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Research Paper

Energy and GHG emissions assessment for biochar-enhanced advanced biofuels value chains

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ABSTRACT

Biochar has an enormous potential to store carbon in the long-term. Differently than BioEnergy Carbon Capture and Storage (BECCS) technologies, biochar incorporates biogenic carbon in a solid form that offers multiple benefits as carbon sink, soil improver or for advanced materials production. The present study proposes an innovative approach, where carbon sequestration through biochar is obtained through the integration of slow pyrolysis with fast pyrolysis in decentralised biorefining systems, and then converted producing drop-in fuels from pyrolysis oil hydrotreating or gasification and Fischer-Tropsch (FT) synthesis. The scope is either to achieve negative GHG emissions assigned to advanced biofuels, or to export the generated carbon credit for the carbon markets (i.e. outside the biofuels carbon intensity). The innovative concept entails process integration and optimisation for the different stages of biomass drying, conversion and upgrading into biofuels in a way to reduce fossil-based inputs, applying a full value chain approach. Methodological choices for the assumptions on life cycle emissions calculation are discussed, evaluating the environmental performances by comparing the new concept to traditional biofuels value chains. Using a tailored lifecycle accounting methodology, this paper demonstrates that high GHG emissions savings can be achieved. The improved scenario shows how the carbon sequestration with biochar further reduces the carbon intensity up to –4.2 $gCO_2e\ MJ^{-1}$ for pyrolysis oil-based fuels, and to -20.2 $gCO_2e\ MJ^{-1}$ for FT-based fuels: this demonstrated that carbon negative sustainable biofuels can be obtained. The study demonstrates that an integrated biorefinery of 100 MW capacity can deliver additional 13.3 and 6.8 ktons of CO_2e of GHG savings of per year, from drop-in fuels made of hydrotreated pyrolysis oil and FT synthesis, respectively.

1. Introduction

Carbon is one of the most important building blocks for life, given its ability to form complex molecules as part of the life cycle and to release energy generating carbon dioxide. A sustainable use of carbon would imply preserving the original balance between carbon dioxide in the atmosphere and maintaining solid carbon deposits sequestered in geological sites (in the form of e.g. coal, crude oil, natural gas), through a global effort to gradually transforming the current economy based on fossil sources into a sustainable and carbon neutral economy based on recycling and circularity [1]. The European Green Deal [2] introduced by the European Commission (EC) since 2019 forms the cornerstone of pivotal regulatory frameworks aimed at curbing carbon emissions across diverse sectors. These measures encompass several key pillars: the “Sustainable Carbon Cycles” Regulation prioritizes carbon farming and sustainable agricultural practices, emphasizing the need for environmentally conscious approaches [3]; the updated Renewable Energy Directive (EU) 2023/2413 establishes ambitious decarbonization targets

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Abbreviations: GHG, Greenhouse Gas; BECCS, BioEnergy Carbon Capture and Storage; LCA, Life Cycle Assessment; LCI, Life Cycle Inventory; EC, European Commission; JRC, Joint Research Centre; TF, Task Force; BIP, Biomethane Industrial Partnership; DG ENER, Directorate-General for Energy; IEA, International Energy Agency; IPCC, Intergovernmental Panel on Climate Change; SMR, Steam Methane Reforming; ETS, EU Emissions Trading System; ILUC, Indirect Land Use Change; RED II, Renewable Energy Directive (EU) 2001/2018; JECv5, Joint Research Centre-EUCAR-CONCAWE collaboration for a Well-to-Tank study; WTT, Well-To-Tank; FPBO, Fast Pyrolysis Bio-Oil; HPO, Hydroprocessed Pyrolysis Oil; FT, Fischer-Tropsch; EFG, Entrained Flow Gasifier; CDR, Carbon Dioxide Removal; EU MED, European Union Mediterranean; RR, Random Reflectance; BECOOL, Brazil-EU Cooperation for Development of Advanced Lignocellulosic Biofuels; HVO, Hydrotreating of Vegetable Oils; GWP, Global Warming Potential; PV, Photovoltaic; m.c., moisture content.

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specifically developed for the energy and transport sectors, setting a significant push towards cleaner energy sources [4]; the Carbon Removal certification framework seeks to expand carbon capture solutions, focusing on certifying and scaling up technologies that effectively remove carbon from the atmosphere [5]. These collective actions underline the EU's commitment to fostering bio-based carbon sequestration as a crucial strategy in the broader initiative to achieve decarbonization goals.

Specifically, biochar production represents a well-established solution that leads to biocarbon sequestration in the form of a porous arrangement of carbon atoms, creating a stable matrix [6]. This porous structure gives to biochar peculiar properties, providing a large surface area that allows it to effectively interact with the environment, contributing to retain carbon and in some cases, to improve soil quality [7]. Biochar is produced through pyrolysis or gasification of organic material in low-oxygen conditions, which are thermochemical processes well-developed for bioenergy and biofuels production [8]. Today the predominant utilization of biochar involves its use as charcoal for bioheat provision. However, when produced from specific lignocellulosic biomasses under controlled thermochemical conditions, it is possible to optimize the internal geometry in terms of specific pores and internal surfaces dimension: this customization aligns with the intended final application of the biochar [9,10]. For instance, these characteristics give to biochar, when applied to the soil, the ability to retain water and nutrients that are afterwards slowly released over time. It is also a precursor to advanced sustainable materials production [11].

Biochar has an enormous potential to permanently capture and store carbon in the long-term, since it contains high fractions of fixed and recalcitrant carbon [12]. It has been already recognized by scientific community as one of the most promising options to mitigate climate change [13]. Recent studies [14] demonstrated that biochar have similar chemical structure to fossil-coal, which indicates by analogy that would represent the most stable form of carbon that can be found at geological level. Through the study of the "random reflectance" of biochar, which indicates the degree of aromatization of its organic carbon structures, and a benchmark with the one measured in fossil coal, it has been possible to assess a similarity to inertinite [15]. However, this should be confirmed by investigating the response of biochar subjected to abiotic oxidation in soil. According to Woolf et al (2021) [16], molar hydrogen to organic carbon ratio (H/C_{org}) is the indicator determining the biocarbon permanence into soil, since a direct indication about the degree of aromatization of the chemical structure of biochar. By means of the experimental measurement on recalcitrant (or fixed) carbon present in biochar, it is possible to roughly estimate the carbon permanence into soil [17,18]. When biochar is properly produced using slow pyrolysis at slow heating rates, longer residence time and temperature (typically between 500 – 700 °C), experimental trials show carbon contents ranging between 80 and 90 % [19], of which the 70 – 90 % is recalcitrant [12]. The combination of such experimental methodologies can help to determine the biochar' permanence factor into soil.

Therefore, by recognizing the dual role of biochar as both soil enhancer and potential carbon sink, new European policies have been recently developed. For instance, biochar has been recently approved for its use as fertilizer in the updated EU rules on fertilising products (Regulation 2019/1009) [20] through the addition of a category for pyrolysis and gasification materials (Commission Delegated Regulation 2021/2088 [21]). As soil carbon enhancer, biochar can generate emission savings from soil carbon accumulation via improved agricultural management (e_{sca}), according to the rules set in the Commission Implementing Regulation (EU) 2022/996 [22] for the Renewable Energy Directive (EU) 2018/2001 [23] (RED II). However, such emissions savings can be claimed only if biochar is used in the lands cultivated for biofuels production. According to the RED II GHG emissions calculation methodology for biofuels and bioenergy [23], there are no other options to claim a GHG emissions credits producing and labelling biochar as a BECCS solution, since the emissions for carbon capture and storage (e_{ccs}) shall refer to CO_2 captured and stored according to the Directive 2009/31/EC [24].

Under the different viewpoint of Carbon Dioxide Removal (CDR), the Innovation Fund methodology for calculating the GHG emissions of large-scale demonstration projects of innovative, low-carbon technologies [25] provides an emission credit for the medium-term carbon storage of biochar (co-produced within the main process), calculated as 50 % of its biogenic carbon content. For long-term storage of biochar into the soil, further assessment is needed in view of future implementations as bio-carbon removal solutions according to the Directive 2009/31/EC on the geological storage of carbon dioxide [24].

Given to the enormous potential of biochar for carbon storage [1], together with the need for sustainable biofuels ensuring low carbon emissions, this paper investigates the potential GHG emission mitigation and biogenic carbon sequestration through biochar co-produced within lignocellulosic biofuels value chains. The novelty of this research stems from examining a synergistic approach that generates both biochar and drop-in fuels: a concept originally developed from the findings of an EU-funded project [26]: specifically, it investigates the possible integration between fast and slow pyrolysis technologies within stand-alone biofuels value chains [27]. The innovative contribution of this article relates to the use of slow pyrolysis to generate bioheat for biomass drying and at the same time to produce biochar for CDR and carbon sequestration. Conversely, fast pyrolysis serves as a pivotal method for densifying biomass and converting it into a liquid form, facilitating transportation to a centralized biorefinery for drop-in biofuels production.

Process integration and optimisation in biomass drying, biofuel, bioenergy and biochar production are studied: mass and energy balances for these processes are undertaken to estimate potential performances in respect to energy efficiency and GHG emissions reduction. The LCA approach used in this study has been developed on the bases of the JECv5 Well-To-Tank (WTT) [28] and RED II requirements, so to quantify the carbon emissions and energy performances of the processes: this is in line with the current EU requirements to produce sustainable biofuels. A supplementary LCI, including the most recent data as regards the innovative technology proposed package within the case studies, is reported. Methodological choices are discussed considering different inputs/outputs such as: initial moisture content of biomass, liquid or gaseous fossil-based fuels supply, H_2 supply, low-carbon electricity, bioenergy and biochar production. The aim is to show to what extent the relative GHG saving potential, attributed to the various outputs, changes with methodological choices, mainly when attributing emissions and credits. For this scope, a specific methodology to estimate the CO_2e credit for co-producing biochar and then sequestering carbon, is proposed. Finally, a first assessment of the carbon intensity of the advanced biofuels is carried out, attributing carbon storage potential to biofuels and considering the maximum amount of co-produced biochar that could be potentially traded to the voluntary carbon markets.

2. Processes and energy data input

Within this study, two main case studies have been selected for advanced drop-in fuels production, based on the pathways available in the JECv5 [28]. The first case study involves the conversion of lignocellulosic feedstock into a drop-in fuel (renewable diesel) through fast pyrolysis and hydrotreatment (WFPD1) while the second one involves gasification, syngas upgrading and Fischer-Tropsch synthesis (WFSD1), as shown in Fig. 1.

Biomass-based renewable diesel pathways can therefore be produced also feeding waste wood (which is included as well in JECv5). If the wood is mostly recovered as residue from sawmills, this is already dried and in the form of sawdust, so pre-treatment before biofuels conversion is not needed. In the present work, the type of lignocellulosic biomass selected for biofuel production is coppice wood deriving from sustainable wood managed in line with RED sustainability criteria. Being fresh

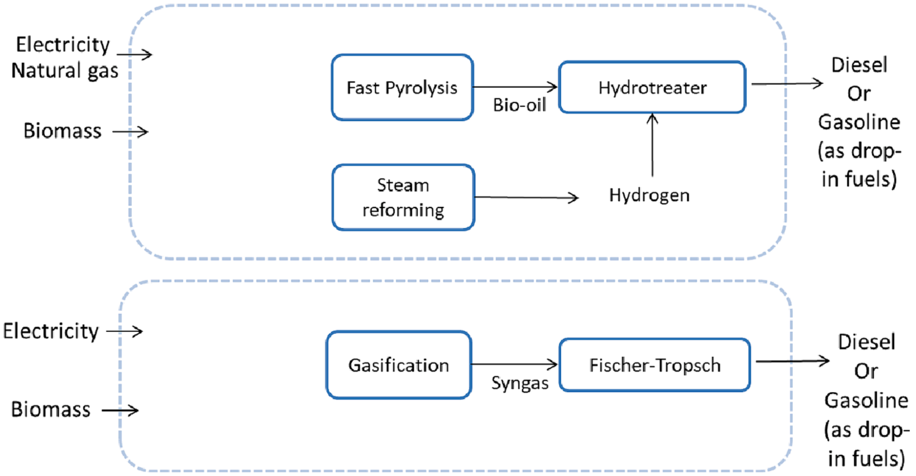

Fig. 1. System boundaries for LCA modelling.

biomass, pre-treatment for thermochemical processes becomes extremely relevant for the overall sustainability. Therefore, this work aims at investigating new technology options on these pathways (described in detail in the next section), using recent figures on conversion processes from industrial trials and biomass moisture content representing real field conditions at harvesting and during the following conversion steps. This leads to a new plant outline suitable for large-scale projects, and considering the actual biomass' water content based on the geographic location and the practical issues related to handling large volumes of biomass supply. This stems on the fact that a facility with a capacity of 100 MW would require to transport several hundred thousand tons of biomass per year at the thermochemical conversion site, requiring complex logistics including drying, transport and storage. Therefore, biomass energy densification into bio-intermediate energy carries, closer to the harvesting sites, facilitates storage and transport operations and improves the overall sustainability [29,30]. It is worth also noting that the use of energy crops would generate the same scenario (as studied in BECOOL project [26]).

2.1. Case studies

A calculation model has been developed within this study to investigate combined advanced biofuels and biochar production, integrating biomass pre-treatment, fast and slow pyrolysis as first conversion step with liquid biofuels upgrading as second conversion step. The new plant outline considers a combined pyrolysis plants' layout, integrating a slow pyrolysis unit into the fast pyrolysis facility, aimed at improving the use of energy within the system boundary: the slow pyrolysis unit is properly sized to cover the energy demand needed to dry the initial biomass for fast pyrolysis requirements, leading also to biochar production. The Fast Pyrolysis Bio-Oil (FPBO) plant is assumed having a design capacity of 40,000 tons per year biomass input (dry) [31]: this is today the size of one of the few commercially operated facilities fed by wood residues from the wood industry (e.g. sawdust, coming with very low moisture content). The new plant configuration assumes lignocellulosic biomass as feedstock, which would require the use of comminution and drying, being powered by diesel or natural gas as mandatory pre-treatment for the thermochemical processes. The innovation proposed within this work is to replace such fossil-based drying with additional biomass coming from the same location, so to supply additional energy for biomass drying and at the same time generate a biogenic carbon product (biochar). Therefore, the new model expands the system boundaries of the original JECv5 considered pathways, also integrating the use of electricity and hydrogen from other systems.

Case Study 1 (CS1, Fig. 3) proposes a new route based on the pyrolysis-based diesel pathway (WFPD1 in JECv5) combined with biochar production. This entails the production of Fast Pyrolysis Bio Oil (FPBO) in a commercial plant combined with slow pyrolysis providing the necessary heat to dry biomass and produce biochar. FPBO is the intermediate bioenergy carrier that undergoes toward further upgrading and hydrotreatment to produce drop-in fuels.

Case Study 2 (CS2, Fig. 3) proposes a second route based on the renewable diesel production pathway through gasification and Fischer-Tropsch (WFSD1 in JECv5) combined with biochar production. Fast and slow pyrolysis have the same dimension as in CS1, but here a slurry (made of bio-oil and biochar from fast pyrolysis only) is the intermediate bioenergy carrier, that goes to the gasification and Fischer-Tropsch synthesis units.

Both case studies allow to decentralize the primary conversion step, where biomass is converted into bio-intermediate energy carriers, which are then transported to a centralized liquid biofuel conversion site of 100 MW capacity. Decentralised biomass energy densification offers multiple advantages in terms of logistics, reduced transport costs [32,33] and optimized conversion steps in biofuel production [34]. When energy densification is based on thermochemical processing, higher energy densification and therefore reduced transport costs can be achieved compared to mechanical densification (such as pelletising), as studied within the EU Horizon 2020 BECOOL project [26] by some authors of this study [29]. For instance, as calculated in Boymans et al (2019) [29], the energy density of slurries composed by FPBO and biochar is about five times higher than raw biomass.

2.2. Biomass pre-treatment

The initial biomass moisture content differs significantly among different bio-feedstocks [35] and depending on different storage practices and locations for the same biomass type [36]. JECv5 assumes that thermochemical processes converting biomass into biofuels are operated with woody biomass at 30 % moisture content (as mass fraction) at the inlet. No drying steps have been considered since it is assumed that biomass is naturally dried (i.e. wood seasoning). However, as explained in section 2.3, thermochemical processes need further biomass drying to perform at the highest conversion yields, therefore additional drying is needed.

In EU, and in particular in the EU MED Southern countries, biomass moisture can be reduced below 20 % after 60 days natural drying in summer conditions [37], while for Northern EU countries this figure cannot be lower than 40 % even after 10 months and considering the best storage techniques [36]. Moreover, considering that biomass supply is expected to happen all year long, the use of artificial drying for the designed value chains is inevitable. For this study, biomass naturally dried up to 30 % moisture content at the pyrolysis plant inlet has been considered for the baseline scenario (also to benchmark data with JECv5), and at 50 % for the alternative scenario (e.g. assuming biofuels plants located in EU Northern countries). The study of Havlík and Dlouhý (2020) [38] has been considered to estimate the specific energy consumption of indirect dryers for biomass, where the average drying energy demand is about 2.8 MJ per kg of water evaporated.

For biomass chipping, data from JECv5 have been used, but in the case of fast pyrolysis, further biomass reduction is required (milling). The calculations assume 223 kJ per kg (as electricity) for milling wood up to 1.5 mm screen size at 11.3 % moisture content, as suggested by Liu et al (2016) [39].

2.3. *Thermochemical processes and assumptions*

2.3.1. *Slow pyrolysis*

Biomass slow pyrolysis (or carbonization) is a thermochemical conversion process based on low heating rates that maximises the production of biochar, which comes together with pyrogas [40]. There are many fully commercial technology options to produce biochar [41], operated in both continuous and batch modes. This process has as energy outputs about equal parts of biochar and bioenergy through pyrogas combustion [19]. Having a high exergy content, this energy flow can serve multiple uses, such as industrial application, district heating and/or biomass drying.

For this study, rotary kiln pyrolysis reactors have been considered for their flexibility on feedstock type and characteristics, following similar other studies on bio-coal supply for steel making process [42,43]. Plant size can be adjusted to integrate this technology into an existing liquid biofuel supply chain (from JECv5, as described in 2.1) to cover the bioheat demand necessary to dry the raw biomass. Input data for this process have been sourced from recent studies [42,44] as well as sizing criteria and process assumptions, then adapted to the system studied within this work. Input data for slow pyrolysis are reported in Table 1.

2.3.2. *Fast pyrolysis*

Differently from slow pyrolysis, the fast pyrolysis process targets the maximisation of the liquid biocrude phase production at the expense of the production of incondensable gases and biochar [8]. The Fast Pyrolysis Bio-Oil (FPBO) can then be upgraded to liquid biofuels through hydrotreatment, necessitating higher hydrogen addition than the traditional Hydrotreating of Vegetable Oils (HVO) process since the bio-oil contains about 45–50 % of oxygen (as mass fraction, [45]) compared to lipids, which have around 12 % oxygen content [46]. Fast pyrolysis requires also very dry biomass (up to 3 % moisture content at the reactor inlet) and low ash content to perform well [47]. For this reason, as of today EU commercial plants are using residues from pellets or residual biomass from sawmills (i.e. dry sawdust) as feedstock.

2.3.3. *Hydrotreating of fast pyrolysis oil*

In this study, JECv5 WFPD1 (pyrolysis-based diesel) has been selected as baseline conversion pathway to renewable diesel (as Hydroprocessed Pyrolysis Oil – HPO). JECv5 model used data sourced from a modelling elaboration [48], which assumed data from a pilot scale fluidized bed reactor operated by VTT in 2012 [49] for fast pyrolysis and FPBO hydroprocessing (details of the process are provided in other works [50–53]). For this study, a commercial fast pyrolysis plant based on a rotating cone-reactor is selected as reference bio-intermediate conversion step, sourcing data from recent H2020 projects studying the LCA of the new commercial plants [44,54,55]. For hydrotreatment of FPBO, the process BTG-Next [56] has been considered as bio-oil upgrading step, and process data have been sourced from recent Horizon 2020 project reports [44] and peer-reviewed papers [57]. Input data for fast pyrolysis are reported in Table 1.

2.3.4. *Gasification and Fischer-Tropsch*

Biomass gasification converts woody feedstock to a gas, which can be cleaned and upgraded to syngas for further FT-synthesis, if the target product is a liquid fuel [58]. JECv5 WFSD1 is based on the study by Tijmensen et al, 2002 [59] and Hamelink and Faaji, 2002 [60], where syngas comes from the Battelle Columbus Laboratory (BCL) gasifier (atmospheric indirectly fired gasifier, 200 MWth size modelled from pilot-scale data). However, this study assumes a decentralized scheme for biomass pre-treatment and conversion to bio-intermediate energy carriers: therefore, the type of gasification technology must be revised in the present work. Specifically, the use of a slurry composed by FPBO and biochar requires (as in the present study) a pressurized Entrained Flow Gasifier (EFG) technology, as suggested in literature [61,62]. This technology leads to a clean syngas that can be coupled with Fisher-Tropsch (FT)-process to liquid fuels production [63]. For this modelling exercise, the study proposed by Leibbrand et al, 2013 [64], case study EG1, has been used as data source. This study considers the combined production of fuels and electricity from a dedicated steam turbine (powered by the heat surplus at the outlet of the conversion process). Differently from FPBO hydrotreating, this process does not require additional hydrogen for syngas upgrading to drop-in diesel.

3. Energy and GHG emissions accounting methodology

3.1. *Energy accounting*

The methodology for the energy assessment of biofuels value chains co-producing biochar is the same used within JECv5 WTT [28]. Both mass and energy flows are accounted for and evaluated for the whole value chain, enabling a comprehensive analysis of the whole energy conversion process. The calculation model is developed by first

Table 1

Data for thermochemical processes considered in the LCA study.

Parameter	Unit	Slow Pyrolysis	Fast Pyrolysis	Upgrading through hydroprocessing	Upgrading to gasification and Fischer-Tropsch
Feedstock	–	Biomass, < 30 mm	Biomass, < 3 mm	FPBO	FPBO + char
Moisture content at the inlet	% mass	10	3	–	–
Conversion to main product	–	Biochar	FPBO	HPO	FT-liquids
	% energy	43	67	75⁑	35.5
LHV main product	MJ per kg	28	16.5	43.7	43.9
Electricity demand	MJ per kg of product	0.61	1.53	0.83	−8.65†
Hydrogen demand	MJ per kg of product	–	–	6	–
Thermal energy demand	% energy*	10	18	‡	36.8

*Fraction of chemical energy absorbed from the input to run the process; ⁑including hydrogen; † generated; ‡exothermic process but powered by hydrogen.

considering the real size of commercial biofuel plants (fast pyrolysis and hydrotreatment/FT-plants), and then then the sizing of the slow pyrolysis unit and consequently the total number of plants is iterative depending on the initial biomass moisture content, to achieve a net-zero bioheat balance. The energy conversion is evaluated as the ratio between the overall system energy output and the energy inputs, incorporating both fossil- and bio-based sources. This parameter allows for benchmarking the results with other studies (in this work with JECv5 pathways) and evaluate the effectiveness of the considered value chains.

3.2. LCA approach and methodological options

The GHG emissions accounting in this paper follows the Well–To-Tank (WTT) approach according to the methodology presented in the JECv5 WTT [28]. Compared to a comprehensive environmental Life Cycle Assessment (LCA) approach studying the Global Warming Potential (GWP) of fuels, WTT can be described as a simplified analysis studying energy and GHG emissions accounting. In the WTT approach, emissions related to the hardware construction, maintenance and decommissioning of fuel and electricity producing facilities, including materials cycles, are usually not considered. The present study - investigates the co-production of biochar from fast and slow pyrolysis processes, evaluating two different allocation criteria for co-products: the energy allocation according to the RED II guidelines, where emissions are allocated between the main product (fuel) and co-products (e.g. biochar) according to their energy content (as Low Heating Value – LHV), and an emission credit attributed to biochar as a nature-based solution for carbon sink. This last option differs from the most common BECCS technologies since biocarbon is captured and geologically stored in its solid form, and not as compressed or liquid CO_2 as for the other BECCS technologies.

As regards the possibility to use biochar in soil as nature-based carbon sequestration solution, IPCC suggested that the stable carbon fraction of the biochar can be considered a durable form of carbon storage [65]. Therefore, the present work considers the following equation to calculate the emissions to produce liquid, drop-in biofuel (e_{fuel}) per energy unit, that includes an emission credit defining the carbon storage potential of biochar:

$$e_{fuel} = e_{fe\_p} + e_{fe\_t} + e_{fef\_p} + e_{bcr\_p} + e_{fu\_t} - C_{bc} \tag{1}$$

$\mathbf{e_{fe\_p}}$: emissions from feedstock production and cultivation;

$\mathbf{e_{fe\_t}}$: emissions from feedstock transport up to the conversion facility (combined pyrolysis plant);

$\mathbf{e_{fef\_p}}$: emissions from processing feedstock to bio-intermediates (pyrolysis oil and biochar);

$\mathbf{e_{bcr\_p}}$: emissions from processing bio-intermediates to commercial grade fuels (biofuels conversion plants);

$\mathbf{e_{fu\_t}}$: emissions from fuel transportation and distribution;

$\mathbf{C_{bc}}$: net credit for carbon storage into biochar (see eq.2).

All emissions sources are calculated as CO_2 equivalent emissions referring to the energy unit of fuel (MJ). The definition of C_{bc} acknowledges the carbon dioxide (as GHG) captured into biochar in a durable form. By considering the carbon sequestration potential of biochar within this LCA systems, an emission credit is assigned to the carbon intensity of the biofuel. As for other BECCS technologies, a GHG emissions credit refers to a unit of measurement that represents a direct reduction of CO_2 equivalent emissions [66]. The methodology to determine the carbon dioxide permanently captured into biochar is described in the next section. This credit can be used in various ways, such as offsetting emissions from other sources or being traded on carbon markets (hence outside the system boundaries of the present LCA model, as depicted in Fig. 2) with the requirement to avoid double counting of GHG emissions.

3.3. Estimation of an emission credit for biochar

In order to estimate the long-term stability to consider biochar as nature-based carbon sink, the temporal dimension of carbon storage needs to be addressed: only the durable carbon sequestration should be considered [66–68]. Clearly, experiments on carbon sequestration can only address short-term verifications: this happens with biochar too, where for experimental investigation on biochar use in soil usually refers to time-frame around 10 years [69]. A recently proposed new method to prove the recalcitrant and durable nature of biochar permanence into soil is the Random Reflectance (RR) analysis, which compare the biochar characteristics with inertinite, among the most stable forms of carbon in the geological crust. As observed in recent geological studies on biochar properties [14,15], a similarity with inertinite has been observed by measuring the RR of many biochar samples: this measurement expresses the degree of aromatization of the organic carbon structures.

By assuming the Hydrogen-to-Carbon (H/C) ratio as the indicator to determine the carbon structure, this work assumes the methodology proposed by Woolf et al (2021) [16] for calculating the biochar permanence into soil through a correlation based on the molar H/C_{org} ratio, which estimates the factor F_{perm} (eq. (2). In order to determine carbon and hydrogen fractions, properties of slow pyrolysis biochar has been assumed from literature [42], i.e. 0.35 as H/C_{org} and pyrolysis temperature of 550 °C. Hence the resulting fraction of biocarbon remaining in soil after 100 years is estimated at 82 % (i.e. 65 % of the mass of biochar). This figure can be even higher, according to some recent findings proposed by Rodrigues et al, 2023 [70], but for simplicity and to keep a precautionary approach, this study assumes the value previously reported. Therefore, the net credit for carbon storage into biochar (C_{bc}) is calculated as follows:

$$C_{bc} = F_{perm} {}^{*} e_{bc\_u} - e_{bc\_p} - e_{bc\_t} \tag{2}$$

$\mathbf{F_{perm}}$: Fraction of biochar carbon remaining in soil after 100 years as defined by Woolf et al (2021) [16]

$\mathbf{e_{bc\_u}}$: CO_2 that can be potentially generated by the full oxidation of

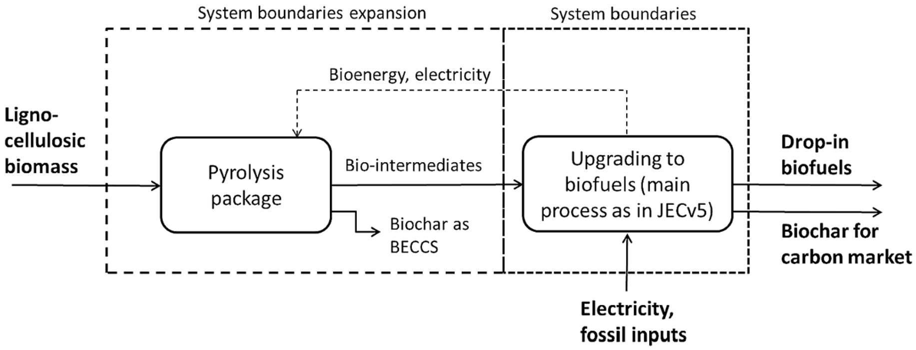

Fig. 2. Concept scheme integrating biomass slow pyrolysis within JECv5 for GHG emission assessment of liquid biofuel pathways.

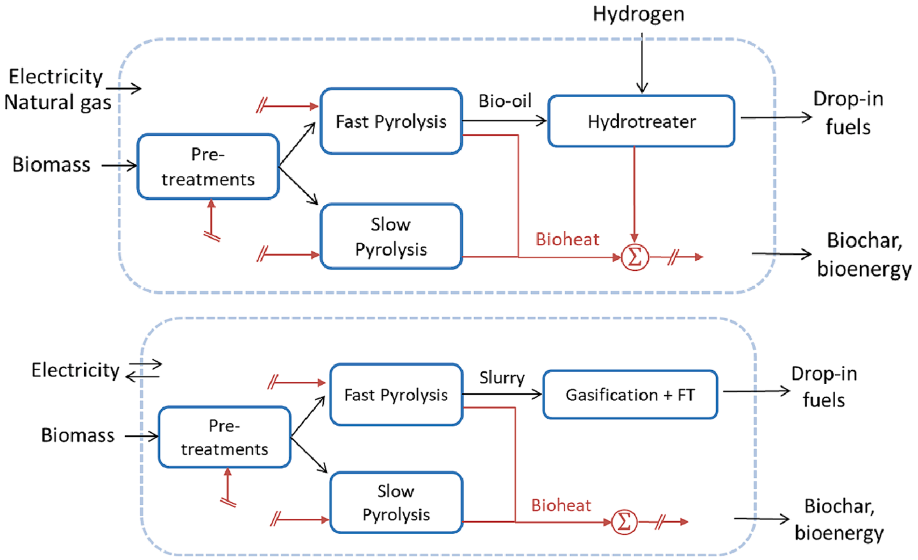

Fig. 3. CS1 – pyrolysis-based diesel pathway combined with biochar production (top); CS2 – renewable diesel through gasification and FT combined with biochar production (bottom).

biochar;

$\mathbf{e_{bc\_p}}$: emissions allocated from biochar production;

$\mathbf{e_{bc\_t}}$: emissions from biochar transportation and intake to the geological site;

The term e_{bc_u} is calculated by considering the carbon content of biochar multiplied by the molecular mass ratio C:CO_2, which is 3.67. Emissions from biochar production are calculated by using the energy allocation criteria described in section 3.1. Emissions from transportation and intake to the storage site (e_{bc_t}) are set to zero since this work assumes that biochar can be stored in the nearby area to the production site (for example in former coal mines, available almost everywhere, as proposed by Dufour, 2013 [71]).

3.4. *Life Cycle Inventory*

In conducting the life cycle inventory for thermochemical processes considered within this study, a specific attention is dedicated to reporting comprehensive input data. The present approach encompassed a rigorous collection of information on feedstock properties, energy inputs, process emissions, and technological specifications explained in section 2.3. Detailed information on energy, electricity, heat, and auxiliary materials, are documented in Table 1 to ascertain the requirements of the thermochemical processes. For the biomass cultivation, transport and biofuels distribution (e_p, e_{tc}, and e_{td} respectively, Equation (1) this study uses the data given in JECv5 for the pathways WFPD1 and WFSD1 [28].

As regards emissions for transport of biomass, this study considers 100 km radius supply area for biomass around each pyrolysis plant (as assumed in BECOOL' project [27]), and 200 km distance from the fuel conversion plant.

Electricity carbon intensities have been sourced from JECv5 [28], assuming the values of the EU Mix (2016). Hydrogen carbon intensities have been sourced from JECv5 through steam methane reforming (GMCH1 pathway). In order to show the impact of the carbon intensity of electricity and hydrogen within EU scenario [72], this study considers the use of hydrogen from electrolysis and electricity with different carbon intensities. Carbon intensities of renewables as PV and wind have been sourced by Scarlat et al, 2022 [73]. Hydrogen carbon intensity from electrolysis is determined by multiplying the energy requirement for H_2 production (assumed as 1.538 MJ electricity for 1 MJ H_2 produced, equivalent to an efficiency of 65 % for electrolysis) and carbon intensity of the electricity used.

4. Results

4.1. *Impact of biomass at 30 % moisture content*

The impact of biochar production within the liquid biofuels has been evaluated for both case studies, assuming biomass is supplied at 30 % moisture content (as in JECv5 study) at the bio-intermediates production site. Energy flows are shown in Fig. 4, which reports the inputs on the left side, and the outputs on the right side. On the horizontal axis of each chart, the effect of different share of biomass supply between the fast pyrolysis plant and the auxiliary slow pyrolysis plant is shown as follows: on the left side, all biomass supply is used for slow pyrolysis; on the right side, all biomass supply is used for fast pyrolysis. The energy requirements of natural gas are considered for start-up operations of plants and biomass drying (when needed), while diesel input is used for vehicles collecting and transporting biomass, bio-intermediate and fuels, and electricity is needed to run the plants.

By increasing the share of biomass fed to the fast pyrolysis plant, the amount of biomass available for the slow pyrolysis plant reduces, and thus the SP plant size, lowering the production of bioheat and biochar at the bio-intermediate conversion site (as seen on the charts on the right side). In this case, fast pyrolysis oil production increases, as well as fuel and heat (as outlet at the fuel conversion facility). Energy requirements such as diesel, natural gas and electricity, increase as well as the fuel production. Moreover, CS1 requires a growing hydrogen demand when targeting the production of pyrolysis oil-based drop-in fuels. CS2 leads also to electricity production due to the high-temperature heat surplus at the Fischer-Tropsch reactor, which is re-used inside the system to cover the internal electricity demand: this effect is therefore reflected in a reduced net electricity demand on the left (inputs) side rather than as an output on the right side.. The biochar at the pyrolysis plants gate consists of both biochar derived from fast pyrolysis and biochar derived from slow pyrolysis. In the CS2, it is assumed that the Fischer-Tropsch reactor

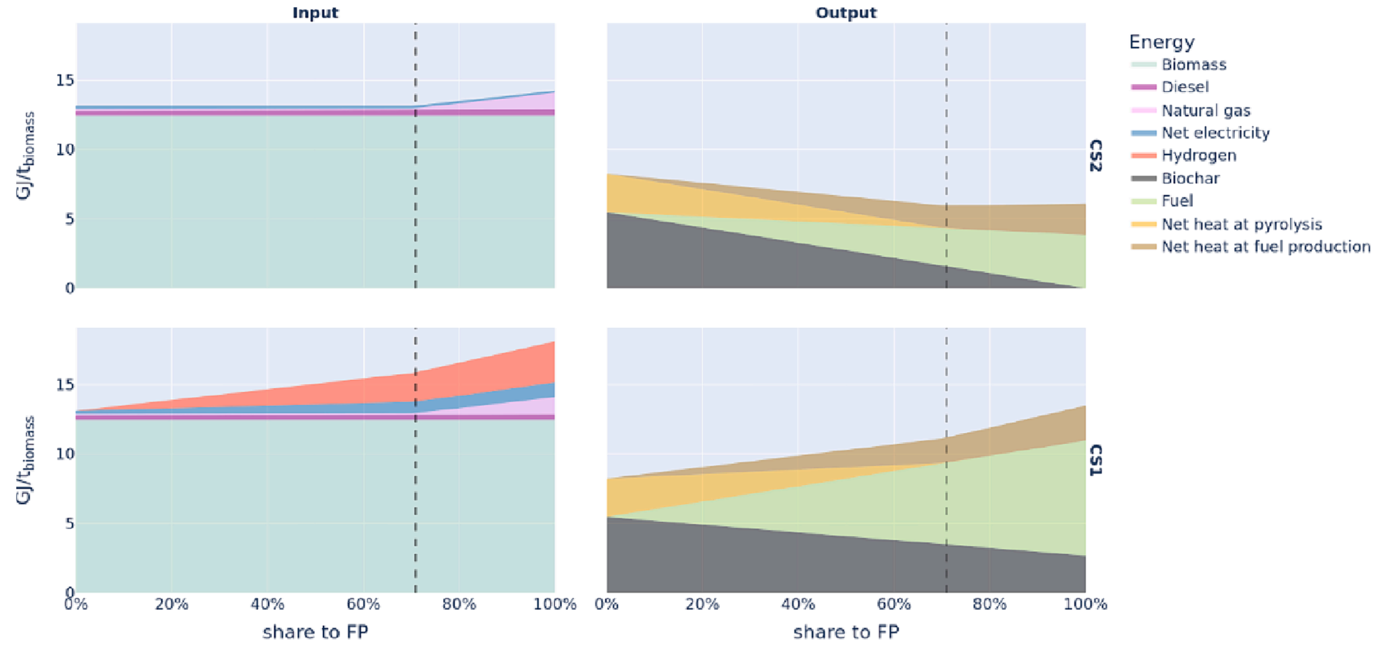

Fig. 4. Energy inlet and outlet varying the share of biomass feeding to the fast pyrolysis unit (assuming 30% biomass moisture content).

is supplied with a slurry containing the entire output of the fast pyrolysis plant, which includes its derived biochar. This is why, when 100 % of the biomass is allocated to the fast pyrolysis plant, no biochar is available for CS2, while the biochar from fast pyrolysis remains available to be combined with biochar from slow pyrolysis in the CS1.

This chart also shows how the biomass input has been tuned to optimize the heat surplus at the bio-intermediate conversion facility: when net heat surplus generated by the pyrolysis plants reduces to zero, slow pyrolysis provides enough heat for covering the heat demand for biomass drying necessary for feeding the fast pyrolysis plant (down to 3 % moisture content for fast pyrolysis). This happens at 70.9 % of biomass feeding to fast pyrolysis, and above this point the demand for natural gas to dry the biomass increases with a steeper slope. At this level the size of the slow pyrolysis plant is estimated at approximately 10 MW, producing 4,200 tons per year of biochar. Therefore, multiple units of combined slow and fast pyrolysis plants are needed to build up the innovative configuration proposed. The results of this sizing exercise are reported in the Table 2, which recaps the main data for drop-in fuel and biochar production, reporting all relevant inputs and outputs on yearly mass and energy basis.

The model calculated that five decentralized units of combined pyrolysis plants are required to supply about 100 MW of bio-intermediates (as hypothesized in Chapter 2) to the centralized biofuel conversion plant. This is valid for both case studies assuming the dimension of the slow pyrolysis unit as calculated from this sizing exercise (since the fast pyrolysis plant is fixed). Overall, the energy conversion rate of this pathway is 31.7 % for fuel and 50.6 % including biochar in the CS1, compared to 44.4 % in the scenario proposed within JECv5, which considers only fuel production at the outlet. Differently, for CS2 this parameter drops to 17.3 % for fuel (27.4 % including biochar) compared to 42.9 % in JECv5, which is much higher due to different data inputs assumed to model the conversion efficiency at the FT-plant (i.e. 45.1 % as wood-to-fuel ratio, compared to 35.5 % as slurry-to-fuel ratio, which is the assumption done in this work [64]). However, the improved scenario enables a considerable amount of energy conversion to biochar and the reduction of about 203,400 GJ per year of natural gas consumptions, which would have been needed to dry the biomass for the fast pyrolysis plant requirements. This improved scenario also allows a reduced fossil-diesel demand of about 40 % compared to JECv5 for both case studies due to the biomass energy densification in bio-intermediates performed in the decentralized plants.

Table 2
Overall mass and energy flows for the proposed case studies at 30% biomass moisture content for the optimal sizing scenario.

		CS1	CS2
Nr. combined pyrolysis plants	–	5	
Biomass supply (30 % m.c.) to each combined pyrolysis plant	t/y	73,165	
	GJ/y	1,098,360	
Nr. combined pyrolysis plants	–	5	
Biomass input (30 % m.c.)	GJ/y	5,491,798	
Diesel input	GJ/y	184,911	161,812
Electricity input	GJ/y	299,600	62,428
Natural gas input	GJ/y	39,450	
Hydrogen input	GJ/y	766,500	–
Net heat surplus at fuel production plant	GJ/y	651,005	586,696
Electricity production	GJ/y	–	196,416
Fuel production	GJ/y	2,151,036	996,108
	t/y	49,223	22,690
Biochar production	t/y	48,217	20,842

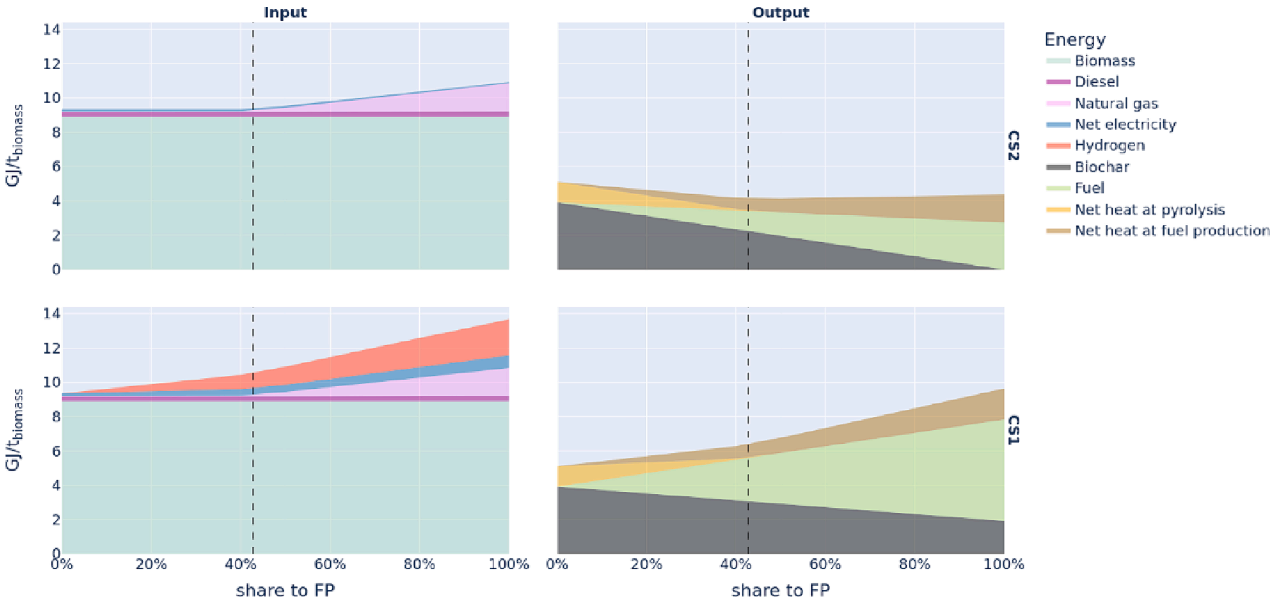

Fig. 5. Energy inlet and outlet from the process varying the share of biomass input into fast pyrolysis at 50% biomass moisture content.

Table 3
Overall mass and energy flows for the proposed case studies at 50% biomass moisture content for the optimal sizing scenario.

		CS1	CS2
Biomass supply (50 % m.c.) to each combined pyrolysis plant	t/y	169,716	
	GJ/y	1,819,843	
Nr. combined pyrolysis plants	–	5	
Biomass input (50 % m.c.)	GJ/y	9,099,217	
Diesel input	GJ/y	280,755	257,656
Electricity input	GJ/y	328,395	91,223
Natural gas input	GJ/y	42,300	
Hydrogen input	GJ/y	766,500	–
Net heat surplus at fuel production plant	GJ/y	651,005	586,696
Electricity production	GJ/y	–	196,416
Fuel production	GJ/y	2,151,036	996,108
	t/y	49,223	22,690
Biochar production	t/y	95,267	67,892

4.2. *Impact of biomass at 50 % moisture content*

When biomass comes at 50 % moisture content, additional bioheat is needed for drying. Therefore, a larger share of wood is needed for slow pyrolysis to cover the additional energy demand for drying, and consequently more biochar is produced in comparison with the scenario when biomass has lower moisture content.

Utilizing the same sizing criteria presented in the previous section, the optimal share of biomass to fast pyrolysis section now drops down to 42.8 % (as shown in Fig. 5). All other considerations considered for the previous section remain applicable to this scenario.

However, it is noteworthy that to maintain the same fast pyrolysis plant size, this model proportionally increased the capacity of the slow pyrolysis plant up over 30 MW, i.e. above 14,000 tons per year of biochar. Therefore, to maintain the same reactor size for the slow pyrolysis unit as in the previous scenario, the present layout assumes the use of three units working simultaneously for the same fast pyrolysis plant. Summarizing, Table 3 recaps the main results from this updated sizing exercise, maintaining the same parameters evaluated in Table 2.

The considerations done in the previous section as regards the number of plants and the energy efficiency of the supply chain remain still valid. However, the energy conversion rate is much lower than the previous scenario, i.e. 45.2 % for CS1 and 30.4 % for CS2 (including biochar), with an incrementing biochar production of almost 50,000 tons per year for each case study, given the higher (but often very realistic) moisture content of biomass feeding to the biorefinery. Nevertheless, this configuration allows for a substantial reduction in

Table 4
Model results for case studies at different levels of biomass moisture content.

Moisture content of initial biomass		30 %		50 %	
		CS1	CS2	CS1	CS2
Liquid biofuels (energy allocation)	g CO$_2$eq/ MJ$_{fuel}$	47.1	29.6	42.3	30.2
Biochar (energy allocation)	g CO2eq/ MJ$_{biochar}$	3.5	7.2	6.0	8.1
Liquid biofuels (including biochar GHG credit)	g CO$_2$eq/ MJ$_{fuel}$	−4.2	−20.2	−56.1	−132.3
Share of biochar for carbon market*	%	70	100	84	100
Carbon intensity of biochar for carbon market (C_{bc}**)	g CO$_2$eq/ MJ$_{biochar}$	−78.4	−70.8	−79.3	−69

*to keep the carbon intensity of biofuels within the minimum GHG emissions requirements (i.e. above 65% GHG emissions savings for advanced biofuels).
** referring to Equation (2).
Note: negative emissions mean that removal or reduction of GHG emissions is performed, effectively creating a carbon sequestration.

natural gas demand (i.e. 484,000 GJ per year) if the scenario proposed in the JECv5 study would have been fed by biomass at 50 % moisture content at the inlet.

4.3. Carbon intensity of fuels and biochar credit

The GHG emissions assessment for both case studies, based on the conditions outlined in the previous paragraphs, has been summarized in Table 4 (details of calculations are reported in the supplementary material). This table first attributes emissions to biofuels and biochar based on their LHV (energy allocation criteria) for each case study according to the RED guidelines. It then calculates the carbon intensity of the fuel if all biochar is used as a durable carbon storage product within the biofuel supply chain. It is important to note that FT-process generates heat excess, therefore at the centralized biofuel conversion plant there is additional bioenergy production (referred to as “net heat surplus at fuel production plant”, reported in both Table 2 and Table 3), which is a co-product that can be utilized for other purposes. Therefore, the energy allocation criteria, along with the application of the Carnot factor for bioheat, have been applied.

The calculation of the GHG emissions of liquid biofuels show that CS2 lead to carbon intensities lower than the RED II’ threshold for advanced biofuels set at 32.9 $gCO_2e\ MJ^{-1}$ (corresponding to 65 % GHG emissions reduction compared to the Fossil Fuel Comparator of 94 $gCO_2e\ MJ^{-1}$). This allows for two possible options: selling all the biochar to carbon markets and generating, or achieving further greenhouse gas reductions within the biofuel production system and gaining benefits to support the achievement of emission reduction targets for the transport sector set by RED III. As regards the first option, biochar may be potentially valorised as BECCS, being a carbon negative vector s (CO2 removed from the atmosphere) as presented in Table 4.

Differently, CS1 generated carbon intensities of liquid biofuels higher than the RED II’ threshold for both scenarios at different biomass moisture levels. In this case, the options consist in: selling only part of the biochar to carbon markets and maintaining the minimum part within the biofuels system to generate sufficient negative emissions to achieve the maximum allowed carbon intensity set at 32.9 $gCO_2e\ MJ^{-1}$; or maintaining the whole biochar credit within the biofuels system to generate the maximum GHG emissions reduction. Table 4 is reporting the results for each of these possibilities, including the share of biochar that can be potentially traded for the voluntary carbon markets.

Finally, comparing these results with their JECv5 counterparts (26.65 and 14.04 $gCO_2e\ MJ^{-1}$ for WFPD1 and WFSD1 respectively, standard scenario at 30 % biomass moisture content), this study shows larger GHG emissions saving generated by improved scenarios calculated within CS1 and CS2 (when considering the biochar GHG credit), which allow a further emissions reduction of 34.3 $gCO_2e\ MJ^{-1}$ for pyrolysis based-fuels and 30.9 $gCO_2e\ MJ^{-1}$ for Fischer-Tropsch based-fuels. For an integrated biorefinery (as in this study, 100 MW capacity) this means to deliver additional savings of 13.3 and 6.8 ktons of CO_2e per year, respectively for CS1 and CS2.

4.4. Improved scenario with increased renewable energy use

This section shows additional opportunities for further reducing GHG emissions generated from the production of liquid biofuels through the new proposed process configurations. By using electricity and hydrogen (differentiating from that produced by SMR, as assumed in JECv5) produced from renewable sources or simply greener electricity grid mix, new scenarios are outlined with the aim of studying the carbon intensity of the biofuels produced by greening fossil-based inputs. Considering the LCA methodological framework, both the carbon intensity of electricity (i.e. considered as a marginal input) and the selected GHG emissions allocation methodology are parameters that strongly impact on the results. Therefore, an impact assessment of such parameters is provided as graphical elaboration, aiming at providing an accurate understanding of how these variables may affect the overall results. Fig. 6 shows the variation of the carbon intensity of liquid biofuels from CS1 depending on the carbon intensity of the electricity, ranging from 0 to values corresponding to grid mix well above the European average (which lacks energy generated from renewable sources). In the graph, the red area defines the zone outside the threshold corresponding to the 65 % GHG emissions reduction necessary to produce sustainable fuels according to RED II requirements. The diagonal lines

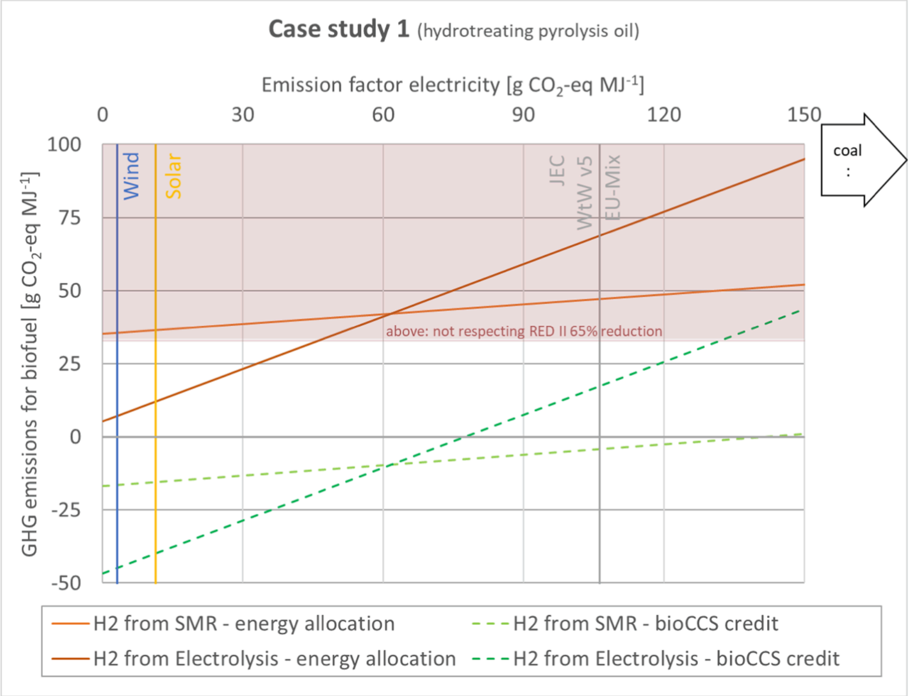

Fig. 6. GHG emission intensity of drop-in diesel produced within CS1.

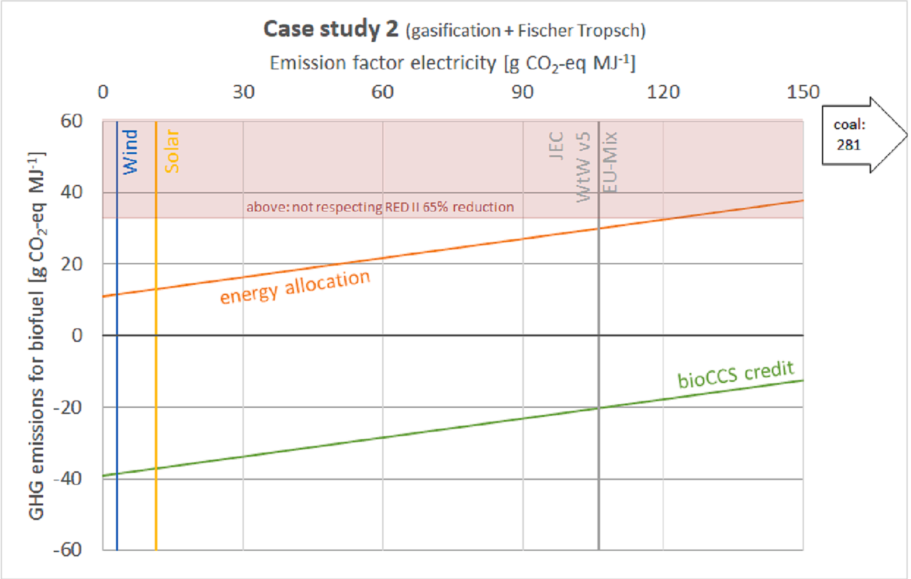

Fig. 7. GHG emission intensity of drop-in diesel produced within CS2.

(orange and blue) represent the trend of the carbon intensities of the pyrolysis-based biofuels considering two different methodologies of hydrogen production, i.e., from SMR or from electrolysis using electricity from the grid, respectively. The graph also features parallel dashed lines that account for the emissions' credit associated with biochar, which is intended to create additional negative emissions, while the solid line considers the energy allocation of liquid biofuels and the use of biochar as a product for other scopes (e.g. voluntary carbon markets). The blue line, corresponding to the carbon intensity of liquid biofuels made of hydrogen coming from electrolysis, has a steeper slope than the orange line, since electricity demand is much higher when hydrogen is produced through electrolysis for the plant instead of producing hydrogen through SMR, and therefore has a larger impact on the GHG emissions.

The carbon intensities of biofuels are given by intersecting the orange and blue lines with the vertical line representing the value of carbon intensity of EU electricity grid mix: considering the yellow line intersecting the grey line, values presented in Table 4 are obtained. Summarizing Fig. 6 shows that achieving a carbon intensity for liquid fuels compatible with REDII sustainability criteria necessitates: the use of the emissions' credit from biochar within the system if hydrogen derives from SMR; electricity supply generated below approximately 40 $gCO_2e\ MJ^{-1}$ when hydrogen is produced from electrolysis regardless of applying the biochar-generated credit; electricity supply generated below approximately 130 $gCO_2e\ MJ^{-1}$ when hydrogen is produced from electrolysis and the emissions' credit from biochar is considered within the system. Fig. 7 depicts the variation of carbon intensity of liquid biofuels depending on the carbon intensity of electricity used in the process for CS2. The same considerations done for Fig. 6 are also applicable to this scenario, with the only difference that this process does not necessitate of additional hydrogen. Therefore, in this scenario, the use of high-RES grid mixes does not notably impact the reduction of biofuel emissions. Consequently, for almost all instances, the emissions remain consistently below the sustainability thresholds set by the REDII. Lastly, it becomes evident that the co-production of biochar for carbon storage purposes is unnecessary unless the grid electricity mix exceeds 120 $gCO_2e\ MJ^{-1}$, thus making it available for the voluntary carbon markets.

By adding the biochar GHG credit, carbon negative sustainable biofuels are always obtained in the considered range of electricity emission factor.

5. Discussion

5.1. Energy optimization and emissions

The proposed approach, which improves lignocellulosic biofuels value chains by including biobased carbon sequestration in the form of biochar, offers a significant opportunity to meet the updated Renewable Energy Directive [4,23] targets for the transport sector by 2030. Specifically, the proposed conversion technologies may contribute to supply large volumes of advanced biofuels with high GHG emission reductions, well below the sustainability threshold imposed by RED II. Moreover, the proposed solution offers the opportunity to store a significant amount of durable carbon in the form of biochar, which ranges from one to three tonnes of biochar produced per tonne of liquid biofuels, depending on the case study.

The present study incorporates the results from experimental work done in European projects, focused on optimizing the development of value chains for advanced fuel production. Unlike other studies, this work considered all relevant issues related to biomass pre-treatment for thermochemical conversion processes and transportation challenges, aspects that are often undermined or marginally considered in most of the assessments: firstly, the necessity to dry the biomass up to 3 % moisture content as required by biomass fast pyrolysis; secondly, the need to consider biomass at higher moisture content where the climate conditions make difficult to perform efficient open air drying, as in many Northern European countries, or when harvesting time do not match with open air seasoning.

The proposed case studies, based on the co-production of biochar alongside biofuels, showed multiple benefits for the EU decarbonization scenario: despite they resulted in lower biomass-to-liquid biofuels conversion rate, fossil inputs demand for drying and transporting feedstock has been strongly reduced.

Two main outcomes have been demonstrated: (1) bioheat excess from slow pyrolysis, opportunely tuned for each system, offer the opportunity to use bioenergy for drying requirements that need abundant, continuous energy supply; (2) the introduction of intermediate bioenergy carriers such as fast pyrolysis bio-oil and slurry into the biomass logistics system, allowed to decentralize the first thermochemical conversion phase in remote areas. This approach allows substantial savings in diesel oil for biomass transportation and remarkably reduced feedstock storage requirements at the centralized conversion unit, employing the concept of bioenergy densification. Specifically, this issue has a significant importance for large-scale projects as depicted by the results (i.e., several hundred thousand tons of biomass per conversion facility to generate 25–50 thousand tons drop-in liquid biofuels per year).

This concept also brings the benefit of enabling the integration of the biorefinery within a conventional oil refinery. This is due to the incoming bio-feedstock being a bio-derived liquid (or bio-slurry) rather than pure biomass, which would require specific pre-treatment and storage infrastructures. Therefore, it becomes more reasonable to allocate the GHG emissions generated within the system to all other co-products, such as electricity and bioheat, which would be readily reused within an integrated infrastructure.

Despite the advancements made in this system, there are unexplored opportunities left for further environmental improvements: for instance, re-purposing the use of drop-in bio-derived diesel into the transports and farm vehicles assumed within these value chains, or proposing more efficient biomass-to-biofuels conversion processes.

5.2. Towards biochar GHG crediting systems

The possibility of claiming biochar as bio-based CCS may trigger numerous opportunities to remove a consistent amount of CO_2 from the atmosphere, but this pathway is still under development within the European legislation. In order to define specific conditions and monitoring requirements to certify biochar as CCS in the same way as CO_2 storage, specific rules for biochar storage in geological deposits should be developed. On the other hand, the current efforts in developing EU Carbon Removals' certification guidelines [74] may unlock soon new economic business models for carbon removal based on biomass solutions. Currently EU negotiators reached a provisional political agreement for establishing the EU-level certification framework for permanent carbon removals, carbon farming, and carbon storage in products [75]. This framework aims to facilitate and accelerate the deployment of high-quality carbon removal and soil emission reduction activities in the EU, and may lead the space to introduce, under certain circumstances, the storage of carbon in biochar as permanent. However, the agreement reached between the EU Council and Parliament is provisional and pending formal adoption by both institutions.

Currently, biochar has the potential to be considered as a substitute for fossil carbon in hard-to-abate sectors, offering a way to mitigate carbon emissions. This could make it a tradeable commodity in the EU ETS market, for example, in the steel industry where it could replace fossil coal. However, it's important to note that biochar cannot be used as a carbon "removal unit" to offset mandatory ETS emissions, due to the reasons outlined earlier. As a result, organizations and initiatives within the voluntary carbon markets are in the process of establishing regulations for certifying biochar production and storage as tradeable carbon units. Their goal is to align these regulations as closely as possible with current policy developments, especially considering that the ETS does not allow any offset with voluntary carbon market credits yet.

The main scope is to ensure that bio-based carbon contained into biochar is safely stored avoiding carbon leakages. For instance, the potential utilization of alternative storage sites than the ones intended for CO_2 (e.g. other than depleted hydrocarbon reservoirs, saline formations, etc.) may be an intriguing possibility. Nonetheless, challenges exist, including the need for comprehensive assessments on the status of these sites, their storage potential and safety issues related to their use. As regards carbon farming or the incorporation of biochar into long-lasting products, robust certification schemes should ensure long-term permanence and no side effects. Particularly, the application of biochar in the first 30 cm of soil of cultivated lands needs further assessment to ensure soil health and long-term verified agricultural performance [76,77].

Summarizing, the methodology proposed within this study can be considered as a robust foundation for policy makers to develop the necessary legislative updates for carbon capture and storage considering biochar within the bioenergy and biofuels sector, starting from geological carbon storage opportunities and then exploiting the significant hidden opportunities to generate potential carbon credits for other sectors.

6. Conclusions

This study investigated the potential of improved lignocellulosic biomass-to-liquid biofuels value chains by integrating fast and slow pyrolysis technologies as biomass pre-treatment in a decentralized approach. This work specifically addressed biomass drying and transportation challenges, suggesting the opportunities for bioenergy substituting fossil-based inputs. The analysis quantifies the benefits resulting by the bioenergy densification, allowed by the production valorisation of bio-intermediate energy carriers, such as fast pyrolysis bio-oil and slurry. These bio-intermediate enhance for effectively reduce logistics costs and storage requirements, enabling potential integration with existing biorefineries. The optimum balance between bioenergy needs for biomass drying and biochar production and liquid biofuel production has been calculated, estimating the ratio of biomass input allocated to the pyrolysis units. When biomass reaches the decentralised bio-intermediate conversion site at 30 % moisture content, 70.9 % of the whole supply goes to fast pyrolysis, generating 49 and 23 hundred tons of liquid biofuels per year for CS1 and CS2 respectively, along with corresponding biochar production of 48 and 21 hundred tons per year. At higher biomass moisture content (50 %), additional biomass is needed to maintain liquid biofuels production unaltered (due to the fixed size of the fast pyrolysis and biofuel conversion plants), but biochar production increased up to almost 95,000 tons per year.

The innovative conversion technologies not only contribute to the production of large volumes of advanced biofuels but also enable significant bio-based carbon sequestration in the form of biochar. Carbon intensity of liquid biofuels was estimated, showing compliance and deviation in comparison with the sustainability threshold set by REDII. CS2 demonstrated lower carbon intensities below the RED' maximum threshold to produce advanced biofuels (65 % GHG emissions reduction), while CS1 exceeded the threshold when fossil-based hydrogen was used. Therefore, to further reduce the GHG emissions and demonstrate the potential of biochar as carbon sink, the results in applying the proposed GHG emissions calculation methodology have been incorporated within the carbon intensities of the liquid biofuels. Comparing these results with the JECv5 pathways, the improved processes show larger GHG emissions saving generated in both CS2 and CS1, allowing a further emissions reduction of 30.9 $gCO_2e\ MJ^{-1}$ for Fischer-Tropsch based-fuels (CS1) and 34.3 $gCO_2e\ MJ^{-1}$ for pyrolysis based-fuels (CS2). For an integrated biorefinery (as in this study, 100 MW capacity) this means to deliver additional savings of 13.3 and 6.8 ktons of CO_2e per year, respectively for CS1 and CS2. Also the possibility of achieving negative carbon sustainable biofuel production through biochar carbon sequestration has been shown within the analysis.

The proposed methodology also allowed to calculate the carbon credit generation potentially tradable in voluntary carbon markets, offering opportunities for decarbonizing hard-to-abate sectors, and stimulating carbon removal solutions in agriculture. The potential of biochar as BECCS solution could unlock new opportunities for carbon removals, developing or improving existing biofuels value chains with combined pyrolysis plants. There is ongoing development of legislative frameworks and certification schemes to recognize and certify biochar production and storage that may trigger new business opportunities for the proposed solutions.

The results also show the pivotal role played by renewable energy sources, particularly greener electricity grids, as significant influencing factors on the GHG emissions associated to biofuels production.

In conclusion, the study underscores the enormous potential of biochar production for carbon removal and storage within existing advanced biofuels conversion pathways, which should be considered to rapidly act against the climate change.

7. Disclaimer

The views expressed here are purely those of the authors and may not, under any circumstances, be regarded as an official position of the European Commission.

CRediT authorship contribution statement

M. Buffi: Writing – review & editing, Writing – original draft, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **O. Hurtig:** Writing – original draft, Validation, Resources, Formal analysis, Data curation. **M. Prussi:** Writing – original draft, Methodology. **N. Scarlat:** Writing – review & editing, Supervision. **D. Chiaramonti:** Writing – review & editing, Validation, Supervision, 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.enconman.2024.118450.

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