Wastewater management towards a sustainable future Dairy production: a study case

R. Castro-Amoedo[a], Anna S. Wallerand [a], Hür Büntün [a], Ivan Kantor [a] and François Maréchal[a]

[a] *Industrial Process and Energy Systems Engineering (IPESE), École Polytechnique Fédérale de Lausanne, Switzerland, rafael.amoedo@epfl.ch*

Abstract:
The increasing demand for heat, electricity, fuels and chemicals is pushing natural resources towards a non-reversible situation. Current solutions have to be adapted, and alternative (desirably sustainable) sources have to be found. With growth assured due to an increasing global population, waste is able to provide a plethora of components in the near future. This work approaches waste management, by using wastewater from a dairy production. The current state-of-the-art which concerns industrial and municipal wastewater treatment focuses on single process design and optimization or, at most, on a set of competing unitary processes. In this study, a superstructure-based model for industrial wastewater integration and valorization is presented. It is formulated as a MILP problem with the objective of minimizing operational costs, while constrain investment costs. It comprises traditional waste conversion roots, but more importantly it proposes greener solutions in order to recover the intrinsic chemical and energetic potential of industrial waste.
Starting with a reference scenario of 23.4 M€ of operating costs and an exergy efficiency of 25 %, corresponding to a typical (optimized) wastewater treatment plant, with proper investment, exergy efficiency can go as high as 70 %, which as a direct link to environmental impact. The compromise solution that minimizes total cost, shows external electricity reduction by 70 %, providing an investment of 27 M€, recoverable in 12 years. Innovative solutions, like solid oxide co-electrolysis cells and methane synthesis from syngas are, with the present costs assumptions, non-profitable. Nevertheless, with incentives for bio-SNG production, as well as a reduction in electricity prices, an innovative and highly efficient solution is proposed, yielding an exergy efficiency of 86 %. The current work provides operating and investment costs of new technologies, as well as relevant technical data.

Keywords:
MILP, Energy integration, Dairy production, WWTP, Biogas, Exergy, Sustainability.

1. Introduction

The increase in demand for energy is undeniable, with global demand expected to be as high as 6 times the present values by the end of the century [1]; allied with continued fossil fuel extraction and consumption, concerns about climate change and environment in general are rising. In addition, industry, governments and societies are becoming more alert; new rules and regulations as well as new sources of energy are being discussed and introduced. A major concern is to be able to find sustainable (thus renewable) sources of heat, electricity, fuels and chemicals.

Waste treatment has seen in the past few years an incredible development concerning its valorization as a useful resource. From optimization in gasification processes, compounding new catalysts and reactors, to the production of bio-fuels, waste management is promoting efficiency in industrial processes and an opportunity to turn a liability into an asset. With waste generation expected to increase 4-fold by 2050 [2], proper waste management is not only an option, but a major necessity; being wastewater a waste sub-category, its numbers are also expected to increase. In addition, with increasing pressure on water resources worldwide, waste water treatment plants (WWTP) will have to address the need for potable water, while dealing with the associated sludges and environmental impact.

Scientific research on waste has been primarily focused on municipal solid waste, due to its greater potential to energy and heat recovery, when compared to wastewater. Indeed, labelled as waste-to-energy (WTE), it has attracted attention of several researchers, exploring thermal and biological approaches [2, 3]. Concerning wastewater, sludge handling and treatment is highly addressed, due to its similarity with biomass (or waste biomass). Chen et al. [4] report the production of biodiesel; Grobelak et al. [5] focus on small and medium scale plants, that besides upgrading sludge for energy recovery, promote also its use as a fertilizer.

When it concerns WWTP as a full unit, studies are predominantly based on Life Cycle Analysis (LCA) [6, 7], that are able to picture all the classical thermal and biological approaches. Gu et al. [8] discuss technologies for energy self-sufficiency in WWTPs, while Tang et al. [9] review new technologies such as electrochemical techniques for electricity recovery and greenhouse gases reduction.

Comparing different routes for wastewater treatment and valorization is best achieved with comprehensive, superstructure-type approaches. To this end, the present work has focused on:

a) development of an extensive superstructure-based MILP model, accounting not only for the heat requirements of a dairy production, but also to the full downstream chain of dairy wastewater, resulting in a Pareto front confronting operational and capital expenditure.

b) inclusion of different technologies for waste processing, including state-of-the-art correlations and values.

c) several scenarios accounting for uncertainty and incentives on electricity and gas prices.

Proposed solutions are not only relevant for developing countries, where a systematic wastewater management system is lacking [2], unsanitary landfill is still the main option and where, in the following years, the majority of produced waste (and wastewater) will come from, but also for developed countries, where current standard practices are re-thought. Therefore, this study configures itself as a key contribution for wastewater management valorization.

2. Process Description and general metrics

A Dairy plant is a set of complex chemical and physical processes that transform milk (labelled as raw milk) into a set of different products, like yoghurt, desserts, among others. Previous studies on dairy productions focused on CO_2-emissions reduction scenario [10] and further expanded by means of an heat pump superstructure and solar energy [11]. Both works consider 10 kg/s of raw milk as input. A reference case was designed based on the model from [10, 11] with the typical destination of wastewater coming from dairy productions, using the same input. A 2.5 folder multiplier is common in terms of wastewater production (thus 25 kg/s), according to a recent publication on dairy wastewater [12]; taking average values, and according to the same reference, total solids (TS) amount to 3.9 $g/kg, wastewater$ and biological oxygen demand (BOD) to 3.07 $g/kg, wastewater$.
A waste water treatment plant (WWTP) is a common destination for primary effluents of industrial plants, with dairy production being no exception. Under several regulations, which are country dependent, the wastewater stream must obey some characteristics in order to be treated jointly with municipal wastewater.
Economic metrics of all technologies discussed in the following sections and respective references are summarized in both Table 1 and 2; only if particularly relevant are they discussed in more detail. The same applies for performance data in Table 3.

2.1. Wastewater treatment plant

When entering a WWTP, wastewater undergoes a series of operations until it is suitable for discharge. The process is complex by nature and, despite new technologies, sizing and process design are semi-empirical. There is, from the process engineering perspective, huge room for improvement. A typical plant comprises a pre-treatment and at least two main treatments (primary and secondary), being one aerobic and other anaerobic [13]; biogas is produced and air and/or oxygen are supplied (when available area is a prime factor, O_2 is preferred to air, due to higher efficiencies, albeit the price). In a typical scenario biogas is burned and used in a boiler to provide heat for all the process. According to [13] this production might be enough to supply all the heating demand.
Digesters (aerobic or anaerobic) and dryers are the main demanding heat units. An average temperature must be kept in the digesters in order to have good yields of biogas and biomass degradation. 35 °C (mesophilic digestion) is the average used temperature [13]. Although the reactions are slightly exothermic, there might be a need to supply heat, specially in cold climates; radiation losses are also important and must be accounted. Furthermore, the feed must be warmed up to avoid breaks of productivity in the bioreactors. As the latent heat of vaporization needs to be supplied, drying might be the single operation in the complex chemical engineering world that requires the largest amount of energy. For subsequent applications the residual sludge humidity is crucial [13] with: a) Sludge to incineration – solids content between 30-35 % b) Landfill disposal – solids content of approximately 65 % c) Farming retail sale – solids content equal or higher than 90 %.

Although a viable option predominantly in developing countries, landfill will not be considered as a possible destination of treated sludge. Difficult operation control and maintenance as well as the associated environmental impact, make it *a priori* excluded. In general direct or indirect dryers are available. For the former, air is put into contact with the sludge, while for the latter it resembles an heat exchanger, where the hot fluid (steam, typically) circulates in a close loop. Regardless the type, the outcoming gas fluid must be treated to avoid particulate and odour contamination of the surroundings. This thermal drying (opposing open bed treatment) provides the single advantage of biological stabilisation for further processing. As the drying temperature is close to 100 °C, potential pathogens cannot survive, resulting in a safe disposal sludge. Thermal drying is one of the primary objectives of retrofitting in WWTP, with several cases of success [13]. From all the disposable options, farming retail sale (as a class-A biosolid) allows for some reimburse of process expenses, nevertheless attending the compromise between heat/biogas spent in the drying process.

Despite the inherently difficulty associated with operating costs, a study on wastewater treatment plants was considered [14], which reported an average value of 50 €/ton of wastewater. Nevertheless, as electricity accounts for a big fraction (around 50 %), half of value was neglected [14]; electricity input was set to 0.05 kWh/kg of wastewater.

Similar to municipal waste, a "gate fee" or "tipping fee" can be assigned to the treatment of wastewater. In EU gate fees are considerably higher when compared to developing countries, where typical values are around 15 €/Ton [3] of received waste. Due to this extreme dependency on geographical location, political decisions and environment awareness of a country, gate-fees were disregarded in both reference and superstructure scenario, which for the sake of comparison presents no obstacle.

The reference scenario (Fig. 1) shows the main processual connections as well as the considered utilities. Not represented, but obviously underlying the utility system, are steam pipes and a refrigeration cycle. The former is essential to recover the boiler's heat while the latter is needed to cool the below-ambient dairy process streams.

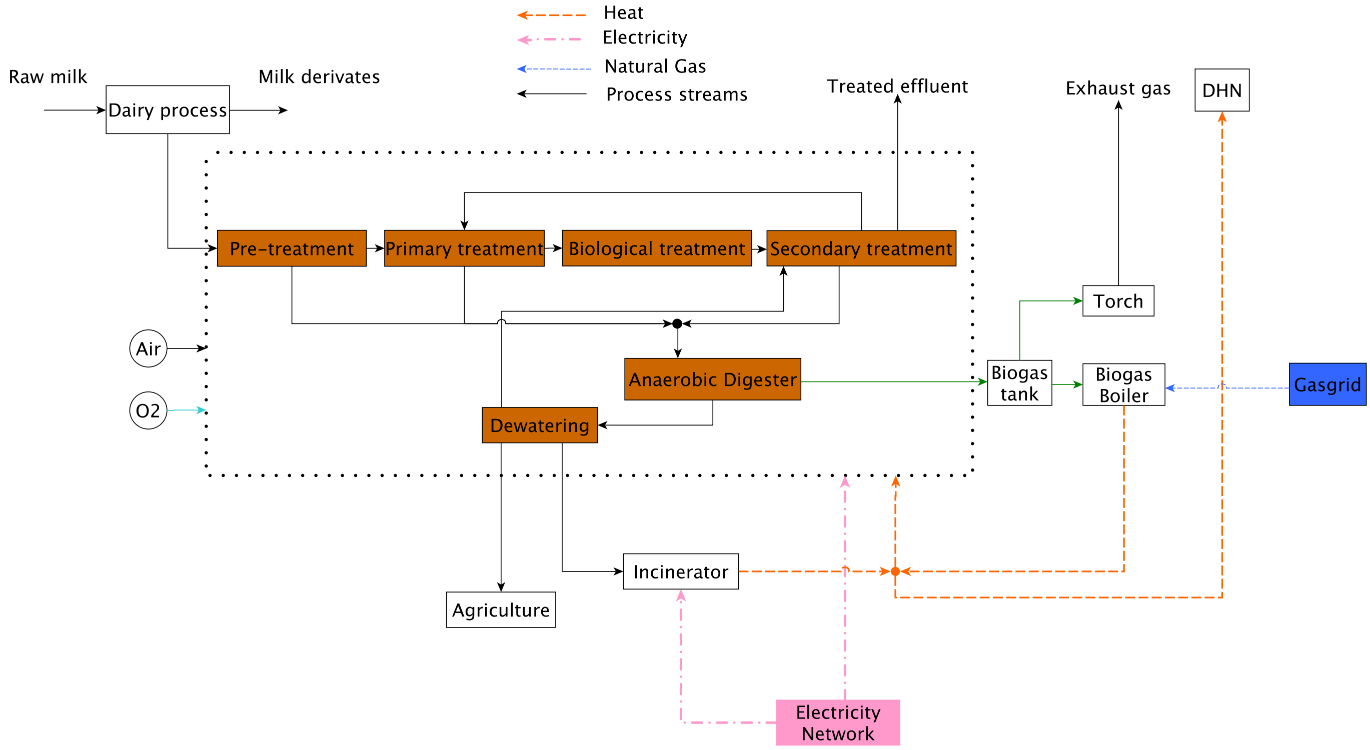


Figure 1: Reference scenario for dairy wastewater treatment

2.2. Exergy efficiency

Exergy is a key parameter to evaluate a system's performance, with higher exergy efficiencies linked to reduced environmental impacts [15]. Any material, heat and/or work stream, has an exergy content associated, supposing composition, pressure and temperature are known, and the reference state (typically the environement) is defined. However, exergy efficiency is far from being consensual [16], with many ways of defining it. The ratio between exergy output and exergy input, also known as second law efficiency, albeit one of the most used, does not take into consideration external destruction of exergy that is associated with heat losses, in particular those of flue gases. In this work, the utilizable exergy efficiency (or coefficient) as first described by Sorin et al. [17] was used; it takes into account not only destruction but also transferred exergy (1). E^{pu} and E^{c}, account for produced utilizable exergy and consumed exergy, respectively.

$$\eta_u = \frac{E^{pu}}{E^{c}} = \frac{E_{DHN} + E_{Dairy} + E_{Elec.}^{produced} + E_{Gas}^{produced}}{E_{WW} + E_{Elec.}^{demand} + E_{Gas}^{demand}} \tag{1}$$

Electricity and shaft work have their exergy value equal to their nominal value, as they can be totally converted into useful work. Heat streams have their exergy computed according to (2), where E_u^H, Q_u, T_{ref} and T_u, account for heat exergy of stream u, heat content of stream u, reference temperature and temperature of stream u, respectively.

$$E_u^H = Q_u \cdot (1 - \frac{T_{ref}}{T_u}) \qquad (2)$$

To compute fuel material streams exergy, extremely rigorous methods are available [18]. Considering an average relative atmospheric humidity of 70 %, the specific molar exergy of natural gas (assumed to be only CH_4) is 832.3 kJ/mol. Biogas was assumed to be 65 % methane and the remaining part (mainly CO_2) as exergetically neglectable. Biogas density (ρ_{CH_4}) was assumed to be 1.15 kg/m^3. Besides fuel material streams, only the dairy wastewater was exergetically considered. These streams are particularly difficult to account, as their composition is variable. Nevertheless, organic matter accounts for the largest share in exergy [19]. The same reference established a relationship between exergy and BOD: $E_{WW} = 13.6 \cdot BOD$ in kW.
The remaining streams, composed essentially of water and air can be neglected in exergetic terms [20]. T_{ref} was considered as 25 °C.
A general schematic representation of the exergy boundaries is depicted in Fig. 2

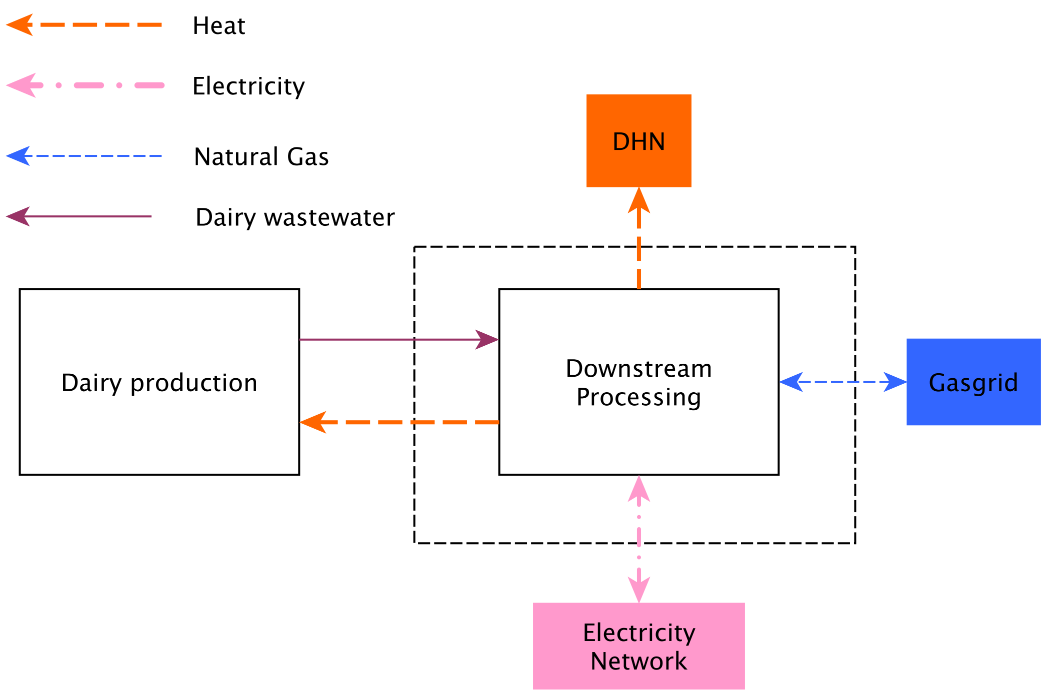


Figure 2: Exergy boundaries

2.3. District Heating Network

In a scenario where excess heat is available, supplying a district with heat is considered a win-win situation. Not only some profit can be made by supplying heat but also cooling water duty is avoided or minimized. An important question refers to the price the city is willing to pay to have heat supplied, as well as its temperature range. A standard that can be applied is impossible to find, as several external factor are conditioning the price; for example decisions are often dependent of local authorities approval and rates, which are geographicaly dependent. However, following former research involving demand for low temperature heat [21], the price the city a willing to pay lies on 1/4 of electricity price. For the electricity price assumption, district heating is rated at 20 €/MWh.

Temperature range suffers from a similar problem. More than geographical, seasonal effects play a role at least in the initial temperature of water. Nevertheless, the return temperature was set to 20 °C while outlet temperature was set to 80 °C. This last value is clearly more than enough for any urban application, including a buffer for possible losses during transport and use.

2.4. Incineration

Incineration is the main way of disposing waste in developed countries, competing only with landfill in developing ones. It is a mature technology, in which the combustion of organic materials is the underlying principle. It is most suitable for high calorific content waste, with subsequent steam production from the released heat.

The process comprises several advantages, as for example being relatively sterile, noiseless and odourless. Also, land requirements are minimal, at least when compared to landfills. The mass-burn incinerator is the most common among industrial areas, as it intakes waste "as received", avoiding any pre-treatment. Depending on the hazardous content of the waste, ash disposal range from landfill to incorporation in the construction industry. However, the latter case, is heavily dependent on national legislation, and thus not considered as a possible cost recovery [22]. The same reference points to the possibility of metal recovery, despite the relatively small scale production and consequently low potential.

Major concerns about incineration are related to the emission of particulate matter, heavy metals, sulphur dioxide, acid gases, carbon monoxide, dioxins and furans, among others. It is crucial to install flue gas cleaners, such as gas scrubbers and electrostatic precipitators. Largely due to potential environmental pollution, some social resistance to incineration is present [23].

Concerning costs, due to strict legislations on emissions and pollutants, the investment cost is typically high. Operational costs follow the same pattern, as auxiliary fuel (assuming natural gas) and electricity are needed in order to run the plant. For natural gas, $6.22\, m^3/ton\, of waste$ is used, while for electricity the requirements are at $123\, kWh/ton\, of waste$, according to a basic reference scenario reported in [24]. Furthermore there are operating costs related to maintenance, in particular scrubbers and filters, as well as labor costs that need to be taken into consideration. These costs are debated in the literature [25], and a value of 15 €/ton of waste was taken.

The heat obtained from the combustion logically depends on the burned waste average composition. Taking average values for a sludge with 30 % water content, $1.44\, kWh/kg\, of waste$ is obtained [13]. An efficiency of 75 % was used for the furnace/incinerator [26].

2.5. Gas Engine

The incorporated gas engine provides mechanical energy that a generator uses to produce power. The combustion gases are used for steam generation, although lower temperatures, around 550

°C [27], are achieved when compared to a boiler; this double production of heat and power constitutes a co-generation unit. Furthermore, gas engines are flexible, since they can intake gas with different compositions, including biogas [28] after a minimal cleaning stage to remove components that otherwise would damage the engine structure, like hydrogen sulphide H_2S.

2.6. Photovoltaic panels

The introduction of renewable energy in energy systems aims primarily for the reduction of operating costs, by reducing and eventually eliminating the electrical dependency from the grid. Photovoltaic panels are a mature technology, largely implemented not only at domestic but also at industrial level. Two main parameters are to be defined: the maximum area of implementation and the average irradiation.
Mainly due to large tank and reactor' areas, a WWTP does not have, in general, a reasonable area for photovoltaic installation. The potential of solar-thermal energy use in the dairy industry was discussed on a previous publication [29], in which an average specific useful roof area was computed, yielding $0.1053\, m^2/(t_{milk}.year^{-1})$.
Concerning solar irradiation values, and since multi-period optimization was not used, an annual average was computed [30]. This value accounts for seasonal changes and different daylight intensity, providing $160W/m^2$ (taken for a central European country). Regarding costs, photovoltaic modules are a mature technology, intensively produced and thus getting cheaper; literature on the topic [11,31] only consider a variable investment cost, function of the installed area, that also incorporates a installation cost factor. The average value of 360 €$/m_2$ was used.

2.7. Co-electrolysis SOEC and Electrolyser

Co-electrolysis in solid oxide electrolysis cells (SOEC) is attracting increasing attention, as an energy storage technology, mainly due to an increase in energy demand associated with a higher share of renewable; due to fluctuations and a mismatch of supply and demand of electricity, co-electrolysis offers a promising solution which allows to transform surplus electricity in chemical energy.
SOEC, although working at high temperatures (around 800 °C), have proved to promote energy savings of up to 20 % when compared to low temperature electrolysis [21]. One of the main advantages is that co-electrolysis can convert H_2O and CO_2, producing syngas and O_2, following the reaction $H_2O+CO_2 \rightarrow CO+H_2+O_2$. Syngas could be directly used in a solid oxide fuel cell (SOFC), forming (in combination with the SOEC) a reversible solid oxide cell (RSOC). Oxygen can be, in the present situation, directly used in the WWTP, promoting higher efficiencies in the aerobic treatment. Furthermore, syngas can be easily converted to methane, dimethyl ether (DME) or more complex hydrocarbons through Fischer-Tropsch conversion.
The major concern when working with SOEC is to ensure that their heating rate is slow enough to avoid cracking, that would result in considerable costs. Thus, it is necessary to guarantee a stable operation, meaning a constant supply of power, H_2O and CO_2. This necessarily means that, besides intaking electricity from renewable sources, the grid must be available to supply electricity when needed.
Concerning costs, syngas production using SOEC is mainly energy and feedstock intensive, rather than capital [21], which was expected due to the huge amounts of electricity needed to

run the technology.
An electrolyser is also considered as an available technology to produce H_2. Hydrogen is essential to promote the conversion of syngas to SNG in a methanation reactor, and due to safety concerns must be produced *in situ*. A commercial Polymer membrane electrolyte is chosen, as it is commercialy available and presents good performance indicators [32]. The same reference provides different sources for cost and size, allowing linearisation with a fixed investment cost of 24,800 € and a variable of 1,975 €/kW. As the working temperature, around 80 °C, is considerably lower than that of a SOEC, operating costs were neglected.

2.8. Methanation

Methanation of syngas in order to produce SNG to be injected in the natural gas grid is extremely complex. Several reactions take place and the final composition is determined not only by the initial composition, but also by temperature and pressure in the reactor. Additionally, the catalyst used and the H_2/CO ratio are crucial [33]. Values around 3:1 are advised, as H_2 is used in more than one reaction, including the water-gas shift.
Overall the process has been heavily studied, with several configurations of equipment in order to achieve the highest CH_4 composition. In this work, "Methanation" includes, besides the main catalytic reactor, the downstream process of cleaning and drying, responsible for removing undesirable components (using fixed bed reactors) and drying the gas for possible injection in the network.
Concerning the energetic point of view, the reactor should be kept at 400 °C, corresponding to a typical value found in the literature [33]. The reactions are extremely exothermic and thus the importance of temperature control, avoiding dangerous run-aways. A value of 220 kJ/mol of CO reacted was assumed [34]. The incoming syngas compostion is defined by the SOEC and assumed to be 1:1. To ensure the desired ratio of 3:1, for every 1 kg/h of equimolar syngas, $\frac{2}{15}$ kg/h of extra hydrogen must be injected.
For costs, there seems to be a general overestimation of methanation units [35]. Typical investment costs comprise not only the reactor but also an electrolyser for in situ production of H_2. In the present situation, as the electrolyser is considered an independent unit, the investment cost is reduced. To account for this situation, and in line with previous investment studies, where electrolysis is the main responsible for the price [35, 36] – close to 75% - the price of the methanation unit was chosen by taking only 25% of the price with most industrial support [37]. Fixed investment cost was set 1,670,000 € and the variable part to 166 €/kW of SNG produced. As the process is catalytic, operational expenses must be included to account for catalyst recovery and/or replacement. The cost was set to 0.036 €/kWh of SNG produced [31], and it also includes gas cleaning/drying after the reactor.

2.9. Pressure swing adsorption

Pressure swing adsorption (PSA) is a widely spread gas separation technique, commonly used in waste-to-methanol projects [38]. It consists of a material (molecular sieves like zeolites, activated carbon or even carbon molecular sieves) that promotes different adsorption affinities among the treated gas stream. By changing the pressure, certain gases are adsorbed or desorbed, allowing their separation. Biogas from anaerobic digestors were studied in detail [39] with a four-bed, seven-step PSA process yielding high CH_4 purity, with an overall efficiency of 97 %.

2.10. Steam Network

A steam network has a double application in a industrial setting. Besides producing steam, which is the primary form of industrial heat, it allows also for the production of electricity (co-generation), by expansion of steam in a series of turbines. It is represented in the flowsheet as RC, which stands for Rankine cycle, being the underlying thermodynamic cycle. The main equipment are turbines, pumps and heat exchangers, where the first clearly dominates over any of the other piece of equipment. The steam network was based on the MILP model presented in [40], which relies on a superstructure approach to size the individual components.

2.11. Heat Pumps

The use of heat pumps for energetically improve industrial processes is well described, inclusively applied to a dairy production [11]. The underlying principle is the use of electricity to promote heat from a lower to a higher grade. It is represented as HP in the flowsheet. Multi-stage heat pumping was modeled based on a MILP superstructure approach by [41].

2.12. Heat Exchangers Network

Heat exchangers are crucial pieces of equipment allowing heat recovery in any plant, thus being part of the skeleton of any industrial process. The area is the sizing parameter of an heat exchanger, which was estimated using the vertical intervals approach first developed by [42]. For each interval, the area is estimated based on the overall heat transfer coefficient, the logarithm mean temperature difference, and logically the amount of heat in the interval. All the areas are added up, and divided by the minimum number of heat exchangers, determined by following the method suggested by [43]. The network total investment will be given by (3), in which A is the area of one heat exchanger, determined under the assumption that all the heat exchangers are equal. They were assumed to be of floating head type.

$$HEX_{cost} = (c_{HEX}^{inv1} \cdot y_u + c_{HEX}^{inv2} \cdot Area) \cdot N_{HEX}^{min} \tag{3}$$

2.13. Investment costs

When taking decisions, economic indicators take special relevance as one of the key criteria. Table 1 summarizes all the values discussed and their respective references. All the values were updated to 2017 using the CEPCI index (567.5), EUR/USD and EUR/CHF were set to 0.9.

2.14. Utility costs and technical data

Utilities' prices are typically difficult to estimate; they encompass an all supply chain of production and transformations thus their final price depends on geographical location, seasonal events, and even political decisions. Intra-daily fluctuations are also very common, making it difficult to have an average value. Table 2 summarizes the values assuming, when applicable, a 75 % selling price compared to the market value. Table 3 resumes operating and technical assumptions.

Table 1: Fixed and variable part of investment and operating costs for different technologies; s.p. - sizing parameter

Technology	C_u^{inv1}, €	C_u^{inv2}, €/s.p.	C_u^{op2}, €/s.p.	Reference
WWTP	-	-	25 €/Ton	[14]
Incinerator	-	-	15 €/Ton	[25]
PV panels	-	360 €/m^2	-	[11,31]
GasEngine	1,554,000 €	3,580 €/kW	-	[44]
Electrolyser	24,800 €	1,975 €/kW	-	[32]
SOEC	118,000 €	9,400 €/kW	0,001 €/kWh	[21]
Methanation and gas cleaning	1,670,000 €	166 €/kW	0.036 €/kWh	[31,37]
Separation (PSA)	882,000 €	1,750 €/$m^3.h^{-1}$	-	[45]
Heat Exchanger	21,860 €	150 €/m^2	-	[26]
Turbines	816,000 €	164 €/kW	-	[26]
Compressors	107,200 €	50.22 €/kW	-	[26]
Pumps	6,380 €	120 €/kW	-	[26]

Table 2: Utility costs assumptions

Parameter	Unit	Value	Reference
Cost of buying electricity	€/MWh	80	[46]
Cost of selling electricity	€/MWh	(-60)	-
Cost of buying natural gas	€/MWh	40	[46]
Cost of selling natural gas	€/MWh	(-30)	-
Cooling water cost	€/m^3	0.06	[47]
Deionized water cost	€/m^3	1.15	[47]
O_2 supply cost	€/m^3	0.07	[35]
Air supply cost	€/m^3	0.0014	[26]

3. Methods

A MILP formulation based on [51] is applied for the optimal utility selection. The overall system contains units ($u \in U$), consisting of process units ($up \in UP$) and utility units ($ut \in UT$). The main difference is that process units are added with a fixed size to the problem, while utility units are sized accordingly. It thus implies both binary (y_u) and continuous (f_u) variables associated with each utility unit. Since the problem is formulated for a single-period, there is no time dependency in any of the variables.

The main objective is minimizing operational costs (OP_{Total}) according to (4), in which t^{op} is

Table 3: Technical and operating assumptions

Technology	Parameter	Value	Unit	Reference
WWTP	Electricity Input	50	kWh/ton WW	[14]
Incinerator	Electricity Input	123	kWh/ton Waste	[24]
	Gas input	6.22	m^3/ton Waste	[24]
	Heat obtained	1440	kWh/ton Waste	[13]
	Efficiency	75	%	[26]
Gas Boiler	Maximum working Temperature	1200	°C	[48]
	Efficiency	80	%	[26]
Gas Engine	Maximum working Temperature	550	°C	[27]
	Electrical Efficiency	31	%	[27]
	Heat Efficiency	55	%	[27]
PV panels	Area	0.1053	$m^2/(ton_{milk}.year^{-1})$	[29]
	GHI	160	W/m^2	[30]
SOEC	Operating Temperature	800	°C	[21]
	Energy efficiency	85	%	[37]
Electrolyser	Operating Temperature	80	°C	[32]
	Energy efficiency	70	%	[35]
PSA	Transformation efficiency	97	%	[39]
	Electricity Input	0.25	kWh/m^3 input gas	[45]
Steam Network	Steam production pressure	150	bar	-
	Steam superheating	200	°C	-
	Steam utilization level	20/2	bar	-
	Condensation level	1.5	bar	-
	Efficiency, backpresure	80	%	[49]
	Efficiency, condensation	70	%	[49]
	Efficiency (isentropic), pump	95	%	[49]
Heat pump structure	Evaporator Temperature	-8	°C	-
	Condenser Temperature	40 / 33 / 25	°C	-
	Efficiency (isentropic), compressor	76	%	[50]
Methanation	Operating Temperature	400	°C	[33]
	Heat of reaction	220	kJ/mol CO	[34]
	$H2 : CO$ ratio	3 : 1	-	[33]
	Chemical efficiency	78	%	[35]
General				
Natural gas	LHV	47100	kJ/mol	[26]
	Specific Exergy	832.3	kJ/mol	[18]
Biogas	CH_4 composition	65	%	[13]
	density	1.15	kg/m3	[13]
DHN	Supply temperature	80	°C	-
	Return temperature	20	°C	-
Dairy	Heating requirements	1987	kW	-
	Cooling requirements	1552	kW	-

the operating time, settled to 8,760 hours.

$$min \sum_{u}^{U} \left(c_u^{op1} \cdot y_u + c_u^{op2} \cdot f_u\right) \cdot t^{op} \Leftrightarrow min\ OP_{Total} \tag{4}$$

Grid utilities such as electricity, natural gas, air and water, are defined as utility units and thus are not explicitly represented in the operational costs. The particularity of all of them is having a fixed operating cost (c_u^{op1}) of 0.
Several constraints are added to the problem. Heat transfer constraints are ensured by the heat cascade (5),(6), in which heat is only allowed to flow from high temperature level streams to lower level ones; the energy balance must also be closed (7).

$$\sum_{u}^{U} f_u \cdot \dot{Q}_{u,k} + \dot{R}_{k+1} - \dot{R}_k = 0 \qquad \forall k \in K \tag{5}$$

$$\dot{R}_k \geq 0 \qquad \forall k \in K \tag{6}$$

$$\dot{R}_1 = \dot{R}_{k+1} = 0 \tag{7}$$

There are also constraints associated with the choice of utilities, linked to their size and existence (8), as well as constraints associated with process units (9).

$$f_u^{min} \cdot y_u \leq f_u \leq f_u^{max} \cdot y_u \qquad \forall u \in U \tag{8}$$

$$y_u, f_u^{min}, f_u^{max} = 1 \qquad \forall up \in UP \tag{9}$$

Furthermore, in order to provide trade-offs between capital and operational expenditures, an $\varepsilon - constraint$ is added to the MILP following (10).

$$\varepsilon \geq \sum_{u}^{U} \left(c_u^{inv1} \cdot y_u + c_u^{inv2} \cdot f_u\right) \cdot \tau \Leftrightarrow \varepsilon \geq INV_{Total},\ \varepsilon \geq 0 \tag{10}$$

in which τ is the annualization factor calculated according to (11), in which i is the interest rate assumed as 0.08 and n is the equipment lifetime taken as 20 years.

$$\tau = \frac{i * (1+i)^n}{(1+i)^n - 1} \tag{11}$$

Table 4 summarizes the parameters used in the MILP formulation and the corresponding description.

Table 4: MILP parameters and description; s.p. - sizing parameter

Parameter	Description	Unit
c_u^{op1}	Fixed operating cost of unit u	€/h
c_u^{op2}	Variable operating cost of unit u	€/s.p.
c_u^{inv1}	Fixed investment cost of unit u	€
c_u^{inv2}	Variable investment cost of unit u	€/s.p.
y_u	Integer variable for use unit u	-
f_u	Sizing factor of unit u	-
f_u^{min}	Minimum sizing factor of unit u	-
f_u^{max}	Maximum sizing factor of unit u	-
t^{op}	Operating time	h
$\dot{Q}_{u,k}$	Heat to or from unit u	kW
$\dot{R}_k$	Cascaded heat in temperature interval k	kW

4. Results and Discussion

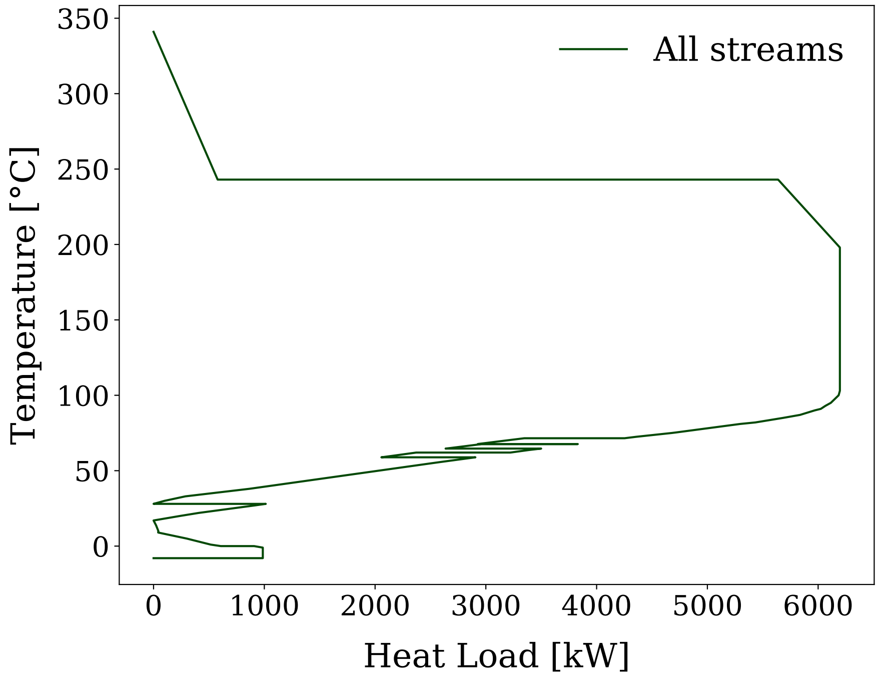
A set of scenarios was generated in order to understand different energy efficiency options and technology choices for the wastewater treatment of a dairy plant. Starting from a reference case, a superstructure scenario was derived, expanding its boarders to incorporate different technology options. Several restrictions were placed on the capital expenditure, in order to mimic typical process/economic restrictions in a real plant.

4.1. Reference scenario

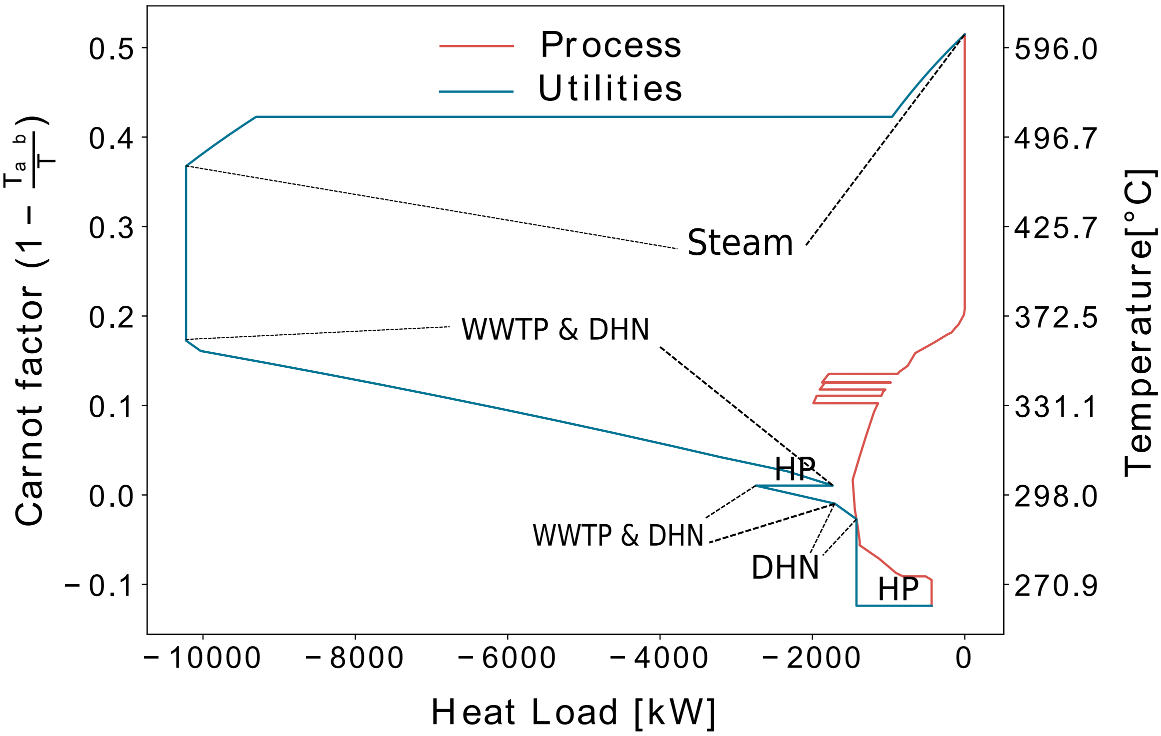
The reference scenario comprises several considerations that impact the values obtained:

- No investment cost was considered, as all the equipment was previously in place;
- The system is cost optimized, meaning that the value obtained is the minimum possible with the considered set of flows;
- A DHN is already in place and uses excess heat from the industrial setting;
- No gate-fee was considered for the wastewater input;

Figure 3 shows the grand composite and the Carnot composite curves of the reference scenario, where the use of steam, coming from the biogas boiler, is the main source of process heat for the dairy plant and the downstream wastewater conversion. There is large exergy destruction, highlighted by the large pocket in the Carnot factor vs entahlpy diagram, in particular between the steam temperature and the remaining low temperature processes; steam is thus being used for providing low-temperature heat. Nevertheless, this is the normal situation in many industrial units, where a boiler is the main (and typically only) provider of heat.



(a)



(b)

Figure 3: Grand composite curve (a) and Carnot composite curves (b) for reference scenario

The main reference results, concerning mass, energy, exergy and economics are shown in Table 5. The main operating costs concern the regular work of a WWTP, as expected. Indeed, large sums are needed for costs associated with labor, maintenance, transport and logistics. Albeit economies of scale can be expected for a large plant, they were not considered. Additionally, the cost of electricity is also heavy on the overall bill, and is a major target for energy integration. On the revenue side, DHN is the major source of income, with an annual total of more than 1 M€, followed at great distance by a contribution of sludge used in agriculture. Despite this last revenue, incineration is the preferred path for sludge disposal, as it needs substantially less drying energy, while providing a source of heat for steam generation. Overall the industrial complex needs close to 23.5 M€ (of operational costs) to run annually, as no gate-fee or any kind of environmental tax was considered. It corresponds to approximately 110 €/ton of wastewater received.

Regarding exergy calculations, the procedure adopted followed section 2.2., in particular (1) and (2). For wastewater, and for the amount of BOD considered (3.07 g/kg of wastewater), an exergy input of 1,044 kW is present. As already stated in section 2.1., wastewater organic composition is highly unpredictable, with a typical range of values of one order of magnitude apart. In addition, in a real WWTP the income wastewater has different origins and compositions (typically it is a mixture of industrial and domestic), which makes values less accurate. Concerning temperatures, heating requirements in the dairy are between 60 and 100 °C, and 5°C steps were used for temperature discretization. Cooling requirements, ranged from 0 to 25 °C, and a step of 5 °C was also taken. Concerning DHN, the value of 80 °C was assumed. Globally, from an energy and exergy perspective, the system presents a exergy efficiency of approximately 25 %. This value drops to 8 % if not accounting for DHN. It seems clear that the system is exergetically poor. As a remark, part of the energy that enters the downstream process is transformed into non-useful forms; among them, flue gases and radiative heat losses are the most significant. However, as the purpose is to compute exergy efficiencies, those flows were on purpose disregarded and thus not included in Table 5.

Table 5: Reference scenario main results. *: internal exergy flow

Parameter	Nominal flow	Units	Operating costs, $kEUR/year$	Exergy, kW
Material flows				
Wastewater	90	ton/h	20,510	1,044
Treated effluent	85.9	ton/h		-
Sludge to agriculture	0.26	ton/h	-93	-
Sludge to incineration	2.89	ton/h	380	-
Biogas production	1,077	m^3/h	-	11,130*
Air supply	3,606	m^3/h	32	-
O_2 supply	0	-	-	-
Energy flows				
Electricity (Net)	5,335	kW	3,740	5,335
NG/SNG (Net)	-	-	-	
DHN	7,000	kW	- 1,165	1,091
Dairy needs	3,539	kW	-	497
Total Cost	-	-	23,400	-
Exergy efficiency	-	-	-	24.9 %

4.2. Superstructure scenario

The superstructure flowsheet (Fig. 4) considers the set of technologies previously described.

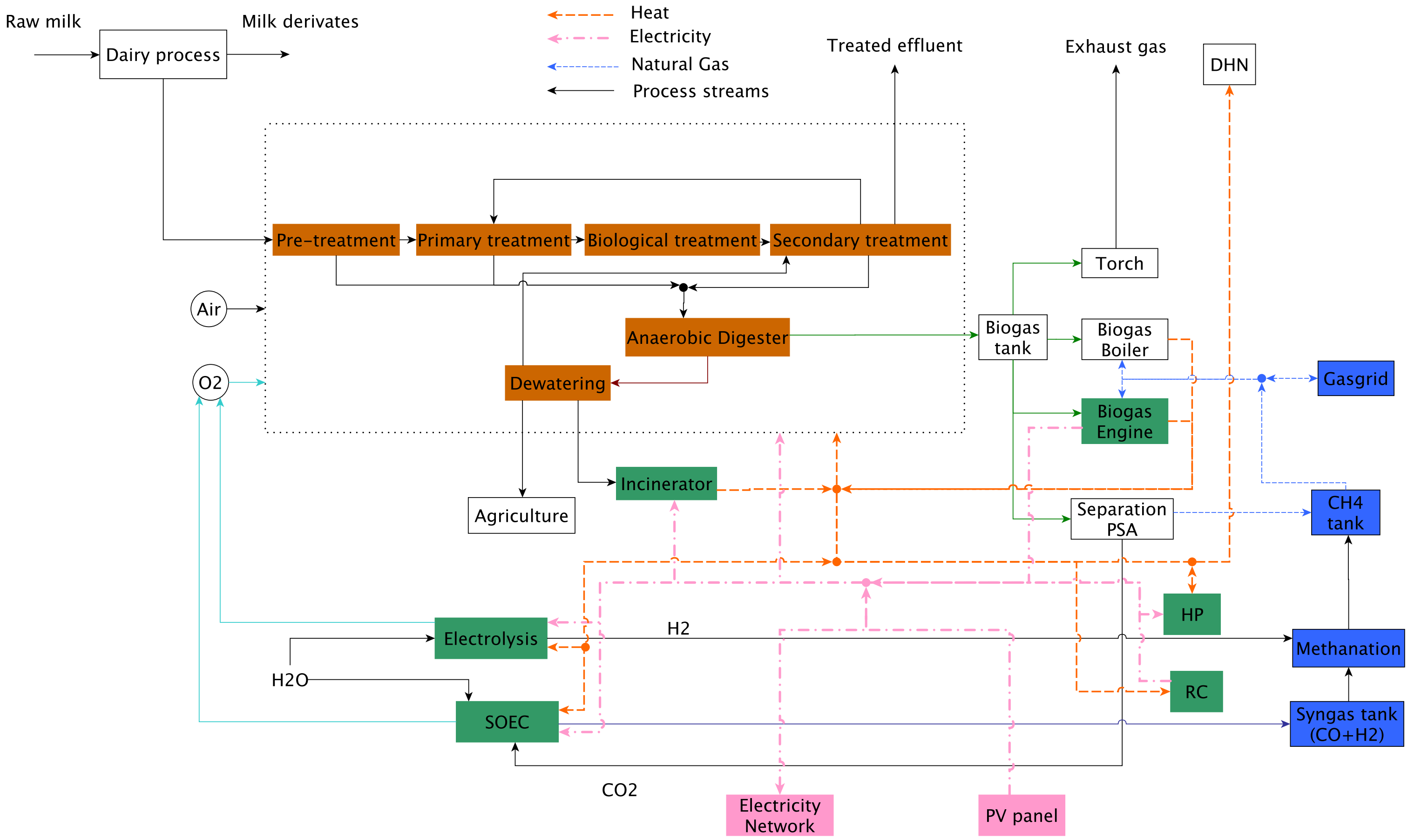


Figure 4: Superstructure scenario for dairy wastewater treatment

The scenario comprises several assumptions that have an impact in the overall results:

- The heat exchanger network was considered to be new, meaning that no retrofitting and no cost of selling equipment was considered;
- A steam network was considered as basic equipment, replacing the steam pipes; thus there is always a minimum capital expenditure to account for.
- The heat pump superstructure was retrofitted, being the 1-stage heat pump compressor re-used. Furthermore, a project decision of limiting the number of compressor stages to 2 was made.
- Temperature levels, for the condenser and evaporator in both steam network and heat pump structure, were chosen by empirical fitting with the integrated composite curves, and were not part of the optimization.

The investment that needs to be made is considerable. The absence of a proper steam network that is able to co-generate electricity will constitute one of the major expenditures. In order to

analyse the trade-off between capital and operating expenditures, ϵ-constrained optimization was carried out. In Figure 5, the resulting solutions are depicted. It is immediate the trade-off between capex and opex, in which the former increases while the latter decreases. In addition, pay-back times are consistently above 10 years, which might hinder several investment decisions. Nevertheless, exergy efficiency is also continually increasing, which could mean a potential profit, if CO_2 taxes were to be considered.
As electricity is one of the main operational expenditures in the overall process, the MILP will try to minimize its consumption; to do so it maximizes the investment in the steam network, which is a high-efficiency co-generation unit.

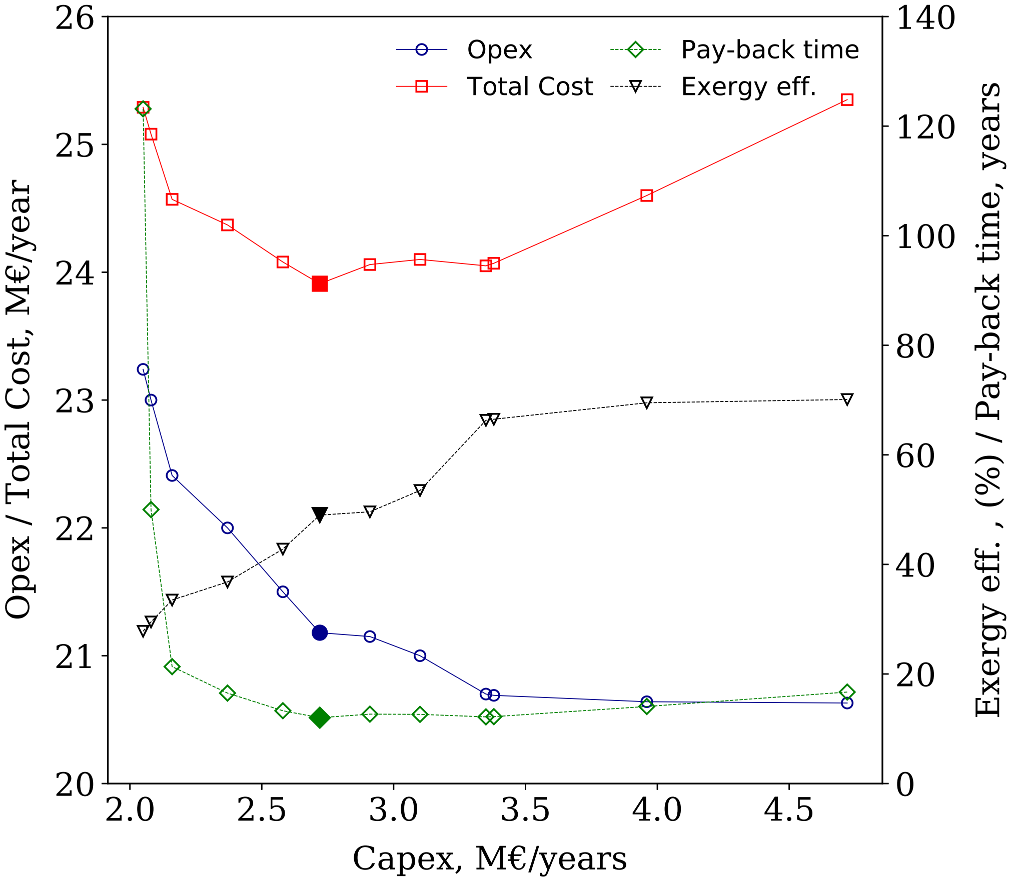


Figure 5: Pareto curve. Lines were added for visual tracking and do not represent a trend or correlation. Larger points, represent the combination with the lowest total cost.

The choice of technologies and respective yearly investment costs are depicted in Fig. 6. The trade-offs above discussed are confirmed, with a considerable investment in the steam network and gas engine, as capital expenditure is allowed to be higher. The driving force is, as already stated, the reduction of electricity consumption from the grid, which in turn increases the exergy efficiency of the overall process. With decreasing operating costs and increasing exergy efficiency, a reduction in environmental impact can be associated, with less non-renewable

energy sources used from the grid.
The reduction of investment to a minimum yields electricity and exergy efficiency close to the values obtained for the reference scenario, albeit slightly higher, due to the presence of a steam network that produces electricity. In reality the sole introduction of a proper steam network can improve the overall energy and exergy efficiency of the plant.

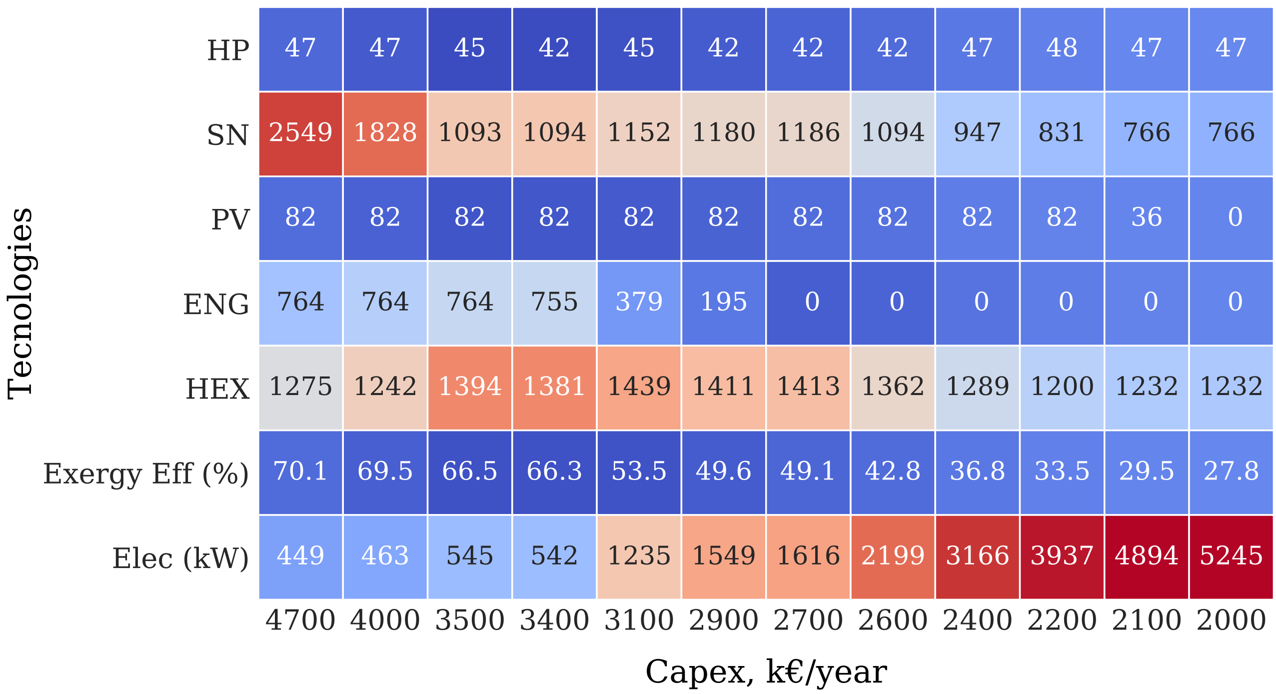
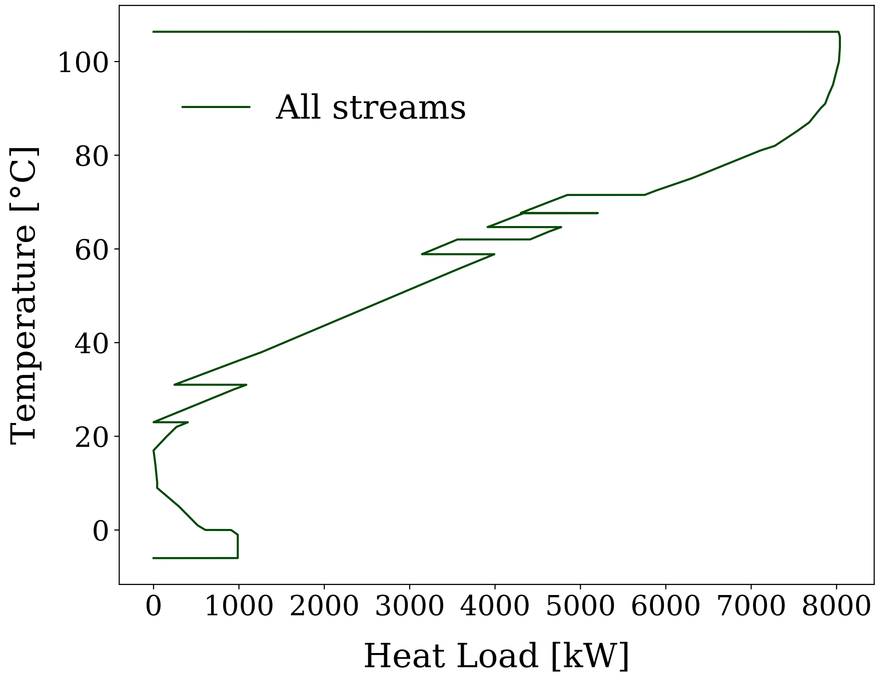


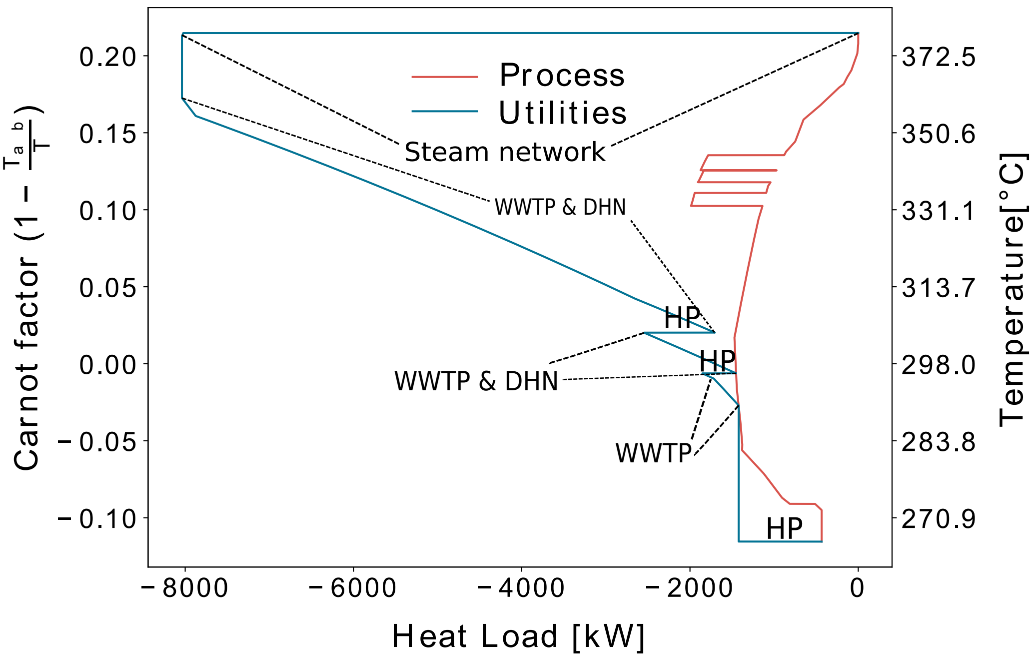
Figure 6: Capital expenditures for several technologies' combinations in k€/year; HP - Heat pump structure; SN - Steam network structure; PV - Photovoltaic panels; ENG - Gas/Biogas engine; HEX - Heat exchangers network; Elec - Electricity consumed from the grid.

The solution that minimizes total cost (24 M€/year) is depicted with larger markers at Fig 5 and corresponds to an annualized investment cost of 2.7 M€/year and operating cost of 21.3 M€/year. It reduces electricity consumption from 5,335 kW to 1,616 kW (corresponding to 70 % reduction) and increases exergy efficiency from 24.9 % to 49.1 %. It presents one of the highest investments in the heat exchange network, alongside with a moderate investment in the steam network, yielding a total pay-back time of 12 years, without considering any incentives that could be given by reducing the environmental footprint.
In general, PV panels are implemented as they have present a good trade-off between investment and electricity gain. The HP superstructure has a curious behaviour, that might be correlated with the heat recovery investment. For higher values of heat recovery the need for heat pumping is reduced, meaning that part of the heating demands are no longer covered by the HP; the opposite is also true.
Fig. 7 shows composite curves for the minimum total cost. When compared to the reference scenario (Fig. 3), the exergy destruction is improved, which is qualitatively displayed by the

area in the Carnot composite curves.



(a)



(b)

Figure 7: Grand composite curve (a) and Carnot composite curves (b) for minimum total cost

It is interesting to note that, among the technologies selected in the pareto points, none of the less established technologies (including PSA, electrolysis, SOEC and methanation) were present. The reason is linked to the increasing investment and higher electrical requirement. The decision concerning investment is always a trade-off between capital expenditure and operational gains, observing environmental, social and political matters.

4.3. Extra scenarios

It would be interesting to let the system choose alternative technologies besides the ones presented in the previous section. Carbon dioxide is an excellent source of carbon, and together with hydrogen, are the pillars of all the main basic chemicals. With increasing regulation concerning CO_2 emissions, technologies that are able to treat and convert carbon dioxide to useful products are strongly encouraged not only economically but also politically. For that purpose, in the superstructure flowsheet (Fig. 4) biogas is forced to be cleaned in a PSA for further treatment, activating a set of technologies that were not the first choice, mainly due to high electric consumption and the lower price of gas compared to electricity. Several extra scenarios are developed.

1. Scenario 1: Biogas to PSA with current prices for both electricity and gas
2. Scenario 2: Biogas to PSA with discount price of electricity. The increasing amount of renewable, as well as specific daily changes in power price would allow electricity to be bought at a discount rate. Indeed, several countries in Europe have periods in which electricity is extremely cheap, due to a large supply and low demand. This scenario is thus considered with a discount of 25% on the reference value, resulting in 60 €/MWh.
3. Scenario 3: Biogas to PSA with premium value for SNG. Countries all over Europe, are implementing policies in which SNG produced from sustainable sources is economically incentivized. The same value as in [52] was considered. The publication reports a value of 120 €/MWh for SNG originated from waste biomass (a sustainable source).
4. Scenario 4: Besides the considerations in Scenario 3, also a 50 % discount on electricity price.

Table 6 shows the MILP results when considering the minimization of operating costs according to (4), for the described scenarios. As biogas is forced to follow a specific path, the investment in SOEC, Meth., Electro. and PSA is the same for all the scenarios.
As expected, with the assumed reference parameters for both electricity and gas, new technologies are clearly inefficient, both in economic and exergetic terms. For the former, the high capital expenditure associated with new technologies, and for the latter the electricity consumption, drive the system to negative pay-back times, which translates into a non-recovery of investment.
When the electricity price is cut by 25% (Scenario 2), there is a slight reduction of investment in the gas engine, without changing dramatically the system. Interesting to notice is that, with increasing valorization of SNG (or bio-SNG), the system stops using gas to feed the engine and boiler (belonging to the steam network) and starts its injection in the grid, providing a

Table 6: Extra scenarios compared; SOEC - Solid oxide electrolysis cell; Meth. - Methanation and gas cleaning unit; Electro. - Electrolyser; PSA - Pressure swing adsorption; PBT - Pay-back time;

		Scenario 1	**Scenario 2**	**Scenario 3**	**Scenario 4**
CAPEX (k€/year)	HP	47	47	48	48
	SN	3261	3261	3081	1630
	PV	82	82	82	82
	ENG	710	552	0	0
	HEX	1619	1567	1813	1608
	SOEC	4264	4264	4264	4264
	Meth	373	373	373	373
	Electro.	559	559	559	559
	PSA	172	172	172	172
	Total Capex (M€/year)	11.1	10.9	10.4	8.7
OPEX (k€/year)	Elec.	5615	4211	7545	3797
	NG/SNG	0	0	-4220	-4220
	Water	7	7	16	7
	Air	5	5	5	5
	DHN	-549	-549	-1	-29
	WWTP	20392	20392	20392	20392
	Incinerator	482	482	482	482
	SOEC	38	38	38	38
	Meth.	33	33	33	33
	Total Opex (M€/year)	26.0	24.6	24.3	20.5
	Total Cost (M€/year)	37.1	35.5	34.7	29.2
	PBT (years)	-41.6	-88.1	-115.4	29.6
	Exergy eff. (%)	11.2	11.2	86.8	86.5

sound economic profit and a considerable increase in exergy efficiency. This happens for both Scenario 3 and 4, albeit for the last one, electricity costs are also reduced, which allows for an investment with a return due in 30 years time, which is still a very long period.
It could be stated that, with proper incentives to (bio)-SNG production allied with cheap and abundant electricity, conditions are met for a profitable and efficient system. For SNG, proper incentives are heavily dependent on policy makers, due to its connection with environmental metrics as well as commercial agreements. Concerning electricity resources, it is perfectly achievable in today's context, considering the growing levels of renewable and taking advantage of seasonal and intra-daily availability. Besides, as technology is being developed and new materials made available, the reported technologies are expected to become more affordable, allowing faster capital recovery. Lastly, though not considered, CO_2 credits/taxes are also a strong possibility for reducing overall costs.

5. Conclusions

Waste management and wastewater treatment in particular are one of the major areas of development in industry and academia; with increasing demographics, GDP and subsequent consumption, waste production rate will increase. Its proper treatment is of crucial importance for a sustainable planet. A general set of technologies is proposed starting from a dairy wastewater input, that goes into a WWTP, yielding a treated effluent, sludge and biogas. A reference case was defined with well integrated technologies found in today's WWTP. Operational expenses are considerable, as no subsidies or gate-fees were considered. As heating demand in a dairy is essentially low grade heat, substantial exergy improvements are possible. A superstructure scenario was developed, built on top of the reference case by incorporating other technologies. As a major remark the use of a proper steam network (with co-generation) and a heat pump superstructure, allow for major savings, although depending on investment. A trade-off between operational and capital expenditures was obtained, accompanied with other metrics such as exergy efficiency and pay-back time. The solution with the lowest total cost, 24 M€/year, allows capital recovery after 12 years, external electricity reduction by 70%, and a substantial exergy efficiency increase from 25 to 49 %.
Aiming for the activation of a set of alternative technologies, some scenarios were created forcing biogas to be treated in a pressure swing adsorption unit and undergoing a series of steps to produce SNG. Albeit non-viable with the assumed reference prices, with proper incentives and policies for both electricity and gas prices, economic viability is achieved for a scenario of CO_2 conversion to SNG, yielding extremely high exergy efficiencies. Furthermore, equipment acquisition values are expected to become lower, due not only to technical and scientific developments but also due to mass production. The fact of using a renewable resource for producing heat, electricity and SNG, is also a sound environmental indicator for future implementation.

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Future work

- Multiperiod approach, to account for seasonal variations; particularly relevant for photovoltaic panels, but also for the dairy production and the WWTP. In reality, dairy facilities change their production depending on the season, which in turn changes the average wastewater composition.
- CO_2 credit/tax on emissions, would have the potential to strongly influence operating costs and thus investment decisions. As emitted CO_2 is from sustainable sources, the accounting of CO_2 is differently processed, which might allow the industrial complex to buy CO_2 credits from external entities.

Nomenclature

$WWTP$, Waste water treatment plant

WW, Wastewater

TS, Total Solids

BOD, Biological oxygen demand

$SOEC$, Solid oxide electrolysis cell

$SOFC$, Solid oxide fuel cell

DME, Dimethyl ether

SNG, Synthetic natural gas

LHV, low heating value, kJ/kg

HHV, high heating value, kJ/kg

$CEPCI$, Chemical Engineering Plant Cost Index

$CAPEX$, Capital Expenditures

$OPEX$, Operational Expenditures

PBT, Pay-back time, years

HP, Heat pump structure

SN, Steam network structure

PV, Photovoltaic panels

GHI, Global Horizontal Irradiation

ENG, Gas engine

HEX, Heat exchanger network

$Meth.$, Methanation reactor and gas cleaning unit

$Electro.$, Electrolyser

PSA, Pressure swing adsorption

$Elec.$, Electricity

NG, Natural Gas

SNG, Synthetic natural gas

DHN, District heating network

T_{ref}, reference temperature, $^{\circ}C$

T_u, temperature of stream u, $^{\circ}C$

Q_u, heat content of stream u, kW

E_u^H, heat exergy of stream u, kW

$s.p.$, sizing parameter, $J/(kgK)$

Greek symbols

η exergy efficiency

τ annualization factor

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