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Published in:
Journal of Cleaner Production

DOI (link to publication from Publisher):
10.1016/j.jclepro.2021.129239

Publication date:
2021

Document Version
Publisher's PDF, also known as Version of record

Journal of Cleaner Production 324 (2021) 129239

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Potential of the sewage sludge valorization in Scandinavia by co-digestion with other organic wastes: A techno-economic assessment

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ARTICLE INFO

Handling editor: Zhen Leng

Keywords:
Biorefinery
Techno-economic analysis
Sewage sludge
Carboxylates
PHA
Anaerobic digestion

ABSTRACT

The aim of this study was to compare the techno-economic feasibility of five different scenarios to valorize sewage sludge in co-digestion with another organic waste. The first three scenarios were based on real full-scale processes in Scandinavia, furthermore two original biorefinery scenarios for carboxylates and biogas production were proposed. Data from the actual plant of one scenario was used in order to calibrate the simulation, creating five realistic models for sludge valorization. Two downstream technologies were simulated for the valorization of carboxylates: anion exchange chromatography and biological separation (through PHA production). Even though none of the models was profitable without subsidies, the biorefinery scenario with biological separation was the most promising one, with the operating costs (2.79 million €) closest to the total revenues (0.8 million €). The use of anion exchange chromatography created an economic bottleneck in the model (57% of the operating costs), suggesting the need for more simple and cost-effective downstream technologies, when developing biorefineries for the valorization of waste streams. Moreover, the importance of public subsidies, gate fees for organic wastes and the amount of treated material were found to be key aspects for the economic feasibility of these facilities treating waste, increasing the total revenues between 2 and 15 times, depending on the modelled scenario.

1. Introduction

In recent years, sewage sludge (SS) generation has increased, ligated to population growth and changing lifestyle. This organic waste constitutes the major by-product from wastewater treatment (Peccia and Westerhoff, 2015) and consists of organic and inorganic compounds. Nevertheless, the specific composition and generated amount of sludge can differ significantly and depend on the type of wastewater and the applied treatment technology (Healy et al., 2015). Different treatments can be applied to sewage sludge in order to valorize it and to reduce its volume; some examples are incineration or anaerobic digestion (AD), which can recover its energy content. Moreover, the remaining fraction after AD contains nutrients (mainly P and N) that can be applied as soil fertilizer. However, energy production from wastes may not always economically feasible. For example, the economic feasibility of waste incineration (waste to energy) depends heavily on the water content of the wastes: the higher the water content, the lower the energy that is recovered (due to the input energy necessary to dewater the waste). Mono incineration of sludge, for example, was found to require more energy than the energetic content of its organic matter (Frijns et al., 2013). For that reason, research on the production of higher value compounds from sludge, such as carboxylates (i.e. volatile fatty acids, VFA) or bioplastics, has increased in recent years (Wu et al., 2021).

Nonetheless, the recalcitrant nature of sludge can hinder the digestion of this substrate and therefore its valorization (Zhou et al., 2015). For that reason, different pretreatment technologies such as thermal (Qiao et al., 2011), chemical (Devlin et al., 2011) or biological (Yang et al., 2010) have been evaluated, in order to optimize AD of the sludge. In fact, the application of thermal pretreatment in sludge before AD increased the revenues from electricity up to 25% in a full-scale wastewater treatment plant, compensating the costs of the pretreatment (Campo et al., 2018).

Another approach to improve sludge valorization via AD is the addition of another organic waste, in order to exploit the microbial synergy of co-digestion (Xie et al., 2017). For instance, previous studies have used organic wastes, such as the organic fraction of municipal solid waste (OFMSW, i.e. food waste) (Gottardo et al., 2015), agricultural or lignocellulosic residues (Elalami et al., 2019), fish sludge (Estevez et al., 2019) or glycerol (Nghiem et al., 2014) in co-digestion with sewage sludge. In fact, sludge co-digestion with OFMSW led to an increase in the methane yield up to 47%, around 380 ml CH_4/gVS (Cabbai et al., 2013),

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https://doi.org/10.1016/j.jclepro.2021.129239
Received 21 April 2021; Received in revised form 22 September 2021; Accepted 2 October 2021
Available online 4 October 2021

Abbreviations

AD	Anaerobic digestion
CHP	Combined heat and power
COD	Chemical oxygen demand
DEA	Diethanolamine
MCCA	Medium chain carboxylic acids
MDEA	Methyldiethanolamine
MEA	Monoethanolamine
NPV	Net present value
OFMSW	Organic fraction of municipal solid waste
OIW	Organic industrial waste
PHA	Polyhydroxyalkanoates
ROI	Return on investment
SS	Sewage sludge
VFA	Volatile fatty acids
VS	Volatile solids
WWTP	Wastewater treatment plant

while co-digestion with *Chlorella vulgaris* achieved a methane potential of 463 ml CH_4/gVS, a 26% increase compared to mono digestion of the sludge (Mahdy et al., 2015). Moreover, co-digestion of organic wastes can reduce the presence of inhibitors, provide buffering capacity and/or balance the C/N ratio, key parameters for an optimal AD.

Scandinavian countries present a good opportunity for co-management of sewage sludge with other organic wastes, since co-digestion for biogas production has been performed for several years (Sund Energy AS, 2010). However, in order to design a waste management strategy based on co-digestion of two residues, it is imperative to take into account the type and amount of available wastes. In Scandinavia, some of the available organic wastes are animal manure (Foged, 2012), food waste, sludge from fish farming (Torrissen et al., 2016) or agricultural residues (Kumar et al., 2020). In Denmark, for example, food waste and organic industrial waste (OIW, i.e. fat from the food processing industry) have been successfully co-digested with sewage sludge since 1997 (Billund Vand and Energi, 2021a), producing a 50% surplus of energy (Billund Vand and Energi, 2020). A similar example can be found in Sweden, where the same organic wastes are co-digested; however, the produced biogas is chemically upgraded via amine scrubbing, using the Cooab® technology (Gryaab, 2020), and sold as vehicle fuel. In Norway, sludge from fish farming has become a potential organic substrate for co-digestion with sewage sludge (Estevez et al., 2019), due to its increase in recent years. A next step in the co-digestion scenario is represented by the Billund Biorefinery, which was established in Denmark in 2017. The project focuses on the co-digestion of sewage sludge, food waste and OIW for the production of bioplastics, biogas and fertilizer in a circular economy perspective (Billund Biorefinery, 2020).

One of the drawbacks of sludge valorization is its high water content, which implies a larger equipment and higher transportation costs. Co-digestion could be a good strategy, as it can increase and balance the organic matter content. On the other hand, sewage sludge valorization could be coupled to already existing wastewater treatment plants (WWTP), and this stream is available throughout the year. Previous studies have investigated the economic feasibility of bioplastics production from sewage sludge, coupled to a WWTP (Crutchik et al., 2020) or incineration of sludge (Samolada and Zabaniotou, 2014). However, despite the high research interest on carboxylic acids production from sewage sludge (Atasoy et al., 2018), there is a lack of knowledge about the economic feasibility of the full-scale process.

The aim of the current study was to compare the techno-economic feasibility of five different strategies to valorize sewage sludge in Scandinavia by addition of different organic wastes. The first three scenarios corresponded to full-scale processes from Denmark, Sweden and Norway, respectively. Additionally, two original biorefinery scenarios, based on experimental data and literature, were proposed for the joint production of carboxylates, biogas and fertilizer. Two different downstream technologies were modelled for the recovery of carboxylates; an innovative method based on anion exchange chromatography and biological separation comprising polyhydroxyalkanoates (PHA) production. To summarize, the modelled scenarios were:

- Scenario A, based on Billund Vand (Denmark). Co-digestion of sewage sludge with food waste and OIW for combined heat and power (CHP) production.
- Scenario B, based on Gryaab (Sweden). Co-digestion of sewage sludge with food waste and OIW followed by biogas upgrading via amine scrubbing.
- Scenario C, based on IVAR (Norway). Co-digestion of sewage sludge with fish sludge followed by biogas upgrading via amine scrubbing.
- Scenario D, proposed original biorefinery scenarios. Co-fermentation of sewage sludge and food waste for the production of carboxylates, biogas and fertilizer. Recovery of carboxylates using two different technologies: biological separation coupled with PHA production (D1), and anion exchange (D2).

The different scenarios were compared in terms of direct capital cost, operating costs and total revenues, using scenario A as a reference case, with data from the actual plant. In addition, the different bottlenecks in the biorefinery scenarios were identified and studied. Sludge valorization was compared based on the price per ton of treated sludge and revenues per ton of sludge. Moreover, the effect of economy of scale and potential public subsidies were considered and discussed.

2. Materials and methods

2.1. Software for simulation

Simulations were performed using the software SuperPro Designer (v. 11, Intelligen Inc. (2020). This software was designed for the modelling and evaluation of different industrial processes, such as biotech, pharmaceutical, wastewater treatment, etc. and can be operated both, in batch and continuous mode. It consists of a series of unit operations and procedures, which can be used to simulate the different industrial process steps. Kinetic and/or stoichiometric equations are used to calculate the rates and/or conversion yields of the reactions, in each step. Moreover, the software's extensive database (including chemicals, costs, equipment and materials, which are regularly updated) allows the simulation of processes and the performance of economic analysis, thus compensating eventual lack of available data in literature. In our study, the design mode was selected in a way to estimate the size of the equipment based on the amount of material treated. In fact, the software contains built-in functions for the estimation of the equipment purchase cost, based on the size, material of construction, etc. In this way, the capital investment and annual operating costs are estimated using multipliers, based on the process specifics and associated costs (Intelligen Inc., 2021). In the SuperPro Designer database (Intelligen Inc., 2020), the facility-dependent costs are estimated based on the capital investment parameters (i.e. maintenance of the equipment, depreciation, insurance, etc.). Table S1 summarizes the basic equations used by the software to perform the economic evaluation.

2.2. Process description

For simplicity, all scenarios were assumed to work in continuous mode 330 days a year (7920 h), as valorization of sewage sludge could be coupled to a WWTP running daily (Table 1). The project lifetime, construction and start-up period were based on literature (Achinas et al., 2019). Sewage sludge was used as the reference stream (with two

Table 1
Assumptions for all the scenarios.

Items	Assumptions	Unit
Process	Continuous	
Annual operating time	7920	h
Sewage sludge	100	t/d
	300	t/d
Year of analysis	2020	
Project life time	25	years
Construction period	12	months
Start-up period	4	months

reference units of 100 t/d and 300 t/d, based on the real case scenarios), and the different organic wastes were added in the same ratio as in the real scenarios. All the calculations were based on the volatile solids (VS) content of the organic wastes, obtained from literature. Further information about the sludge characteristics and quality can be found in Fernando-Foncillas et al. (2021b).

A summary of all the scenarios is presented in Fig. 1, in the form of simplified block diagrams. In all scenarios, sewage sludge was generated during treatment of wastewater, which consisted of a mechanical primary treatment (to remove larger solids) and a biological treatment, in which carbon, nitrogen and phosphorus were removed during the activated sludge process (IVAR, 2021). Moreover, in scenarios A (Billund Vand and Energi, 2021b) and B (Videbris, 2021), an additional step for chemical precipitation of the remaining phosphorus was performed. In scenario D, the same steps for sewage treatment were assumed as in scenario A. Scenario A (Wittrup, 2012) consisted in the co-digestion of sewage sludge with food waste and OIW (i.e. a combination of wastewater and fat from the food processing industry), followed by biogas conversion into electricity and heat. The remaining fraction after anaerobic digestion, the digested sludge, was dewatered in a screw press.

Scenario B (Nunes et al., 2017) consisted of co-digested sewage sludge with food waste and OIW as well, but following the real case scenario of Sweden. Therefore, the methane yield and co-digestion ratio

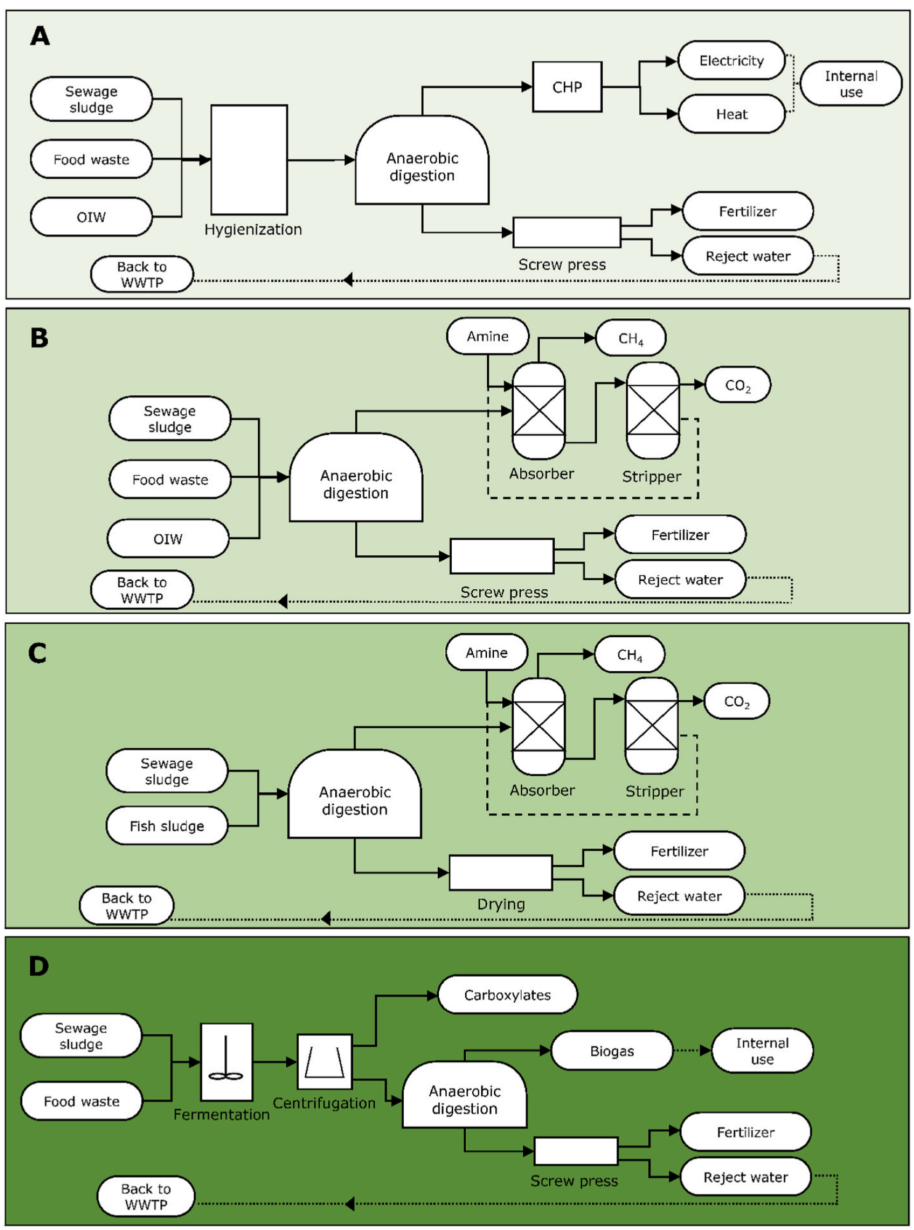

Fig. 1. Schematic overview of the simulated scenarios. A: Denmark; B: Sweden; C: Norway; D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography; CHP, Combined heat and power; OIW: Organic industrial waste.

differed from scenario A. On the other hand, in scenario C (Estevez et al., 2019) sewage sludge was co-digested with fish sludge, simulating the real case scenario of Norway. Both scenarios (B and C) generated biogas that was upgraded via amine scrubbing and produced fertilizer from the digestate fraction. However, in scenario B the digestate was dewatered with a screw press (similar to scenario A), while in scenario C it was dried and subsequently pelletized. In both scenarios, the dewatered digested sludge was sold as fertilizer.

Lastly, scenario D simulated co-fermentation of sewage sludge with food waste for carboxylates production. The fermentation broth was centrifuged, in order to separate the carboxylates (in the soluble fraction) from the solid fraction, which was sent to anaerobic digestion. In scenario D1, the stream with carboxylates was used for polyhydroxyalkanoates (PHA) production (biological separation) and the remaining fractions sent to the anaerobic digester (section 2.6). In scenario D2, the stream with carboxylates was pretreated by microfiltration and purified via anion exchange chromatography, followed by CO_2-expanded methanol desorption (Fernando-Foncillas et al., 2021a). The remaining fractions after microfiltration and anion exchange were sent to anaerobic digestion as well. Finally, the digestate was dewatered in a screw press to generate a fraction used as fertilizer. In all scenarios, the reject water generated after dewatering of the digested sewage sludge was modelled to be sent back to the original WWTP (not included in the simulation).

2.3. Anaerobic digestion

In order to simulate biogas production from co-digestion of the organic wastes, methane yields from each scenario were used, as presented in Table 2. Additionally, a ratio of 60% CH_4 and 40% CO_2 was assumed, in order to estimate the final biogas volume. In scenario D1, the remaining fraction with organic matter (VS) after centrifugation was sent to the anaerobic digester. Additionally, the cell debris after PHA production was also digested anaerobically. Assuming a microbial biomass composition of $C_5H_7O_2N$, the theoretical methane yield was calculated according to Buswell's equation (Eq. (1)):

$$C_nH_aO_bN_c + \left(n - \frac{a}{4} - \frac{b}{2}\right)H_2O \rightarrow \left(\frac{n}{2} + \frac{a}{8} - \frac{b}{4}\right)CH_4 + \left(\frac{n}{2} - \frac{a}{8} + \frac{b}{4}\right)CO_2 + cNH_3 \quad \text{(Eq. 1)}$$

The remaining organic matter after carboxylate centrifugation (Rest VS) was estimated based on the initial chemical oxygen demand (COD) in the reactor and the theoretical COD of the produced carboxylates. Anaerobic digestion of Rest VS was modelled using a methane yield from literature, with similar feed composition (Bolzonella et al., 2006). In scenario D2, biogas production was modelled from a) the remaining carboxylates after their purification, b) the remaining organic material after other processes such as centrifugation or microfiltration and c) part of the recirculated methanol after ion exchange chromatography. The theoretical methane yield from each compound was calculated according to Buswell's equation, and Rest VS was estimated according to literature (Cabbai et al., 2013).

Table 2
Methane yields for the different scenarios.

Scenario		CH_4 yield (ml/gVS)	Reference
A		356	Wittrup (2012)
B		267	Nunes et al. (2017)
C		450	Estevez et al. (2019)
D1	Rest VS	280	Bolzonella et al. (2006)
	Cell debris	570	
D2	Rest VS	365	Cabbai et al. (2013)
	Methanol	524	
	Acetate	373	
	Propionate	529	
	Isobutyrate	636	
	Butyrate	636	
	Isovalerate	713	
	Valerate	713	
	Hexanoate	771	
	Heptanoate	817	

2.4. Combined heat and power

In scenarios A and D, the produced biogas after anaerobic digestion was used for cogeneration of electricity and heat. This section was modelled according to literature (Achinas et al., 2019), using the pertinent process units. Firstly, biogas was compressed and burned in a gas turbine, in which electricity was generated. The efficiency of this unit operation was 38%, and the temperature of the exhausted gas was around 520 °C. Then, a steam generation unit was used to capture the heat from the exhaust stream, using an input of water and another for air. The efficiency of this unit procedure was 34%, requiring 0.8 kWh of heat per kg of generated steam. Therefore, 1 m^3 of biogas generated in total 2.1 kWh of electricity and 1.9 kWh of heat (Wittrup, 2012).

2.5. Biogas upgrading

The generated biogas in scenarios B and C was upgraded using an amine scrubbing process, according to literature (Vo et al., 2018). The process consisted of two steps: absorption of carbon dioxide in an amine liquid followed by stripping of the carbon dioxide. In the first place, biogas entered the absorption column, which was also fed with the amine liquid. This solvent absorbed the CO_2 and the purified biomethane exited the column. Secondly, the saturated solvent was fed to the stripper, in which the solution was heated by steam addition. The CO_2 was then released and exited the stripper, which was followed by a condenser, in order to separate it from the steam (Bauer et al., 2013).

Some of the most common amine solvents used during biogas upgrading are methyldiethanolamine (MDEA), diethnolamine (DEA) and monoethanolamine (MEA) (Bauer et al., 2013). In this study, MEA was selected due to its low cost, rapid reaction rate and high absorbing capacity (Vo et al., 2018). Additionally, a 30%wt MEA solution was chosen due to its high CO_2 removal efficiency (Ma'mun et al., 2005). Moreover, the columns were assumed to be filled with packing material to increase the contact time between solution and CO_2. Plastic pall rings (packing constant 170 and surface area 128 m^2/m^3) were selected for the absorber, and stainless steel pall rings (packing constant 170 and surface area 341 m^2/m^3) for the stripper, as described in literature (Vo et al., 2018).

2.6. Production and valorization of carboxylates

In scenarios D, sewage sludge and food waste were co-fermented at 37 °C with hydraulic retention time of 2 (Fernando-Foncillas and Varrone, 2021) and 4 days (Fernando-Foncillas et al., 2021a), in order to produce carboxylates, as described in literature. Due to the different composition of the fermentation effluents, two different strategies were selected for carboxylate valorization (Fig. 2) after centrifugation. In scenario D1, the fermentation stream consisted in a complex mixture of carboxylates (from acetate to heptanoate) at a low concentration (around 9 g/l) (Fernando-Foncillas et al., 2021a), which were used as carbon source for PHA production. In scenario D2, the carboxylate mixture consisted mainly in butyrate (25%) and hexanoate (63%), as described in literature (Fernando-Foncillas and Varrone, 2021). These carboxylates were upconcentrated via anion exchange chromatography.

In scenario D1, the centrifuged stream with carboxylates was converted into PHA by mixed culture microorganisms, assuming a yield of 0.45g PHA/g carboxylate (Valentino et al., 2018). The same PHA yield was assumed for all carboxylates, as suggested by literature. Additionally, the remaining VS were considered as cell debris that was digested anaerobically. A fixed cost of 2.2 €/kg PHA was assumed, which includes

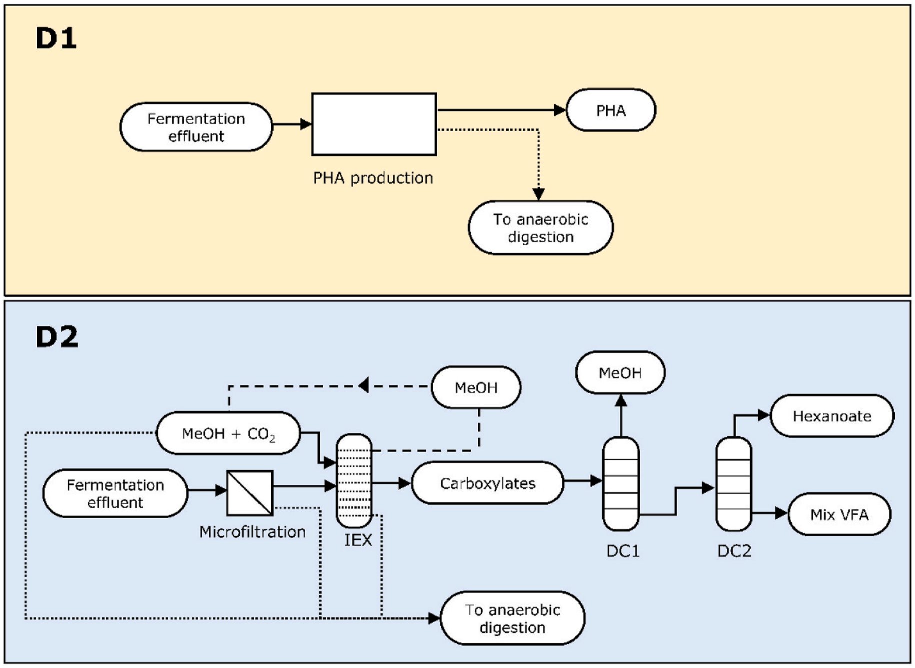

Fig. 2. Scenarios for recovery of carboxylates. D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography; IEX, Ion exchange; DC1, Distillation column 1; DC2, Distillation column 2; PHA: Polyhydroxyalkanoates.

the utilities, raw materials, equipment cost, etc. for PHA production (Fernández-Dacosta et al., 2015).

In scenario D2, instead, the fermentation effluent was centrifuged and pretreated by microfiltration, in order to remove impurities. The stream was then adsorbed using an anion exchange resin, followed by desorption with CO_2-expanded alcohol (Fernando-Foncillas et al., 2021a). Firstly, the stream was pumped to the chromatography column containing the strong anion exchange resin. Once the carboxylates were loaded on the column, methanol was pumped to the column to exchange the solvent, and then the carboxylates were desorbed using CO_2-expanded methanol (Cabrera-Rodríguez et al., 2018). The adsorption percentage and concentration factor varied depending on the specific carboxylate, increasing along with the number of carbons (Fernando-Foncillas et al., 2021a). This downstream technology was selected, due to promising results especially with medium-chain carboxylates (MCCA, i.e. hexanoate). After solvent exchange, the methanol could contain some carboxylate traces; therefore, part of the methanol was recirculated to the column, and mixed with new methanol for the new solvent exchange cycle. The remaining methanol was sent to anaerobic digestion (representing 10% of the initial methanol, and 1.2% of the total inlet to the anaerobic digester), together with other waste streams in the process (i.e. solids after centrifugation and microfiltration, and waste from adsorption), in order to produce biogas. After recovery of carboxylates via anion exchange, the stream (consisting in a mixture of carboxylic acids and methanol) was distilled in two steps to recover the different fractions with methanol, hexanoate and a remaining mixture of VFA. A simple column with one feed, a top and a bottom product was modelled, and no esterification was assumed due to lack of precise data. The first distillation column consisted in 20 stages with 80% stage efficiency and an operating temperature of 64.8 °C, while the second column consisted in 26 stages with 80% stage efficiency and an operating temperature of 166.7 °C. Additionally, default values from the SuperPro Designer database (Intelligen Inc., 2020) were used to model the distillation section, such as the ratio of the minimum to effective reflux ratio (R/Rmin, 1.25). A detailed mass balance with the composition of each stream is presented in Tables S3, S4 and S5.

2.7. Economic analysis

In all scenarios, the cost of waste streams was assumed zero. In addition, energy recovery opportunities were maximized by coupling operations requiring cooling (i.e. generating heat) with operations requiring heating. MEA solvent price, estimated at 1500€/t, was assumed a one-time expenditure since it is recycled in the system (Vo et al., 2018). An overview of the raw materials, utilities, revenues and other economic assumptions is presented in Table S2, based on literature and the default values in the software SuperPro Designer database (Intelligen Inc., 2020). Disposal costs of the dewatered digested sludge was assumed zero, since it was provided free of charge for the farmers in scenarios A, D1 and D2 (according to the real case of Billund Vand). In scenarios B and C, however, this fraction was sold as fertilizer (real case from Sweden and Norway), with the corresponding selling price according to its quality (Table S2).

Additionally, the purchase cost of the equipment was estimated by the software, based on the size and material. However, the price of the anaerobic digester, absorber and stripper was assumed from literature (Vo et al., 2018). The design mode was selected, in all scenarios, in order to size the equipment (and therefore the purchasing cost), according to the amount of material treated. Moreover, the other cost factors (such as instrumentation and installation of the equipment) were set as default by SuperPro Designer, in order to determine the capital and operating costs for each scenario. Data from a real operating full-scale plant was used in order to create an initial calibrated model (scenario A), based on operating costs and total revenues, provided by Billund Vand (DK). This calibration allowed to evaluate our assumptions and process design, making the models more realistic in terms of equipment purchase cost and operating expenses, thus improving the quality of the simulations for the original biorefinery (scenario D) presented in this study.

3. Results and discussion

3.1. Mass balances

In scenario A, electricity and heat were produced from biogas via CHP, generating 2416 MWh of electric power every year (Table 3).

Table 3
Mass flow in and out in the five scenarios.[a]

	Input		Unit	Output		Unit
Scenario A	*Material*			*Products*		
	Sewage sludge	33,000	t/y	Electricity	2416	MWh/y
	Food waste	1447	t/y	Steam	2670	t/y
	OIW	3524	t/y	Fertilizer	3016	t/y
	Total (4.5% VS)	37,971	t/y			
Scenario B	*Material*			*Products*		
	Sewage sludge	33,000	t/y	Biomethane	2.49E+05	m^3/y
	Food waste	396	t/y	CO_2	1.12E+05	m^3/y
	OIW	297	t/y	Fertilizer	1260	t/y
	Total (4.9% VS)	33,693	t/y			
Scenario C	*Material*			*Products*		
	Sewage sludge	33,000	t/y	Biomethane	3.13E+05	m^3/y
	Fish sludge	6758	t/y	CO_2	1.34E+05	m^3/y
	Total (3% VS)	39,758	t/y	Fertilizer	126	t/y
Scenario D1	*Material*			*Products*		
	Sewage sludge	33,000	t/y	PHA	164	t/y
	Food waste	10,451	t/y	Electricity	2113	MWh/y
	Total (5% VS)	43,451	t/y	Steam	2342	t/y
				Fertilizer	6764	t/y
Scenario D2	*Material*			*Products*		
	Sewage sludge	33,000	t/y	Hexanoate	238	t/y
	Food waste	21,410	t/y	Mix VFA	166	t/y
	Total (7% VS)	54,410	t/y	Electricity	5464	MWh/y
				Steam	5946	t/y
				Fertilizer	8439	t/y

A: Denmark; B: Sweden; C: Norway; D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography; OIW; Organic industrial waste; PHA: Polyhydroxyalkanoates; VFA: Volatile fatty acids; %VS, Volatile solids content.

[a] All the scenarios were modelled assuming a daily treatment of 100 tons of sewage sludge, which corresponds to 33,000 tons per year. The different organic wastes in each scenario were added in the same ratio presented in the literature.

Furthermore, 3016 tons of fertilizer were also produced per year. On the other hand, in scenarios B and C (with comparable mass input flow), a similar amount of biomethane and CO_2 were produced, using the same biogas upgrading system (Table 3). However, in scenario B, the generated amount of fertilizer was 10 times higher than in scenario C. This difference was mainly due to the water content: while the fertilizer in scenario B had a water content of 70%, the one in scenario C contained only 15% water (and therefore had a different selling price). On the other hand, a fertilizer fraction with 85% water was produced in scenario A; hence, the higher amount compared to scenarios B and C. Nevertheless, the fertilizer produced in scenario A was provided to the farmers free of charge, according to the real case scenario, implying no revenues from this fraction. In scenarios B and C, on the other hand, the produced fertilizer was sold.

Additionally, 33,000 t/y of sewage sludge and 10,451 t/y of food waste were treated in scenario D1 (Table 3). As a result, 164 tons of PHA and 6764 tons of fertilizer were produced every year. Similarly, in scenario D2, 33,000 tons of sewage sludge and 21,410 tons of food waste were treated per year, but recovering 238 t/y of hexanoate and a mixture of 166 t/y of VFA (Table 3). Moreover, the highest amount of fertilizer was produced in scenario D2, compared to the other scenarios. The final water content was also 85%, but the input material contained a higher amount of organic matter (VS). In scenarios D1 and D2, based on the Danish real-case scenario (A), the generated fertilizer was provided to the farmers nearby free of charge. In all scenarios, the main difference between total tons in the inlet and the outlet depended on the water (and the remaining water in the process went back to the WWTP, in order to be treated together with the incoming wastewater to the plant). A more detailed mass balance for scenarios D1 and D2 is provided in the supplementary material (Tables S3, S4 and S5).

The addition of the reject water can potentially cause an ammonium overloading in the WWTP, leading to a decrease in the nutrient removal efficiency and therefore affecting the quality of the discharged water from the WWTP. Moreover, its recycling could imply extra costs for additional nitrification/denitrification systems (Kim et al., 2020). An alternative for the reject water generated during sludge dewatering is to reuse it for anaerobic digestion, an approach successfully modelled for continuous operation, using an anaerobic granular sludge internal circulation reactor (Feldman et al., 2018). However, the high nitrogen content of the reject water should be taken into consideration, to avoid potential ammonium inhibition during anaerobic digestion (Kim et al., 2020). Simulation of the reject water recycling was not within the framework of the current study, and therefore not included in the models.

3.2. Economic analysis

3.2.1. Comparison scenarios

As presented in Table 4, none of the five scenarios were profitable under the current simulation (disregarding local subsidies and detaxation system), with negative return on investment (ROI) and net present value (NPV). Scenarios D1 and D2 required the highest capital investment and operating costs, due to more complex processes, higher investment in equipment and associated costs. The total capital investment consisted of equipment purchasing, installation, engineering and construction costs, among others. In addition, it was assumed that the entire plant was built from scratch, which can lead to higher capital investment and operating costs, compared to the integration into an existing WWTP for example.

Notably, previous studies have modelled biogas production followed by CHP (Achinas et al., 2019) or biogas upgrading via water (Mel et al., 2015) and amine scrubbing (Vo et al., 2018), resulting in lower capital investment and operating costs and a positive NPV. These differences can be in part explained by the different characteristics of waste streams used for anaerobic digestion, such as dairy slurry, grass silage or sugar beet pulp. These different waste streams contain for example varying amounts of organic matter, which can affect the equipment size or the yearly utilities and transportation costs. In fact, sewage sludge, which was used as main substrate in the current study, consists mainly of water

Table 4
Overall comparison of the five scenarios.

		Scenarios				
		A	B	C	D1	D2
Total capital investment	M€	4.37	3.76	4.44	9.97	16.42
Operating costs	M€/y	1.37	2.36	2.58	2.85	5.11
Net operating costs	M€/y	1.34	1.65	1.94	2.79	4.94
Total revenues	M€/y	0.06	0.39	0.49	0.80	0.99
ROI	%	−19.9	−24.3	−23.5	−9.0	−12.4
NPV (5% interest)	M€	−16.4	−25.6	−27.1	−22.6	−45.6
Unit processing cost	€/t SS	41.5	71.4	78.1	86.4	155.0
Net unit processing cost	€/t SS	40.7	49.9	58.8	84.6	149.7
Unit processing revenue	€/t SS	1.9	11.8	14.8	24.2	29.2

A: Denmark; B: Sweden; C: Norway; D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography; M€: million €; NPV: Net present value; SS: Sewage sludge; ROI: Return on investment.

(and contains recalcitrant material (Koch et al., 2016)). The equipment size is dependent of the total mass flow, and is therefore higher than for other substrates with less water content. Consequently, costs associated to the equipment purchase cost, such as installation or maintenance, are also higher, increasing the total investment and operating costs. Hence, the importance of co-digestion with other organic wastes in the case of highly diluted substrates, such as sewage sludge. Besides increasing the organic matter content, co-digestion can be helpful to balance some of the nutrients and reduce potential inhibitory compounds.

Previous studies evaluated the economic feasibility of treating sewage sludge in mono-digestion or in co-digestion with food waste, in a WWTP. Co-digestion increased the operating costs up to 44%, but it also proved to triple the revenues (Vinardell et al., 2021).

Another approach to decrease the costs and improve the economic feasibility of these plants could be further dewatering of the incoming sludge, or the integration into an existing WWTP. In addition to WWTP, the use of biogas production plants could also improve the economic feasibility of the co-management of sludge and other organic wastes.

It is worth noting that scenarios A, B and C should not be taken as a direct comparison between biogas conversion to electricity and heat, on one side, and biogas upgrading. Each model was developed according to the real-case scenarios, which use different ratios (and different VS content) to co-digest sewage sludge with organic wastes. Therefore, the total amount of material treated differed in each scenario, affecting the operating costs and revenues. In order to be able to compare the economic feasibility of all the processes, a throughput of 100 t/d sewage sludge was used as standard unit, while each scenario presented an alternative strategy for sludge valorization. Hence, the treatment cost or the revenues per ton of sludge can be used as economic indicators to standardize the comparison between the different scenarios, as presented above. Nevertheless, there is a lack of studies that evaluate the economic feasibility of large-scale plants for sludge (co-)management, hindering the comparison of different organic wastes and scenarios. Therefore, the models in the current paper were also standardized with real data provided by Billund Vand (scenario A), used to create an initial calibrated model that made the models more realistic (section 2.7).

In Table 4, the net operating cost was calculated by subtracting the savings, due to power and heat integration, to the operating costs. Therefore, the scenarios that can match operations requiring heating with operations producing it, could decrease the operating costs. For example, the condenser from biogas upgrading via amine scrubber (section 2.5) generated a heat stream, which can be used during anaerobic digestion of the wastes. In fact, scenarios B and C (with biogas upgrading) had the highest difference between total and net operating costs. This is in line with previous observations that reported how biofuel production from waste streams strongly depends on the amount of heat and power consumed during the process. For this reason, the use of energy flows generated inside the process is considered an important strategy, which allows reducing external energy consumptions (Varrone et al., 2013).

Scenarios D1 and D2, on the other hand, required the highest net unit processing cost (expressed in € per ton of SS), in agreement with their operating costs. Likewise, their unit processing revenues were also the highest. The operating costs consisted mainly of facility-dependent, labor-dependent, utilities and raw materials costs, as presented in Fig. 3.

There can be some differences among the Scandinavian countries, but in general, the costs are high and comparable. Therefore, all the scenarios were modelled assuming the same factors for labor, facility-dependent and laboratory costs, which are correlated to the quantity of equipment, size and purchase cost. The facility-dependent was the highest operating cost in most of the scenarios, compared to other operating costs such as laboratory and consumables costs. The fact that SuperPro Designer was initially developed to model processes for the costly chemical and biochemical industry can explain the high importance of this cost in the models.

Additionally, scenarios A and D, which used biogas for electricity and heat production, had almost no utilities expenses. This can be explained by the heat integration in these plants: the generated steam was used as heating agent in the plant, in addition to the power savings, due to electric power generation. On the other hand, utilities were the highest operating cost in scenarios B and C (biogas upgrading), due to the use of heating and cooling agents (despite the heat integration in the plant). Similar findings were reported by previous studies, comparing a CHP plant and a biogas upgrading facility, using biogas from co-digested sewage sludge and municipal solid waste. Despite both scenarios presenting similar annual operating costs, utilities expenses comprised

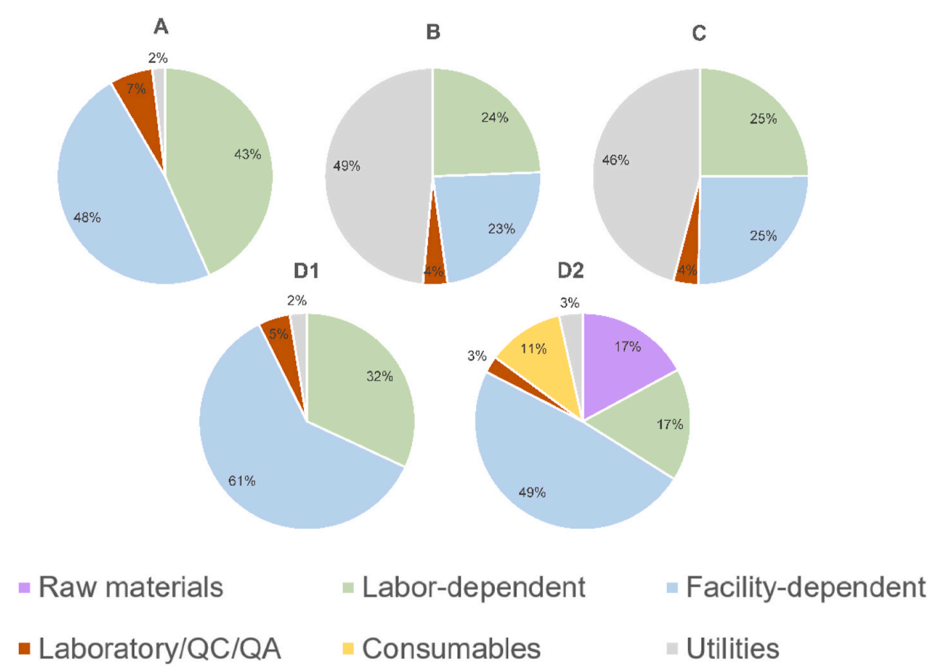

Fig. 3. Allocation of operating costs in each scenario. A: Denmark; B: Sweden; C: Norway; D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography.

0.34% of the operating costs in the CHP scenario, while this percentage increased up to 13% in a biogas upgrading plant (Morero et al., 2017).

Furthermore, raw materials and consumables were an important operating cost in scenario D2, resulting from the recovery of carboxylates via anion exchange chromatography (i.e. purchasing cost of methanol, CO_2 and bicarbonate, as well as membrane filters and anion exchange resin). A detailed analysis of the different sections in scenarios D, representing more complex biorefinery processes, is provided in the next sub-paragraph (3.2.2).

3.2.2. Biorefinery scenario

The biorefinery approaches presented in scenarios D1 and D2 focused on the production of carboxylates/PHA and biogas. As presented in section 2.6, the different metabolites distribution and fermentation titers were assumed from literature (Fernando-Foncillas et al., 2021a; Fernando-Foncillas and Varrone, 2021). Fig. 4 presents the itemized cost of both scenarios, divided in three sections: fermentation, downstream and anaerobic digestion. The first section contained the fermentation reactor and centrifugation, while anaerobic digestion consisted of the anaerobic digester and combined electricity and heat production. The downstream section comprised the valorization of carboxylates after fermentation: PHA production in scenario D1 and upgrading via anion exchange chromatography in scenario D2 (section 2.6).

As presented in Fig. 4, downstreaming of the carboxylates represented the highest costs in scenario D2, for both the capital investment (43% of the total) and the operating costs (57% of the total). As previously mentioned in section 3.2.1, this was caused by the use of costly raw materials and consumables for the anion exchange chromatography. Additionally, the operating costs included facility dependent costs, which were correlated to the type of equipment and purchase cost, higher in section D2. Interestingly, in scenario D1, the sections fermentation and anaerobic digestion were the most expensive, compared to the downstream (PHA production). The high operating costs of anaerobic digestion in scenario D1 can be explained by the higher amount of equipment in the model, which included biogas production and conversion to electricity and heat.

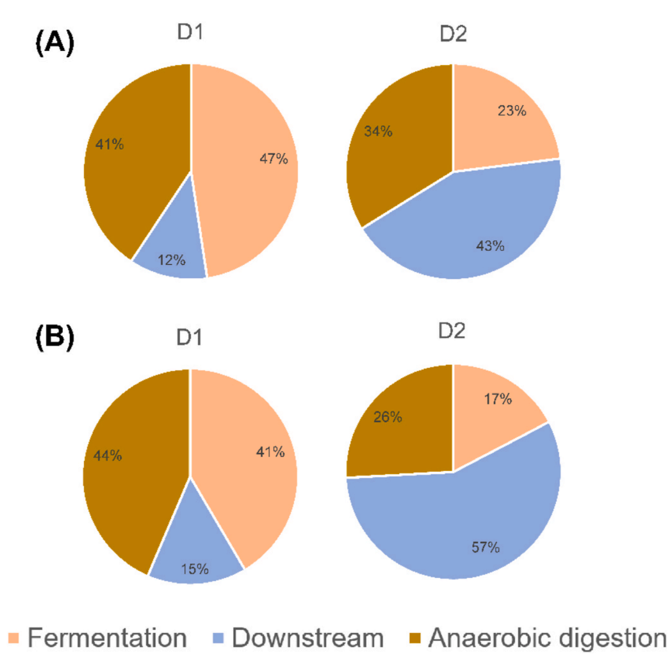

Fig. 4. Allocation of capital investment (A) and operating costs (B) in scenarios D for fermentation, downstream and anaerobic digestion sections. D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography.

In scenario D2, carboxylate valorization presented a two times higher capital investment and operating costs compared to D1 (Table 4), mainly due to the use of anion exchange chromatography. To reduce operating costs, part of the methanol, after solvent exchange, was recirculated in the column and sent to the anaerobic digester (section 2.6). In principle, the methanol obtained after desorption, as well as the sodium bicarbonate from column regeneration, could also be recirculated to a) decrease the raw materials operating costs and b) prevent costs of waste disposal.

It is worth noting that ion exchange is commonly applied in the pharmaceutical industry for purification of higher value products (Jenke, 2011). In this scenario, production of carboxylates was modelled due to their higher market value compared to biogas. Nevertheless, despite the higher revenues compared to scenarios A, B and C (Table 4), none of the biorefinery scenarios were profitable, as a result of the higher capital investment and operating costs. The main bottleneck in scenario D2 was the recovery of the bioproducts, caused by the complex mixture of carboxylates and the expensive technology. As modelled in scenario D1, production of PHA from low-titer carboxylates solutions is a promising technology for their valorization, in good agreement with literature (Moretto et al., 2020). The biodegradability and thermoplastic properties of PHA make them a potential substitute for fossil-based plastics (for example as packaging material) (Reddy et al., 2003), contributing to a more circular economy. However, the metabolite distribution in the carboxylate mixture or the feeding strategy affect the PHA composition and properties (Albuquerque et al., 2011) and therefore its market price, typically ranging between 4€/kg (Kourmentza et al., 2017) and 16€/kg depending on the composition (Platt, 2006). Therefore, the valorization strategy for carboxylates (i.e. converting them carboxylates into PHA, with additional costs, or directly purify and sell them on the market) will depend on several considerations, such as the fermentation titer, metabolite distribution, need for purity, cost of overall process, as well as the value, properties and final application of the recovered products.

In the two proposed biorefinery scenarios, the generated biogas was used for electricity and steam production, reducing the utilities expenses. However, biogas can also be upgraded to biomethane, as presented in scenarios B and C. For example, in scenario D2, the potential biogas upgrading (instead of heat and electricity production) would generate a CO_2 stream, which could be combined with recycled methanol for the desorption. This strategy would reduce the raw material costs, but in turn increase the utilities expenses. Therefore, each scenario should be carefully evaluated in order to select the most suitable biogas utilization technology.

3.2.3. The importance of public subsidies and economy of scale in the biobased economy

In a biobased economy, energy is classified as the least profitable product from biomass, and therefore public subsidies are often granted in order to promote the production of green energy. For example, the Danish Energy Agency offers a varying support scheme for the production of green electricity (i.e. from biogas, wind power, solar panels, etc.) (Energistyrelsen, 2020). As a consequence, the plant in Billund Vand is (in real life) profitable, thanks to the public subsidies for green electricity production (around 80% of the electricity revenues are based on subsidies, 0.11€/kWh) (R. H. Nielsen, personal communication, December 14, 2020), the gate fees for treatment of organic wastes and a larger throughput of treated waste. In fact, another important and well-known aspect is the economy of scale, as the total amounts of treated waste also influence the economic feasibility of the plant. As presented in Table 5, the current models treating 33,000 tons of sewage sludge per year did not constitute a feasible process, despite the support of subsidies. On the other hand, the same models treating three times the amount of sewage sludge (based on the actual amount in Billund Vand and keeping their corresponding co-digestion ratios), reached economic feasibility, but when combined with the Danish subsidies scheme

Table 5
Comparison of the operating costs and total revenues assuming the Danish subsidies and three times higher amount of treated waste for the five scenarios.

		Scenario				
		A	B	C	D1	D2
Treated SS: 33,000 t/y						
AOC	M€	1.34	1.65	1.94	2.79	4.94
Revenues no subsidies	M€	0.06	0.39	0.49	0.80	0.99
Revenues with subsidies	M€	0.88	0.93	1.62	1.86	2.78
Net UPC	€/t SS	40.7	49.9	58.8	84.6	149.7
Treated SS: 100,000 t/y						
AOC	M€	1.94	2.72	3.01	5.36	11.33
Revenues no subsidies	M€	0.20	1.21	1.42	2.45	3.02
Revenues with subsidies	M€	2.68	2.84	3.77	5.75	8.25
Net UPC	€/t SS	19.4	27.2	30.1	53.6	113.3

A: Denmark; B: Sweden; C: Norway; D1: Biorefinery scenario with PHA production; D2: Biorefinery scenario with ion exchange chromatography; AOC, Annual operating cost; M€: million €; SS, Sewage sludge; UPC, Unit processing cost.

(assuming the same scheme for all Scandinavian countries, for simplicity). Scenario D2 was the only exception, as the total revenues, including public subsidies, were lower than the annual operating cost, despite the higher amount of waste material treated. This can be explained by the high consumption of raw materials and consumables in the process, related to some operations such as microfiltration or anion exchange chromatography. Therefore, the revenues from high value products in scenario D2, such as hexanoate, were not enough to run a waste treatment process with this type of expensive downstream technology, more common in the pharmaceutical industry.

Currently, in order to promote a greener and circular economy, public subsidies (still) play a fundamental role to achieve economically feasible bioprocesses for the valorization of different waste streams. For that reason, each valorization process should be customized according to different variables, including (among other things) the type of wastes, their availability, the target product, but also the local policies and subsidies in place.

The influence of gate fees for food waste treatments was also found to have a noticeable economic impact in previous co-digestion studies, treating sewage sludge and food waste in a WWTP (Vinardell et al., 2021).

The use of other raw materials, such as lignocellulosics or brewing and food processing waste streams (Zhang et al., 2020), for the co-management with sewage sludge, could also be further investigated and evaluated.

Another important aspect is for instance the cost of materials selected for the construction of the large-scale reactors, which can be significantly decreased when dealing with co-fermentation of waste streams in non-sterile conditions (compared to the expensive materials needed for the chemical or pharmaceutical industry). Moreover, integration of new bioprocesses with already existing plants could also help reducing investment and operating costs.

The models from this paper represent some current waste (real-case) management scenarios from Scandinavian full-scale plants, in which anaerobic digestion and final sludge disposal on the fields is the most common sludge management methodology (Fernando-Foncillas et al., 2021b). Nevertheless, alternative scenarios could be studied to optimize sludge disposal and its cost, especially considering the more stringent regulations regarding disposal of biosolids on the fields. Such scenarios should also consider the potential savings for WWTP, when sludge is not disposed in landfills or incinerated, which would also contribute to the overall economic feasibility of the plants. These potential savings were not considered in the current paper, as the modelled (real-case) scenarios do not apply these final disposal alternatives.

An important challenge to develop a biobased economy is therefore to raise awareness regarding the importance of public subsidies and private investments. Even though (uncontrolled) subsidies might lead to situations of a distorted market (see for instance the situation of cheap biodiesel sold by some countries at costs below that of the raw materials to produce it), it is possible to implement smart strategies that would support and boost the development of new and more sustainable technologies (initially less viable). Topics such as step-wise decreasing support with increasing maturity of the technology and market, or the guarantee of a stable policy over a certain period of time, could help attracting also private investors and reduce the risk of distorted markets. Last but not least, different activities such as the creation of educational reports, engaging the media and policy-makers, and more science dissemination will be necessary.

4. Conclusions

In this study, the economic feasibility of five different scenarios for sewage sludge valorization were modelled. None of the scenarios was profitable in absence of subsidies, but both biorefinery scenarios presented higher revenues per ton of valorized SS. Facility and labor dependent costs represented more than 50% of the operating costs in most of the scenarios, while at least 40% corresponded to utilities in scenarios with biogas upgrading. As expected, the biorefinery scenario with anion exchange chromatography presented the highest capital investment and operating costs, due to the more expensive equipment and use of raw materials. On the other hand, the scenario with PHA production from a mixture of carboxylates (biological separation) was the most promising one, with the operating costs closest to the total revenues. This study confirmed that, despite the highly selective recovery of carboxylates via anion exchange chromatography, the use of this technology creates an economic bottleneck in a biorefinery scenario for waste valorization. Therefore, it is fundamental to carefully evaluate and identify the proper downstream processing strategy, based on the composition of the carboxylate mixture and market value of the target product. Moreover, the economic feasibility of the five scenarios was highly dependent on public subsidies, gate fees for organic wastes and the amount of treated waste, leading to profitable processes with larger plants.

CRediT authorship contribution statement

C. Fernando-Foncillas: Investigation, Methodology, Writing – original draft. **C. Varrone:** Supervision, Conceptualization, Methodology, Writing – review & editing, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This study was carried out as part of the COWI Fonden Project A092762-001 Wastewater and Organic Waste Treatment Facilities – Net Green Energy, Nutrients and Bioproducts Producers. The authors would also like to thank Billund Vand & Energi for data provision.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2021.129239.

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