1)</td>
<td>59 (2)</td>
<td>61.0 (1)</td>
</tr>
<tr>
<td>pH</td>
<td></td>
<td>7.6 (0.2)</td>
<td>7.6 (0.28)</td>
<td>7.84 (0.04)</td>
<td>7.83 (0.02)</td>
<td>7.88 (0.02)</td>
</tr>
<tr>
<td>NH₃-N</td>
<td>mg/L</td>
<td>718 (30)</td>
<td>715 (48)</td>
<td>1722 (18)</td>
<td>1517 (79)</td>
<td>1724 (24)</td>
</tr>
<tr>
<td>Total VFA</td>
<td>mg/L</td>
<td>25 (7)</td>
<td>28 (1)</td>
<td>328 (13)</td>
<td>124 (13)</td>
<td>590 (13)</td>
</tr>
<tr>
<td>Acetate</td>
<td>mg/L</td>
<td>21.05 (6.32)</td>
<td>25 (1)</td>
<td>287 (12)</td>
<td>100 (12)</td>
<td>355 (7)</td>
</tr>
<tr>
<td>Propionate</td>
<td>mg/L</td>
<td>0.27 (0.24)</td>
<td>0 (0)</td>
<td>35.42 (1)</td>
<td>22 (1)</td>
<td>223 (3)</td>
</tr>
<tr>
<td>Butyrate</td>
<td>mg/L</td>
<td>3.24 (0.63)</td>
<td>3 (0.3)</td>
<td>6.1 (0.4)</td>
<td>3 (0.05)</td>
<td>10 (0.6)</td>
</tr>
<tr>
<td>Valerate</td>
<td>mg/L</td>
<td>0.0 (0.0)</td>
<td>0 (0)</td>
<td>0 (0)</td>
<td>3 (2)</td>
<td>12 (2)</td>
</tr>
</tbody>
</table>

Methane yield obtained from the CSTR experiments was lower than both the BMPs estimated theoretically and by means of batch assays (Figure 1). This is of course as expected, because by definition the BMP represents maximum practically achievable methane production at an “indefinite” degradation time. Moreover, the methane yield in CSTR experiments will always permit some degradable organic matter to leave the reactor with the effluent. Continuous reactor experimentation is the best way to test if the substrate mixture has an optimal composition. For example, if the substrate mixture lacks some...
nutrients/minerals, or has too low N, the effect of these deficiencies may not be seen in the batch test due to optimised media composition (BA media), or due to sufficient nutrient residue from the healthy inoculum. In a continuous reactor operation, the supplement from inoculum will diminish after 2 HRT, and the effect of nutrient limitations may be noticed.

The results for specific methane yields are comparable to values previously reported for the AD of WWTP sludge in Malmo, which was reported as 270 NmL CH4/g VS (Jansen et al., 2004). Co-digestion of sewage sludge and the organic fraction of municipal solid waste (75:25 v/v) in CSTR and UASB provided a yield of 400 – 600 NmL CH4/g VS (Sosnowski et al., 2003), which shows that the results are comparable even though yield in the latter case was higher, albeit this may be due to the nature of the substrates.

The concentration of ammonia and total VFA was considerably higher during co-substrate AD (both R1 and R2) compared to the digestion of sludge as a single substrate (Figure 3). After a significant increase when passing from phase 1 (only mixed sludge) to phase 2 (co-digestion), the concentration of ammonia decreased slightly as HRT decreased, possibly owing to the adaptation of the microbial community to changes in process conditions. On the other hand, for the same HRT but with different mixing ratios, the ammonia concentration in R1 was 4-12% higher than in R2, an exception being the 10 days HRT (phase 5) where there was no significant difference. The high content of ammonia in R1 relative to R2 in all phases could be due to the composition of substrates (Table 2), since the feedstock in R1 has higher protein content than R2. The obtained results demonstrate that the total VFA and ammonia-N concentrations were below the inhibition limit of 3100 mg-N/L (Mata-Alvarez et al., 2000), thus indicating the occurrence of good microbial activity.

As shown in Figure 3, methane content in the produced biogas dropped significantly in both reactors when passing from phase 1 (~70%) to phase 2 (~60%); however, it remained constant at ~60% in both R1 and R2 during the subsequent changes in HRT. The pH values increased in both R1 and R2 when the feedstock was changed (from phase 1 to phase 2), while small changes were then seen during the following phase 2 through to phase 5. The increase in pH values during a change in feedstock (from phase 1 and phase 2) could be attributed to the combined effect of VFA and ammonia accumulation.

The content of VFA components is shown in Figure 4 for both R1 and R2 and for all experimental operation phases. In phase 1, the concentration of total VFA more or less corresponded to the concentration of acetate, whereas the concentrations of propionate, butyrate and valerate were insignificant. Feeding of co-substrates in phase 2 caused an increase in acetate concentration, followed by increasing propionate concentration. The most dominant VFA components during phase 2 through to phase 5 were acetate and propionate, whereas the concentration of butyrate and valerate values remained insignificant throughout the whole experiment. In general, the concentration of total VFA in R1 was higher than in R2 due to the higher fraction of food waste in the feedstock.

The trade-off between methane productivity and specific methane yield is shown in Figure 5. In R1, the specific methane yield was rather constant during phases 2, 3 and 4 (30, 20 and 15 days HRT respectively), whereas a slight drop was recorded for R2 during the same phases. In both reactors R1 and R2, the specific methane yield declined significantly at 10 days HRT, even though the results demonstrate an increase in methane productivity. It can thus be concluded that, in steady-state conditions, optimum reactor performance in terms of methane productivity and specific methane yield was obtained at 15 days HRT, when other parameters, such as the concentration of ammonia and VFA, pH and methane content in biogas, also showed as being rather stable.

The co-digestion of urban waste (food, grass and garden waste) with wastewater treatment plant sludge increased methane production and considerably improved methane yield in comparison to the AD of 100% sludge. Hence, greater utilisation of organic fractions of urban waste may boost the efficiency of AD in wastewater treatment plants while achieving sustainable resource utilisation. This will change the future of many struggling WWTP companies, as they will be able to receive locally available urban waste into the existing system, without any dramatic changes in infrastructure, to produce sustainable energy that could benefit society by providing clean electricity and heating.
Figure 3 – Co-digestion of mixed sludge with food and green waste: development of NH3-N, pH, VFA and methane content in R1 and R2 during phase 1 (0-75 day), phase 2 (76-130 days), phase 3 (131-164), 4 (165-203) and phase 5 (204-230) days.
Figure 4 – Co-digestion of mixed sludge with food and green waste: development of the composition and concentration of volatile fatty acids.

Figure 5 – Co-digestion of mixed sludge with food and green waste: trade-off between methane yields versus productivity in steady-state conditions.
4. Conclusion

The addition of food waste, grass clippings and garden waste to mixed sludge as co-substrates increased methane yield by 48% and 35% in reactors R1 and R2 compared to the AD of only mixed sludge at 30 days HRT, with the difference being attributable to the different shares of food/green waste in the feedstock. However, methane yield remained constant as HRT decreased (30, 20, 15) but dropped markedly when HRT was decreased to 10 days. The trade-off between yield and productivity was achieved at 15 days HRT with stable process parameters.

Acknowledgments

We would like to thank Hector Garcia and Hector Diaz for their technical support throughout the research work. Besides, we would also like to thank Jonas Høeg Hansen (Teknologisk Institut, Stendrup, Kolding), David Croft and Martin Riis Weisbjerg (Aarhus Universitet Foulum, Foulumgård), Preben Haulund (Vogelsang A/S, Denmark), Susanne Rasmussen and Dorthe Hovland (Lundtofte WWTP), Thomas Guildal (Avedøre WWTP), Jonas Nedeskov (Amager Research Center), Ylva Eriksson (Va Syd, Sjölunda WWTP) and Jan Dahl (DTU Lyngby Campus Canteen) for helping us during sample collection and pre-treatment. The authors would also like to acknowledge the DTU-Environment student Nikolaj From Petersen for his contribution during the experiments in this study.

The project was supported by the Danish Council for Strategic Research (DSF) under the “Strategic Research in Sustainable Energy and Environment” research programme through the project “Optimisation of value chains for biogas production in Denmark (BioChain)”. 
List of references


Supplementary Information

Figure S1 – Co-digestion of mixed sludge with food and green waste: development of NH$_3$-N and CH$_4$ concentrations during phase 1 (0-75 day), phase 2 (76-130 days), phase 3 (131-164), 4 (165-203) and phase 5 (204-230) days.

Figure S2 – Co-digestion of mixed sludge with food and green waste: development of pH and total volatile fatty acid (VFA) concentrations during phase 1 (0-75 day), phase 2 (76-130 days), phase 3 (131-164), 4 (165-203) and phase 5 (204-230) days.
Figure S3 – Experimental set-up for the co-digestion process in a continuously stirred tank reactor.

Figure S4 – Substrates used in the co-digestion process (food waste, grass clippings, garden waste and sludge).
Figure S5 – Sampling and pre-treatment of substrates (food waste, grass clippings and garden waste).