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Biogas-to-biomethane upgrading: A comparative review and assessment in a life cycle perspective

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ABSTRACT

The study reviews and compares the most utilised techniques to obtain high quality biomethane by upgrading biogas from anaerobic digestion of the organic fraction of municipal solid waste. Environmental and economic aspects of membrane separation, water scrubbing, chemical absorption with amine solvent, and pressure swing adsorption have been quantified in a life cycle perspective. An attributional environmental Life Cycle Assessment has been implemented with the support of a Material Flow Analysis and in combination with a complementary environmental Life Cycle Costing. The analyses are based on data largely obtained from Italian existing plants but they can be generalised to the whole European Union, as demonstrated by a companion sensitivity analysis. The comparative assessment of the results indicates all the examined options as fully sustainable, also identifying the "win-win" situations. In particular, the membrane separation technique appears to have the best performances, even though in some cases with limited differences. With reference to base case scenarios, this technique shows better results for the respiratory inorganics potential (up to 34%, i.e. up to 328 $kg_{PM2.5eq}$/y), global warming potential (up to 7%, i.e. up to 344 t_{CO2eq}/y), and non-renewable energy potential (up to 12%, i.e. up to 6400 $GJ_{primary}$/y) as well as for life cycle costs (up to 3.4%, i.e. about 60 k€/y). The performances of the examined techniques appear anyway dependent on site-specific conditions (such as the injection pressure in the gas grid or the existence/amount of local economic incentives) and commercial strategies for the market of interest.

1. Introduction

1.1. Background

In recent years, public attention on environmental concerns, and in particular on climate change, resource depletion and air quality, is soaring. Several European directives have been accordingly issued, such as the Directive (EU) 2018/2001 "RED II" [1], which provide for measures to reduce fossil fuel consumptions and related greenhouse gas (GHG) emissions. The role of biomethane in the decarbonisation of the energy sources in the coming decades appears then of crucial importance. Biomethane (also defined BioSNG) is fully substitutive of fossil natural gas, since it has a methane content higher than 97%$_{vol}$, and is obtainable from biomass or biowaste through appropriate biochemical or thermochemical treatments that make it suitable as energy carrier or transportation fuel. Biomethane can be produced by means of a "syngas road" [2], which is based on the biomass gasification process coupled with a series of complex stages of syngas cleaning and conditioning, catalytic methanation, and final upgrading and compression [3]. On the other hand, the production of biomethane by means of the upgrading of

Abbreviations: AD, Anaerobic Digestion; AwR, alkaline Absorption with Regeneration; BABIU, Bottom Ash for BIogas Upgrading; BM, Biological Methods; CA, Chemical Absorption; CIC, (Italian: *Certificati di Immissione in Consumo*) Tradable certificates based on the quota obligation for fossil fuel traders; CS, Cryogenic Separation; DEA, DiEthanolAmine; DGA, DiGlycolAmine; EBA, European Biogas Association; EC, European Commission; E.E., Electric Energy; FU, Functional Unit; GHG, Greenhouse Gas; GWP, Global Warming Potential; HPC, Hot Potassium Carbonate; ISO, International Organisation for Standardisation; LCA, Life Cycle Assessment; LCC, Life Cycle Costing; LCI, Life Cycle Inventory; LCIA, Life Cycle Impact Assessment; LHV, Low Heating Value; MDEA, MethylDiEthanolAmine; MEA, MonoEthanolAmine; MFA, Material Flow Analysis; MISE, (Italian: *Ministero dello Sviluppo Economico*) Ministry of Economic Development; MS, Membrane Separation; NREP, Non-Renewable Energy Potential; OFMSW, Organic Fraction of Municipal Solid Waste; OPS, Organic Physical Scrubbing; PSA, Pressure Swing Adsorption; PtG, Power-to-Gas; RINP, Respiratory INorganics Potential; TE, Thermal Energy; TRL, Technology Readiness Level; VF, Variation Factor; VOCs, Volatile Organic Compounds; WS, Water Scrubbing.

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biogas obtained from anaerobic digestion (AD) is more convenient in terms of technological reliability, economic feasibility and environmental sustainability [4]. This "biogas road" has a Technology Readiness Level (TRL) that already reached a value equivalent to the market availability, that is the maximum level of development of a technology [5]. Biogas is a mixture of methane, with contents between 55% and 70%, and carbon dioxide for the remaining part, with presence of trace elements, such as H_2O, H_2S, NH_3 and siloxanes [6]. It is generated from the anaerobic digestion of wet biomass (with a minimum moisture content of 30%), such as the organic fraction of municipal solid waste (OFMSW), energy crops, agricultural residues (mainly manure and straw), sewage sludge and other organic waste (such as landfilled waste or industrial waste from food and beverage industry) [7]. The production and utilisation of biomethane is favoured by important environmental advantages, such as the reduction of GHG emissions (more than 80%) with reference to those deriving from the use of conventional fossil fuels [8] as well as the intrinsic saving of non-renewable energy resources. These advantages appear even wider if biomethane is used as transport fuel [9]. All the carbon dioxide obtained from biomethane combustion is "biogenic", that is "carbon-neutral" because it originates from a biomass that has absorbed a comparable quantity of carbon dioxide during its growth. Moreover, when obtained from OFMSW, biomethane is one of the so-called "advanced" biofuels, i.e. not in competition with food crops [10], making its production and utilisation more environmentally and ethically sustainable than those of first-generation biofuels [11]. It is finally noteworthy that the injection of biomethane into the natural gas grid is considered an efficient energy strategy, also when it is distributed over long distances [12]. In 2017, over 500 plants produced about 20,000 GWh of biomethane from anaerobic digestion in Europe [13], with the number of plants that has considerably grown in recent years (43 new units only from 2016 to 2017). European biomethane production is actually based mainly on the treatment of agricultural feedstock, including both residues and energy crops (more than 13,000 GWh/y), and for the remaining part on that of biowaste (>4900 GWh/y) and sewage sludge (>1200 GWh/y) [13].

1.2. Life cycle perspective

Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) are two assessment tools, widely recognised as among the most objective and reliable to analyse and quantify the environmental and economic performances of goods, processes and services. The assessments refer to the whole life cycle, then they can include all the stages from the production till the end-of-life treatments. This allows to individuate the positive or negative contribution of each phase to the overall environmental and economic sustainability of the analysed system, then suggesting possible

Table 1
Main scientific papers focused on technical, environmental and economical performances of biogas-to-biomethane upgrading. (AwR, alkaline Absorption with Regeneration; BABIU, Bottom Ash for Biogas Upgrading; BM, Biological Methods; CA, Chemical absorption; CS, Cryogenic Separation; HPC, Hot Potassium Carbonate; MS, Membrane Separation; OPS, Organic Physical Scrubbing; PSA, Pressure Swing Adsorption; WS, Water Scrubbing).

Reference	Type of the study			Analysed upgrading technologies									
	Review study	Environmental assessment	Economic assessment	MS	WS	CA	PSA	OPS	CS	HPC	BM	BABIU	AwR
Pertl et al., 2010 [25]	–	LCA	–	x	x		x					x	
Adelt et al., 2011 [26]	–	LCA	–			x							
Patterson et al., 2011 [11]	Technical review	–	Economic analysis		x	x	x	x	x				
TUV, 2012 [18]	Technical review	–	Economic analysis		x	x	x	x					
Bauer et al., 2013 [19]	Technical review	–	Economic analysis		x	x	x	x	x				
Rehl & Müller, 2013 [33]	–	LCA	LCC		x								
Starr et al., 2014 [27]	–	LCA	–	x	x	x	x	x	x			x	x
Ravina & Genon, 2015 [35]	–	Carbon footprint	–		x								
Morero et al., 2015 [36]	–	LCA	–		x	x		x					
Sun et al., 2015 [20]	Technical review	–	Economic analysis	x	x	x	x	x	x		x		
Budzianowski, 2016 [21]	Technical review	–	–	x	x	x	x	x	x		x		
Leonzio, 2016 [28]	–	LCA	–			x							
Awe et al., 2017 [23]	Technical review	–	–	x	x	x	x	x	x				
Collet et al., 2017 [29]	–	LCA	Economic analysis	x	x	x	x						
Khan et al., 2017 [24]	Technical review	–	Economic analysis	x	x	x	x	x	x		x		
Koido et al., 2017 [37]	–	LCA	Economic analysis										
Miltner et al., 2017 [22]	Technical review	–	–	x	x	x	x	x	x				
Angelidaki et al., 2018 [7]	Technical review	–	Economic analysis	x	x	x	x	x	x		x		
Ardolino et al., 2018 [6]	–	LCA	–	x									
Khoshnevisan et al., 2018 [38]	–	LCA	–		x	x	x					x	x
Ardolino & Arena, 2019 [4]	–	LCA	–	x									
Baena-Moreno et al., 2019 [39]	Technical review	–	–	x	x	x	x	x	x				
Barbera et al., 2019 [32]	–	–	Techno-economic analysis		x		x			x			
D'Adamo et al., 2019 [30]	–	–	Socio-economic analysis	x									
Ferella et al., 2019 [31]	–	–	Techno-economic analysis				x						
Kapoor et al., 2019 [40]	Technical review	–	–	x	x	x	x	x	x		x		

improvement actions. The use of environmental LCA is largely widespread to individuate and quantify the potential benefits or burdens to the environment, but so far it is almost always used as a stand-alone tool [14,15]. Coupling LCA with LCC allows to obtain an objective and exhaustive assessment from both environmental and economic points of view along the whole life cycle perspective, which should be crucial to define whether a good, process or service is really sustainable [16] and to identify the win-win situations [17].

1.3. Literature review

Different technical reviews, LCA studies and economic assessment have been focused on biogas production and its upgrading to biomethane (Table 1). In particular, the first published reports [11,18] investigated the technical aspects of the main biogas upgrading technologies available on the market and provided some economic data, such as specific investment and maintenance costs, with reference to units with different treatment capacities [19]. Sun et al. [20] analysed both commercial and innovative upgrading techniques, in order to compare characteristics of the obtained biomethane with those required for its utilisation in different final applications. Other reviews [21,22] focused on innovative solutions for the biogas-to-biomethane upgrading sector, highlighting the potential advantages of some techniques still under development [23], such those implying the adoption of biological methods [7]. Khan et al. [24] indicated the crucial role of a better knowledge of the environmental performances related to the different upgrading units, anyway none of currently published technical reviews (listed in Table 1) reported such investigation with a life-cycle approach. On the other hand, LCA has been often adopted as the appropriate tool to evaluate the reduction of GHG emissions obtained by biogas upgrading with reference to the whole biomethane production chain [25], starting from different types of feedstock, such as energy crops, manure and municipal biowaste [26]. Starr et al. [27] investigated two innovative biogas upgrading technologies, namely alkaline absorption with regeneration (AwR) and bottom ash for biogas upgrading (BABIU), considering different process configurations and comparing them with well-developed technologies. Environmental performances of BABIU technology appear to be better than those of AwR and similar to those of commercial upgrading technologies. Anyway, both these innovative processes are still at pilot-plant scale, then need a further and positive scale-up before their commercialisation. Leonzio [28] focused on chemical absorption with different solvents, finding out that even though mono-ethanolamine solution has the highest upgrading efficiencies, it still needs some technical and environmental improvements. Collet et al. [29] investigated biomethane production by means of anaerobic digestion coupled with a Power-to-Gas (PtG) technology, by using sewage sludge as substrate. They found out that environmental sustainability of the PtG technology is strongly affected by the availability of renewable energy as source of electricity. They also demonstrated that the economic feasibility of the process cannot be reached currently, but possible improvements can be obtained in future decades thanks to the progress of the technology and those in the energy sector. The study by Ardolino et al. [6] quantified the advantage of the utilisation of biomethane as transport fuel, also highlighting the limited contribution of biogas upgrading to the overall environmental performances of the anaerobic digestion process. Ardolino and Arena [4] compared the “biogas road” and the “syngas road” for biomethane production, demonstrating the high potentials in terms of energy efficiency and carbon utilisation of the syngas road. The latter, on the other hand, still suffers from its low TRL and, above all, from an economic feasibility obtainable only with large-scale plants. Few studies evaluated economic aspects of the upgrading technologies [30], quantifying some conventional indexes such as Net Present Value [31] and investment payback time [32]. At the authors’ knowledge, only one paper [33] quantified the economical performances of a single upgrading technology by means of an LCC carried out in combination with an environmental LCA. Other papers focused on LCC of waste management systems have a very different scope with respect to that of the present study [16,34].

1.4. Aims and innovative aspects

The scope of the study is to investigate and compare, in a life cycle perspective, the environmental and economic aspects of the main commercial techniques for biogas-to-biomethane upgrading: membrane separation, water scrubbing, chemical absorption with amine solvent, and pressure swing adsorption. The pros and cons of each of them have been identified and quantified under different operative conditions by means of environmental LCA and LCC. There are important innovative aspects of this study. First, the LCA analysis has been developed with the synergy of a material flow analysis (MFA) specifically applied to each of the considered technological options. Moreover, it is the first time that different biogas-to-biomethane upgrading techniques are investigated by a combination of LCA and LCC analyses. The obtained results have been used to individuate the win-win situations, which are those that better optimise environmental and economic aspects [17]. There are only few examples of these combined assessments in the literature of solid waste management, since the environmental and economic aspects are often developed separately, and generally based on different crucial assumptions, such as the functional unit and system boundary definitions [16]. The structure of the paper and the proposed methodological approach is schematically summarised in Fig. 1, with the indication of the input/output to/from all the steps, and that of the links between them. The paper reports first an essential description of the main commercially available upgrading techniques, together with the MFAs developed for each of them. Hence, the environmental LCA is implemented in compliance with the standard procedure [41]. The environmental LCC is then proposed, together with the comparative analysis of the results obtained with the two life cycle tools.

2. Technical aspects of biogas-to-biomethane upgrading

There is an increasing interest towards the anaerobic digestion process aimed to produce biogas and upgrade it to biomethane, especially from a negative-value feedstock, such as biowaste. The obtained biomethane can be used for several final applications, such as heating or transportation fuel in shipping and automotive sectors [9,11]. There are several techniques for biogas upgrading available on the market at different development stages. Four of them are well-established on the market: membrane separation, water scrubbing, chemical absorption and pressure swing adsorption. Some others are less widespread, such as organic physical scrubbing, or still at demonstrative stage, like cryogenic separation technologies, hot potassium carbonate and biological methods [7]. This study is focused on these most widespread upgrading techniques, which represent more than 85% of the whole market [13].

The Membrane Separation (MS) technique utilises special membranes in the form of a bundle of hollow fibres made of polymeric materials such as polysulfone, polyimide or polydimethylsiloxane, incorporated in a stainless-steel tube [42]. These materials have a strong selectivity in methane/carbon dioxide separation. More specifically, they are permeable to CO_2, H_2O, NH_3, less permeable to O_2 and H_2S and very little permeable to CH_4 and N_2 [43]. In this way, a flow (called “permeate”) composed mainly by CO_2, H_2O, NH_3 and other residues penetrates through the micro-pores, while the CH_4 rich gas (called “retentate”) passes through the membranes without being removed. To provide a sufficient surface area, these membranes are used as a set of multiple modules [42]. In the analysed configuration (top of Fig. 2), raw biogas is preliminary cleaned in a dryer and a scrubber to remove water and hydrogen sulphide, which could cause a worsening of the upgrading performance. The scrubber utilises a mixture of caustic soda, polyvinyl alcohol and water as desulfurization solvent. Then, an activated carbon filter allows removing the remaining traces of H_2S and VOCs (Volatile

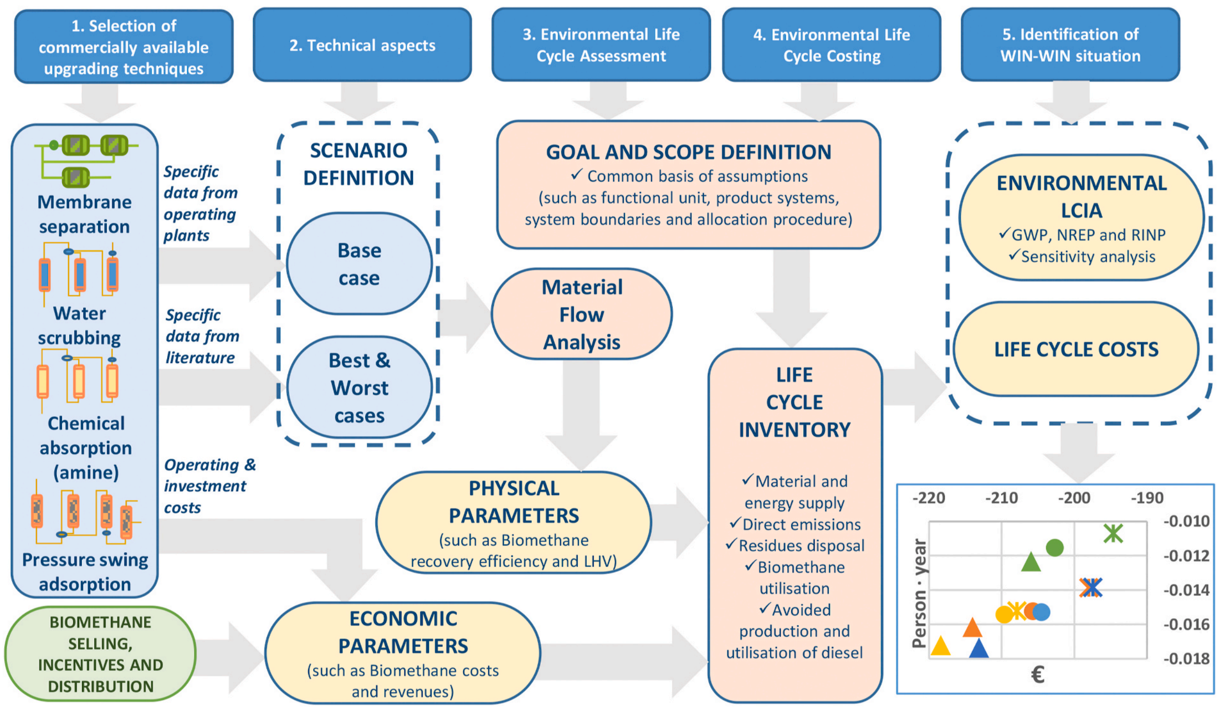

Fig. 1. Structure of the methodological approach of the paper, with the indication of each step and related inputs/outputs and links.

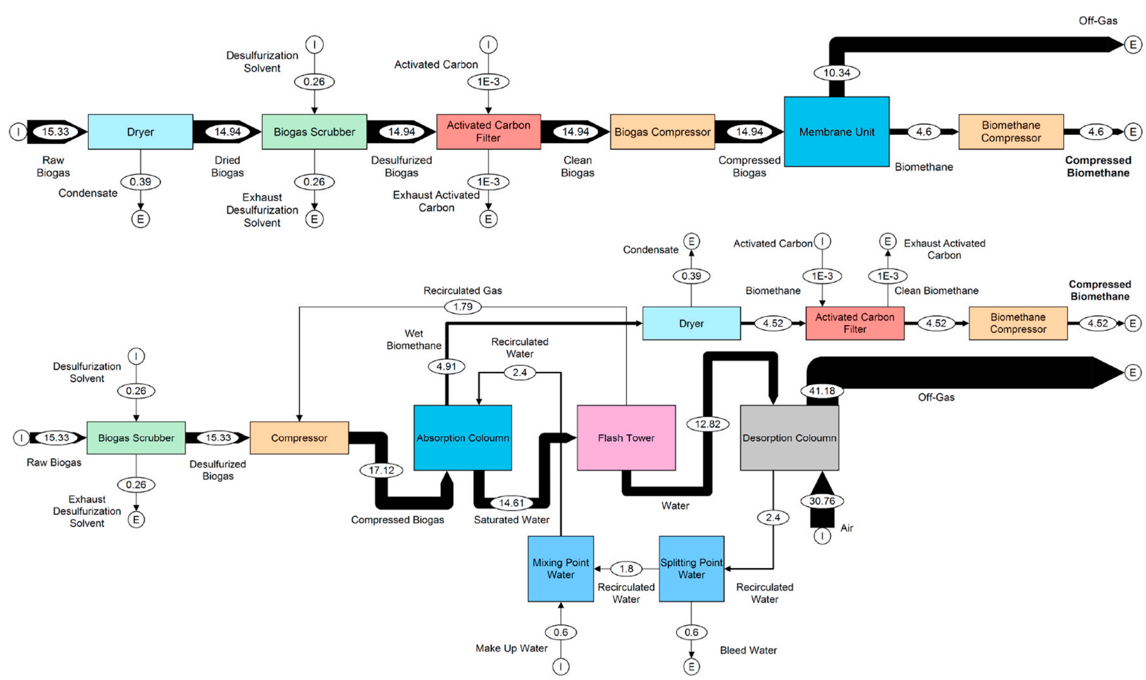

Fig. 2. Quantified flow sheet of the MS base scenario (top) and WS base scenario (bottom). Mass flow rates are reported in t/d, with reference to the functional unit (500 $m^3_{N\ raw\ biogas}$/h, which corresponds to 15.33 t/d). I = import stream; E = export stream.

Organic Compounds). Successively, biogas is compressed up to pressures of 10–16 bar and sent to the entrance of the permeation unit. The internal gas recirculation in a 3-stage polyimide membrane unit allows achieving high CO_2 removal efficiencies (about 98%), with very low methane slips (around 0.69% for the base case) [6].

The Water Scrubbing (WS) technique is based on different solubility of carbon dioxide in water with respect to that of methane: according to Henry's law, carbon dioxide has a solubility 26 times higher than methane in water at 25 °C. The WS process is favoured by low temperatures and high pressures [19,44]. In the configuration analysed in this study (bottom of Fig. 2), raw biogas is first treated in a scrubber to remove hydrogen sulphide, utilising the same type and amount of desulfurization solvent utilised for the MS unit. Desulfurized biogas is compressed to around 4–6.5 bar and introduced into the washing column from the bottom, where it meets water injected from the top. CO_2 is absorbed by water while CH_4 comes out from the top of the washing column and, after a drying step and a refining stage in an activated carbon filter to remove VOCs traces, can be compressed into the natural gas grid. The saturated water coming out of the scrubber is rich in CO_2 but it also contains about 5–6% of the methane content in the compressed biogas (Fig. 2 bottom) [44], which cannot be lost nor emitted into the atmosphere. For this reason, water is directed to a flash column, where a pressure drop up to 2–4 bar allows methane separation and recirculation [19,44]. After that, water stream is sent to a stripping column, where CO_2 is removed, making water reusable in the process, reducing the required make-up to 0.05 kg/$m^3_{N\ raw\ biogas}$. WS technology shows a CO_2 removal efficiency always higher than 98% and a range of methane slip from 1 to 2% [7].

The Chemical Absorption (CA) with amine solvents is a variant of the scrubber techniques, since it uses organic amines as solvent, such as monoethanolamine (MEA), diethanolamine (DEA), methyldiethanolamine (MDEA) and diglycolamine (DGA) [28]. The operating principle is similar to that of WS, but amine solvents are more selective in absorbing CO_2 with respect to water, which make them able to remove larger amount of CO_2 per unit volume, ensuring smaller upgrading units [11]. Furthermore, amines solvents are efficacious at almost atmospheric pressure [18], thus consuming low quantity of electric energy. On the other hand, they require a certain amount of thermal energy for the amine regeneration inside the stripper [19], together with amine solvent make-up to avoid loss of process efficiency. In the configuration analysed in this study and reported on the top of Fig. 3, raw biogas is desulfurized (analogously to MS and WS) and fed to the absorber where it meets the amine solvent (MEA), which absorbs carbon dioxide from biogas. This exothermic reaction increases the temperature of the amine solvent. Biomethane exits from the top of the absorber and enters a refining phase and then the final compression. The "rich" amine solvent, so called because of its CO_2 content, is first heated in a heat exchanger, exploiting the heat of the "lean" amine solvent (which comes from the stripping column and is directed to the absorber), and then is fed to the stripper. The latter is provided with a reboiler to ensure the necessary heat for the stripping reaction. In this way the recovered CO_2-free amine solvent can be sent to the heat exchanger mentioned above, after an appropriate make-up phase. Steam and amine solvent traces are separated from the obtained off-gas and recirculated to the stripping column [19], while the remaining part (mainly CO_2) is released into the atmosphere. The CA technology allows the highest methane recovery thanks to its lowest methane slip (up to 0.04%) [18].

Finally, the Pressure Swing Adsorption (PSA) technology utilises the capability of a porous adsorbent medium to adsorb some target molecules out of a gas mixture, then released by applying different values of pressure [19]. For the biogas upgrading process, PSA units take advantages from the different molecular dimensions of CO_2 (0.34 nm) and CH_4 (0.38 nm). Consequently, the utilisation of an adsorbent material with cavities of 0.37 nm allows retaining CO_2 in the pores, while CH_4 flows without being retained [11]. Zeolites and activated carbons represent the most utilised types of available adsorbent materials, thanks to their efficiency [45,46]. The specific configuration, reported on the bottom of Fig. 3, includes a pre-treatment phase with activated carbon to remove H_2S and a drying step to remove water, and then a compression to around 4 bar towards the PSA unit. The latter is provided with four columns in series (Skarstrom cycle), packed with zeolites as adsorbent material. The four columns configuration allows the continuity of the operations, with each of them involved in a different phase of the process: in the first, compressed biogas is fed in a column in which carbon dioxide is adsorbed by the adsorbent material while methane passes through without being adsorbed; in the second, a pressure drop allows carbon dioxide desorption; during the third phase, the column is cleaned from residual CO_2 injecting a part of biomethane and, finally, the column is re-pressurized during the fourth phase [19]. Residual VOCs are removed from biomethane in another activated carbon filter and then biomethane is compressed to 24 bar. Due to uncertain data related to adsorbent material substitution, the full replacement of zeolite has been considered once a year, as suggested by Ferella et al. [31]. PSA technology shows the lowest efficiency of recovery with methane slip varying from 1.8% to 2% [18,19].

3. Environmental life cycle assessment

3.1. Goal and scope definition for LCA and LCC

The first phase of any standardised LCA [41] is the definition of the goal and scope of the study. This step is in common for the

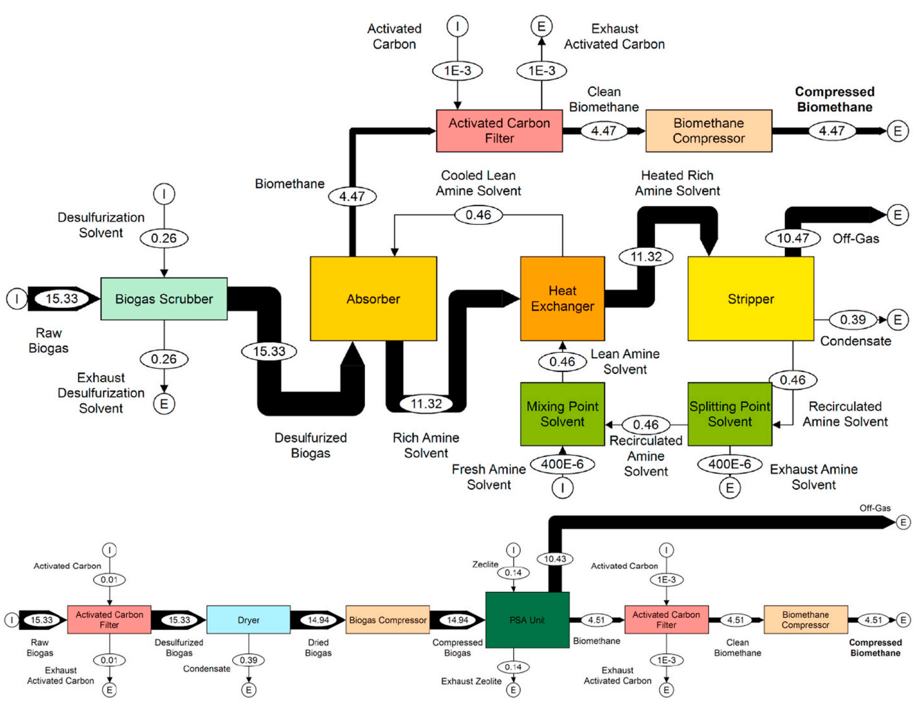

Fig. 3. Quantified flow sheet of the CA base scenario (top) and PSA base scenario (bottom). Mass flow rates are reported in t/d, with reference to the functional unit (500 m^3_N raw biogas/h, which corresponds to 15.33 t/d). I = import stream; E = export stream.

environmental LCA and LCC analyses described here. An attributional approach has been utilised for the process-based LCA, then accounting for impacts directly related to the system of interest and attributing them to the activities within the system in the current perspective [47]. An environmental approach has been instead utilised for the LCC, then accounting all internal costs (i.e. monetary flows) related to the life cycle of the product system of interest that are directly covered by one or more of the involved actors, with the inclusion of taxes and duties [17].

The *intended application* of the study is the comparison of the environmental and economic performances of market available biogas upgrading techniques utilised to produce biomethane from anaerobic digestion of the OFMSW. The *main reason for carrying out the study* is the quantification of the environmental and economic sustainability of biomethane production through different biogas upgrading techniques, taking into account the necessity of different pre-treatments, energy demands, water and material consumption as well as methane slips, biomethane purity, CO_2 removal efficiencies. The *intended audience* of the study is that of different kinds of operators in (bio)waste management, such as decision-makers involved in waste management planning, where biowaste treatment plants should be included.

The *product systems to be studied* are the biogas upgrading units aimed at biomethane production and based on different processes (absorption, adsorption, membrane separation), with different process sub-units and pre-treatments (scrubbers, dryers, heat exchangers), and a final biomethane pressurization. The *function of the product systems* is then the upgrading of raw biogas from anaerobic digestion of OFMSW for biomethane production. The *functional unit* is the upgrading of 500 m^3_N/h of raw biogas, having a fixed composition, to produce biomethane for road transport. The value of 500 m^3_N/h was chosen as functional unit because this flow rate is one of the most frequent in commercial applications. It corresponds to the biogas generation from an AD plant with a capacity of 30–35 kt/y of OFMSW, that is a common size in Europe [6]. All the analysed upgrading units are commercially available for this specific flow rate. Biogas composition strictly depends on feedstock from which it is produced. Here, a mesophilic anaerobic digestion phase (37–39 °C) has been considered to produce the biogas [6], with the composition reported in Table 2.

The selected *comparative scenarios* are the mentioned main upgrading techniques of biogas produced from OFMSW. For each of them, three configurations (base, worst and best) have been analysed and quantitatively compared both in the LCA and LCC. The *system boundaries* are those generally indicated as a "gate-to-wheel", where the input gate is the raw biogas produced by anaerobic digestion of OFMSW, and the final gate is the combustion of biomethane as fuel for automotive purpose, as illustrated in Fig. 4.

Processes and flows in the foreground system are those to which the analyses mainly focus on, while those in the background system are those necessary for an appropriate working of the foreground system [48]. Most of data for the foreground derive from Italian existing plants, even though some of them have been obtained from manufacturers or scientific literature. Data for the background derive from Ecoinvent v.3.3 [49], utilising the databank processes reported in Table A.1 of Annex A, with reference to each specific upgrading technique. The AD stage has been excluded from the system under analysis because it is the same for each scenario, then implying an equal set of direct, indirect and avoided burdens. On the contrary, the use of biomethane in cars (and therefore the avoided utilisation of diesel) is not the same for each upgrading unit, being dependent on their specific performance parameters (methane slip, CO_2 removal efficiency, etc.), then it has been included.

The *allocation* issue deriving from the multi-functionality of the analysed systems is avoided by means of the system expansion methodology, then identifying which market-available product is substituted by the obtained co-product, and evaluating the avoided burdens and impacts to be considered in the analysis [48]. As in previous studies on the same field of interest [6], the avoided impacts linked to biomethane utilisation as substitute of fossil fuel have been calculated, choosing diesel as replaced product and following the procedure indicated by Vadenbo et al. [50]. The latter calculates the possible substitution of a conventional product already present on the market (diesel) with a secondary resource (biomethane), as the product:

$$\gamma = U^{\text{biomethane}} * \eta^{\text{biomethane}} * \alpha^{\text{biomethane: diesel}} * \pi^{\text{diesel}} \qquad \text{Eq. 1}$$

where $U^{\text{biomethane}}$ is the possible quantity of the secondary resource; $\eta^{\text{biomethane}}$ is the recovery efficiency of this resource, which is related to the performances of each single upgrading unit; $\alpha^{\text{biomethane: diesel}}$ is the substitutability, which depends on the functionality of the biomethane compared with the functionality of diesel when used as automotive fuel, quantified as the required amounts of these two fuels necessary to travel the same distance, taking also into account the generated emissions; and π^{diesel} is the related acceptability showed by the market [6]. Eq. (1) gives the distances (expressed in km) travelled by using biomethane and avoiding diesel utilisation.

The adopted *Life Cycle Impact Assessment* (LCIA) *methodology* is Impact 2002+ [51], and the impact categories of main interest are those of Respiratory Inorganics Potential (RINP), Global Warming Potential (GWP) and Non-Renewable Energy Potential (NREP). The *data quality,* evaluated in an LCA study as geographical, temporal and technological consistency, is high, since almost all data have been collected from existing plants or directly provided by manufacturers. The analyses have been developed with reference to the Italian market, considering both its national electricity mix [52] and legislation related to the promotion of the use of biomethane in the transportation sector [53]. With reference to the main *assumptions and limitations*: the study must be considered valid within the specific conditions assumed, in particular for the composition of reference biogas and the examined techniques.

3.2. Life cycle inventory

The study collected all the data necessary to quantify the direct and avoided burdens linked to the different activities considered in the system boundaries (biogas pre-treatment, upgrading and pressurization as well as biomethane/diesel utilisation). The technical description of each scenario is reported in section 2, while data and input parameters for the quantification of environmental burdens related to all scenarios are listed in Table 3. Output parameters are calculated based on the MFAs summarised by Figs. 2 and 3. Base cases identify real situations for which data from Italian existing plants [54–56] were available or obtainable by manufacturers [57–60]. Best and worst scenarios are instead defined by identifying the key parameters (energy consumption, carbon dioxide removal efficiency, methane slip, etc.), and defining a range of variation based on information collected from scientific literature [7,19,28,29,61,62]. Different solutions in pre-treatments and refining phases (i.e. scrubbers and/or pre- and post-activated carbon filters) have been chosen based on different real plants configurations. The electricity necessary to compress the biomethane to the pressure

Table 2
Composition of the raw biogas utilised as reference [6].

Raw biogas	
Composition, $\%_{vol}$	
CH_4	50.83
CO_2	44.59
H_2S	0.01
N_2	0.38
O_2	0.10
NH_3	<0.01
H_2O	4.09
Pressure, mbarg	5
Temperature, °C	37–39
Low Heating Value, MJ/m^3_N	18.2

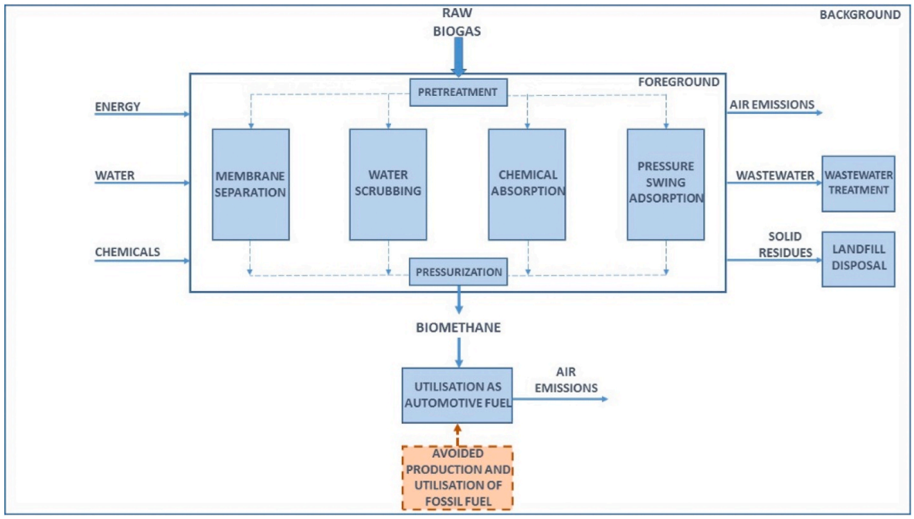

Fig. 4. System boundaries of LCA and LCC analyses. Blue dashed lines refer to the analysed alternative scenarios; red dashed lines indicate the avoided burdens. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 3
Specific data and parameters utilised to evaluate the environmental burdens of the analysed scenarios. Data for base cases derive directly from existing plants [54–56] or from manufacturers [57–60]. Data for best and worst scenarios are mainly derived from scientific literature [7,19,28,29,61,62].

	MS			WS			CA			PSA		
	WORST	BASE	BEST	WORST	BASE	BEST	WORST	BASE	BEST	WORST	BASE	BEST
INPUT parameters												
Pre-treatment phase												
Desulfurization solvent, $g/m^3_{N\ raw\ biogas}$		21.4			21.4			21.4			–	
Activated carbon for pre-treatment, $g/m^3_{N\ raw\ biogas}$		0.08			-			–			0.81	
Upgrading and refining phases												
CH_4 slip, %	1.0	0.69	0.5	2.0	1.5	1.0	0.1	0.1	0.04	2	1.8	1.8
CO_2 removal, %	97	98	99	98	98.5	99	99	99.5	99.9	98	98.5	99
Operating pressure, bar		14			4			1			4	
Electric energy, $kWh/m^3_{N\ raw\ biogas}$	0.30	0.29	0.18	0.33	0.25	0.20	0.15	0.10	0.03	0.30	0.25	0.20
Thermal energy, $kWh/m^3_{N\ raw\ biogas}$		–			–		0.75	0.55	0.27		–	
Zeolite, $g/m^3_{N\ raw\ biogas}$		–			–			–			11.9	
Water, $kg/m^3_{N\ raw\ biogas}$		–			0.05			0.03			–	
Amine solvent, $g/m^3_{N\ raw\ biogas}$		–			–			0.03			–	
Activated carbon for refining, $g/m^3_{N\ raw\ biogas}$		–			0.08			0.08			0.08	
Injection into the grid phase												
E.E. for biomethane compression, $kWh/m^3_{N\ raw\ biogas}$		0.05			0.09			0.14			0.09	
OUTPUT parameters												
Biomethane recovery efficiency $\eta^{biomethane}$, $m^3_{N\ biomethane}/m^3_{N\ raw\ biogas}$	0.521	0.518	0.515	0.512	0.512	0.512	0.517	0.515	0.513	0.512	0.510	0.508
Biomethane LHV, $MJ/m^3_{N\ biomethane}$	34.59	34.89	35.15	34.85	35.01	35.16	35.17	35.32	35.44	34.85	35.00	35.16

required at the injection point of the natural gas grid (24 bar) has been quantified considering the different operating pressure of each upgrading unit. Table 4 reports the direct and avoided burdens, calculated starting from all the above reported data. With reference to the main direct burdens, the consumptions relate mainly with electric and thermal energy, desulfurizing solvents, activated carbon, water and amine solvent (where necessary). Emissions to water are mainly due to wastewater and to the amine solvent make-up, while solid residues are linked to spent activated carbon and zeolites. Air emissions are related to biogenic CO_2 emissions (linked to CO_2 removal efficiency of each upgrading technology) and CH_4 lost through off-gas. The CH_4 emissions are related to methane slip of the single technology, therefore CA shows the lowest emissions followed by MS. The impacts linked to biomethane utilisation as automotive fuel in place of diesel have been quantified based on the procedure indicated by Vadenbo et al. [50], already reported as Eq. (1). The potential amount of biomethane ($U^{biomethane}$) is equal for all analysed units, since the same raw biogas flow rate and composition have been considered and established as functional unit. The biomethane recovery efficiencies ($\eta^{biomethane}$), which strictly depend on the upgrading units, have been quantified as the ratio between the volumetric flow rate of obtained biomethane and that of biogas, thus taking into account specific CH_4 slip and CO_2 removal efficiencies. The obtained values for each scenario are shown in Table 3. The substitutability ($\alpha^{biomethane:\ diesel}$) takes into account the functionality of biomethane compared to that of diesel in the same type of vehicles. A vehicle fleet composed by passenger cars has been assumed with a consumption of 159 MJ of biomethane or 2.62 kg of diesel for travelling 100 km. Different air emissions linked to the use of biomethane and diesel (including biogenic/fossil CO_2, CO, CH_4, NOx, PM2.5, N_2O, SO_2, NMVOC, and Hydrocarbons) have been considered, as reported by Ardolino et al. [6]. The market response (π^{diesel}) has been set equal to 1 for all scenarios under analysis, considering that all the obtained biomethane is marketable, since the Italian legislation [53] establishes that economic benefits for biomethane can be received by producers only if its utilisation as transportation fuel is demonstrated. The avoided burdens (distance covered with diesel and related production) calculated by Eq. (1) for each analysed scenario are shown in Table 4.

Table 4
Environmental burdens (direct and avoided) for the analysed scenarios. All the data refer to the functional unit (500 $m^3_{N\ raw\ biogas}$/h).

	MS			WS			CA			PSA		
	WORST	BASE	BEST	WORST	BASE	BEST	WORST	BASE	BEST	WORST	BASE	BEST
Consumption												
Electric energy, kWh	175	170	115	210	170	145	145	120	85	195	170	145
Thermal energy, kWh		–			–		375	275	135		–	
Desulfurization solvent, kg		10.7			10.7			10.7			–	
Activated carbon, g		40			40			40			445	
Zeolite, kg		–			–			–			5.95	
Water, kg		–			25			15			–	
Amine solvent, g		–			–			15			–	
Waste to treatment												
Wastewater, kg		–			25			15			–	
Exhaust amine solvent, g		–			–			15			–	
Exhaust desulfurization solvent, kg		11			11			11			–	
Condensate, kg		16			16			16			16	
Activated carbon, g		45			45			45			525	
Zeolite, kg		–			–			–			5.95	
Air emissions												
$CH_{4\ biogenic}$, kg	1.8	1.3	0.9	3.6	2.7	1.8	0.2	0.2	0.1	3.6	3.3	3.3
$CO_{2\ biogenic}$, kg	424.5	428.9	432.7	428.9	431.1	433.3	433.3	435.5	437.2	428.9	431.1	433.3
Biomethane utilisation												
Distance covered with biomethane, km	5665	5682	5693	5607	5636	5665	5716	5716	5719	5607	5619	5619
Avoided burdens												
Distance covered with diesel and related production, km	5665	5682	5693	5607	5636	5665	5716	5716	5719	5607	5619	5619

3.3. *Life cycle impact assessment*

The normalised results of LCIA [63] shown in Fig. 5 (only base cases) and Fig. 6 (base, best and worst cases) quantify the potential impacts of the midpoint categories that have a crucial role in the environmental performances of the analysed systems. The same results related to all the midpoint categories are reported in Figs. B1–B.4 of Annex B. The normalised results are reported in terms of person·year, that is the average impact in a specific category caused by a person during one year in Europe. The total values of all the main impact categories are negative, showing always benefits in the environmental impacts related to all the examined upgrading techniques, because the avoided burdens, associated with biogas upgrading to biomethane and its subsequent use as fuel for transport, are greater than the related direct and indirect burdens. MS always shows the best environmental performances.

For RINP (Fig. 5), there is a significant contribution related to biomethane utilisation in the direct potential impacts. This is mainly due to the NO_X and particulate emissions during biomethane combustion [9]. On the other hand, the avoided potential impacts are larger and related to the higher emission levels during avoided diesel utilisation and production phases. PSA shows the worst performances, due to the negative contributions related to activated carbon and, especially, zeolite consumption (material supply). For GWP, the main contribution to the high avoided potential impacts are related to diesel utilisation phase, which implies high fossil CO_2 emissions [9]. Among the direct potential impacts, there is a small contribution related to direct emissions for MS, WS and PSA, which are mainly due to the methane slip. For CA, this contribution is negligible (Table 4) but there is a higher contribution related to energy supply, since this technique requires important amounts of thermal energy. PSA again suffers from the consumptions of

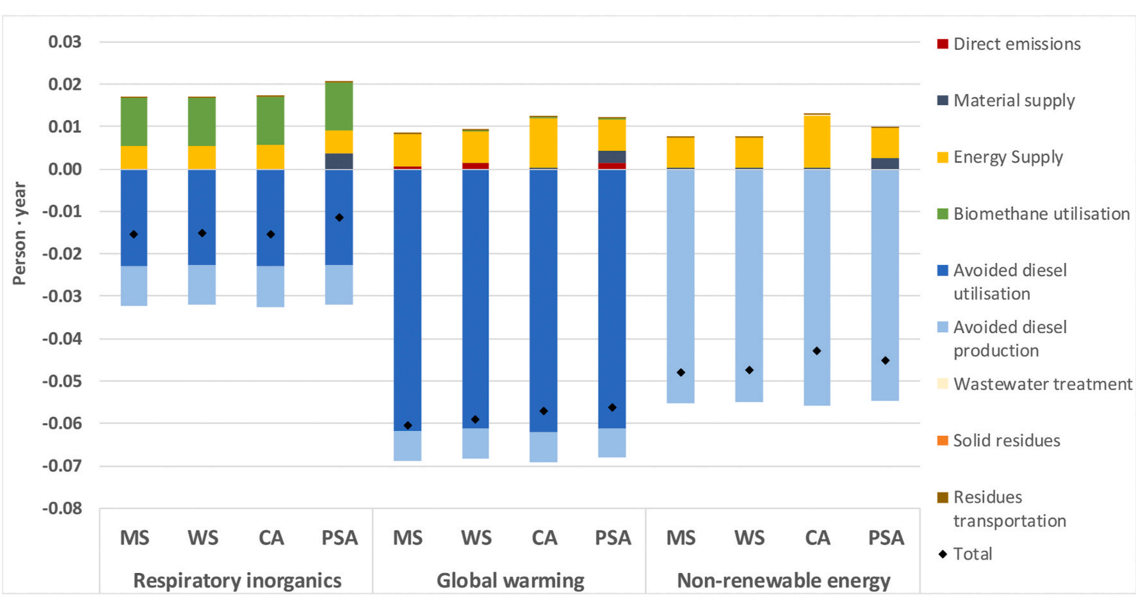

Fig. 5. Normalised results of impact assessment for the main impact categories (only base scenarios). The shaded rhombus represents the total value for each analysed technology. Results are expressed as "Person*year", i.e. the average impact in each midpoint category generated by a person during an entire year in Europe.

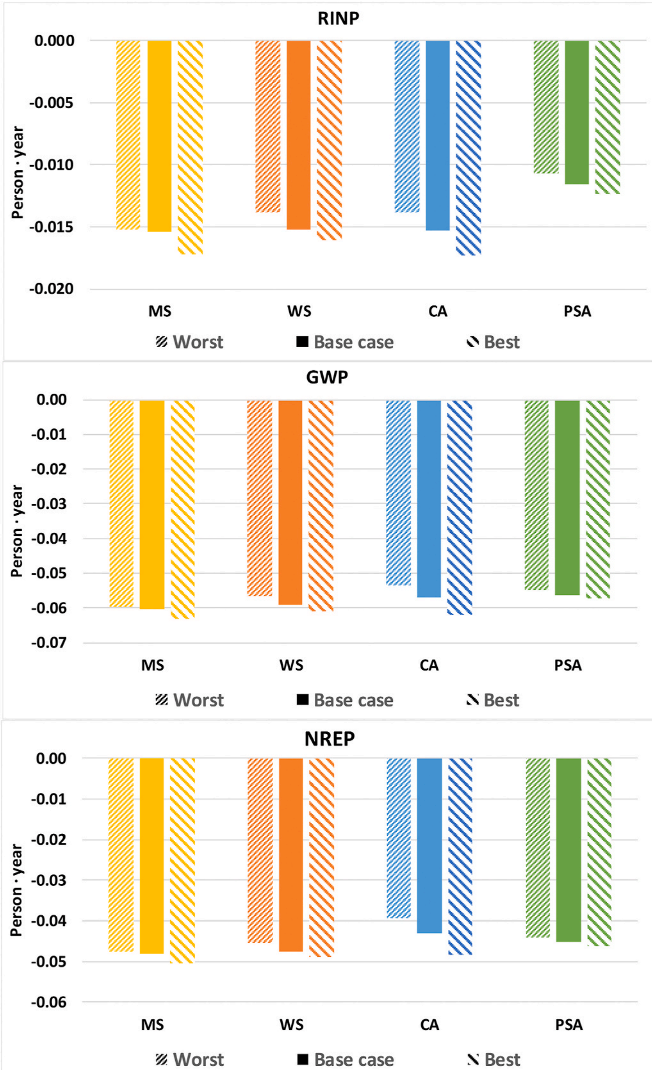

Fig. 6. Normalised results of impact assessment for all scenarios in the three impact categories of interest. Results are expressed as "Person* year", i.e. the average impact in each midpoint category generated by a person during an entire year in Europe.

zeolite and activated carbon. For NREP, there is a similar situation with large avoided impacts related to the diesel production phase, while direct impacts are determined by energy consumption (with higher values for CA, due to the thermal energy requirement) and material supply (for PSA only). Then, taking into account the contributional analyses reported in Figs. B5–B.16 of Annex B, the technique of

Table 5
Description of the sensitivity scenarios under analysis with the indication of the assumed values for each changed parameter.

ID_Name	Parameters	Value in sensitivity scenarios	Value in base case scenarios
5 bar 60 bar	Grid pressure at the injection point	5 bar 60 bar	24 bar
Worst methane slip Best methane slip	Methane slip	Worst value of each upgrading unit, shown in Table 3 Best value of each upgrading unit, shown in Table 3	Base value of each upgrading unit, as shown in Table 3
EU mix	Electricity mix	European, 2016	Italian, 2016

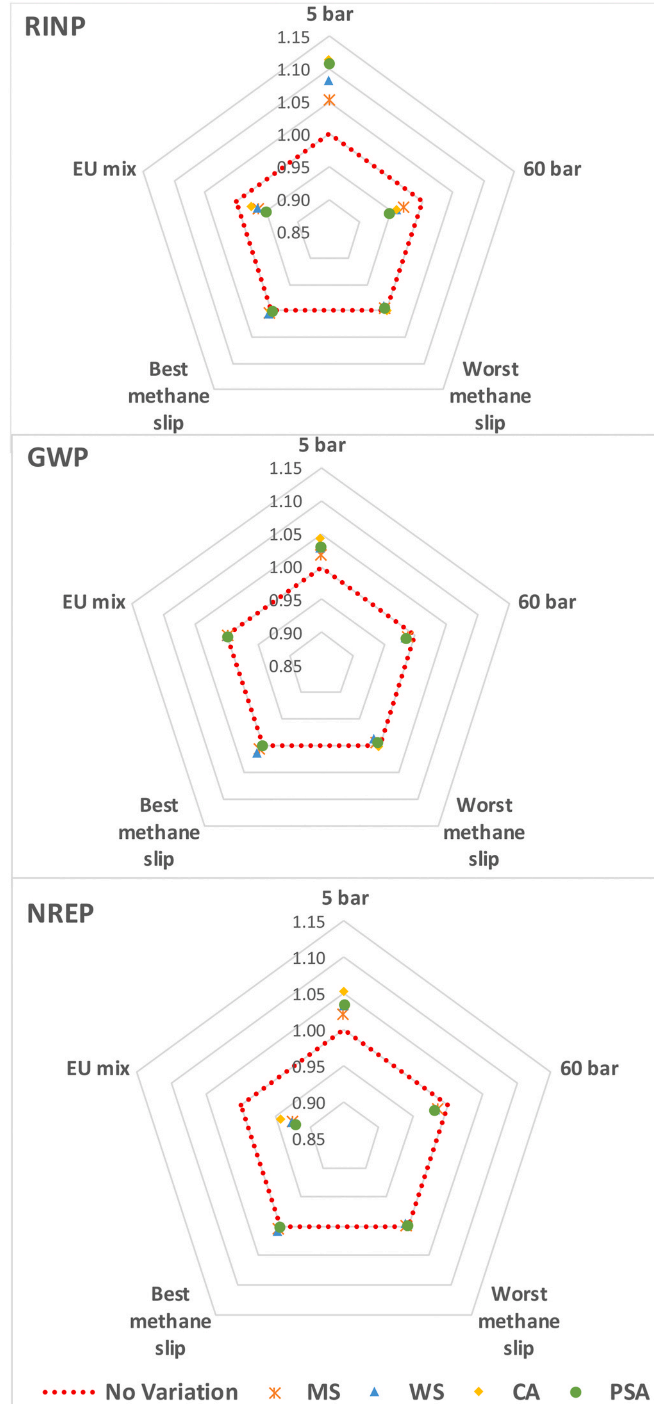

Fig. 7. Normalised results of impact assessment for sensitivity scenarios in the three impact categories of interest. Results are expressed in terms of a variation factor (VF), which is quantified as the ratio between the LCIA result obtained in the sensitivity scenario and that of the base case scenario (VF = 1 indicates no variation; some variations occur when VF is <1 or VF > 1).

membrane separation shows improvements for RINP (from 1% up to 34%, i.e. from 8.4 $kg_{PM2.5eq}$/y to 328 $kg_{PM2.5eq}$/y), for GWP (from 2% up to 7%, i.e. from about 101 t_{CO2eq}/y to 344 t_{CO2eq}/y) and for NREP (from 1% up to 12%, i.e. from about 604 $GJ_{primary}$/y to 6400 $GJ_{primary}$/y).

Fig. 6 refers to all the scenarios (base, worst and best) in each of the analysed impact categories, while further details can be found in Tables B.1-B.3 of Annex B. MS again shows the best performances, with improvements with respect to WS and CA that increase from 0.6% for the base case to more than 10% for the best or worst cases, and with respect to PSA that increase from about 34% for the base case to more than 42% for the best or worst cases. With reference to the selected impact categories, it can be noted that for GWP, the techniques of MS, WS and PSA show limited variation between base, best and worst scenarios (in a range of about 5% with respect to the base case), while CA shows a more remarkable improvement in the best case (up to about 9%) thanks to the reduction of thermal energy consumption (Table 4). This effect for Chemical Absorption also applies for NREP (up to about 12% of improvement in the best case) and RINP (up to about 13%) categories. For PSA, this is valid only for NREP while for RINP this technique shows the worst performances among all that examined.

3.4. Interpretation

Five alternative scenarios for the base case of each upgrading unit have been quantified in a sensitivity analysis [64]. The alternative scenarios have been obtained by changing three key parameters: pressure at the injection point of the natural gas grid, methane slip and energy mix (Table 5). In particular, with reference to the injection pressure, the alternative values of 5 bar and 60 bar have been chosen as representative of the lowest and highest injection pressures in the Italian natural gas grid [65].

Fig. 7 shows the results of the sensitivity analysis in terms of a variation factor (VF), defined as the ratio between the LCIA result for each scenario of the sensitivity analysis and the LCIA result obtained for the base cases (then VF = 1 indicates no variation). The sensitivity analysis show that the investigated parameters affect the base cases results only to a limited extent (VF always very close to 1, in the range of 0.90–1.11). The pressure of natural gas grid affects the electricity necessary to compress biomethane from the operating pressure of the upgrading unit to that required at the injection point. The consumption of electric energy is higher when the grid pressure is higher, and vice versa. In particular, the first compression steps (1–10 bar) are the most energetically expensive [19]. This parameter shows different effects on upgrading units based on their different operating pressure. For all midpoint categories under analysis, WS, CA and PSA show the highest improvements (in particular for RINP, VF > 1.05) with a low injection pressure (5 bar), because this value is near to their operating pressures (around 1 bar for CA and 4 bar for WS and PSA). For the same reasons, these technologies show also the largest worsening for a high injection pressure (60 bar). The obtained results show that the choice of the most suitable technique can be site-specific, as it depends on the conditions of the local gas network.

The variation of the methane slip affects both direct air emissions deriving from the off-gas and biomethane yield (and then travelled kilometres). The LCA results for this parameter show a limited relevance for all midpoint categories under analysis (VF very close to 1). Finally, electricity mix affects the direct impacts linked to the electricity supply. European electricity mix [52] has a lower share of non-renewable sources than the Italian one (35% vs 45%), implying higher direct impacts for all upgrading units, and then worse overall results. In particular, it affects all upgrading units in terms of RINP and NREP, with values of VFs of about 0.9. In summary, the sensitivity analysis shows that the selected key parameters have a rather limited effect on the obtained results, for the assumed ranges of variation. The biomethane injection pressure into the gas grid is certainly the most important, since it could affect the selection of the more suitable upgrading technique for each specific situation.

4. Environmental life cycle costing

4.1. Methodology

Three types of Life Cycle Costing analysis can be used to assess the economic performance of a product (good or service) throughout its entire life cycle. The Conventional LCC is "based on purely economic evaluation, considering various stages in the life cycle" [17], then it is close to the traditional financial assessments, accounting for marketed goods and services. It generally neglects external costs and does not always consider the complete life cycle. The Environmental LCC is a financial assessment too, but the costs derive by all the stakeholders involved in the life cycle of a product and it is the only one that can be developed in parallel to LCA. In fact, it utilises system boundaries and functional unit equivalent to those of the complementary LCA, and both are steady-state methods [16]. The unique feature of Societal LCC is the inclusion of externality costs: in other words, it takes into account environmental and social impacts by assigning monetary values to the respective effects. It is a stand-alone method, then is not coupled with a complementary LCA and/or social LCA [17].

An environmental LCC has been here implemented for all scenarios already examined in the LCA, by employing the same assumptions (such as functional unit, product systems, system boundaries and allocation procedure) and physical parameters (such as material and energy flows). The Equation (2) has been utilised to quantify the life cycle costs in terms of €/FU, i.e. euros related to the functional unit, as better detailed in the Annex C:

$$\textbf{Life Cycle Costs}\ (€/FU) = (\textbf{Upgrading}_{Costs} - \textbf{Biomethane}_{Revenues} + \textbf{Biomethane Utilisation}_{Costs} - \textbf{Diesel}_{Costs}),\ (€/FU) \qquad \text{Eq. 2}$$

In particular, the upgrading costs are those related to the biomethane production (i.e. investment and operating costs of each upgrading units), and have been calculated by Equations C.2-C.5 of Annex C. The investment costs have been allocated equally between the total flow rate of biogas treated by a specific technology during its economic lifetime, converting the initial budget costs into annuities as reported by Martinez-Sanchez et al. [16]. For all upgrading units, an economic lifetime of 20 years with an availability of 8400 h/y have been considered and an interest rate equal to 5% has been assumed, according to Ferella et al. [31]. The operating costs take into account the different contributions for each upgrading unit of general maintenance, required consumables and electric/thermal energy consumptions. The biomethane revenues are instead those deriving from biomethane selling and incentives, and have been calculated by Equations C.6-C.8 of Annex C, by considering the biomethane flow rate and its calorific value. The costs for biomethane utilisation have been calculated by Equations C.9-C.11 of Annex C, and are related to the infrastructures for biomethane distribution and its utilisation. They have been quantified as marginal costs with reference to those of diesel in the same type of vehicles, based on data reported by Ricardo-AEA [9]. Finally, the diesel costs have been calculated as avoided costs by Equation C.12 of Annex C. They relate to diesel production, considering the same distances travelled with biomethane, analogously to the allocation procedure adopted for the LCA. The main physical and economic parameters utilised for LCC calculations of base cases are shown in Table 6, together with the indications of their sources.

4.2. LCC results

LCC results are shown in Fig. 8 with reference to base case scenarios. The total life cycle costs are negative for all the analysed upgrading units

Table 6
Physical and economic parameters utilised for LCC calculations of base cases, with the indication of data source.

	MS	WS	CA	PSA	*Source of Data*
		PHYSICAL PARAMETERS			*LCI of Environmental LCA*
Biogas IN, m^3_N/h		500			
Biomethane OUT, m^3_N/h	259.0	256.0	257.3	255.2	
Biomethane LHV, MJ/m^3_N	34.89	35.01	35.32	35.00	
Biomethane utilisation, km	5682	5636	5716	5619	
		ECONOMIC PARAMETERS			
	Upgrading unit				[19,30,31,54,58]
Investment costs, €	1,200,000	1,700,000	1,500,000	1,150,000	
Investment costs, €/($m^3_{N\ biogas}$/h)	2400	3400	3000	2300	
Operating costs, €/y	240,000	217,000	273,000	280,000	
Operating costs, (€/y)/($m^3_{N\ biogas}$/h)	480	435	545	560	
	% of total operating costs				
Maintenance costs	*14.4*	*5.5*	*21.8*	*12.4*	
Consumable costs	*8.2*	*9.1*	*18.1*	*21.3*	
Energy costs	*77.4*	*85.4*	*60.1*	*66.3*	
Biomethane selling, €/kWh		13.54			[53]
Biomethane incentive, €/CIC[a]		375			[53]
Diesel production, €/km		0.021			[9]
	Marginal Costs Biomethane - Diesel				
Vehicles, €/km		0.001			[9]
Infrastructure, €/km		0.01			

[a] CIC are tradable certificates based on the quota obligation for fossil fuel traders (1 CIC is released for each 5 Gcal of biomethane produced starting from OFMSW).

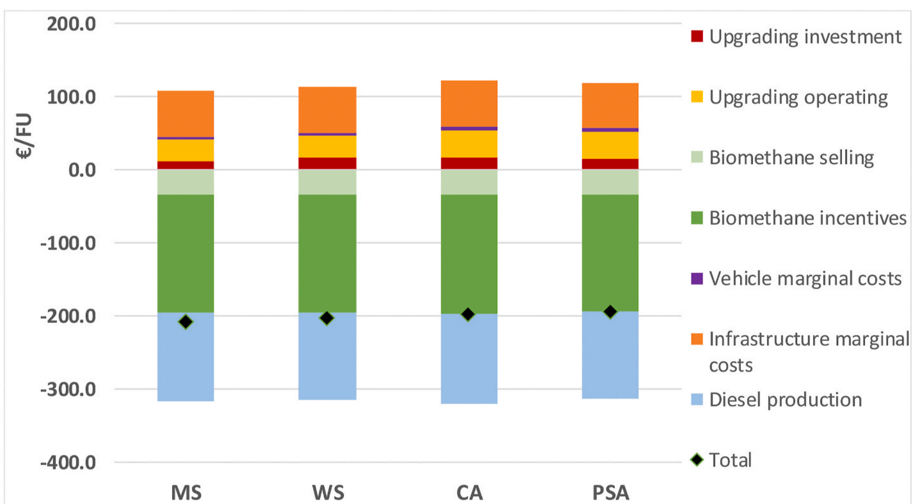

Fig. 8. LCC results for base cases of the upgrading technologies under analysis, with the contributions of different life cycle stages.

(varying from −203 €/FU to −210 €/FU for PSA and MS, respectively), indicating that biomethane revenues and diesel avoided costs are always larger than direct and indirect costs. This means that the biogas-to-biomethane upgrading stage can provide a life cycle saving of more than 1764 k€/y, while the difference between the analysed techniques can reach a value of about 60 k€/y. It is noteworthy that, for each upgrading unit, the contributions of the investment costs (varying from 11 €/FU to 16 €/FU for PSA and WS, respectively) and maintenance ones

Table 7
LCC results for best and worst scenarios of the upgrading technologies under analysis, together with the indication of modified economic parameters utilised as input.

	MS			WS			CA			PSA		
	Worst	Base	Best	Worst	Base	Best	Worst	Base	Best	Worst	Base	Best
INPUT PARAMETERS												
Specific investment costs, €/($m^3_{N\ biogas}$/h)	2500	2400	2200	3500	3400	2600	3500	3000	2600	3200	2300	2300
Specific operating costs, (€/y)/($m^3_{N\ biogas}$/h)	491	480	360	567	435	380	624	545	435	615	560	505
% operating costs												
Maintenance costs, %	*14.1*	*14.4*	*18.6*	*12.2*	*5.5*	*6.2*	*19.0*	*21.8*	*27.2*	*11.3*	*12.4*	*13.7*
Consumable costs, %	*8.1*	*8.2*	*10.6*	*7.0*	*9.1*	*10.4*	*15.8*	*18.1*	*22.7*	*19.4*	*21.3*	*23.7*
Energy costs, %	*77.9*	*77.4*	*67.6*	*80.8*	*85.4*	*83.4*	*65.1*	*60.1*	*50.1*	*69.3*	*66.3*	*62.7*
Biomethane revenues, €/m^3_N	0.75	0.76	0.76	0.75	0.76	0.76	0.76	0.77	0.77	0.76	0.76	0.76
LCC RESULTS												
Life Cycle costs, €/FU	**−208**	**−210**	**−218**	**−198**	**−206**	**−214**	**−197**	**−205**	**−213**	**−195**	**−203**	**−206**

(varying from 26 €/FU to 33 €/FU for WS and PSA, respectively) are limited with reference to the total life cycle costs. Biomethane revenues have a crucial role, with a total value of about 200 €/FU for each upgrading unit, mainly thanks to the incentives, which are about 160 €/FU. The comparison between the upgrading units shows that differences are so limited (always lower than 4%) that they cannot be considered significant, since market dynamics are affected by several factors and strategies, which can lead to different bids. LCC results for worst and best scenarios are shown in Table 7, together with the indication of different economic parameters (investment and operating costs as well as biomethane revenues) utilised for the calculations. Analogously to the base scenarios, physical parameters (such as biomethane flow rate, LHV and utilisation) adopted for LCC calculations are those obtained from LCI of best and worst scenarios. Obtained results confirm that there are no significant differences between the life cycle costs of the upgrading units, which range between −195 €/FU for the worst scenario of PSA and −218 €/FU for the best scenario of MS.

A comparative analysis of the results obtained by the combined

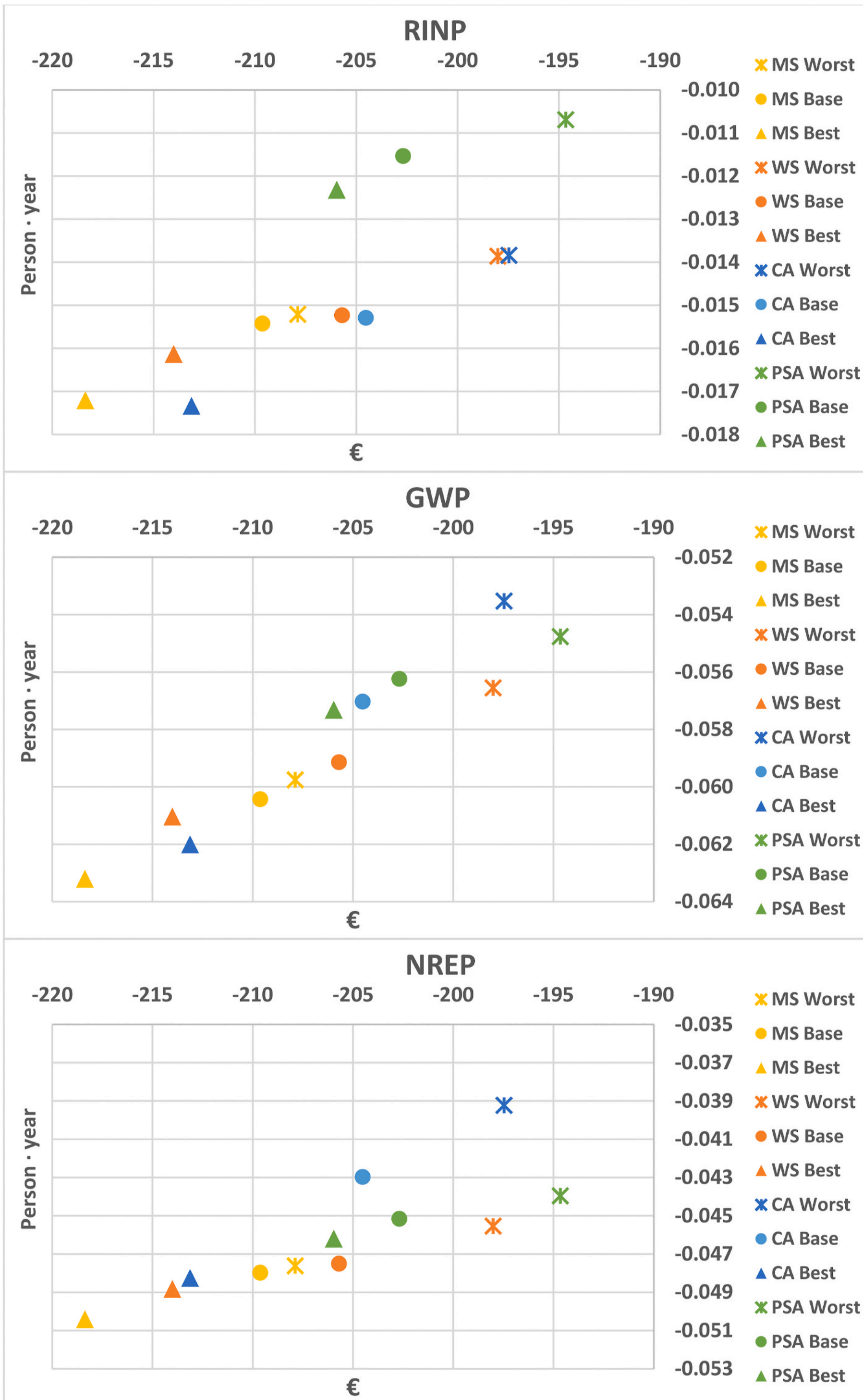

Fig. 9. Combined results of LCA (in terms of normalised LCIA data) and LCC, for all the base, best and worst scenarios in the three impact categories of interest. All values refer to the functional unit.

utilisation of LCA and LCC has been carried out in order to individuate the win-win situations, that is the scenarios that better balance the environmental and economic performances along the whole live cycle of the considered system. Fig. 9 reports these combined results of LCA (in terms of normalised LCIA data) and LCC, for all the base, best and worst scenarios for the three impact categories of interest. Data are all negative, indicating the positive environmental and economic effect of the analysed configurations. The win-win situations are those in the left and lower part of the diagram. It appears that the membrane separation technique (in its best, but also base and worst, configurations) provides some advantages with respect to all the other techniques.

5. Conclusions

The study reviews and compares environmental and economic aspects of the most utilised techniques to obtain high quality biomethane by upgrading biogas from anaerobic digestion of the organic fraction of municipal solid waste.

Three scenarios (base, worst and best) for each of selected upgrading techniques (membrane separation, water scrubbing, chemical absorption with amine solvent, and pressure swing adsorption) have been assessed by means of the combined utilisation of an attributional environmental Life Cycle Assessment and a complementary environmental Life Cycle Costing.

The results are negative for the main environmental impact categories (GWP, NREP and RINP) as well as for the life cycle costs, highlighting that all the examined upgrading technologies imply a substantial reduction of the overall environmental and economic impacts. These good performances over the whole life cycle are confirmed by the quantification of the best and worst scenarios and by the related sensitivity analysis, both showing very limited variations from the base case results.

The combined results of LCA and LCC analyses indicate the win-win situations, which all suggest that membrane separation provides the best performances. With reference to base case scenarios, this technique shows the largest improvements for RINP (from 1% up to 34%, i.e. from 8.4 $kg_{PM2.5eq}$/y to 328 $kg_{PM2.5eq}$/y) together with better performances for GWP (from 2% up to 7%, i.e. from about 101 t_{CO2eq}/y to 344 t_{CO2eq}/y), NREP (from 1% up to 12%, i.e. from about 604 $GJ_{primary}$/y to 6400 $GJ_{primary}$/y) and life cycle costs (from 1.9% up to 3.4%, i.e. from 34 k€/y to about 60 k€/y). Pressure swing adsorption shows worse performances for all the examined categories, mainly due to activated carbon and zeolite consumptions.

In any case, the selection of the best upgrading technique has to take into careful account some factors, such as site-specific conditions (like the injection pressure in the natural gas grid or the existence and the amount of local economic incentives) and commercial strategies for the market of interest.

Future work will be focused to extend the combined LCA/LCC analysis to other emergent technologies that have so far a too low Technology Readiness Level to be considered in this study.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2020.110588.

Author contribution

Filomena Ardolino: Conceptualization; Methodology; Investigation; Software; Writing-Original Draft. **Giovanni Francesco Cardamone:** Investigation; Data curation; Visualization; Writing-Original Draft. **Francesco Parrillo:** Investigation; Data curation; Visualization; **Umberto Arena:** Conceptualization; Validation; Supervision; Writing-Reviewing and Editing.

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