

Research Paper

Life cycle assessment and techno-economic comparison of methane production routes from sewage sludge: Incineration vs. Hydrothermal Gasification and Anaerobic Digestion

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ABSTRACT

The disposal of sewage sludge poses significant challenges due to storage difficulties and the presence of pollutants and pathogens. Conventional treatment methods, such as incineration and anaerobic digestion (AD), are compared to advanced technologies like hydrothermal gasification (HTG) with syngas upgrading for methane production. Various indicators — i.e. environmental impacts, exergy efficiency, capital expenditures, and operational expenses — are assessed to evaluate these pathways. The scope of the life cycle assessment (LCA) accounts for the waste recovery, the treatment infrastructure, and material and energy flows. It additionally account for the substitution of valorized products. For instance, the methane produced is assumed to replace fossil methane with a substitution rate ranging from 100% (entire replacement of fossil methane) to 0%, considering either no substitution or future decarbonized energy systems. As a result, HTG achieves an exergy efficiency as low as 10.2 %. Yet, carbon management strategies, such as co-electrolysis (co-SOEC), can improve the exergy efficiency up to a value of 62.2 %. The most favourable routes in terms of GHG emissions are those mineralizing CO₂ from sludge gasification (−1,100 kg CO₂-eq/FU) or maximizing sludge-to-methane conversion (e.g., HTG with co-SOEC, −790 kg CO₂-eq/FU). However, this benefit reverses under a 0 % substitution scenario (+770 kg CO₂-eq/FU). In contrast, AD-based routes with lower energy demand show impacts between − 328 and + 70 kg CO₂-eq/FU, also being more competitive in terms of costs. Beyond GHG emissions, trade-offs emerge across other impact categories, notably water scarcity, ecosystem quality, and fossil and nuclear energy use, particularly for routes involving CO₂ mineralization and co-SOEC due to their high energy demand.

1. Introduction

Economic growth and resource consumption are tightly interlinked, leading to resource scarcity and environmental impacts such as greenhouse gas (GHG) emissions, pollution, and biodiversity loss [1,2]. This relationship is particularly evident in the linear economy model, in which economic expansion directly correlates with rising waste production [3]. Waste not only contributes to environmental degradation through soil and water pollution, but also generates significant GHG emissions [4]. In 2016, municipal solid waste (MSW) alone produced around 1.6 Gton of CO₂-eq emissions [5]. Concerning wastewater production per capita, the trend tends to stabilize over time in Switzerland.

Sewage sludge production is following a similar evolution [6].

Even with a constant sewage sludge per capita production, its disposal poses significant challenges due to storage difficulties and the presence of pollutants and pathogenic organisms [7]. These health concerns encouraged authorities, such as the Swiss government, to implement stricter regulations in 2016, prohibiting the direct use of sludge as fertilizer [8]. Instead, incineration became the preferred disposal method, except when valuable constituents can be recovered [9]. Indeed, it remains the most used technology for sewage sludge disposal in Europe, representing 37 % of the applications, followed by agricultural use with 35 % [10]. However, the high moisture content of sewage sludge makes incineration energy-intensive and poorly effective, unless paired with effective energy recovery systems. Incineration also causes a

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Nomenclature			
Abbreviation/Symbol	Definition		
AD	Anaerobic Digester	ISO	International Organization for Standardization
c	Specific cost (€ per kg or kWh)	KPI	Key Performance Indicator
CAPEX	Capital Expenditures (€)	LCA	Life Cycle Assessment
CC	Carbon Capture	LHV	Lower Heating Value (MJ/kg)
CCS	Carbon Capture and Storage	m	Mass flow rate (kg/h)
CE	Circular Economy	MILP	Mixed-Integer Linear Programming
CHP	Combined Heat and Power	n	Size of the sample
CH ₄	Methane	N	Lifetime
CO ₂	Carbon Dioxide	op	Operational
CO ₂ -eq	Carbon Dioxide-equivalent	OPEX	Operating Expenditures (€)
co-SOEC	co-electrolysis Solid Oxide Electrolysis Cell	r	Resource
CP	Compressor	RE	Reference Environment
$\Delta_f G$	Gibbs standard free energy (kJ/mol)	RSME	Root Mean Square Error
E _{ch}	Specific chemical exergy (MJ/kg)	SMR	Steam Methane Reforming
EI	Environmental impact	T	Time Period
Env.	Environment	TC	Total Carbon
f _u	Size of the technology	t _{op}	Total Operating Time (h/y)
GHG	Greenhouse Gas	TOTEX	Total Expenditures (€)
h _{lv}	Enthalpy of evaporation (kJ/kg)	u	Unit or technology
HTC	Hydrothermal Carbonisation	W	Power (kW)
HTG	Hydrothermal Gasification	WGS	Water Gas Shift
i	Interest rate (%)	η	Efficiency
		ξ	Emission Factors
		φ	Ratio

scattering of carbon atoms (mainly biogenic) into the atmosphere in the form of CO₂, which is contrary to the principles of circular economy (CE) [11]. Paradigm shifts as CE invite a reconsideration of the multiple transformation paths carbon-based wastes can undergo (such as production of fertilizers and conversion to fuels and gases). Moreover, the defossilization of the energy system introduces a need to produce alternative fuels and storable forms of energy, integrating waste management solutions with broader environmental and energy goals.

Sewage sludge is also rich in nutrients such as nitrogen and phosphorus, two critical elements to major biogeochemical flows, which cycles are associated with planetary boundaries. Anthropogenic use of phosphorus causes a leak of this atom to the oceans, which may cause an anoxic event. Nitrogen affects the resilience of ecosystems by increasing acidification and eutrophication [12]. The two most important leaks are caused by agriculture and wastewater discharge [13]. From a broader perspective – beyond the planetary boundaries indicators, phosphorus is also a critical and non-substitutable element that guarantee worldwide food security. Reducing nutrients leaks by improving sewage sludge treatment not only is an effort towards remaining within the safe operating space of planetary boundaries, but also supports long-term food security through the conservation of essential resources.

A potential pathway for long-term sustainable treatment of sewage sludge is hydrothermal gasification (HTG), a process that converts wet biomass into a syngas mainly composed of CO₂, CH₄, H₂, and CO. HTG does not necessarily require an external catalyst, as the reaction occurs under supercritical water conditions (above 374 °C and 22.1 MPa). In this state, water itself acts as a reaction medium and can exhibit catalytic behavior [14,15,16]. The high temperature and pressure operating conditions facilitate the precipitation of minerals like phosphorus and potassium, that can be extracted and re-used [17]. HTG involves three steps. Firstly, the feedstock preparation stage, by taking biomass with a dry matter content ranging from 1 % to 50 % and preparing it accordingly, which may involve dilution and grinding if necessary. The biomass is converted in the reactor, which heats and conserves the pressure of the feedstock. This step precipitates the minerals whereas a mix is formed in the gasifier. It goes through a first gas/liquid separation, at high pressure. Afterwards, the syngas is purified, and the

remaining liquid/gas solution goes through a second separation unit, at low pressure. Methane can be extracted from the syngas using specific membranes, and injected into the grid.

This process has several benefits, including the residence time of the process (10 min), a high carbon conversion rate, a strong reduction of ultimate waste, and the elimination of bacteria and viruses [18]. The main drawback of the technology is the quantity of water that needs to be heated up at high temperature and pressure [16], making it energy-intensive. Proper heat integration can help mitigate this issue. Several types of biomass can be converted using HTG, such as wood [19], digestate [20], MSW [21], chicken manure [15], and sewage sludge [22]. Different works have studied HTG processes, in which syngas upgrading to hydrogen has been the focus of the analysis. For instance, Hantoko et al. [22] is simulating on Aspen Plus the production of hydrogen from sewage sludge, at different temperature and biomass to water ratio. More recently, Inuwa et al. [23] are co-producing hydrogen and electricity from poultry litter using HTG operating at 850 °C.

Although the studies are pertinent, they solely focused on thermodynamic or environmental indicators, overlooking the environmental and economic analysis. Another major drawback is the validation of the Aspen model, which is achieved considering one set of data. For example, Hantoko et al. [22] are modelling the HTG of sewage sludge and calibrated the model using glycerol HTG dataset from Byrd et al. [24]; Fadilla N. et al. [20] builds upon experimental data from Antal Jr et al. [25], e. g. cornstarch gasification to calibrate a model used on digestate. Moreover, the conversion of syngas into methane has not been investigated, although the syngas produced at the gasifier outlet are mainly composed of methane, especially when considering sewage sludge as feedstock [22]. Finally, HTG technology has not been compared with other thermochemical conversion processes for biomass, simultaneously considering economic, thermodynamic, and environmental criteria. Heat integration can mitigate negative externalities of the HTG as thoroughly studied by Gassner and Maréchal [26]. They considered the integration of an organic rankine cycle for recovering waste heat from HTG. In the present work, the Pinch Analysis has shown that a transcritical CO₂ cycle is more efficient for this purpose, as it features a gliding heating profile for the working fluid that closely fits

the cooling requirements of the HTG system.

Several studies have investigated sewage sludge treatment technologies using LCA approaches. Mayer et al. [27] conducted an LCA comparing four pathways considering AD as core technology, further integrated with hydrothermal carbonisation (HTC), pyrolysis, and incineration as treatment pathways for sewage sludge in Germany. Although they evaluated 17 impact categories based on the ReCiPe midpoint method, their analysis lacked thermodynamic and economic assessments. Moreover, technologies were assessed independently, without considering heat integration between the HTC, pyrolysis, and incineration, which could improve system efficiency and reduce overall impact score. Pimenta-Ocampo et al. [28] focused on the characterization of pyrochars and hydrochars from anaerobically digested sludge, comparing structural and stability properties. While insightful in terms of material properties, their study did not address environmental or economic aspects. Mannarino et al. [29] performed an LCA on HTC and AD, evaluating three conversion pathways. However, their study did not include thermodynamic performance or economic feasibility. Li et al. [30] extended the LCA scope to biochar and hydrochar production from straw manure. They focused on endpoint impacts and soil conditioning effects, limiting applicability to sewage sludge management. Hosseini et al. [31] compared pyrolysis, AD, and composting in Swedish municipalities, assessing LCA performance across four scenarios. While they acknowledged the agronomic benefits of digestate, the study remained limited to impact scores and did not explore integrated system configurations.

Despite the relevance of the studies, several research gaps remain. First, prior models often lack rigorous validation, particularly for core technologies like HTG, whose syngas composition is highly sensitive to the modelling assumptions. Second, gas treatment technologies, such as syngas upgrading favouring methane conversion, or CO₂ sequestration have not been considered. Third, the role of heat integration, a key lever to mitigate energy consumption in HTG and incineration systems, has been largely overlooked [19].

Fourth, most studies evaluated only one or two indicators, usually environmental or thermodynamic, without integrating a comprehensive multi-criteria analysis combining thermodynamic performance, environmental impacts (including endpoint categories), and cost analysis. Thus, in this work, a holistic approach is proposed, comprising heat integration, economic analysis, life cycle impact assessment over the five endpoint categories of the IMPACT World + method [32], and exergy efficiency. The validation of the HTG model is assessed considering the results reported in the literature, which were obtained either from laboratory-scale experiments [15,18,25] or modeling studies [20,22], for five types of biomass feedstock, with different operating conditions (temperature, pressure and biomass-to-water ratio).

This paper aims to develop a consistent and adaptable methodology to evaluate the environmental, economic, and exergetic relevance of technology combinations for the treatment of carbon-based waste. To achieve this, the following steps are proposed:

1. Identify the most relevant technologies to be assessed either as alternatives or in complement with the main existing treatment routes, namely anaerobic digestion (AD) and incineration.
2. Implement proper heat integration within the processes in order to mitigate their main drawback—high energy intensity—and to enable a fair comparison between the different configurations.
3. Evaluate the different routes using multiple key performance indicators (KPIs), including: exergetic performance, through theoretical calculations; economic viability, based on techno-economic analysis (CAPEX and OPEX); environmental impact, using endpoint indicators and the LCA methodology.
4. Incorporate an innovative parameter into the LCA approach: the methane substitution (or avoidance) rate, which reflects societal scenarios and the evolving energy mix—from 100 % substitution (assuming full replacement of fossil methane in the current mix), down to 0 % (assuming a future where methane is no longer used as an energy

carrier).

2. Methods

This section describes the model and the optimization methods applied to perform the process integration that aims to minimize the energy requirements, total cost, and impact score. To this end, the waste heat recovery is maximized through the energy conversion system based on the pinch analysis. The studied process routes have been modeled using Aspen® Plus V11 software to carry out the mass and energy balances, as well as to model the complex interactions occurring during the HTG [63] and the electrolysis system [62]. Each transformation process will be described in this section. The OSMOSE platform is used to solve the energy integration problem and the multi-objective optimization on costs and impacts [65]. For the calculation of environmental impacts, the Ecoinvent database is used with the IMPACT World + method [32]. Moreover, OSMOSE platform also allows to graphically determine the minimum energy requirement, while enabling the calculation of both capital and operational expenditures.

2.1. Treatment process and carbon conversion routes

In this work, nineteen conversion routes for sewage sludge treatment are compared, as shown in Table 1. Three core technologies are studied: HTG, incineration, and AD. Building upon these core technologies, different gas management options are considered: first, direct CO₂ emission into the environment, then CO₂ sequestration via mineralization, and finally the use of electrolysis and methanation to enhance the conversion of carbon-based gases into methane. The digestate can either be landfilled, gasified, or incinerated. All possible combinations of these approaches result in the nineteen treatment routes presented here. The block flow diagram summarizing the combinations can be found in Fig. 1 – whereas the detailed process flow diagram with utilities can be found in the supplementary information S11.1.

Conversion routes listed from (a) to (e) are based on gasification. Route (a) considers HTG with methanation of the syngas and membrane separation to recover CH₄, but without carbon capture. However, the CO₂ recovered by the membranes can either be mineralized (b), or further transformed into methane using co-SOEC (c). To avoid the use of CH₄ separation membranes, an alkaline electrolyzer producing hydrogen to enhance methane production with the methanation system is considered, either with emissions of CO₂ to the environment (d) or with its mineralization (e). The digestion-based routes complete the list of alternative configurations considered for sewage sludge treatment and valorization. Some of them do not consider treatment of the

Table 1
Conversion routes analyzed.

Name	Conversion Route
a	HTG
b	HTG, mineralisation
c	HTG, co-SOEC
d	HTG, alkaline electrolyser
e	HTG, alkaline electrolyser, mineralisation
f	Digester
g	Digester, mineralisation
h	Digester, co-SOEC
i	Digester, HTG
j	Digester, HTG, mineralisation
k	Digester, HTG, co-SOEC
l	Digester, HTG, alkaline electrolyser
m	Digester, HTG, alkaline electrolyser, mineralisation
n	Digester, incineration
o	Digester, incineration, CC, mineralisation
p	Digester incineration, CC, co-SOEC
q	Incineration
r	Incineration, CC, mineralisation
s	Incineration, CC, co-SOEC

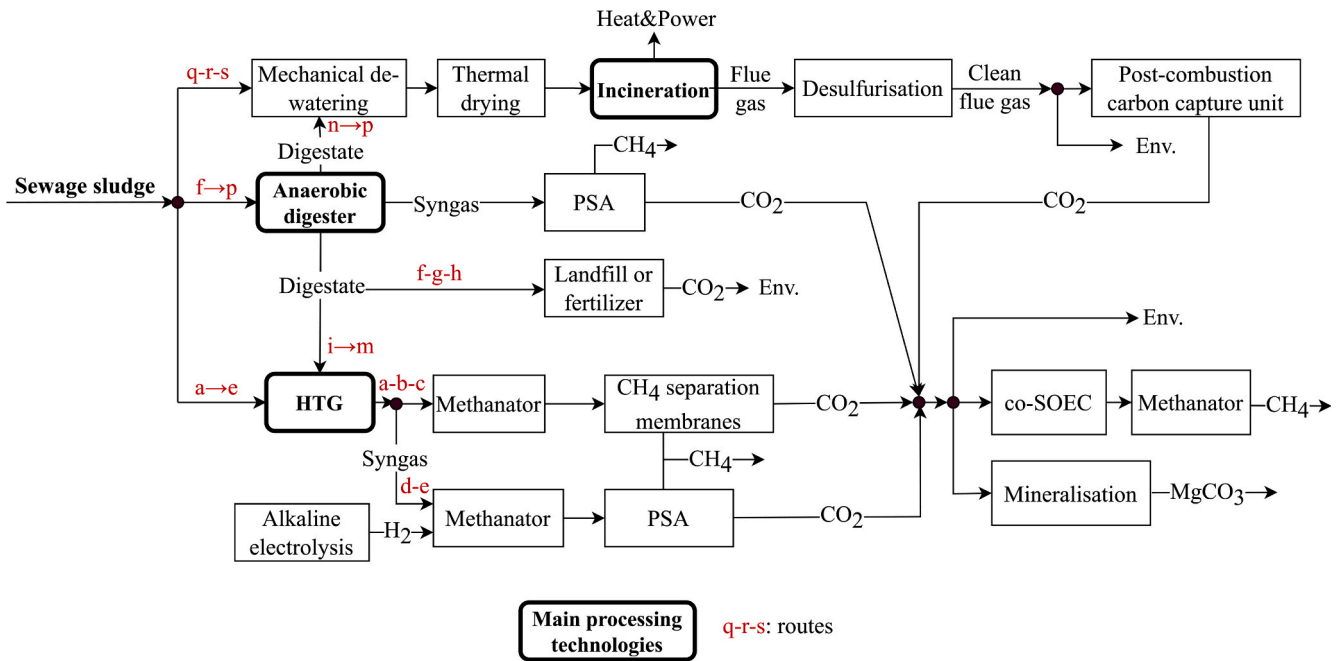


Fig. 1. Integrated flowsheet of sewage sludge treatment technologies with upgrading syngas units and CO₂ management processes. The routes studied are in red, and described in Table 1. Env: environment.

digestate: (f) evaluates the digester alone, (g) includes upgrading of the biogas to methane and converting the CO₂ into syngas using co-SOEC and methanation unit, and (h) involves mineralization of the CO₂. Routes (i) to (p) also build on the digestion process but the digestate management incorporates gasification steps (i–m) or incineration (n–p) of the digestate, with CO₂ management units as described earlier in this paragraph. Finally, the conventional incineration (q) method, without CO₂ recovery and with only basic desulfurization treatment using a limestone scrubber, has been modeled. This route can be revamped with point-source carbon capture of the CO₂ in the flue gas, which can either be upgraded to methane via co-SOEC (r) or permanently stored through mineralization (s).

2.2. Heat integration of the routes

In order to mitigate the main drawback (high energy intensity) of the nineteen routes and to enable a fair comparison between the different configurations, heat integration is performed according to the pinch analysis. The process integration technique is used to calculate the maximum heat recovery. The method applied is the following: first, a list of the units involved in the heat recovery is defined, targeting the minimum energy requirement and giving temperature enthalpy profiles. Then, a set of possible utilities is sized and integrated to the process [33,34].

2.3. Process modeling and simulation

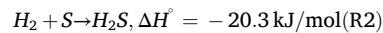
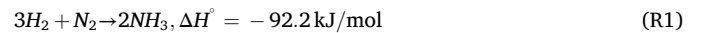
The modeling of the technologies compared in this work is explained in this section. The ultimate and proximate composition of the sewage sludge used for the modeling can be found in SI1.2.

2.3.1. Hydrothermal gasification

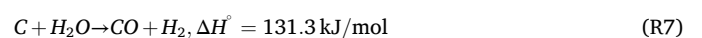
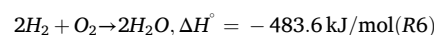
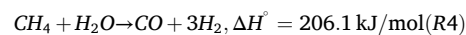
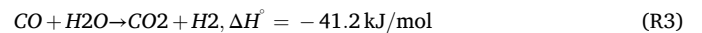
The HTG process is modeled on the Aspen® Plus software using the Peng-Robinson thermodynamic method. Firstly, the water is pumped and heated to supercritical conditions, using a pump with an isentropic efficiency of 85 % and several. It simulates a supercritical steam generator, which is a boiler operating at supercritical pressure and temperature (i. e. above 220 bar and 374 °C). It allows to turn the liquid

water in a supercritical steam, with no phase change (there is no mixed phase between water and steam), until the water reaches 500 °C and 250 bar.

The sewage sludge is pumped and heated to 400 °C. The gasification is simulated using first the RYield reactor, which decomposes biomass into atoms and simple molecules (e. g. C, H₂, O₂, N₂, Ash and S), and two RGibbs modules that emulate the conformation of the syngas components, based on the mechanism proposed by Okolie et al. [18]. In the first reactor, nitrogen, and traces of ammonia and hydrogen sulfide are formed, according to Reactions (R1) and (R2):



Ammonia, nitrogen, and hydrogen sulfide are separated from the other components using a classic separator unit. In the second reactor the syngas is formed according to a stoichiometric approach. The set of equations used in this work is the water gas shift (WGS) and the steam methane reforming (SMR) (Reactions R3 and R4), which are the most common in literature (as referenced in SI1.3). The steam reforming, water formation, and co-hydrogenation are also considered (R5–7). The gasification reaction is based on equilibrium, using the RGibbs reactor in Aspen Plus v11, with temperature approach $T = 0^\circ\text{C}$. This set of reactions has been validated according to the composition of the syngas produced, as discussed in the following section.



2.3.2. Validation of the model

The HTG model has been validated using literature data at different

pressures, temperatures, biomass-to-water ratio, and biomass composition. To this end, studies based on sewage sludge [22], chicken manure [15], soybean straw [18], cornstarch [25], and digestate [20] have been considered. The disparity of feedstock biomass makes it challenging to accurately approximate the gas composition. In that sense, a first calibration of the model and sensitivity analysis has been achieved using Hantoko et al. [22] results on the effect of feedstock concentration, pressure and temperature over the molar composition of syngas. Next, the calibrated gasifier has been tested with the four other feedstock, at their respective reaction conditions. The root mean square error (RMSE), measuring the average difference between the values taken from literature and the values from this work is calculated. The RMSE is 0.11, 0.08, and 0.07 for methane, carbon dioxide, and hydrogen respectively. The results and operating conditions can be found in Fig. 2. The RMSE values reflect an adequately calibrated model suitable for simulating syngas composition across different biomass feedstocks. While improvements are possible, particularly for methane predictions, the current level of accuracy is likely sufficient for the intended applications in this work. Additionally, the mean percentage of error for Fig. 2 is 20 % for methane, 9 % for carbon dioxide and 7 % for hydrogen.

In the following section, the details on the processes to transform the syngas produced are further elaborated.

2.3.3. Syngas treatment and upgrading units

The syngas produced by the HTG, with a molar fraction of 10 % H_2 , pass through the shift reactors and subsequently, the methanation system, to convert the CO and CO_2 into methane [35]. Two conversion routes are compared. The first is increasing the carbon oxide (CO, CO_2) gases conversion into methane using an additional amount of hydrogen (produced using an alkaline electrolyzer operating at 80 °C) at the required stoichiometric ratio. The second one uses only the hydrogen already present in the syngas to partially convert the carbon oxide gases into methane, with a subsequent separation of excess CO_2 via CH_4 separation membranes. The membrane system is modeled considering a double-step system configuration with permeate recycle, allowing a CH_4

recovery higher than 85 % [36,37]. The temperature of the membrane operation is 60 °C. For the recirculation, two compressors at 4 bar with intercooling are used. Then, a compressor to 5 bars is used to increase the methane pressure to the level required for the export to the methane grid.

2.3.4. Anaerobic digester with purification of the biogas

The sewage sludge can also be transformed into a methane-rich gas in an AD plant. AD is a biological process in which organic material is broken down and converted into biogas in an oxygen-free environment, facilitated by microorganisms. The biogas obtained is a mixture primarily consisting of methane and carbon dioxide. The residual, non-gaseous byproduct, known as digestate, can serve as a fertilizer, either in liquid form or after drying [38]. The product considered consists of 75.2 % moisture, 54.7 % volatile matter, 7.2 % fixed carbon, and 41.5 % ash in the proximate analysis, while the ultimate analysis shows 31.5 % carbon, 4.3 % hydrogen, 4.8 % nitrogen, 1.7 % sulfur, 16.2 % oxygen, and no chlorine [39]. In this setup, a thermophilic reactor is utilized, operating at 55 °C. This temperature is chosen to reduce retention times, enhance the breakdown of substrates, and ensure more effective pathogen elimination [40]. During the initial phase, the organic feedstock is fed into the digester using a hopper. The biomass is consistently heated to 55 °C and stirred within the reactor. The biochemical transformations occurring over a retention period of around 30 days convert the organic waste into biogas, mainly comprising methane, carbon dioxide, and traces of hydrogen sulphide. The methane potential for sludge is estimated at 366 Nm^3 per ton [41]. The remaining digestate is commonly applied as a fertilizer for specific crops [38]. The methane produced is directly cleaned and upgraded using water scrubbing, which is a cost-effective and mature method for CO_2 and H_2S removal in the biogas removal field [42]. In the water scrubbing process, the pressurized biogas is injected at the bottom of the absorption column and washed with water injected in the overhead of the column. The counterflow contact between the gas and the liquid phases allows reaching high mass transfer, effectively absorbing CO_2 [43]. The pressure of the absorber is

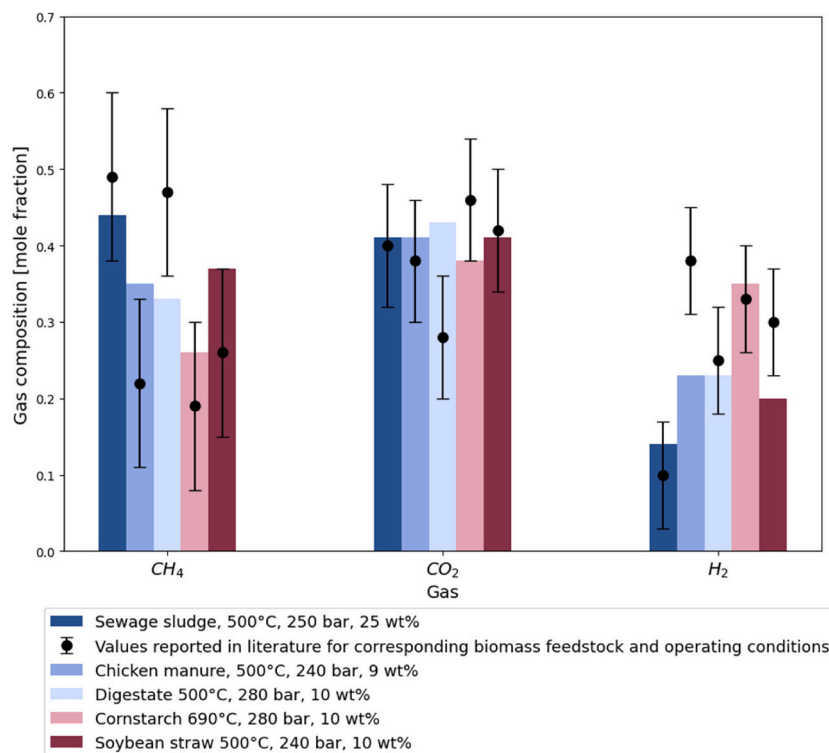


Fig. 2. Comparison of the syngas composition calculated for the HTG gasification model in Aspen® Plus in this work with the values and operating conditions reported in literature for other biomass feedstock. Error bars on the literature data correspond to the RSME.

set at 30 bars to increase the capture efficiency and allows reaching proper composition for the methane to be sent to the grid.

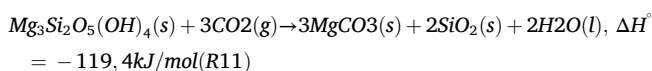
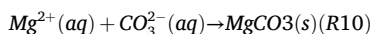
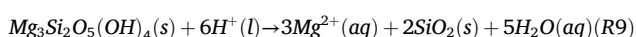
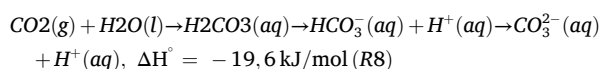
2.3.5. Incineration

The incineration process considers waste pre-treatment using first a mechanical crusher followed by thermal drying [39]. The incineration reactor has been modeled on Aspen® Plus using RYield and RGibbs reactors. After incineration of the feedstock, the ashes are separated from the flue gas for disposal, and the gas is desulfurized using a limestone scrubber, which has an efficiency of 97 % and is suitable for this flue gas having a SO₂ molar concentration 3.5 % [44]. Flue gases from biomass incineration, although considered as biogenic and not contributing to the net emissions balance, can be captured more easily at the source due to higher concentration compared to that in the atmosphere. It disincentivizes the use of less efficient options for carbon abatement like direct air capture. In that sense, a carbon capture unit for post-combustion gases is evaluated, allowing for the recovery of 90 % of the CO₂ from the flue gases [45]. The CO₂ captured, whether it is from the post-combustion unit or the CH₄ separation membranes, can be utilized or stored.

2.3.6. CO₂ management units

The CO₂ flows coming out of the processes have a high purity. They can be upgraded using a co-SOEC, converting carbon dioxide and water into a mixture of CO, CO₂, and H₂. This model has been adapted from Domingos et al. [46], which was based on the equations described by Ni et al. [47]. The mixture can be converted into methane in the methanator system based on the TREMP® process, in which a series of methanation beds are intercooled by recycling or indirect inter-cooling in order to achieve higher conversion [48]. The co-SOEC operates at 800 °C, requiring steam generation and fuel cooling.

Another CO₂ upgrading route considered is permanent storage using serpentine-based CO₂ mineralization. The captured CO₂ is dissolved in water, and the solution is added to the pre-treated rocks. Pre-treatment of the rocks consists of magnetic separation, a grinding process, and a thermochemical pre-treatment [49]. The mineralization process, including the pre-treatment of the rocks, follow several chemical reactions. The first reaction is the dissolution of CO₂ in water, forming carbonic acid (H₂CO₃), which subsequently dissociates into bicarbonate and carbonate ions, according to (R8) [50]. Next, the serpentine is dissolved in water as per (R9), where M represents an alkaline metal of group I or II of the Periodic Table of the Elements. Finally, the dissolved CO₂ precipitates, according to (R10). The overall reaction is represented by (R11).



2.4. Definition of the scope of analysis, approaches and adopted performance indicators

This section presents the scope analysis, including the definition of system boundaries and the functional unit (FU) for each route. In this study, the different treatment pathways are assessed and compared using key performance indicators (KPIs) from thermodynamic, economic, and environmental perspectives. The methodology applied to calculate and interpret these KPIs is also described in detail.

2.4.1. System boundary and functional unit

The function of the modeled system is to treat sewage sludge. To compare the routes, the FU ‘one ton of sewage sludge, treated in Switzerland, in 2024’ is used. The system boundaries can be found in Fig. 3. For consistency, the colors used in Fig. 3 to identify the processes and elementary flows are the same as those used in the impact assessment results, in SI2.3 and in Section 3.2.1.

2.4.2. Total syngas yield and carbon conversion yield

The performance of the conversion processes is established by the calculation of the methane yield and the carbon conversion efficiency. The methane yield ($\text{Yield}_{\text{CH}_4}$) refers to the amount of methane produced per unit mass of sewage sludge processed. The carbon conversion efficiency represents the fraction of carbon in the sludge that is effectively converted into methane. Detailed definitions and calculation formulas are provided in SI1.4.

2.4.3. Exergy analysis and exergy efficiency

To evaluate the thermodynamic performance of the processes, indicators based on the First and Second Laws of Thermodynamics are employed. Exergy, a metric associated with the quality of energy, is defined as the maximum theoretical work achievable by a thermodynamic system during its interaction with the environment through reversible processes, until equilibrium is established with the referred components in terms of mechanical, thermal, and chemical states [51]. Using exergy efficiency to compare thermochemical conversion pathways for sewage sludge highlights how effectively the systems utilize the energetic content of the feedstock. The system under analysis is described in section 2.4.1. The details of the calculation (determination of exergy values for material and energy flows) can be found in SI1.5.

Finally, the exergetic efficiency η of the process is determined according to Equation (1):

$$\eta = \frac{\text{Useful exergy output}}{\text{Total exergy input}} \quad (1)$$

In this expression, the total exergy input represents the exergy content of the consumed energy and material resources, expressed in MJ/kg. The useful exergy output refers to the exergy content of the primary utilizable products, also in MJ/kg, including value-added products such as methane. However, emissions of CO₂ in conventional waste treatment pathways are not accounted for as an environmental burden in this calculation.

To internalize the GHG emissions and promote accountability for avoiding carbon dioxide emissions, a modified reference environment is proposed. This reference environment is designed to internalize the climate impact and is detailed in the following section.

2.4.4. Addressing atmospheric CO₂ emissions in the reference environment

The exergy calculations in this work are based on Szargut’s reference environment, which assumes the Earth’s current state as the reference. This approach implicitly hypothesizes that CO₂ can continue to be emitted. However, the release and dispersion of chemical components have significant consequences on the Earth’s ecosystems. The emission of carbon dioxide alters the atmospheric composition, contributing directly to global warming, as highlighted by the Intergovernmental Panel on Climate Change [52]. Moreover, these emissions impact the hydrosphere by driving ocean acidification [53]. Valero et al. [54] emphasized the limitedness of mineral resources at their current concentrations. As extraction rates increase, resource concentrations will diminish, leading to higher energy costs for material recovery.

The proposed reference environment, closely aligned with Earth’s current state, incorporates an atmospheric threshold for anthropogenic CO₂ emissions – beyond which significant climatic impacts occur – to ensure the preservation of life-sustaining conditions. Maintaining these conditions necessitates a stable atmospheric CO₂ concentration.

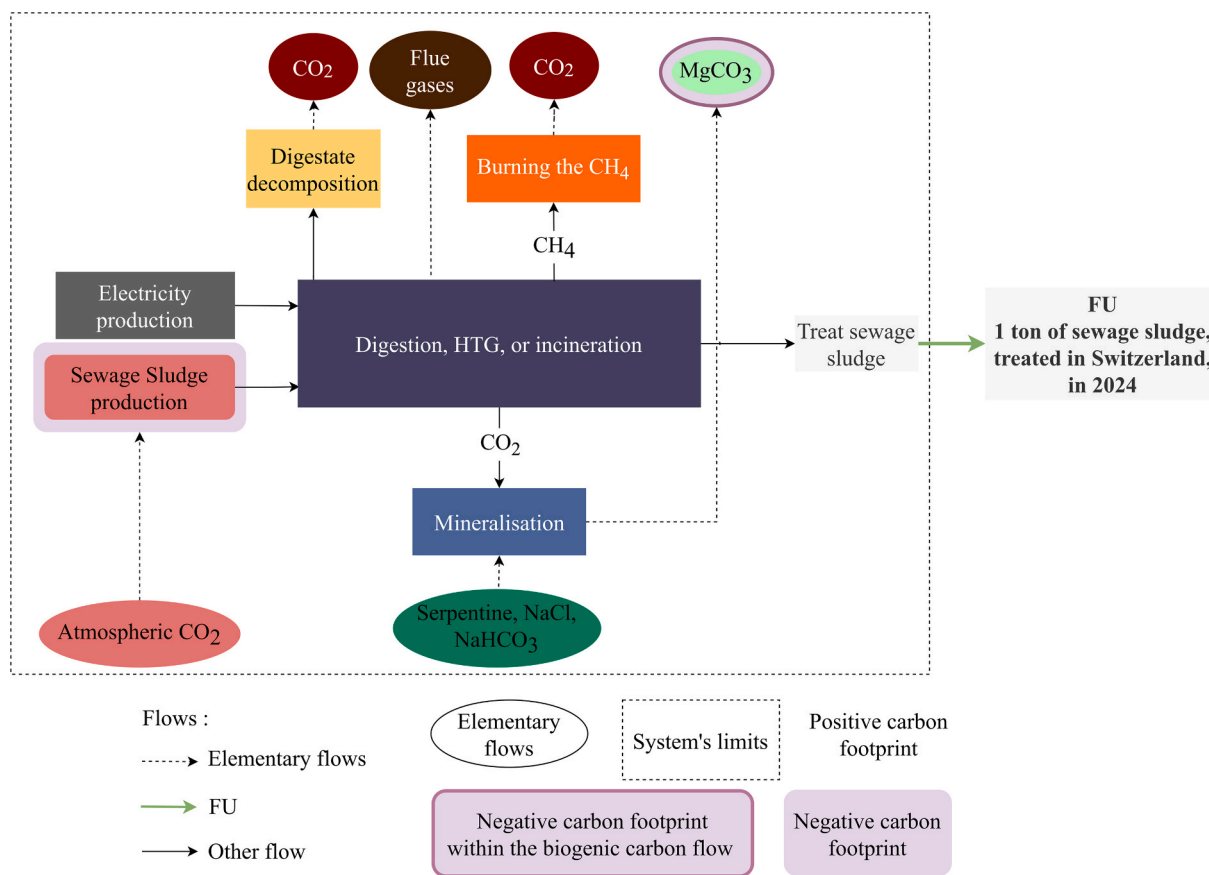


Fig. 3. System boundaries of the LCA and decomposition of the carbon footprint accounting.

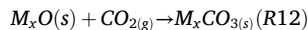
Therefore, carbon dioxide should not be treated as a dispersed component in the atmosphere but rather as a sequestered form within minerals, specifically carbonates such as calcium carbonate (CaCO_3) and magnesium carbonate (MgCO_3). Indeed, Ahrendts [55] emphasized that a reference environment must account for all substances through the production and consumption of its components. Since carbonate minerals are integral to the global carbon cycle, they provide an appropriate reference state for CO_2 in exergetic evaluations. Among the Earth's primary carbon reservoirs – the atmosphere, lithosphere, and oceans – the lithosphere serves as the most significant storage system, holding four to five times more carbon than the other two combined [56]. Carbon within the lithosphere predominantly exists as minerals, making carbonate compounds the largest natural carbon repository on the planet. Morris and Szargut [51] established four criteria for selecting a reference species, all of which are satisfied by carbonates (e.g., MCO_3 , where M represents a metal from the periodic table). First, the reference species for each chemical element must be defined, with carbonates acting as the natural reference for carbon dioxide. Second and third, the reference species must be a prevalent substance in the relevant environment, a criterion met by naturally abundant carbonates such as calcium and magnesium carbonates [56]. Finally, the fourth condition requires that the chosen reference species have sufficient thermodynamic data available to enable precise calculations – a requirement also fulfilled by carbonates due to their well-documented properties.

2.4.5. Definition of standard chemical exergy for CO_2 as carbonate

The new exergy efficiency calculation considers the proposed hypothetical reference environment, in which the reference species of carbon dioxide is a carbonate, and is complementary to the one of Morris and Szargut [51] for the other material flows involved.

The updated standard chemical exergy for the carbon dioxide component has been calculated using the formation of carbonate,

following (R12) as reference reaction, where M is a metal.



It is possible to establish the standard chemical exergy, using Equation (3) in SI1.5, of carbon dioxide when considering that carbonates are the reference species for CO_2 in the reference environment (Table 2).

The updated reference state for carbon dioxide is integrated into the exergetic efficiency calculation for each reference route of sewage sludge treatment and is illustrated for route (a) (HTG + methanator + CH_4 separation membranes, with direct CO_2 emissions) in Fig. 4. In the proposed reference environment, where CO_2 carbonation is enforced, the associated exergetic cost is incorporated into the input calculations

Table 2

Standard chemical exergy of carbon dioxide when considering that carbonates are the reference species for CO_2 in the reference environment.

Carbonate	ΔG° (kJ/mol)	Exergy (kJ/mol)
Ag_2CO_3	−437	481
BaCO_3	−1,140	923
CaCO_3	−1,130	1,025
CdCO_3	−671	630
CoCO_3	−637	616
FeCO_3	−667	397
Hg_2CO_3	−468	519
K_2CO_3	−1,060	722
MgCO_3	−1,030	968
MnCO_3	−816	765
Na_2CO_3	−1,040	775
NiCO_3	−618	618
PbCO_3	−625	589
Rb_2CO_3	−1,050	701
SrCO_3	−1,140	966
ZnCO_3	−732	719

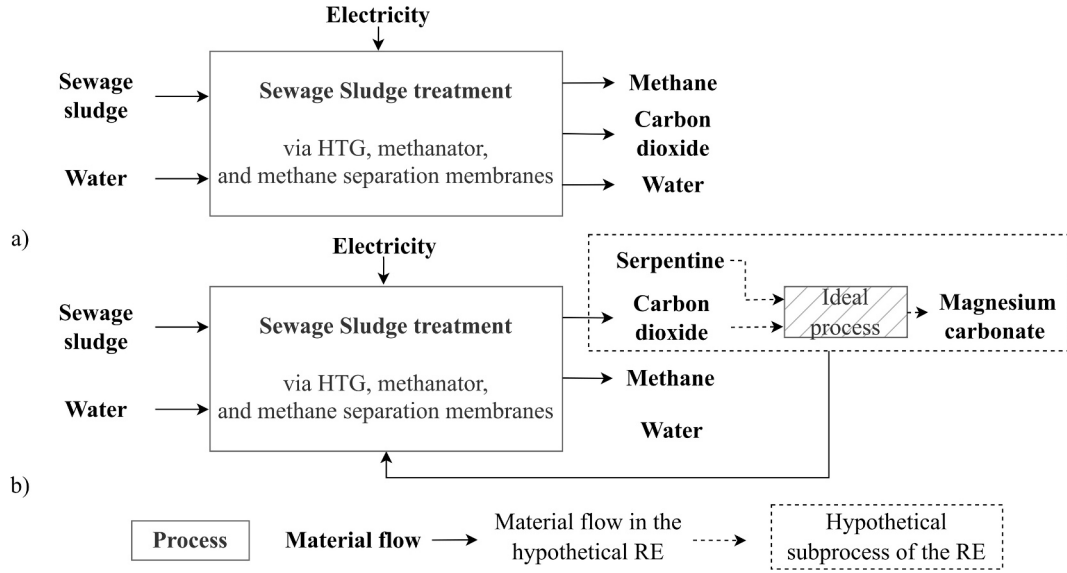


Fig. 4. Control volume of the reference route (a) of sewage sludge treatment, considering Morris and Szargut [51] reference environment (RE), and the one proposed in this work (b).

for the sewage sludge treatment route (Fig. 4, b). This approach revises the conventional control volume typically used in exergy efficiency calculations (Fig. 4, a).

2.4.6. Life cycle assessment (LCA) methodology

Impact score analysis focuses on the five areas of protection of the IMPACT World + v2.0.1 method (carbon footprint, human health, ecosystem quality, fossil and nuclear energy use, and water use) [32]. The methodology follows the Life Cycle Assessment (LCA) framework, in compliance with ISO 14040 and ISO 14044 standards. The function of the modeled system is to treat sewage sludge. To compare the routes, the FU '1 ton of sewage sludge, treated in Switzerland, in 2024' is used. The system boundary is cradle-to-grave and system boundaries can be found in Fig. 3.

The accounting of biogenic GHG contained in the feedstock follows the $-1/+1$ method, as applied within the SCORE LCA framework [57]. This method inventories all biogenic CO_2 flows. Emissions of CO_2 or other biogenic (or equivalent) GHG are characterized and accounted as fossil-based, in order to reflect their actual contribution to climate change. In that sense, the CO_2 released during biomass transformation steps (e.g. GHGs from incinerator flue gases or digestate degradation) is treated as fossil emissions. The biogenic carbon in the sludge inherit a negative value corresponding to the stoichiometric amount of CO_2 stored into the biomass by the photosynthesis. If it will be permanently sequestered the life cycle balance remain negative. In that sense, the permanent storage (in mineralisation routes) of the biogenic carbon contained in the sludge is considered as a negative elementary flow.

In this context, two biogenic carbon flows are accounted for as negative contributions: the first corresponds to the carbon captured in the sludge, and the second is the portion of that same carbon which is permanently sequestered through mineralization. To avoid double-counting the carbon, the permanently sequestered fraction via mineralization is represented within the biogenic carbon flow of the sewage sludge. This method ensures a comprehensive representation of carbon flows in the carbon footprint analysis of the different treatment routes.

System boundaries have been extended for routes that co-produce methane, incorporating both CO_2 emissions from methane combustion and potential CH_4 leaks into the atmosphere, using appropriate characterization factors.

The methane produced by the sludge is modeled with an avoidance method in the carbon footprint analysis, meaning that a fossil methane

process is considered as a negative flow. Yet, the notion of avoidance is nuanced, as some studies have shown that this alternative CH_4 production may not replace current CH_4 consumption, but rather supplement it because of economic energy-market effects [58,59]. Moreover, while fossil methane is widely used in today's Swiss energy mix, this may not hold true in the coming decades. For this reason, it has been chosen to introduce a fossil methane avoidance rate related to sludge-based methane production. This rate ranges from 100 % avoidance, assuming a complete substitution of fossil methane in the actual energy mix, down to 0 % avoidance, considering this methane as an additional source of energy in the market. This method gives a range for the GHG impact of the routes studied. The final value depends on the conditions of the energy system in which the sludge conversion process takes place. Therefore, it is important to emphasize that current results are strongly dependent on future societal and energy system needs. Routes that appear most beneficial today in terms of GHG emissions may not necessarily remain so if energy mixes or societal priorities evolve.

The calculation of the carbon footprint impact score and the integration in the OSMOSE platform can be found in SI1.6.

2.4.7. Economic evaluation of the proposed configurations

The problem is formulated as a mixed-integer linear programming problem (MILP), in which the total costs are minimized using optimization and heat integration. The total costs, or TOTEX (in €/y), are defined as the sum of the capital expenditure (CAPEX) and operational expenditure (OPEX), as presented in Equations (2) to (4). The economic assessment methodology is based on Turton et al. [60].

$$\text{CAPEX} = \sum_{u \in U} \frac{i(1+i)^N}{(1+i)^N - 1} (c_u^{\text{inv}} f_u) \quad (2)$$

Where:

$$f_u^{\text{min}} \leq f_u \leq f_u^{\text{max}} \quad \forall u \in U \quad (3)$$

$$\text{OPEX} = \left(\sum_{r \in R} c_r^+ m_r^+ + \sum_{r \in R} c_r^- m_r^- + \sum_{u \in U} c_u^{\text{op}} f_u + c_e^+ E^+ - c_e^- E^- \right) t^{\text{op}} \quad (4)$$

where, for the CAPEX, $u \in U$ are the technologies considered, such as the auxiliary furnace or the HTG. N is the lifetime or expected project lifetime in years, i is the interest rate, c_u^{inv} is the fixed investment cost of the

unit u , in €. The CAPEX is constrained by the sizing factors f_u^{min} and f_u^{max} , which ensure that the selected technology is neither oversized nor undersized, taking u into account the associated costs and material flows. Concerning the OPEX, it corresponds to the specific cost c_r^+ / c_r^- of purchasing/selling the resource $r \in R$ (in €/kg or €/kWh), multiplied by the flow of the given resource $\dot{m}_r^+ / \dot{m}_r^-$, which represents the mass flow of the resource r consumed/produced. This is added to the operating specific cost of unit u per mass flow, minus the c_e^+ / c_e^- price for purchased/sold electricity $\dot{E}^+ / \dot{E}^-$ (€/kWh). All terms are multiplied to the total operating time of the plant, t^{op} (h/y).

The OPEX of the process considers market prices, as presented in SI1.7 and SI2 (Data). The annual capacity factor is 91 %, which is equivalent to 8,000 operating hours per year, for a plant that has a feeding rate of 1 t/h of sewage sludge. The total capital investment for an HTG plant is 14.1 M€, for a 20-year operating plant, with an interest rate at 6 % [16]. The HTG plant considered in this work and by Gasafi et al. [16] both present the same characteristics, using a high-pressure pump, a reactor, a product cooler, and a gas-liquid separator.

Concerning the membrane, the CAPEX have been established according to Hao et al. [61], taking into account a cost of 10 €/ft² at installation, and 5 €/ft² to change the membrane every four years. The biogas treated has a molar concentration close to 40 % of CO₂, and the membrane system is a two-stage in series with CH₄/CO₂ selective membranes. 3.43 ft² of the membrane is required for a flow of 1 m³/h. The cost model assumed to scale linearly with the methane production capacity. Details of the market costs and technology costs can be found in SI1.8 and SI2 (Data).

3. Results and discussion

First, energy integration and carbon conversion analyses are conducted on the HTG route to ensure proper heat recovery and process efficiency. Next, the nineteen proposed routes are assessed using thermodynamic principles—specifically, through exergy analysis. A broader

analysis is then introduced, centered on two key performance indicators: environmental and economic metrics. Finally, the most promising routes according to the KPIs considered are discussed. All results are consistently expressed per FU.

3.1. Technological and thermodynamic evaluation

3.1.1. Heat and energy integration

The heating and cooling requirement of the different heat streams for the route (a), (HTG, methanator, and CH₄ membrane separation) are shown in the form of the composite curve, in Fig. 5a. The setup has a minimum heating and cooling requirement of 4,400 kW and 1,250 kW, respectively, per ton of treated sewage sludge. The grand composite curve, displaying the heat flow of the HTG, methanator and CH₄ separation membranes according to the temperature of the processes, can be found in Fig. 5b. The heating requirement is above 500 °C and can be met using an auxiliary furnace operating at 550 °C. Water is required to cool the different process streams to ambient temperature using a cooling tower. A CO₂ transcritical power cycle is also integrated, allowing the conversion of medium to high-temperature waste heat into electricity, and reducing energy imports and exergy losses of the plant.

The heating requirement exceeds the cooling requirement because the gasification process is net endothermic, absorbing more heat than it releases. This is due to the high-temperature demands of the process (>500 °C), required to ensure the complete conversion of sludge into syngas, as well as the significant energy needed to vaporize the moisture in the sludge. The heating requirement for the process is 4,400 kWh/FU, representing approximately 73.6 % of the total energy content of the methane produced, which amounts to 5,977 kWh/FU.

The heat is provided using an electrical furnace, partially powered by the CO₂ transcritical cycle. This technology maximizes heat recovery and allows for the conversion of waste heat into 188 kWh of electricity (per ton of sewage sludge treated), thereby improving the system's overall efficiency. However, the system still requires an additional electricity consumption of 414 kWh/FU, and self-consume 90 % of the

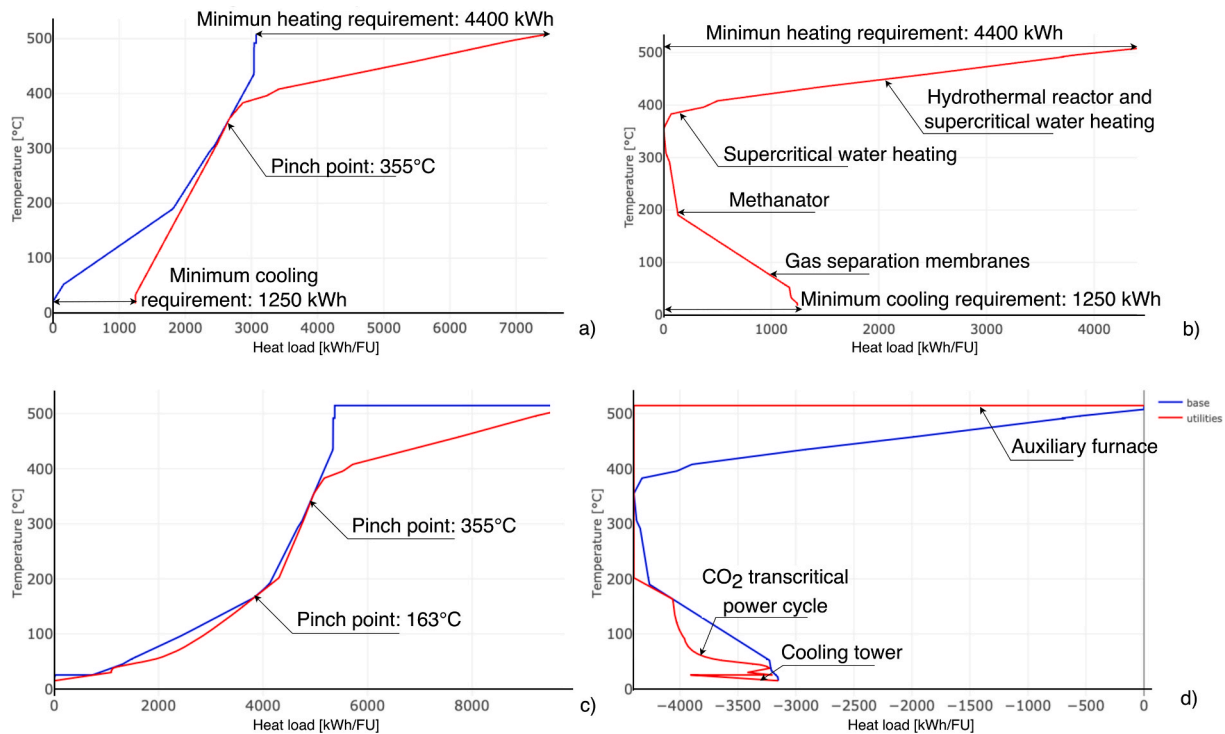


Fig. 5. (a) Composite and Grand Composite Curve (open demand) (b) of the process without heat integration, indicating the minimum heat requirement for heating and cooling. Composite (c) and Integrated Composite Curve (closed demand), including utilities (d) of the process, with heat integration and complete power recovery. Results are presented in kWh/FU.

methane produced to meet its heat demand. This high energy requirement is due to the supercritical conditions of the HTG process, as well as the compressors needed for gas separation in the membranes unit. Additionally, waste heat recovery from the methanator contributes a total of 330 kWh/FU, further reducing the system's overall energy demand.

Finally, the CO₂ transcritical cycle has proven to be the most efficient option for integrating HTG. Designed to align with the superheated CO₂ cooling profile, the transcritical cycle minimizes heat losses and ensures effective energy recovery across the entire temperature range. Conversely, a heat pump (<200 °C) cannot be installed, as it would create a plateau in the heat integration curves, disrupting the efficient recovery of heat between process streams.

3.1.2. Syngas composition and carbon conversion yield

The sewage sludge is fed to the HTG at a rate of 1 t/h. Syngas is obtained at a mass flow rate of 1.40 t/h, 1.69 t/h, and 1.80 t/h for temperatures 500 °C, 600 °C, and 700 °C respectively. The syngas composition variation according to temperature and feed concentration is shown in SI2.1. Methane is the predominant gas in the syngas produced by the gasifier, with its highest mole fraction observed at 500 °C with a water-to-biomass ratio of 40 %. The methane content in the syngas and the importance and versatility of methane as an energy vector make HTG an attractive conversion route for sewage sludge deactivation/upgrading. At 500 °C, 600 °C, and 700 °C, the total gas yield is 40 %, 47 %, and 53 %, and the carbon conversion rate is 81 %, 95 %, and 98 %, respectively.

3.1.3. Exergy efficiency of the sewage sludge conversion routes

The results of the comparative exergy analysis of the different conversion and upgrading routes for the sewage sludge are summarized in Table 3. Energy-intensive systems such as the co-SOEC and the HTG can benefit from energy integration via power generation and waste heat recovery for steam and supercritical water production. The results indicate that high-pressure and high-temperature processes such as HTG and incineration, which must evaporate the large amount of water contained in the sludge – partly using the gas produced – and do not recover all the carbon atoms in the output present a low exergy

efficiency. Conversely, processes that enable substantial methane production, such as those involving the co-SOEC, exhibit higher exergy efficiency (> 50 %) due to the chemical recovery of the exergy of the carbon atoms, which would be otherwise oxidized to CO₂ and released to the environment.

For instance, the highest exergy efficiency is achieved by coupling HTG with co-SOEC (route c), which converts most carbon atoms from the feedstock into a high exergy-content gas while facilitating partial heat recovery. In this process, the methane initially recovered from the HTG-derived syngas is directly injected into the grid. The separated CO₂ is converted to methane through co-SOEC and methanation, enhancing the exergy efficiency of standalone HTG by 50 %. The high exergy efficiency of this route is further enhanced by efficient heat integration between the HTG and the co-SOEC, both operating at elevated temperatures. Finally, enhancing methane generation using an alkaline electrolyzer (routes d and e) improves the exergy efficiency of the HTG by 20 %.

Routes based on sewage sludge digestion have the lowest power consumption; however, the methane yield remains low, resulting in exergy efficiencies of around 15 %. Similar to the HTG-based routes, adding electrolyzers and a methanation system, which facilitate the conversion of carbon atoms to more methane, yields additional valuable products and raises the process overall exergy efficiency by 15 %. Finally, although incineration produces combined heat and power (CHP), the substantial energy requirement to evaporate water from the feedstock – mainly due to its high moisture content – keeps the exergy efficiency at 0 %. The implementation of flue gas cleaning with membrane separations to recover CO₂ and convert it into methane achieves an exergy efficiency gain of nearly 60 %, due to the recovery and valorization of carbon atoms. This gain is plausible when considering the integration of a 50 % efficient electrolyzer and the incinerator's contribution of sufficient heat to offset part of the power consumption.

Considering routes that are HTG-based (a and b, c and e, l and m), the results show that carbon capture with mineralization has a lower exergy efficiency compared to the standalone waste treatment technologies, even considering the by-products (e. g. rocks produced) as useful exergetic content in output, Equation (1). It can be explained by the definition of exergy efficiency which does not consider the CO₂ emitted in the waste treatment configurations as an environmental burden. To internalize the GHG emissions in the exergy efficiency calculation and encourage the accountability for carbon dioxide emissions avoidance, a distinct reference environment is used for exergy calculations.

The implementation of the proposed reference environment leads to a reduction in exergy efficiency for routes emitting CO₂ into the atmosphere. The most significant decrease, 12.6 %, is observed in the HTG with alkaline electrolyser route (d). This is followed by the HTG-only route (a), which has a reduction of 7.8 %. The control volumes of the reference environments (conventional and the one proposed in this work) are illustrated in SI2.2. The most substantial reductions are occurring in routes characterized by higher initial efficiencies or greater CO₂ emissions.

3.2. Life cycle assessment and economic analysis of the routes

3.2.1. Life cycle assessment of the conversion routes

For clarity purposes, the LCA of five of the nineteen routes studied (a: HTG, b: HTG and mineralisation, c: HTG and co-SOEC, f: AD, and q: incineration) will be presented here, and in Fig. 6. Results for all the routes and for the other areas of protection can be found in SI2.3. The processes used from the Ecoinvent database can be found in the SI2 (Data).

Regarding route (a), the carbon footprint impact score is relatively low, ranging from 15 kgCO₂-eq/FU (for 0 % fossil methane avoidance) to −32 kgCO₂-eq/FU (100 % fossil methane avoidance). This is due to two main factors: the process consumes almost all the methane produced to self-sustain, and the impact of transformation technologies remains

Table 3
Conversion routes analyzed with exergy efficiency of the routes.

Name	Conversion Route	Exergy Efficiency (%)	Exergy Efficiency with Proposed RE (%)
a	HTG	10.7	2.9
b	HTG, mineralisation	2.1	2.1
c	HTG, co-SOEC	62.2	62.2
d	HTG, alkaline electrolyser	30.4	17.8
e	HTG, alkaline electrolyser, mineralisation	29.8	29.8
f	Digester	15.8	13.6
g	Digester, mineralisation	16.1	16.1
h	Digester, co-SOEC	25.7	25.7
i	Digester, HTG	4.3	4.3
j	Digester, HTG, mineralisation	5.7	5.7
k	Digester, HTG, co-SOEC	59.5	59.5
l	Digester, HTG, alkaline electrolyser	33.5	33.5
m	Digester, HTG, alkaline electrolyser, mineralisation	33.5	33.5
n	Digester, Incineration	9.7	2.7
o	Digester, Incineration CC mineralisation	17.6	17.6
p	Digester, Incineration, CC, co-SOEC	59.8	52.0
q	Incineration	0.0	0.0
r	Incineration, CC, mineralisation	4.9	4.0
s	Incineration, CC, co-SOEC	59.1	55.3

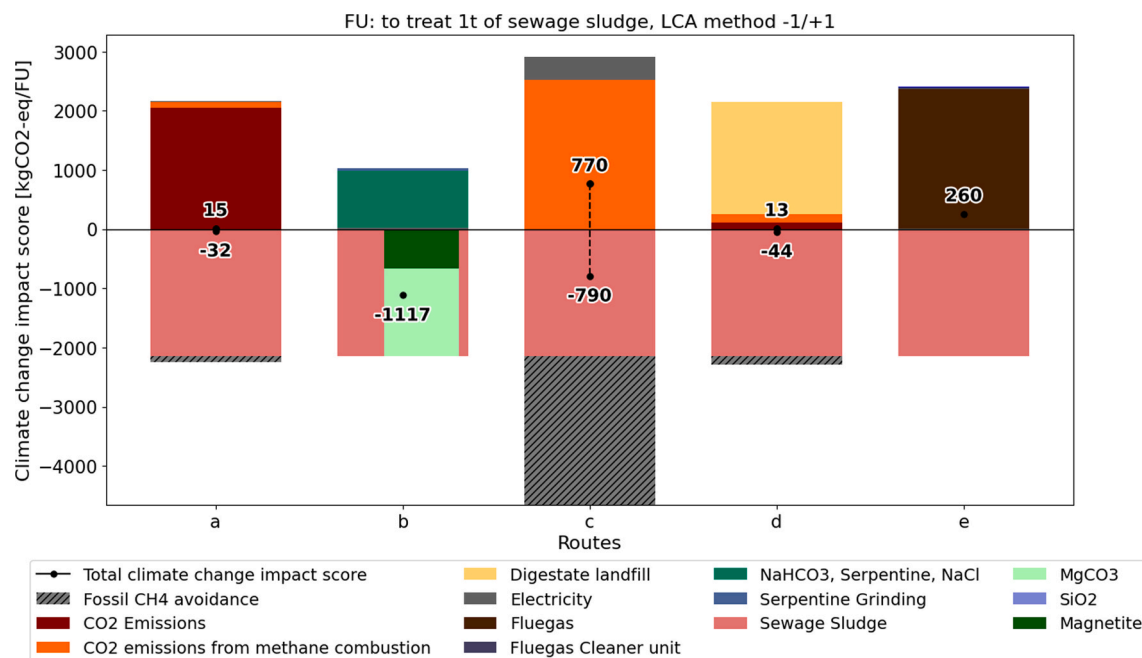


Fig. 6. CO₂ emissions assessment, in kgCO₂-eq for the five route considered. The climate change impact score varies according to the importance of methane for society, with 100 % to 0 % avoidance considered. The routes are decomposed as follows: (a) HTG + direct CO₂ emissions; (b) HTG + mineralisation; (c) HTG + co-SOEC + methanator; (f) AD; (q) Incineration.

low. Moreover, this route performs relatively well compared to the others considering all the endpoint categories, by consistently falling within the lower or very lower ranges (see SI2.3). Concerning route (b), all the methane produced is used to power the HTG and the CO₂ mineralisation processes. The biogenic CO₂ in the sludge is mineralised and permanently stored as MgCO₃, resulting in a net-negative impact of −1,117 kgCO₂-eq/FU. The ecosystem quality category shows the best performance for this route, due to the large amount of CO₂ captured, which helps mitigate marine acidification. However, the rocks used to mineralise the CO₂ leads to a shift in environmental impacts towards the endpoint categories of water scarcity and fossil and nuclear energy use (cf. SI2.3). Route (c) has the widest range of carbon footprint impact, due to the large quantity of methane produced (12.56 MW/FU). Considering a scenario where methane as 100 % avoidance in the mix, the impact is −790 kgCO₂-eq/FU. If methane does not substitute fossil fuels in the energy system, the impact becomes 770 kgCO₂-eq/FU. This route shows the highest sensitivity to the assumptions made about the future energy system, resulting in significant variability in its environmental performance. In addition, it is the most dependent on the carbon footprint of the energy system, with the actual mix contributing to 390 kgCO₂-eq/FU. More broadly, the high energy consumption of this route, due to the combination of HTG and co-SOEC, makes this route – and all those based on co-SOEC – the least attractive in terms of fossil and nuclear energy use, ecosystem quality, and water scarcity footprint endpoint categories. Route (f) has a carbon footprint impact score ranging from 13 to −44 kgCO₂-eq/FU. The impact shows minimal variation due to the low amount of methane produced through digestion. With low energy consumption, this route is also the least sensitive to the energy mix's impacts. Although it has the lowest impacts across most endpoint categories, it exhibits the highest impact score in terms of residual human health damage. Finally, route (q), based purely on incineration is the least favorable option in terms of carbon footprint, with 260 kgCO₂-eq/FU. This is due to the energy required for incineration, ash treatment, and the presence of impurities in combustion gases. Indeed, this process is energetically unfavorable, as an additional electricity input is required to run the incinerator and evaporate the water contained in the feedstock. This route performs low to moderately

compared to others across the other four endpoint categories.

In conclusion, considering only the carbon footprint indicator, mineralisation is the best process to handle CO₂, particularly when the routes allow for a high conversion of the sludge in CO₂ (i.e., without electrolyzers). Indeed, mineralisation-based routes are the only ones with consistently negative impacts, due to co-products and permanent carbon storage from the sludge. However, the full LCA reveals a shift in the other endpoint categories (except for ecosystem quality), thereby mitigating the advantage of this route over the others. The routes that produce the most methane (based on electrolysis) are those with the widest and most variable impact range if methane becomes irrelevant in the future energy mix. While they have the highest negative impacts under 100 % avoidance, they also have a high positive impact (from 550 to 980 kgCO₂-eq/FU) if no avoidance is considered. Though efficient in energy markets where additional methane produced replaces fossil methane, they may not remain so in different economic or societal contexts. Even with proper heat integration, the energy intensity of these routes results in the highest impact scores in the water scarcity, ecosystem quality, and fossil and nuclear energy use categories. AD-based routes consume less energy than the thermochemical ones (HTG, incineration), and show lower carbon footprint impact score (between −87 to +74 kgCO₂-eq/FU for those not using electrolyzers or mineralisation). They are considered robust over potential economic and societal evolution due to the slight fluctuation of the carbon footprint impact score. However, the very high impact of the digestate (around 4,200 μDALYs/FU), when it is neither incinerated nor gasified, in the human health damage category undermines the viability of routes f, g, and h. Finally, the incineration-based route has a high carbon footprint impact score compared to the other routes, mainly due to impurities in the flue gases and the energy demand of the process. It also demonstrates low to moderate performance compared to other routes across the other four endpoint categories.

3.2.2. Economic analysis

The CAPEX is broken down per technology and illustrated in Fig. 7. Routes integrating co-SOEC are the most expensive, with a CAPEX exceeding 2,500 €/FU. Digester-based routes without valorisation of the

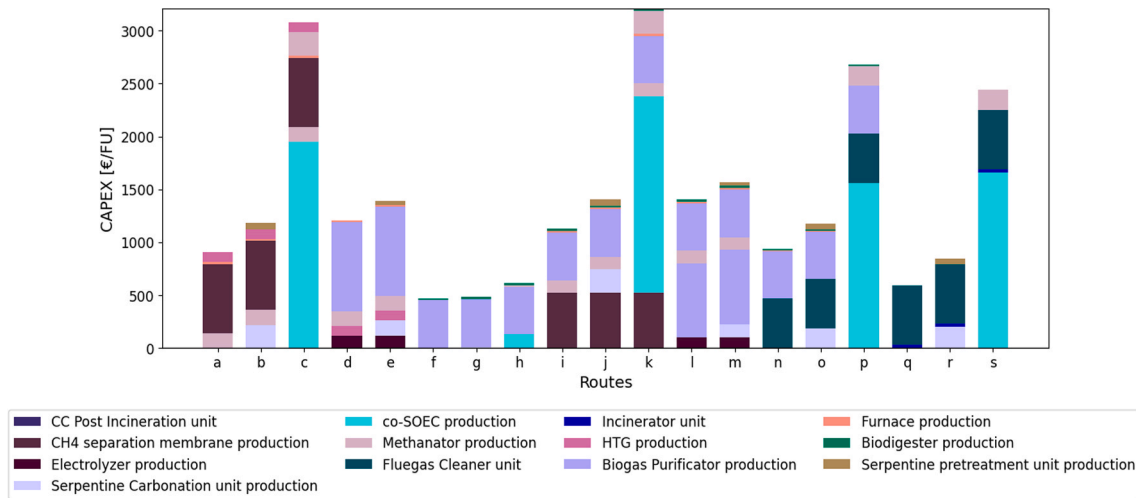


Fig. 7. Breakdown of the CAPEX of the different routes, in €/FU. Routes can be found in Table 1.

digestate (*f* to *h*) have the lowest CAPEX range (470 to 613 €/FU), however those solutions are not legally feasible in countries like Switzerland [8]. The business-as-usual route *q*, incineration alone, is the pathway with the lowest CAPEX after the AD-based ones, at 591 €/FU. The majority of its costs is from the flue gas cleaning unit. Each route underlines the high share in the investment costs for the treatment of gases technologies (either CH_4 separation membranes, biogas purification and flue gas cleaning from incineration), representing nearly 80 % of the investment costs for routes without any co-electrolysis. Finally, integrating mineralisation to the base processes adds from 10 % (for digestion and alkaline electrolyzer, routes *l* and *m*) to 40 % investment costs (for incineration-based processes *q* and *r*). In most scenarios, the integration of mineralisation leads to CAPEX increases of up to 20 %.

The OPEX are broken down per resources consumed or produced in Fig. 8. Despite the fact that value-added methane is produced in most of the configurations, this advantage is offset by both the extensive electricity consumption and self-consumption of methane (*a-e*; *k-m*; *p*, and *s*). In that sense, HTG with an co-SOEC (*c*) is showing the highest OPEX, with 3,139 €/FU. The amount of minerals produced by route (*b*), HTG with mineralization, are responsible for a negative OPEX at −152 €/FU. Digester-based routes (*f*, *g*) exhibit low OPEX (3–12 €/FU), due to valorisation of the methane and the low energy-intensity of the process's

routes. Incineration alone (*q*) also present a relatively low OPEX of 77 €/FU. Overall, the OPEX is very dependent on market prices such as electricity and methane. Coupling high carbon tax, renewable cheap electricity and high price for methane on the market tends to support low-emission conversion routes.

The CAPEX-OPEX trade-offs are illustrated in Fig. 9. The solutions are clearly divided into three main groups: high CAPEX and OPEX configurations, integrating co-SOEC (*s*, *p*, *c*, and *k*), medium CAPEX and OPEX, integrating alkaline electrolyzer (*d*, *e*, *l*, and *m*), and the other remaining routes, with low OPEX (<500 €/FU), and low to moderate CAPEX (from 450 to 1,500 €/FU). Co-SOEC-based configurations are energy-intensive, driven by the requirements of the gasification and incineration reactors with significant heat demands. The least favorable solutions combine HTG with co-SOEC, resulting in high OPEX due to their energy consumption and elevated CAPEX due to their technological complexity. Conversely, widely adopted solutions, such as incineration-based systems, or those operating at low temperatures and on a smaller scale, i.e. AD-based routes, hold a privileged position within the studied solution space, as shown in Fig. 9.

Other studies have found that to address the uncertainty of the market prices for the commodities can reveal important aspects regarding the likelihood of loss of risky projects [35]. By imposing

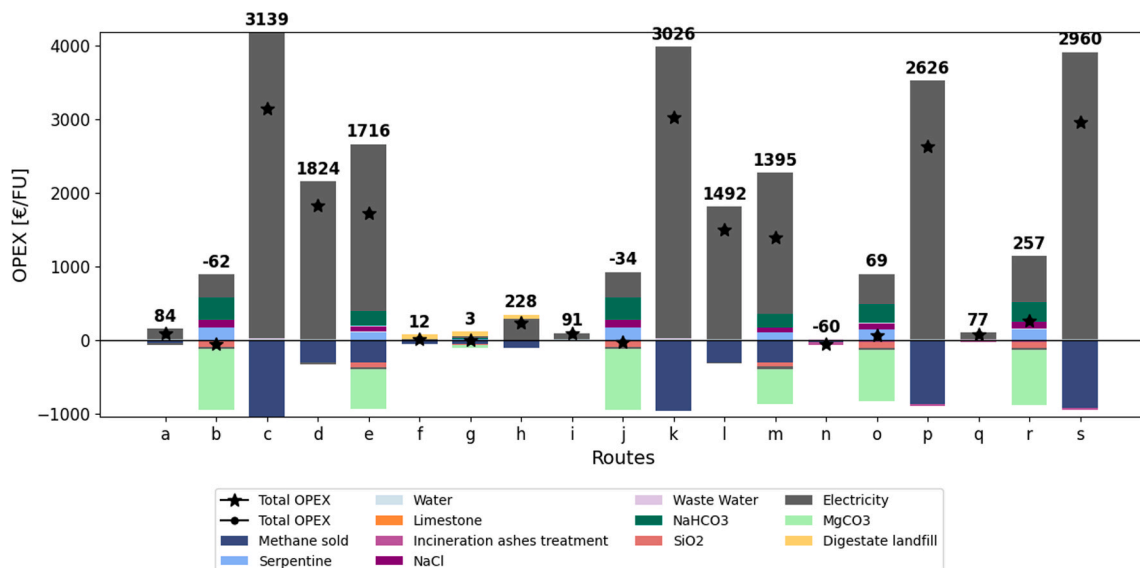


Fig. 8. Breakdown of the OPEX of the different routes, in €/FU. The black stars represents the total OPEX. Routes can be found in Table 1.

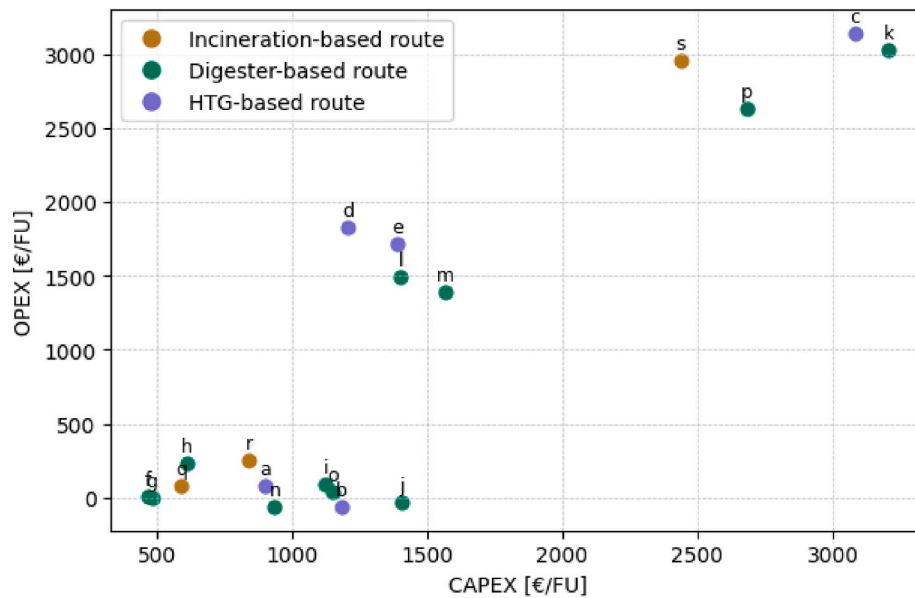


Fig. 9. CAPEX and OPEX solution space of the conversion routes studied. Results are per ton of sludge treated. Routes can be found in [Table 1](#).

stricter carbon taxes and improving technological readiness, financial risks could be reduced, enabling decision-makers to implement biorefineries and explore new business opportunities aimed at decarbonizing essential commodities, like methane. In a future study, the uncertainty of the market prices for the commodities will also be conducted to compare the routes evaluated on this study, as it has been suggested in similar works [35,64].

3.3. Cross-analysis of the performance indicators

3.3.1. Cost-impact analysis

Fig. 10 provides a cost-carbon footprint impact score analysis, illustrating the trade-offs over those two KPIs between the routes. Three solution clusters emerge, depending on the integration technologies

used: low-cost, low and with small impact score variation solutions without CO₂ management (direct biogenic CO₂ emissions) or with mineralization (permanent storage of biogenic CO₂); medium-cost, low-impact solutions with partial upgrading of the syngas using alkaline electrolysis; and high-cost, highly varying impact solutions maximizing methane production using co-SOEC.

The first cluster includes low-cost, low-impact routes, ranging from 500 € to 1,500 €/FU. Digester and HTG-based options without carbon management exhibit impacts centered around zero, with little variation under the assumption of methane avoidance. The total costs for these solutions remain below 1,000 €/FU. Among these, pathways incorporating mineralization stand out, particularly routes (b) and (j), which show slightly higher costs (approximately 1,100 €/FU) and a negative impact of around $-1,100$ kg CO₂-eq/FU. However, the rocks used to

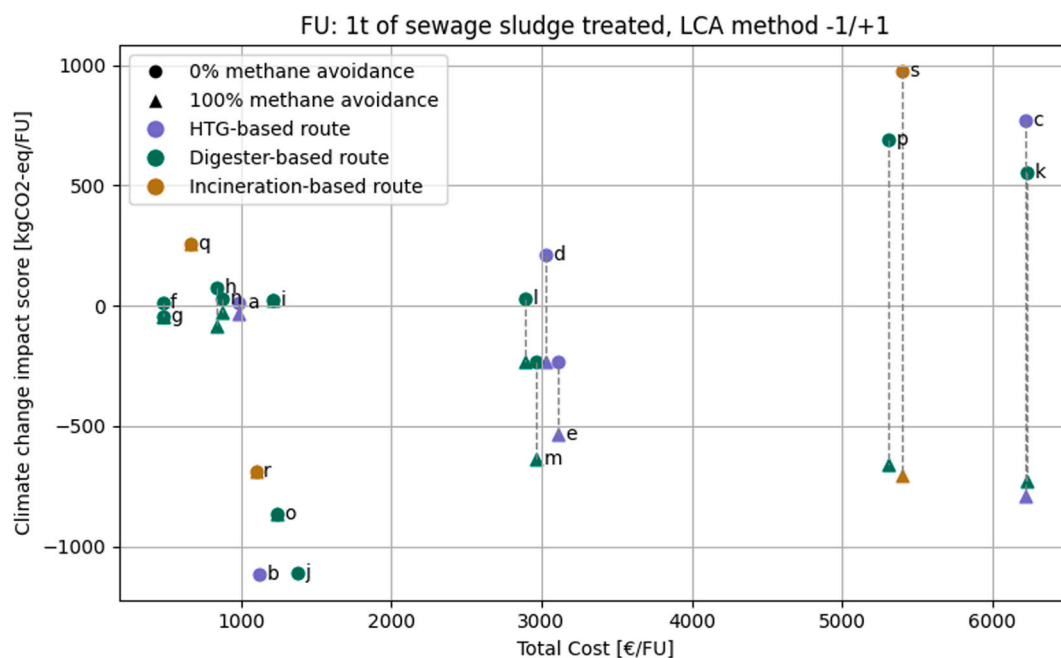


Fig. 10. Total cost *versus* carbon footprint impact score solution space of the studied routes, considering 0% and 100% methane avoidance. Routes can be found in Table 1.

mineralise the CO₂ leads to a shift in environmental impacts towards the endpoint categories of water scarcity and fossil and nuclear energy use (cf. SI2.3). AD-based technologies with management of the digestate via HTG or incineration present the best tradeoff between total cost and carbon footprint impact. Routes (f) and (g) seem optimal in terms of cost and carbon footprint impact, but rely on open-air digestate degradation. This leads to the highest impact in terms of human health damage (cf. SI2.3). Moreover, in countries like Switzerland, using sludge-derived digestate as fertilizer is forbidden, rendering such routes inapplicable [8]. HTG-based route (a) is more expensive than routes based solely on digestion, due to the energy required to evaporate sludge moisture and the costs of flue gas treatment. However, it is the only one to minimize all endpoint impact categories across the LCA. Finally, the treatment technology currently most used in Switzerland, route (q) – incineration without carbon recovery – is one of the lowest-cost solutions, and has an impact score of 260 kgCO₂-eq/FU. While the climate change impact remains high, the route demonstrates low to moderate performance across the other four endpoint categories.

The second cluster covers medium-cost, low-impact solutions (routes l, m, d, e), which maximize syngas production via gasification (of sludge or digestate), coupled with alkaline electrolysis to improve the methane yield. Under the actual energy mix (therefore assuming 100 % methane avoidance), these routes show strong negative impacts (–233 to –637 kgCO₂-eq/FU), with average costs around 3,000 €/FT. The impact score of this cluster varies significantly depending on the methane avoidance hypothesis, going from –637 to + 212 kgCO₂-eq/FU. Routes (e) and (m), which integrate CO₂ mineralization, remain net-negative even if methane is no longer needed in the future energy mix—making it the most robust solution facing uncertain societal changes in this cluster. However, the high energy demand and the use of rocks for mineralization result in moderate to high impacts across the areas of protection, particularly for routes (m) and (e), in the categories of water scarcity, human health damage, and fossil and nuclear energy use.

The third cluster involves high-cost routes (>5,000 €/FU), maximizing methane production by integrating co-SOEC and methanation to the main processes (incineration, AD, HTG). These routes have highly variable impacts scores depending on the methane avoidance assumption, ranging from + 977 kgCO₂-eq/FU (incineration and co-SOEC, with 0 % avoidance) to –790 kgCO₂-eq/FU (HTG and co-SOEC, with 100 % avoidance). Therefore, assuming that the current energy will not greatly change in the upcoming decades, these routes have the advantage of offering both a low impact score and an energy product that is widely used in the energy mix, such as methane. The best trade-offs in this cluster appear to be routes (s) and (p) (incineration-based and AD-based, respectively), which are less expensive than routes (c) and (k) that rely on gasification. Considering the other endpoint categories (cf. SI2.3), these routes exhibit the highest impacts in terms of water scarcity, fossil and nuclear energy use, and ecosystem quality. However, they remain relatively favorable compared to the others regarding the human health impact.

3.3.2. Discussion

The analysis of various KPIs revealed three distinct clusters of solutions across the exergy efficiency, the carbon footprint impact score, and the total costs. First, routes with low energy consumption and limited methane production—typically based on digesters or systems without electrolyzers—tend to exhibit low exergy efficiency and low to medium costs. They also show near-zero climate change impact scores with limited influence from the variation of the evolving energy mix. Only the solutions integrating mineralisation exhibit a net negative and fully stable climate change impact. This cluster of solutions has been considered robust over the years, as their performance in terms of both cost and impact has only been marginally influenced by the evolution of the energy mix over time.

The second cluster of solutions, which integrates an alkaline electrolyzer, shows an exergy efficiency of around 30 % and a variable

climate impact ranging from approximately – 640 to + 250 kg CO₂-eq/FU, depending on the methane avoidance assumption rate. These systems show a moderate total cost (around 3,000 €/FU), and their climate change impact scores are relatively less variable and less sensitive to the evolution of the energy mix than those based on co-SOEC. However, the high energy demand and the use of rocks for mineralization result in moderate to high impacts compared to the other routes across the areas of protection, particularly for routes (m) and (e), in the categories of water scarcity, human health damage, and fossil and nuclear energy use.

The third cluster includes routes integrating co-SOEC, allowing to the achievement of high exergy efficiencies (around 60 %) due to the valorisation of most of the carbon atoms contained in the feedstock in methane. However, they are associated with high CAPEX and OPEX due to their technological complexity and substantial energy requirements. These solutions demonstrate a favorable carbon footprint impact score under the current energy mix (around – 750 kg CO₂-eq/FU), assuming the methane effectively substitutes the fossil-based one. However, this impact score is also the most sensitive to variations in the fossil methane avoidance rate. Moreover, co-SOEC-based routes exhibit a shift in impacts across the other endpoint categories, presenting the worst performance in water scarcity, ecosystem quality, and fossil and nuclear energy use, due to their high energy demand.

Results suggests that an increase in exergy efficiency is generally associated with a decrease in the climate change impact score for these technologies when considering 100 % methane avoidance in the impact evaluation. This correlation has also been identified by other authors, such as Domingos et al [35]. In fact, exergy-efficient pathways (>50 %, using co-SOEC) tend to valorize most of the carbon-based atoms by converting them into methane, thereby avoiding GHG emissions and the associated environmental impacts.

4. Conclusions

The thermo-economic evaluation and the life cycle impact assessment of nineteen different routes to treat sewage sludge have been compared. The evaluation of KPIs highlights three distinct groups of sewage sludge treatment solutions, each characterized by trade-offs between efficiency, costs, and environmental impacts. A direct correlation has been observed between the exergy performance of the technologies, their total cost, and their carbon footprint impact score.

Overall, the HTG treatment process coupled with CO₂ mineralization shows the most promising long-term potential for achieving net-zero, and even net-negative, life cycle global warming potential per ton of treated sewage sludge. Meanwhile, HTG alone exhibits the best compromise across the five endpoint categories in the LCA, with a total cost of 990 €/FU.

Routes based on digestion exhibit low costs and near-zero carbon footprints, although at the expense of increased human health impacts if the digestate is neither gasified nor incinerated. Solutions integrating CO₂ mineralization to complement gasification of the sludge or the digestate achieve a net-negative carbon footprint (around –1,100 kg CO₂-eq/FU) at a cost of approximately 1,200 €/FU. However, the use of rocks for mineralization results in less favorable impact scores compared to other routes in the categories of fossil and nuclear energy use and water scarcity.

Solutions incorporating alkaline electrolyzers offer moderate exergy efficiencies (around 30 %) and moderate impacts across all categories compared to the others, although at higher costs (around 3,000 €/FU). In contrast, technologies based on co-SOEC achieve the highest exergy efficiency – around 60 %. However, their carbon footprint performance is highly sensitive to the methane avoidance rate considered, and they involve substantial trade-offs in other environmental categories due to their high energy demand. These routes maximize methane production and appear promising when methane can be fully substituted (100 %) into the energy mix. Despite proper heat integration, the high costs (>5,000 €/FU) and energy demands currently limit their feasibility.

CRediT authorship contribution statement

Soline Corre: Writing – original draft, Software, Methodology, Formal analysis, Conceptualization. **Meire Ellen Gorete Ribeiro Domingos:** Writing – review & editing, Methodology, Formal analysis, Conceptualization. **Daniel Florez-Orrego:** Writing – review & editing, Methodology, Formal analysis, Conceptualization. **Gabriel Magnaval:** Writing – review & editing, Methodology. **Manuele Margni:** Writing – review & editing, Supervision, Formal analysis. **François Maréchal:** Writing – review & editing, Supervision, Formal analysis.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.enconman.2025.120015>.

Data availability

Data will be made available on request.

Life cycle assessment and techno-economic comparison of methane production routes from sewage sludge: incineration vs. hydrothermal gasification and anaerobic digestion (Reference data) (Zenodo)

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