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An environmental assessment of green gases production routes: biogas-to-biomethane vs biogas-to-hydrogen

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Abstract: To meet the 1.5°C global warming limit, net-zero emissions targets have been set all over the world. In this context, two green gases, i.e., biomethane and hydrogen, are of utmost relevance. Biomethane is generally obtained through the upgrading of biogas produced by anaerobic digestion. It represents a renewable alternative to natural gas and has several applications in sectors such as transport and energy. Similarly, hydrogen is a key element for decarbonising the global economy. It has applications in sectors such as energy, transport, and construction. Most hydrogen is produced from non-renewable sources generating significant emissions. Therefore, finding alternatives to produce low-carbon hydrogen is an urgent challenge. Although the "green hydrogen" route (i.e., electrolysis fuelled by electricity from renewable sources) is very promising from an environmental perspective, its high electricity consumption represents a barrier to its large-scale implementation. In this regard, the so-called "steam biogas reforming" route represents a viable alternative. It consists of producing hydrogen from biogas obtained from anaerobic digestion. Hydrogen production from biogas replaces the production of biomethane. To this concern, this paper aims to identify the best green gas production route among biogas-to-biomethane and biogas-to-hydrogen from an environmental point of view. Consistent with this purpose, an analytical model was developed to assess each alternative based on the direct, indirect, and avoided emissions. The results showed that the decarbonisation of the green gas production routes and the environmental convenience of either alternative is strongly affected by multiple aspects related to the energetic assets of the country considered.

Keywords: environmental assessment, decarbonization, biogas, biomethane, hydrogen, anaerobic digestion.

I. INTRODUCTION

In the current context of climate change tackling and energy crisis, the identification of renewable-based fuels and energy vectors is a key issue [1]–[3]. To this concern, green (i.e., low carbon) gases such as biomethane (bio-CH_4) and hydrogen (H_2) are of utmost relevance. They are indeed key elements for reaching the "net-zero emissions" targets set worldwide to meet the Paris Agreement's 1.5°C global warming limit [4]–[6].

Bio-CH_4, also known as "renewable natural gas", is a near-pure source of methane (CH_4). It is generally produced by upgrading (i.e., by removing CO_2 and other contaminants) the biogas (BG) obtained from the anaerobic digestion (AD) of organic wastes (OWs) or biomasses [7]. Bio-CH_4 has almost the same LHV as fossil natural gas (around 36 MJ/m^3 [8]) and find its same several applications (e.g., electricity and heat production, and as a fuel in the transport sector) [9]. Most of all, Bio-CH_4 can be used with no changes in transmission and distribution infrastructure or end-user equipment with respect to the natural gas [8]. This green gas has therefore the potential for meeting the requirements of natural gas-based applications with the same effectiveness as the fossil fuel, but without the associated emissions. Emissions generated from the use of fossil natural gas for electricity and heat production were indeed 3.2 $GtCO_2$ in 2021 [10], a value not consistent with a decarbonized scenario. Moreover, Bio-CH_4 could be helpful in reducing the emissions from the transport sector, which reached 7.7 $GtCO_2$ in 2021. Consistent with the "net-zero" scenario, indeed, emissions from this sector must be reduced by 20% by 2030 [11]. It is noteworthy that also the main Bio-CH_4 production process, i.e., AD of organic wastes (OWs), shows a great environmental potential. This process allows to valorize wastes, at the same time reducing methane emissions mainly from OWs' decomposition and agriculture [12], which generated 1.49 $GtCO_{2eq}$ and 3.49 $GtCO_{2eq}$ in 2019, respectively [13]. CH_4 is indeed the second major greenhouse gas after CO_2. Although it persists in nature for fewer years than CO_2 (i.e., 12 years compared to centuries for CO_2), CH_4 has the capacity to absorb much more energy, generating 28-36 times more impact on global warming over a 100-year time horizon [14]. Consistently, CH_4 emissions reduction can provide significant climate benefits in the near-term [15]. It can be therefore stated that the production of Bio-CH_4 has a threefold decarbonization potential; it is useful to reduce CO_2 emissions from the energy, industry and transport sectors, it helps reducing the fugitive methane emissions (FMEs) generated from natural gas supply, and it helps reducing CH_4 emissions from waste and agriculture sectors. Actions and strategies to foster the widespread use of Bio-CH_4 are indeed subject of current studies. To this concern, in [16], starting from the economic and environmental potentials of Bio-CH_4, the opportunities and barriers for the implementation of a European Bio-CH_4 market are analyzed. In a previous study, starting from the experience of European countries, the large-scale development and drivers of BG and Bio-CH_4 production are explored. At the same time, issues of future interest such as policy recommendations and supply chain risks are analyzed [17]. Similarly, by considering the Bio-CH_4 as a virtuous example of circular bioeconomy, in [18] a framework for evaluating Bio-CH_4 communities is proposed. As for the optimization of the bio-CH_4 production process, in [19] the integration of AD with hydrothermal gasification is proposed, in order to maximize the bio-CH_4 yield. In [20] the AD-based bio-CH_4 production process is analyzed in order to identify the operational variables that most affect the greenhouse gases emissions from the process. Similarly, in [21] the environmental impacts associated with bio-CH_4 production from AD are assessed through a Life Cycle Assessment methodology. Process optimizations are also provided in [22]–[25].

Along with bio-CH_4, H_2 is a promising energy vector that plays a key role in decarbonizing the global economy [26]. Indeed, it has an energy density about three times higher than gasoline [27] and its combustion generates water vapour only [28]. H_2 is already employed in many industrial processes (e.g., in crude oil refining, and in the ammonia production [26]) and, due to its high environmental and energy potential, it is being considered in many innovative applications. Indeed, H_2 is currently proposed as a low-carbon fuel and energy vector in the transport sector, in the building sector, and in the power generation sector [29]. H_2 adoption, moreover, is considered as the most effective decarbonization solution for the so-called "hard-to-abate" sectors (e.g., iron and steel, cement and concrete, chemicals, etc.) [30]. This strong interest in innovative H_2 applications will lead to an exponential growth in hydrogen demand. To this concern, the global H_2 demand, which was 75 Mt/y in 2019, will increase by 593%, reaching 520 Mt/y by 2070 [31]. However, environmental concerns arise when considering the main H_2 sources. Indeed, almost all the H_2 produced currently comes from unabated fossil fuels, generating 900 $MtCO_2$/y [32]. Therefore, the large-scale adoption of environmentally sustainable processes is crucial to achieve the expected decarbonization goals. H_2 produced through water electrolysis fuelled by renewable electricity, also known as “green hydrogen”, is the most promising alternative from an environmental perspective [33]. It is indeed zero emissions [34]. However, the large-scale implementation of this technology faces major barriers mainly from an environmental point of view. Indeed, electrolyzers are characterized by very high energy demand (about 5 kWh/Nm^3H_2 [35]), and there is currently not enough renewable electricity to produce large amounts of green H_2. Therefore, huge indirect emissions would be generated by the supply of electricity from the national power grid. For large-scale green H_2 production, it is therefore necessary to accelerate the current energy transition phase, in order to increase the availability and reduce the cost of renewable electricity [36]. During this transition phase, an interesting hydrogen production process could be the so-called Steam Biogas Reforming (SBR) route. It consists of producing H_2 from the dry reforming of the biogas (i.e., the reactions between CH_4 and H_2O, and CO_2 and H_2O to obtain syngas [37]) produced by the AD treatment of OWs and biomasses [38]. This H_2 production route has a threefold benefit; it allows producing low-carbon H_2, at the same time valorizing wastes and reducing the H_2 dependency on fossil sources. These features make this process as an enabler for the development of a low-carbon H_2 market. To this concern, many studies are currently focusing on this topic. In [39] the potential of the SBR process is illustrated from the ecological, economic, and environmental perspective. Similarly, in [40] the effectiveness of the SBR route is highlighted focusing on the reduction of fossil natural gas consumption. In [41], the SBR route is analyzed from a technical perspective by investigating the effect of BG composition on the performance of the process. Although the SBR route has multiple environmental benefits, it is noteworthy that the production of bio-CH_4 by biogas upgrading is being foregone in this scenario with all the related environmental benefits. To this concern, the objective of the present paper is to identify the best green gas production route among the biogas-to-biomethane (BG-bio-CH_4), i.e., AD with biogas upgrading, and biogas-to-hydrogen (BG-H_2), i.e., SBR process, from an environmental point of view. Consistent with this purpose, an analytical model was developed to assess each alternative based on total greenhouse gases emissions.

The rest of the paper is organized as follows: in section 2 the plant configurations considered for the BG-bio-CH_4, and BG-H_2 routes are described, and the developed analytical model is illustrated. In section 3 the results obtained from the numerical application of the model are provided and discussed. Finally, in section 4 the conclusions of the work are provided with insights for future studies.

II. MATERIAL AND METHODS

This section describes the plant configurations considered for the BG-bio-CH_4 (Fig. 1) and BG-H_2 routes (Fig. 2). The analytical model developed to compare them is subsequently described along with the data used in the numerical application.

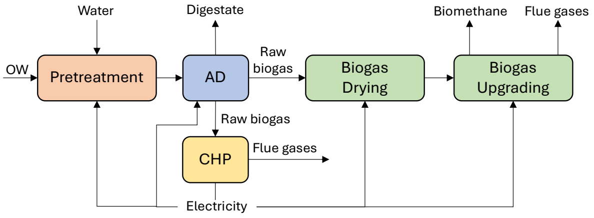

Figure 1. Plant configuration considered for the BG-bio-CH_4 route.

The configuration considered for the BG-bio-CH_4 route was adapted from [42] (Figure 1). OWs are first subjected to a mechanical pretreatment to remove substances not compatible with AD. Next, the substrate undergoes AD treatment in a mesophilic temperature regime (i.e., 37-39°C). The chemical stabilization process produces two main by-products, i.e., BG and digestate (i.e., the solid by-product). The digestate is removed from the system for disposal in landfills or recovered as soil fertilizer; it is noteworthy that the management of this by-product was not considered within the scope of this work. About 70% of the raw BG is then sent to an upgrading unit and the remainder to a CHP unit to partially satisfy the needs of the pretreatment unit, AD reactor, and upgrading unit (the remaining energy demand is satisfied by the supply from the national grid). The BG sent to the upgrading unit undergoes a preliminary stage of drying and compression, and H_2 sulfide removal. Finally, the purified BG undergoes the CO_2 removal process through a three-stage membrane separation system with an efficiency of 98%, thus obtaining bio-CH_4.

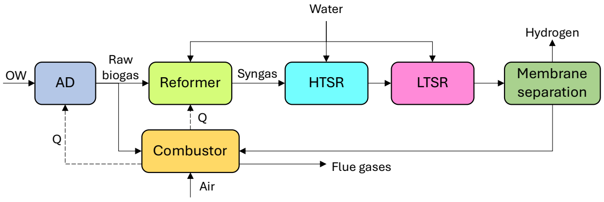

Figure 2. Plant configuration considered for the biogas-to-hydrogen route.

The plant configuration considered for the BG-H_2 route is adapted from [43] (Figure 2). The BG obtained from AD treatment is for the main share (i.e., almost 74%) sent to a catalytic reformer, and for the remaining share to a combustor to provide the heat to sustain the endothermic reactions occurring in the process. Within the reforming reactor, the BG undergoes the so-called "dry reforming" process. It consists of reacting the BG with water vapor to obtain syngas, mainly composed of H_2 and CO. The obtained syngas then undergoes a water-gas shift reaction within the high-temperature (i.e., 300-400°C) and low-temperature (i.e., 200-300°C) shift reactors (HTSR and LTSR, respectively). The objective of the water-gas shift reaction is to increase the H_2 content in the syngas. Finally, the gas mixture is subjected to a CO_2 separation process using a membrane system, obtaining >99%vol pure H_2. The gas obtained from the separation unit, containing unreacted CH_4, is sent to the combustor, while the exhaust gas from the combustor is sent to the AD reactor to maintain the reaction temperature before being released into the atmosphere.

The developed environmental analytical model allows to evaluate the total emissions associated with each green gas production route. It is expressed according to equation 1.

$$\varphi\left[\mathrm{kgCO}_{2_{eq}}/t\mathrm{OW}\right] = \varphi_{\mathrm{indirect}} - \varphi_{\mathrm{av}} \qquad (1)$$

$\varphi_{\mathrm{indirect}}\left[\mathrm{kgCO}_{2_{eq}}/t\mathrm{OW}\right]$ are the emissions associated with electricity supply from the national grid (equation 2) and $\varphi_{\mathrm{av}}\left[\mathrm{kgCO}_{2_{eq}}/t\mathrm{OW}\right]$ are the avoided emissions provided by each green gas production route (equation 3). As it can be observed from equation 1, direct emissions were neglected. This is because both green gas routes generate biogenic emissions only, which are considered to be carbon-neutral [44].

$$\varphi_{\mathrm{indirect}}\left[\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{tOW}\right] = \mathrm{EL}_{\mathrm{cons}} \cdot \mathrm{f}_{\mathrm{grid}} \tag{2}$$

Indirect emissions were calculated as the product between the electricity consumption of the considered process ($\mathrm{EL}_{\mathrm{cons}}$ [kWh/tOW]) and the national grid emission factor ($\mathrm{f}_{\mathrm{grid}}$ [$\mathrm{kgCO}_{2_{\mathrm{eq}}}$/kWh]). It is noteworthy that the value of the $\mathrm{f}_{\mathrm{grid}}$ variable directly reflects the composition of the national energy mix.

$$\varphi_{\mathrm{av}}\left[\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{tOW}\right] = \varphi_{\mathrm{av}_{\mathrm{CH}_4}} + \varphi_{\mathrm{av}_{\mathrm{H}_2}} \tag{3}$$

Avoided emissions were calculated as the sum between avoided emissions from bio-CH_4 production ($\varphi_{\mathrm{av}_{\mathrm{CH}_4}}$ [$\mathrm{kgCO}_{2_{\mathrm{eq}}}$/tOW] and avoided emissions from H_2 production ($\varphi_{\mathrm{av}_{\mathrm{H}_2}}$[$\mathrm{kgCO}_{2_{\mathrm{eq}}}$/tOW]). They are expressed according to equations 4 and 5, respectively.

$$\varphi_{\mathrm{av}_{\mathrm{CH}_4}}\left[\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{tOW}\right] = \eta_{\mathrm{bio}_{\mathrm{CH}_4}} \cdot \mathrm{HHV}_{\mathrm{CH}_4} \cdot \eta_{\mathrm{pow}} \cdot \mathrm{f}_{\mathrm{pow}} \tag{4}$$

Avoided emissions from bio-CH_4 production were considered as the emissions that would be generated from electricity production by employing fossil natural gas. They were calculated as the product between the bio-CH_4 yield of the process ($\eta_{bio_{CH_4}}$[kgCH_4/tOW]), the higher heating value of natural gas ($\mathrm{HHV}_{\mathrm{CH}_4}$ [kWh/kgCH_4]), the power efficiency of a natural gas power plant (η_{pow} [%]) and the emission factor of a natural gas-based power production process ($\mathrm{f}_{\mathrm{pow}}$ [$\mathrm{kgCO}_{2_{\mathrm{eq}}}$/kWh]).

$$\varphi_{\mathrm{av}_{\mathrm{H}_2}}\left[\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{tOW}\right] = \varphi_{\mathrm{av}_{SMR}} + \varphi_{\mathrm{av}_{el}} \tag{5}$$

Avoided emissions from H_2 production were calculated as the sum between avoided emissions from fossil-based H_2 production ($\varphi_{\mathrm{av}_{\mathrm{SMR}}}$ [$kgCO_{2_{eq}}/tOW$]) and electrolysis based H_2 production (equation 7). The overall amount of H_2 produced by the BG-H_2 route (η_{H_2} [$\mathrm{Nm}^3\mathrm{H}_2$/tOW] is weighted according to the share of global H_2 production from electrolysis (α [%]). It is noteworthy that emissions from the Steam Methane Reforming (SMR) process, i.e., currently the most widespread H_2 production route [45], were considered in the case of fossil-based production process. They are expressed according to equation 6.

$$\varphi_{\mathrm{av}_{SMR}}\left[\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{tOW}\right] = (1-\alpha) \cdot \eta_{\mathrm{H}_2} \cdot (\varphi_{\mathrm{d}_{\mathrm{SMR}}} + \mathrm{EL}_{\mathrm{cons}_{\mathrm{SMR}}} \cdot \mathrm{f}_{\mathrm{grid}} + \mathrm{NG}_{\mathrm{cons}_{\mathrm{SMR}}} \cdot \mathrm{FME} \cdot \mathrm{GWP}_{100}) \tag{6}$$

Emissions from the SMR process were calculated as the sum between direct emissions $\varphi_{\mathrm{d}_{\mathrm{SMR}}}$[$\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{Nm}^3\mathrm{H}_2$]), indirect emissions from electricity supply ($\mathrm{EL}_{\mathrm{cons}_{\mathrm{SMR}}} \cdot \mathrm{f}_{\mathrm{grid}}$ [$\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{Nm}^3\mathrm{H}_2$] and the FMEs generated from natural gas supply. They were calculated as the product between the natural gas consumption of the process ($\mathrm{NG}_{\mathrm{cons}_{\mathrm{SMR}}}$ [$\mathrm{kgCH}_4/\mathrm{Nm}^3\mathrm{H}_2$]), the factor which quantifies the amount of FMEs for each unit mass of CH_4 consumed (FME [#]) and the CH_4's impact factor on the global warming potential over a time horizon of 100 years (GWP_{100} [$\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{kgCH}_4$].

$$\varphi_{\mathrm{av}_{el}}\left[\mathrm{kgCO}_{2_{\mathrm{eq}}}/\mathrm{tOW}\right] = \alpha \cdot \eta_{\mathrm{H}_2} \cdot \mathrm{EL}_{\mathrm{cons}_{\mathrm{el}}} \cdot \mathrm{f}_{\mathrm{grid}} \tag{7}$$

Emissions from electrolysis were finally calculated as the product between the electricity consumption of the process ($\mathrm{EL}_{\mathrm{cons}_{\mathrm{el}}}$[$\mathrm{kWh}/\mathrm{Nm}^3\mathrm{H}_2$] and the national grid emission factor.

The developed analytical model was then numerically applied to the current scenario. In this regard, the global average values of the grid emission factor and the share of hydrogen production from electrolysis were employed. Table 1 shows the data used in the analysis.

TABLE I. DATA EMPLOYED FOR THE NUMERICAL APPLICATION OF THE DEVELOPED ANALYTICAL MODEL

Variable	BG-bio-CH₄	BG-H₂
EL_{cons}	48.7 [42]	32.34 [43]
f_{grid}	0.342 [46]	
η_{bioCH_4}	32.72 [42]	-
HHV_{CH_4}	15.4	-
η_{pow}	60 [47]	-
f_{pow}	0.506 [48]	-
FME	-	3.5% [49]
GWP_{100}	-	32 [50]
α	-	0.04 [45]
η_{H_2}	-	215.6 [43]
$\varphi_{d_{SMR}}$	-	0.91 [51]
$EL_{cons_{SMR}}$	-	0.12 [52]
$NG_{cons_{SMR}}$	-	0.3 [52]
$EL_{cons_{el}}$	-	6 [51]

III. RESULTS AND DISCUSSIONS

The results obtained from the numerical application of the developed analytical model are illustrated in Figure 3.

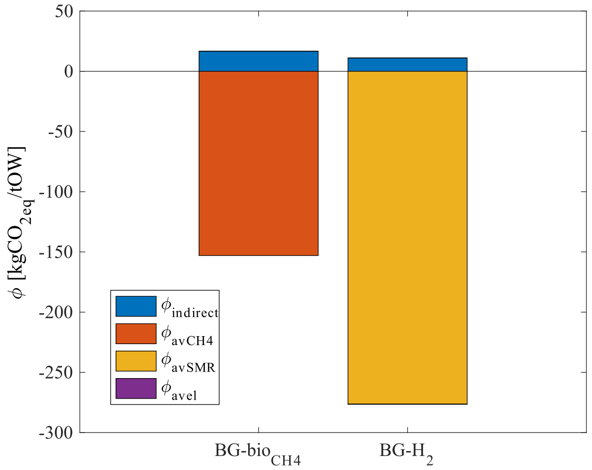

Figure 3. Total emissions for the BG-bio-CH₄ and BG-H₂ routes.

As it can be observed, both green gas production routes provide an environmental benefit, i.e., negative overall emissions. The alternative with a better overall balance is the BG-H₂ route. It has total emissions of -285.5 kgCO$_{2eq}$/tOW. This value is lower than the total emissions of BG-bio-CH₄ route (i.e., -136.32 kgCO$_{2eq}$/tOW). As for the BG-H₂ route, the major contribution is provided by avoided emissions from fossil-based H₂ production ($\varphi_{av_{SMR}}$= -276.38 kgCO$_{2eq}$/tOW). In the BG-bio-CH₄ route, avoided emissions from power generation from fossil natural gas ($\varphi_{av_{CH_4}}$) are -153 kgCO$_{2eq}$/tOW, a value lower than avoided emissions from the BG-H₂ route. It is noteworthy that the contribution provided by avoided emissions from H₂ production by electrolysis ($\varphi_{av_{el}}$) is almost negligible. This is because this alternative is currently barely employed in the H₂ production mix (α=0.04%). It can be therefore concluded that, although H₂ yield (i.e., 19.38 kgH2/tOW) is lower than bio-CH₄ yield (i.e., 32.72 kgbio-CH4/tOW) in the considered processes, BG-H₂ route currently shows a higher decarbonization potential.

Since, as pointed out, the major barrier to large-scale implementation of green H₂ relates to indirect emissions generated by the need of national grid electricity supply, a sensitivity analysis of total emissions was conducted with respect to the f_{grid} and α variables. The objective was to jointly capture any effects caused by an energy transition (i.e., decreasing f_{grid} values) and changes in the H₂ production mix (i.e., increasing α values). The results obtained are represented in figure 4.

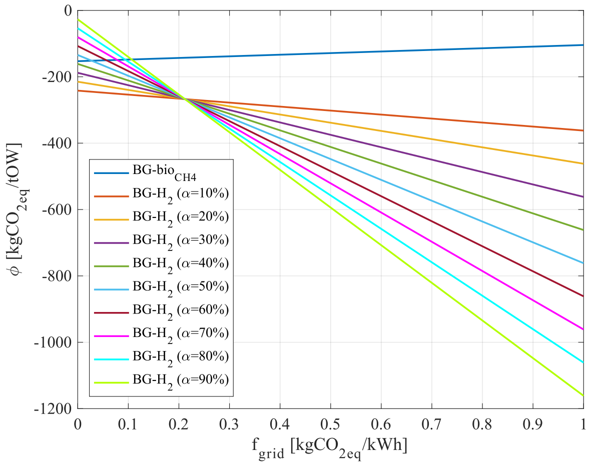

Figure 3. Sensitivity analysis with respect to the f_{grid} and α variables.

As it can be observed, as f_{grid} value increases, the total emission functions related to the two green gas production routes have an opposite trend. To this concern, the environmental benefit provided by the BG-bio-CH₄ route decreases as emissions from the national power grid increase. Indeed, the emissions generated by electricity consumption become greater than the avoided emissions. On the contrary, the environmental benefit provided by the BG-H₂ route increases as both the f_{grid} and α variables increase. It can also be observed that emissions from BG-H₂ route are equal for each value of α at f_{grid}=0.21[kgCO$_{2eq}$/kWh]. For lower values of f_{grid}, it is observed that the lowest emissions are recorded at minimum α (10%), and for higher values the highest emissions are recorded at maximum α (90%). This result highlights the BG-H_2 route's high decarbonization potential in the current transition phase. Once decarbonization targets will be met, this environmental benefit will be reduced, given the advantage provided by the green electrolysis route. Finally, as it can be observed, for α values greater than 50% and f_{grid} values up to 0.11 kgCO2eq/kWh, there are intersections between the total emission curves relative to the two green gas production routes. This implies that, in a decarbonized scenario, also the BG-bio-CH_4 route will represent a viable alternative.

IV. CONCLUSIONS

The objective of the present work was to identify the best green gas production route among BG-bio-CH_4 and BG-H_2 from an environmental point of view. To this concern, an analytical model was developed to assess each alternative based on total (i.e., direct, indirect and avoided) greenhouse gases emissions. The results obtained from the numerical application of the model to the current scenario, showed that the BG-H_2 route offers the best decarbonization potential. Indeed, H_2 production from SBR process provides an environmental benefit of -285.5 $kgCO_{2eq}$/tOW, unlike the BG-bio-CH_4 route, which offers a benefit of -136.32 $kgCO_{2eq}$/tOW. As for the BG-H2 process, the major contribution is provided by avoided emissions from fossil-based H_2 production. It is noteworthy that avoided emissions from the electrolysis process are almost negligible, due to the near absence of this route within the current H_2 production mix. A sensitivity analysis also allowed to understand that the decarbonization potential of the BG-H_2 route increases as emissions from the national power grid increase, in contrast to the BG-bio-CH_4 route. Moreover, it was possible to conclude that the BG-H_2 route offers a real decarbonization potential in the current energy transition phase, but this potential will be reduced when emissions from the grid will decrease, and electrolysis will turn out to be environmentally convenient. Finally, it was found that the BG-bio-CH_4 route could be effective in some decarbonized scenarios. Although the developed model is a useful tool for the evaluation of green gas production routes, the present work shows some limitations. In this regard, further green gas production routes such as gasification are not considered. Moreover, the effect of operational variables on bio-CH_4 and H_2 yields is neglected. Avoided emissions from bio-CH_4 production can also be further investigated by assessing emissions generated by the transportation sector. Future studies can then focus on overcoming these limits, also considering the economic aspect of the problem.

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