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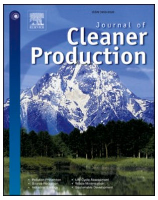

Economic and environmental implications of carbon capture in an olive pruning tree biomass biorefinery

Stylianos Fanourakis [a], Juan Miguel Romero-García [b], Eulogio Castro [b], Laureano Jiménez-Esteller [a], Ángel Galán-Martín [b,*]

[a] Department d'Enginyeria Quimica, Universitat Rovira i Virgili, Av. Països Catalans 26, 43007, Tarragona, Spain
[b] Department of Chemical, Environmental and Materials Engineering, Center for Advanced Studies in Earth Sciences, Energy and Environment (CEACTEMA), University of Jaén, Campus Las Lagunillas s/n, 23071, Jaén, Spain

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ABSTRACT

This study explores the integration of bioenergy with carbon capture and storage (BECCS) in a biorefinery system that converts olive tree prunings into bioethanol and antioxidants. With a capacity to process 1,500 tons of prunings daily, the biorefinery yields an annual production of around 12,000 tons of antioxidants (purity >60 %) and 78,000 tons of bioethanol. Utilizing a holistic approach involving process simulations and life cycle assessment, our analysis covers technical, economic, and environmental dimensions across two scenarios differing in design and heating source: natural gas or a BECCS system using olive prunings. Our findings reveal the potential for BECCS to drastically reduce the carbon footprint, potentially achieving net-negative emissions (−84.37 kg CO_2eq per 1.00 kg of bioethanol and 0.15 kg antioxidants produced). However, these environmental gains are counterbalanced by economic and environmental challenges, with investment and operating costs nearly doubling and leading to complex environmental trade-offs related to eutrophication (+75 %), increased water consumption (+45 %), and expanded land use (+80 %). Nevertheless, the premium nature of carbon-negative products, coupled with growing awareness and supportive policy frameworks, may overcome these economic barriers. This study highlights the importance of holistic evaluation when integrating CCS into biorefineries facilitating informed decision-making to address unintended adverse effects and promoting sustainability.

1. Introduction

The European (EU) Green Deal, with its commitment to achieving climate neutrality by 2050, is built upon three fundamental pillars: fostering energy efficiency, promoting renewable energy, and incorporating clean mobility systems (Calvin et al., 2023). A pivotal player in achieving the long-term climate goals will be the deployment of carbon dioxide removal (CDR) strategies. CDR encompasses practices and technologies aimed at removing carbon dioxide (CO_2) from the atmosphere, thereby compensating for emissions from industries and sectors that are difficult to decarbonize. CDR should be understood as an indispensable complement to the primary focus on rapid decarbonization because the extent of the CDR required hinges on delayed reductions in emissions (Calvin et al., 2023; Galán-Martín et al., 2021b). Estimates range from hundreds of billions to several trillion metric tons of CO_2 removal depending on factors such as residual emissions, emission reduction rates, natural carbon sinks, and technological advances (Galán-Martín et al., 2021b). Given the scale of this unprecedented challenge, governments and businesses must start promoting and scaling up CDR strategies sustainably (Calvin et al., 2023).

The CDR portfolio encompasses various technological solutions and practices, including afforestation, reforestation, soil carbon sequestration, biochar production, direct air capture and sequestration (DACCS), enhanced weathering, ocean fertilization, and bioenergy with carbon capture and storage (BECCS), among others (Matuštík et al., 2020; Minx et al., 2018). BECCS stands out for its ability to produce energy and marketable bio-based products while removing CO_2, provided the removed CO_2 exceeds emissions across the value chains (Bello et al., 2020; Negri et al., 2021; Tanzer and Ramírez, 2019). Notably, these bio-based products generated such as bioethanol, hydrogen, or electricity can replace fossil-based equivalents, providing further environmental benefits. BECCS involves absorbing CO_2 through biomass growth which is subsequently released during the biomass conversion processes

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* Corresponding author.
E-mail address: galan@ujaen.es (Á. Galán-Martín).

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Abbreviations

BECCS	Bioenergy with Carbon and Capture System
DACCS	Direct Air Carbon and Capture System
OPTB	Olive Pruning Tree Biomass
LCA	Life Cycle Assessment
ETEA	Environmental and Techno-economic Analysis
CDR	Carbon Dioxide Removal
GHG	Green House Gas
CO_2eq	Carbon Dioxide Equivalent
BIOR CCS	Biorefinery with Carbon and Capture System
BIOR NG	Biorefinery with Natural Gas
CAPEX	Capital Expenditure
OPEX	Operational Expenditure
AF	Annualized Factor
TAC	Total Annual Cost
LCOB, NG	Levelized Cost of BECCS or Natural Gas
NPV	Net Present Value
ROI	Return on Investment
PBT	Payback time
IRR	Internal rate of return

and captured and securely stored in underground geological sites, ensuring its long-term sequestration out of the atmosphere. Despite interest and its expected future role, there is only one large-scale BECCS industrial facility in operation today, situated in Illinois and producing bioethanol, with the capability to capture one million tons of CO_2 per year (Global CCS Institute Ltd, 2019). Meanwhile, there are several initiatives currently operating on a small scale and others in various stages of deployment (Global CCS Institute Ltd, 2019). Nevertheless, numerous challenges still need to be addressed to unlock the potential of BECCS, including the high costs, environmental considerations, and governance issues.

The BECCS concept lies at the heart of biorefineries, industrial facilities designed to produce multiple products from different biomass feedstock. These facilities can integrate carbon capture and storage (CCS) technologies to capture the biogenic CO_2 emissions generated during the conversion processes (e.g., fermentation, gasification, or direct combustion) that would be otherwise released back into the atmosphere. Then, the captured CO_2 is stored ensuring its long-term sequestration, while delivering bio-based energy, biofuels, and other chemicals with multiple applications (Galán-Martín et al., 2022). There are also opportunities and markets for utilizing the captured CO_2 (Xu et al., 2010). However, carbon capture and utilization (CCU) applications differ from CDR because the CO_2 will eventually return to the atmosphere.

Traditionally, biorefineries suffer from economic competitiveness, and integrating or retrofitting them with CCS technologies will add an extra cost. Nevertheless, the current record prices of carbon permits and the emergence of incentives for CCS are opening new avenues for BECCS projects (Laude et al., 2011). Consequently, a comprehensive understanding of the technical, economic, and environmental implications surrounding the large-scale deployment of biorefineries coupled with CCS is crucial to ensure their sustainable implementation and contribution to the climate goals (Pérez-Almada et al., 2023).

This study focuses on the valorization of olive pruning tree biomass (OPTB), which presents significant opportunities for utilization in biorefinery schemes. OPTB is an abundant biomass resource in the EU Mediterranean region (Galán-Martín et al., 2022), arising from the pruning of olive trees in the late winter or early spring in the form of leaves and branches. This pruning serves to enhance olive tree structure, stimulate new growth, and increase fruit production. Spain alone accounts for more than 2.70 million hectares of olive groves, yielding up to 4.0 tonnes of OPTB per hectare annually (Galán-Martín et al., 2022). Notably, this lignocellulosic biomass has traditionally been underutilized and regarded as waste, often burned in the field or incorporated crushed, and chipped into soils. Hence, there is a great opportunity to valorize this residual biomass and transform it into a potential feedstock for biorefineries that can be integrated with CCS technologies and contribute to national and global CDR commitments (Galán-Martín et al., 2022).

Numerous studies have explored the value of utilizing OPTB in various contexts, including experimental investigations and research delving into technical feasibility, economic aspects, and environmental performance. Romero-García et al. (2016) analyzed the economic and efficiency aspects of a biorefinery producing bioethanol and antioxidants from OPTB, finding that small-capacity biorefineries focusing on natural antioxidants displayed strong economic attractiveness. Similarly, Suzmosas et al. (Susmozas et al., 2019) conducted a techno-economic assessment of an OPTB-based biorefinery, showing its viability in producing ethanol, xylitol, antioxidants, and electricity, achieving energy self-sufficiency. More recently, Servian-Rivas et al. (2022) expanded on this by performing a life cycle assessment of two distinct OPTB-based biorefinery schemes. One configuration produced ethanol, xylitol, and antioxidants, while the other solely focused on the production of antioxidants. Their findings underscored the economic importance of antioxidants and the reduced environmental impacts of bio-based products compared with their business-as-usual production methods. The study also emphasized the importance of optimizing heating, water, and energy requirements, particularly through the combustion of residual biomass for steam and power cogeneration. Another line of research delved into the environmental trade-offs associated with biorefinery systems, such as the ecological negative side-effects arising from carbon reduction benefits in biomass-based power plants (Pang et al., 2017) while other studies proposed strategies to mitigate collateral environmental damages through recycling and integration (Martinez-Hernandez and Hernandez, 2018; Ramirez-Contreras et al., 2020).

Building upon this previous research on OPTB-based biorefineries, our study takes a comprehensive approach by conducting an integrated environmental and techno-economic assessment (ETEA) of a multi-product OPTB-based biorefinery coupled with a CCS system to produce bioethanol and antioxidants. To this end, we rely on a combination of process simulation (validated with experimental data) and life cycle assessment (LCA). Within this framework, the OPTB undergoes a cascading transformation, yielding high-value-added products, such as antioxidants and bioethanol. To meet the energy demands of the entire biorefinery (including heating and cooling), a BECCS cogeneration plant is integrated, utilizing OPTB as its primary fuel source. The novelty of this work is twofold. Firstly, it stands as the pioneering study that integrates an OPTB-based biorefinery with a BECCS system, presenting the potential to deliver carbon-negative bio-based products contributing to the CDR efforts. Secondly, from an environmental perspective, we conduct a comprehensive assessment across several environmental categories, extending beyond the scope of climate change impacts. This holistic approach allows exploring the potential occurrence of burden-shifting —a phenomenon where resolving one environmental challenge inadvertently gives rise to another.

2. Olive pruning tree biomass-based biorefinery. Scenario definition

This study centers on a biorefinery that converts OPTB into two valuable bio-products: antioxidants and bioethanol. Antioxidants are high-value compounds with potential applications in the food, cosmetic and pharmaceutical sectors. Bioethanol serves as an eco-friendly substitute for fossil gasoline in vehicles or chemical plants, contributing to reducing fossil fuel dependency and advancing energy independence.

Our ETEA study considers the entire supply chain of the biorefinery system, encompassing all the stages that ensure the timely and efficient delivery of feedstocks and products. The initial phase involves the preparation and acquisition of OPTB, which consists of branches and leaves resulting from olive tree pruning. These prunings are collected and stacked in piles, and chipped before their transportation to the biorefinery facility. The transportation of the feedstock typically relies on trucks, assuming an average distance of 100 km to the biorefinery. Ideally, the biorefinery should be strategically located in proximity to the feedstock source, ensuring sufficient and steady supply. Upon arrival at the biorefinery facility, the OPTB feedstock undergoes various processing and conditioning stages, involving equipment like shredders, grinders, and extractors, which vary according to the biorefining pathway. The pretreated OPTB is subsequently processed in parallel in the antioxidant and bioethanol production subsystems, each equipped with specialized machinery (see section 3.1 for a detailed description of the process flowsheet). Ultimately, the antioxidants and bioethanol are produced, which are prepared for distribution from biorefinery plants. The distribution of the bioproducts to customers and their usage phase are not considered in our study as we follow a cradle-to-gate approach.

In our study, we examine two alternative scenarios of the OPTB-biorefinery system outlined below (Fig. 1). These scenarios differ in the design and the energy source employed to power the processes of the biorefinery plant. The first scenario, denoted as "BIOR NG", represents the conventional or business-as-usual approach utilizing natural gas to fulfill the heating and cooling requirements necessary for the various biorefinery processes. In contrast, the alternative scenario, labeled as "BIOR CCS", integrates a BECCS technology to capture the biogenic CO_2 released from the fermentation unit (bioethanol subsystem) and the cogeneration unit, which burns OPTB and residual biomass to meet the energy requirements of the plant. The BIOR CCS scenario thus includes CO_2 capture, compression, transportation via pipeline, and the CO_2 geological injection ensuring secure long-term storage. These two scenarios are assessed and compared regarding technical, economic, and environmental performances.

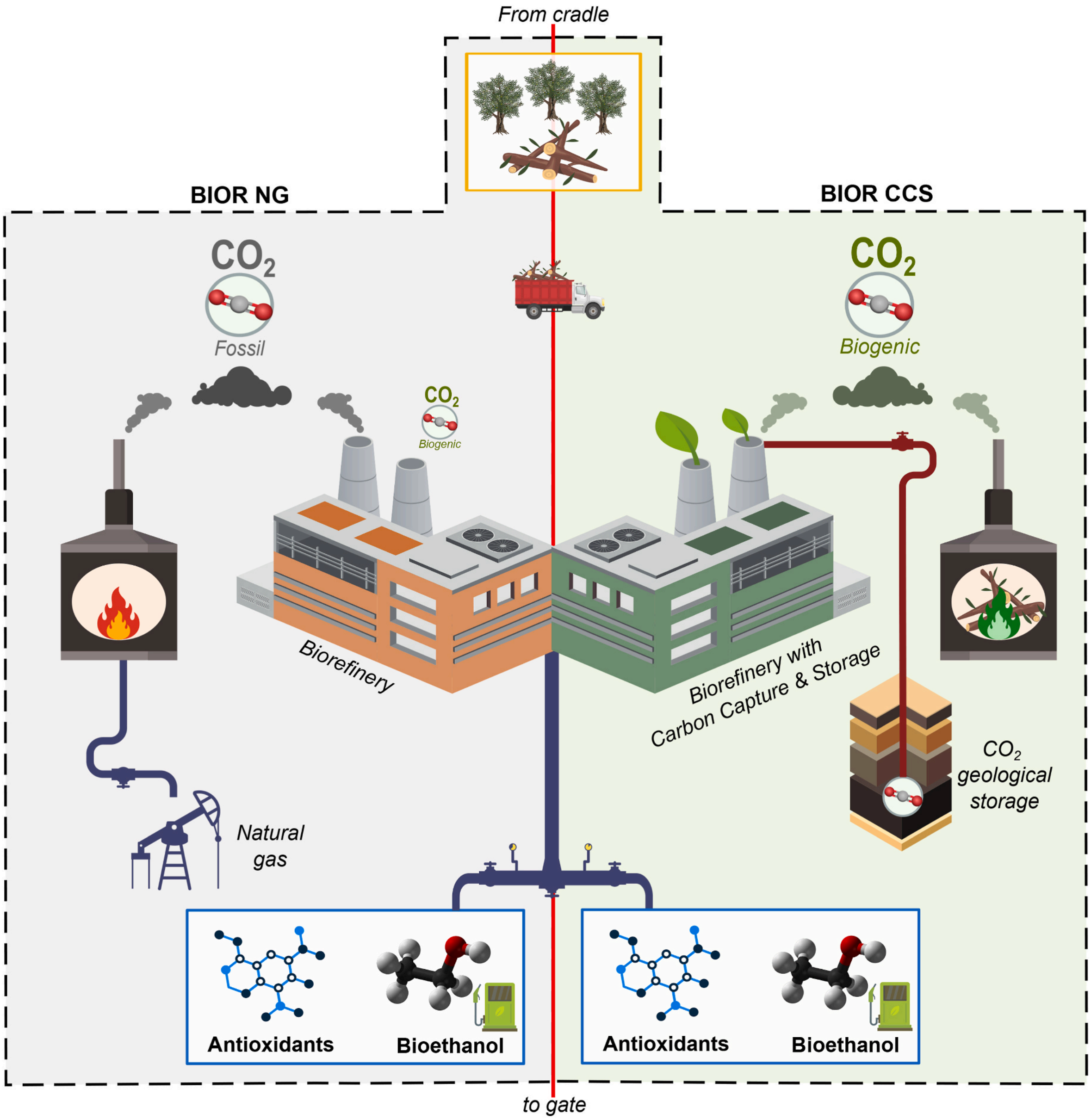

Fig. 1. Graphical overview of the two scenarios for the olive pruning tree biomass-based biorefinery. On the left, the fossil BIOR NG scenario, which relies on natural gas to power the biorefinery. On the right, the alternative scenario BIOR CCS shows the integration of bioenergy with carbon capture and storage subsystem.

3. Methods description

The comprehensive ETEA assessment of the OPTB-based biorefinery entails the integration of process simulation and LCA. The next subsections offer an in-depth explanation of these methodologies.

3.1. Modeling of the biorefinery with CCS

The biorefinery system comprises five interconnected subsystems, as illustrated in Fig. 2: the feedstock (SS1), the antioxidant (SS2), the bioethanol (SS3), the wastewater treatment (SS4), and the cogeneration unit (SS5).

The primary biomass feedstock utilized is OPTB, serving dual purposes in the production of bioethanol and antioxidants, as well as in direct combustion for heat generation.

The OPTB-based biorefinery plant was simulated in SuperPro Designer v9.0. The model was validated with experimental data (Romero-García et al., 2016). The system contains 55 unit operations, 7 reactors, and 22 heat exchangers (details of the simulation, refer to Appendix A1 and A2). The BIOR CCS scenario include the BECCS additional subsystem to capture, compress, and transport the biogenic CO_2 captured, which was modeled as in (Galán-Martín et al., 2021a). The potential site for the biorefinery is the Andalusia region in southern Spain, known for having the highest number of olive groves globally, resulting in a substantial amount OPTB available (Galán-Martín et al., 2022). However, it is important to note that determining the optimal location, design, and operational considerations of the biorefinery are beyond the scope of our current study. The process begins with the collection of the OPTB at the olive grove, followed by chipping before transportation to the biorefinery plant. Upon arrival, a series of pretreatment steps are executed, including the conditioning phase (removing impurities like soil, small stones, leaves, etc.), as well as grinding and sieving to homogenize the particle size from 4 to 10 mm. For the antioxidant subsystem (SS2 in Fig. 2), the daily feed comprises 1, 500 t of OPTB in dry weight (dw). The choice of this production capacity was based on several factors, including the availability of biomass in the proposed region to locate the biorefinery (Jaén, Spain), industry standards, and the scale at which the biorefinery model was found to operate optimally according to previous work (Romero-García et al., 2016). The biomass enters the aqueous extraction system, resulting in the separation of liquid and solid streams. Approximately 20 % (w/w) of the material is solubilized (Martínez-Patiño et al., 2015) with a 20 % (w/v) solid/liquid ratio (Conde et al., 2009). The liquid phase is then processed in a non-ionized adsorption chromatography column to purify the natural antioxidants This purification process involves the addition of a substantial amount of non-ionic hydrophobic resin and ethyl acetate at 3:1 (v/v) liquid-solvent ratio. The ethyl acetate operates in a closed circuit, which subsequently undergoes ultrafiltration to separate sugars (oligomers and mannitol). The remaining solution is evaporated and distilled, resulting in a product with a purity of 99 % (w/w). This filtered liquid contains phenolic compounds, with a concentration of 3.1 g gallic acid equivalent per 100 g of OPTB, representing a high-value-added product that includes the initial presence of mannitol (Martínez-Patiño et al., 2015).

Under these conditions, the recovery rate of phenolic compounds reached 55 %, with an extract purity of 36.95 % (g gallic acid equivalent/g ethyl acetate extract). In the ethyl acetate extracts, most of the natural antioxidants including hydroxytyrosol, oleuropein, tyrosol, and 3,4- dihydroxybenzaldehyde can be found. These antioxidants have widespread applications in the pharmaceutical, food, medicine, and cosmetic sectors (Romero-García et al., 2016). The outcome of this process is a mixture of antioxidants (12,082 t/yr) with a purity greater than 60 % (w/w) achieving an antioxidant recovery rate higher than 80 %. The chromatography column was loaded with a stream of 125 g ethyl acetate extract/L, using a ratio of 45.5 % (v/v) load stream/resin. The eluent for this process consisted of slightly acidic water (0.01 % H_2SO_4) at a ratio of 8 (v/v) (Romero-García et al., 2016).

The extracted solids undergo a filtration process and enter the bioethanol production subsystem (SS3 in Fig. 2) (Romero-García et al., 2016). The OPTB solids were subjected to a 30 % (w/v) successive solid loading pretreatment, using a mixture of water and phosphoric acid (0.5 % (w/v)). The hydrolysis pretreatment is conducted at 170 °C for a duration of 10 min. The addition of an overliming agent, calcium hydroxide, helps to maintain the pH level at 10. This pretreatment effectively breaks down the hemicellulose and cellulose structures.

Following the pretreatment, the dissolved sugars are further processed through co-saccharification and enzymatic fermentation, resulting in the recovery of nearly 70 % of the sugars in both the hydrolysate and the saccharification. In the subsequent co-fermentation stage, a modified microorganism (*Escherichia coli* MS04) was used to ferment all

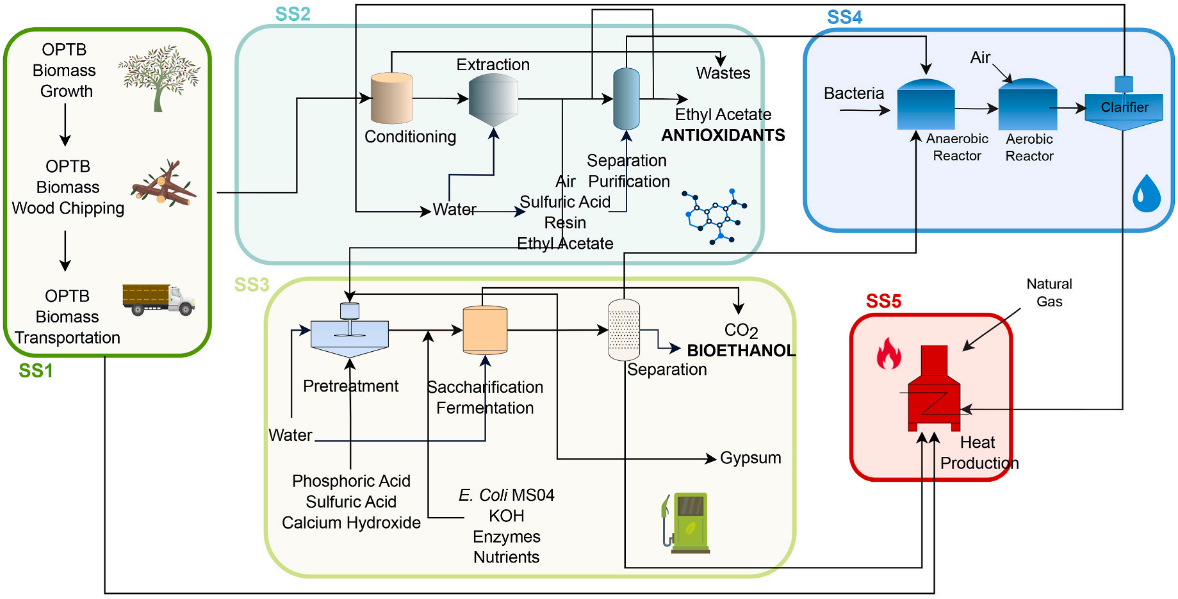

Fig. 2. Schematic flowsheet of the olive pruning tree biomass biorefinery system comprising five subsystems: SS1: Feedstock subsystem, SS2: Antioxidant system, SS3: Bioethanol subsystem, SS4: Wastewater treatment subsystem, SS5: Cogeneration subsystem.

the glucose and over 93 % of the xylose (Romero-García et al., 2016). Nutrients were added to feed the microorganisms, and KOH was added to maintain the pH level at 7. The ethanol produced achieves a yield of 90.20 % of the theoretical maximum (Martínez-Patiño et al., 2015). The final separation stage involves the extraction of ethanol through a double-column sequential rectifying-stripping distillation system. This process achieves an ethanol concentration of 91.00 % and a purity of 99.70 % after molecular sieves (azeotropic distillation). In total, the annual production of bioethanol amounts to 78,074 t/yr.

The wastewater generated from SS2 and SS3 enters SS4 for treatment within a sequential continuous anaerobic-aerobic reactor system. In the initial anaerobic reactor, biogas is produced consisting of 41.00 % methane and 59.00 % carbon dioxide by volume. Subsequently, a clarifier separates the solids from liquids. The separated solids are directed toward a vacuum filter and to the cogeneration unit for combustion, while the treated liquids are recycled for use as process water. Within the cogeneration unit (SS5 in Fig. 2), natural gas or residual biomass along with OPTB are combusted at high temperatures, resulting in the production of high and low-pressure steam (depending on the scenario assessed). This steam will generate the heat and cooling energy demands to cover the needs of the plant. In the BIOR NG scenario, process biomass residues and natural gas are combusted to cover the energy requirements of the biorefinery. On the other hand, in the BIOR CCS, the cogeneration unit is powered by the utilization of process residues and OPTB as feedstock. In both scenarios, 69.50 % of heating demands are covered by process residual biomass, while 30.50 % is covered either by natural gas or OPTB.

A detailed report on the simulation results for the biorefinery is provided in the appendix, sections A1 and A2.

3.2. Economic assessment

In the economic evaluation, the goal is to compare the scenarios based on a common FU, i.e., to compare the total annual cost (TAC) to produce 1 kg of bioethanol and 0.15 kg of antioxidants. To perform these calculations, we assume a plant's lifetime of 25 years, taking into account the simulation results for a processing capacity of 1,500 t OPTB dw/day. The *TAC* includes both the capital expenditures (*CAPEX*) and operational expenditures (*OPEX*) associated with biorefinery production as detailed in reference (Ganat Tarek Al Arbi Omar, 2020) and described in Eq. (1)

$$TAC = AF \ast CAPEX + OPEX(fix, var) \tag{1}$$

CAPEX includes one-time long-term investments (Pritha Banerjee, 2022) which are annualized using the capital annualization factor (*AF*) and an interest rate of 6.00 %. The *AF* is calculated as in Eq. (2).

$$AF = \frac{i(1+i)^n}{(1+i)^n\text{-}1} \tag{2}$$

The *OPEX* encompasses both fixed and variable costs (*fix, var*) associated with ongoing operations, including expenses related to raw materials, utilities, labor, and maintenance costs. As for the *CAPEX*, the equipment costs were determined using correlations from Sinnot and Towler (Sinnott and Towler, 2009). See details on the calculations in Appendix section A.3.

Finally, we employed Equation (3) to estimate the levelized cost of the biorefinery (*LCOB*) per FU (Lehtveer and Emanuelsson, 2021) by considering the CAPEX, the variable and fixed operating expenditures ($OPEX_{fix}$ and $OPEX_{vr}$, respectively) and the total annual production of the biorefinery (*TAP*, in kg).

$$LCOB = \frac{CAPEX \ast AF + OPEX_{fix} + OPEX_{var}}{TAP} \tag{3}$$

Furthermore, the economic evaluation also includes economic metrics to assess the financial viability of the biorefinery, including the net present value (*NPV*), the payback time (*PBT*), and the return on investment (*ROI*). A detailed description of the economic calculations and the *CAPEX* and *OPEX* parameters can be found in the Appendix (section A3).

3.2.1. Sensitivity analysis and breakeven point

A sensitivity analysis of the economic assessment was performed by defining optimistic and pessimistic scenarios considering maximum and minimum costs for the parameters involved (costs of raw materials and utilities). The parameters defined on the cost parameters are detailed in Table A.5. The results of the sensitivity analysis are presented with error bars in Fig. 3 depicting the pessimistic and optimistic uncertainty cost ranges.

Moreover, a breakeven point analysis was conducted to determine the minimum selling price of bio-based products for the BIOR CCS scenarios that make the NPV equal to one for the BIOR NG scenario. The breakeven point serves as a pivotal metric, indicating the price threshold at which the revenues generated from selling the bio-based products of the alternative BIOR CCS (premium pricing considerations) become competitive compared to the fossil counterparts (BIOR NG scenario). To conduct the breakeven point analysis, the selling price of the bioethanol and antioxidants in the BIOR CCS are increased (without altering those of the BIOR NG scenario) until equilibrium was achieved in NPV between both scenarios. Moreover, a complementary analysis was performed to determine the tax credit that should be provided per CO_2 sequestred to incentivize the BIOR CCS scenario.

3.3. Life cycle assessment

To comprehensively evaluate the environmental impacts of the OPTB-based biorefinery, we employed LCA, a standardized methodology to evaluate the potential environmental impacts of a system considering its entire life cycle. Our LCA adheres to the steps and guidelines outlined in ISO 14040 (ISO, 2006a) and 14044 (ISO, 2006b).

The first phase involves defining the goal and scope. In this study, the objective is to assess and compare the environmental impacts associated with the production of bioethanol and antioxidants, considering the OPTB-based biorefinery scenarios described in section 2. Our approach adopts a cradle-to-gate scope, with the system boundaries from the acquisition of raw materials, growth of biomass, pretreatment of olive tree prunings, its transportation to the plant, and all subsequent processes at the biorefinery facility, up to the delivery of the bio-based products at the biorefinery gate (excluding the end-use phase). For illustration, we consider a cradle-to-grave scenario assuming that the

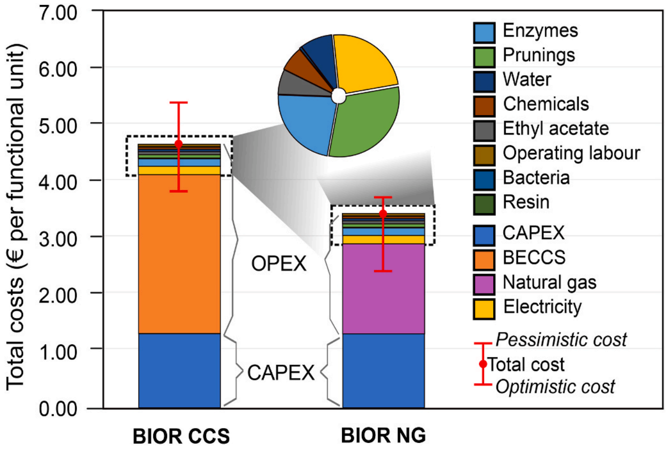

Fig. 3. Costs breakdown of the two scenarios. The column on the left BIOR CCS integrates bioenergy with carbon capture and storage (BECCS) within the biorefinery. The column on the right corresponds to the BIOR NG scenario, which relies on natural gas to power the biorefinery. Error bars depict the pessimistic and optimistic cost estimates according to the sensitivity analysis conducted.

bioethanol is incinerated resulting in emissions equivalent to 1.90 kg CO_2 per kg of bioethanol combusted. These direct emissions are estimated assuming the complete combustion of the ethanol. Note that determining if a BECCS system achieves net negative emissions requires a comprehensive cradle-to-grave assessment of all greenhouse gas emissions. Furthermore, we followed an attributional approach that assigns all impacts to the bio-based products considering that the life cycle of the product system remains fixed. The functional unit (FU) entails the simultaneous and coupled production of 1 kg of bioethanol and 0.15 kg of antioxidants, derived from the division of the total feedstock daily input by the total daily production of bioethanol and antioxidants (results from the simulation). This multi FU ensures a fair comparison of the BIOR NG and BIOR CCS scenarios which is the main objective of this study. Note that we avoided dividing the biorefinery into mono-functional systems and have refrained from utilizing allocation procedures, as these can introduce significant uncertainty and greatly influence the results.

The second phase of the LCA involves the Life Cycle Inventory (LCI), which entails identifying and quantifying all inputs and outputs of the system over its entire life cycle (e.g., raw materials, energy, emissions to air). Data for the foreground system (the biorefinery production site) were obtained from the mass and energy balances of the plant simulation implemented in SuperPro Designer v9.0 (Table A.1 and Table A.7 in Appendix). Data for the background system were retrieved using datasets from Ecoinvent v3.8 (Wernet G. et al., 2016) and completed with literature to fill the gaps. Note that we account for the impacts associated with the pruning raw material employed in the biorefinery, including all relevant processes of growing olive trees, including fertilizers, pesticides, nutrients, land occupation techniques, and irrigation among others. For this, we used the 'Olive fruit production – ES' dataset from Ecoinvent v3.8 and allocated all inputs and outputs between the olive fruit and the prunings based on an economic factor as detailed in Appendix section A.4. We also considered chipping and mulching pretreatment activities as well as transportation using a lorry for a 100 km distance from the olive groves. The complete inventory of the OTPB is provided in Table A.6 in Appendix. Moreover, for the cogeneration unit (SS5) in the BIOR CCS, we primarily rely on previous work modeling a BECCS system (Galán-Martín et al., 2021a) by adapting it to our specific context by considering the olive pruning as input. In essence, the BECCS system comprises a heat and power cogeneration plant integrated with post-combustion CO_2 capture technology using monoethanolamine solvent (with a 90 % capture efficiency). In total, 1.69 kg of biogenic CO_2 is captured per kWh delivered with the BECCS system, calculated based on an OPPTB carbon content of 49.40 %, a chip-to-kWh ratio of 0.84 kg, and accounting for the own energy requirements of the CCS system. Once captured, the CO_2 is compressed to 110 bar and transported via a pipeline 200 km for secure storage in a deep saline aquifer. Note that our study incorporates illustrative assumptions regarding the supply chain (e.g., biomass and CO_2 transportation distances), which may not apply to all regional contexts. Therefore, each project needs to consider its unique characteristics and regional aspects when implementing our model. All the detailed LCIs tables for the entire biorefinery system can be found in section A4 of the appendix (Tables A6-7).

Concerning the third phase, the life cycle impact assessment (LCIA), we implemented the inventory in SimaPro v9.0. To transform the inventory entries into environmental impacts, we employed the ReCiPe 2016 (Hierarchist perspective) (Huijbregts et al., 2017). In particular, we used the midpoint categories indicatos that allow assessing the climate change impact through the Global Warming (GWP) indicator (carbon footprint measured in kg CO_2 eq). Notably, as we deal with a BECCS system, the biogenic CO_2 embodied in the OPTB is modeled as a negative entry from fossil CO_2 emissions in the system. Hence, a negative input of -1.52 kg CO_2 per kg of pruning was estimated based on a water content of 27.60 % and a carbon content, on a dry basis, of 49.40 %, as per the Phyllis2 database (TNO Biobased and Circular Technologies, 2024). Additionally, we considered seventeen other indicators referring to particular environmental problems including stratospheric ozone depletion (SOD), ionizing radiation (IR), ozone formation, human health (OF, HH), fine particulate matter formation (FPMF), ozone formation, terrestrial ecosystems (OF, TE), terrestrial acidification (TA), freshwater eutrophication (FER), marine eutrophication (MER), terrestrial ecotoxicity (TE), freshwater ecotoxicity (FE), marine ecotoxicity (ME), human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HNCT), land use (LU), mineral resource scarcity (MRS), fossil resource scarcity (FRS) and water consumption (WC).

This holistic approach considering the midpoint categories offers a comprehensive assessment of the broad environmental implications of the biorefinery, providing an understanding of its overall environmental sustainability.

4. Results and discussion

4.1. Economic performance

The results of the economic analysis are presented in Fig. 3. Note that the key distinction between the two scenarios lies in the costs of the heating needs. In the case of the BIOR NG scenario, which relies on natural gas, the heating cost is 89.70 €/MWh, whereas for the BIOR CCS scenario, the costs escalate to 160.00 €/MWh. Considering the central values for the cost parameters, due to the increased investment and operational costs associated with CCS technology, the BIOR CCS scenario emerges as a more expensive alternative compared to the business-as-usual BIOR NG scenario, with costs of 4.70 vs. 3.48 € per FU, respectively. These cost values are aligned with those reported in the literature for bioethanol production from biomass, ranging from 0.86 up to 1.50 €/kg (Romero-García et al., 2016; Sánchez et al., 2013). Notably, our costs are higher due to antioxidant production, which mounts up to 7.33 €/0.15 kg (Romero-García et al., 2016).

Even under the most optimistic cost data, the cost of the BIOR CCS scenario (3.93 € per FU) would be higher than the pessimistic estimates for the BIOR NG scenario (3.74 € per FU). This is attributed to the substantial upfront investments necessary for establishing the CCS infrastructure, which pose a financial barrier for stakeholders contemplating BECCS deployment. These investments entail not only the development of CCS technology within the biorefinery but also the construction of CO_2 transportation and storage infrastructure. Furthermore, BIOR CCS necessitates additional energy and biomass feedstocks to sustain the BECCS process, further increasing the economic burden.

Regardless of the scenario, the OPEX outweighs CAPEX, comprising 64.13 % and 73.46 % of the BIOR NG and BIOR CCS scenarios, respectively. It is worth noting that the primary contributor to total costs in both scenarios is heating and cooling, accounting for over 44.83 % and 59.18 % in each respective scenario. Following this, the costs associated with the OPTB biomass raw material and enzymes contribute 30.00 % and 23.00 % in such scenarios, while other factors such as ethyl acetate, other chemicals, and bacteria collectively contribute 14.00 % of the total OPEX (Fig. 3, pie chart).

The CAPEX represents a substantial portion of the overall cost, amounting to 35.87 % and 26.54 % in the BIOR NG and BIOR CSS scenarios, respectively. The primary cost driver within the CAPEX is the total equipment cost, accounting for over 50.00 % of the total, followed by the costs associated with the inside battery limits that contribute nearly half of the total CAPEX (49.00 %). The remaining 1.00 % of CAPEX can be attributed to various factors including outside battery limits and other contingencies (see parameters in the Appendix, section A3).

The NPV in the BIOR NG scenario stands at 1.00 billion €, whereas for the BIOR CCS worsens up to −74.12 million € (detailed economic parameters are provided in Table A4 of the Appendix). Similarly, the BIOR CCS underperforms compared to the BIOR NG in other financial metrics. For example, the PBT for the BIOR CCS is more than double (14.00 vs 6.17 years), while the ROI is less than half (7.16 % vs 16.20 %).

The worse economic performance in the BIOR CCS scenario may be attributed to the higher upfront costs or excessive operating expenses, with the heating requirements from BECCS as the main driver of the overall costs (Fig. 3). However, it is important to acknowledge the uncertainties inherent in these calculations, particularly concerning the dependency on volatile natural gas prices, which can be influenced by geopolitical tensions, supply disruptions, and shifts in energy policies. Additionally, other factors such as technological advancements, new regulatory frameworks, and market dynamics could also impact the economic viability of the BIOR CCS scenario. Therefore, while the current analysis provides valuable insights, ongoing monitoring and adaptation to evolving circumstances are essential for robust decision-making in transitioning toward more sustainable energy systems.

The results of the break-even point analysis show that increasing slightly the price of the antioxidants to 11 €/kg (Alibaba Group, 2023; Servian-Rivas et al., 2022) and the price of bioethanol to 3.85 €/kg (METRO, 2023) would bring the BIOR CCS scenario close to the break-even point where the NPV equals zero. This price for bioethanol is aligned with current market prices, which typically range from 3.85 to 4.60 € per liter (Laboratorio SYS, 2023; METRO, 2023). Moreover, increasing the selling price of the bio-based products by 25.50 % would make the BIOR CCS competitive compared to the BIOR NG scenario (NPV of BIOR CCS equal to that of the BIOR NG). Hence, a prior investigation showed that carbon-negative products attract consumers, potentially increasing sales by an average of 60 % (Hong et al., 2023). Both willingness to pay a premium for carbon-negative products and for carbon credits reflect growing environmental awareness and changes in consumer preferences. This shift in consumer behavior could be pivotal in promoting carbon-negative business models and overcoming economic barriers. Moreover, the introduction of carbon-negative bio-products presents an opportunity for market differentiation and competitive advantage, which may position the industry at the forefront of sustainability-driven innovation.

In addition to the premium nature of the products, the regulatory environment will play a pivotal role in shaping market dynamics for carbon-negative products. Various policies and incentives have already been implemented to create a conducive market environment for carbon-negative products. Hence, recently the EU approved the EU carbon removal certification framework which aims to develop a voluntary certification to incentivize scale-up carbon removal activities sustainably (European Commission, 2022). This regulatory framework will encourage the implementation of carbon removal business models by stimulating both public and private financing options as well as voluntary carbon markets and eco-labeling initiatives which will contribute to scaling up production effectively. Furthermore, the CDR efforts are being incentivized and rewarded through various means such as direct subsidies, and taxes, which can help offset the additional expenses. Hence, we found that a subsidy or tax credit of 9.75 € per tonne of CO_2 sequestred would render the NPV of the BIOR CCS scenario positive while increasing it up to 132.35 € per tonne of CO_2 sequestred would make it economically attractive compared to the BIOR NG, reaching the break-even point. Notably, the tax credit value to make the BIOR CCS profitable is lower than the Q45 tax credit provided by the United States for carbon sequestration, highlighting the potential to promote and support biorefineries with CCS.

4.2. Environmental performance

We start by assessing the carbon footprint using the GWP indicator from ReCiPe (2016) (Huijbregts et al., 2017). Fig. 4 provides a detailed breakdown of the carbon footprint of both scenarios. In the BIOR CCS scenario, the carbon footprint reaches net negative emissions, totaling −84.37 kg CO_2eq for the production of 1 kg of bioethanol and 0.15 kg of antioxidants. Conversely, the business-as-usual scenario (BIOR NG) shows a positive carbon footprint of +28.22 kg of CO_2eq per FU. The negative outcome of BIOR CCS is due to the heating requirements of the biorefinery (280 MJ per FU) being met by burning residual biomass and OPTB in the cogeneration plant with CCS. During the combustion process, the biogenic CO_2 released is captured and stored deep underground. This energy will be provided with a carbon intensity of −0.30 CO_2eq/MJ. Furthermore, the almost pure biogenic CO_2 released during the fermentation is also directed to the CCS systems and captured. Note, that all these biogenic CO_2 are modeled as a negative flow in our LCA calculations which compensates for all the positive GHG of the biorefinery system. Additionally, our results in Fig. 4 account for end-use phase emissions, assuming that the bioethanol is combusted and contributes to positive emissions in the"bioethanol combustion” category. To put context, considering that our biorefinery at scale would produce 213,900 kg of bioethanol and 33,100 kg of antioxidants per day, this equates to around 7.60 million tonnes of CO_2 eq that would be captured annually. This amount of CDR solely provided by the biorefinery plant would represent approximately 2.80 % of the annual GHG emissions in Spain (Macrotrends LLC, 2022).

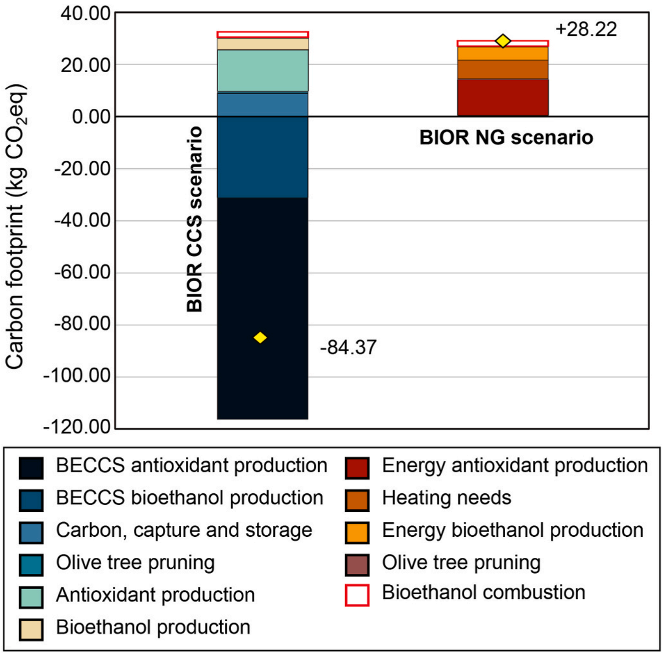

Fig. 4. Breakdown of the carbon footprint for the BIOR NG and BIOR CCS scenarios for the production of 1 kg of bioethanol and 0.15 kg of antioxidants.

Analyzing the carbon footprint breakdown, the antioxidant subsystem (SS2 in Fig. 2) is the main contributor to the carbon footprint representing 55.80 % and 48.70 % of the positive GHG in BIOR NG and BIOR CCS scenarios, respectively. This can be attributed to the utilization of ethyl acetate, its recycling loop after a double stripping distillation column, as well as the energy demands required for the purification of polyphenols. After the antioxidant subsystem, in the BIOR NG, subsystems SS3 and SS5 represent 16.54 % and 19.43 % of the total carbon footprint, respectively. In contrast, in the BIOR CCS scenario, the positive GHG emissions from the CCS system contribute substantially to the carbon footprint (29.73 % of the positive emissions). Additionally, in the BIOR CCS, we observe carbon-negative contributions from subsystems SS2, SS3, and SS5, contingent on their energy requirements being supplied by the carbon-negative heating from BECCS.

We next report the results of the 17 remaining midpoint impact categories of the ReCiPe 2016 methodology to compare the environmental performance of the BIOR NG and BIOR CCS scenarios. Fig. 5 shows the relative environmental impact of the scenarios across the 17 categories while the absolute values in the specific units are provided in Table A7 in the Appendix. Although the BIOR CCS scenario outperformed BIOR NG in the Global Warming category (as shown in

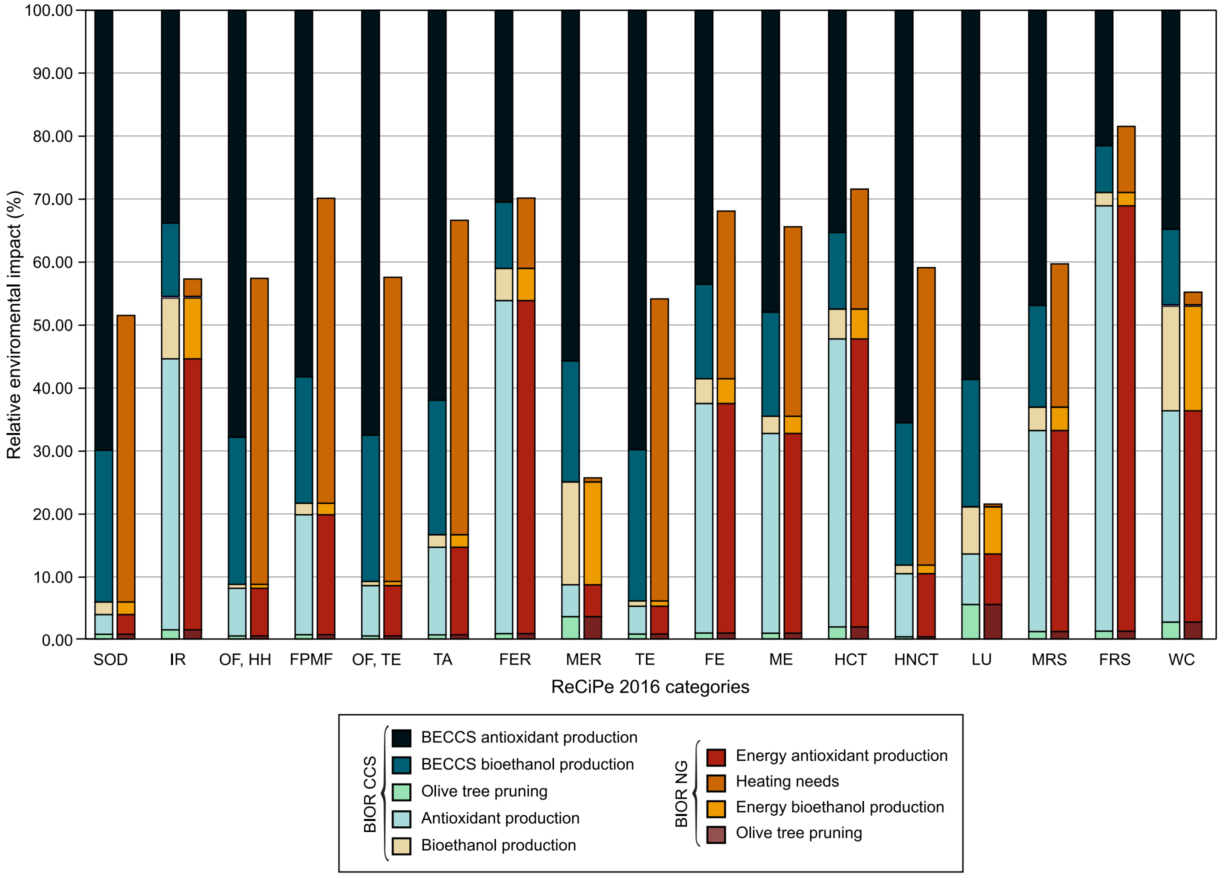

Fig. 5. Comparative environmental implications of the two biorefinery scenarios. The column on the left BIOR CCS scenario integrates bioenergy with carbon capture and storage within the biorefinery. The column on the right corresponds to the BIOR NG scenario, which relies on natural gas to power the biorefinery. SOD: stratospheric ozone depletion, IR: ionizing radiation, OF, HH: ozone formation, human health, FPMF: fine particulate matter formation, OF, TE: ozone formation, terrestrial ecosystems, TA: terrestrial acidification, FER: freshwater eutrophication, MER: marine eutrophication, TE: terrestrial ecotoxicity, FE: freshwater ecotoxicity, ME: marine ecotoxicity, HCT: human carcinogenic toxicity, HNCT: human non-carcinogenic toxicity, LU: land use, MRS: mineral resource scarcity, FRS: fossil resource scarcity and WC: water consumption.

Fig. 4), a different picture emerges when analyzing the remaining categories. Notably, the BIOR CCS exhibits higher environmental footprints than BIOR NG across all the impact categories. This indicates that the advantages gained in terms of climate change mitigation might come at the expense of exacerbating other environmental issues, some of which display notable discrepancies between the scenarios. The increase in the impact is largely attributed to the higher need for OPTB raw materials required to cover the heating requirements of the biorefinery and energy required for the CCS system. Note that despite the OPTB being mainly considered today as residue, we allocate environmental burden to this biomass waste generated at the agricultural phase of olive production (see appendix, section A4).

The environmental impact is of particular concern in the BIOR CCS scenario when compared to the BIOR NG in terrestrial ecotoxicity (TE), marine eutrophication (MER), land use (LU), and fossil resource scarcity (FRS). In the case of eutrophication (MER), BIOR CCS presents an impact particularly high, 75 % higher than the BIOR NG scenario. This increase is mainly attributed to the nitrogen and phosphorus fertilizers associated with olive cultivation (OPTB) and the production of monoethanolamine. Moreover, land use (LU) is nearly 80 % more extensive in the BIOR CCS scenario due to the land transformation and occupation attributed to olive tree cultivation, which is linked to the OPTB feedstock. Similarly, water consumption (WC) is 45 % higher in the BIOR CCS scenario, again attributed to the increased irrigation demands. In our assumptions, we consider that only 25 % of the agricultural area is irrigated (1,500 m^3/ha year). Additionally, the impact in the toxicity-related categories (TA, TE, HNCT, and FE) is also higher, likely due to the various chemicals involved in the process.

Analyzing the breakdown of impacts presented in Fig. 5, the antioxidant subsystem (SS2) combined with BECCS are the main drivers in most impact categories, representing between 21.53 % and 69.98 % of the total impact depending on the specific category. The high-energy demand with the antioxidant subsystem, which accounts for 73.65 % of the total energy requirements of the biorefinery, leads to the attribution of all impacts associated with the energy source to this particular subsystem. For some impact categories, such as stratospheric ozone depletion, terrestrial ecotoxicity, and human non-carcinogenic toxicity, the antioxidant separation is the main contributor.

The presence of environmental trade-offs associated with the BIOR CCS underscores the importance of preventing or minimizing them. Most of the collateral damages are linked to the increased consumption of olive prunings in the BIOR CCS scenario to satisfy the heating demands. However, it is important to note that attributing these unintended consequences solely to the biorefinery strategy due to the allocation of burdens to the pruning residues may overlook the value derived from their valorization. Otherwise, these prunings would be burned or chopped and used as mulch in the olive orchard, incurring both economic and environmental burdens. Nevertheless, to mitigate eutrophication, which is primarily driven by nutrient runoff (attributed to olive pruning from olive production), targeted measures such as buffer zones and precision agriculture techniques may help reduce nutrient loading in adjacent water bodies. Similarly, for water consumption and land use, implementing improved and sustainable agricultural practices during the agricultural phase will aid in mitigating these footprints. Some strategies may include the implementation of drip irrigation systems to minimize water loss through evaporation and runoff or the application of organic mulch around olive trees to conserve soil moisture. Concerning land use, increasing density or transitioning to agroforestry systems may help maximize land use efficiency and biodiversity. All of these considerations play a pivotal role in determining the optimal site for our biorefinery. By evaluating potential sites against criteria related to environmental impact mitigation, including proximity to water bodies, existing land use patterns, and biodiversity hotspots, the biorefinery could be strategically positioned to minimize its environmental footprint while maximizing its socio-economic benefits.

5. Conclusions

In this work, we rely on process simulation and LCA methodologies to delve into the interplay of the technical, economic, and environmental implications surrounding the integration of BECCS into a biorefinery that converts OPTB into bioethanol and antioxidants.

On the economic front, the adoption of BECCS comes with substantial financial implications. It entails higher upfront investments and operational costs due to the intricate CCS infrastructure. In our analysis, we observed that the BIOR CCS scenario ultimately incurs a 30 % higher cost compared to the business-as-usual scenario (BIOR NG). Nonetheless, it is important to note that the costs associated with BECCS projects are expected to decrease over time as technology advances and economies of scale come into play. To overcome these economic barriers and logistical challenges, the EU has introduced various funding mechanisms, legislative frameworks, and strategic initiatives such as the Innovation Fund, the Connecting Europe Facility, the Just Transition Fund, and the EU certification system for carbon removals which collectively represent an opportunity to facilitate the adoption of large-scale BECCS projects. Hence, our results demonstrated that integrating insights from consumer behavior, policy interventions, and regulatory frameworks is crucial. The higher production costs associated with the carbon-negative bioproducts delivered may find a receptive market among environmentally conscious consumers who are increasingly willing to pay premium prices to support carbon offset projects. Furthermore, strategic policy interventions, such as carbon taxes can play a pivotal role in offsetting the initial capital costs, rendering BECCS economically appealing in the long run and expediting CCS infrastructure development.

On the environmental dimension, integrating CCS into the biorefinery allows for reducing substantially the carbon footprint even reaching a negative emissions balance due to the energy-intensive processes being met with carbon-negative energy. This achievement could be a game-changer in the ongoing efforts to combat climate change and promote CDR initiatives in the biorefinery context. However, these environmental gains are not without their own set of challenges. Hence, our study sheds light on a complex trade-off. Notably, we have identified areas of concern related to eutrophication, water consumption, and land use, most of them related to the higher utilization of OPTB. These environmental trade-offs are crucial in selecting the best location for the biorefinery aimed at mitigating environmental footprint and enhancing socio-economic advantages.

Additionally, scaling up a biorefinery coupled with CCS will involve technical and logistical complexities throughout the entire value chain. It will require significant infrastructure development, including the biomass processing and conversion processes itself, storage facilities for securing a reliable biomass supply, CO_2 capture system, and transportation network and storage sites for CO_2. Identifying and addressing technical, economic, and environmental challenges across the value chain before full-scale deployment is key. It requires optimal siting strategies, considering the proximity to biomass sources and availability of suitable geological formations for CO_2 storage, adherence to regulatory requirements, effective logistical planning including partnerships with biomass suppliers to ensure uninterrupted operations, and community engagement to ensure project viability and social acceptance.

In conclusion, our research underscores the importance of taking a holistic view in shaping decision-making and policy formulation for the integration of CCS into biorefinery systems. While this integration holds immense promise in terms of reducing carbon emissions, it also poses technical, economic, and environmental challenges. It is imperative for decision-makers, policymakers, and stakeholders to recognize the multidimensional nature of these challenges and to prioritize sustainable practices. By integrating insights from consumer behavior research, designing effective regulatory frameworks, and competitive positioning, scalability challenges might be overcome towards unlocking the CDR potential and fostering long-term business resilience by delivering carbon-negative bio-products.

CRediT authorship contribution statement

Stylianos Fanourakis: Writing – original draft, Visualization, Investigation, Formal analysis. **Juan Miguel Romero-García:** Writing – review & editing, Investigation, Formal analysis, Data curation. **Eulogio Castro:** Writing – review & editing, Validation, Investigation, Data curation. **Laureano Jiménez-Esteller:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Investigation, Formal analysis, Data curation, Conceptualization. **Ángel Galán-Martín:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Project administration, Methodology, Investigation, Formal analysis, Data curation, 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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2024.142361.

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