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Framework for consequential life cycle assessment of pyrolysis biorefineries: A case study for the conversion of primary forestry residues

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Abstract

In an endeavor towards GHG neutrality, alternatives to fossil carbon resources are explored, such as the use of biomass carbon. Pyrolysis, a thermochemical process that can convert residual biomasses into liquid (bio-oil), solid (biochar) and gaseous (non-condensable gases) products is gaining increased attention, as it both supplies alternatives to fossil carbon while potentially inducing so-called negative emissions through the biochar. Yet, the pyrolysis technical and environmental performance is heavily dependent upon process operational parameters and biomass types. In the perspective of national strategic bioeconomy planning, this study presents an easily replicable consequential life cycle assessment (LCA) framework to quantify the environmental performance of a pyrolysis biorefinery where process conditions are documented, and boundaries expanded to consider the current use of the residual biomass. Results obtained from this method are intended to provide insights for evidence-based decision making towards investments in the low fossil carbon future. The proposed LCA framework was applied to a national case study for the use of primary forestry residues (PFR). Results showed that as compared to the reference scenario in which PFR are left on soil to decay, pyrolysing PFR to biocrude oil, wood vinegar, biochar and gas presents trade-offs in six out of the 16 impact categories studied. These results highlighted that the biomass feedstock supply, the pyrolysis technology, the co-products yields, properties and uses, as well as the choice of marginal technologies have an influence on the environmental performance of pyrolysis biorefineries.

Highlights

- A consequential LCA framework for pyrolysis biorefineries was developed
- A case-study for the use of primary forest residues (PFR) was presented
- Life cycle inventory data for pyrolysis were compiled and presented
- The environmental performance was assessed for 16 impact categories
- Pyrolysis of PFR is climate-efficient as 80% of biochar's carbon is sequestered

Keywords

Pyrolysis, Biochar, Bio-oil, Consequential LCA, Primary forestry residues, Biofungicide

Word count

7633 words (excluding title, author names and affiliations, keywords, abbreviations, table/figure captions, acknowledgements and references).

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Abbreviations

% d.b.	Percentage on a dry basis
%vol.	Volumetric concentration
ACD	Acidification
a.i.	Active ingredient
A-LCA	Attributional life cycle assessment
C	Carbon
Ca	Calcium
CDR	Carbon dioxide removal
CED	Cummulative energy demand
CFC11 eq	Trichlorofluoromethane equivalent
CH ₄	Methane
CHP	Combined heat and power
C-LCA	Consequential life cycle assessment
C ₂ H ₄	Ethylene
C ₂ H ₆	Ethane
CO	Carbon oxyde
CO ₂	Carbon dioxide
CO _{2e}	Carbon dioxide equivalent
C _{org}	Organic carbon
CSE	Carbon sequestration efficiency
CTUh	Comparative toxic unit for human
DM	Dry matter
EF	Environmental footprint
eq	Equivalent
EUT	Eutrophication
F _{perm}	Fraction of biochar carbon remaining after 100 years
g	Gram
Gg	Gigagrams
GWP	Global warming potential
h	Hour
ha	Hectare
H	Hydrogen
H ⁺	Hydrogen ion
H ₂	Dihydrogen
HH	Human Health
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
K	Potassium
kBq	Kilobecquerel
kg	Kilogram
km	Kilometer
kW	Kilowatt
kWh	Kilowatt hour
L	Liter
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LHV	Lower heating value
m ³	Cubic meter
Mg	Megagram

Mg	Magnesium
Mj	Megajoule
Min	Minute
mm	Millimeter
MW	Megawatt
N	Nitrogen
N ₂	Dinitrogen
NCGs	Non condensable gases
NMVOG	Non-methane volatile organic compounds
NO _x	Nitrous oxides
O ₂	Oxygen
ODP	Ozone depletion
P	Phosphorus
PJ	Petajoule
PFR	Primary forestry residues
RESP	Respiratory effects
S	Sulfur
s	Second
Sb	Antimony
seq.	Sequestration
SMOG	Smog formation
SO _x	Sulfur oxides
TRACI	Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts
U	Uranium
µm	Micrometer
USA	United States of America
USD	United States dollar
w.c.	Water content
wt%	Percentage by mass

1. Introduction

Bioeconomy development for the conversion of residual biomass into bio-based products, food, feed and bioenergy is needed to accelerate the transition towards low fossil carbon use. A study by Hamelin et al. [1] revealed the importance of residual biomass as a feedstock for the European bioeconomy, where a theoretical potential of 8500 PJ year⁻¹ was estimated. Among the biomass sources, primary forestry residues (PFR) which include logging residues, stumps, roots and dead or damaged trees represented 3200 PJ year⁻¹. In France alone, it is estimated that at least 2.7 Million Mg DM year⁻¹ (~ 42 PJ) of logging residues (branches, treetops, leaves, needles and bark) could be harvested without reducing soil fertility [2]. Moreover, recent work indicates a theoretical potential (i.e. without considering environmental or economic constraints) of 8.4 Million Mg DM year⁻¹ (4.4 – 13.9 Million Mg DM year⁻¹) in the country [3]. Currently, most of logging residues are left on soil. This contributes to increasing soil's organic carbon and minerals (Ca, Mg, K, P, and N) stocks, to maintaining biodiversity and to avoiding soil compaction [4], among others. Removal of logging residues can alter soil productivity under some site and stand conditions [5, 6] and negatively affect biodiversity [7]. On the other hand, harvesting PFR can provide advantages such as reducing risk of pest infestation [8], reducing the risk of forest fires and facilitating reforestation work [9]. The implementation of harvest and management guidelines based on the specific local conditions are often promoted as a measure to limit the negative impacts of forestry residues removal [10].

Biological and thermochemical processes are two alternatives for biomass feedstock conversion to produce bio-based products and bioenergy. Thermochemical conversion processes use heat to convert lignocellulosic biomasses such as forestry residues, agricultural crop residues and perennial crops. Among the thermochemical conversion processes, pyrolysis is used to convert dry biomass (generally < 10% water content [11]) at elevated temperature (350-700°C) in limited oxygen environment, into a solid biochar, a liquid bio-oil and non-condensable gases (NCGs). Pyrolysis products yields and properties highly depend on biomass characteristics as well as process operational parameters. In a pyrolysis biorefinery, bio-based products (e.g. biochar and wood vinegar) and renewable energy (bio-oil and NCGs) combinations can be produced from biomass by controlling the operational parameters of the plant [12]. Until now, fast pyrolysis plants have been used essentially to supply renewable fuels for heating purposes. Bio-oil produced from pyrolysis as a source of energy can be stored and transported easily as compared to raw biomass feedstock. However, some issues may arise from bio-crude oil combustion in heating appliances due to the high content of reactive oxygen-containing compounds [13]. High-quality bio-oil with low water content, low oxygen content and low acidity can be produced from biomasses with high content of cellulose and low ash content [14], by selecting appropriate pyrolysis parameters. Besides the use of bio-crude oil for combustion purposes, the production of terrestrial transport fuels and even jet fuels by upgrading bio-crude oil is maturing [15].

The global environmental impact of a pyrolysis plant depends on many factors, both linked to the upstream biomass supply chain, and the downstream activities such as the pyrolysis technology itself or the way the generated co-products are used. Life cycle assessment (LCA) is a widely used and normalized [16] environmental assessment methodology to quantify the environmental performance of products and services. In the context of climate emergency, such tools for evidence-based decision making are key for ensuring the environmental performance of future investments in low fossil carbon economy. Only a few LCA studies on pyrolysis systems have been published so far. Albeit valuable, these suffer from short-comings. Notably, they all use allocation techniques where the overall environmental impact is randomly partitioned between the product studied (typically bio-oil) and its co-products (e.g. non-condensable gases, biochars), whether according to the market value of these at a certain point in time, or their dry/wet mass, etc. In other words, the exact use of the co-products is completely disregarded in most studies, which influences the environmental performance of the pyrolysis system. In LCA, this particular way of handling multi-functionality of products is typically referred to as attributional LCA (A-LCA), while a full inclusion of the co-products along with

an understanding of their fate and what they replace on the market is often referred to as consequential LCA (C-LCA) [17]. In other words, a consequential LCA model includes all the activities that are expected to change as a consequence of a change in demand for the functional unit [18]. Sharifzadeh et al. [19] reported that there is a critical knowledge gap regarding C-LCA evaluation of fast pyrolysis. Examples of A-LCA studies evaluating the environmental performance of pyrolysis of forestry biomass are shown in Table 1. Other limitations of the current published studies include a lack of transparency about life cycle inventory data, the exclusion of capital goods in the analysis and the evaluation of a narrow set of environmental impact categories.

The aim of this study is to provide a generic consequential LCA framework for the quantification of the environmental performance of pyrolysis biorefineries, along with a transparent and detailed life cycle inventory of the pyrolysis process itself. The objective is to supply the analytical tools needed in order to figure out if it is environmentally viable to use residual biomass as an input in a pyrolysis biorefinery to produce bio-based products, biochar and bioenergy. To illustrate the proposed framework, a case study for the use of PFR in an auger pyrolysis reactor is presented.

Table 1. Non-exhaustive list of publications about life cycle assessment of pyrolysis systems using forestry biomass as feedstock

Ref.	Background data ¹	Goal and aim of pyrolysis	Pyrolysis technology	Biomass	Functional unit	System Boundaries	LCIA method and Impact categories studied ⁽²⁻¹⁰⁾
[20]	Ecoinvent 3.3 database	Producing bio-oil and biochar for the displacement of fossil fuels (heat) in cement manufacturing.	50 Mg per day mobile plant using steel-shot fluidized bed technology and feedstock drying process at the front-end.	Dry wood chips (2% w.c.)	300000 Mg of cement	From feedstock acquisition to the use products for energy generation.	TRACI (v.2.1) method. ACD, EUT, GWP, ODP, SMOG, RESP. CED method 1.08.
[21]	Aspen modeling software (pyrolysis) / USA LCI database.	Substituting fossil fuels by bio-oil for heat production.	Fluidized bed pyrolysis feed. 2000 Mg (dry biomass) per day.	Wood chips /pine, 7% w.c., <3 mm particle.	1 MJ of energy (bio-oil)	Cradle-to-grave: From collection of forestry residues to bio-oil combustion.	TRACI (v.3) method. GWP, ACD, aquatic EUT, SMOG, ODP, RESP.
[22]	Database in Simapro	Converting biomass from waste wood (residential, institutional, commercial and municipal) to bio-oil.	Flash pyrolysis (500°C). 1783 kg of bio-oil per hour.	Waste wood (residential, institutional, commercial, municipal), < 3mm particle.	Production of 1783 kg _{bio-oil} h ⁻¹	From waste management center to combustion of bio-oil.	EDIP 200X method. GWP, ODP, ACD, EUT, SMOG, ecotoxicity (water, chronic or acute, soil chronic), human toxicity (air, water, soil), waste (bulk, hazardous, radioactive).
[23]	GREET model (GREET.nt vq.3.0.12844), database version 12384.	Producing renewable jet fuel.	Circulating fluidized bed reactor, 500°C. Bio-oil is converted to a mixture of hydrocarbons by hydrodeoxygenation (H ₂).	Poplar, willow, corn stover and forestry residues.	1 MJ of fuel	Well-to-wake, from feedstock cultivation to products end use.	GWP ₁₀₀
[24]	Ecoinvent v.2.2	Gasoline and diesel produced from fast pyrolysis and hydroprocessing for use of the fuel in a vehicle.	Fast pyrolysis: circulating fluidized bed.	Forestry residues	1 km traveled by a light-duty passenger vehicle.	Well-to-wheel	Climate change, Net energy value.
[25]	Literature sources, Ensyn/UOP (for pyrolysis)	Pyrolysis oil combustion for power generation.	Fast pyrolysis integration with power plant.	Woodchips	1 kWh electricity generated	Cradle-to-Grave	GWP ₁₀₀

¹ Ensyn/UOP : <http://www.ensyn.com/honeywell-uop.html>; ²LCIA: Life cycle impact assessment; ³ACD: Acidification potential; ⁴EUT: Eutrophication potential; ⁵GWP: Global warming potential; ⁶GWP₁₀₀: Global warming potential on a 100 years time horizon; ⁷ODP: Ozone depletion potential; ⁸SMOG: Photochemical oxidant formation; ⁹RESP: Respiratory effects; ¹⁰CED: Cumulative energy demand.

2. Methodology

2.1 Overall consequential LCA framework

The LCA methodology follows the ISO international standards: ISO 14040 [26] and 14044 [27]. The consequential LCA approach was selected over the attributional method since it is recognised as the most suitable to support decision-making [28].

Background life cycle inventory (LCI) data were retrieved from Ecoinvent v.3 [29] and Agri-footprint [30,31] databases, while foreground data were taken from literature, pyrolysis experiments and characterisation of co-products. Conformingly with the consequential LCA rationale, marginal processes were used, which can be defined as the technologies installed due to expected long-term changes in demand [32]. The life cycle impact assessment (LCIA) is carried out to translate all flows into 16 characterized impact category metrics using the Environmental Footprint (EF) Method 2.0 as implemented in the Simapro LCA software, version 9.0 (PRé Consultants B.V., The Netherlands). The EF method was selected since it is in accordance with the recommendations of the European Commission [33,34]. Data quality was evaluated for all flows of the model (Table S19), conforming with ISO standards 14040 and 14044 [26,27]. Moreover, the importance of data for each flow in the 16 impact categories was qualitatively assessed (Tables S20 and S21 in the supporting document).

2.2 Goal and scope definition

Besides the production of a framework for pyrolysis LCA, the goal of this illustrative case study is to quantify the environmental performance of pyrolysis biorefineries fed with PFR, where the pyrolysis process is driven by the production of bio-crude oil. The vision is to compare this environmental performance with the current system in which PFR are left on soil to decay (reference case, also referred to as counterfactual case). It will allow determining whether it is environmentally viable or not to harvest PFR for conversion by pyrolysis to produce bioenergy, biochar and bio-based products (herein biopesticide). As the results of the LCA to be performed are intended for strategic investment decisions, it is the longer-term temporal scope (30 years) that is considered herein to select the marginal technologies. The time horizon for the impact assessment is 100 years. The geographical scope of interest is metropolitan France.

The functional unit, i.e. the unit to which input and output flows are related, corresponds to the management of 1000 kg of dry biomass. This is selected in the perspective of using the framework essentially with residual biomass, but also for future comparison with other bioeconomic uses for this feedstock.

2.3 Scenario description and system boundaries

The system boundary (Figure 1) includes the biomass supply chain (harvest, chipping, transport), the conditioning of biomass (e.g. storage, grinding, drying), the pyrolysis plant construction and operation (including the fractional condensation of pyrolysis gases) and the use of the pyrolysis co-products. The system boundaries are expanded to include what is affected by the use of PFR for pyrolysis and the use of the biorefinery co-products.

A mass balance (Table S1 and S2) was established considering the harvest of 1000 kg of dry PFR (conifer species) from forests of France. The quality and partition between the different co-product of pyrolysis are highly dependent upon process conditions, among other the temperature, the residence time of biomass in the reactor and the flowrate and type of carrier gas [35,36]. The quantity and properties of each pyrolysis co-product (bio-crude oil, wood vinegar, non-condensable gases and biochar) were determined based on published and on-going experimental data, as further described by Brassard et al. [37]. Accordingly, the auger reactor technology was selected for the pyrolysis process.

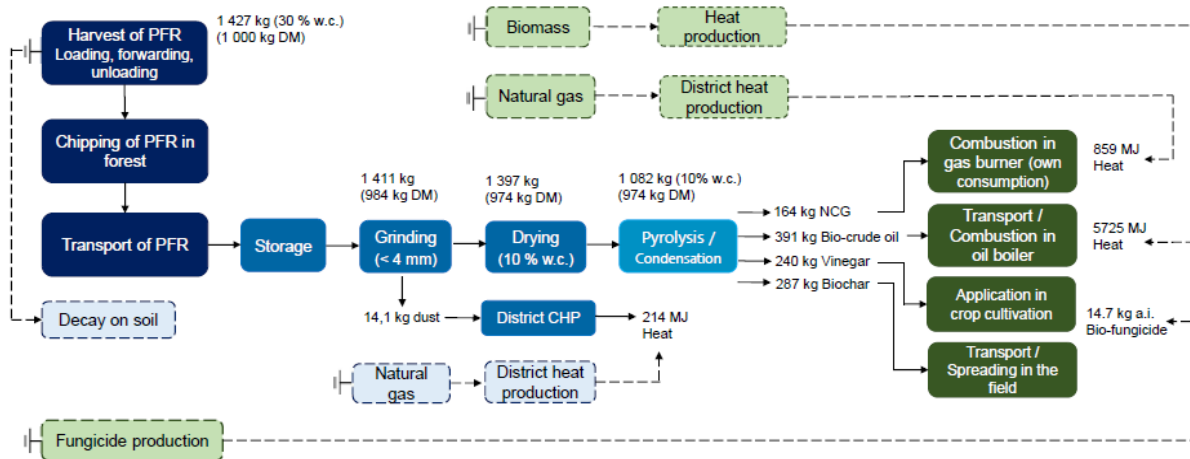


Figure 1. Process flow diagram of the pyrolysis scenario studied. Dotted lines represent avoided processes (reference scenario). Processes without flow indicate no losses. PFR: Primary forest residues; NCG: Non-condensable gas; CHP: Combined heat and power.

2.4 Substitution of products and related services

As illustrated in Figure 1, harvesting PFR avoids its counterfactual management, which is the decay on soil. Here, the vision is to produce bio-crude oil to be used in small-scale oil boilers in areas without access to the gas grid. Other uses of the bio-crude oil could of course be considered, for instance as a feedstock for bio-based aviation fuels following hydro-processing and hydro-cracking [38]. Here, the purpose was to illustrate that the selected use of the bio-crude oil conditions the whole system boundary for the LCA. In the short-term, bio-crude oil would likely substitute fossil fuel oil. For instance, fuel oil represents 12.8% of energy sources used for building heating systems in France [39]. In the longer-term, it may rather prevent the deployment of biomass boilers [40], which is considered herein.

Biochar is applied to soil as amendment and it is considered that no additional process is avoided by this, as no soil amendment would have been added otherwise. The wood vinegar is used as a bio-fungicide and chemical fungicides production is therefore avoided.

It is considered that non-condensable gases are burned in a natural gas industrial furnace (> 100 kW) for district heat production. The equivalent amount of heat is considered avoided from the combustion of natural gas in the same industrial furnace. The vision is that the heat could be used at the pyrolysis plant which is nearby a city where natural gas grid is accessible. Similarly, the heat produced by district cogeneration (CHP) is considered to substitute the heat produced by the combustion of natural gas.

The marginal electricity mix used in all processes (high and medium voltages) is taken from Ecoinvent 3.5 for consequential modeling. Accordingly, the power sources for France are wind (84%), wood (13.2%), geothermal and hydroelectricity.

2.5 Scenario modelling and life cycle inventory

2.5.1 Biomass supply chain

The PFR feedstock can be defined as the by-products of wood harvesting. In this study, it is considered that only logging residues (treetops and small branches less than 7 cm; stumps are excluded) are harvested for pyrolysis. The biomass supply chain considered herein includes the harvest of PFR in forest by forwarder, transport to the roadside, chipping to 10-100 mm using a mobile chipper consuming diesel and transport to the biomass depot by van. This corresponds to the

most widespread supply chain for logging residues after final fellings in Europe, including France [41]. The operation time of forwarder and chipper for processing 1 Mg of dry biomass was calculated [42, 43] as detailed in the supporting document (Table S3 and S4).

Meta-analyses of field studies did not report significant reduction of soil organic carbon (SOC) following removal of forestry residues [10]. Therefore, changes in forest carbon stocks due to forestry residues removal are assumed to be negligible [14].

The chemical composition of PFR at harvesting (Table 2) was determined based on a compilation of literature data. The moisture content of wood residues collected in the forest is variable and depends on many parameters. Typical moisture content for fresh forestry residues is near 50% [44], and can vary from 48 to 57% [45]. In a study by Pettersson and Nordfjell [46], moisture content of logging residues stored in small piles at the felling site dropped from approximately 50% to 28.6% in 3 weeks in June. According to Thörnqvist [47], the biomass can reach 25% moisture content by the end of summer when they remain in the logging area. Routa et al. [48] reported that logging residues can dry by up to 30% in the forest during a 6-week period if weather conditions are favorable. Accordingly, it is here considered that fresh PFR at 50% moisture content are left on soil in dry weather conditions for passive drying during at least a six weeks period to reach 29.9% moisture content before harvesting. This reliance on passive drying implies that PFR are harvested only between May and October.

Table 2. Elemental composition of primary forestry residues at harvest

Element	Unit	Value
Carbon (C)	% (d.b)	51.5
Oxygen (O)	% (d.b)	40.5
Hydrogen (H)	% (d.b)	6.0
Nitrogen (N)	% (d.b)	0.44
Sulfur (S)	% (d.b)	0.01
Ashes	% (d.b)	1.5
Water content	% (w.b)	29.9

2.5.2 Biomass storage and conditioning

The surface area of the building needed to store the biomass was calculated based on considerations detailed in the supporting document. Storage of wet biomass can cause microbial decomposition and chemical reactions at high temperatures [43,44], which increases the dry matter loss (and therefore emissions to air) besides causing a decline in fuel quality. The dry matter loss can be associated with methane (CH₄) emissions from the chips pile [44]. According to the review performed by Hofmann et al. [45], the maximum moisture content for storage stability is between 15 and 30 %. Drying in forest to less than 30% water content prior to chipping and storage therefore allows stable storage conditions with few losses of dry matter [48]. A 2% dry matter loss during storage was selected in the present case study [42]. It is assumed that only CH₄ and CO₂ were emitted due to the degradation of dry matter, based on Tonini et al. [49] (see calculation in the supporting document, Table S6).

Auger pyrolysis reactors, as considered in this study, require biomass with small particle size to favor the heat transfer and to ensure complete reaction [14]. A range of particle size of 1-3 mm is typically recommended for pyrolysis [21, 22, 50]. In the study by Brassard et al. [51], wood particle size between 1 and 3.8 mm was used for pyrolysis in an auger reactor. The pyrolysis scenario studied here considers that wood chips are grinded using a stationary electric chipper (Table S7). The mass loss into dust following grinding was estimated to 1% of total mass at the inlet of the grinder. This dust is here burned for heat and power co-generation (Table S9).

Biomass physicochemical properties will influence both the pyrolysis process and the various properties of co-products. More specifically, the moisture content of biomass has high importance as it influences the physicochemical properties of bio-oil [52]. Bridgwater et al. [11] recommend a water

content below 10 wt% prior to feeding the pyrolyzer in order to produce a bio-oil with a water content below 30 wt%. Therefore, it is here considered that PFR are further dried to 10 wt% moisture prior to pyrolysis [20,21] with a rotary dryer [53]. The temperature generally employed is around 200°C but should not be above 250°C in order to prevent degradation of dry matter [54]. In these conditions and with a biomass water content < 30 wt% prior to drying, degradation of dry matter and consequently gas emissions are considered to be negligible [55] at the drying stage. The energy needed to evaporate 1 kg of water from biomass was estimated to 4 MJ kg⁻¹_{water}, according to literature [56,57]. Therefore, the total heat needed was calculated based on the quantity of water to evaporate (Table S8). It is considered that the energy source to heat the rotary dryer is natural gas burned in an industrial furnace.

2.5.3 Pyrolysis and condensation

Mass balance of the pyrolysis biorefinery (Table S2) was established based on experiments carried out with a semi-pilot scale auger reactor of 1 kg biomass per hour capacity [37]. These experiments were performed using pyrolysis parameters (temperature, residence time and nitrogen flowrate) that were determined using the response surface methodology approach to produce the highest yield of bio-oil [51]. Nitrogen is used as a carrier gas to prevent the injection of oxygen in the pyrolysis system. The residence time represents the duration that biomass stays in the reactor.

Further pyrolysis experiments on the set-up described by Brassard et al. [37] were carried out by Álvarez-Chávez [58] to study the fractional condensation of bio-oil in order to obtain a bio-crude oil with lower water content and an aqueous fraction. The condensation system consists in two double shell stainless tubes in which water/glycol (50:50 mixture in the first condenser and 100:0 in the second condenser) circulates counter flow to pyrolysis gases. The heavy phase (bio-crude oil) is obtained in the first condenser in which the cooling fluid is set at high temperature and the light phase (wood vinegar) is obtained in the second condenser at a lower temperature. The pyrolysis and condensation operational parameters and product yields considered in this case study are presented in Table 3. Electricity supply to the pyrolysis and condensation units was calculated (Table S10 and S11) based on the energy consumption of the semi-pilot pyrolysis unit described by Brassard et al. [37].

Table 3. Parameters and product yields considered for the pyrolysis of PFR (as soft wood residues)

Pyrolysis parameters		
Temperature	°C	559
Biomass residence time	s	61
N ₂ flow rate	L min ⁻¹	3
1 st condenser temperature	°C	120
2 nd condenser temperature	°C	4
Product yields		
Bio-crude oil	wt%	36.1
Wood vinegar	wt%	22.2
Biochar	wt%	26.5
Non-condensable gases	wt%	15.2

A few industrial scale pyrolysis units were developed in the last few years. Perkens et al. [15] listed twenty pyrolysis plants around the world operating at feed rates of 1 to 274 Mg per day. Industrial auger pyrolyzers with capacities up to 50 Mg day⁻¹ are available on the market [15,59].

On this basis, the full-scale pyrolysis plant modelled in this study is expected to process 20 Mg of biomass (10% w.c.) per day and 4800 Mg per year. It is considered that products yields and properties will be similar to the results obtained in the semi-pilot reactor described above [37] (Table 3). The construction of the pyrolysis plant was modeled using the infrastructure process "Synthetic gas factory" from Ecoinvent 3.5 (see the details in the supporting document, Table S10). This process

includes buildings and facilities (including dismantling) of a typical biomass gasifier as well as land use. Its lifetime is assumed to be 50 years.

2.5.4 Combustion of bio-crude oil and avoided combustion of wood chips

The produced bio-crude oil has a water content of 18.8 wt% and an energy content of 18.3 MJ kg⁻¹ [37]. The heat produced by bio-oil burning (5725 MJ, which considers 80% conversion efficiency for the oil burner) is considered to replace the equivalent heat provided by the combustion of softwood chips in a central or small-scale furnace (50 kW).

Transport distance from pyrolysis plant to the burner is conservatively estimated to 200 km. In general, the emissions from bio-oil combustion are between those of light (fossil) oil and heavy oil, but particulate emissions can be higher and SO_x emissions are very low [60]. Similarly, Hou et al. [61] experimented the combustion of a blend of 2.5% bio-oil / 97.5% heavy fuel oil and found that emissions were not different as compared to the combustion of pure heavy fuel oil. Ren et al. [13] also reported that raw bio-oil had comparable combustion performance to fuel oil. Accordingly, the partition of C, N and S compounds in the gaseous emissions from bio-crude combustion was modeled using the partition of gases provided by the process "Heat, central or small-scale, other than natural gas {Europe without Switzerland} | heat production, light fuel, at boiler 100 kW, non-modulating | Conseq, U" retrieved from Ecoinvent database version 3.5 (Table S12).

2.5.5 Combustion of non-condensable gases and avoided combustion of natural gas

The non-condensable gases (NCGs) composition considered herein is based on the sampling and characterization work described by Brassard et al. [37]. In this study, hydrogen (H₂) has not been analyzed and was therefore added to all other compounds reported. The volumetric concentration (%vol.) of H₂ was fixed at 5.2%vol. based on simulations made from the equilibrium reactions presented by Ferreira et al. [62] and the remaining volumetric composition was adjusted on the basis of the results reported by Brassard et al. [37]. The resulting composition and lower heating value (LHV) of the pyrolysis NCGs is presented in Table 4.

It is considered that NCGs are burned in a natural gas industrial furnace (> 100 kW) for district heat production. This results in 858.7 MJ of heat produced, which was calculated from the calorific value of gases (Table 4) and the heat conversion efficiency of syngas estimated at 75% by Roberts et al. [57]. The equivalent amount of heat is considered avoided from the combustion of natural gas in the same industrial furnace, but with an efficiency of 85%.

The partition of C, N and S compounds in the gaseous emissions from non-condensable gases combustion was modeled using the partition of gases provided by the process "Heat, district or industrial, natural gas {Europe without Switzerland} | heat production, natural gas, at industrial furnace > 100 kW, | Conseq, U" retrieved from Ecoinvent database version 3.5 (Table S14).

Table 4. Characterization of pyrolysis non-condensable gases

Composition	Unit	Value
CO	%vol.	43.2
CO ₂	%vol.	42.9
CH ₄	%vol.	7.34
C ₂ H ₄	%vol.	0.58
C ₂ H ₆	%vol.	0.74
H ₂	%vol.	5.2
Lower heating value	MJ kg ⁻¹	7.0

2.5.6 Biochar as a soil amendment

The biochar elementary composition (Table 5) was determined based on pyrolysis experiments with woody biomass [37] and adjusted according to the chemical composition of the PFR considered herein (Table 2). It is considered that biochar is transported on a 100 km distance from the pyrolysis plant to a farm with a freight lorry, mixed with manure, and applied with a conventional manure spreader (see detailed process in Table S15).

Biochar applied to soil increases soil C stocks, resulting in net CO₂ removals from the atmosphere. A large fraction of C in biochar is retained in soil over several decades, but the time it remains will vary depending on the type of biochar and soil conditions [63]. According to 12 studies published after 2010, the C sequestration potential by 2050 ranges between 0.03 and 6.6 Gt CO₂e yr⁻¹ [64]. Schmidt et al. [65] demonstrated that pyrolytic carbon capture and storage can attain carbon sequestration efficiency (CSE) of more than 70% when C in biochar and bio-oil are sequestered, CSE being defined as the percentage of the feedstock C being captured [66]. According to Budai et al. [67], a biochar H/C_{org} molar ratio in the range 0.4 – 0.7 would indicate that the fraction of biochar C remaining after 100 years (F_{perm}) is equal to 50%. The IPCC [68] indicated that the F_{perm} is 80% (± 11%) for biochar produced at medium temperature (450-600 °C). In a LCA of large-scale biochar production, Azzi et al. [69] have set the carbon stability of biochar to 80%, but also considered lower and higher values of 70 and 90% in a sensitivity analysis. Therefore, F_{perm} was set to 80% in the present case study.

Table 5. Characterization of biochar (% d.b.)

Parameter	Value
Carbon (C)	70.8
Nitrogen (N)	0.44
Hydrogen (H)	4.27
Oxygen (O)	19.4
Sulfur (S)	0.008
Ashes	5.09

2.5.7 Wood vinegar used as a biofungicide

The composition of the aqueous fraction of bio-oil (wood vinegar) was derived from unpublished data produced from fractional condensation experiments with the pyrolysis set-up described by Brassard et al. [37]. Accordingly, it has a water content of 53.1 wt%, it contains sugars (5.7 wt%), phenolic compounds (8.2 wt%) and acetic acid (11.4 wt%). Some studies reported that phenolic compounds in the wood vinegar have an antifungal potential [70,71,72,73,74]. According to Jung [75], phenols in combination with methanol and carboxylic acid are the major anti-fungal compounds found in bio-oil. Using the aqueous fraction of bio-oil as a bio-fungicide is consistent with the increase of the global biopesticide market that was valued at 3 Billion USD in 2017 [76] and 4.4 Billion USD in 2019 [77]. Among all biopesticides, biofungicides hold the largest market share [77].

In this case study, wood vinegar is expected to replace a typical fungicide on the market. Generic fungicide LCI data were taken from Agri-Footprint database (2017; Table S17) but the process was modified by integrating the marginal sources of energy considered herein. In that process, the application rate of fungicide is 0.292 kg a.i. ha⁻¹ by mixing 1 kg of fungicide to 370 kg of water. The same total volume but with a different proportion of wood vinegar (16.3 kg) and water (354 kg) is applied in the pyrolysis scenario. In this solution, the acetic acid concentration is limited to 0.5% in order to avoid damage to the plants [37].

2.5.8 Counterfactual fate of PFR

It is considered that the counterfactual fate of PFR is the decay on soil, i.e. the PFR are left on-site and never harvested. All the C from biomass decay on soil is considered to be released as CO₂ because forest soils are generally aerobic [10]. The decomposition rate of PFR within 100 years was

estimated at 91% based on a study by Repo et al. [78]. The authors developed a model simulating the decomposition of forestry residues and reported that 2-16% of the initial branch (Norway spruce species) remains after 100 years. Accordingly, the decay of 1 Mg of PFR on soil would emit 1715 kg CO₂e and would sequester 169.6 kg CO₂e.

2.6 Uncertainty analysis

The sources of uncertainty in LCA are numerous and are related to data (parameter uncertainty), choices made in the construction of scenarios and impact assessment modelling choice [79]. In this study, uncertainty is assessed using sensibility analysis for the potentially sensitive parameters identified after a contribution analysis. The parameters selected for a sensitivity analysis are the one that are uncertain and for which a change can cause a different conclusion in the impact assessment for a given impact category.

3. Results and Discussions

3.1 Impact assessment

3.1.1 Climate change impact

The pyrolysis scenario shows a reduction of 906.4 kg CO₂e Mg⁻¹ dry PFR as compared to the reference (or counterfactual) scenario in the climate change impact category (Figure 2). The modeled pyrolysis plant processing 4800 Mg of biomass (10% w.c.) per year would therefore contribute to a reduction of 4264 Mg CO₂e year⁻¹. This corresponds to ca. 645 average passenger cars (a mix of car sizes and fossil fuel use) travelling 20,000 km, based on the Ecoinvent process "Transport, passenger car, EURO 4 | Conseq, U".

The difference between both scenarios is mainly due to the sequestration of ~142 kg C_{biochar} Mg⁻¹ dry PFR in the pyrolysis scenario, considering that 80% of the biochar-C remains in soils for more than 100 years. The CO₂ emissions from the combustion of bio-crude oil (674 kg CO₂e) is the biggest contributor to the climate change category as its importance was defined as crucial (Table S20). It is followed by the emissions from PFR storage and pretreatment (295 kg CO₂e) which includes 206 kg CO₂e from wood chips storage (88.2% from CH₄ and 11.6% from CO₂), and 87 kg CO₂e from drying using heat from natural gas combustion. The contributions of the PFR supply chain processes (harvest and transport) and of the pyrolysis & condensation processes are relatively small.

Considering the objective of limiting the increase in the global average temperature to well below 2°C above pre-industrial levels [80], PFR are thus clearly better managed as a feedstock to pyrolysis for bio-oil and biochar production than left unharvested to decay in forests (reference). However, Giuntoli et al. [10] demonstrated that the climate mitigation efficiency of bioenergy production from forest logging residues depends on the decay rate of biomass left on soil. The result obtained for the case study presented herein is based on the hypothesis that 91% of the forest residues left on soil will decompose and emit CO₂ within 100 years [78].

The reduction of 906.4 kg CO₂e Mg⁻¹ dry PFR stands in the range of 0.4 – 1.2 Mg CO₂e Mg⁻¹ (dry feedstock) reported in other studies on the life cycle climate change impact of biochar systems [81]. In the present study, only the C sequestration potential of biochar was considered as a GHG emission mitigation effect. However, Azzi et al. [69] reported that considering cascading effects of biochar could double the benefits provided by biochar in terms of CO₂ emissions mitigation. This additional effect can be obtained when biochar is used to feed cows to reduce enteric CH₄ emissions, mixed with manure to reduce emissions from manure storage and applied to soil to reduce N₂O emissions from nitrogen fertilization. A meta-analysis [82] revealed that biochar overall reduces N₂O emissions by 38% (compared to situations without biochar application), but that reductions are greater immediately after application. Biochar can provide additional benefits when added to soil and have positive effect on soil physical, chemical and biological properties, which can increase plant growth

[83]. However, these effects are specific to biochar type, manure and soil properties and environmental conditions.

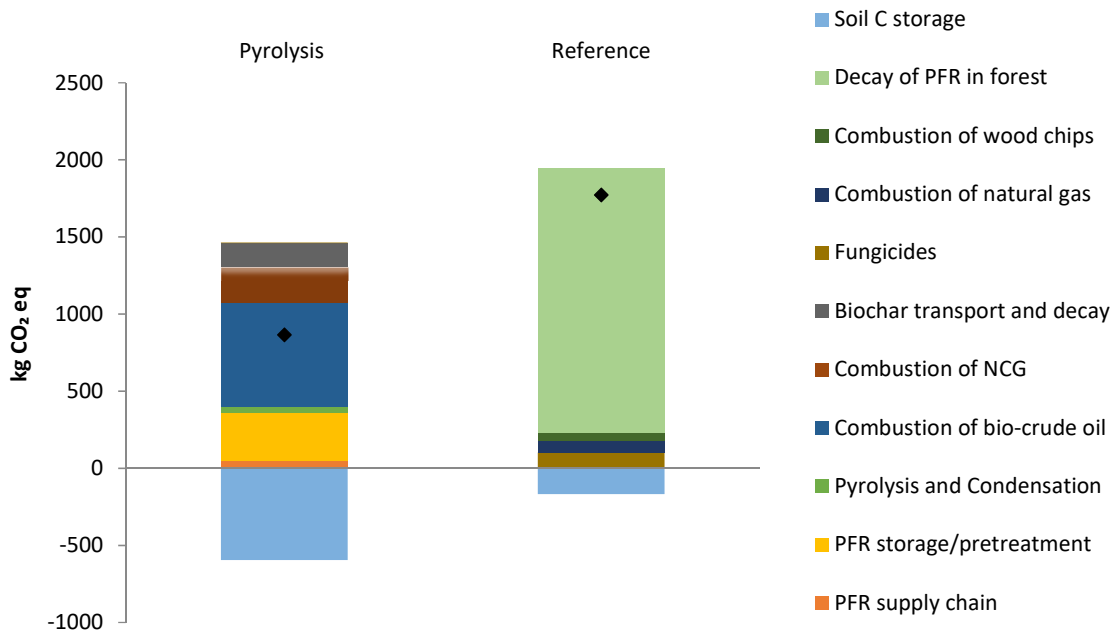


Figure 2. Contribution analysis for climate change impact category (kg CO₂e Mg⁻¹ dry PFR). PFR: primary forestry residues; NCG: Non-condensable gases; CHP: Combined heat and power.

3.1.2 Trade-offs

Trade-offs were observed for six impacts, i.e. impacts for which the reference scenario presents a higher performance than for the pyrolysis case. These are “cancer human health effects”, “freshwater eutrophication”, “resource use (minerals & metals)”, “ozone depletion”, “Acidification, terrestrial and freshwater”, and “water scarcity” (Figure 3). The pyrolysis and condensation process is the highest contributor in the first three impact categories mentioned above. Most of the contribution (82 to 85%) of this process is from the use of electricity as a source of energy in the pyrolysis process. In the consequential electricity mix used (France), 84% of electricity is produced from wind turbines.

The main substance contributing to “Cancer human health effects” is chromium, emitted to water and air from the steel production for the construction of wind turbines. Similarly, the main substance contributing to “Freshwater eutrophication” is phosphate, which is emitted by the copper production used for the construction of wind turbines. The contributions to “Resource use, mineral and metals” essentially stem from three processes: pyrolysis (metals used in the construction of wind turbines for electricity production), storage of biomass (metals used in the construction of the building hall), and combustion of bio-crude oil (chromium and copper used for the boiler and storage tanks).

Though it was unimportant for climate change, electricity consumption of the pyrolysis process is thus a critical spot for these impacts. The consumption of the pyrolysis reactor (781 kWh Mg⁻¹ biomass) measured in experiments with a semi-pilot scale reactor could be slightly overestimated for a full-scale pyrolysis plant. For instance, Tews et al. [14] established the energy balance of a fast pyrolysis plant with a capacity of 3000 Mg of dry biomass per day, modeled based on a compilation of experimental data. Authors indicated that the need in heat and power of the pyrolysis unit is ca. 12% of energy content of biomass input, which would be equivalent to 564 kWh Mg⁻¹ biomass in the present scenario.

The trade-off (i.e. delta between the pyrolysis and reference scenario) in the impact category “Acidification terrestrial and freshwater” is only 0.018 mol H⁺eq Mg⁻¹ PFR. The main contributing substances to this impact in the pyrolysis scenario are nitrogen oxides (NO_x, 60.9%) and Sulfur dioxide (SO₂, 30.8%). About 45% of these substances are emitted from the combustion of bio-crude oil. In the reference scenario, 64.7% of the contribution to this impact comes from NO_x emissions from wood chips combustion.

The contribution to ozone depletion is, in the pyrolysis scenario, mainly due to the harvest of forestry residues (PFR supply chain) and the drying of wood (PFR storage/pretreatment). Methane (bromochlorodifluoro-, Halon 1211) emitted from the transport through pipeline of natural gas used as an energy source to dry the wood to 10% w.c. contributes for 40.3% of the total emissions in this category. Similarly, in the reference scenario, combustion of natural gas (marginal energy replacing the energy produced by dust and NCGs combustion) contributes to the “ozone depletion” potential mainly because of the emission of Halon 1211. Finally, about 25% of the total CFC111 eq. of the pyrolysis scenario is due to methane (bromotrifluoro-, Halon 1301) emitted from the diesel production used by the forwarder for PFR harvest and by the lorry for the transport of the wood chips.

The process consuming the highest amount of water is, in both scenarios, the application of pesticides (37 m³ for biofungicides in the pyrolysis scenario and 49.5 m³ for fungicides in the reference scenario). However, the use of water is higher in the pyrolysis scenario (+5.7 m³ because 13.6 m³ of water is needed for condensation).

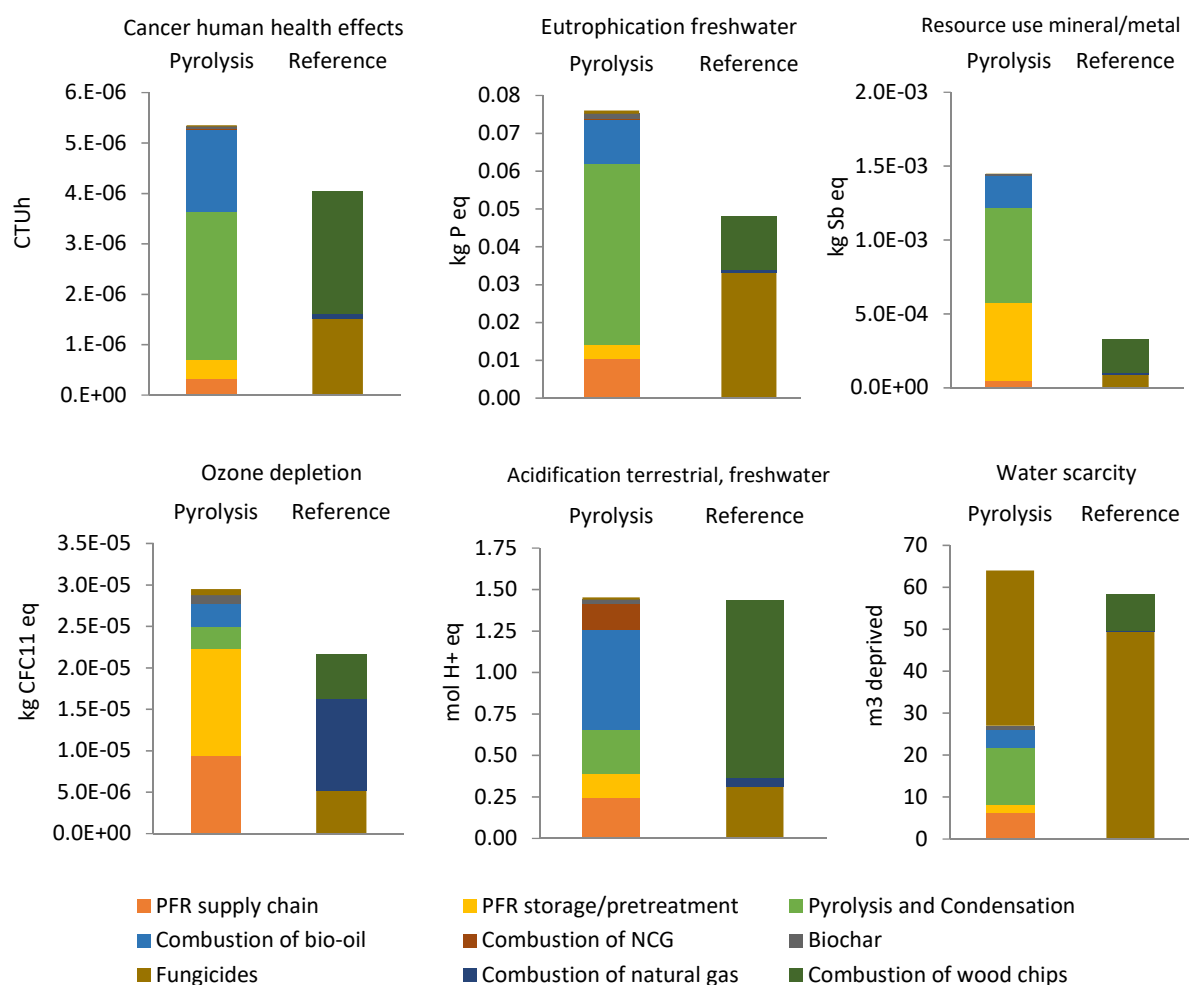


Figure 3. Contribution analysis (per Mg dry PFR) for six impact categories presenting a trade-off for pyrolysis scenario. PFR: primary forestry residues; NCG: Non-condensable gases.

3.1.3 Remaining impact categories

In the nine additional impact categories (Figure 4), the environmental performance of the pyrolysis scenario is better (lower impact) than in the reference scenario. The combustion of wood chips is the major contributor in the reference scenario for many impact categories. In the impact categories “photochemical ozone formation (HH)”, “eutrophication marine” and “eutrophication terrestrial”, the main contributing substance is nitrogen oxides (NO_x), which is mainly emitted from the combustion of wood chips (reference scenario), and bio-crude oil and NCGs (pyrolysis scenarios). NO_x emissions from the combustion of softwood chips at a furnace (50 kW; marginal heat in areas without access to the gas grid) is relatively high (0.143 g MJ^{-1}) as compared to the emission of NO_x calculated for the combustion of bio-crude oil (0.07 g MJ^{-1}) in a small-scale boiler (100 kW). The quality of this data (i.e. NO_x emission for these combustion processes) is acceptable (Table S19) but could be improved by experimental measurement of emissions from the combustion of bio-crude oil. Moreover, NO_x emissions from combustion can be reduced by adopting different types of measures like energy efficiency improvements or specific mitigation technologies [84]. However, such technologies like selective catalytic reduction, which can reduce NO_x emissions by 80-95%, are applied only on large combustion plants (> 100 MW) as it would not be cost-effective on smaller combustion units.

The combustion of wood chips also contributes highly to the “ecotoxicity freshwater” and “non-cancer human health effects” impact categories. The main contributor substance is zinc accumulated in soil in ashes and emitted to air.

According to the model, particulate matter (< 2.5 µm) emissions from the combustion of woodchips are higher than from combustion of bio-crude oil for the same amount of heat produced. This is why the number of disease increase is higher in the reference scenario for the “respiratory inorganics” impact category (Figure 4).

The production of wood chips used for combustion contributes to the higher “land use” impact in the reference scenario, due to occupation of forest land (intensive). In the Ecoinvent process, it is considered that 27577 m² x year are occupied and 674 m² are transformed per tonne of wood chips. Indirect land use changes are not even included. Pyrolysis also contributes to land use impact due to the considered electricity mix, produced partly from wood.

Carbon-14 emitted to air during the fabrication of fungicide is the main contributor to the ionizing radiation impact category in the reference scenario. In the pyrolysis scenario, the biomass supply chain contributes mostly to the ionizing radiation. Emissions of Carbon-14 come from diesel production, which is used as an energy source for harvest and transport of residues.

Finally, the “Resource use, energy carriers” impact is higher in the reference scenario because the fabrication of fungicides consumes a lot of energy, mostly from natural gas, uranium, oil and coal. Combustion of natural gas and wood chips also contribute to the high energy consumption of the reference scenario. In the pyrolysis scenario, biomass drying is the most energy consuming process (natural gas).

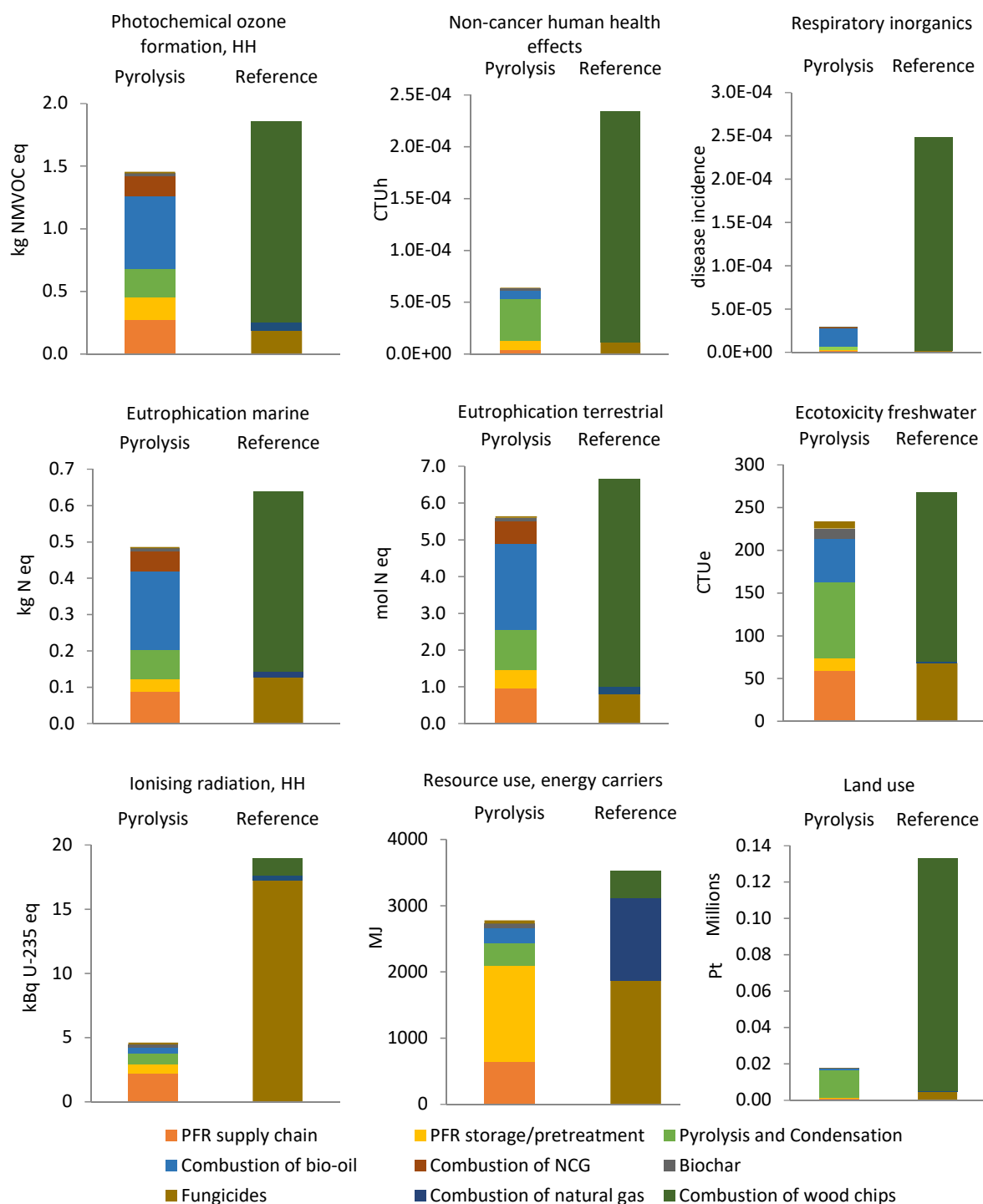


Figure 4. Contribution analysis (per Mg dry PFR) for 9 impact categories. PFR: Primary forestry residues; NCG: Non-condensable gases; CHP: Combined heat and power.

3.2 Sensitivity analysis

The potential for C sequestration by biochar is a key parameter for the impact category climate change (Fig. 2). Therefore, based on literature data, a sensitivity analysis for C sequestration values (F_{perm}) of 70, 80 (baseline) and 90% (over 100 years) was performed (Figure 5). It shows that even with a lower C sequestration potential (70%), the pyrolysis scenario still presents a lower GHG impact than the reference scenario. Considering a C sequestration potential of 90%, the GHG savings reaches 1.06 Mg CO₂e per Mg of dry PFR harvested.

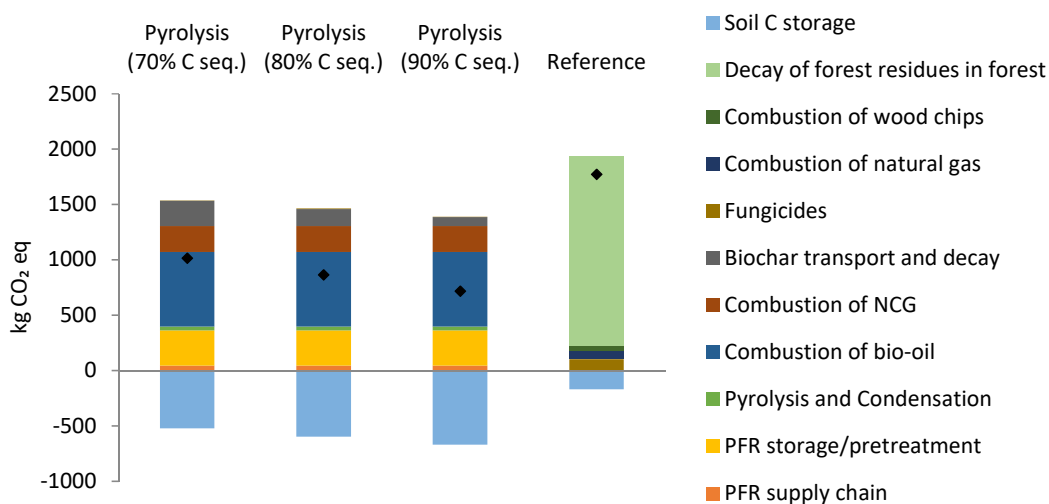


Figure 5. Sensitivity analysis for the climate change impact category ($\text{kg CO}_2\text{e Mg}^{-1}$ dry PFR) for different values of carbon sequestration potential (C seq.). PFR: primary forestry residues; NCG: Non-condensable gases.

The process “pyrolysis and condensation” is the major contributor to “Cancer human health effects” and “eutrophication freshwater” impact categories, contributing to the trade-off for the pyrolysis scenario. A decrease of the overall impact from this process could contribute to reduce or avoid a trade-off. The electricity consumption of the pyrolysis reactor, which contributes mostly to this process, could have been overestimated due to a lower efficiency of the semi-pilot scale reactor as compared to an industrial scale reactor. Therefore, a sensitivity analysis considering a reduction of 15% and 30% of electricity consumption of the pyrolysis reactor was performed. Even with the reduction of the electricity consumption of up to 30%, the pyrolysis scenario presents a trade-off of 12% for the cancer human health effect (Figure S1) and 24.9% for the eutrophication freshwater (Figure S2). The trade-off in the impact category “acidification terrestrial, freshwater” is very low in the baseline case (1.8%) and is therefore avoided with reductions of 15% and 30% of electricity consumption (Figure S3).

Fungicides production is the process contributing the most to the environmental impact of the reference scenario in the following impact categories: “Eutrophication freshwater” (69.2%), “ionizing radiation” (90.9%), “water scarcity” (85.1%) and “resources use, energy carriers” (52.9%). Moreover, fungicides have a non-negligible contribution in the “acidification terrestrial, freshwater” impact category in which a small trade-off is observed. However, the dose of wood vinegar that could be applied in the field to replace fungicides is still uncertain, as literature data are not available at large scale. A 0.5% acetic acid concentration in the biofungicide solution was selected for the baseline scenario, which could be sufficient to observe a fungicide effect, according to an experiment described in [37]. As this experiment was carried out in incubation conditions, the required concentration in the field could be higher. Therefore, a sensitivity analysis considering 0.5% (baseline), 0.75% (sensitivity 1 – S1) and 1% (sensitivity 2 - S2) concentrations of acetic acid was performed for the impact categories mentioned above. Increasing the concentration of acetic acid means that the amount of wood vinegar produced in the pyrolysis scenario (240.2 kg) will be spread on a reduced surface area. Consequently, less fungicide will be substituted (14.7 kg, 9.8 kg and 7.4 kg for the baseline, S1 and S2, respectively). Results of the sensitivity analysis show that an increased dose of wood vinegar (reduced quantity of fungicides avoided) increases the trade-off in the impact category “Eutrophication freshwater” and “acidification terrestrial, freshwater” (Figure S4 and S5). In the “ionizing radiation” impact category, the reduction of the emission of kBq U-235 eq. in the pyrolysis scenario as compared to the reference scenario is decreased from 14.33 in the baseline to 5.73 for S2 (Figure S6). Regarding the “resources use, energy carriers” impact category, the pyrolysis scenario presents a better environmental performance in the baseline and for S1 (Figure S7).

However, S2 results in a trade-off, as the energy use is 186 MJ lower in the reference scenario. It means that the energy use for fungicide production is a key parameter in this impact category. This sensitivity analysis was also done in the “water scarcity” impact category (Figure S8), as an increased dose of wood vinegar will reduce the use of water for dilution in the pyrolysis and reference scenarios. The trade-off was increased from 5.67 m³ (baseline) to 11.35 m³ (S2).

4. Conclusion

A consequential LCA framework for the evaluation of the environmental performance of a pyrolysis biorefinery was developed and applied to a case study in which primary forestry residues are used. The method presented requires extending the system boundaries in order to consider the counterfactual use of biomass. It also requires to consider marginal technologies (i.e. technologies avoided in the pyrolysis scenario). In ten out of sixteen impact categories, the pyrolysis scenario presented a better environmental performance than the reference scenario in which forestry residues are left on soil to decay. Among others, pyrolysis of PFR was shown as climate-efficient, in particular based on the consideration that 80% of biochar’s carbon is sequestered in soils for at least 100 years. The results highlighted that the environmental performance of pyrolysis can vary according to many factors, including the biomass feedstock supply, the pyrolysis technology, the co-products yields, properties and uses and the identification of marginal technologies. Assumptions such as the C sequestration potential of biochar, the energy consumption of the pyrolysis reactor and the efficient dose of wood vinegar to substitute fungicide have an impact on the results. Therefore, foreground data from pyrolysis, combustion and agronomic experiments are needed to refine LCA models. Finally, techno-economic studies considering all the processes and products included in the LCA system boundary are needed to validate the economic feasibility of pyrolysis.

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