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Process modelling and life cycle assessment of a carbon capture and conversion technology for methane production in indoor air and bioenergy environments

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ABSTRACT

The climate change crisis demands urgent action to prevent crossing tipping points that could jeopardize societal well-being. This research presents the process modelling and life cycle analysis (LCA) of a novel carbon capture and conversion system referred to as the "De-cent concept" (decentralised mitigation technology of CO_2 emissions). This innovative system is designed to capture CO_2 and convert it into methane suitable for grid injection, also providing high quality air. The technology consists of an absorption/desorption unit for CO_2 concentration and a bio-electrochemical system for methane conversion. The potential implementation of this system is first modelled using process simulation software (SuperPro Designer® v11) to obtain the mass and energy balances of methane production, considering two environments: indoor air and apple-based bioethanol production. Next, the environmental impacts of methane production, as well as the integration of the conversion technology into a bioethanol biorefinery, are assessed using the LCA methodology. Additionally, a sensitivity analysis is conducted to enhance the environmental performance of the products (methane and bioethanol) by identifying critical hotspots in these scenarios. The results indicate that methane production in indoor environments could represent a promising approach to low or even negative carbon emissions (in the 'cradle to gate' scope) when renewable energy sources such as photovoltaic systems support the technology's electricity supply. Furthermore, the environmental impact of bioethanol production could be reduced by approximately 55% to 84% by integrating carbon capture and conversion technology, together with renewable energy sources, into the design of the biorefinery. This research highlights the potential for energy generation in indoor environments and highly rich in CO_2 industrial streams, and the development of bioenergy models that simultaneously capture carbon and generate methane.

1. Introduction

The Sixth Assessment Report by the IPCC demonstrates that global surface temperature reached 1.1 °C above the 1850–1900 baseline during the 2011–2020 period due to human activities [1]. Urgent actions are needed to reduce greenhouse gas (GHG) emissions to meet the 1.5 °C target of the Paris Agreement and to minimize the likelihood of extreme weather events and the risk of crossing tipping points. In this context, balancing anthropogenic emissions from sources and removals by sinks in the European Union by 2050 has been the objective of the European Climate Law (European [2].

The necessity for reducing the CO_2 concentration in the air, rather than solely curtailing carbon emissions promote a group of technologies referred to as negative emission technologies [3]. From them, Carbon Capture and Utilization (CCU) technologies emerge to contribute to climate change mitigation, where carbon dioxide (CO_2) emitted, e.g.,

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from industrial facilities and power generation, can be captured and reused in various applications [4], serving as a valuable feedstock to produce fuels, chemicals, and materials in the transition to circular carbon systems Wen and Ierapetritou [5]. Additionally, Direct Air Capture (DAC) technologies can capture CO_2 from the air (e.g., using sorbents), aiming to achieve negative emissions and reduce atmospheric CO_2 concentrations [6,7]. This technology is also effective in indoor environments, improving air quality and human health [8], as well as the broad application of this technology is likely to improve the carbon footprint of buildings in the future [9]. In this regard, the combination of DAC with the utilisation of the captured CO_2 (DAC-U) represent a promising strategy toward carbon neutrality and efficient carbon circulation [10].

Carbon capture, utilization, and storage (CCUS) systems can be promising technologies for decarbonizing energy-intensive industries and reducing carbon emissions in the context of climate change. However, recommendations for implementing these systems should be contingent upon demonstrating the achievement of environmental benefits (e.g., emissions reductions) in the contexts where they can be implemented. As a technology focused on carbon emissions, and to prevent potential increases in other relevant environmental impacts (e.g., acidification, ozone depletion, particulate matter, etc.); life cycle assessment (LCA) is an effective and well-established methodology for evaluating and identifying the potential environmental benefits of implementing carbon capture and use/conversion technologies. The literature on the environmental assessment of CCU systems, through a life cycle perspective, has focused on their implementation coupled with energy production systems to reduce carbon emissions, mainly. For example, capturing CO_2 from electricity/cement production [11]; in municipal waste incineration system to produce chemicals or fuels production, where carbon capture reduces up to 50 % of the energy recovered [12]; the carbon intensity of CCU-derived e-fuel [13]; methanol and formic acid production from coal-fired power plants [14],the environmental evaluation of biogas management technologies (e.g., combustion, upgrading to natural gas, direct utilisation of CO_2, among others) where CCU provide one of the largest carbon savings [15]; consequential LCA on carbon capture and storage and CCU for combined heat and power plants, hydrothermal liquefaction, and gasification [16]; and the carbon footprint of carbon capture utilisation and storage technologies in coal chemical industry [17]). Additionally, Garcia-Garcia et al. [18] conducted a literature review of LCA application in CCU technologies, identifying environmental benefits in chemicals production compared to standard system, and suggesting an evaluation of various environmental impacts to avoid burden shifting to other impact categories. On the other hand, the LCA literature on DAC technologies have been less explored in the literature, although identifying resource-flows that require attention in the environmental evaluation of carbon capture systems. For example, electricity demand dominates carbon emissions in a hybrid DAC system, which integrates moisture swing adsorption and atmospheric water extraction technologies for the capture of CO_2 and water from ambient air [19]; negative carbon emissions per tonne of CO_2 captured can be obtained in renewable-based DAC technologies due to power demand is covered by renewable energy sources (e.g., solar, biomass) [20]; or that the CO_2 extracted from the atmosphere by DAC is crucial for the conversion of solar energy into synthetic natural gas imported from the Atacama Desert (Chile) to Belgium, despite the high contribution that DAC and land and sea transport have on carbon emissions [21].

In carbon capture and conversion technologies, bio-electrochemical systems (BES) have aroused the interest of the scientific community for the last two decades, because of their flexibility as they offer a sustainable pathway to capture and convert CO_2 into biofuels and valuable energy carriers [22]. In particular, the production of methane (CH_4) from CO_2 occurs at the biocathode, in presence of methanogenic and hydrogenotrophic biomass, in a process also known as microbial electromethanogenesis (MEM) [23]). Being the technology still developed at a laboratory scale, scarce LCA studies on BESs and EM have been reported in literature so far [24,25]; for example: i) the comparison of the environmental impact of microbial fuel cell in various configurations [26]; ii) the design of a continuous auto circulating bio-electrochemical reactor for wastewater treatment [27]; and the comparison of microbial fuel cell, microbial electrolysis cell, and microbial desalination cell systems (J. [28]. This technology presents great potential in terms of carbon conversion into valuable biofuels and commodity chemicals; however, its technology readiness level still stands low (e.g., 3–5), achieving at best pre-demo size and application. This issue is mainly caused by the difficulty of scaling up the size of the reactors, which might cause increased ohmic losses and decrease the overall efficiency of the process [29].

BES technologies represent a paradigm shift by valuing pollutants to convert them into energy, which, combined with DAC technology, can represent a promising path towards circular energy and construction sectors. However, the limited exploration of their environmental advantages or disadvantages must be evaluated before promoting their implementation in real-world settings. In this regard, this manuscript assesses the potential environmental impacts, through a life cycle perspective, of a novel carbon capture and conversion technology for methane generation through a two-step system: adsorption/desorption unit for CO_2 concentration and microbial electro-methanogenesis (i.e., BES) for its conversion into methane, considering two distinct sources of CO_2: i) indoor air and ii) emissions from a biorefinery facility. To our knowledge, no previous study has assessed the environmental impacts of this novel carbon capture and conversion technology developed at laboratory scale. Thus, the research combines process simulation and life cycle assessment approaches with two aims: i) to evaluate the potential environmental impacts when the technology is applied in indoor environments to determine the critical factors that may limit its implementation in the building sector; and ii) to assess the technology's ability to reduce the environmental burdens of bioethanol on an industrial scale, which has diverse market interest (e.g., the energy or chemical industries). We work with the hypothesis that a thorough LCA would target methane production from indoor and industrial environments as environmentally friendly practice, and the analysis will help identify further improvements that can be incorporated into the technology.

2. Materials and methods

The carbon capture and conversion (CC&C) technology proposed, the De-Cent (decentralised mitigation technology of CO_2 emissions) prototype, is evaluated in two environments. Firstly, an indoor air environment towards the implementation of green buildings, which aims e.g., a sustainable site design, indoor environmental quality, and conservation of materials and resources, among others. The indoor air pollutants generated like CO_2 can be captured and transformed into biofuels for internal use, displacing fossil fuels demand [9]. Secondly, the carbon capture and conversion technology is modelled in an apple-based bioethanol production, which produces a high-purity CO_2 stream (99 %) from the fermentation unit [30]. The biorefinery design is assumed to be in Chile, the leading exporter of apples in the Global South [31]. Additionally, a sensitivity analysis will be conducted to identify opportunities for improving the environmental performance of the carbon capture and conversion to methane technology in both indoor and bio-industrial settings. A general framework of this research is display in Fig. 1.

2.1. Carbon capture and conversion technology for methane production

The development of an innovative CC&C technology is the framework for the De-Cent project (see Fig. 2), which aims to develop a standalone and decentralized unit for converting CO_2 into methane of sufficient quality for grid injection, as well as providing purified air. The prototype consists of two units: i) an adsorption/desorption unit for CO_2

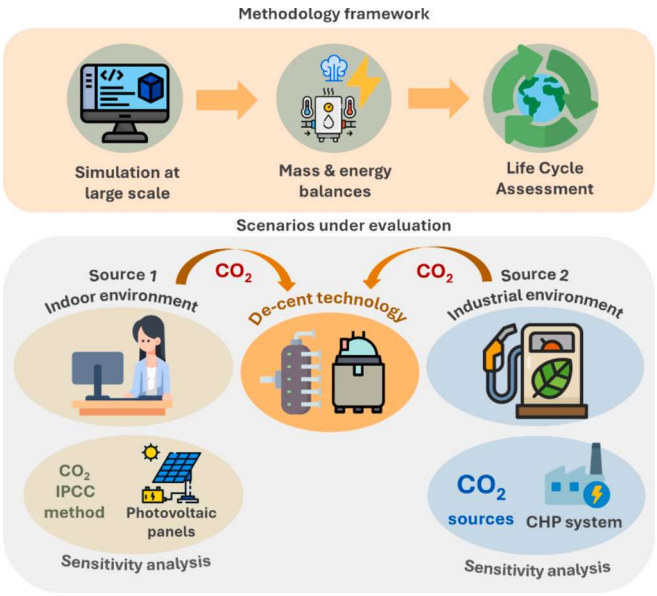

Fig. 1. Methodological framework and scenarios evaluated in this research.

concentration and ii) a microbial electro-methanogenesis unit for its conversion into methane.

The adsorption/desorption unit consisted of a CO_2-MCM (micro concentrator module) prototype. The adsorption phase consisted of indoor CO_2 capture using 4 adsorption columns filled with adsorbent material (50 % fumed silica – 50 % PEI LMW). The exhaustion time marked the instant when 95 % of the inlet CO_2 concentration was reached at the column outlet: thus, depending on the CO_2 load of the environment, the length of the adsorption phase lasted between 6 h and 1 d. Once exhaustion time was reached and the adsorbent saturated, the desorption chamber was heated at 80 °C by using heating plates to release the CO_2 captured from the indoor environment until complete desorption. The system and absorption/desorption cycles were controlled by an Arduino-based control system.

The microbial electromethanogenesis (MEM) unit consisted of a two-chambered, low gap commercial electrochemical reactor (Micro Flow Cell®, ElectroCell, Denmark), with a biocathode (carbon felt, projected surface of 0.001 m^2, Thermo Scientific, Germany) inoculated with anaerobic digester biomass. The MEM was operated in a two-electrode configuration, with a potentiostat (Bio-Logic SP50, France) controlling the current provided (galvanostatic mode) and recording the cell voltage and power consumption at different fixed current values (5–50 A m^{-2}). Experiments were conducted at room temperature (25 ± 3 °C), with no need for external heating. The cathodic chamber was fed with the indoor CO_2 gas captured by the CO_2-MCM or synthetic CO_2 (99.9 %, AirLiquide, Spain) as only carbon source. Experimental values obtained throughout the laboratory validation of the prototype were provided as a baseline for simulation. Bio-electrochemical CO_2 conversion into methane has demonstrated CO_2 conversion over 90 %, and a potential for biogas upgrading up to 95 %, with characteristics compatible with in-grid injection. A detailed description of the two units and the experimental tests carried out prior to this simulation can be find in [32]. The two units together consisted in the De-Cent prototype, also referred as CC&C technology in the scenarios simulated.

2.2. Process simulation

To simulate the carbon capture and conversion technology, firstly, to address the adsorption/desorption step, a GAC adsorption column was used to capture CO_2 from indoor air environment (e.g., room, office) or CO_2 emitted by industrial processes. Furthermore, due to the unavailability of a bio-electrochemical unit, a bioreactor was used to convert the captured CO_2 into methane with a purity of > 95 %. To connect both units, storage was also considered. The simulation was modelled using Superpro designer® software v11 Inc [33].

2.2.1. CC&C technology in indoor environment

In the case of indoor conditions, the simulation assumed the operation of a building with 20 rooms (total volume of 800 m^3), each occupied by five people. It is assumed that each person emits, on average, 16.2 L/h of CO_2 under sedentary conditions, as reported by Sakamoto et al. [34]. Based on the experimental analysis of the technology, it is considered an air composition of 2,000 ppm (i.e., 0.2 % of CO_2), which is the inlet flow of the adsorption stage. According to the literature, indoor CO_2 concentrations should generally be kept below 1000 ppm, although depending on the exposure time, the acceptable CO_2 concentration in healthy environments can vary between 500 and 3000 ppm [35]. In this regard, CO_2 levels can easily exceed 2000 ppm depending on the number of occupants and the volume of the indoor space (e.g., classrooms, transportation) [36,8]. Therefore, it is worth noting that, in addition to the conversion of carbon into methane, another objective of decent technology is to reduce CO_2 concentrations to levels below those harmful to health in indoor environments, providing purified air with a CO_2 content of 0.1 %.

Then, the inlet CO_2 content of the BES system ranges from 9 % to 20 % (an average value of app. 15 % was consider for simulation), with the remaining composition being O_2 and N_2 in the same ratio of atmospheric air. The electricity consumption of the bio-electrochemical system for methane production was 15.9 ± 5.7 kWh Nm^{-3}. High purity biomethane has an energy content of approximately 10.34 kWh Nm^{-3}. The produced methane has an overall higher energy cost than its energy power, but this value can be considered neglegible when implementing

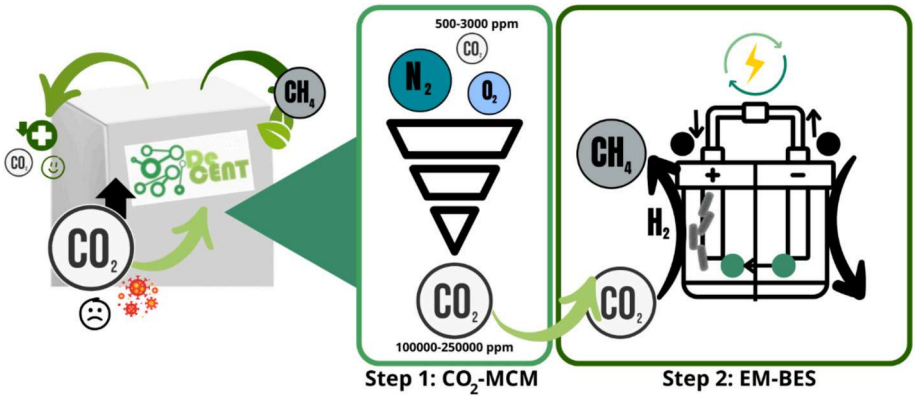

Fig. 2. De-Cent prototype scheme: 1) carbon capture unit (CO_2-MCM); 2) bio-electrochemical system for methane production (EM-BES).

the use of renewable energy. In either case, the process could grant an output of up to 9.86 kWh Nm^{-3} at the purity desired.

Although the quality of methane produced with the capture and conversion technology was not consistent over time, the values achieved —close to the target for reinjection into the gas pipeline network (<2% CO_2, <1% O_2, and > 95 % methane)—were within acceptable limits for captured CO_2.

2.2.2. CC&C technology in industrial context: bioethanol production

The biorefinery design is based on processing 4,000 tonnes of apple pomace, annually, which represents the amount generated by one of the major dehydrated apple processing plants in Chile, where the designed platform is intended to be integrated. The platform includes four main stages: i) pre-treatment of the pomace, ii) fermentation, and iii) purification of bioethanol, and iv) a carbon capture and conversion system.

The bioethanol production process using apple pomace is based on the lab-scale work conducted by Hernández et al. [31]. In the pre-treatment phase, the apple pomace is milled and pressed to obtain the liquid phase, which is then sent to the fermentation unit. The solid phase is assumed to be directed to a boiler unit for energy recovery (i.e., steam generation). The fermentation process takes place over 144 h at 30 °C [31], after which ethanol is recovered and purified through distillation columns. The vinasse by-product, obtained from the bottom of the columns, is sent to a biogas generation unit (i.e., anaerobic digestion process), modelled following the work of Estévez et al. [37]. After distillation, the ethanol is dehydrated to achieve a purity of 99.5 %. For more details regarding bioethanol production, please see the work by Hernández et al. [31].

The CC&C technology is located next to the fermentation section, where the off-gas from this process, which is highly concentrated in CO_2, is processed without additional refinement. Furthermore, CO_2 emissions from the boiler, which generates steam from the solid biomass stream (after the pressing step), are also considered, providing another carbon source for the CC&C technology.

2.3. Life cycle assessment

2.3.1. Aim and scope

This attributional life cycle assessment aims to evaluate the potential benefits of a carbon capture and conversion technology for methane generation in both indoor and industrial environments, exemplifying the latter with a bioethanol biorefinery. For both carbon capture and conversion pathways, a *cradle-to-gate* approach is followed as the system boundary. Fig. 3a illustrates the boundaries of the production of methane and purified air, through the CC&C technology, in an indoor environment, while Fig. 3b shows the boundaries of bioethanol production with the CC&C technology.

In an LCA study, the functional unit (FU) serves as a reference unit for reporting results and quantifying the function of the product system being analysed [38]. Hence, due to the two contexts of application of the CC&C technology, two alternatives were evaluated. Firstly, when the CC&C technology is used in the indoor environment, the FU is 1 m^3 of methane produced. Secondly, in an industrial context, where CC&C technology is joined with bioethanol production, the FU is defined as 1 kg of bioethanol (the primary product of the system). Thus, it is expected to estimate the potential impact reduction related to bioethanol production, through the methane generation with the CC&C technology, which is used to reduce the energy demand of the biorefinery. For comparison purposes of the industrial context, a baseline scenario is established, which involves the production of bioethanol without CC&C technology, and using fossil sources (e.g., natural gas for steam production).

2.3.2. Life cycle inventory

The life cycle inventory (LCI) of the foreground system was developed using a *"bottom-up"* approach, incorporating the mass and energy balances of the biorefinery design through the Superpro designer® software v11 Inc [33]. LCIs of methane production in indoor environments and bioethanol production are displayed in Table 1 and Table 2, respectively. For methane production in an indoor environment, an average European electricity generation mix was considered in the modelling, sourced from the Ecoinvent® v3.10 database [39]. Furthermore, absorbent materials used in the absorption column were excluded from the LCI due to a lack of data in the Ecoinvent® database. No allocation procedure of environmental loads was assumed between

Table 1
Mass and energy balance of methane production in indoor environment.

Stage	Input	Value	Output	Value
Adsorption	Air (0.2 % CO_2) (m^3)	800	Purified Air (0.1 % CO_2) (m^3)	799.2
	Electricity (kWh)	2.47		
Bio-electrochemical	Air (~15 % CO_2) (m^3)	0.8	Biogas (95 % CH_4) (m^3)	0.72
	Electricity (kWh)	11.44	Water (kg)	0.02

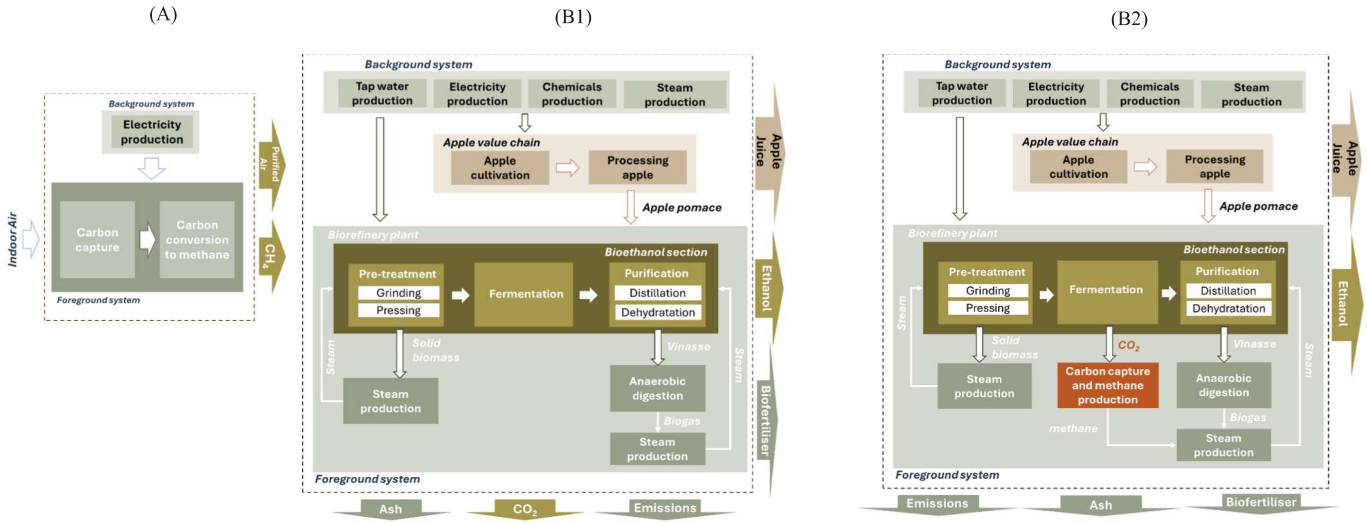

Fig. 3. System boundary of the methane production in indoor (A) and industrial sources (B): bioethanol production without (B1) and with (B2) the carbon capture and conversion technology.

Table 2
Life cycle inventory of CO_2 industrial).

Section of the biorefinery	Inputs from Technosphere	Value	Outputs to Technosphere	Value
Pre-treatment	Pomace (t)	18.45	Residual biomass (t) [10]	0.76
	Electricity (kWh)	375.97		
Fermentation	Nutrient (t)	0.01	CO_2 (t) [4]	1.11
	Water (t)	0.02		
	Chilled water (t)	170.19		
	Electricity (MWh)	1.72		
Purification	Cooling water (t)	1634.2	Ethanol (t)	1
	Steam (t) [1,2, 3]	9.39	Vinasse to treatment (t)	15.62
	Chilled water (t)	8.29		
	Electricity (kWh)	2.92		
Steam generation (from solid flow) [6]	Water (t)	0.45	CO_2 (t) [5]	0.25
	Electricity (kWh)	0.07	Ash (t)	0.12
Vinasse treatment (anaerobic digestion and steam generation	Water (t)	7.1	Fertiliser sludge (m^3)	16.3
	Sodium bicarbonate (g)	33.5	*Emissions to air*	
	Polymer (kg)	2.1	CO_2 (kg)	99.6
	Electricity (kWh)	23.7	NH_3 (g)	119.3
			N_2O (g)	1.1
			CH_4 (g)	69.6
			H_2O (kg)	0.9
			Emissions to water	
			COD (g)	13.9
			Total N (g)	3.1
			Total P (mg)	3.5
			Total solids (mg)	28.3
Carbon capture and cogeneration system	Electricity (MWh) [7]	10.06	Steam (t) [9]	5.64
	Steam (t)	0.11	CO_2 (t)	1.12
	Water (t) [8]	6.35	H_2 (kg)	2.07
			H_2O (air) (t)	0.90

CCBF + CHP: Bioethanol production with CC&C technology using CO_2 from fermentation and boiler gas streams to the CHP system.
[1] Steam quantity without CC&C technology.
[2] Steam quantity with CC&C technology change to 3.10 t per FU.
[3] Steam quantity with CC&C technology change to 0 kg per FU in sub-scenario CCBF + CHP.
[4] Amount of CO_2 sent to CC&C technology.
[5] Amount of CO_2 sent to CC&C technology in sub-scenario CCBF + CHP (plus the CO_2 from the fermentation unit).
[6] No implemented in sub-scenario CCBF + CHP.
[7] Change to 27.33 kWh in CCBF + CHP.
[8] Change to 15.6 t in CCBF + CHP.
[9] Co-product in sub-scenario CCBF + CHP.
[10] Amount in CCBF + CHP.
Source: bioethanol production (based on 1 tonne

methane and purified air, allocating all impacts to the first one, as air is considered a "free good" (for a potential economic allocation), and a mass allocation would not properly represent the main function of the system (i.e., methane production).

For the bioethanol scenario, the Chilean electricity mix generation for the year 2023 was modelled [40]. The LCI for apple juice production was taken from Cheng et al. [41]. The apple orchards and manufacturing plant are located in the Maule region in the Central Valley of Chile. The transportation distance of fresh apples from the orchards to the manufacturing facility was assumed to be 300 km, based on previous studies [42,43]. The transport of the pomace to the biorefinery was neglected, assuming that the biorefinery is co-located with the juice manufacturing plant. An economic allocation approach was used to allocate loads between the juice product and the pomace. The average market price of apple juice during 2018–2022 was 1.5 $\$\cdot kg^{-1}$ [44], while the price for pomace was 0.01 $\$\cdot kg^{-1}$ based on information from the manufacturing plant.

Infrastructure-related processes were not considered in the environmental modelling, as the environmental impacts associated with the construction and installation of biorefinery plants during their lifetime are negligible in both cases [45]. The LCI data were modelled using SimaPro® v10.0.0.2 software [46], and background processes, such as the apple cultivation stage in Chile, were obtained from the Ecoinvent® v3.10 database [39]. No allocation of environmental loads was made between bioethanol and the biofertiliser (i.e., digestate) produced from anaerobic digestion, as the economic contribution of biofertiliser is marginal (≤1%) compared to bioethanol [47,48]. Thus, all burdens were attributed to the bioethanol.

2.3.3. *Life cycle impact assessment*

The characterisation factors from the ReCiPe 2016 (H) V1.07 / World (2010) (H) method [49]) were used to translate the inventory data from the scenarios analysed. The selection of this method is based on its use of global characterisation factors, and its frequent updates, making it one of the most widely applied impact assessment methods [50]. The impact categories selected for the analysed scenarios were Global Warming (GW), Stratospheric Ozone Depletion (SOD), Particulate Matter (PM), Terrestrial Acidification (TA), Freshwater Eutrophication (FE), Marine Eutrophication (ME), Terrestrial Ecotoxicity (TET), Freshwater Ecotoxicity (FET), Human Carcinogenic Toxicity (HT); Land Use (LU), Fossil Resource Scarcity (FRS), and Water Consumption (WC). Furthermore, the Cumulative Energy Demand (CED) (LHV) indicator was also considered to provide an energy balance of the system.

2.3.4. *Sensitivity analysis*

Sensitivity analysis was conducted after estimating the environmental burdens and identifying those stages with the most significant impacts for both methane production in an indoor and industrial context. Regarding the indoor environment, as electricity is the key factor demand for the technology, we consider a photovoltaic system to be coupled with the CC&C technology, as well as different European countries that have a relevant contribution of renewable sources such as Sweden and Spain. Furthermore, the IPCC 2021 GWP100 (incl. CO_2 uptake) method V1.01 was also considered to focus on the CO_2 emissions balance in the indoor context.

In the case of the industrial environment (see Fig. 4), two changes were considered in the biorefinery design of the bioethanol production, while the electricity demand of the CC&C technology is supplied by photovoltaic source: i) the sub-scenario CC&C + CHP (see Fig. 4a), where the methane is sent to a cogeneration (CHP) system to obtain electricity and steam; and ii) the sub-scenario CCBF + CHP (see Fig. 4b), where the CO_2 is capture from fermentation and boiler units, and the methane is sent to a CHP system to obtain steam and electricity. In this sub-scenario, the solid mass flow after the pressing process is not burned and used as animal feeding. Therefore, there are no related emissions and no ash disposal by combustion. Additionally, as the steam generated from the CHP excess the demand of the biorefinery, the excess amount is considered a co-product. Thus, an economic allocation is considered to distribute the loads between ethanol and steam. The price of steam considered was 12 $/t Inc [33], while for the ethanol a price of 0.73 $/kg was assumed (Bussiness [51].

3. Results and discussion

3.1. *CC&C technology applied in an indoor environment*

As mentioned in Section 2.3.2, although the CC&C generates both methane and purified quality air as process outputs, all emissions were

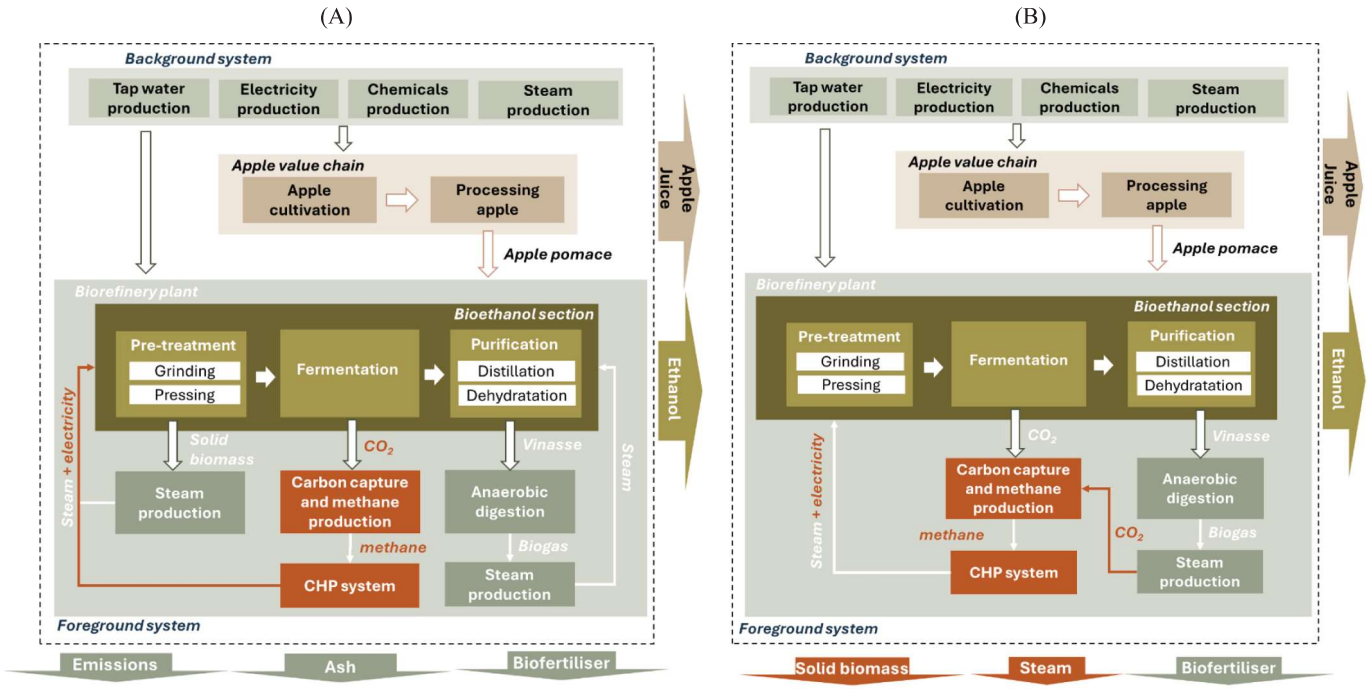

Fig. 4. System boundaries of the sensitivity analysis of industrial environmental: CO_2 capture from fermentation section used for CHP system (A), and CO_2 capture from fermentation and boiler unit used for CHP system (B).

allocated to methane, connected to its global warming potential and the potential emissions deriving from its further use in cogeneration/heating facilities. The environmental profile of methane production using indoor air is presented in Table 3, which shows that the GW category is about 6.22 kg of CO_2-eq per m^3. In this system, the primary contributor to environmental impact across all categories is the BES stage, due to the high electricity demand of the system (15.9 kWh average per m^3 of methane).

Different environmental profiles can be obtained from the Ecoinvent® database v3.10 for biomethane (high pressure) production alternatives, for example, biomethane from synthetic gas; biomethane (96 %) from biogas purification by amino washing, membrane technique, and by pressure swing adsorption. In the case of GW profile, the impacts varied from 0.17 (biomethane from biogas purification by membrane technique) to 0.34 (biomethane from synthetic gas, wood, fluidised technology) kg CO_2-eq per m^3. As shown in Fig. 5, methane production through CC&C technology has the highest impact in 8 out of 14 categories evaluated, except for TET, FET, MET, HT, HNT, and LU categories, where the impacts are significantly lower than those of these pathways. By considering an average European electricity generation mix, the main sources of the environmental impacts are countries such as Germany, Poland, and Italy, which are mainly based on fossil sources (e.g., hard coal, lignite, and natural gas). So, renewable sources for electricity supply will be essential for the promotion of this carbon capture and conversion technology to be competitive compared to the biomethane production alternatives presented.

3.1.1. *Sensitivity analysis of methane production in an indoor environment*

The influence of the electricity generation profile and renewable energy use is shown in Fig. 6, which depicts the variation in the electricity generation mix and the use of a photovoltaic source for methane production in indoor environments. Comparing electricity mix generation, results indicate that the Swedish electricity generation performs better in most categories, except for LU and WC. For instance, the Swedish and Spanish profile results in about 91 % and 41 % reduction in the GW category (0.50 and 3.36 kg CO_2-eq, respectively) compared to the average European mix. Sweden encompassed notable reductions in

Table 3
Environmental profile of methane in indoor environment (FU: 1 m^3) and bioethanol production without and with the CC technology (FU: 1 kg ethanol).

Impact category	Unit	Indoor source	Industrial source	
		Methane in indoor environment	Bioethanol without the CC&C technology	Bioethanol with the CC&C technology
Global warming	kg CO_2 eq	6.22E + 00	3.95E + 00	4.55E + 00
Stratospheric ozone depletion	kg CFC_{11} eq	2.78E-06	5.85E-07	1.57E-06
Fine particulate matter formation	kg $PM_{2.5}$ eq	8.19E-03	8.45E-03	3.24E-02
Terrestrial acidification	kg SO_2 eq	2.03E-02	1.03E-02	1.60E-02
Freshwater eutrophication	kg P eq	6.58E-03	1.51E-03	3.76E-03
Marine eutrophication	kg N eq	4.37E-04	5.02E-05	1.24E-04
Terrestrial ecotoxicity	kg 1,4-DCB	2.26E-03	1.55E-01	8.82E-01
Freshwater ecotoxicity	kg 1,4-DCB	7.14E-06	1.03E-05	2.55E-05
Marine ecotoxicity	kg 1,4-DCB	1.34E-05	1.00E-04	5.04E-04
Human carcinogenic toxicity	kg 1,4-DCB	5.20E-04	1.12E-03	1.73E-03
Human non-carcinogenic toxicity	kg 1,4-DCB	5.59E-04	1.05E-02	5.58E-02
Land use	m^2a crop eq	1.61E-01	3.89E-02	1.95E-01
Fossil resource scarcity	kg oil eq	8.21E-01	2.63E-01	5.01E-01
Water consumption	m^3	8.90E-02	1.83E + 00	1.88E + 00

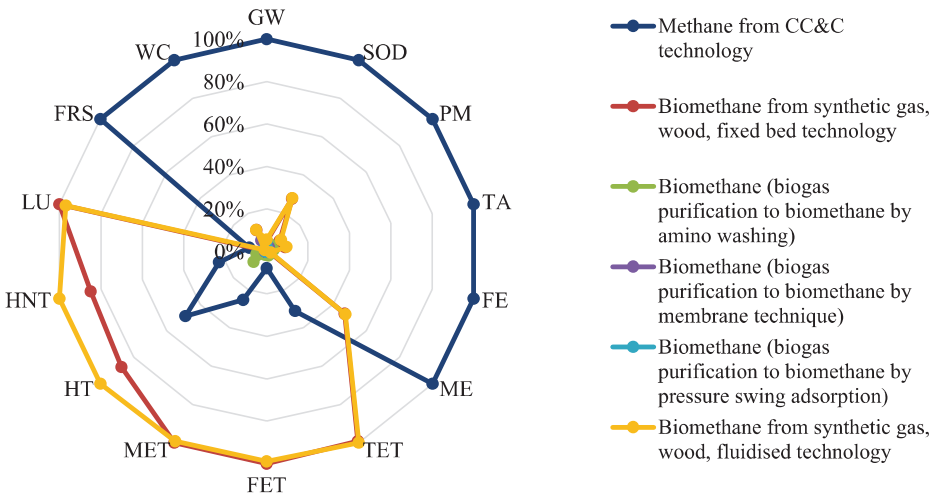

Fig. 5. Comparison of the environmental profiles of different methane production routes. AD: Anaerobic digestion. GW: Global Warming; SOD: Stratospheric Ozone Depletion; PM: Particulate Matter; TA: Terrestrial Acidification; FE: Freshwater Eutrophication, ME: Marine Eutrophication; TET: Terrestrial Ecotoxicity; FET: Freshwater Ecotoxicity; HT: Human Carcinogenic Toxicity; LU: Land Use; FRS: Fossil Resource Scarcity; WC: Water Consumption; AD: Anaerobic digestion.

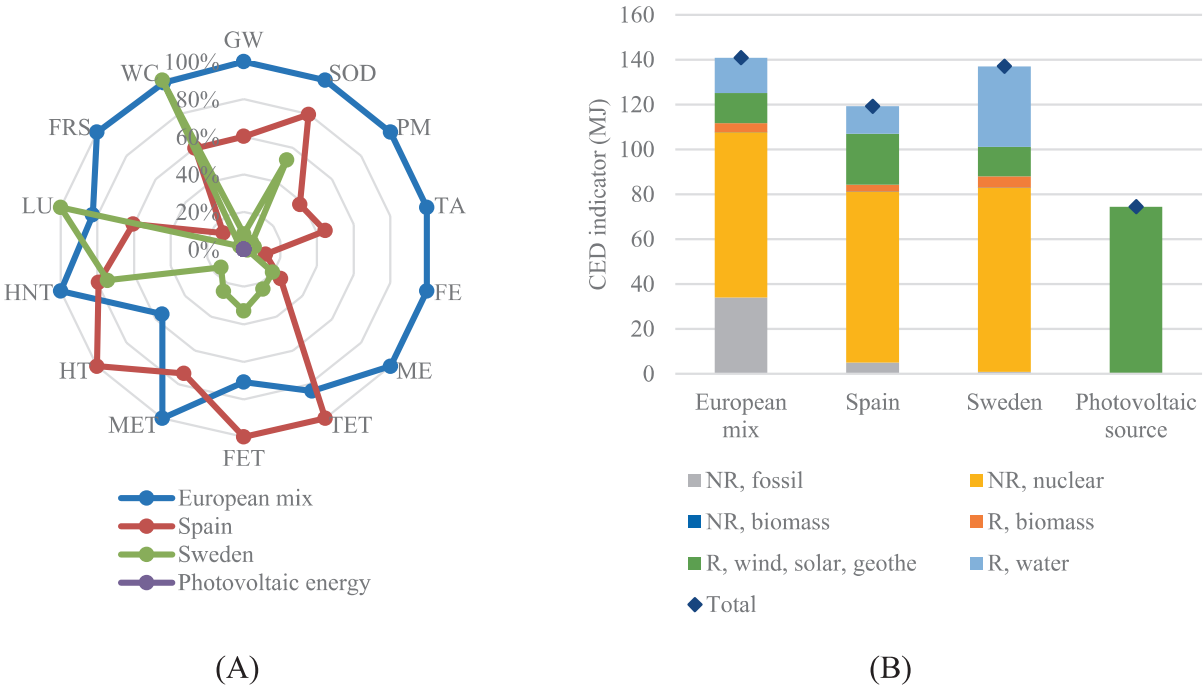

Fig. 6. Sensitivity analysis of the influence of electricity profile of the CC&C technology in indoor environment (A), and CED indicator of electricity mix generation (B). GW: Global Warming; SOD: Stratospheric Ozone Depletion; PM: Particulate Matter; TA: Terrestrial Acidification; FE: Freshwater Eutrophication, ME: Marine Eutrophication; TET: Terrestrial Ecotoxicity; FET: Freshwater Ecotoxicity; HT: Human Carcinogenic Toxicity; LU: Land Use; FRS: Fossil Resource Scarcity; WC: Water Consumption. NR: Non-renewable; R: Renewable.

FE (98 %), FRS (97 %), PM (94 %), TA (94 %), while Spain highlights in FE (88 %), FRS (86 %), ME (75 %), and PM (62 %).

When a photovoltaic system (mounted on a building's slanted roof) is used to supply electricity to the CC&C technology, all environmental impact indicators are significantly reduced compared to using grid electricity. For instance, the GW indicator reached a value of 27.75 mg CO_2-eq per m^3 of methane produced. The use of renewable sources demonstrates that carbon capture and conversion technology offer even better environmental performance than the traditional biogas production routes mentioned in previous section 3.1. This shows that the use of renewable energy is essential to support the implementation of this technology. Furthermore, the GW profile per kg of methane in an indoor environment based on an electricity photovoltaic source (27.75 mg CO_2-eq) is competitive regarding the natural gas (70–98 % methane) production with a value of 0.46 kg CO_2-eq per m^3, according to the ecoinvent® database, as a fossil reference system.

Regarding the CED indicator, per m^3 of methane generated, European mix generation encompassed the highest energy demand (140.9 MJ), followed by Sweden (137.1 MJ), and Spain (119.3 MJ), where nuclear source plays a relevant role. The photovoltaic energy source represents a reduction of 47 % with respect to the European mix.

Another sensitivity analysis was conducted by changing the impact assessment method for the Global Warming indicator, using the IPCC 2021 method. Fig. 7 shows the GW profile for methane generation using the European electricity grid, Spanish, and Sweden electricity generation, as well as the use of a photovoltaic system. Results, per m^3 of

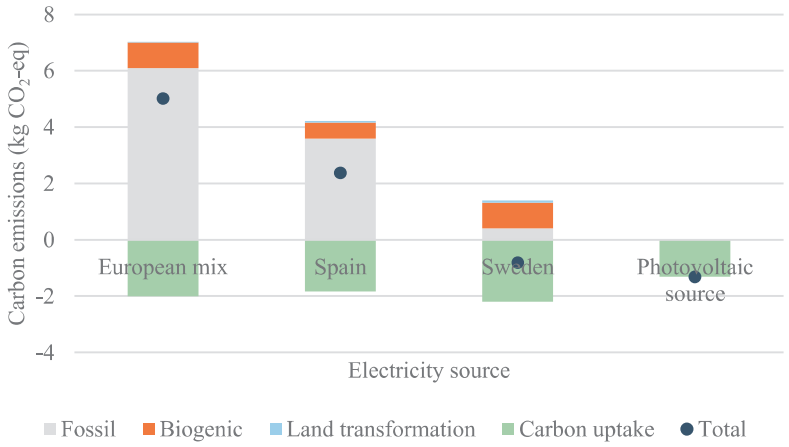

Fig. 7. Carbon balance in methane generation in indoor environment using the IPCC 2021 (Global Warming 100) method with the European, Spanish electricity profile and a photovoltaic system.

methane, indicate that the GW values were 5.02, 2.38, −0.81 and − 1.31 kg CO_2-eq for the European, Spanish and Swedish electricity mix, and the photovoltaic system, respectively. As shown in Fig. 7, the carbon emissions from methane production using the CC&C technology based on the European and Spanish electricity grid are mainly due to fossil fuel sources in the generation mix, which contrasts with the advantages of a high contribution of renewable energy to the electricity grid for the operation of CC&C technology. Thus, in addition to a photovoltaic source, a cleaner electricity generation grid also presents a competitive scenario for the implementation of CC&C technology compared to its fossil fuel-based counterpart.

3.1.2. Comparison of environmental profile of methane production in indoor environment with findings of the literature

Only the study carried out by Fan et al., [52] assess the global warming impacts of a membrane-based DAC technology for methane generation in three indoor environments such as: ambient air (400 ppm CO_2 concentration), office (800 ppm CO_2 concentration) and classroom (1000 ppm CO_2 concentration). The electricity required to operate the technology was sourced from solar power, and hydrogen was externally supplied for the thermal conversion of CO_2 to methane. The operation phase ranges between 1455.8 and 1654 kg of CO_2-eq for the classroom and ambient air scenarios, respectively. Based on this work, if we consider the reported emissions (and credits) of the manufacturing, operation and maintenance stages, as well as the annual amount of methane generated (46.38 m^3), the carbon emissions ranges between 3.7 and 4.0 kg CO_2-eq per m^3 of methane. This is aligned with the GW profile of the CC&C technology with a Spanish electricity grid (3.74 kg CO_2-eq). The main differences with this manuscript are: i) a cradle-to-grave approach, where landfill and incineration were the end-of-life stages; ii) the functional unit: 1 kg of CO_2 captured per day (i.e., the capacity of the technology); iii) the assumption of environmental credits due to the avoided import of natural gas and the electricity recovered; iv) electricity consumption: 5.5 kWh of solar electricity to produce 91.1 g CH_4 per day.

Regarding related studies considering direct air capture technology to produce methane, Choe et al. [53] evaluated methane production through syngas methanation, where CO_2 is collected using DAC, and syngas is produced by solid oxide electrolysis (SOE) with renewable-based electricity. According to these authors, results depend on the system scale, reaching net emissions, per MJ, for onshore wind, offshore wind, and solar PV are − 0.030, −0.030, and − 0.015 kg CO_2-eq at 1 MW SOE system scale, and − 0.031, −0.030, and − 0.016 kg CO_2-eq at 10 MW SOE system scale, respectively. Additionally, Badger et al., [54] performed a cradle-to-gate LCA of synthetic methanol production, integrating low-temperature solid sorbent DAC systems with renewable energy sources and green hydrogen. They found that hydroelectric and wind power were the most sustainable options, showing the lowest GWP impacts at − 2.53 and − 2.39 kg CO_2-eq per kg methanol produced. Finally, Huang et al. [55] performed a LCA of an integrated DAC and methanation system using dual function material (i.e., catalytic and adsorbent components), reaching a minimum amount of 0.706 kg CO_2-eq per kilogram captured CO_2 in the operating conditions analysed. These findings support the relevance of using renewable energy sources for carbon and conversion technologies similarly to this study.

3.2. CC&C technology applied in an industrial context: bioethanol production

3.2.1. Bioethanol production without and with the De-Cent concept

Table 3 and Fig. 8a displays the environmental profile and contribution analysis of the bioethanol production when the CC&C technology is not considered, to compare it when the technology is applied. From this, purification is the critical process representing the main contributor in 8 out of 14 categories analysed. This is motivated mainly by the demand of heating and cooling agents in the distillation process, i.e., steam and cooling water, respectively. The fermentation stage is the hotspot in categories such as PM, FET, TET, HNT, and LU due to electricity demand.

When the technology is introduced in the biorefinery design to capture and convert CO_2 from the fermentation section, the environmental profile of this product is increased in all impact categories analysed (see Table 3). In the case of GW category, the burdens were increased by about 13 %, reaching a profile of 4.55 kg CO_2-eq per kg of product. The carbon and conversion stage is the main contributor to the environmental profile of bioethanol in the CC&C technology scenario (see Fig. 8b), in most of the categories studied (see Fig. 8b). The high electricity demand during the BES phase is the main reason for these results, as well as the significant contribution of fossil fuels to the European electricity grid, as mentioned in section 3.1.1.

3.2.2. Sensitivity analysis of bioethanol production with the CC&C technology

3.2.2.1. Sub-scenario CC&C + CHP: bioethanol production with CC&C technology and CHP system for electricity and steam generation. Since the steam and electricity are key elementary flows in the biorefinery design, the CO_2 captured and converted from the fermentation section was used for obtaining these components through a CHP system.

Results indicate that, regarding the baseline scenario production of

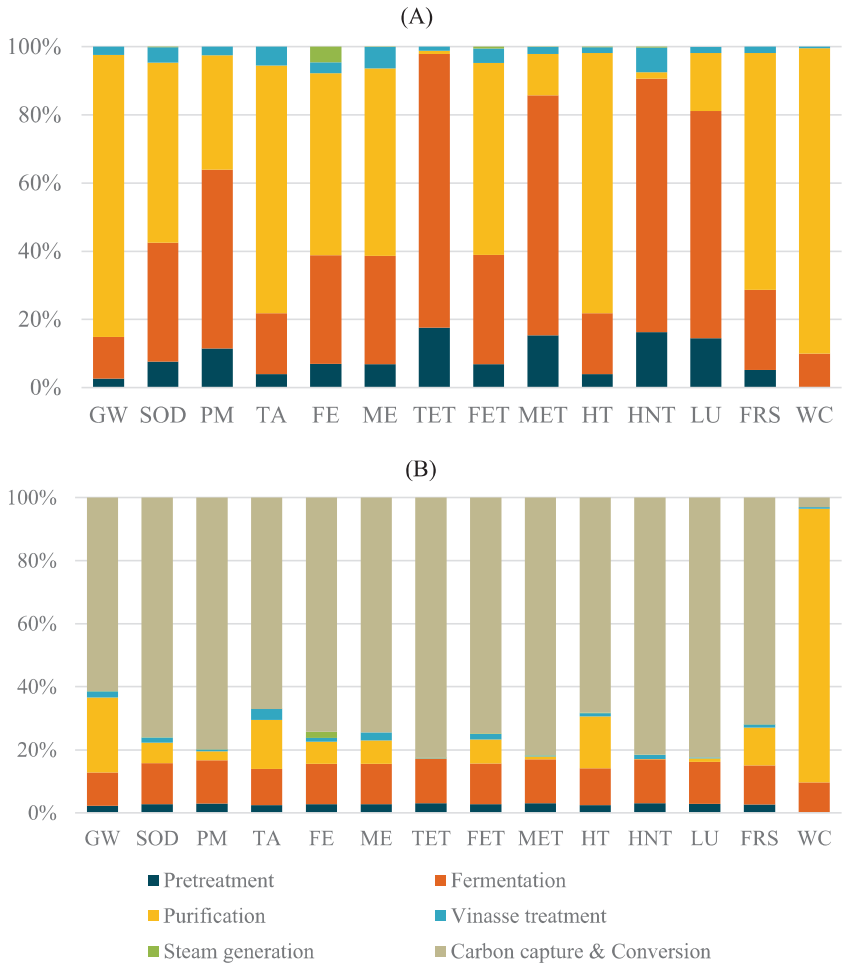

Fig. 8. Contribution analysis of bioethanol production without (A) and with (B) the CC&C technology. GW: Global Warming; SOD: Stratospheric Ozone Depletion; PM: Particulate Matter; TA: Terrestrial Acidification; FE: Freshwater Eutrophication, ME: Marine Eutrophication; TET: Terrestrial Ecotoxicity; FET: Freshwater Ecotoxicity; HT: Human Carcinogenic Toxicity; LU: Land Use; FRS: Fossil Resource Scarcity; WC: Water Consumption.

bioethanol (without the technology), this strategy improved the environmental performance in almost all impact categories analysed, except for the LU category, due to the steam demand of the CC&C technology is supplied by cogeneration systems. In the case of the GW category, carbon emissions were reduced from 3.95 to 1.77 kg CO_2-eq per kg of bioethanol (i.e., about 55 %). Other large reductions were observed in categories such as HT (50 %), TA (48 %), and FRS (46 %). Furthermore, the contribution analysis (see Fig. 9a) indicates that the fermentation section is the main contributor in most of the impact categories with contributions ranging from 44 % (FRS) to 81 % (TET). Exceptions occur in categories such as GW, TA, HT, and WC, where purification stage remains as a hotspot.

3.2.2.2. Sub-scenario CCBF + CHP: bioethanol production with CC&C technology using CO_2 emissions from fermentation and boiler flue gas streams to the CHP system. Previously, CC&C technology considered CO_2 capture and conversion only from the fermentation section. Here, the results presented consider the carbon capture and conversion from both the fermentation section and the CO_2 emitted by the boiler unit after anaerobic digestion.

In this sub-scenario, the environmental impacts were drastically reduced compared to the baseline of bioethanol production without the CC&C technology. For instance, the GW is reduced about 84 %, from 3.95 to 0.64 kg CO_2-eq per kg of bioethanol. Other relevant decreases were observed in categories such as HT (77 %), TA (74 %), FRS (71 %), FE and FET (60 %). A marginal increase of about 3 % is observed in WC, due to the cooling water demand for decreasing the temperature of streams from the boiler to the 80 °C required by the carbon capture technology. Since the steam demand by the biorefinery is completely supplied through the methane produced by the CC&C technology, changes in the contribution of the stages are observed (see Fig. 9b), where the purification section is the hotspot only in WC category due to cooling water demand. Fermentation appears to be the area requiring further improvement, with a focus on electricity demand.

Additionally, when using a photovoltaic energy source, the CC&C technology improved all environmental impact categories of the bioethanol production, as highlighted in Fig. 10a with all sub-scenarios evaluated. Further strategies can be implemented to improve environmental performance of the bioproduct analysed throughout its life-cycle. However, the CED indicator (see Fig. 10b) suggests that the use of CC&C technology, although it reduces the energy demand from fossil sources, also leads to higher energy demand than the reference scenario. This is particularly significant when electricity demand is met by renewable energy sources (solar in this case), where the demand increases five-times. This demonstrates the multi-criteria perspective of the results of a life cycle analysis, where improvements in certain categories may imply worse results in other environmental burdens.

3.3. Comparison of environmental profile of bioethanol with CC&C technology with literature

The estimation of environmental impacts, following an LCA

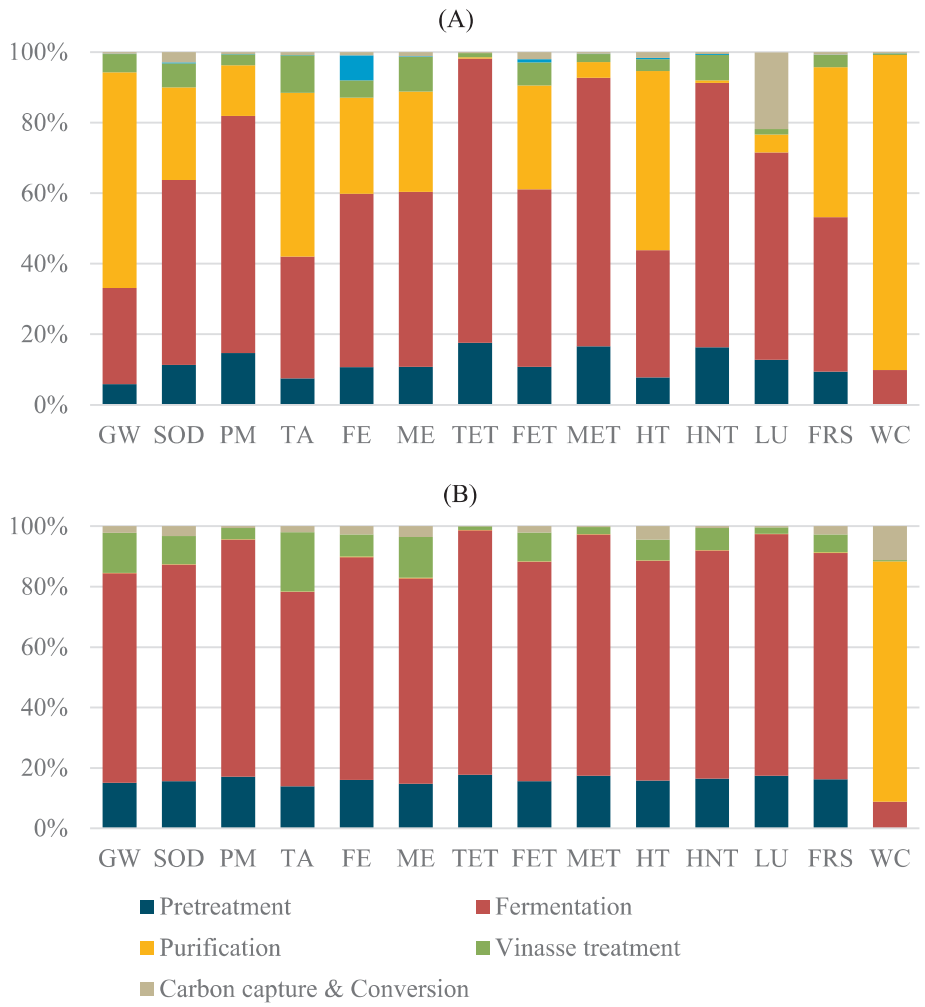

Fig. 9. Contribution analysis in sensitivity analysis of bioethanol production: (A) CC&C technology using methane in a CHP system, based on CO_2 from fermentation; and (B) CO_2 from fermentation and boiler units. GW: Global Warming; SOD: Stratospheric Ozone Depletion; PM: Particulate Matter; TA: Terrestrial Acidification; FE: Freshwater Eutrophication, ME: Marine Eutrophication; TET: Terrestrial Ecotoxicity; FET: Freshwater Ecotoxicity; HT: Human Carcinogenic Toxicity; LU: Land Use; FRS: Fossil Resource Scarcity; WC: Water Consumption.

approach, of bioethanol production based on different biomass sources has been reported in the literature, particularly focused on GHG emissions, and using different types of functional units such as mass, volume, and energy-based approaches. Regarding a mass-based FU, the environmental profiles of bioethanol using maize grain and stover, sugarcane, sugar beet and wheat, were reported by Muñoz et al. [56]. They estimated GHG emissions ranging from 0.7 to 1.5 kg CO_2-eq per kg ethanol, under a cradle-to-gate approach. High impacts were obtained using barley straw and brewer's spent grains by González-García et al. [57], who estimated an overall GW of 7.39 kg CO_2-eq per kg product, with autohydrolysis pre-treatment as the hotspot stage. Additionally, molasses-based bioethanol, literature values range from 0.37 to 3.26 kg CO_2-eq/kg ethanol [58,59], whereas Thapa et al. [60] reached a GW value of 0.044 kg CO_2 per kg of bioethanol, when it is produced together with methane and hydrogen. In that system, bioethanol is the second most important contributor after hydrogen production. Using the whole-plant corn for bioethanol production can represents impacts from1.15 to 1.39 kg of CO_2-eq per kg of product, based on the scenario evaluated [61]. These results contrast significantly with our findings in the Global Warming category, demonstrating the benefit of using the CC&C technology integrated with a renewable energy source, for improving the environmental performance of the bioethanol production.

Considering a volume-based FU, it has been reported that using cassava root, cassava straw and whole-plant cassava resulted in emissions of 1.74, 2.93, and 1.55 kg CO_2-eq per L of ethanol, respectively [62]. Similar results were achieved by Wang et al. [63] using sweet potato, where a GW profile of 1.47 kg CO_2-eq per L of product. The authors reported that their sensitivity analysis indicates that steam demand during bioethanol conversion has the most significant impact, reflecting the opportunity of the CC&C technology to reduce energy consumption in bioethanol production. A close value of 1.5 kg CO_2-eq per L of product was reached using rice straw by [64], although lower carbon impacts can be obtained with Cassava (0.425 kg CO_2-eq) and Cane molasses (0.558 kg CO_2-eq) per L of bioethanol, although using non-renewable fuel sources for process energy could increase the environmental profile. Furthermore, Santoyo-Castelazo et al. [65] used sugarcane bagasse through a gasification process to achieve a profile of about 8.4 and 26.7 kg CO_2-eq per L, when a cradle-to-gate or cradle-to-grave are considered, respectively. Recently, Patel & Singh [66] reported a GW value per L of ethanol of 0.37 and 1.1 kg CO_2-eq using corn stover and cassava straw, respectively. The cassava straw represents the highest impacts in this category, because of emissions released during the feedstock production (67 %) and hydrolysis process (31 %), mainly. Whether 1 L is used as FU in this research (considering an ethanol density of 0.789 kg/L), a GW profile of about 1.41 and 0.50 kg of CO_2-eq is estimated for the scenarios CC&C + CHP and CCBF + CHP, respectively, which is aligned with previous mentioned studies and lower than them in some cases.

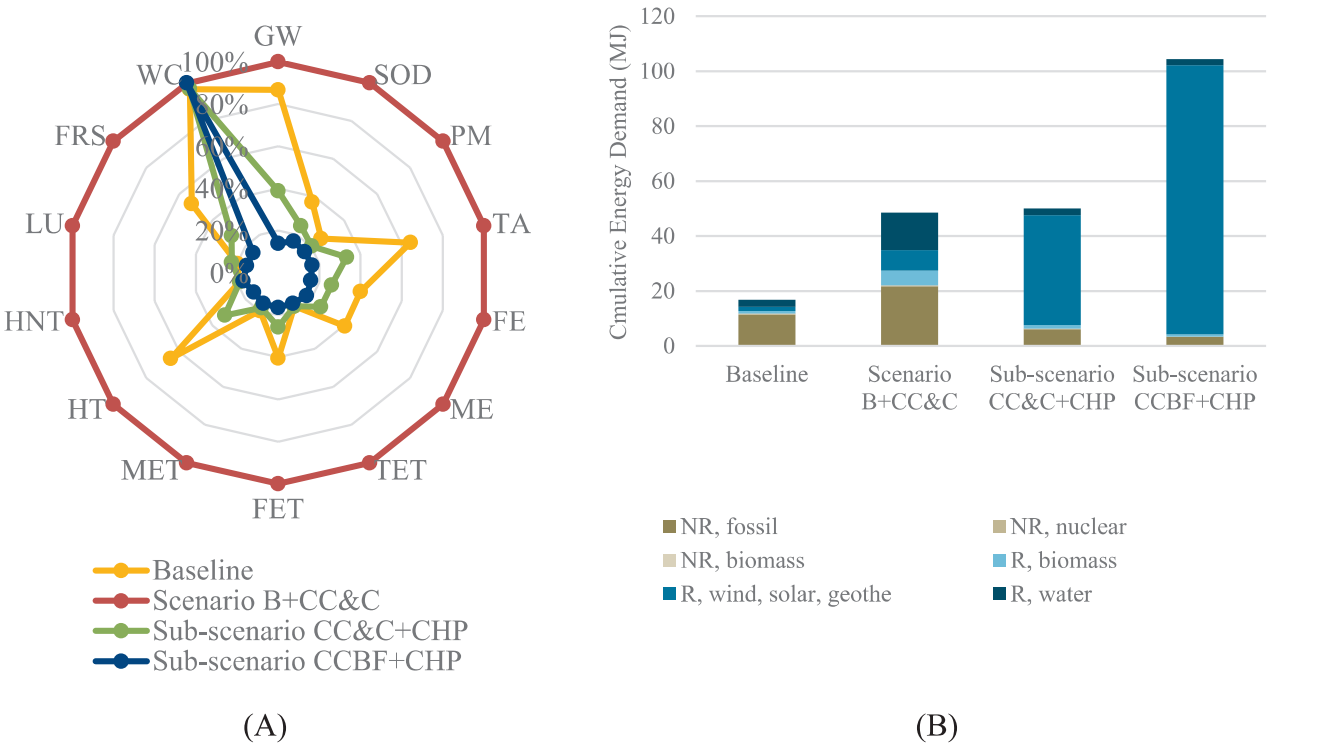

Fig. 10. Comparison of environmental profiles when the CC&C technology is applied for bioethanol production in the industrial context (A) and Cumulative Energy Demand of these scenarios based on 1 MJ bioethanol (B). Acronyms: Baseline: bioethanol without the carbon capture and conversion technology; B + CC&C: bioethanol with carbon capture technology (from fermentation section); CC&C + CHP: carbon capture and conversion with CHP system for electricity and steam production; CCBF + CHP: carbon capture from fermentation and boiler unit with a CHP system for electricity and steam production. GW: Global Warming; SOD: Stratospheric Ozone Depletion; PM: Particulate Matter; TA: Terrestrial Acidification; FE: Freshwater Eutrophication, ME: Marine Eutrophication; TET: Terrestrial Ecotoxicity; FET: Freshwater Ecotoxicity; HT: Human Carcinogenic Toxicity; LU: Land Use; FRS: Fossil Resource Scarcity; WC: Water Consumption; NR: Non-renewable; R: Renewable.

By using an energy-based functional unit (i.e., MJ), García et al. (2011) estimated values of 36.8 and 38.4 g of CO_2-eq per MJ using sugarcane juice produced during sugar making process. The difference of these values is motivated by the consideration or not of surplus electricity. Espada et al. [67] reported about 60 g of CO_2-eq per MJ of ethanol by using cardoon biomass (leaves and stalks) with steam explosion and steam explosion plus alkaline extraction. Recently, Priadi et al. [68] estimated a value of 61 g CO_2-eq per MJ of ethanol, using palm tree fronds and catalytic hydrothermal hydrolysis pre-treatment, although an enzymatic hydrolysis reached the half of the impacts. Additionally, higher profiles such as 168 and 98 g CO_2/MJ ethanol were obtained with supercritical water treatment and CO_2-water hydrolysis with, respectively. In the latter case, the carbon emitted in the heating and fermentation processes was reused as supercritical CO_2, reducing the impacts associated with this hydrolysis pathway, demonstrating the benefits of reusing carbon emissions at an early stage of biorefinery designs. Using 1 MJ as functional unit, the GW profile for bioethanol production from apple pomace using CC technology in the CC&C + CHP and CCBF + CHP scenarios is approximately 76.1 and 27.5 g CO_2-eq, respectively, which is within the range of values reported in the aforementioned studies.

4. CC&C technology application perspectives

The De-Cent prototype could find application in sustainable buildings construction, which are designed with environmental awareness and resource efficiency in mind, aiming to minimize the ecological footprint while maximizing energy efficiency of their components, water resources conservation, and material sustainability. These structures already incorporate features such as built-in renewable energy sources (solar panels, wind turbines), high-performance insulation, and water-saving technologies to reduce energy consumption and waste. The De-Cent prototype could help meet the goal of creating spaces that promote the health and well-being of their occupants, while reducing the overall environmental impact of the built environment [9]. Furthermore, the modularity of the prototype makes it ideal for a decentralized application, containing losses in performance connected to scaling up the BES technology [29].

Carbon capture and storage and utilization technologies are increasingly being integrated into industrial facilities to capture CO_2 emissions produced during the manufacturing process, preventing them from being released into the atmosphere. Of particular interest, fermentation processes gas streams are highly rich in CO_2. Additionally, biorefineries are adopting renewable energy sources such as solar, wind, or biogas to power their operations, further reducing their reliance on fossil fuels. By using waste materials like apple pomace and incorporating energy-efficient, low-carbon technologies, biorefineries will help lower the overall carbon footprint of bioethanol production, contributing to more sustainable and circular bioeconomy practices (Abdul Kareem [69]. In this context, CC&C technology as the De-Cent prototype, could potentially be beneficial to any fermentation process, and it could be inserted into existing production plants to enhance resource recovery, while minimizing their carbon emissions and producing fuel to be used directly on-site.

5. Conclusion

This manuscript presented the potential environmental impacts of a novel carbon capture and conversion technology to produce methane in indoor and bio-industrial environments. Findings demonstrated that the technology could provide environmental benefits in both contexts simulated, based on renewable electricity generation sources, mainly.

First, the methane produced by the technology in indoor environments, although it adds the benefit of a healthier and improved air quality, reached higher environmental burdens in 8 out of 14 impact categories analysed than other methane production pathways (e.g., biomethane from synthetic gas, biomethane from biogas purification), with exceptions in categories such as eco-toxicity-related, human toxicity-related, and land use. This disadvantage is motivated by the high electricity demand of the technology (specifically in the BES system), and the relevant role of fossil-sources in electricity generation grid. However, significant emission reductions could be achieved if the technology is coupled with a photovoltaic system. Additionally, focusing on the CO_2 balance, and considering the CO_2 uptake, the De-cent prototype could be a candidate for negative carbon emissions (−0.8 and −1.3 kg CO_2-eq per m^3 of methane), when renewable energy sources play a relevant role in the electricity supply and the cradle-to-gate scope is taken into account.

In the case of the industrial context (i.e., bioethanol production), introducing the carbon capture and conversion technology in the biorefinery design represented higher impacts compared to the baseline scenario (bioethanol production without the technology), due to the electricity demand of the bio-electrochemical unit, mainly. However, switching to an electricity grid with a significant contribution from renewable sources can reduce the environmental impact of the carbon capture and conversion technology, decreasing, for example, the global warming potential from 3.95 to 0.64 kg of CO_2-eq per kg of bioethanol, which represents an attractive environmental performance for achieving the goal of low carbon emissions.

Overall, while carbon capture and conversion technology holds great potential for addressing energy security and mitigating climate change issues supported by renewable energy sources, political support will be needed in the early stage of development to drive its deployment towards an urgent need for climate change mitigation actions and prompt its implementation in sustainable buildings or industrial environments.

CRediT authorship contribution statement

Ricardo Rebolledo-Leiva: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Silvia Bolognesi:** Writing – review & editing, Visualization, Validation, Investigation, Conceptualization. **Lluis Bañeras:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Methodology, Investigation, Conceptualization. **Sebastià Puig:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Investigation, Funding acquisition, Conceptualization. **Sara González-García:** Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Conceptualization.

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.enconman.2026.121087.

Data availability

Data will be made available on request.

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