Synthesis of Biogas Supply Network Based on Experimental Data from Lab-Scale Anaerobic Digestion of Sewage Sludge and Organic Waste

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The aim of this work was to perform synthesis of biogas supply network based on the experimental data from laboratory-scale anaerobic digestion (AD) experiments. Experiments were performed with various types of organic waste and sewage sludge, such as different types of sewage sludge from municipal and industrial wastewater treatment plants, waste from vegetable oil, wine, dairy and meat processing industry, lignocellulosic waste and bio-plastic waste. The synthesis problem was formulated as a mixed-integer linear program (MILP) and was solved by using General Algebraic Modelling System (GAMS) software with the main objective of maximizing economic profit. The synthesis is demonstrated on an illustrative case study of up to three biogas plants in Slovenia. Three scenarios were considered with different prices of waste materials. The study showed good agreement between the model and experimental results. Results demonstrated that biogas production could be profitable by using considered waste materials only if biogas plant was paid for processing the substrates. Up to 6 GWh/y of electricity could be produced with the profit of about 2·10^6 $/y where prices of waste materials were around 80 $/t.

1. Introduction

The world’s energy supply is still predominantly based on fossil fuels (Chua et al., 2015). The use of fossil energy sources in the future is uncertain due to their non-renewability and unsustainability, fluctuations in prices, and the pollution that they cause to environment (Soria-Verdugo et al., 2017). Among alternatives to fossil fuels are biofuels including biogas, which can efficiently contribute to reduction of greenhouse gas (GHG) emissions and sustainable development (Bora et al., 2020). Biogas is mainly produced by the decomposition of biomass and waste during anaerobic digestion (AD), whereby biogas yield and methane content in biogas are highly dependent on the type of substrate, as well they are crucial for economic operation of biogas plants. Efficient supply chain of feedstocks and optimization of operating parameters are important factors for designing profitable biogas production (Kucharska et al., 2018). Especially storage and transportation of substrates and digestate create high costs (Plana and Noche, 2016). To produce products with higher added value, the digestate could be separated into solid and liquid fractions. Solid fraction could then be used for biochar production via pyrolysis process, while liquid fraction could be used for nutrient recovery in the form of fertilizers (Petrovič et al., 2021).

A supply network is a network of suppliers, manufacturing facilities, warehouses, and distribution centers, that includes several activities through which the products are distributed to customers (Santoso et al., 2005). In the last years, there is increasing demand for designing green supply chains that integrate sustainability and environmentally-friendly activities (Banasik et al., 2018). Several studies could be found in the literature in relation to sustainable supply chain network design. Egieya et al. (2020) synthesized optimal biogas supply networks with different sustainability objectives, while Correia et al. (2017) reviewed the supply chain maturity models with sustainability concerns. Most of the studies were performed on literature data for chosen substrates, while studies considering real data for the substrates and their location, are less common.

Identified research gaps and the novelty of this work is biogas supply chain optimisation based on real data. Where possible, the data were obtained from laboratory AD experiments, otherwise data specific to considered

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location were considered. The synthesis of biogas supply network was performed applying and slightly modifying the mixed-integer linear programming (MILP) model called BIOSOM (Egieya et al., 2019). The model was solved using CPLEX solver in GAMS on the case study of up to three biogas plants in Slovenia. The data regarding the biogas potential and methane content were obtained from lab-scale AD experiments performed with various feedstocks, such as different types of sewage sludge and organic waste that are available at selected locations. AD experiments were conducted under mesophilic conditions, while fermentation was additionally enhanced by adding the cow rumen fluid microorganisms into digestate mixtures. All other data required for the model, such as amounts, prices, harvesting period, etc. were determined based on the information available for each location.

2. Materials and methods

In this section, first lab-scale anaerobic digestion (AD) experiments are described, and further mathematical model used for the synthesis of biogas supply networks is presented.

2.1 Experimental work - Anaerobic digestion experiments

To obtain the data required for the mathematical model of biogas supply network synthesis, batch AD experiments with 17 different feedstocks were firstly performed. The following feedstocks were analyzed for their biogas production potential, methane content, density, dry matter and volatile matter (VM) content: i) different sewage sludge (SS) types (SS from municipal wastewater treatment plant (WWTP), SS from WWTP of vegetable oil industry, SS from WWTP of food processing industry and SS from WWTP of meat processing industry), ii) waste from vegetable oil industry and oil extraction (bleaching earth, filtration additive, grape seeds press cake, pumpkin seeds press cake), iii) lignocellulosic waste (T. latifolia grass, sawdust, hemp stalks, paper mill sludge), iv) bioplastic waste (bioplastic bags and bioplastics cutlery), v) waste from wine industry (grape pomace), vi) waste from dairy industry (buttermilk), and vii) waste from meat processing industry (filtration sludge obtained from dissolved air flotation). Due to recalcitrance of lignocellulosic waste, sawdust, hemp stalks and paper mill sludge were chemically pre-treated by 10 v.% citric acid (2 h, 100 °C) to improve fermentation process. To enhance degradation and biogas production, bioplastic waste was dissolved by 2 M NaOH solution (48 h), and the pH was then neutralized by 2 M HCl solution. Substrates such as grape seeds press cake, pumpkin seeds press cake, T. latifolia grass, bioplastic bags and bioplastics cutlery were pretreated only physically (by grinding and cutting) to obtain smaller pieces of sample. The mono-digestion experiments were conducted in 0.25 L batch reactors (~180 mL of working volume) with a retention time of 40 days. The reaction mixtures (160 g of wet basis) were prepared in two parallels with the inoculum/substrate ratio of 2:1 and dry matter content of 4.5 wt.%. Mixtures contained 2.4 g of the substrate and 4.8 g of inoculum on a dry basis, and a buffer solution (Angelidaki et al., 2009) was added to dilute the reaction mixtures to the selected dry matter content. To enhance fermentation process, 25 mL of rumen fluid was added to each reaction mixture. Before AD experiments, the reaction mixtures were flushed with inert argon gas for 30 s to achieve anaerobic conditions and the reactors were then placed in a heating bath at a constant temperature of 42 °C. The flasks were hand-mixed daily for approximately 20 s. Besides feedstocks, blank assays of inoculum and rumen fluid were tested at the same digestion conditions. All the assays were performed in duplicate, and the average values are reported in the continuation. Biogas production was measured daily by a water displacement method. The CH4 content was determined by Optima7 biogas analyzer. To check the stability of the reactor system, the pH value was measured occasionally. Experimental setup was as described in the paper by Bedoć et al. (2019).

2.2 Mathematical modeling - synthesis of biogas supply network

The synthesis of biogas supply network was performed based on the experimental data from lab-scale AD experiments. The main purpose of the supply network is to generate electricity from biogas profitably by considering monthly variations in feedstocks availability. Supply network model was slightly adapted from the previous work (Egieya et al., 2019) and was formulated as a mixed-integer linear programming (MILP) model solved in GAMS. The MILP model includes four layers (Egieya et al., 2019): i) harvesting and collection of feedstocks, ii) primary conversion such as AD, iii) secondary conversion such as cogeneration and digestate dewatering, and iv) demand for electricity and other products. The schematic representation of biogas supply network is shown in Figure 1. The model selects among different types and quantities of feedstocks and products, the land area required for growing feedstocks, different potential locations of biogas plants, sizes of conversion technologies, transportation modes and logistics. The detailed description of the model and all the equations used can be found in work by Egieya et al. (2019).

The synthesis of biogas supply chain is represented as an illustrative case study of biogas plants located in northeast Slovenia on three different locations, such as in Egieya et al. (2019). The total area of each harvesting...
site of substrates is 250 km². Besides three different locations and sizes of conversion facilities, the model could select among 17 different feedstocks and two transportation modes (road transport or pipeline) for substrates delivering to biogas plants.

Substrates can be selected within a radius of up to 9 km from the biogas facilities and digestate can be used at the same locations as fertilizers for crops. Dry matter (DM) content in AD reactors was specified to be between 2 and 13 wt.% as in (Egieya et al., 2019). Water recovered from digestate dewatering is recycled in the anaerobic digestion unit, while dewatered digestate with DM content > 23 wt.% is used for agricultural purpose as a soil enhancer. Each biogas plant can produce up to $999$ kW, with the price of electricity sold to the grid 182.9$/MWh. Three scenarios were shown based on the prices of waste materials (from the most negative to 0 $/t).

The input data used in mathematical model are the amounts of the substrates, their availability at each location, dry matter contents, densities, prices of substrates, biogas yields and average methane contents and are presented in Table 1 and Figure 2.

3. Results and Discussion

This section first presents the main results obtained from experimental work, and further the results from mathematical modeling are described.

3.1 Results of experimental work

The biogas yield per dry mass (d.m.) and average methane content (v.%) measured for tested samples are shown in Figure 2. The highest biogas yields in laboratory experiments were obtained with flotation sludge ($1,343$ m$^3$/t), filtration additive ($928$ m$^3$/t) and pumpkin seeds press cake ($740$ m$^3$/t), and the lowest in the cases

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**Figure 1: Schematic presentation of biogas supply network (diagram modified from Egieya et al. (2019))**

**Figure 2: Biogas yields and average CH$_4$ contents obtained from laboratory experiments**
of sawdust (82 m³/t), spent bleaching earth (96 m³/t) and SS from food processing industry (111 m³/t). The poor performance of the last three substrates may be due to various reasons. Low content of volatiles was characteristic also for spent bleaching earth and for SS from oil industry (Table 1). In general, a C/N ratio of 15–35 should be ensured to run AD process without incurring the risks of lack in nutrients (Pellera and Gidarakos, 2017). Although for tested substrates (except for bleaching earth, hemp stalks, SS from oil industry and T. Latifolia grass) relative low C/N ratios were characteristic, the AD process according to the obtained results was not affected. But to achieve higher biogas yields the co-digestion with substrates with higher carbon content would be reasonable. The substrate with the lowest biogas yields also gave the lowest methane content. The highest average methane content (>55 v.%) was measured in biogas obtained from SS from WWTP of meat processing industry, grape pomace, bioplastic bags, filtration additive acquired from vegetable oil industry and flotation sludge from meat processing industry.

Table 1: The results of experimental work (left) and assumed input data (right) for mathematical model

<table>
<thead>
<tr>
<th>Substrate</th>
<th>DM (wt.%)</th>
<th>VM</th>
<th>C/N ratio</th>
<th>Density (kg/m³)</th>
<th>Substrate (t/y)</th>
<th>Price1 ($/t)a</th>
<th>Availability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bioplastics bags</td>
<td>100</td>
<td>99.8</td>
<td>2.6</td>
<td>416.6</td>
<td>28.6</td>
<td>81.0</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Bioplastics cutlery</td>
<td>100</td>
<td>71.9</td>
<td>2.1</td>
<td>688.6</td>
<td>28.6</td>
<td>81.0</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Bleaching earth</td>
<td>68</td>
<td>37.1</td>
<td>50.0</td>
<td>736.2</td>
<td>/</td>
<td>/</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Buttermilk</td>
<td>9</td>
<td>92.7</td>
<td>4.0</td>
<td>1,044.4</td>
<td>/</td>
<td>1187</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Filtration additive</td>
<td>94</td>
<td>72.1</td>
<td>3.1</td>
<td>667.8</td>
<td>/</td>
<td>/</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Flotation sludge</td>
<td>11</td>
<td>95.4</td>
<td>4.2</td>
<td>996.8</td>
<td>/</td>
<td>/</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Grape pomace</td>
<td>25</td>
<td>94.6</td>
<td>2.1</td>
<td>2,245.8</td>
<td>1,800</td>
<td>1,800</td>
<td>Sep – Oct</td>
</tr>
<tr>
<td>Grape seeds cake</td>
<td>92</td>
<td>96.6</td>
<td>4.4</td>
<td>764.3</td>
<td>450.0</td>
<td>450.0</td>
<td>Sep - Oct</td>
</tr>
<tr>
<td>Hemp stalks</td>
<td>27</td>
<td>96.6</td>
<td>44.3</td>
<td>408.1</td>
<td>51.6</td>
<td>98.5</td>
<td>Aug – Oct</td>
</tr>
<tr>
<td>Paper sludge</td>
<td>47</td>
<td>96.9</td>
<td>2.8</td>
<td>191.5</td>
<td>/</td>
<td>/</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Pumpkin seeds cake</td>
<td>93</td>
<td>89.1</td>
<td>4.3</td>
<td>678.8</td>
<td>222.0</td>
<td>222.0</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>Sawdust</td>
<td>68</td>
<td>99.1</td>
<td>3.1</td>
<td>276.5</td>
<td>540.0</td>
<td>540.0</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>SS-food industry</td>
<td>51</td>
<td>33.2</td>
<td>2.8</td>
<td>879.6</td>
<td>/</td>
<td>/</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>SS-meat industry</td>
<td>27</td>
<td>86.4</td>
<td>3.8</td>
<td>1,138.0</td>
<td>/</td>
<td>10.0</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>SS-municipal WWTP</td>
<td>21</td>
<td>80.9</td>
<td>5.8</td>
<td>940.0</td>
<td>7,200</td>
<td>7,200</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>SS-oil industry</td>
<td>37</td>
<td>27.2</td>
<td>13.3</td>
<td>795.0</td>
<td>/</td>
<td>/</td>
<td>Jan – Dec</td>
</tr>
<tr>
<td>T. Latifolia grass</td>
<td>13</td>
<td>88.3</td>
<td>13.4</td>
<td>1,140.7</td>
<td>8,750</td>
<td>8,750</td>
<td>May – Oct</td>
</tr>
</tbody>
</table>

a L - Location
b Positive price represent cases when biogas plant needs to pay for substrates, while negative price indicates that biogas plant gets paid for collecting the substrates

1Prices for the first scenario (SC 1). The prices for the second (SC 2) and third scenarios (SC 3) were the same except for the substrates with the negative sign. For SC 2 the prices were halved as compared to SC 1, while for SC 3 price were 0 $/t.

3.2 Mathematical modeling - case study of a biogas supply network

To synthesize biogas supply network, mathematical programming was used with the objective to maximize economic profit. Three scenarios (SC 1–SC 3) were considered where feedstock prices were changed. SC 1 represents the base case scenario where the prices of the feedstocks were as presented in the last column in Table 1. In SC 2, prices for feedstocks with negative prices were increased and were halved as compared to SC 1, while in SC 3, the price of those waste materials were assumed to be 0 $/t. The results obtained are presented in Figure 3 and Table 2. In both SC 1 and SC 2 the same feedstocks were considered with similar amounts. The following feedstocks were selected from the highest to the lowest amounts: SS-municipal WWTP (20,792.7 t/y), grape seeds cake, buttermilk, pumpkin seeds cake, paper sludge, filtration additive, SS-oil industry, bleaching earth, flotation sludge, SS-food industry, SS-meat industry, bioplastics bags and bioplastics cutlery (18.9 t/y). In SC 3, the same feedstocks were selected as in SC 1 and SC 2, only grape pomace was chosen instead of buttermilk, while amounts of bleaching earth and SS-municipal WWTP were significantly reduced. Figure 3 shows the amounts of selected feedstocks for all the scenarios. For clarity, the values of some feedstocks, such as buttermilk, grape pomace, grape seeds cake, paper seeds cake and SS-municipal WWTP should be multiplied (e.g., buttermilk by 4) to get the amounts of selected feedstocks. The total amounts of feedstocks used were maximal for SC 2 and were about 31.1 kt/y, slightly lower were for SC 1 (25.6 kt/y), while for SC 3 only about 11.8 kt/y of feedstocks were used as shown in Table 2. In SC 1 and SC 2, location 2 was selected as the optimal location for the biogas plant and in SC 3, location 3 was selected.
In SC 1, about 5.0 GWh/y of both electricity and heat were produced, in SC 2 6.0 GWh/y of both electricity and heat, and in SC 3 only 3.2 GWh/y of electricity and 3.1 GWh/y of heat were produced. As the highest amounts of waste materials were used in SC 2, also the highest amount of dry digestate was produced in SC 2 (21.3 kt/y). Similar, the lowest amount of waste materials was used in SC 3 (10.3 kt/y).

The amount of biogas produced in SC 1 was $2.5 \times 10^6$ m$^3$/y which contained 51.9 % methane, in SC 2 $3.0 \times 10^6$ m$^3$/y biogas was produced (methane content of 52.4 %), and in SC 3 the amount of biogas produced was $1.6 \times 10^6$ m$^3$/y with 52.1 % methane. SC 1 and SC 2 yielded positive profit, while SC 3 negative which was mainly due to cost of feedstocks. Investment cost was in correlation with the amount of waste materials and biogas and digestate produced; the highest for SC 2 and the lowest for SC 3. Table 2 also shows that DM in fermenter was between 12.1 for SC 2 and SC 3 and 12.7 % for SC 1.

![Figure 3: Amounts of selected feedstocks for each scenario](image)

**Table 2: The results of mathematical modelling**

<table>
<thead>
<tr>
<th>Waste material (kt/y)</th>
<th>Electricity (GWh/y)</th>
<th>Heat (GWh/y)</th>
<th>Dry digestate (kt/y)</th>
<th>Optimal location</th>
<th>DM in the fermenter (wt.%)</th>
<th>Investment cost ($10^6$)</th>
<th>Profit before tax ($10^6$ $$/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SC 1 25.6</td>
<td>5.0</td>
<td>4.9</td>
<td>18.0</td>
<td>2</td>
<td>12.7</td>
<td>3.8</td>
<td>3.8</td>
</tr>
<tr>
<td>SC 2 31.1</td>
<td>6.0</td>
<td>6.0</td>
<td>21.3</td>
<td>2</td>
<td>12.1</td>
<td>4.2</td>
<td>2.1</td>
</tr>
<tr>
<td>SC 3 11.8</td>
<td>3.2</td>
<td>3.1</td>
<td>10.3</td>
<td>3</td>
<td>12.1</td>
<td>2.8</td>
<td>-0.2</td>
</tr>
</tbody>
</table>

The distribution of cost, which include cost of purchased materials, feedstock cost, storage, transport, maintenance, depreciation, labour, miscellaneous, and other cost is shown in Figure 4 for SC 1 and SC 3, while the values for SC 2 were in between. As it can be seen the highest contribution in SC 1 was due to feedstock revenue (negative cost, mainly due to using waste materials with negative price). On the contrary, in SC 3 feedstocks represented small contribution to the cost (2.1 %). The largest contributions to the cost in both SC 1 and SC 3 were depreciation (8.2 and 34.6 %), maintenance (4.2 and 17.8 %), transportation (3.4 and 4.5 %) and miscellaneous cost (2.7 and 14.2 %). Labour, other, storage cost and cost for purchased materials (mainly electricity) formed small contributions to the total annual cost.

### 4. Conclusions

The results of synthesis of biogas supply network based on the real experimental data, obtained during AD of 17 different substrates, were presented in this work. The results have shown that maximal economic profit for SC 1 and 2 could be obtained in location 2, while for SC 3 in location 3. Between 5 and 6 GWh/y of electricity was suggested to be profitably produced in fermenters with DM content between 12 and 13 wt.% and investment cost of about $4 \times 10^6$. Among available feedstocks, sewage sludge from municipal WWTP, oil, food and meat industry, bioplastics bags, filtration additive, grape pomace, flotation sludge, bleaching earth, paper sludge, pumpkin seeds cake and buttermilk were selected for biogas production achieving optimal supply chain. These feedstocks have also shown promising results in the laboratory experiments. The results obtained indicated that synthesis of biogas supply network using proposed MILP model provide satisfactory solutions. Models could be used as a good decision support tool for preliminary analysis in biogas sector.

Future laboratory work could include pretreatment of waste materials and addition of different agents such as biochar to improve methane yield. Further, supply network model could additionally include parameters, such as C/N ratio and nutrient content in waste materials and digestate, and could consider pretreatment technology.
Figure 4: Cost breakdown a) first scenario, b) third scenario

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