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From Secondary Biomass to Bio-Methanol through CONVERGE Technology: An Environmental Analysis

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Abstract: Owing to residual biomass availability, the share of advanced biofuels produced from secondary biomass is forecasted to increase and significantly contribute towards achieving net-zero emissions. The current work investigates bio-methanol production through a new process configuration designed to improve the environmental performance when compared to the state-of-the art technologies (Base Case). The environmental evaluation is conducted according to the Life Cycle Assessment (LCA) methodology. ReCiPe was employed as an impact assessment method with the aid of GaBi software. Depending on the plant geographical location, wooden biomass and exhausted olive pomace were evaluated as biomass sources. A scenario analysis targeting different energy sources was performed as well. The outcome of the environmental evaluation highlights a better performance in eight of a total of nine impact categories studied in the wooden biomass scenarios compared to the exhausted olive pomace. Moreover, two of the CONVERGE technology cases were compared against the Base Case. As the results show, CONVERGE technology registers a lower score in at least six of the impact categories studied. Concerning the total CO_2 emissions, CONVERGE exhibits a better performance compared to the Base Case, if the additional amount of CO_2 is either stored, sold as a by-product or vented into the atmosphere.

Keywords: residual biomass; CO_2 valorization; bio-methanol; advanced biofuels; life cycle assessment

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1. Introduction

In consideration of the increasing energy requirements in the domestic and industrial sectors [1], fossil fuels (e.g., coal, gas, crude oil, etc.) continue to play a significant role in fulfilling the global energy demand [2]. Based on the current energy consumption trends, it is expected that the limited fossil reserves will be depleted within the next 50 years [3], while also affecting the economic wealth through the increasing price of natural energy [4]. Moreover, it is well recognized that fossil fuel combustion generates large quantities of greenhouse gas (GHG) emissions. Among the GHG released, carbon dioxide is of high importance as it is the primary contributor to global warming and climate change, leading to a rise in both global surface temperatures and sea levels [5].

Given the above considerations, the attention of the scientific community has moved towards finding more renewable and sustainable solutions in order to overcome the energy crisis [6]. As stated in the revised version of the Renewable Energy Directive (RED), the European Union (EU) states must provide 32% share of total energy consumption as renewable energy. Furthermore, a minimum of 14% of the energy used in the transportation sector has to be produced from renewable sources by 2030 [7]. To achieve this ambitious target, it is essential to replace the fossil energy sources with low-carbon, sustainable alternatives [8]. Several studies assessing the potential replacement of fossil energy sources with renewable alternatives such as wind, hydro, solar, or biomass have been carried out [6].

In spite of the fact that all sources present positive aspects, biomass is regarded as the best option since several types of biomass are available regardless of the geographical location [9]. At the moment, an approximately 10% share of the overall energy demand is supplied through biomass. Additionally, the amount of carbon dioxide produced during the biofuel combustion is captured in the process of biomass growth, thus reaching carbon neutrality and potentially even resulting in negative emissions [10]. Waste biomass may be provided through various types, from municipal, agricultural and industrial wastes up to forest residues [11].

Among the most auspicious alternatives, the use of green fuels (e.g., bio-ethanol, bio-methanol, biodiesel, etc.) or fuel blends, under certain conditions, would result in a decrease of GHG emissions and a more sustainable environment [12]. In addition, Shadidi and co-authors have shown that the use of fuel blends led to increased engine power [13]. Considering that the use of food crops for biofuel production would directly affect the food industry by reducing the agricultural area, the EU committee imposed multiple restrictions in regard to the feedstock used to avoid the food vs. fuel competition. As a result, the share of biofuels produced from residual biomass has increased [14], and according to Yadav and co-authors [15], a large amount of forest residues is available across Europe and can be utilized by converting it to biofuels. In addition, olive pomace is generated as a by-product of the olive oil industry, and it is further treated to extract the residual oil, resulting in a solid residue called exhausted olive pomace. The exhausted olive pomace could potentially be used as a renewable resource for biofuels production [16].

Across a wide diversity of biofuels that might be obtained through the biomass conversion, such as, for example, bio-ethanol, bio-butanol or bio-methanol, bio-methanol is considered the most appropriate option in the long-term due to its GHG reduction capability [17]. Bio-methanol shows significant value both as a direct fuel or fuel blends [18], while it is also an essential raw material in the chemical industry as it might be further used to synthesize formaldehyde, methyl tertiary butyl ether (MTBE), dimethyl ether (DME) or biodiesel [19]. Given the amount of interest methanol received due to its energy storage capacity [20] and to the attention moved towards the Carbon Capture and Utilization (CCU) topic [21], approximately 110 Mt of methanol are produced in more than 90 methanol production plants all over the world [22]. Moreover, as stated by Choe and collaborators, it is forecasted that the global methanol market size will nearly triple by 2026 as compared to 2015, reaching around USD 92 billion [23].

Commonly, methanol is produced from fossil sources such as natural gas or coal through either a steam methane reforming (SMR) process [24] or by coal gasification [25]. Regardless of the production route, methanol synthesis usually consists of the syngas production, syngas conditioning and methanol production and purification stages. As SMR or gasification occur in the first step, part of the gas is burnt with oxygen to reach the heat demand, thus an air separation unit (ASU) is employed for oxygen generation. One can observe that besides the amount of fossil resources required and GHG emissions generated from the main process, an increased power consumption is registered in the upstream stages, specifically for the ASU [26]. However, power-to-gas (PtG) techniques could be implemented to mitigate global warming and climate change since PtG assumes the use of captured CO_2 to produce value-added chemicals or fuels. According to Chauvy and co-authors, one such process approaching commercial scale is CO_2 to methanol, having reached a technology readiness level (TRL) of 8–9 [27].

Life Cycle Assessment (LCA) is an efficient instrument used to determine the environmental footprint of a specific process or product by means of specific software (e.g., GaBi, SimaPro, openLCA, etc.) considering all stages, from raw materials extraction up to final disposal [28]. Several studies were carried out to evaluate the environmental performance of various methanol production routes using the Life Cycle Assessment (LCA) methodology. Qin and co-authors performed a case study of China for methanol production through coal gasification. The authors concluded that methanol production stands as the largest emission source (i.e., roughly 93% share of total carbon footprint), while the gasification unit and water–gas shift step are the main contributors. Moreover, the outcome also highlights the potential benefits brought by the addition of a carbon capture unit [29]. Gao and collaborators also investigated the coal to methanol production in China, aiming to determine large emission points and possible reduction methods. It was proved that high quantities of both water and energy are used, thus releasing vast CO_2, SO_2 and NO_x emissions [30]. Matzen and Demirel evaluated green methanol and DME production through the use of electrolytic H_2 and captured CO_2. The authors showed that a better environmental performance is achieved either in the production of methanol or DME compared to the traditional fuels [31]. Methanol and DME production starting from natural gas were studied by Lerner and co-authors. The investigation aims to identify the best conditions towards the lowest environmental impact. The authors concluded that the use of low-emission energy sources would result in a more environmentally friendly process [32]. Li and collaborators compared the potential utilization of coke oven gas as a raw material for methanol production against both coal and natural gas-based methanol. As the authors point out, a better environmental impact is achieved when using coke oven gas as feedstock compared to the coal pathway [33].

Recent studies evaluate different approaches for bio-methanol production through the utilization of captured CO_2 as raw material. Khojasteh-Salkuyeh and collaborators explored three different methanol production routes and concluded that the employment of renewable energy sources is imperative if considering the CO_2 hydrogenation route [34]. Rigamonti and Brivio examined a new process design by evaluating the potential utilization of steel mill flue gases as feedstock for methanol production and power generation. The outcome of the LCA analysis shows an improved environmental performance for the new designed system [35]. Ryoo and collaborators investigated four methanol production routes across different TRL classes. The authors found that the CO_2 hydrogenation process could be of major importance towards net zero emissions if renewable energy sources are used [36]. The key role of renewable energy sources is highlighted as well in the study made by Cordero-Lanzac and co-authors [37]. The authors concluded that a decrease of around 1.75 tons of CO_2 per ton of methanol could be registered, only if renewable energy sources are to be used.

The CONVERGE system is based upon the concept of developing and integrating new technologies such as catalytic cracking of tars (CCT) from an indirectly heated gasifier, recovery of refinery products including aromatics for green BTX fraction, sorption-enhanced reforming (SER) for excess-carbon removal and H_2 production, highly efficient electrochemical green H_2 compression (EHC), and membrane-based reactor for efficient methanol synthesis. The CCT design offers several benefits such as heat transfer through the catalyst from the regeneration to the cracking zone and no need for inlet gas nozzles that would potentially be clogged by the high dust content in the gas. The main advantages of the SER process compared to the conventional CO_2 separation are the requirement of fewer reaction vessels, higher H_2 yields (>95 mol%, dry basis) obtained in one single process step, no shift catalysts and CO_2 solvents required and energy efficiency improvement due to the integration of heat between the exothermic carbonation and endothermic reforming reactions. The membrane system for the methanol synthesis can enhance the per pass production resulting in a notable reduction of the equipment size.

Therefore, the present study is focused on the bio-methanol production from secondary biomass through a more innovative process that takes advantage of cutting-edge technologies with the aim of achieving an improved environmental performance, given the innovative syngas cleaning and conditioning steps or membrane-based reactor for methanol synthesis, when compared to the state-of-the-art technology [38].

2. Materials and Methods

The following CONVERGE technology scenarios are considered in the present work:

Case 1: Bio-methanol production from wooden biomass baseline;

Case 2: Bio-methanol production from wooden biomass with enhanced CO_2 resistance;

Case 3: Bio-methanol production from exhausted olive pomace with enhanced CO_2 resistance.

In spite of the fact that both Case 1 and Case 2 assess the bio-methanol production starting from wooden biomass, the main difference between the two consists of the process configuration. To better present the investigated scenarios, the block flow diagrams of each configuration are further displayed in Figure 1. As illustrated, biomass is fed to a drying system to adjust the moisture content for the MILENA gasifier. The gasification island includes the MILENA gasifier and both flue gas and solids management. The outlet stream of the gasifier is sent to the cleaning and conditioning section, where tars are removed and syngas composition is adjusted for the subsequent sections. As previously mentioned, tar removal stands as one of the innovative processes developed as it involves a conversion step to achieve BTX, which are then separated and utilized as a by-product. Afterwards, the clean syngas is sent to the SER section to separate the CO_2 from the main stream. Offgas combustion is used to generate the required steam and heat for BTX production, as well as for sorbent regeneration. To reach the required pressure for methanol synthesis, the main stream from the SER section is sent to the EHC to separate and compress pure H_2, while the CO_2 is compressed in a conventional multistage intercooled compressor. H_2 and CO_2 are mixed and fed to membrane reactors to produce methanol.

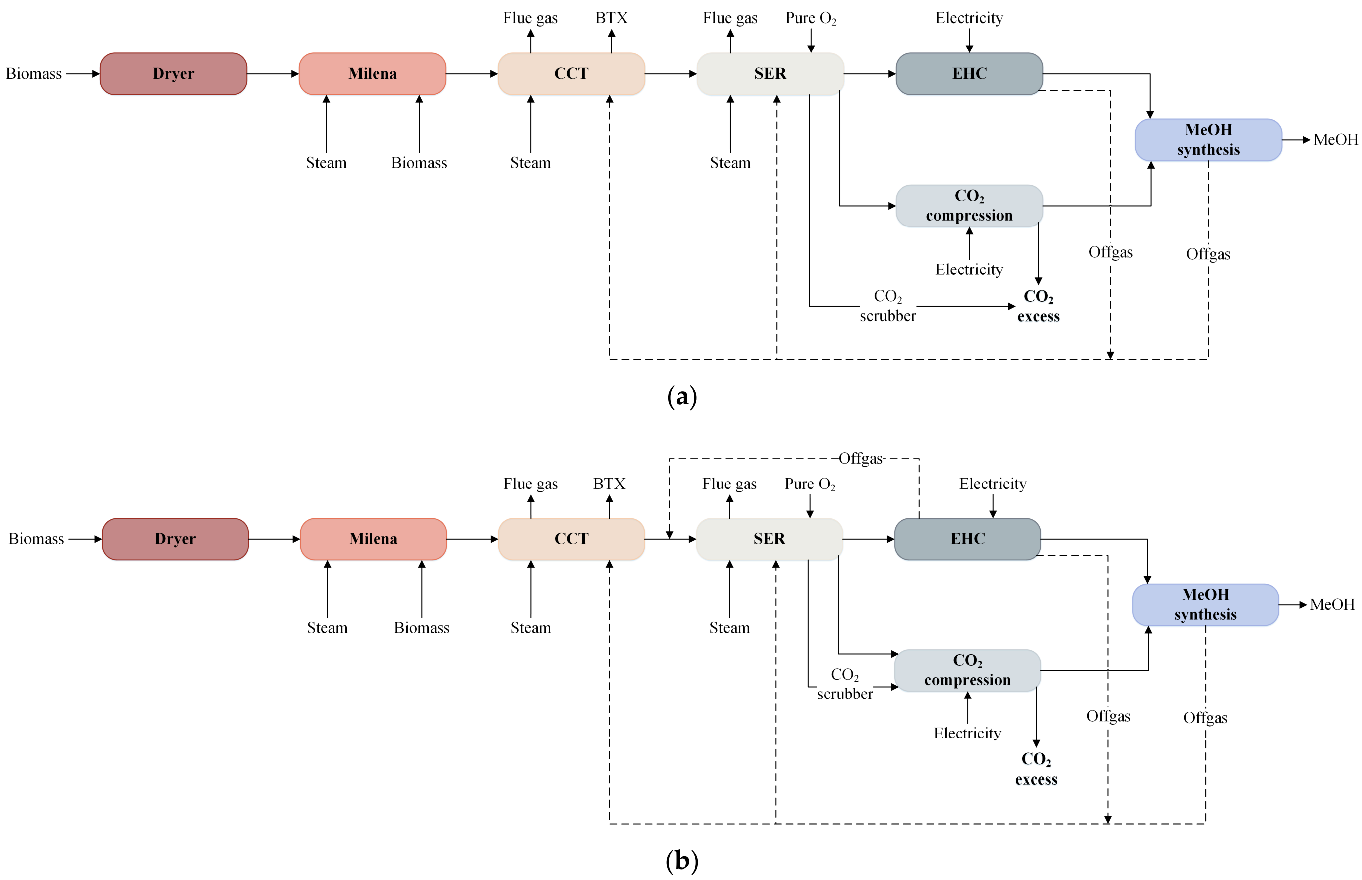


Figure 1. Block flow diagrams for the CONVERGE technology scenarios. (**a**) CONVERGE baseline, (**b**) CONVERGE with enhanced CO_2 resistance.

Moreover, considering that the best CONVERGE scenario is eventually compared to the state-of-the-art technology (i.e., Base Case), Table 1 presents the main differences and the benefits of CONVERGE against the Base Case. As one can observe, the most significant differences between the two technologies start from the tar removal section. On the one hand, catalytic cracking of tars is used within the CONVERGE system, resulting in a stream of BTX that is considered as a by-product. On the other hand, the Milena gasifier is coupled with the OLGA tar removal in the Base Case. For the syngas cleaning and conditioning, water scrubbing is employed both in the CONVERGE and Base Case. However, the CO_2 capture is performed using the Sorption enhanced reforming process in the CONVERGE scenario, while a Methyl-Di-Ethanol-Amine (MDEA)-based reactive gas–liquid separation system is utilized in the Base Case. Lastly, methanol synthesis is performed using a membrane reactor, employing both high- and low-pressure separators for methanol purification in the CONVERGE system, while the Base Case is using a boiling water reactor.

Table 1. Main differences between the Base Case and CONVERGE process.

Process	CONVERGE	Base Case
Biomass drying	Tube bundler drier	Tube bundle drier
Synthesis gas production	Indirect gasification (Milena); Atmospheric pressure; Air and steam added; Filtration included.	Indirect gasification (Milena)l; Atmospheric pressure; Air and steam added.
Tar removal	Catalytic cracking tar removal (CCT); Air and steam added.	Oil scrubbing (OLGA).
Syngas cleaning and conditioning	Water scrubbing; BTX scrubbing; BTX considered as a by-product; Sorption enhanced reforming (SER) considered for CO_2 removal; H_2 compression using electrochemical hydrogen compression (EHC) up to 80 bar.	Water scrubbing; Compression up to 22 bar; Tubular reforming; Water gas shift (WGS) bypassed; MDEA based acid gas removal (AGR); Compression up to 72 bar.
Methanol synthesis and purification	Membrane reactor; High (i.e., 80 bar) and low pressure (i.e., 1.5 bar) separators.	Boiling water reactor; Stripping of light gases and water separation.

The present study follows the requirements and recommendations provided by the International Organization for Standardization (ISO) through ISO 14040:2006 [39] and ISO 14044:2006 [40], covering all four stages: (i) Goal and scope definition, (ii) Life Cycle Inventory (LCI), (iii) Life Cycle Impact Assessment (LCIA), and (iv) Interpretation of the results.

2.1. Goal and Scope Definition

The goal of the current study is to quantify, evaluate, and compare the environmental burden of the CONVERGE technology for the bio-methanol production against the state-of-the-art technologies for biomass conversion and bio-methanol synthesis. As previously presented, the CONVERGE system combines five innovative technologies aiming to improve the conversion steps of secondary biomass to biofuels in an efficient and cost-effective manner. A functional unit is used as a reference point for all impact assessment calculations; thus, one ton of bio-methanol (t_{MeOH}) was chosen as functional unit in the current work.

The system boundaries are referred to as the interface between the set of unit processes under investigation and environment. Based on the goal of the study, there are four options to define the limits of the system: (i) cradle-to-grave, (ii) cradle-to-gate, (iii) gate-to-gate, and (iv) gate-to-grave. The present work is a cradle-to-gate LCA study, as it covers all production steps from the raw materials supply chain (i.e., biomass) up to the finished product (i.e., bio-methanol).

As illustrated in Figure 2, the system boundaries include the following: (i) upstream processes: biomass supply chain, catalysts production and supply, membrane production and supply, sorbent production and supply chain; (ii) main processes: bio-methanol production using different biomass sources (i.e., wooden biomass and exhausted olive pomace); (iii) downstream processes: disposal of wastes (i.e., catalyst, sorbent, etc.).

In regards to the geographical limitations, the CONVERGE system assumes Northern Sweden and Italy as the plant location for the wooden biomass and exhausted olive pomace scenario, respectively. A plant lifetime of 20 years was considered as well.

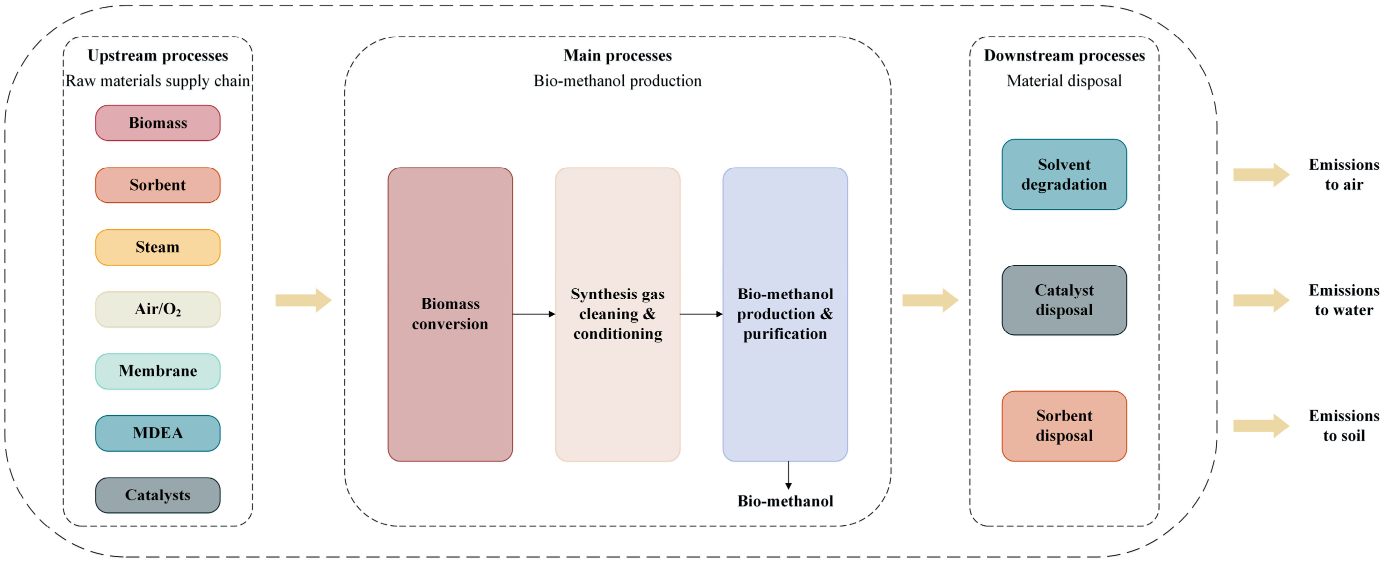


Figure 2. System boundaries for the CONVERGE technology.

To appropriately provide conclusions and recommendations, it is of major importance to clearly state the limitations of the study. The current investigation does not take into account the following items: (i) construction and decommissioning of the plant; (ii) repair and maintenance operations; (iii) building the infrastructure (i.e., roads, railways, etc.); (iv) installation of unloading facilities; (v) human activities associated to labor tasks; (vi) low-frequency, high magnitude, unpredictable events (i.e., fugitive, accidental releases, etc.).

Table 2 presents the data collection and the most relevant assumptions considered for the biomass supply chain (i.e., both for the wooden and exhausted olive pomace).

Table 2. Assumptions for the biomass supply chain.

Assumption for Wooden Biomass	Unit	Value
Biomass (M = 50%) as forest residues	t	4.52
Biomass (wood chips), losses included (5%)	t	3.65
Biomass (M = 35%) as wood chips, at plant gate	t	3.48
Residues' bundling and handling from forest site to the road side [41]		
Distance	km	6.79
Bulk density of residues	t/m^3	0.15
Handled forest (residues/t_{MeOH})	t	4.52
Tractor and trailer	tkm	30.70
Diesel consumption	kg_{diesel}/tkm	0.044
	kg_{diesel}/t_{MeOH}	1.34
Chipping phase of dried residues		
Diesel consumption	$L_{diesel}/t_{residue}$	2.40
	kg_{diesel}/t_{MeOH}	7.45
Road transport of wood chips [42,43]		
Distance	km	100
Lorry > 32t EURO3	tkm	365.40
Diesel consumption	kg_{diesel}/tkm	0.025
	kg_{diesel}/t_{MeOH}	9.36

Table 2. *Cont.*

Assumption for exhausted olive pomace	Unit	Value
Input raw material (M ≤ 12%)	t	2.34
Biomass at the facility (M ≤ 12%)	t	2.34
Biomass at the facility, handling losses (5%)	t	2.36
Road transport		
From the point of extraction to conversion plant	tkm	472.68
Road transport from field to conversion plant	tkm	425.41

The assumptions considering other up-stream processes are illustrated in Table 3.

Table 3. Assumptions considering upstream and downstream processes.

Process	Assumption		
Upstream stages			
Olive extraction & transportation [44]	Water consumption	6037.76	$kg/t_{olivine}$
	Electricity consumption	145.67	$MJ/t_{olivine}$
	Fuel consumption	9.95	$kg_{NG}/t_{olivine}$
	Gaseous emissions	5.28	$kg_{CO2}/t_{olivine}$
	Waste water	6037.76	$kg/t_{olivine}$
	Transportation by rail from Norway	1200	km
	Transportation fuel type	Diesel	
Reforming catalyst [45]	Composition (wt.%): 18% NiO, 0.1% SiO_2, 0.05% SO_3, 81.85% Al_2O_3.		
	Amount: 8.934×10^{-9} m^3/kg_{H2}.		
	Transportation mode: Ship + Rail (from UK).		
	Transportation distance: 1120 km (ship) + 1420 km (rail).		
	Transportation fuel type: heavy fuel oil/diesel and electricity.		
Water gas shift catalyst [46]	Composition (wt%): 88% Fe_2O_3, 9% Cr_2O_3, 2.6% CuO, 0.4% Al_2O_3.		
	Amount: 9.927×10^{-9} m^3/kg_{H2}.		
	Transportation mode: Ship + Rail (from UK).		
	Transportation distance: 1120 km (ship) + 1420 km (rail).		
	Transportation fuel type: heavy fuel oil/diesel and electricity.		
Membrane production & transportation	Raw materials ceramic support (wt.%): 40% Clay materials, 20% Marble dust, 20% Olive stone, 20% Fired tile scrap.		
	Coating: there are two intermediate layers of alfa and gamma alumina on the ceramic suport and, afterwards, the membrane coating comes on top.		
Downstream stages			
Ash & Char disposal	Considered to be transported to the neareast cement plant in order to be used as raw material for the cement production.		
Olivine disposal	Landfill disposal was assumed for the used olivine.		
Sorbent disposal	Sorbent is modelled as a consumed product (100 years assumed as sorbent lifetime); thus, no end-of-life stage was assumed.		

The main assumptions considered for the bio-methanol production following the CONVERGE technology are as follows:

(i). 7889 annual operating hours assumed;
(ii). 80–90% total carbon conversion in Milena gasifier and 100% char combustion in the combustor;
(iii). O_2 produced through an Air Separation Unit (ASU) according to data provided in the scientific literature [47];
(iv). all higher hydrocarbons are lumped in C_2H_4, including BTX;
(v). CCT completely converts higher hydrocarbons (>C8);
(vi). single operating point for EHC is considered and a purity higher than 99.5% H_2 was set;
(vii). membrane working at 250 °C and 80 bar;
(viii). electricity imported from the grid mix considering the electric grid mix specific to Sweden for woody biomass, and Italy for the exhausted olive pomace.

2.2. Life Cycle Inventory

Life cycle inventory (LCI) is the second step of an LCA and it implies the set-up of an inventory consisting of input and output flows for a specific product system. It consists of detailed tracking of all the input and output streams, including raw materials, resources, energy by type, water, and emissions to air, water and soil.

The data sets used within an LCA study can be categorized as primary data (i.e., data sets collected from interviews, questionnaires, on-site measurements, etc.) and secondary data (i.e., derived from modelling and simulation activities, estimation, scientific literature, etc.). The current study utilizes both primary and secondary data sets, as the data required in the LCI of the current investigated systems were retrieved from interviews (i.e., biomass supply chain), process modelling and simulation, and scientific literature (i.e., membrane and catalysts production), where it could not be provided by project partners. Experimental activities and laboratory measurements were performed to provide the required data sets and aid in modelling activities.

The most significant data sets for the investigated CONVERGE technology scenarios are presented in Supplementary Materials.

2.3. Life Cycle Impact Assessment

Life cycle impact assessment (LCIA) aims to create a link between the system's inventory of elementary flows and their potential environmental impact. In the case of generally accepted and straightforward impact categories, for which characterization factors have already been derived, all inventory results are pre-classified to pre-selected impact categories already available in different LCA software tools (e.g., GaBi, SimaPRO).

ReCiPe was selected as the impact assessment method given that it is one of the most recent and updated assessment methods available to the LCA practitioners [48]. The life cycle inventories are converted into a number of harmonized impact scores at both midpoint and endpoint level. On the one hand, midpoint characterization methods lead to more accurate results and reduce the uncertainty. On the other hand, endpoint impact categories are easier to interpret, but exhibit higher modeling uncertainty. The main environmental impact categories considered in the present work, along with their description and relevant contributors, are outlined in Table 4.

For the purpose of this work, GaBi version 10.5, developed by Sphera™ [47], was chosen as LCA software tool. GaBi was developed by the German company initially called thinkstep and more recently, Sphera™.

Table 4. ReCiPe method–impact categories definition.

Impact Category	Definition
Climate change/Global Warming Potential (GWP)	Impact of anthropogenic emissions enhancing the radiative forcing of the atmosphere, causing the temperature at the earth's surface to rise.
Freshwater Eutrophication Potential (FEP)	Quantification of phosphorus and nitrogen from the inland waterways into phosphorus equivalents.
Ozone Depletion Potential (ODP)	Thinning the stratospheric ozone layer due to anthropogenic emissions.
Fossil fuel Depletion Potential (FDP)	Surplus energy per extracted MJ, kg or m^3 fossil fuel.
Freshwater Ecotoxicity Potential (FETP)	Potential impact of toxic substances on aquatic ecosystem.
Human Toxicity Potential (HTP)	Potential impact of toxic substances on human health.
Mineral Depletion Potential (MDP)	Surplus energy needed for future extraction of ore.
Photochemical Oxidant Formation Potential (POFP)	Formation of reactive compounds by sunlight action on primary air pollutants.
Terrestrial Ecotoxicity Potential (TETP)	Potential impact of toxic substances on terrestrial ecosystems.

3. Results and Discussion

The LCA results for the three evaluated CONVERGE technology scenarios are reported in Table 5. As previously mentioned, the results were obtained using the ReCiPe impact assessment method based on the above-described assumptions and limitations of the current study.

Table 5. LCA results for the CONVERGE technology scenarios.

KPI	Units	Case 1	Case 2	Case 3
GWP	kg CO_2 eq./t_{MeOH}	1436.19	1288.04	2511.22
FEP × 10^3	kg P eq./t_{MeOH}	4.15	7.69	8.73
ODP × 10^9	kg CFC−11 eq./t_{MeOH}	2.86	7.09	5.85
FDP	kg oil eq./t_{MeOH}	31.24	32.39	263.86
FETP	kg 1,4-DB eq./t_{MeOH}	0.28	0.26	0.07
HTP	kg 1,4-DB eq./t_{MeOH}	13.58	13.98	22.53
MDP	kg Fe eq./t_{MeOH}	4.95	5.05	10.08
POFP	kg NMVOC/t_{MeOH}	0.28	0.29	1.05
TETP × 10^3	kg 1,4-DB eq./t_{MeOH}	7.93	7.69	19.34

As one can observe, the outcome of the environmental impact assessment points towards Case 1 as the most sustainable scenario among the evaluated cases. Case 1 presents the lowest score in six out of a total of nine impact categories (i.e., FEP, ODP, FDP, HTP, MDP and POFP). Moreover, Case 1 ranks second in terms of GWP and TETP impact. The worst-case scenario is represented by Case 3 as it records the highest values in seven impact indicators. Case 3 assumes that the main plant is located in Italy, while exhausted olive pomace is used as biomass source. Therefore, the main reason behind the increased environmental impact is represented by the electricity generation since the electric grid mix for Italy is mainly based on fossil fuels. Another cause would be the additional amount of biomass required as additional fuel within the process.

Freshwater eutrophication potential (i.e., FEP) presents a quantification of nutrients that are present in the inland waterways through their conversion into phosphorus equivalents. As shown in Table 5, the highest FEP score is registered in Case 3 (8.73×10^{-3} kg P eq./t_{MeOH}), followed by Case 2 (7.69×10^{-3} kg P eq./t_{MeOH}). The ODP impact category is mainly influenced by the wastewater treatment process (i.e., at least 98% contribution to the total impact) and, as previously mentioned, the lowest score is achieved in Case 1 (2.86×10^{-9} kg CFC−11 eq./t_{MeOH}), succeeded by Case 3 and Case 2, respectively (5.85×10^{-9} kg CFC−11 eq./t_{MeOH} and 7.09×10^{-9} kg CFC−11 eq./t_{MeOH}). Further, FETP category shows the potential impact of toxic substances on the aquatic ecosystem. The lowest score is registered in Case 3, followed by Case 2 and Case 1, respectively (0.07 kg 1,4-DB eq./t_{MeOH} vs. 0.26 kg 1,4-DB eq./t_{MeOH} vs. 0.28 kg 1,4-DB eq./t_{MeOH}). For the first two investigated cases (i.e., Case 1 and Case 2), electricity generation for H_2, as well as CO_2 compression, rank as first and second contributor, respectively. In the third scenario, Case 3, the highest share to the total impact (i.e., around 28%) comes from the sorbent supply chain. POFP category refers to the formation of reactive chemical compounds (e.g., ozone) by the action of sunlight on primary air pollutants. The lowest impact is obtained in Case 1, while a similar value is registered in Case 2 (0.28 kg NMVOC/t_{MeOH} vs. 0.29 kg NMVOC/t_{MeOH}). Case 3 displays a more than 3.5 times higher POFP score compared to the wooden biomass scenarios (i.e., Case 1 and Case 2).

Details and discussion regarding the sub-processes that exhibit the highest influence on the most relevant impact categories are presented in the next subsections.

3.1. GWP Impact Category

The detailed distribution of the GWP impact category for all CONVERGE technology scenarios is displayed in Figure 3. As illustrated, Milena (i.e., gasification section) followed by SER section show the highest influence on the GWP impact in either of the three evaluated cases. When looking at Case 1, Milena contributes with approximately 50% to the total share, releasing around 718 kg CO_2 eq./t_{MeOH}, while the SER section generates 544 kg CO_2 eq./t_{MeOH} as gaseous emissions. Therefore, about 89% of the total greenhouse gas emissions in Case 1 are due to the two processes. Tar removal and electricity generation for the electrochemical H_2 compression rank as third and fourth contributors, respectively. Similar to Case 1, Milena and SER section remain the main greenhouse gas sources with a combined share of nearly 86% of the total emissions released in Case 2 (i.e., 641 kg CO_2 eq./t_{MeOH} are produced from Milena and 462.641 kg CO_2 eq./t_{MeOH} are released from SER). Compared to Case 1, tar removal and electricity generation for electrochemical H_2 compression show slightly higher contributions, 6.90% and 2.29%, respectively.

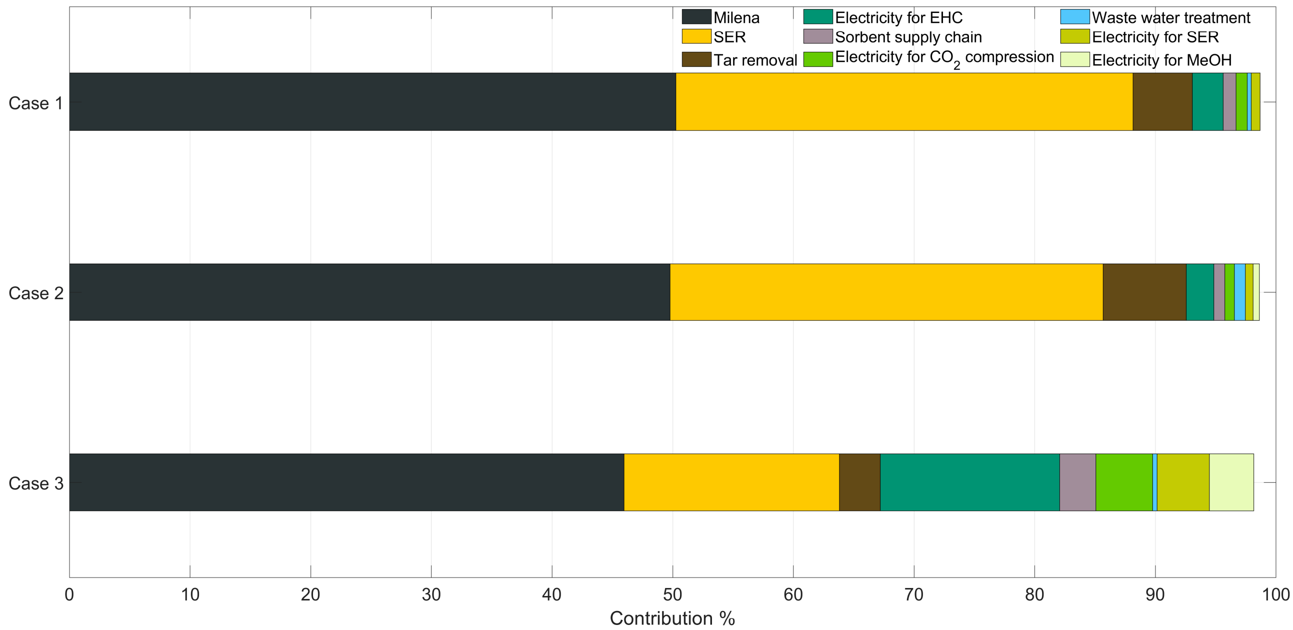


Figure 3. Detailed distribution of the GWP impact category for the CONVERGE scenarios.

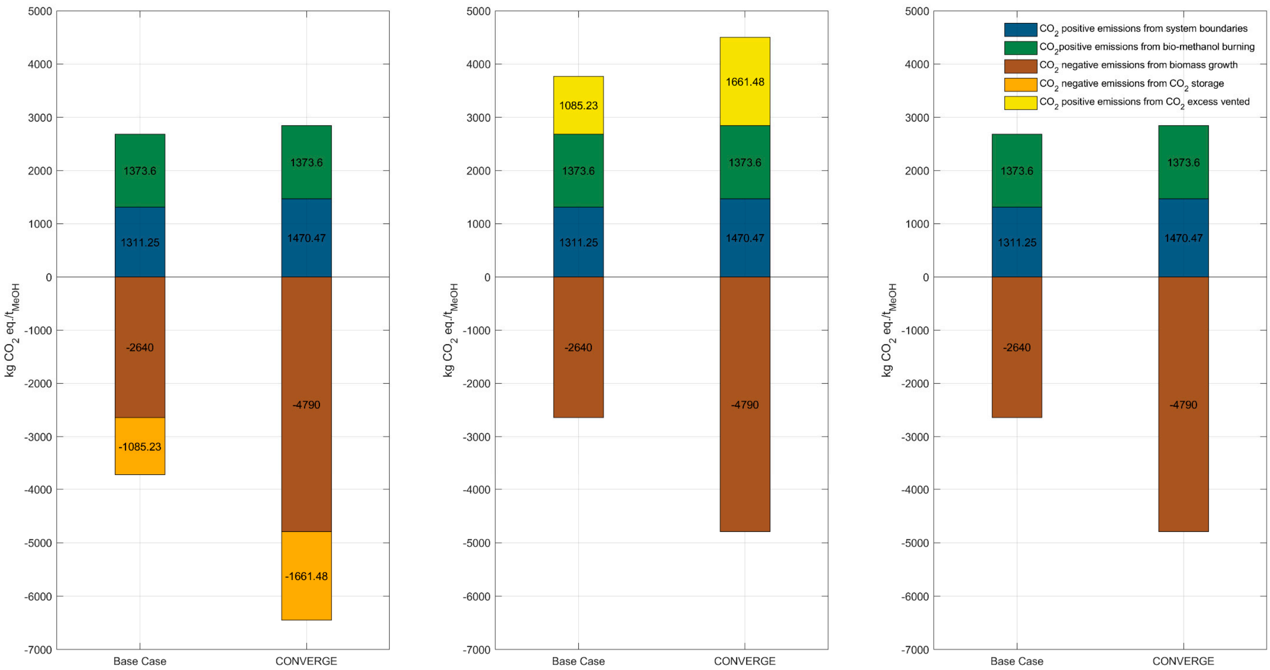
In the third evaluated case (i.e., Case 3), the gasification section remains the primary contributor towards the GWP impact with nearly 46% out of the total share, thus generating around 1154 kg CO_2 eq./t_{MeOH} out of the total of 2511.22 kg CO_2 eq./t_{MeOH}. As already mentioned, the high score highlights the impact brought by the additional amount of biomass required as additional fuel. Since Italy is assumed as plant location, the electricity production has a considerable influence due to the fact that the electric grid mix is mainly based on fossil fuels.

Compared to the first two cases, the impact of the SER section is much lower (i.e., 17.86% in Case 3 vs. 35.88% in Case 2 vs. 37.88% in Case 1), yet the electricity generation for H_2 and CO_2 compression show an important increase. On the one hand, the power production for the electrochemical H_2 compression ranks third with a 14.87% share; therefore, 373 kg CO_2 eq./t_{MeOH} are released. On the other hand, the electric power production for the CO_2 compression provides 4.70% out of the total impact, and it is followed by the power generated for the SER and MeOH section, respectively.

However, by considering and quantifying all emission sources, negative emissions can be obtained. The total GWP obtained by adding all the emissions absorbed during the biomass growth, CO_2 emissions released from the processes considered within the LCA boundaries, CO_2 released by the bio-methanol combustion and the different scenarios considering the separated CO_2 from AGR (i.e., in the Base Case) and the CO_2 separated from the SER and CO_2 compression stages (CONVERGE technology) are presented in Figure 4. As illustrated, the amount of CO_2 removed from AGR (i.e., in the Base Case) and the CO_2 removed from SER and CO_2 compression in the CONVERGE case, could be considered as negative emission if stored. Therefore, an additional amount of 1085.23 kg CO_2 eq./t_{MeOH} for the Base Case and 1661.48 kg CO_2 eq./t_{MeOH} for the CONVERGE case might result as negative emissions (see Figure 4). The CO_2 derived from the combustion of one ton of bio-methanol is 1373.60 kg CO_2/t_{MeOH}. The total CO_2 emissions for the Base Case will reach −1040.39 kg CO_2 eq./t_{MeOH} and −3607.72 kg CO_2 eq./t_{MeOH} for the CONVERGE case. As a general conclusion when taking into account the total GWP impact, the CONVERGE technology exhibits a better performance when compared to the Base Case. As is according to the LCA calculations, the contribution of the compression and transportation stage of the CO_2 from AGR (i.e., for Base Case), and the CO_2 from SER and CO_2 compression (i.e., CONVERGE) were found to be negligible to the total impact (i.e., less than 0.5%).

Depending on the fate of the separated CO_2 from the AGR (i.e., for Base Case) and the CO_2 separated from SER and CO_2 compression in the CONVERGE case, two more scenarios can be approached. On the one hand, if the separated CO_2 is released into the atmosphere, the scenario depicted in the middle part of Figure 4, the total CO_2 eq. released would be around 1130.08 kg CO_2 eq./t_{MeOH} for the BC and −284.45 kg CO_2 eq./t_{MeOH} for the CONVERGE case. The negative emissions obtained in the CONVERGE scenario are due to a higher quantity of biomass required as compared to the Base Case. On the other hand, the separated CO_2 can be considered as a valuable by-product and, therefore, it can be sold (see Figure 4). As a result, it can be considered close to zero emissions from the AGR section, as well as from the SER and CO_2 compression. These assumptions lead to a total of nearly 44.85 kg CO_2 eq./t_{MeOH} and −1945.93 kg CO_2 eq./t_{MeOH} for the Base Case and CONVERGE technology, respectively.

As a general conclusion regarding the GWP impact indicator, one can state that the CONVERGE technology provides better environmental performance compared to the BC both if the separated CO_2 is stored or if it is either sold or vented into the atmosphere.



(**a**) (**b**) (**c**)

Figure 4. Detailed GWP impact indicator considering all emission sources: (**a**) excess CO_2 is sent to storage; (**b**) excess CO_2 is vented into the atmosphere; (**c**) excess CO_2 is modelled as intermediate component, ready to be used as raw material in another process/section.

3.2. FDP Impact Category

As presented in Table 5, Case 1 and Case 2 register a similar FDP score (i.e., 31.24 kg oil eq./t_{MeOH} vs. 32.39 kg oil eq./t_{MeOH}), while Case 3 shows an approximately eight times higher value (e.g., 263.85 kg oil eq./t_{MeOH}). The high difference between the first two cases and the third one consists of the difference between the electricity grid mix when comparing Sweden and Italy; thus, the plant location significantly influences the environmental performance. When referred to Sweden, most of the electric power used is produced from hydro and nuclear sources, while Italy relies on natural gas and oil.

Figure 5 shows the main sub-processes contributing to the FDP score for each evaluated scenario. For the two wooden biomass scenarios (i.e., Case 1 and Case 2), the biomass supply chain, particularly the diesel consumption within various steps of the biomass pre-processing, contributes the most. For Case 1, more than half (i.e., 54.41%) of the total of 31.24 kg oil eq./t_{MeOH} is brought by the biomass supply chain. The electricity generation for H_2 compression is followed by olivine disposal rank second and third contributors, respectively (i.e., 14.60%, and 10.98%). In the second case, Case 2, the biomass supply chain remains the primary contributor with a 53.73% share, while the wastewater treatment section and electricity production for H_2 compression rank second and third, respectively. For the Case 3 using exhausted olive pomace, the electricity generation for several sections, such as electrochemical H_2 compression, CO_2 compression, SER and the methanol synthesis section, present the highest influence, since the grid mix, when assessing Italy as plant location, is mainly based on fossil fuels.

3.3. HTP Impact Category

The HTP impact category refers to the potential effects of toxic substances upon human health. As shown in Table 5, Case 3, the scenario employing the use of exhausted olive pomace thus results in the highest environmental impact. The two scenarios assuming the use of wooden biomass (i.e., Case 1 and Case 2) lead to a similar score (13.58 kg 1,4-DB eq./t_{MeOH} vs. 13.98 kg 1,4-DB eq./t_{MeOH}).

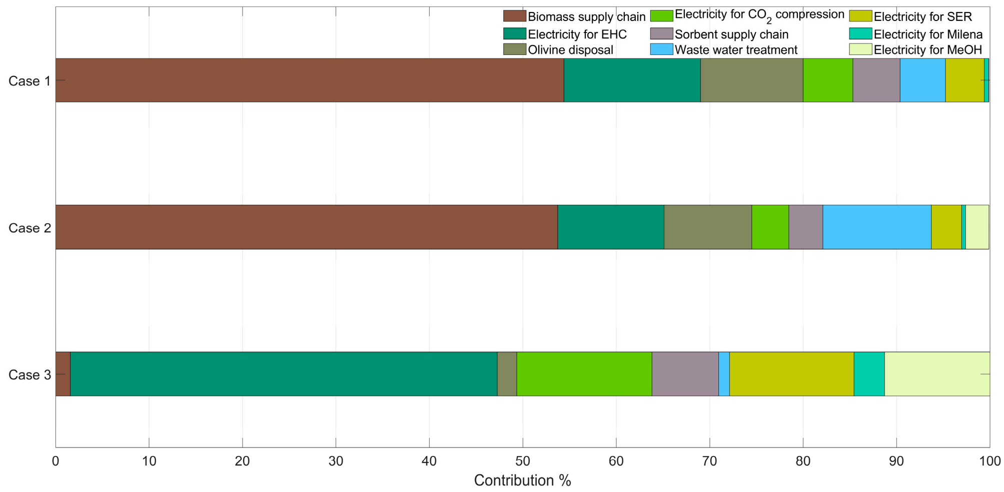


Figure 5. Detailed distribution of the FDP impact category for the CONVERGE scenarios.

Figure 6 displays the HTP distribution for all three investigated CONVERGE technology scenarios. As highlighted, the power production for electrochemical H_2 compression stands as the largest contributor to HTP in either case, contributing with at least 26% share to the total impact. Olivine disposal and electricity generation required for the CO_2 compression rank as second and third contributors, respectively, for Case 1 and Case 3. When evaluating Case 2, the wastewater treatment process comes as second contributor with an 18.35% share, thus providing 2.56 kg 1,4-DB eq./t_{MeOH} out of the total score.

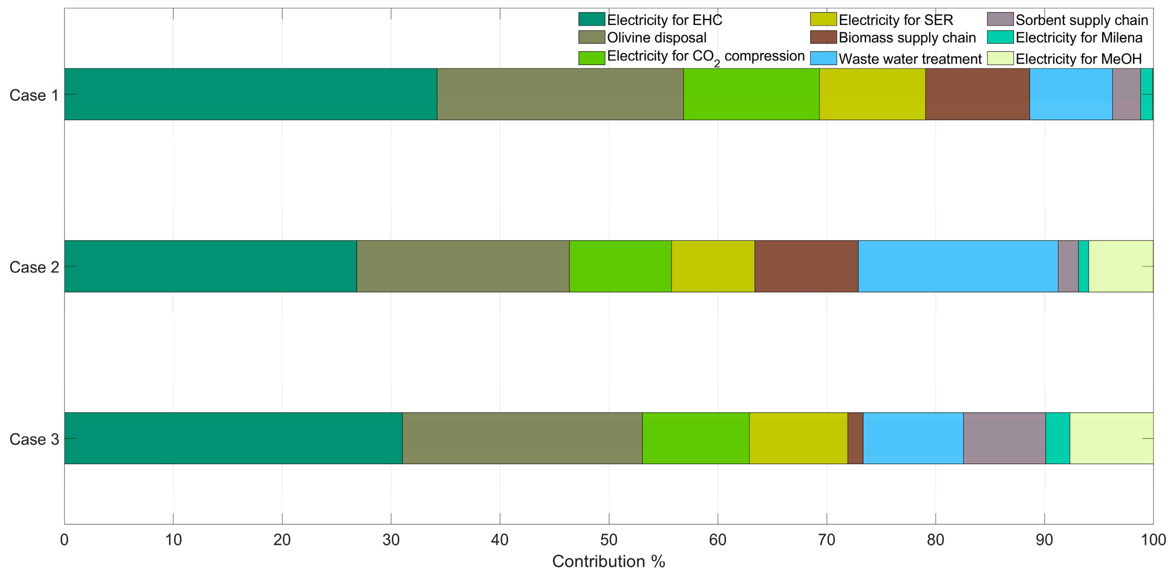


Figure 6. Detailed distribution of the HTP impact category for the CONVERGE scenarios.

The best environmental performance was obtained when assessing the use of wooden biomass (i.e., Case 1 and Case 2); thus the cases assuming Sweden as a plant location. The electricity generation for the electrochemical H_2 compression significantly impacts five out of nine impact categories, and therefore, a scenario analysis was further performed. Based on the fact that the majority of electricity production in Sweden relies on hydro and nuclear power, Table 6 presents the results for the scenarios when the electric power required for H_2 compression is supplied either from nuclear or hydro power sources.

Table 6. LCA results for the CONVERGE cases using nuclear and hydro power for H_2 compression.

KPI	Units	Nuclear Power		Hydro Power	
		Case 1	Case 2	Case 1	Case 2
GWP	kg CO_2 eq./t_{MeOH}	1403.52	1261.66	1413.53	1269.74
FEP × 10^3	kg P eq./t_{MeOH}	3.31	7.01	3.28	6.98
ODP × 10^9	kg CFC−11 eq./t_{MeOH}	2.86	7.09	2.86	7.09
FDP	kg oil eq./t_{MeOH}	28.32	30.04	26.98	28.95
FETP	kg 1,4-DB eq./t_{MeOH}	0.47	0.42	0.12	0.14
HTP	kg 1,4-DB eq./t_{MeOH}	14.29	14.55	9.33	10.55
MDP	kg Fe eq./t_{MeOH}	5.29	5.33	4.24	4.48
POFP	kg NMVOC/t_{MeOH}	0.20	0.22	0.19	0.21
TETP × 10^3	kg 1,4-DB eq./t_{MeOH}	10.70	9.86	4.29	4.75

On the one hand, a slightly better environmental performance can be seen when comparing the results obtained in the nuclear power scenario against the results obtained when supplying the electric power for H_2 compression from the grid mix (see Table 5). Five out of nine impact categories register similar or lower values (i.e., GWP, FEP, ODP, FDP, and TETP), while an increase in FETP, HTP, MDP and POFP can be observed. On the other hand, all impact indicators show lower values in the hydro power scenario when compared to the results obtained following the electric power supply from the grid mix, except ODP which is influenced by the wastewater treatment.

Furthermore, a comparison between the CONVERGE technology against the state-of-the-art indirect gasification technology (i.e., Base Case) is presented in Table 7 to better illustrate the potential benefits brought by the implementation of the CONVERGE.

Table 7. Environmental impact comparison between CONVERGE technology and Base Case.

KPI	Units	Base Case	Case 1	Case 2
GWP	kg CO_2 eq./t_{MeOH}	1311.25	1413.53	1269.74
FEP × 10^3	kg P eq./t_{MeOH}	4.32	3.28	6.98
ODP × 10^9	kg CFC−11 eq./t_{MeOH}	5.87	2.86	7.09
FDP	kg oil eq./t_{MeOH}	25.74	26.98	28.95
FETP	kg 1,4-DB eq./t_{MeOH}	0.51	0.12	0.14
HTP	kg 1,4-DB eq./t_{MeOH}	37.78	9.33	10.55
MDP	kg Fe eq./t_{MeOH}	2.66	4.24	4.48
POFP	kg NMVOC/t_{MeOH}	1.14	0.19	0.21
TETP × 10^3	kg 1,4-DB eq./t_{MeOH}	9.57	4.29	4.75

As observed, Case 2 registers the lowest GWP score (1269.74 kg CO_2 eq./t_{MeOH}), followed by the Base Case and Case 1, respectively. Milena section ranks as the main contributor to the GWP score in either Case 1 or Case 2 with an approximately 50% share, while the Milena and Olga system display a 70% contribution as well in the Base Case. A more detailed explanation regarding the GWP impact, taking into account the CO_2 negative emissions corresponding to the biomass growth is drawn in Section 3.1. Furthermore, it can be noticed from Table 7 that the overall environmental performance for both CONVERGE scenarios (i.e., Case 1 and Case 2) is higher compared to the Base Case scenario. Five out of the total of nine evaluated impact categories (i.e., GWP, FETP, HTP, POFP, and TETP) display a lower score in Case 2 against the Base Case, while Case 1 stands as the best performing scenario. Case 1 registers the best score in six impact indicators studied (i.e., FEP, ODP, FETP, HTP, POFP, and TETP).

4. Conclusions

Three CONVERGE technology scenarios following two different process' configurations, using either wooden biomass or exhausted olive pomace were compared against the state-of-the-art technology for bio-methanol production. The major results of the current research are as follow:

1. The utilization of exhausted olive pomace as biomass source (i.e., CONVERGE Case 3) resulted in the highest score in seven out of nine impact categories studied (i.e., GWP, FEP, FDP, HTP, MDP, POFP and TETP). These high values may be primarily attributed to the fossil fuel use for electric power generation and, as well, to the fact that an additional amount of biomass is required.
2. CONVERGE Case 1 exhibits the best environmental performance among the investigated scenarios as it displays the lowest score in six out of a total of nine impact categories (i.e., FEP, ODP, FDP, HTP, MDP and POFP), while placing second in regard to GWP.
3. A detailed analysis of the three most relevant impact categories (i.e., GWP, FDP and HTP) was carried out to highlight critical issues. As pointed out, the gasification section contributes half of total process GWP impact; thus, small improvements in this area may result in increased environmental performance. The impact of different grid mix scenarios (i.e., Sweden for wooden biomass vs. Italy for exhausted olive pomace) may be seen when looking both at the FDP and HTP since electricity production shows strong influence.
4. Based on the aforementioned aspects and considering that the electricity generation significantly influences several other impact categories, a scenario analysis targeting the power generation was performed. The results indicate that the use of hydro power sources would enhance the environmental performance of the CONVERGE system.
5. A comparison between the best CONVERGE configurations and state-of-the-art technology (i.e., Base Case) was performed. As concluded, Case 1 registers a 6 times lower POFP impact, 4 times lower FETP and HTP scores, as well as better FEP, ODP and TETP performance. Moreover, eight impact categories display lower values in Case 1 when compared to Case 2.
6. The GWP impact must be computed by taking into account all emission sources, and thus, by also considering the upstream stages besides the main process. The amount of CO_2 captured can either be stored, sold as a by-product and used in other processes, or released into the atmosphere. In terms of total CO_2 emissions, after investigating the previously mentioned options (see GWP discussions), it was pointed out that CONVERGE technology provides better results as compared to the BC, if the captured CO_2 is either stored, sold as a by-product or vented into the atmosphere.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/en16062726/s1, Table S1: Life cycle inventory for the main upstream processes in all CONVERGE scenarios (i.e., Case 1, Case 2, and Case 3); Table S2: Life cycle inventory for the main processes in Case 1; Table S3: Life cycle inventory for the main processes in Case 2; Table S4: Life cycle inventory for the main processes in Case 3; Table S5: Life cycle inventory for the downstream processes in all CONVERGE scenarios (i.e., Case 1, Case 2, and Case 3).

Author Contributions: S.C.G. performed and validated two LCA cases, wrote the first draft of the paper and revised the last version of the manuscript. L.P. provided guidance, supervised the LCA study, revised the manuscript and acquired funding. D.A.C. performed the calculation for one LCA case and for the Base Case and validated the results. C.-C.C. was involved in conceptualization and supervision. M.U. provided useful data for the biomass supply chain. All authors have read and agreed to the published version of the manuscript.

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Nomenclature

GHG	Greenhouse gas
RED	Renewable energy directive
EU	European Union
MTBE	Methyl tertiary butyl ether
DME	Dimethyl ether
CCU	Carbon capture and utilization
SMR	Steam methane reforming
PtG	Power-to-gas
TRL	Technology readiness level
LCA	Life cycle assessment
CCT	Catalytic cracking of tars
SER	Sorption enhanced reforming
WGS	Waster gas shift
EHC	Electrochemical hydrogen compression
AGR	Acid gas removal
MDEA	Methyl-di-ethanol-amine
ISO	International Organization for Standardization
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
GWP	Global warming potential
FEP	Freshwater eutrophication potential
ODP	Ozone depletion potential
FDP	Fossil depletion potential
FETP	Freshwater ecotoxicity potential
HTP	Human toxicity potential
MDP	Mineral depletion potential
POFP	Photochemical oxidant formation potential
TETP	Terrestrial ecotoxicity potential

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