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BECCS based on bioethanol from wood residues: Potential towards a carbon-negative transport and side-effects

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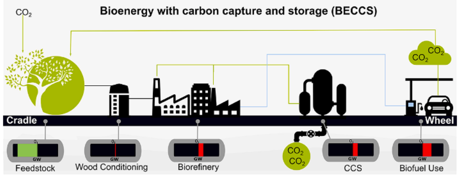
HIGHLIGHTS

- BECCS systems have the potential to deliver carbon-negative wood-based biofuels.
- A carbon footprint of −2.7 kg CO_2 eq./100 km was the best result obtained in an E85.
- Net removal depends on the carbon intensity of electricity and heating consumed.
- Risk of burden-shifting is a reality and should be considered for biofuel policies.

GRAPHICAL ABSTRACT



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ABSTRACT

Bioenergy with carbon capture and storage (BECCS) is gaining broad interest as an effective strategy to go beyond carbon neutrality. So far, most of the work on BECCS focused on power systems, while its application to the transport sector has received much less attention. To contribute to filling this gap, this work investigates the potential of BECCS as a carbon-negative strategy in the transport sector by applying process modelling and life cycle assessment (LCA) to bioethanol production from lignocellulosic waste. The process was analyzed following a cradle-to-wheel approach, i.e., from biomass growth to the combustion of biofuel in the cars, assuming that the CO_2 emitted in the fermentation and cogeneration units is captured, compressed and transported to be stored permanently in geological sites. Several scenarios differing in the bioethanol-gasoline blends (10–85% bioethanol) were considered for a functional unit of 1 km of distance travelled, comparing with fossil-based gasoline. Our results show that blends above 85% (ethanol/gasoline) could have the potential to deliver a net-negative emissions balance of −2.74 kg CO_2 eq per 100 km travelled and up to −5.05 kg CO_2 eq per 100 km using a low carbon electricity source. The final amount of net CO_2 removal is highly dependent on the carbon intensity of the electricity and the heating utilities. Biofuels blends could, however, lead to burden-shifting in eutrophication, ozone depletion and formation, toxicity, land use, and water consumption. This work highlights the potential of BECCS in the transport sector, and the need to analyze impacts beyond climate change in future studies to avoid shifting burdens to other categories.

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1. Introduction

The European Union member states have set targets to achieve a 40% reduction in greenhouse gas (GHG) emissions by 2030 (and proposed an even more ambitious goal of at least 55%) aiming at reaching climate neutrality by 2050. In this context, the European Green Deal is based on a series of strategic goals mainly sustained on three pillars, i.e., encouraging energy efficiency, promoting cleaner energy through the deployment of renewable sources and incorporating clean mobility systems (e.g., use of second or third-generation biofuels) [1].

Very likely, these actions will have to be accompanied by carbon dioxide removal (CDR) strategies, which seem vital to meet the goals stated in the Paris Agreement [2]. The draft of the upcoming EU Climate Law explicitly mentions the necessity of CDR to achieve the EU 2050 climate-neutrality goal [3], which could be delivered through carbon capture and storage (CCS). The portfolio of CDR options available includes afforestation and reforestation (AR), ocean alkalinity enhancement, biochar sequestration, mineralization of carbon dioxide, direct air capture and storage (DACCS) and bioenergy with carbon capture and storage (BECCS). Notably, CCS could be applied to a wide range of fossil-based industries. High emission sources include the cement industry, the iron and steel industry, and fossil refineries [4]. CCS in the fossil-based industry is deemed necessary to reach the decarbonization goals, yet it cannot lead to a net negative carbon balance. On the other hand, BECCS and DACCS, regarded as promising CDR options, have the potential of achieving net negative emissions.

According to estimates, the CO_2 removal capacity for CDR options in 2050 will range from 0.5 to 3.6 $GtCO_2 \cdot yr^{-1}$ for afforestation and reforestation, 2.0 to 4.0 $GtCO_2 \cdot yr^{-1}$ for enhanced weathering, 0.5 to 2.0 $GtCO_2 \cdot yr^{-1}$ for biochar, and reach 5.0 $GtCO_2 \cdot yr^{-1}$ for soil carbon sequestration. DACCS is assumed to be only limited by the geological storage capacity and the availability of energy resources. At the same time, the potential of BECCS varies significantly, between 0.5 and 5.0 $GtCO_2 \cdot yr^{-1}$, depending on the technical assumptions and land availability (i.e., degraded and marginal land and/or abandoned and unused agricultural land) [5–10].

Among all these CDR options, BECCS is receiving significant attention and already emerges as predominant in most of the climate change mitigation scenarios aligned with the 1.5 °C target. BECCS allows removing CO_2 while providing the clean and reliable energy needed to underpin economic growth and development, which makes it particularly appealing [5,11]. Indeed, BECCS is already considered in some Integrated Assessment Models (IAMs), in which other CDR engineered options are rarely contemplated mainly due to lack of maturity [8,12,13]. Hence, alongside with AR, the broad deployment of BECCS technologies will very likely play a pivotal role in meeting the climate goals as they represent a good compromise between the carbon removal potential and the associated removal costs [8,14].

Primarily, the BECCS concept refers to technologies converting biomass resources into valuable products in tandem with CO_2 capture systems. The latter prevents the release of the CO_2 absorbed via photosynthesis during biomass growth to the atmosphere. Then, the captured CO_2 is transported and injected into underground geological sites ensuring its long-term storage [15]. Compared with other CDR options, BECCS has the value-added of potentially providing a net negative balance of CO_2 with the atmosphere while delivering renewable energy-based products. The latter can, in turn, displace the use of their fossil-based counterparts, thereby avoiding their associated impacts [8,16].

The BECCS concept emerged in the last decade of the 20th century through conceptual studies addressing the production of biomass-based biofuels combined with CCS (as applied to the hydrogen fuel [17]) and other bio-energy applications that could potentially deliver negative emissions [18,19]. The beginning of the 21st century brought formally the concept of BECCS (initially called biomass-energy with carbon removal and disposal) as a risk management strategy to maintain GHG emissions at a safe level even under conditions hard to predict [20]. In this context, Möllersten et al. addressed the potential CO_2 reductions and associated costs in the chemical pulp and paper mill industry [21] and in sucrose fermentation to produce ethanol [22]. In 2003, the term BECCS was first introduced as a technological solution to convert the energy system into a CO_2 remover [23]. However, the kick-off for BECCS was the special report on CCS published in 2005 by the Intergovernmental Panel on Climate Change (IPCC), which highlighted BECCS as a feasible large-scale option to provide net negative emissions [24]. Since then, due to the continued use of fossil fuels and the steady increase in the associated carbon emissions, BECCS has attracted increasing attention as a key option to meet the climate targets sought [5,25].

Despite their expected pivotal role in climate change mitigation, the deployment of BECCS technologies would, however, face some obstacles. These challenges include constraints given by land availability and CO_2 storage capacity, socio-economic barriers, policy adequacy issues, logistical implementation difficulties, as well as other sustainability concerns [26,27], all of them linked to the specific BECCS technology selected. There are a handful of BECCS technologies implementing several conversion routes and spanning different sectors. These include (among others) biomass feedstocks burned at power or heating plants with CCS [28], gas or liquid biofuels production at biorefineries with CCS [29–31], and pulp and paper mills equipped with CCS [32].

Several studies have delved into the BECCS technologies analyzing its cost-effectiveness, potentials and side-effects [8,25,33–35]. Other authors studied the negative emission potential of biomass co-fired with coal in a power plant coupled with CCS from a life cycle assessment perspective [36]. On the other hand, others focused on the BECCS supply chain optimization to deliver carbon-negative electricity [37–39]. Despite extensive research and the growing interest in BECCS at the industrial level [40], most of the efforts on BECCS have focused on biomass conversion to heat and power. In contrast, the BECCS concept applied to biorefineries that produce biofuels [14,39] remains mostly unexplored [7].

Carbon-negative biofuels could, however, become an appealing alternative to replace conventional fossil-based fuels in the transport sector. By 2050, a 60% reduction in GHG emissions from transport is expected compared to 1990 in order to comply with the recommendations [41]. Accordingly, the use of alternative fuels in transport will need to grow by about 20% to meet the 2 °C scenario of decarbonization [42]. In this context, the use of carbon-negative biofuels could provide significant environmental benefits by reducing the dependence on fossil fuels and curbing the associated GHG emissions. Furthermore, they could also help to accomplish the more ambitious goal of achieving a carbon–neutral or even carbon-negative road transportation sector.

Previous works on carbon-negative biofuels focused only on quantifying the savings in global warming potential (GW) while disregarding the potential collateral damage on other environmental categories such as land use, acidification or toxicity. Some authors estimated the cradle-to-wheel GHG emissions of bioethanol [43–46], while only a few considered CO_2 capture coupled with the biofuel production pathway [29,47–50]. To the best of our knowledge, no single study carried out a full life cycle assessment (LCA) of a bioethanol production system with CCS adopting a "cradle-to-wheel" scope and embracing impacts on human health, ecosystems and resources. This research gap is particularly critical, given the trade-offs between climate change and other environmental impacts inherent to some carbon mitigation strategies [51–55]. These trade-offs are exemplified in the case of first-generation biofuels, where carbon emissions are reduced at the expense of exacerbating impacts on land use and water consumption while posing the issue of competition for land with food crops [12,56]. Overlooking these trade-offs could lead to unwanted collateral damages, thereby potentially hampering sustainable development.

To contribute to filling this research gap, in this study, we investigate the production of bioethanol from residual woodchips covering a range of environmental categories beyond climate change. Our analysis considers direct and indirect emissions throughout the whole supply chain, including biomass residues procurement, transportation, conversion, and the end-use of the biofuel in vehicles. Hence, acknowledging the potential role of BECCS as an effective strategy to go beyond carbon neutrality, we apply LCA to the production of wood-based bioethanol coupled with CCS as a potential negative emission biofuel for transport decarbonization. LCA is a well-established holistic methodology that allows conducting a negative emissions assessment by considering all the carbon emissions in the entire life cycle of the fuel while simultaneously evaluating other environmental categories. Hence, LCA allows us to determine whether technologies can deliver a net negative carbon balance and whether this may happen at the expense of worsening other categories. This holistic analysis is particularly relevant for BECCS technologies, as they have not yet been extensively deployed at large scale. Moreover, LCA also allows us to pinpoint environmental hotspots within complex value chains, thereby assisting in the prioritization of efforts to improve the environmental performance [57].

Eight scenarios differing in the bioethanol-gasoline blending ratios were considered and compared with the fossil-based counterpart, i.e., conventional gasoline. In short, our results show that achieving a net negative emissions balance requires a bioethanol-gasoline blend above 85% and that the sources of electricity and heat consumed by the primary production process play a vital role in the final carbon balance achieved. However, biofuels from lignocellulosic residues could worsen other environmental impacts, including eutrophication, ozone depletion and formation, toxicity, land use and water consumption. Our results could help in the development of future policies aimed at promoting negative emissions technologies and practices, where holistic assessments are critical to ensure sustainable development.

2. Methods

A holistic evaluation of the value chain for bioethanol production was performed through the implementation of the LCA approach, as described in the ISO 14040 and 14044 standards [58,59]. The goal and scope definition, life cycle inventory (LCI), life cycle impact assessment

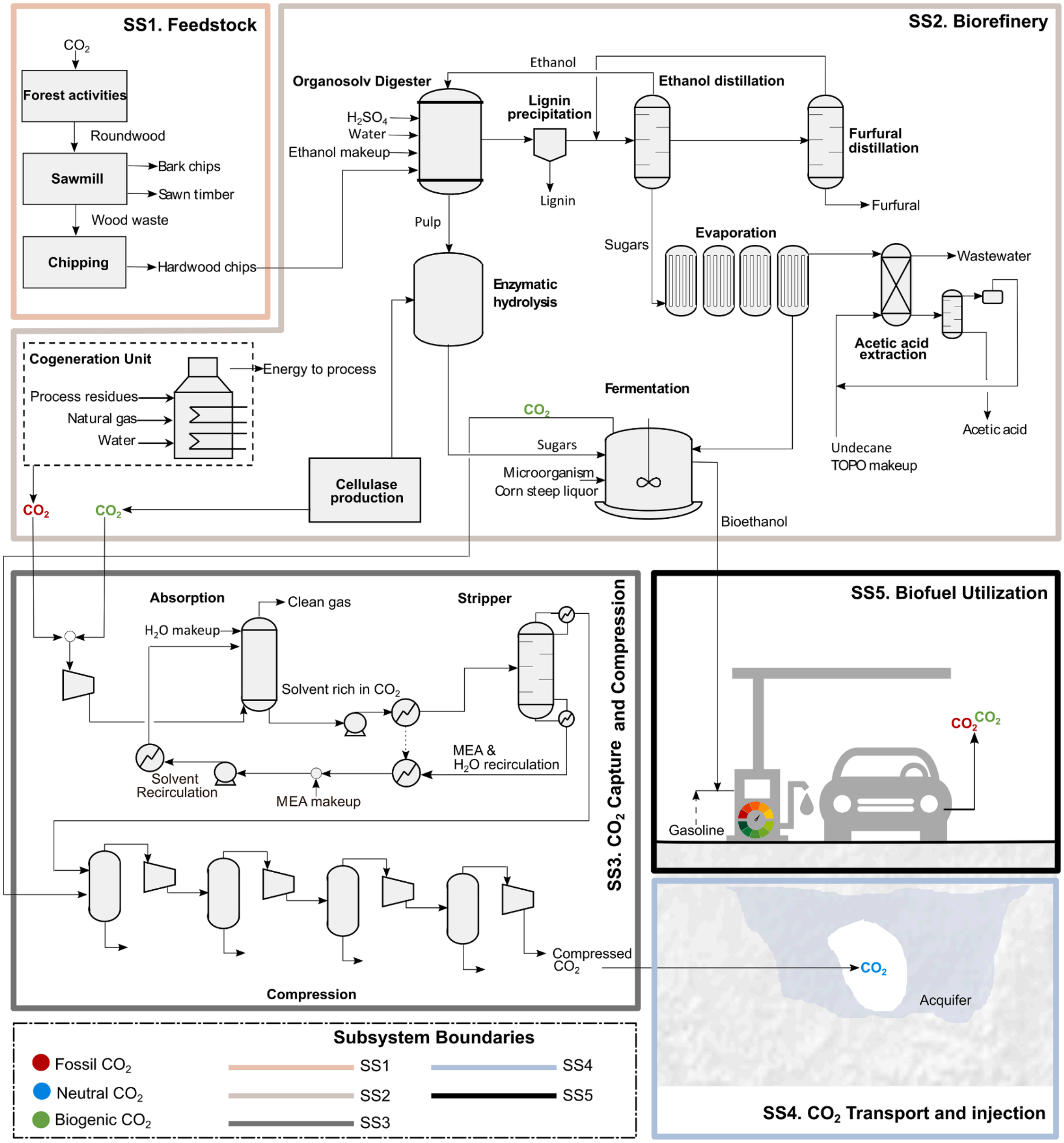

Fig. 1. Cradle-to-wheel system boundaries for the use of bioethanol produced in a biorefinery with CCS (functional unit: 1 km travelled with bioethanol and/or a bioethanol blend with a gasoline-fueled vehicle).

(LCIA), and the interpretation of the results stages were all completed, as discussed in detail in the ensuing sections.

2.1. Goal and scope definition

The goal of the study is to analyze the carbon footprint (CF), together with other environmental impacts, of the complete lignocellulosic bioethanol production and utilization value chain. To this end, our study follows a cradle-to-wheel scope that considers all the impacts from the growth and exploitation of lignocellulosic biomass to the end-use of the biofuel in a passenger vehicle. This scope, therefore, covers direct and indirect CO_2 emissions over the whole life cycle while avoiding double counting . The functional unit that best describes the main operational objectives of the system is 1 km travelled by the bio-fueled vehicle.

2.2. System boundaries

This section describes the system under study (Fig. 1) based on a cradle-to-wheel scope. Five main subsystems (SS) have been defined: SS1 Feedstock; SS2 Biorefinery; SS3 CO_2 Capture and compression; SS4 CO_2 transport and injection; and SS5 Biofuel utilization.

2.2.1. SS1. Feedstock

The biomass feedstock consists of hardwood residues, specifically beechwood chips from a sawmill. This subsystem includes the silviculture activities, comprising the uptake of CO_2 associated with forest growth, soil preparation, and wood extraction activities. The extracted round wood is then further processed in sawmill facilities to obtain the three main products: bark, sawn timber, and wood residues. The latter, corresponding to the waste fraction, is the target feedstock in this process and enters the biorefinery in the form of woodchips [60,61].

2.2.2. SS2. Biorefinery

The biorefinery includes all the process units required for the transformation of the woodchips, which are transported from a sawmill. We assume a transportation distance of 100 km by lorry. Woodchips are first digested in an Organosolv reactor, using ethanol and sulfuric acid as the catalyst at 180 °C. Pulp and liquor fractions are recovered in this first unit. The pulp stream is rich in hydrolyzable celluloses and hemicelluloses. These compounds are transformed into fermentable sugars in an enzymatic hydrolysis unit using cellulases. Lignin is precipitated from the liquor stream, which enters a distillation unit for the recovery of ethanol. A furfural stream is recovered via distillation as well. The sugars fraction is processed in an evaporator that removes water and acids. A liquid–liquid extraction unit separates then the acetic acid from a residual water flow. All lignocellulosic sugars are fed to the fermentation unit, where steep corn liquor and other micro-nutrients are added to produce bioethanol. Products other than ethanol are retrieved from the wood fractioning steps (furfural, lignin, acetic acid), yet our study focuses on bioethanol as the primary fermentation product. Process residues and natural gas are both combusted in the cogeneration unit in order to cover the energy requirements of the system [62].

2.2.3. SS3. CO_2 capture and compression

The CO_2 flue emissions from the biorefinery are captured, purified and compressed in this subsystem. Three main emission streams are the target of this subsystem (see Figure S1 and Table S1 in Supplementary Material). In the biorefinery (SS2), heating needs are supplied by combusting both process residues and fossil fuels. Therefore, the stream leaving the cogeneration unit contains a mix of biogenic and fossil CO_2, both of which are captured. On the other hand, the CO_2 emissions from the ethanol fermentation unit and the production of cellulases are entirely biogenic. The CO_2 streams from the cogeneration unit and the enzyme production process are fed to the capture system. In contrast, the biorefinery off-gas is fed just before the compression stage (due to its higher degree of purity), which reduces the energy and chemicals requirements of the system. Notably, the CO_2 flue gas and the off-gas from cellulase production are directed through a blower towards an absorption–desorption system with an aqueous monoethanolamine (MEA) solution [63]. MEA absorption was selected as CO_2 capture method due to its suitability for post-combustion capture. MEA is highly reactive in contact with CO_2 and is particularly recommended to treat gas streams with low concentrations of CO_2 (such as the one leaving the cogeneration system) [64,65]. In the stripping section, the MEA is desorbed from the CO_2, resulting in a purified CO_2 stream that exits the top of the column at a purity of 13.9% wt., containing 86.1% wt. of residual water; this gaseous stream will later undergo a compression stage. The bottoms stream of the distillation column is recirculated to reuse the lean solvent back in the capture process. Before compression, the target CO_2 stream is directed through a flash unit, in which a fraction of the water is removed. The overall compression ratio of 110 requires four stages, with a constant inter-stage compression ratio of 3.2. Inter-stage cooling between compressors is applied to keep the temperature within the desired range [66]. The flash cooling allows delivering a purified CO_2 stream free of water, reaching the required quality specifications. The conditions of the stream leaving the compression stage should be fixed based on the pressure, temperature and purity conditions required for the transport and injection of CO_2.

2.2.4. SS4. CO_2 transport and injection

CO_2 exits the previous system at a pressure of 110 bar and 50 °C, that is, at a supercritical state that facilitates its transport, geological injection and long-term storage (e.g., in saline aquifers). Purity specifications are relevant to avoid pipeline corrosion, i.e., water limit of 400 ppm, and a concentration below 4% vol. of N_2 and H_2, the main compounds present in the treated streams. A concentration of CO_2 above 95.5% wt. is also recommended (in our case, 99.8% wt. in SS3) [67]. SS4 includes the pipeline for CO_2 transport, considering a distance of 200 km. Based on the physical conditions of the stream and the transport distance, we assume that no further recompression is needed. The LCA covers the drilling of the well and the CO_2 losses during pipeline transport, considering 0.026% of losses per 1,000 km [68].

2.2.5. SS5. Biofuel utilization

The bioethanol produced in the biorefinery is used in internal combustion engine vehicles fueled with bioethanol-gasoline blends. Direct emissions in a vehicle travelling a distance of 1 km (functional unit) were considered. Eight scenarios were studied differing in the biofuel-gasoline blend percentages. Scenarios were also defined according to the heating source employed in the capture and compression system (SS3), i.e., either natural gas or sugar cane bagasse (Table 1) to provide a set of results ranging from fossil- to bio-based resources. The latter resource is only available in specific geographic regions, yet including it in the analysis sheds further light on the extent to which biofuels can deliver negative emissions. Moreover, bio-based heating from sugar cane bagasse was selected following a conservative assumption, as it shows a poor GW performance among all the heating alternatives from biomass available in the Ecoinvent v3.5 [69] database (Figure S2). The fossil-based alternative is based on conventional gasoline since gasoline-fueled vehicles represent the largest share of today's fleet. Our analysis excludes the vehicle infrastructure (i.e., manufacture, assembly, and end-of-life) since all the scenarios consider the same internal combustion engine vehicles.

2.3. Assumptions and limitations

The following assumptions apply to the LCA study. The transport of bioethanol to fueling stations was omitted. In contrast, we considered the transportation of woodchips from the sawmill to the biorefinery, assuming a distance of 100 km with 5% losses in a lorry freight. Electricity and chemical processes are based on a European average, when available, or a global average otherwise. The role of the carbon intensity

Table 1
Scenarios considered based on the biofuel-gasoline blend percentages.

Scenario acronym	Heating source in the ethanol plant	Fuel blend		Vehicle
		Bioethanol(%)	Gasoline(%)	
Gasoline	–	0	100	Gasoline compression ignition, internal combustion engine vehicle (GCI ICEV)
E10 SC	Sugar cane	10	90	Spark ignition, internal combustion engine vehicle (SI ICEV)
E10 NG	Natural gas			
E25 SC	Sugar cane	25	75	Spark ignition, internal combustion engine vehicle, high octane fuel (SI ICEV HOF)
E25 NG	Natural gas			
E40 SC	Sugar cane	40	60	Spark ignition, internal combustion engine vehicle, high octane fuel (SI ICEV HOF)
E40 NG	Natural gas			
E85 SC	Sugar cane	85	15	Spark ignition, internal combustion engine vehicle (SI ICEV dedicated)
E85 NG	Natural gas			

(CI) of the electricity mix was analyzed by considering a wide range of mixes differing in their CFs (below and above the European average). Regarding the heat requirements of the CO_2 capture and compression system, we assumed that the cooling needs are covered using cooling water pumped in a closed circuit. Infrastructure was omitted (installation, construction, and decommissioning), as it can be considered negligible over a typical lifetime of industrial installations of over 30 years [70].

In SS1 -biomass feedstock acquisition- economic allocation was applied to split the total impact among the products and co-products. Notably, impacts from forest activities and sawmill were economically allocated among co-products, while the impacts from chipping were allocated entirely to woodchips [71]. All the impacts from the biorefinery subsystem were allocated to the bioethanol, which represents the most conservative approach.

In this analysis, the impacts from the production, assembly, and end-of-life stages of the vehicle itself were omitted. Note that all the scenarios consider the same conventional gasoline-fueled spark-ignition vehicle (ICEV), so they remain comparable. Direct combustion emissions from the use of bioethanol and gasoline were considered from the GREET 1.3 vehicle cycle model [72], together with the indirect impacts from the production of each fuel.

2.4. Life cycle inventory

The LCA analysis relies on a compendium of different data sources, namely bibliographic-published data, simulation data, as well as databases. For the biomass silviculture [60,61] and the biorefinery facility [62], bibliographic data was used. Data for transport and injection of CO_2 were retrieved from literature sources [68]. The GREET 1.3 database was used for estimating the direct emissions of vehicles, including CO_2, CH_4 and NO_x emissions, by subtracting the well-to-pump emissions from the well-to-wheel emissions, both available in the said database [72].

With regards to SS3, data are based on a process simulation of the CO_2 capture system following the work by Adams II et al. (2014) [63]. Further details regarding the process simulation for the capture and compression of CO_2 are presented in the Supplementary Material file. The inventory data for each subsystem are displayed in Tables S2-S6 in the Supplementary Material.

2.5. Life cycle impact assessment method

An attributional approach was followed to quantify a set of midpoint impact indicators. Characterization factors from the ReCiPe 1.1 Hierarchist method [73] were applied using the SimaPro 9.0 software. The Ecoinvent v3.5 database [69] was used for the modelling of the background processes. Our analysis covers the CF indicator derived from the GW category from ReCiPe [74], expressed in kg CO_2 eq, as well as a set of mid-level impact categories provided by the same impact assessment method. The latter include ozone depletion in kg CFC11 eq, ozone formation in kg NO_x eq, terrestrial acidification in kg SO_2 eq, freshwater eutrophication in kg P eq, marine eutrophication in kg N eq, freshwater ecotoxicity in kg 1,4-DCB eq, marine ecotoxicity in kg 1,4-DCB eq, human toxicity in kg 1,4-DCB eq, land use in m^2a crop eq, fossil resources scarcity in kg oil eq and water consumption in m^3.

2.5.1. Carbon accounting within LCA: Carbon footprint

Standard LCAs of systems involving biogenic inputs with a CO_2 uptake from the atmosphere, such as those involving forests, assume that this CO_2 uptake is released at the end of the product's life cycle. Accordingly, the biogenic CO_2 cycle is assumed to be mass balanced over the life cycle [75]. In contrast, fossil CO_2 emissions (both direct and indirect) contribute to GW because they entail a net release of fossil carbon to the biosphere (atmosphere), which contributes to climate change. Accordingly, most standard LCA methods, such as ReCiPe or CML, assign a zero characterization factor for GW to the biogenic CO_2 emissions [73,76].

In contrast, when assessing the CF in systems that capture CO_2 and store it permanently (CCS), it is critical to consider both the fossil and biogenic carbon flows adequately. A system either capturing fossil CO_2 or consuming biomass resources without CCS can lead, in the best case, to a zero-balance, i.e., carbon–neutral system (Fig. 2). On the other hand, routes consuming biogenic carbon coupled with CCS systems could potentially achieve a net negative balance, provided the CO_2 is stored underground in the long-term [77,78]. More precisely, a system can provide a net negative emissions balance if the biogenic CO_2 uptake exceeds the fossil and biogenic life cycle emissions (considering the capture system) embodied in the biofuel product (Fig. 2). Therefore, to quantify the carbon emissions of CCS systems precisely, the biogenic CO_2 captured via photosynthesis during biomass growth (embodied in the biomass resource) is assigned a negative value to give credit to the CO_2 removed from the atmosphere. The carbon footprint accounting is then performed by considering all of the upstream and downstream activities and their corresponding direct and indirect (both biogenic and fossil) GHG emissions occurring throughout the fuel's value chain. The latter include, as well, the end-of-life direct emissions from burning the biofuel in the engine. Furthermore, to assess the real potential to deliver negative emissions (physical net removal of CO_2 from the atmosphere), we consider a cradle-to-wheel approach (also known as cradle-to-grave or well-to-wheel) [77]. Hence, based on this tailored LCA accounting system, a fuel is deemed carbon-negative if it achieves a negative GHG emissions balance over its life cycle [14,79]. All data employed in this study are included in Table S7 in the Supplementary Material.

3. Results and discussion

The results section presented below focus, firstly, on discussing the CF results, to then extend the analysis to other environmental indicators, investigating the potential occurrence of burden-shifting.

3.1. Carbon footprint assessment: Negativity potential

The CF was analyzed following the methodology explained in

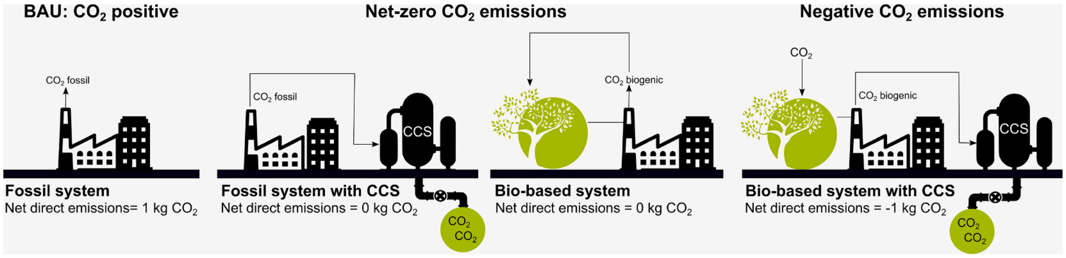

Fig. 2. Carbon accounting of direct CO_2 emissions in fossil and bio-based systems with and without CCS.

Section 2.5.1. Notably, we cover several fuel blends as well as national electricity mixes and renewable technologies (i.e., solar photovoltaic and wind energy), which differ in their CI (i.e., kg CO_2eq·kWh^{-1}). The base-case corresponds to the European average electricity mix. Recall that the electricity is consumed in the sawmill activities, the biorefinery section, and also in the CO_2 capture and compression stage (Fig. 1).

Fig. 3 shows the CF results as a function of the CI of the electricity consumed by the process. Each scenario is depicted by a line whose slope depends on the specific composition of the blend. Similarly, the intercept of the line is given by the concentration of bioethanol in the blend and the heat source in the process. Higher slopes correspond to blends with a higher concentration of bioethanol, in which the contribution of electricity towards the total emissions is higher. For a carbon-free electricity source, it holds that a higher bioethanol content results in a lower CF. Furthermore, the efficiency of the engine increases with the bioethanol content [80] (e.g., the energy consumed per distance travelled for the E40 is 2,677.9 J·m^{-1}, while for the E85 is 2,016.7 J·m^{-1}) [72]. Therefore, increasing the bioethanol content in the blend provides environmental benefits directly related to the lower fuel requirements. In all the bio-based heating scenarios (depicted in green in Fig. 3), it holds that increasing the bioethanol content decreases the CF for the whole range of carbon intensities considered. However, in the scenarios using natural gas as the heating source, some of the lines cross for high carbon intensities. Consequently, higher bioethanol contents can lead to larger CFs, e.g., E40 NG vs. E25 NG for a CI above 0.70 kg CO_2 eq·kWh^{-1}.

For the bio-based heating scenarios, all bioethanol blends, except for E10 SC for carbon-intensities above 0.85 kg CO_2 eq·kWh^{-1}, perform better than the business as usual (BAU) scenario (i.e., conventional gasoline depicted with a horizontal blue line). However, for the scenarios based on natural gas as the heating source for SS3, only the E40 NG and the E85 NG scenarios would outperform the conventional benchmark gasoline for low carbon electricity sources. Notably, the only blend delivering negative emissions is E85 SC, which does so for CIs below 0.91 kg CO_2eq·kWh^{-1}. For the average electricity mix in Europe, E85 SC would deliver −2.74 kg CO_2 eq/100 km, while in Switzerland or France, the CF would be further reduced to −4.62 kg CO_2 eq/100 km and −4.87 kg CO_2 eq/100 km, respectively. Furthermore, wind power could reduce the CF of E85 SC to −5.05 kg CO_2 eq/100 km. In contrast, European countries such as Poland, which plans to maintain coal power

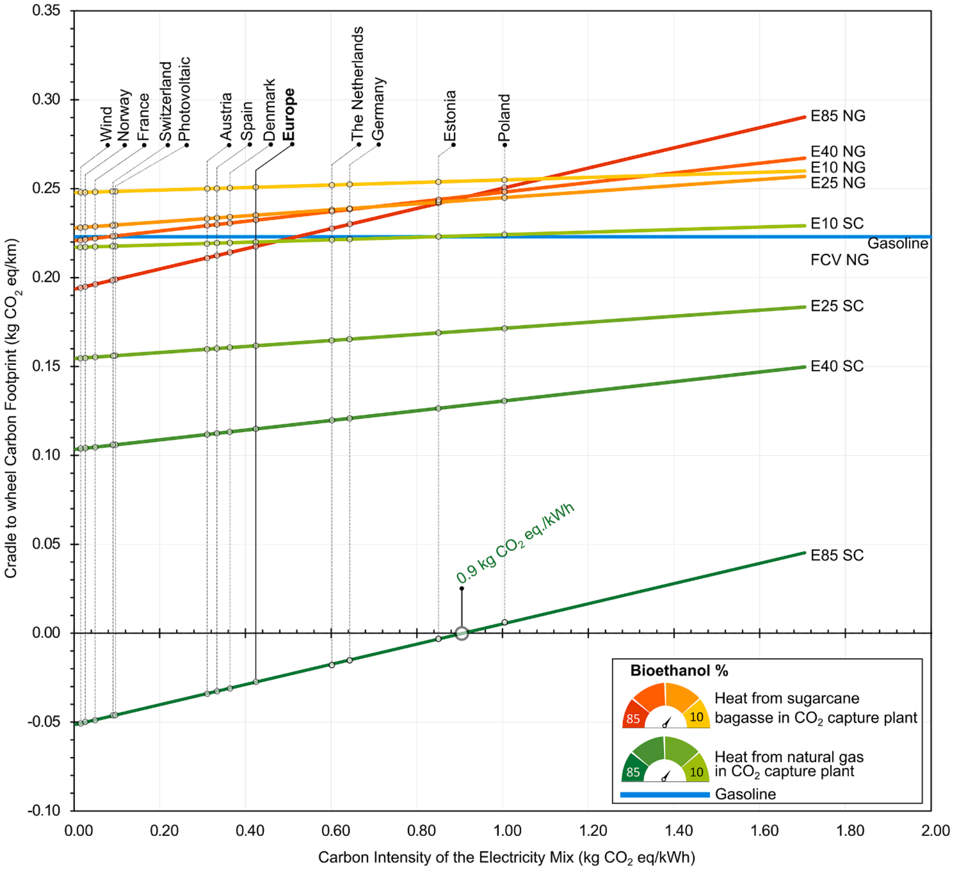

Fig. 3. Cradle-to-wheel carbon footprint (kg CO_2 eq km^{-1}) for eight scenarios as a function of the CI of the electricity mix (kg CO_2 eq kWh^{-1}). Green scenarios use sugarcane bagasse as the heat source in SS3. Red scenarios use natural gas as the heat source in SS3. The darker the shade of the color, the higher the bioethanol content in the blend (E10, E25, E40, E85). Vertical dotted lines denote the carbon intensities of the electricity mixes of some EU countries and renewable electricity technologies. For comparison purposes, gasoline is depicted with a horizontal blue line.

plants to enhance its energy security [81], would be unable to produce biofuels leading to net negative emissions.

Considering that a regular passenger car may typically travel an average of 14,000 km·yr^{-1} [82], the potential for decarbonization of a E85 SC vehicle would be −382.98 kg CO_2 eq·(car·yr)$^{-1}$ assuming an average European electricity mix. The overall savings, however, should also consider the avoided emissions by gasoline replacement (3,121 kg CO_2 eq·(car·yr)$^{-1}$ [72]). Considering, for instance, the average carbon emissions in Spain, i.e., 5,030 kg CO_2 per capita for 2017 [83], the implementation of the E85 SC fuel could reduce 52.98% current per capita emissions. Similarly, reductions of 37.51% in per capita emissions (relative to average values) could be achieved in Europe [83].

Meeting the environmental goals of the European Commission will critically depend on our ability to change the European vehicle fleet. According to the IPCC, the global transport sector could reduce its emissions 4.7 $GtCO_2$ eq·yr^{-1} by 2030 [5]. The implementation of carbon-negative bioethanol fueled vehicles could help to offset emissions from hard-to-abate aviation or shipping transportation [84]. Considering the total passenger-car fleet in 2015 in the European Union [82], replacing gasoline-fueled passenger vehicles by E85 SC vehicles

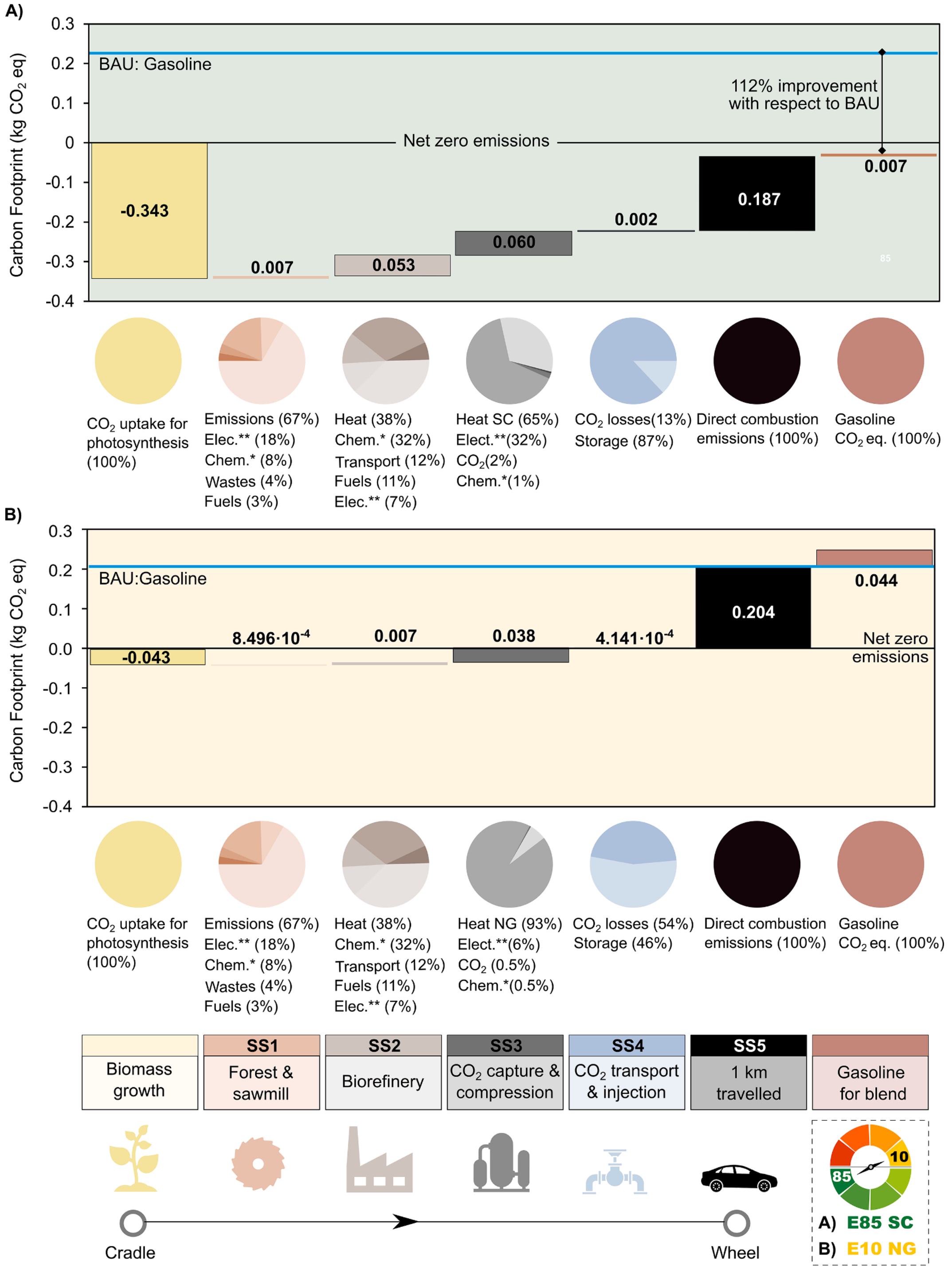

Fig. 4. Breakdown of contributions of each subsystem to the CF for the E85 SC and E10 NG scenarios expressed per 1 km travelled. Subplot A corresponds to the E85 SC, i.e., the best-case scenario, while subplot B corresponds to E10 NG, i.e., the worse-case scenario. Pie charts show the relative CF contributions per activity for each subsystem.

could reduce 0.88 $GtCO_2$ eq yr^{-1}, which represents 18.79% (16.73% from the removal of gasoline cars and 2.06% from the negative emissions in E85 vehicles) of the global transportation sector reduction target for 2030 in the 1.5 °C scenario (4.7 Gt CO_2 eq·yr^{-1}). Note, however, that the final CDR potential required to meet the climate targets remains uncertain as it ultimately depends on the delay of the mitigation actions. Moreover, other BECCS technologies, such as biomass conversion to power and heat, as well as other negative technologies and practices in the portfolio of CDR options, could help to reduce the reliance on BECCS [13]. Notably, the large scale deployment of BECCS will face many challenges, such as sustainability concerns (e.g., land-system change and loss of biodiversity) [51], governance problems, sociopolitical constraints and economic viability barriers [85].

The pathways to avoid overshooting the 1.5 °C target by 2050 require removing globally around 8 Gt CO_2·yr^{-1} by BECCS [5]. Removing this amount of carbon using E85 SC vehicles would require producing 3,400 GL per year of lignocellulosic bioethanol (considering the full displacement of gasoline). The annual world production of bioethanol in 2018 was 110 GL, while only<1% of the global bioethanol production in Europe was second-generation fuel [86]. Hence, the commercialization of lignocellulosic bioethanol with CCS should be dramatically increased for this fuel to play a significant role in combatting climate change. Note, however, that the CDR that would be required to reach the climate goals is expected to be provided by BECCS applied also to the power and heating sector.

To provide a full picture of the CF balance, we next analyze the breakdown of emissions by subsystem for the extreme cases, i.e., the E85 SC and E10 NG scenarios (waterfall plot in Fig. 4, subplot A and subplot B, respectively) in the base case (i.e., European average electricity mix). Due to space limitations, the results for the remaining scenarios are presented in the Supplementary Material (Figures S3-S8).

For the E85 SC scenario (Fig. 4, subplot A), the negative emissions from the CO_2 uptake during biomass growth account for 52% of the total absolute value. The direct emissions in the vehicle engine are the most significant positive contributor to the total CF impact (28%), followed by the capture and compression plant (SS3), and then the production process in the biorefinery (SS2), which account for 9.1% and 8.1% of the total emissions, respectively. In contrast, the contributions of the silviculture and sawmill-related activities (SS1) and the CO_2 pipeline transportation and injection (SS4) are both marginal (1.03 and 0.26% relative contributions, respectively). Overall, the negative emissions exceed the positive ones, thereby resulting in a carbon-negative biofuel providing −0.027 kg CO_2 eq·km^{-1}.

The sensitivity of the CF results to the CO_2 transport distance to the geological site has been studied in the range of 1–400 km, considering that after the first 200 km, recompression of the CO_2 is needed [68] (Figure S9). The CF of the scenarios varies very little with the CO_2 transportation distance, ranging from 0.11% to 8.40% of increase in CF for the E10 NG and the E85 SC scenarios, respectively. Note that the overall conclusions remain qualitatively the same, as the scenarios still lead to a negative balance (although the net carbon efficiency would be reduced). The transport distance from the BECCS plant to the geological site, together with the distance to the areas of larger lignocellulosic biomass availability, will determine the optimal geographical location of the plant. The low emissions of the CO_2 transport through pipelines and the low energy density of biomass make locations near the biomass source more appealing. However, the need of infrastructure for CO_2 transport could hinder a quick deployment of BECCS for biofuels, which might be essential to meet the decarbonization goals [39,87].

Regarding E10 NG (Fig. 4, subplot B), its positive emissions exceed the negative ones linked to the uptake of CO_2 during the biomass growth, thereby making the fuel carbon-positive on a life cycle basis (+0.25 kg CO_2 eq·km^{-1}). Notably, negative emissions from biomass growth represent 13% of the total emissions (−0.043 kg CO_2 eq·km^{-1}), and (in absolute value) lie slightly below the positive cradle-to-gate emissions embodied in the gasoline contained in the blend, 0.044 kg CO_2 eq·km^{-1} (i.e., 90% gasoline, 10% bioethanol). The emissions from the biomass pretreatment and biorefining activities are quite small (<2% of the total). In contrast, the CO_2 capture and compression stage accounts for 11% of the total emissions due to the large amount of energy required to regenerate the amine in the CCS system. Most of the positive emissions correspond to the biofuel combustion in the vehicle, around 60% of the total well-to-wheel emissions; meanwhile, the emissions of the silviculture and sawmill activities and the CO_2 transportation are, again, negligible (<0.5%).

The breakdown of the CO_2 emissions per activity of each subsystem (pie charts in Fig. 4) allows identifying environmental hotspots where potential improvement efforts are most needed. The heat consumed to regenerate the MEA is a major source of CO_2 emissions in SS3. Hence, the CF performance of biofuels could be improved by using low-carbon heating sources or taking advantage of waste heat from industrial activities. Identifying new solvents or developing new catalytic processes to reduce energy consumption in the CCS system (SS3) could also help to reduce this contribution [88,89]. At present, this is the primary hotspot for this subsystem in both the natural gas scenarios (93% share within the subsystem) and the bio-based heating scenarios (65% share within the subsystem). As for the biorefinery plant (SS2), the primary hotspot is given jointly by the consumption of chemicals and heat, with 32% and 38% shares of the total impact, respectively.

Furthermore, the feedstock (SS1) contributes with 1.03% in scenario E85 SC and 0.25% in scenario E10 NG. We note that the impact of beech wood (given by the fertilizers, water use, machinery and associated yield) may vary in forestry residues of other species (e.g., birch, eucalyptus, spruce) [90]. However, these changes might not be that significant unless second-generation biomass (i.e., wood or residues) is replaced by first-generation biomass (i.e., edible crops). Notably, the latter shows worse performance in all of the environmental categories (Figure S10) and also competes with food [91]. Furthermore, the process would need to be adjusted to accommodate other feedstocks, e.g., the biomass pretreatment method might entail a lower environmental impact when dealing with first-generation feedstocks [92]. Specifically, Organosolv or other pretreatment methods for delignification, such as steam explosion or liquid hot water, are generally more energy-intensive due to the recalcitrance of biomass [93].

Regardless of the fuel blend, the capture and compression plant subsystem causes a significant impact (Figs. 4 and 5). With gasoline percentages above 75% in the blend, however, the hotspot shifts from the capture plant to the direct emissions from the gasoline combustion (Figures S3-S8). The development of new sorbents could help to reduce the substantial energy requirements (and costs) of the CO_2 separation, thereby decreasing its impact [89]. Accordingly, Figure S11 in the Supplementary Material provides the results of a sensitivity analysis on the heating demand of the CCS plant for the different scenarios benchmarked against bibliographic heat demands for MEA absorption processes [94–99]. Our CCS system requires 7.5 MJ per kg CO_2 captured, an amount slightly above the values reported in the literature (5.5–3.5 MJ·kg^{-1} CO_2 captured). Note that, for lower heating needs, the E85 NG would be able to achieve carbon-negativity, even when relying on natural gas as the heating source (Figure S11). These results indicate that the CF of biofuels could be further improved by reducing the heating needs for the solvent regeneration and by exploiting waste-heat recovery options and other synergies with other industries [28]. Ultimately, the impact of the heating demand is dependent on its magnitude (MJ·kg^{-1} CO_2 captured) as well as the heating source. As presented in Figure S11 in the Supplementary Material, for bio-based heating, lowering the energy consumption would not affect that much the impact, especially for values below 35 GJ·kg^{-1} bioethanol. On the contrary, heating via natural gas offers more room for improvement.

We note that very pure CO_2 streams from fermentation could be handled via direct dehydration and compression of the gas stream, thereby reducing the energy needs substantially [9]. Flue gas with a lower CO_2 concentration would increase the energy and solvent

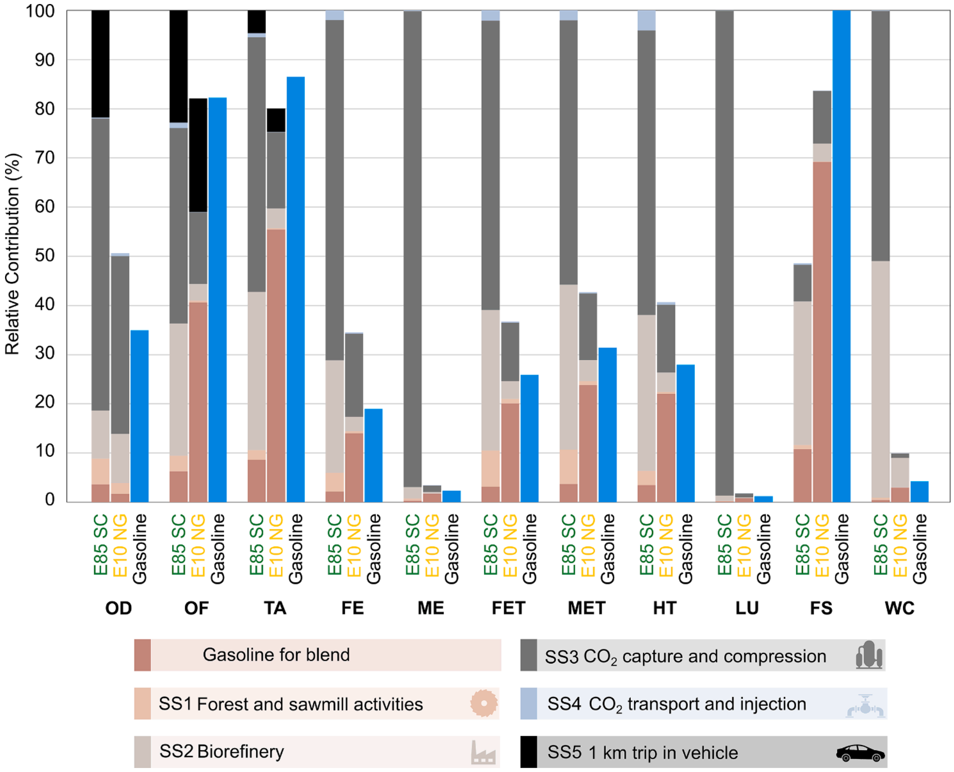

Fig. 5. Comparative evaluation of environmental profiles for the best-case scenario E85 SC, the worst-case scenario E10 NG and the BAU alternative, i.e., conventional gasoline. OD: ozone depletion, OF: ozone formation, TA: terrestrial acidification, FE: freshwater eutrophication, ME: marine eutrophication, FET: freshwater ecotoxicity, MET: marine ecotoxicity, HT: human toxicity, LU: land use, FS: fossil resource scarcity and WC: water consumption.

requirements in CCS, and, consequently, the impact of SS3. Thus, the BECCS potential for net CO_2 removal would be lower in less concentrated streams and higher in more concentrated ones. The CO_2 source, therefore, impacts the net removal efficiency and, thus, needs to be considered in the selection of the capture method [24]. Either way, there is a clear need to cut down the energy needs, mostly through better solvents and, whenever possible, through the use of waste heat (or heat from waste biomass).

3.2. Other environmental implications and burden-shifting

We now turn our attention to the potential occurrence of burden-shifting, that is, the collateral damage to some environmental areas of protection taking place when attempting to mitigate carbon emissions. Accordingly, Fig. 5 (and Tables S8 and S9 in the Supplementary Material), shows the relative performance (compared to gasoline) of the two extreme scenarios (E85 SC and E10 NG) in the midpoint impacts of the ReCiPe 1.1

Indeed, burden-shifting takes place in the E85 SC fuel, which displays a negative CF (Fig. 3) and emerges as the best option in fossil resource scarcity but shows the worst performance in all the other impact categories [100]. . Similarly, E10 NG performs worse than gasoline in all the categories, except for fossil resource scarcity and terrestrial acidification. The latter impacts are strongly linked to fossil fuel combustion and the atmospheric deposition of acidifying compounds. Note that, due to the use of chemicals in SS2 (e.g., sulfuric acid), increasing the bioethanol content worsens the TA and OF categories.

Our results show that burden-shifting is particularly critical in marine eutrophication, land use and water consumption, i.e., E85 SC biofuel with 42.45, 82.91 and 23.59 times higher impact relative to gasoline, respectively (and 1.46, 1.48 and 2.34 times in each category, for the E10 NG benchmarked against gasoline). Furthermore, the E10 NG outperforms the E85 SC biofuel in all the categories except for CF and fossil resource scarcity, where it is inferior due to its higher content of fossil-based resources (gasoline in the blend and natural gas for heating). Therefore, it becomes clear that the potential collateral damage of biofuels should not be overlooked.

Delving into the drivers of burden-shifting, the breakdown of impacts in Fig. 5 allows pinpointing the main hotspots in each impact category. The relative burdens and environmental profile change substantially attending to the scenario analyzed (Fig. 5), which can be further observed in Figures S3 through S8 in the Supplementary Material for the scenarios omitted here. Overall, for blends rich in bioethanol, the biorefinery (SS2) and the CO_2 capture and compression (SS3), are the main hotspots of the system in most of the impact categories.

The ozone depletion category for the bioethanol blends worsens with respect to gasoline, mainly due to the high heating needs in the capture process (SS3) and the marginal increase in the unburned hydrocarbons and nitrogen oxide in the engines [101]. Similarly, in the ozone formation and terrestrial acidification categories, the E85 SC performs worse than the E10 NG and gasoline alternatives due to the large impacts of the biorefinery and the capture activities. Note that the impact of the fuel utilization subsystem (SS5) is negligible in most non-climate change related impact categories, with the exception of ozone-related indicators (ozone depletion and ozone formation) where it represents around 18% of the total impact in both categories. As seen in Fig. 5, the E10 NG fuel performs slightly better than gasoline due to the reduction in the emissions of organic compounds (contributing to the ozone formation burdens), nitrogen oxides and ammonia (main drivers of the acidification category). These emissions are strongly linked to the refining and combustion of fossil fuels.

Freshwater eutrophication and marine eutrophication worsen substantially in the E85 SC and, to a lesser extent, in the E10 NG. The main drivers of these impacts are the use of nitrogen fertilizers (soil N_2O, ammonia and NO_x emissions) and phosphorous fertilizers (phosphoric acid emissions). Both compounds are linked to the production of dedicated bioenergy crops (i.e., the sugar cane bagasse employed for heating in SS3). In the marine eutrophication category, 97% of the impacts of E85 SC are due to the capture and compression plant. This high impact might be linked to the nutrient accumulation in water bodies due to the loss of nitrogen and phosphorus fertilizers (associated with the heat source) and the use of monoethanolamine. The latter is an amine-compound that can act as a driver of nutrient oversupply in marine environments.

Ecotoxicity in the freshwater and marine compartments also worsens in both biofuels (Fig. 5). Particularly, E85 SC increases the ecotoxicity impacts by 74% and 69%, respectively (and 30% and 26%, for the E10 NG). The trend in human toxicity is quite similar, where the best option is, again, gasoline followed by the E10 NG and, finally, the E85 SC fuel. For scenarios rich in bioethanol, the main contributors are the CO_2 capture and compression (SS3) and biorefinery sub-systems (SS2), while for the others, the primary hotspot is the gasoline. This might be due to the pesticides and fertilizers consumed during the biomass growth (e.g., sugarcane cultivation), which evaporate and runoff into freshwater and marine water bodies, and also to some compounds involved in the pretreatment and fermentation of the wood residues [102].

Regarding the land use and water consumption, the E85 SC fuel is by far the worst option, with the E10 NG alternative lying close to gasoline. The negative impact in these categories is mainly due to the contribution of the sugar cane burnt to provide heat in the CO_2 capture system (SS3). The land use impact is mostly linked to the transformation and occupation of land to grow the sugarcane feedstock. Furthermore, the increase in water consumption is due to the irrigation needs and the water required for pulp washing and lignin precipitation (SS2).

4. Conclusions

In this work, we investigated the concept of BECCS applied to a biorefinery coupled with CCS that converts wood waste material into bioethanol. We showed that blends with higher contents of bioethanol have the potential to deliver negative emissions. Moreover, in most of the scenarios, biofuels with CCS reduce the CF of conventional gasoline, more so when using low-carbon electricity and/or biomass as the heating source in the process. Particularly, with an E85 blend, a net balance of -2.74 kg CO_2 eq per 100 km travelled could be attained considering the European average electricity mix and heating for the capture and compression system supplied by biomass resources.

Furthermore, electricity mixes with higher shares of renewable energy (e.g., Switzerland, France or Norway) would double the final net amount of negative emissions provided (e.g., -5.01 kg CO_2 eq/100 km in Norway). Hence, the geographical location of the BECCS facilities becomes a key aspect in the production of net negative biofuels. Ideally, the biorefinery with CCS should be placed near the low-carbon energy resources available (electricity and heat), the biomass resources and the CO_2 geological storage sites. In practice, finding a suitable site might be challenging because these resources tend to be geographically dispersed. Locations near the biomass source might be preferred, which will require pipeline infrastructure yet to be developed.

Policies aiming at the decarbonization of the electricity mix will help to curb the CO_2 emissions in the transport sector. Further improvements in bioethanol production with CCS should focus on minimizing the heating demand of the CCS technologies, opting for heating systems relying on biobased residues, and exploiting opportunities for waste heat from other industries. In this context, process integration concepts and tools could help to use energy more efficiently. Our results show that substantial environmental benefits may be attained in climate change and fossil depletion while simultaneously enhancing energy security, a primary focus of most environmental policies. However, biofuels can lead to burden-shifting, i.e., CF improves at the expense of worsening other categories, which highlights the need to enlarge the scope of current environmental assessments beyond climate change. Notably, policies such as mandates on biofuels consumption solely focused on climate change mitigation may exacerbate impacts on eutrophication, ozone depletion and formation, toxicity, land use, and water consumption. Minimizing energy consumption in the CO_2 capture and compression stages, e.g., via heat integration and the use of biobased residues for heating, could reduce the collateral damage to other environmental areas. Nevertheless, trade-offs will arise in the deployment of biofuels, which should not be overlooked to avoid potential undesirable side-effects.

Overall, the BECCS concept applied to biorefineries offers excellent opportunities to reduce the carbon footprint of the passenger-vehicle fleet in the transition towards a carbon–neutral (or even carbon-negative) mobility system. In this context, the occurrence and severity of burden-shifting should be analyzed in-depth.

CRediT authorship contribution statement

Sara Bello: Formal analysis, Investigation, Methodology, Writing - original draft, Writing - review & editing, Visualization. **Ángel Galán-Martín:** Conceptualization, Methodology, Writing - review & editing, Visualization. **Gumersindo Feijoo:** Methodology, Writing - review & editing. **Maria Teresa Moreira:** Conceptualization, Methodology, Writing - review & editing. **Gonzalo Guillén-Gosálbez:** Conceptualization, Methodology, Writing - review & editing, Supervision.

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.apenergy.2020.115884.

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