

Article



Techno-Economic and Environmental Analysis of a Sewage Sludge Alternative Treatment Combining Chemical Looping Combustion and a Power-to-Methane System

Piero Bareschino ^{1,2,*}, Roberto Chirone ^{3,4,*}, Andrea Paulillo ^{3,5}, Claudio Tregambi ¹, Massimo Urciuolo ², Francesco Pepe ¹ and Erasmo Mancusi ¹

- ¹ Dipartimento di Ingegneria, Università degli Studi del Sannio, P.zza Roma 21, 82100 Benevento, Italy; claudio.tregambi@unisannio.it (C.T.); francesco.pepe@unisannio.it (F.P.); erasmo.mancusi@unisannio.it (E.M.)
- ² Istituto di Scienze e Tecnologie per l'Energia e la Mobilità Sostenibili, Consiglio Nazionale delle Ricerche, P.le V. Tecchio 80, 80125 Napoli, Italy; massimo.urciuolo@stems.cnr.it
- ³ eLoop s.r.l., V.le A. Gramsci 17/B, 80122 Napoli, Italy; and reapaulillo@eloop.consulting
- ⁴ Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale, Università degli Studi di Napoli Federico II, P.le V. Tecchio 80, 80125 Napoli, Italy
- ⁵ Department of Chemical Engineering, University College London, Torrington Place, London WC1 E7JE, UK
- * Correspondence: piero.bareschino@unisannio.it (P.B.); roberto.chirone@unina.it (R.C.)

Abstract: An innovative process layout for sludge waste management based on chemical looping combustion and flue gas methanation is analyzed in this work. The technical performance of the system was assessed by considering that the flue gas is first purified and then mixed with a pure hydrogen stream sourced from an array of electrolysis cells to produce methane. The life cycle assessment (LCA) and life cycle cost (LCC) methodologies were applied to quantify the environmental and economic performances of the proposed process, and a hotspot analysis was carried out to recognize its most critical steps. The proposed system was then compared with a reference system that includes both the conventional waste management pathways for the Italian context and methane production. Finally, to account for the variability in the future economic climate, the effects of changes in landfill storage costs on sewage end-of-life costs for both the proposed and reference systems were evaluated. With respect to 1 kg/h of sewage sludge with 10%wt of humidity, the analysis shows that the proposed system (i) reduces landfill wastes by about 68%, (ii) has an end-of-life cost of 1.75 EUR \times kg⁻¹, and (iii) is environmentally preferable to conventional sewage sludge treatment technologies with respect to several impact categories.

Keywords: sewage sludge disposal; CO₂ capture and utilization; methanation; climate change impact; sensitivity analysis

1. Introduction

The disposal of sewage sludge, one of the solid by-products of wastewater treatment, has become a prerogative to achieve the sustainable development goals defined by the United Nations. Significant quantities of wastewater sludge are generated globally every year, varying from approximately 5 g to 90 g of dry material per capita per day [1]. As reported in the literature [2], global dry sewage sludge production reached 45 million tons in 2017, and forecasting suggests that sewage sludge production will increase in the next few years due to both the rapid growth of the population and the fast rate of urbanization.

The disposal and reuse of sewage sludge are governed by different regulations. The European Directive on Sewage Sludge 86/278/EEC (1986), which encourages reuse in agriculture, requires adequate processes to be conducted to reduce the fermentability of sludge and the consequent risks to human health. Directive 91/271/EEC (1991) states that the sludge from wastewater treatment should be reused wherever possible. Furthermore, Directive 99/31/EC (1999) limits the amount of sewage sludge and other organic waste that



Citation: Bareschino, P.; Chirone, R.; Paulillo, A.; Tregambi, C.; Urciuolo, M.; Pepe, F.; Mancusi, E. Techno-Economic and Environmental Analysis of a Sewage Sludge Alternative Treatment Combining Chemical Looping Combustion and a Power-to-Methane System. *Energies* **2024**, *17*, 901. https://doi.org/ 10.3390/en17040901

Academic Editors: Adam Smoliński and Pouya Ifaei

Received: 5 January 2024 Revised: 10 February 2024 Accepted: 13 February 2024 Published: 15 February 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). can be disposed of in landfills. Within most European countries, the most widely adopted approaches for sewage sludge disposal are land-spreading (after thickening processes) or compost production (after mechanical dewatering) for agricultural use, while deposition in landfills and thermal treatments are only considered as a last option [3].

In Italy, sludge production varies broadly between the southern (about 10 g of dry matter per capita per day) and northern (about 80 g of dry matter per capita per day) regions [4], suggesting that wastewater treatment services are not evenly spread within the country. A similar variability across the country can also be observed for sludge disposal: while in Northern Italy, land-spreading and composting plants are the final destinations of about 45% of total produced sludge (followed by thermal treatments and landfilling, this latter corresponding to about 23% of total production), in the southern regions, landfilling is, by far, the most common [5]. With respect to the Campania region, the disposal of sewage sludge cannot be performed within the region itself due to strict environmental limitations introduced by local laws, and hence, the whole production is sent for treatment outside the region—mostly to landfills [4,6]—increasing both environmental impacts and costs of sludge disposal. As a consequence, alternative disposal routes are actively being examined, spanning from biological to thermochemical treatments with innovative water removing technologies [7–9].

The thermochemical treatment of sludge—such as combustion, pyrolysis, and gasification—represents promising technologies with short reaction times (from seconds to minutes) and high conversion efficiency (less than 20% of unconverted organic constituents at the end of the process) in comparison with biochemical conversion routes such as anaerobic digestion (reaction time from 7 days to 5 weeks; about 40–70% of unconverted organic constituents). Among the above-mentioned thermal treatments, chemical looping systems are becoming very interesting since these technologies are more efficient and cost-effective than the commercially available ones. Chemical looping relies on the cyclic exposure of a solid metal oxide—the oxygen carrier (OC)—to two distinct reactive environments to enable the selective transport of a chemical, such as oxygen or carbon dioxide, between otherwise uncoupled devices or environments. Chemical looping combustion with oxygen uncoupling (CLOU) systems is the most promising technology for sludge combustion, with inherent CO₂ sequestration and almost zero NO_x and dioxin emissions [10].

This work analyzes the techno-economic and environmental performances of an innovative process layout proposed and detailed in a previous work [11]. To the best of the authors' knowledge, no such analysis has been previously carried out in the literature. The technical performance is assessed in terms of the residual amount of sewage sludge treated to be sent to landfills, the thermal power generated, the number of electrolysis cells required to produce hydrogen, and methane production. The economic performance is assessed as the specific sewage end-of-life cost. The environmental performance is expressed in terms of impacts in numerous environmental categories (e.g., climate change, eutrophication, toxicity, etc.) using the life cycle assessment methodology (LCA, e.g., [12]).

2. Materials and Methods

The proposed process layout [11] is a two-step Power-to-Methane (PtM) that integrates chemical looping combustion with oxygen uncoupling, hydrogen production through water electrolysis by polymer electrolyte membranes (PEM), and a methanation unit (MU). The core of the proposed PtM system is the CLOU unit, consisting of multiple interconnected fluidized equipped with a two-stage fuel reactor and a riser used as the air reactor [11,13]. For the methanation unit, several reactor concepts and layouts have been proposed in the literature, ranging from multiple adiabatic packed beds with inter-cooling and optional product recycling [14,15] to micro-structured reactors consisting of multiple micro-tubes, filled by a catalyst and surrounded by a coolant fluid [16]. Recently, sorption-enhanced methanation with the use of zeolites for water adsorption has shown that high methane purity can be achieved for quite long running times [17]. The methanation unit comprises a series of adiabatic fixed-bed reactors with inter-cooling between each stage where CO₂

reacts with H₂ over Ni supported on an alumina catalyst. Two real sewage sludges from the Campania region, chemically characterized in a previous paper [9], are considered fuel to be fed to the CLOU system. Finally, data from commercially available polymer electrolyte membrane cell (PEM) units were considered [18]. A comprehensive description of the equations, assumptions, and limitations of the adopted mathematical model was reported in previously published papers [11] and in the Supplementary Materials, as well as the operating conditions for the system under analysis (Table S7).

2.1. Economic Analysis

The economic performance of the proposed system was compared with that of a real scenario for treating sewage sludge in the Campania region, consisting of mechanical dewatering up to 18% of sewage sludge dry matter content, transportation (\sim 300 km), and final disposal in landfill.

The economic index used in this analysis is the specific (i.e., per kg of sludge) end-oflife cost (C_{EoL}), defined as the following:

$$C_{EoL} = 1.02 \cdot TCR_s + TCL_s - R_s \tag{1}$$

where TCR_s is the specific annualized total capital requirement, TCL_s is the specific total cost for landfilling, and Rs is the specific revenue.

To assess TCR_s , the method presented by Peters [19] was applied, except for the estimation of fuel and air reactor costs, where the methodology developed by Lyngfelt and Leckner (2015) [20] was used instead. To perform annualization, it was considered that the project interest rate and amortization years were equal to 8.75% and 20, respectively, for both the reference and the proposed systems. According to the literature [21], specific fixed (FOM) and variable (VOM) operating and maintenance costs were assumed to be 2% of TCR_s for both systems.

To assess TCL_s , data from the Italian Ministry of Sustainable Infrastructures and Mobility [22] was considered to estimate the specific transport costs, whereas a specific landfill disposal cost equal to 0.5 EUR/kg was assumed. The additional parameter values (base case) needed to perform the economic evaluation are reported in the Supplementary Material (Table S8). Furthermore, a sensitivity analysis was carried out by varying the specific cost of landfill disposal.

2.2. Life Cycle Assessment

The life cycle assessment methodology was first used to identify the main sources of environmental impacts (i.e., "hot-spots") associated with the proposed layout for sewage sludge treatment and to evaluate its life cycle environmental performance compared with the conventional treatment technology used in the Campania region.

The main function of the proposed system is to treat sewage sludge, and the functional unit corresponds to the treatment of 1 kg/h of sewage sludge with a water content of 10%. However, the system also co-produces thermal energy and methane. These additional functions are accounted for by crediting the "avoided" environmental impacts [23] associated with the functions of thermal energy and methane production. In particular, the energy surplus generated from the CLOU system and that generated from burning the methane stream in a conventional boiler with 90% efficiency are assumed to displace the most common source of thermal energy, i.e., natural gas [24]. The choice of burning the methane stream in a conventional boiler instead of direct injection into the gas grid reflects the fact that the hydrogen content in the produced methane stream is close to the limit for its direct injection into the existing infrastructure (around 10%). Furthermore, it was assumed that surplus electricity from onshore wind farms satisfied the required electricity for producing hydrogen from water electrolysis.

The conventional technology for comparative purposes reproduces a real scenario for treating sewage sludge in the Campania region, which consists of mechanical dewatering, transportation, and landfilling; the latter includes gas utilization for electricity production.

It was assumed that the system is credited for the avoided impacts of electricity generated, assuming that it displaces the electricity produced according to the Italian grid mix.

The system boundaries, which are schematically illustrated in Figure 1, are divided into the foreground and background [25], where the foreground is defined as the "processes whose selection or mode of operation is affected directly by decisions based on the study" [25]. In this study, it includes mechanical and thermal drying, the CLOU, the PEM, and methanation units. On the other hand, the background includes all other processes that interact with the foreground; notably, Figure 1 reports as "black boxes" the processes of transportation and disposal of residual of sewage sludge, production, transportation, and disposal of oxygen carrier (OC) used in the CLOU unit and of nickel catalyst required by the methanation unit, production of electricity and chemicals used in the drying step, polymer electrolyte membrane production used for the PEM unit, and generation and distribution of electricity from wind power. Furthermore, the facilitied construction is considered for all units in the foreground and background system, even if it is not reported in the diagram, while decommissioning the facilities in the foreground system is not considered.



Figure 1. Schematic representation of the system boundaries considered, including the reference system.

The life cycle inventory is based on the mathematical model results [11] and literature data. The results of the mathematical model describe the operation of the CLOU reactor and of the methanation unit in terms of mass and energy inputs/outputs. In particular, it was assumed that mechanical dewatering would require an electric consumption of 41.16 kWh per ton of dry matter and 0.72 kWh per kilogram of water evaporated during thermal drying [8]. The HP-PEM unit is based on literature data [18] and on the ecoinvent Database[®], version 3.6 cut-off model [26]. The facilities' construction phase data are taken from ecoinvent for the CLOU reactor (the working assumption is that construction of the CLOU reactor for electricity production is equivalent to that of a coal power plant), on ecoinvent and literature [18] for the HP-PEM cell, and on [27,28] for the methanation reactor using a nickel catalyst. The electric requirement for the PEM is assumed to correspond to 46.6 kWh/kgH₂ [18]. It is considered 50% copper oxide and 50% zirconia as a composition

of the oxygen carrier, and the data on its production are on a laboratory scale [29]. It is assumed that 5% of OC per hour needs to be replaced. The life cycle inventory data for the remaining activities in the background system and for the conventional end-of-life pathway of sewage sludge, i.e., mechanical dewatering and landfill, are obtained from the ecoinvent database. Finally, an average distance of 300 km is considered for transporting the sludge to the landfill site (data provided from a real sewage treatment plant located in Campania). Table 1 reports the complete inventory used for this work.

Table 1. Inventory analysis.

			Sewage Sludge 1	Sewage Sludge 2	ConventionalEnd of Life			
			1 kg–10%wt water content into CLOU system					
Conditionir	ng and mechanical drying							
	Sewage sludge Polyacrylamide Electricity	kg kg kWh	$\begin{array}{c} 9.00 \\ 1.06 \times 10^{-2} \\ 7.44 \times 10^{-2} \end{array}$	$\begin{array}{c} 9.00 \\ 1.06 \times 10^{-2} \\ 7.44 \times 10^{-2} \end{array}$	$\begin{array}{c} 9.00 \\ 1.06 \times 10^{-2} \\ 7.44 \times 10^{-2} \end{array}$			
Thermal dry	ying							
	Energy	kWh	2.36	2.36	-			
CLOU								
	OC Air Produced thermal energy	kg kg kWh	$8.62 imes 10^{-3} \\ 3.86 \\ 2.59$	6.67×10^{-3} 3.86 2.68	- - -			
PEM	07							
	H ₂ O Renewable electricity	kg kWh	6.91 1.79	6.91 1.79	- -			
MU								
	CO ₂ produced in CLOU H ₂ produced in PEM Methane Ni catalyst	kg kg kg kg	$\begin{array}{c} 2.11\\ 3.84\times10^{-1}\\ 7.37\times10^{-1}\\ 1.47\times10^{-5}\end{array}$	$\begin{array}{c} 2.11 \\ 3.84 \times 10^{-1} \\ 7.37 \times 10^{-1} \\ 1.47 \times 10^{-5} \end{array}$	- - - -			
Transport								
	Cargo Diesel Single way	kg kg km	$\begin{array}{c} 3.15 \times 10^{-1} \\ 1.80 \times 10^{-3} \\ 3.00 \times 10^2 \end{array}$	$\begin{array}{c} 2.50 \times 10^{-1} \\ 1.43 \times 10^{-3} \\ 3.00 \times 10^2 \end{array}$	$\begin{array}{c} 4.17\\ 2.44\times 10^{-2}\\ 3.00\times 10^2\end{array}$			
Landfill								
	Ash OC spent Catalyst Sewage Electricity	kg kg kg kg kWh	3.06×10^{-1} 8.62×10^{-3} 1.47×10^{-5}	$\begin{array}{c} 2.43 \times 10^{-1} \\ 6.67 \times 10^{-3} \\ 1.47 \times 10^{-5} \\ - \\ - \end{array}$	4.17 9.66×10^{-1}			

The environmental performance is quantified using the environmental footprint (EF) 3.0 method developed by the Joint Research Centre (JRC) of the European Commission [30,31]. Performance is expressed in terms of 16 impact categories, which are described in the literature [32]. Climate change impacts are only reported as the sum of the contributions from fossil and biogenic greenhouse gases and land-use change.

3. Results and Discussion

The proposed system requires a single HP-PEM cell and produces 0.74 kg/h of methane for both sewage sludges, while the amounts of solid waste sent into a landfill and the generated

6 of 13

thermal power are 0.31 kg/h and 2.59 kW/h for sludge 1 and 0.24 kg/h and 2.68 kW/h for sludge 2, respectively, with respect to 1 kg/h of sludge fed to the CLOU system. A summary of the technical performance of the system is reported in Table S9 of the Supplementary Materials.

Given the different ash and volatile contents of the sludge, slightly different results were evaluated at the exit of the CLOU unit. From this point on, given that, at the inlet of the methanation unit, a 4:1 H_2 :CO₂ ratio is considered, the calculated results overlap.

Table 2 reports the results of the economic analysis. These clearly show that, under the above-mentioned hypotheses, the C_{EoL} of the reference system is slightly less than half of the specific end-of-life cost of the proposed one; the main reason being the costs related to the electric energy consumption in the HP-PEM cell. However, given the fact that existing landfill sites are rapidly filling up, an increase in the cost of landfilling in the near future is very likely. This could change the results of the economic analysis in favor of the proposed system.

Table 2. Economic performances of the proposed and reference systems.

	Sewage Sludge 1	Sewage Sludge 2	Reference		
	EUR/kg	EUR/kg	EUR/kg		
Conditioning and drying					
Polyacrylamide	$1.48 imes 10^{-2}$	$1.48 imes 10^{-2}$	$1.48 imes 10^{-2}$		
Mechanical drying	$5.36 imes 10^{-3}$	$5.36 imes 10^{-3}$	$5.36 imes10^{-3}$		
Thermal drying	$1.68 imes 10^{-2}$	$1.68 imes 10^{-2}$	-		
CLOU system					
Reactors	2.77×10^{-2}	$2.77 imes 10^{-2}$	-		
Cyclone	$6.49 imes 10^{-3}$	$6.49 imes 10^{-3}$	-		
Compressors	$8.46 imes10^{-3}$	$8.46 imes 10^{-3}$	-		
OC (inventory)	$8.74 imes10^{-4}$	$8.74 imes10^{-4}$	-		
Heat exchanger	$2.37 imes 10^{-2}$	$2.37 imes 10^{-2}$	-		
OC (replacement)	$1.51 imes10^{-3}$	$1.17 imes 10^{-3}$	-		
Produced thermal energy	$1.21 imes 10^{-1}$	$1.21 imes 10^{-1}$	-		
Electric energy	$2.46 imes 10^{-3}$	$2.46 imes 10^{-3}$	-		
Hydrogen production system					
PEM	$2.67 imes 10^{-1}$	$2.67 imes 10^{-1}$	-		
Water	$1.34 imes 10^{-2}$	$1.34 imes10^{-2}$	-		
Electric energy	4.83	4.83	-		
Methanation unit					
Reactors	$4.36 imes 10^{-2}$	$4.36 imes 10^{-2}$	-		
Compressors	$3.55 imes 10^{-2}$	$3.55 imes 10^{-2}$	-		
Catalyst (inventory)	$1.40 imes 10^{-2}$	$1.40 imes 10^{-2}$	-		
Methane	$4.00 imes10^{-1}$	$4.00 imes10^{-1}$	-		
Catalyst (replacement)	$6.62 imes10^{-4}$	$6.62 imes10^{-4}$	-		
Electric energy	$9.58 imes10^{-3}$	$9.58 imes 10^{-3}$	-		
Produced thermal energy	2.20×10^{-2}	$2.20 imes 10^{-2}$	-		
Transport					
Diesel	3.28×10^{-3}	$2.60 imes 10^{-3}$	$4.44 imes 10^{-2}$		
Single way	$5.24 imes10^{-3}$	$5.24 imes10^{-3}$	$5.24 imes10^{-3}$		

	Sewage Sludge 1	Sewage Sludge 2	Reference
	EUR/kg	EUR/kg	EUR/kg
Landfilling			
Ash	$1.53 imes10^{-1}$	$1.22 imes 10^{-1}$	-
OC spent	$4.31 imes10^{-3}$	$3.34 imes10^{-3}$	-
Ni catalyst	$7.35 imes10^{-6}$	$7.35 imes 10^{-6}$	-
Sewage	-	-	2.09
Produced electric energy	-	-	6.52×10^{-2}
Total			
$TCR_s + TCL_s$	5.03	5.00	2.15
R_s	$5.21 imes10^{-1}$	$5.21 imes 10^{-1}$	6.52×10^{-2}
C_{EoL}	4.51	4.48	2.09

Table 2. Cont.

Figure 2 reports C_{EoL} as a function of the specific cost of landfill disposal. As expected, the specific end-of-life cost of the reference system increases with rising landfill costs. Conversely, the C_{EoL} of the proposed system, which is almost similar for the two sewage sludges considered, is very marginally affected by the landfilling cost. Accordingly, the economic performances of the two systems are equal to a specific landfill cost of about 1 EUR/kg. Higher landfilling costs make the proposed system cost-effective.



Figure 2. Specific end-of-life costs as a function of specific landfill disposal costs for the proposed and reference systems.

The environmental impacts of the proposed system were calculated using GaBi sustainability software version 10. Figure 3 compares the environmental impacts of the proposed system with those associated with the reference system. The results are expressed in terms of the percentage difference between the impacts of the reference system and those of the proposed one, relative to those of the proposed system.



Percentage difference between conventional scenario Vs proposed layout (relative to proposed layout)



The chart shows that the proposed end-of-life scenario for sewage sludge outperforms the reference system in 7 out of the 16 impact categories analyzed, namely acidification, climate change, eutrophication marine and terrestrial, ozone depletion, photochemical ozone depletion, and resource use–energy carriers for both sewage sludges. Specifically, the system delivers a reduction of about 40% of the impacts in the acidification category due to anthropogenic air pollutants such as SO₂, NH₃, and NO_x, up to 880% of the impacts in the marine eutrophication category due to substances containing nitrogen and phosphorus in the marine environment. However, the proposed scenario yields significant increases in other impact categories, which are as high as ~30% in respiratory inorganic effects and ~140% in human health impacts from ionizing radiation.

Figure 4 shows, as a grouped stacked bars plot, the results of the hot-spot analysis for the process proposed in this work. Within each impact category, analysis outcomes for the conventional end-of-life scenario, the proposed layout fed with sludge 1, and the proposed layout fed with sludge 2 were reported from top to bottom. Clearly, the largest portion of the environmental impacts originate from the PEM unit, and particularly from the electricity required, with contributions ranging from 35% in the category "acidification" up to \sim 95% in the category "cancer human health". By contrast, the categories of eutrophication freshwater and ecotoxicity freshwater are dominated by the impacts associated with the CLOU plant, and in particular, by direct emissions and the production of the oxygen carrier.

As a multi-functional system, the proposed system benefits from credits associated with the generation of surplus thermal energy. These credits are significant, leading to net savings in the acidification, climate change, eutrophication, terrestrial and marine, photochemical ozone formation, resource use, and energy carriers categories.

A sensitivity analysis was performed to establish the required degree of confidence in the results of this study. The analysis covers three parameters: the OC replacement ratio, the sludge transportation distance, and the PEM electricity consumption. Each parameter was increased by 5%, 10%, and 25% compared to the base case assumption. It is worth noting here that the ranges in variation for PEM electricity requirements are representative of alternative commercial PEMs [33], while being arbitrary for the remaining two parameters.



Figure 4. Hot-spot analysis of the proposed process layout and the conventional end-of-life scenario. The label "Construction" includes the construction of all units.

The results of the analysis, reported in Table 3 for sewage sludge 1 and Table S10 in Supplementary Materials for sewage sludge 2, suggest that the LCA model is particularly sensitive to the assumptions made with respect to the oxygen carrier replacement rate and the PEM electricity consumption, while being only minimally affected by the transportation distance. For example, a 25% increase in the OC replacement rate leads to increases in the environmental impacts of up to 52% in the category acidification, 27% in ecotoxicity, terrestrial eutrophication, and ionizing radiation, and 12% in resource use (minerals and metals). The same increment in PEM electricity consumption results in considerable increases in 10 categories, ranging from 17% in the category of non-cancer human health to 74% in the category of marine eutrophication. Notably, in some categories, the percentage variation in the environmental impact is higher than the model parameter variation itself. This non-linear behavior occurs for categories where either CLOU or PEM are major contributors and the credits from thermal energy are significant (see Figure 4). These latter (which are negative) contribute by reducing the absolute value of the denominator, thus boosting the percentage variation.

	Sludge Transportation Scenario		Oxygen Carrier Replacement Ratio Scenario		Energy Consumption PEM Scenario					
	+5%	+10%	+25%	+5%	+10%	+25%	+5%	+10%	+25%	
				variation (from baseline)						
Acidification terrestrial and freshwater	0%	0%	0%	10%	21%	52%	8%	16%	40%	
Cancer human health effects	0%	0%	0%	0%	0%	1%	5%	10%	24%	
Climate change	0%	0%	0%	0%	1%	2%	1%	2%	6%	
Ecotoxicity freshwater	0%	0%	0%	5%	11%	26%	1%	1%	3%	
Eutrophication freshwater	0%	0%	0%	0%	0%	0%	0%	0%	0%	
Eutrophication marine	0%	0%	1%	5%	10%	26%	15%	29%	74%	
Eutrophication terrestrial	0%	0%	1%	6%	11%	28%	13%	26%	65%	
Ionising radiation-human health	0%	0%	0%	1%	2%	5%	4%	9%	21%	
Land use	0%	0%	1%	1%	2%	4%	6%	11%	28%	
Non-cancer human health effects	0%	0%	0%	1%	1%	3%	3%	7%	17%	
Ozone depletion	0%	0%	0%	0%	0%	0%	0%	0%	0%	
Photochemical ozone formation-human health	0%	0%	0%	2%	4%	9%	4%	8%	20%	
Resource use, energy carriers	0%	0%	0%	0%	1%	2%	1%	2%	4%	
Resource use, mineral and metals	0%	0%	0%	2%	5%	12%	3%	5%	14%	
Respiratory inorganics	0%	0%	0%	1%	2%	6%	4%	8%	20%	
Water scarcity	0%	0%	0%	1%	1%	4%	1%	2%	4%	

Table 3. Results as variation from the base line for the alternative sludge transportation distance, O.C. replacement ratio, and PEM energy consumption for sewage sludge 1.

The proposed system may represent a valid alternative with respect to several impact categories when compared to the reference system comprising conventional technology for the treatment of sewage sludge in the Campania region. The results also suggest that efforts to improve environmental performance should be primarily focused on the electricity consumption of the PEM.

The LCA study has two main limitations. First, it did not consider the decommissioning of the units in the foreground system; however, since the construction phase has negligible contributions to all of the environmental impacts (see Figure 4), it is expected that decommissioning also has negligible effects. Second, the comparative analysis did not include technologies other than landfilling, which represents the current end-of-life in Campania. Further comparisons should be carried out considering other technologies that are common practice in European countries and in the north of Italy, including land spreading, incineration, and composting or a mix of these. Even though in the Campania region, at present, the sludges end up in landfills outside the region, we also compared our results with alternative end-of-life strategies available in the literature, such as agriculture use (both land spreading and composting) and incineration. Disposal cost and environmental performance data are taken from [8]. Figure 5 reports the result of the comparison in terms of cost and carbon emissions, while data on the other impact categories analyzed are reported in Table S11 in the Supplementary Materials. In particular, the proposed system would significantly decrease carbon emissions with a relevant increase in cost with respect to conventional end-of-life strategies. Furthermore, it results in the best option in terms of emissions, causing photochemical ozone formation and terrestrial eutrophication impacts, while it represents a comparable solution in terms of acidification in comparison with landfilling, freshwater eutrophication in comparison with incineration, and marine eutrophication in comparison with land spreading. For the sake of clarity, the results of this comparison must be taken very carefully since the sludge characteristics (namely moisture content and/or elemental composition) may significantly change the treatment energy consumption and yields.



Figure 5. Comparison of disposal cost (A) and carbon cycle (B) of different end-of-life strategies.

Nonetheless, it must be noted that the results and conclusions from the LCA study are not limited to the Campania region; rather, they can be extended, with appropriate considerations, to other regions where alternative technologies like land-spreading and composting plants are either not available or prevented by local laws.

4. Conclusions

The analysis presented in this work shows that the proposed system is an effective sewage sludge treatment system, reducing landfill wastes by about 68% with respect to 1 kg/h of sewage sludge with 10% wt of humidity, while producing a net thermal power of 2.7 kWh and a low-purity methane stream.

The economic analysis shows that, under the base case hypotheses, the proposed system has a higher C_{EoL} (1.75 EUR \times kg⁻¹) than the conventional one (0.41 EUR \times kg⁻¹). The proposed system would become economically preferable when the cost of landfill disposal is above 0.4 EUR \times kg⁻¹. Although this means that the cost of landfill disposal would experience a fourfold increase compared to the current costs, this scenario is likely to be realized in the near future, given the fact that landfilling sites are rapidly filling up and, therefore, the associated costs are expected to increase significantly. The LCA results show that the proposed process is environmentally preferable to conventional technology for treating sewage sludges with respect to several categories including climate change, marine and terrestrial eutrophication, ozone depletion, photochemical ozone formation, and resource use-energy carriers. The credits for thermal energy generation and the reduction in the volume of sludge to be treated in landfills play a relevant role in reducing the environmental impacts of the proposed system. The largest source of environmental impacts, according to the hot-spot analysis, is the electricity consumption from water electrolysis, with contributions ranging from 35% up to \sim 90% in most of the impact categories. Finally, the sensitivity analysis suggests that the LCA results are highly affected by the oxygen carrier replacement rate and the PEM electricity consumption but not by the sludge transportation distance.

Supplementary Materials: The supporting information can be dowloaded at https://www.mdpi.com/article/10.3390/en17040901/s1.

Author Contributions: Conceptualization, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; methodology, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; software, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; validation, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; formal analysis, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; investigation, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; resources, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; data curation, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; writing—original draft preparation, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; writing—review and editing, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; visualization, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; supervision, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; supe

F.P. and E.M.; project administration, P.B., R.C., A.P., C.T., M.U., F.P. and E.M.; funding acquisition, P.B., R.C., A.P., C.T., M.U., F.P. and E.M. All authors have read and agreed to the published version of the manuscript.

Funding: This research was founded by the the MUR program PRIN 2022, grant number P2022K2449.

Data Availability Statement: Data available on request from the authors.

Conflicts of Interest: All authors declare that they have no conflicts of interest.

References

- 1. FAO. Information System on Water and Agriculture; FAO: Rome, Italy.
- Bagheri, M.; Bauer, T.; Burgman, L.E.; Wetterlund, E. Fifty years of sewage sludge management research: Mapping researchers' motivations and concerns. J. Environ. Manag. 2023, 325, 116412. [CrossRef]
- 3. Eurostat. Eurostat—European Statistics, Database; European Commission: Brussels, Belgium, 2021.
- 4. ISPRA. Rapporto Rifiuti Speciali, 2nd ed.; ISPRA: Rome, Italy, 2018.
- 5. Mininni, G.; Mauro, E.; Piccioli, B.; Colarullo, G.; Brandolini, F.; Giacomelli, P. Production and characteristics of sewage sludge in Italy. *Water Sci. Technol.* **2019**, *79*, 619–626. [CrossRef]
- Buonocore, E.; Mellino, S.; De Angelis, G.; Liu, G.; Ulgiati, S. Life cycle assessment indicators of urban wastewater and sewage sludge treatment. *Ecol. Indic.* 2018, 94, 13–23. [CrossRef]
- Kacprzak, M.; Neczaj, E.; Fijałkowski, K.; Grobelak, A.; Grosser, A.; Worwag, M.; Rorat, A.; Brattebo, H.; Almås, Å.; Singh, B.R. Sewage sludge disposal strategies for sustainable development. *Environ. Res.* 2017, 156, 39–46. [CrossRef]
- Zhang, H.; Rigamonti, L.; Visigalli, S.; Turolla, A.; Gronchi, P.; Canziani, R. Environmental and economic assessment of electrodewatering application to sewage sludge: A case study of an Italian wastewater treatment plant. J. Clean. Prod. 2019, 210, 1180–1192. [CrossRef]
- 9. Migliaccio, R.; Brachi, P.; Montagnaro, F.; Papa, S.; Tavano, A.; Montesarchio, P.; Ruoppolo, G.; Urciuolo, M. Sewage Sludge Gasification in a Fluidized Bed: Experimental Investigation and Modeling. *Ind. Eng. Chem. Res.* 2021, *60*, 5034–5047. [CrossRef]
- Niu, X.; Shen, L.; Jiang, S.; Gu, H.; Xiao, J. Combustion performance of sewage sludge in chemical looping combustion with bimetallic Cu-Fe oxygen carrier. *Chem. Eng. J.* 2016, 294, 185–192. [CrossRef]
- Bareschino, P.; Mancusi, E.; Urciuolo, M.; Paulillo, A.; Chirone, R.; Pepe, F. LCA and Feasibility Analysis of a Combined Chemical Looping Combustion and Power-to-Methane System for CO₂ Capture and Utilization. *Renew. Sustain. Energy Rev.* 2020, 130, 109962. [CrossRef]
- 12. ISO 14040:2006; Environmental Management—Life Cycle Assessment—Principles and Framework. ISO: Geneva, Switzerland, 2006.
- Bareschino, P.; Mancusi, E.; Urciuolo, M.; Coppola, A.; Solimene, R.; Pepe, F.; Chirone, R.; Salatino, P. Modelling of a combined biomass CLC combustion and renewable-energy-based methane production system for CO₂ utilization. *Powder Technol.* 2020, 373, 421–432. [CrossRef]
- 14. Rönsch, S.; Schneider, J.; Matthischke, S.; Schlüter, M.; Götz, M.; Lefebvre, J.; Prabhakaran, P.; Bajohr, S. Review on methanation—From fundamentals to current projects. *Fuel* **2016**, *166*, 276–296. [CrossRef]
- 15. Soleimani, S.; Lehner, M. Tri-Reforming of Methane: Thermodynamics, Operating Conditions, Reactor Technology and Efficiency Evaluation—A Review. *Energies* 2022, *15*, 7159. [CrossRef]
- 16. Brachi, P.; Bareschino, P.; Tregambi, C.; Pepe, F.; Urciuolo, M.; Ruoppolo, G.; Mancusi, E. Assessing the feasibility of an integrated CLC-methanation system using solar dried and torrefied biomasses as a feedstock. *Fuel* **2023**, *331*, 125951. [CrossRef]
- 17. Gómez, L.; Martínez, I.; Navarro, M.V.; García, T.; Murillo, R. Sorption-enhanced CO and CO₂ methanation (SEM) for the production of high purity methane. *Chem. Eng. J.* **2022**, *440*, 135842. [CrossRef]
- 18. Ferrero, D.; Lanzini, A.; Santarelli, M.; Leone, P. A comparative assessment on hydrogen production from low- and high-temperature electrolysis. *Int. J. Hydrogen Energy* **2013**, *38*, 3523–3536. [CrossRef]
- Peters, M.S.; Timmerhaus, K.D.; West, R.E.; Timmerhaus, K.; West, R. Plant Design and Economics for Chemical Engineers; McGraw-Hill: New York, NY, USA, 2003.
- Lyngfelt, A.; Leckner, B. A 1000MWth boiler for chemical-looping combustion of solid fuels—Discussion of design and costs. *Appl. Energy* 2015, 157, 475–487. [CrossRef]
- Diglio, G.; Hanak, D.P.; Bareschino, P.; Mancusi, E.; Pepe, F.; Montagnaro, F.; Manovic, V. Techno-economic analysis of sorptionenhanced steam methane reforming in a fixed bed reactor network integrated with fuel cell. *J. Power Sources* 2017, 364, 41–51. [CrossRef]
- 22. M.I.T., Pubblicazione del 9.luglio.2015 concernente i valori indicativi di riferimento dei costi di esercizio dell'impresa di autotrasporto per conto di terzi. 2015, 3.
- 23. ISO 14044:2006/Amd 2:2020; Environmental Management—Life Cycle Assessment—Requirements and Guidelines. ISO: Geneva, Switzerland; Amendment 2: London, UK, 2020.
- 24. Deloitte Consulting. Energy Market Reform in Europe European Energy and Climate Policies: Achievements and Challenges to 2020 and Beyond; International Atomic Energy Agency: Vienna, Austria, 2020.

- Clift, R.; Doig, A.; Finnveden, G. The application of Life Cycle Assessment to Integrated Solid Waste Management. Part 1—Methodology. Process Saf. Environ. Prot. 2000, 78, 279–287. [CrossRef]
- 26. Wernet, G.; Bauer, C.; Steubing, B.; Reinhard, J.; Moreno-Ruiz, E.; Weidema, B. The ecoinvent database version 3 (Part I): Overview and methodology. *Int. J. Life Cycle Assess.* 2016, 21, 1218–1230. [CrossRef]
- Zhang, X.; Bauer, C.; Mutel, C.L.; Volkart, K. Life Cycle Assessment of Power-to-Gas: Approaches, system variations and their environmental implications. *Appl. Energy* 2017, 190, 326–338. [CrossRef]
- 28. Mistry, M.; Gediga, J.; Boonzaier, S. Life cycle assessment of nickel products. Int. J. Life Cycle Assess. 2016, 21, 1559–1572. [CrossRef]
- 29. Thorne, R.J.; Bouman, E.A.; Sundseth, K.; Aranda, A.; Czakiert, T.; Pacyna, J.M.; Pacyna, E.G.; Krauz, M.; Celińska, A. Environmental impacts of a chemical looping combustion power plant. *Int. J. Greenh. Gas Control.* **2019**, *86*, 101–111. [CrossRef]
- Fazio, S.; Castellani, V.; Sala, S.; Schau, E.; Secchi, M.; Zampori, L.; Diaconu, E. Supporting Information to the Characterisation Factors of Recommended EF Life Cycle Impact Assessment Method; JRC Technical Reports; New Models and Differences with ILCD Contents; European Commission: Brussels, Belgium, 2018. [CrossRef]
- 31. Joint Research Centre. *Product Environmental Footprint Category Rules Guidance, Version 6.3;* Joint Research Centre: Brussels, Belgium, 2018.
- 32. Hauschild, M.Z.; Rosenbaum, R.K.; Olsen, S.I. *Life Cycle Assessment: Theory and Practice*; Springer: Berlin/Heidelberg, Germany, 2017. [CrossRef]
- Tenhumberg, N.; Büker, K. Ecological and Economic Evaluation of Hydrogen Production by Different Water Electrolysis Technologies. Chem. Ing. Tech. 2020, 92, 1586–1595. [CrossRef]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.