Highlights

Biomass gasification as a key technology to reduce the environmental impact of virgin olive oil production: A Life Cycle Assessment approach

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- A new approach with an integrated gasification plant is proposed for the olive oil sector
- An LCA of the impact of gasification on the olive oil supply chain is performed
- The gasification plant allows a 8.25% reduction in the normalized environmental impact
- The industrial phase becomes a major carbon sink with the integrated gasification plant
- The climate change impact of the olive oil value chain is reduced by 21%

Biomass gasification as a key technology to reduce the environmental impact of virgin olive oil production: A Life Cycle Assessment approach

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Abstract

The olive oil value chain faces nowadays important challenges toward environmental sustainability, both in terms of waste management and energy efficiency improvement. This research work proposes an integrated gasification plant fueled with olive pomace for combined heat and power (CHP) generation and biochar production, which can be installed directly at oil mills. An alternative scenario for olive oil production incorporating the gasification technology was compared to a baseline scenario based on traditional olive oil production. The environmental impacts of producing 1 kg of unpacked virgin olive oil at the farming and industrial phases were estimated for both scenarios by following the Life Cycle Assessment (LCA) methodology under a "cradle-to-gate" approach. The gasification technology applied to the olive oil industry is able to manage all the pomace from the oil extraction process on site, avoiding transportation to pomace oil extraction plants. The proposed gasification plant generates 0.88 kWh of renewable electricity per kg of olive oil and enough heat to abandon the current practice of burning a significant part of the olive pit production. As a result, the alternative scenario contributes to a 8.25% reduction in the normalized environmental impact of olive oil production. In terms of climate change, the environmental impact of the functional unit is reduced from 2.21 to 1.74 kg CO_2 eq. (−21%) and the industrial phase becomes a major carbon sink with −0.51 kg of CO_2 eq. per kg of olive oil. In this regard, the integrated gasification plant is viewed as an attractive option for most olive oil mills to invest in sustainability through waste management and recovery.

Keywords: Olive pomace, Life cycle assessment (LCA), Climate change, Downdraft gasifier, Combined Heat and Power (CHP), Biochar

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1. Introduction

Olive oil production is an ancient practice that shapes the social, economic and environmental activity of many areas surrounding the Mediterranean basin. As a result of the outstanding nutritional properties of olive oil for human consumption, this economic activity is a source of wealth and employment. Despite the numerous benefits of olive oil production, a series of environmental impacts (EIs) are associated to the olive oil value chain. Over the last decades, a growing number of studies are focusing on life cycle assessment (LCA) applied to the olive oil industry. Many LCA studies in the olive grove have taken place in Italy [1–7] or Greece [8, 9], and only a few have provided information about LCA in Spain [10–15]. In particular, Fernández-Lobato et al. [10] performed an LCA of an extensive area of tree crops and different olive oil mills in Andalusia, the largest virgin olive oil producing region within Spain. They developed an inventory for the farming and industrial phases over the period 2015–2020, which can be useful as an average reference for a comparative analysis between different harvests.

There are plenty of studies on LCA applied to the olive oil sector in the scientific literature. However, most of them are focused on the agricultural phase of olive oil production. Only a limited number of scientific publications deal with assessing the environmental performance of biomass conversion technologies applied to the value chain of virgin olive oils (VOOs). Among the different thermochemical conversion processes, biomass gasification is regarded as one of the best available technologies for simultaneous generation of electricity and heat (cogeneration) from by-products of the olive oil supply chain [16–21], since this technology at the same time reduces the amount of undesirable by-products from olive oil production. In gasification processes, a carbonaceous solid fuel such as olive pomace is partially oxidized and converted into a gaseous fuel, typically termed as producer gas or synthesis gas (syngas) [22]. The high-carbon feedstock requires a gasifying agent (air, pure oxygen and/or steam) in order to be gasified as a result of a series of chemical reactions requiring heat (endothermic) and releasing heat (exothermic). Gasification with air as gasifying agent is usually performed under autothermal conditions, which means that the exothermic combustion reactions release enough heat for the endothermic reactions responsible for the producer gas formation to occur. The producer gas from gasification, once cooled and cleaned, can be used as fuel in gas engines or microturbines for electric and/or thermal power generation. In addition to the distributed electricity and heat generation for self-consumption by olive oil mills, another economically and environmentally valuable product of the gasification technology is biochar, a carbonaceous solid material with numerous benefits for the agricultural soil [23–27].

A few number of works have evaluated the environmental performance of thermochemical or biochemical conversion technologies applied to the by-products of the olive oil value chain through the LCA methodology. In particular, Parascanu et al. [13] performed an environmental assessment of olive pomace valorization through two different thermochemical processes for power generation, namely, combustion and gasification. They applied the LCA methodology under a “cradle-to-gate” approach using the software SimaPro 8.2 and considering 1 MJ of energy as functional unit . For their comparative assessment, the combustion and gasification plants were assumed to be located in the proximity of an olive pomace oil extraction plant, and thus, disregarding biomass transportation. In both scenarios, the conventional Rankine cycle operating with water/steam as working fluid was used for power generation in a steam turbine (the turbine inlet temperature was 500 °C and the pressure ratio was 20). The authors concluded that from the point of view of the environmental performance, the combustion process outperformed the gasification process, with the Rankine cycle as the major contributor for all the impact categories they assessed. The reason is that the efficiency of biomass gasification is below that of combustion, with conversion efficiencies up to around 85% [22]. As a steam turbine was proposed in both scenarios for power generation, the gasification process requires a larger quantity of inputs for generating 1 MJ, and hence, their greater EI. However, biomass gasification has a much wider potential for application, since it allows processing raw materials with higher ash and moisture content. Furthermore, gasification generates substantially smaller emissions of particulate matter, carbon dioxide and even nitrogen oxides into the atmosphere, since the gasification temperature is considerably lower than that of combustion [22]. Last but not least, the producer gas from gasification is a gaseous fuel that could be used more efficiently to power a gas engine or a gas turbine with a better environmental performance than that of a conventional Rankine cycle. For that reason, a spark-ignition gas engine with waste heat recovery from the exhaust gases and the cooling water is proposed as power generation unit in the present work.

In a different work, Parascanu et al. [14] carried out an LCA of olive pomace valorization through pyrolysis under a "cradle-to-gate" approach and considering 100 kg of olive pomace as functional unit . The EIs associated with three different stages were evaluated: olive production, olive oil extraction and pyrolysis of olive pomace. They reported that the main factor influencing all the impact categories was electrical energy consumption [28]. In this regard, electricity self-consumption is viewed as the pathway to reduce the EI of the industrial phase in the olive oil value chain.

El Hanandeh [29] presented an LCA of five different alternatives for energy recovery of olive oil industry wastes in Australia. As claimed by the author, this paper was the first to evaluate options for energy utilization of olive solid waste using the LCA methodology and compare it to industry current best practices. The options for energy recovery included manufacturing briquettes as solid fuel for home heating; pellets for domestic or industrial water heating; pyrolysis and composting. The functional unit was the processing of 1 Mg of two-phase olive solid waste or herein referred to as wet olive pomace. The pyrolysis process performed well in the LCA and avoided EI in all impact categories except for acidification potential and photochemical oxidant formation potential. In this comparative study, pyrolysis scored in third place or better in six impact categories and was regarded as one of the alternatives that were most likely to outperform the current best practices.

Christoforou and Fokaides [30] performed an LCA on torrefaction of olive husk, a three-phase olive mill waste with a moisture content of 45% (wet basis). The EI of olive husk torrefaction at temperatures within the range of 200–300 °C was estimated using the software GaBi with one tonne of torrefied olive husk as functional unit . A series of alternative scenarios were examined regarding the thermal energy source applied to the system and considering transported and on site available raw material. Their results highlight the importance of the drying stage and the potential improvement of the olive husk torrefaction process in terms of energy consumption.

Finally, Batuecas et al. [7] compared the EIs of two alternative scenarios for olive pomace management in olive oil production through the LCA methodology. The two alternative scenarios were: (I) Anaerobic digestion and (II) Disposal on soil. Anaerobic digestion is a biochemical conversion process for energy recovery that involves biogas production with high methane content from the wet olive pomace, while disposal on the soil with no preliminary treatment represents the most widespread practice. A cradle-to-gate approach was adopted and 1 L of extra virgin olive oil was used as functional unit in both scenarios. The LCAs were carried out using Simapro 8.5.2 with the Ecoinvent database V3.4, in accordance with the principle of ISO standards. Their results revealed that the highest impact are produced in the cultivation and harvesting phase for both scenarios, while anaerobic biodigestion allows a significant reduction in the EIs with respect to disposal on soil. In particular, the climate change impact category was reduced by about 45%.

There are countless theoretical, experimental or simulation works on thermochemical and biochemical conversion of biomass in the scientific literature. However, as discussed above, very little information can be found specifically on the environmental performance and life cycle assessment of thermochemical conversion technologies, such as biomass gasification, in the olive oil sector. Thus, the primary objective of the present work is to evaluate the environmental effects and implications of incorporating the gasification technology in the EIs of olive oil production under a "cradle to gate" perspective, considering 1 kg of VOOs as functional unit. The EIs of the current and most representative scenario in many areas worldwide, including the overwhelming majority of Spain and many other Mediterranean regions, which is characterized by traditional olive cultivation in the farming phase and two-phase extraction processes in the industrial phase, are compared to those of a novel approach including an integrated gasification plant for combined heat and power (CHP) generation and biochar production. As discussed above, only a limited number of works are focused on the environmental performance of biomass conversion technologies applied to the value chain of virgin olive oils (VOOs). In this regard, Table 1 places the scope of the present work in the context of other related works. Nonetheless, it is noteworthy that the results from this work are not limited to a single region or country, but rather they can be extended to any existing VOOs value chain based on the two-phase extraction process.

Table 1: Scope of the present work in the context of other related works in the olive oil sector.

	Present work	Parascanu et al. [13]	Parascanu et al. [14]	El Hanandeh [29]	Christoforou et al. [30]	Batuecas et al. [7]
Geographical scope	Andalusia, Spain	Toledo, Spain	Toledo, Spain	Australia	Cyprus	Southern Italy
System boundaries	Cradle-to-gate	Cradle-to-gate	Cradle-to-gate	Cradle-to-gate	Cradle-to-grave	Cradle-to-gate
Functional unit	1 kg of VOOs	1 MJ of energy	100 kg of WOP	1000 t of WOP	1 t of torrefied olive husk	1 L of VOOs
Biomass conversion technology	Gasification	Combustion Gasification	Pyrolysis	Pyrolysis	Torrefaction	Anaerobic digestion
Electric power generation unit	Gas engine	Steam turbine	Not applicable	Not applicable	Not applicable	Not specified

2. Methodology

The methodological approach for this work is divided into four subsections. First, the case study is presented and described in depth. Secondly, the functional unit and system boundaries are established. After having reported the procedure for data collection and the main assumptions, a detailed description on the life cycle inventory of VOOs production is presented. Finally, the LCA method that was selected for application is briefly described and justified.

2.1. Case study

The case study is placed in the context of Spain and comprises three parts. The first part describes the olive farming phase. The second part details the current process of olive oil production and waste management in the industrial phase. The third part proposes a novel approach for the industrial phase incorporating the gasification technology.

In order to comply with the Product Environmental Footprint (PEF) Guide of the European Commission Recommendation 2013/179/EU [31], the LCA has followed the guidelines and quality requirements of the third draft of the Product Environmental Footprint Category Rules for olive oil (PEFCR) [32].

2.1.1. *Farming phase*

The largest olive oil production area in Spain is located in the south of the country, in the region of Andalusia, with Jaén being the province with the highest concentration of olive tree crops and oil mills [33–35]. The most representative value chain is the traditional (or extensive) olive grove, with a density below 150 trees per ha and an olive production over 750 kg per ha. In this crop group, around 67% of the land under cultivation has a slope below 20%. For this reason, olive harvesting is commonly mechanized. In addition, due to the negative impact of the dry climate of the Andalusian hinterland during the hot summer, around 43% of the olive grove has an irrigation system, and in the case of the traditional olive grove, this proportion is reduced to 29%.

The environmental impact (EI) assessment was developed for the traditional tree crop type during the harvests 2017–18, 2018–19 and 2019–20. Regarding the period under consideration, the data quality was at the maximum level according to the PEFCR. The data used to characterize this type of crop were obtained from 81 surveys of different farmers, representing a total of 1,485 ha of traditional olive grove, collecting the information available in the PEFCR to develop an LCA. The information generated was organized and allocated in 4 subgroups with different weights within the traditional group, based on the director plan of the olive grove by the Regional Government of Andalusia [36]: low slope rainfed (43.8%), low slope irrigated (23.3%), high slope rainfed (27.5%) and high slope irrigated (5.4%).

2.1.2. *Industrial phase*

Around 90–95% of the olive oil mills in Andalusia are based on the two-phase oil extraction method [36–38]. This process consists of two successive stages. In the first stage, the olive oil is extracted from the fruit solid material and its moisture content in a horizontally mounted centrifuge. In the second stage, the olive oil is washed with water to remove impurities and contaminants. The washing water is separated from the olive oil with a vertical centrifuge [10]. Olive pits are generated in the extraction process as by-products, which are mostly used as a source of thermal energy. Twigs and leaves are other typical by-products, which are used by farmers for livestock feeding or agricultural amendments.

The main by-product of the extraction process is wet olive pomace (also referred to as two-phase olive mill waste or *alperujo* [37], hereinafter abbreviated as WOP), a thick sludge that is characterized by its high moisture content. WOP must be transported to pomace oil extraction plants in cargo trucks, because its accumulation at oil mills can lead to production stoppages and substantial economic losses [39]. The yield of WOP generated in the oil extraction process typically ranges from 70% to 90% of the weight of raw olives. This residue is composed of a small amount of oils, pits and other non-profitable residues of the olive fruit mixed with large amounts of water (about 60–70% by weight). The main physicochemical properties of olive pomace are reported in Table 2 [40]. After an energy-intensive drying stage, the dried olive pomace (DOP) is eventually subjected to a solid-liquid extraction process with an organic solvent, usually hexane [37]. The products of the extraction process are the crude oil contained in the pomace (2–3% by weight) and exhausted olive pomace (EOP) with a moisture content of about 12% (roughly 34% of the initial weight). The crude pomace oil is finally refined and sold as a by-product with a lower price than that of VOOs. Part of the EOP is typically used as an inexpensive fuel in the drying process, whereas the largest remaining part is sold as fuel for thermal applications [40].

A large part of the excess EOP from olive pomace oil extraction plants is usually purchased by electric power generation companies in the biomass sector. As a result, most of the EOP for sale is eventually transported in cargo trucks to conventional power plants based on the Rankine cycle with water/steam as working fluid. Conventional large-scale power plants are centralized and often require electrical energy to be transported over long distances. The electricity produced in these

Table 2: Physicochemical properties of olive pomace (two-phase extraction process).

Proximate analysis (wt. %)		**Ultimate analysis (wt. %, dry basis)**		**Other properties**	
Moisture (W), ar	60–70 (WOP)	Carbon	51.3	LHV, ar (MJ/kg) with W = 12%	17.5
	10–15 (DOP)	Hydrogen	6.4		
Ash, db	5	Nitrogen	2.0	Ash meting point (°C)	>1000
Volatile matter, db	77.4	Sulfur	0.3	Bulk density (kg/m^3)	780
Fixed carbon, db	17.6	Oxygen (by difference)	35.0	Avg. particle size (mm)	2–5

WOP= wet olive pomace, DOP= dried olive pomace

power plants enters a transmission substation, where large transformers convert the generator's voltage up to extremely high voltages in order to travel more efficiently over the transmission lines. However, transmission and distribution of electrical energy over long distances has remarkable drawbacks, such as line and power conversion losses in the transmission and distribution network, which are originated by the physical effects of the electric current flow and can be divided into Joule losses, losses due to the corona effect and losses on insulators. Line and power conversion losses in the Spanish power grid can amount up to 19% of the power generation output [41].

Primary data were obtained from the surveys of olive oil mills with different structures, sizes and organization types for a period of study spanning the harvests from 2017–18 to 2019–20. A total of 16 questionnaires including real information about the main inputs and outputs of the different extraction processes were obtained to model the main features of the extraction processes. As a secondary stage, the drying process of a typical olive pomace oil extraction plant was modeled in Aspen Plus process simulator according to the information contained in the PEFCR and assuming an initial moisture content of WOP equal to 70% (wet basis). The drying process in most olive pomace oil extraction plants consists of a furnace fueled with natural gas or EOP as a source of thermal energy for drying the WOP feedstock in large-scale continuous rotary drum dryers. As the vast majority of industrial rotary dryers operate in a co-current or parallel flow direction, a co-current rotary drum dryer was considered in this work [42, 43]. The moisture content of the WOP feedstock is largely evaporated into the exhaust gas stream and thus reduced down to an acceptable level for the furnace, namely, 12% on a wet basis. The evaporation process requires a substantial amount of thermal energy because moisture has a high specific heat and enthalpy of vaporization [42]. As a result, the energy-intensive drying stage requires the burning of around 23% of the EOP production in furnaces or even the use of fossil fuels such as natural gas in reciprocating internal combustion engines, leading to large amounts of carbon dioxide emissions into the atmosphere [39]. However, it is noteworthy that the majority of olive pomace oil extraction plants use part of the EOP as fuel for the external furnace. The furnace is assumed to operate with a five percent excess air, which means that air is present in an amount equal to 105% times that required for stoichiometric combustion. Under a conservative approach, the thermal efficiency of the furnace was set equal to 80%. As high-temperature direct-heat rotary dryers typically present thermal efficiencies in the range of 55–75% [42], the thermal efficiency of the rotary drum dryer was selected as 65%, the average value. The inlet temperature of the hot drying exhaust gas to the rotary drum dryer was assumed equal to 600 ºC. The temperature of the exhaust gas decreases drastically along the dryer down to about 100 ºC as its humidity increased, leading to a substantial reduction in the moisture content of WOP from 70% to about 12%, hereinafter referred to as DOP.

2.1.3. Gasification plant

This research work proposes a novel approach for olive pomace management and valorization, where the WOP being continuously produced in oil mills at high rates is directly dried on site, instead of being transported over long distances to olive pomace oil extraction plants for eventual drying. Worthy of note is that in the current management approach for the WOP, a substantial amount of moisture is transported over long distances by truck, leading to considerable economic losses and detrimental EIs for olive oil producers. This novel approach involves installing an integrated gasification plant for combined heat and power generation (CHP) at distributed scale from the WOP being continuously produced at massive rates in oil mills. A schematic process flow diagram of the integrated gasification plant is presented in Fig. 1. The proposed plant consists of a DOP pelletizing machine or pelletizer, a downdraft fixed bed gasifier fueled with DOP pellets, a producer gas cooling and cleaning unit, a spark-ignition engine–generator set for electric power generation, an auxiliary furnace and a co-current flow rotary drum dryer for drying the WOP with the hot exhaust gases leaving the gas engine.

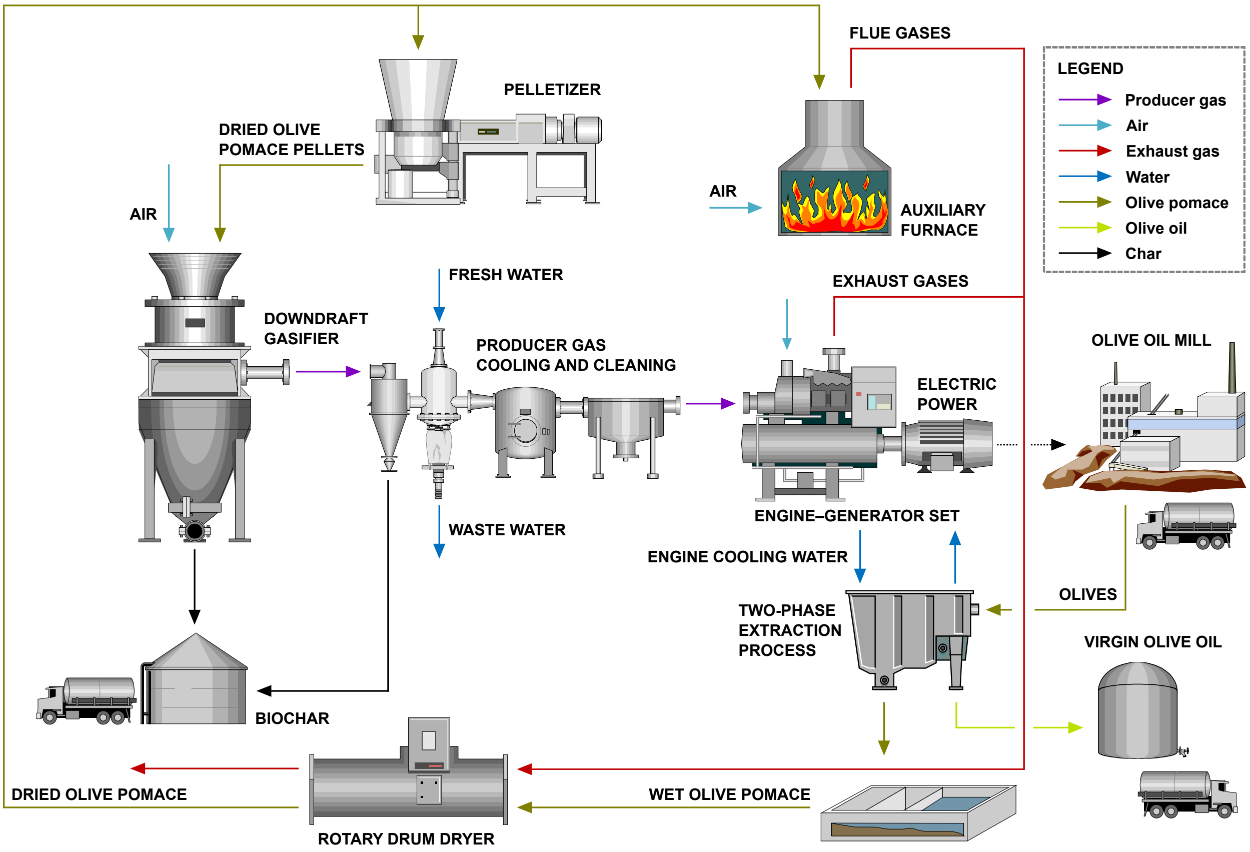

Figure 1: Process flow diagram of the integrated gasification plant for on-site olive pomace management.

The downdraft gasifier is supplied with DOP pellets and air as gasifying agent. Even though gasification with pure oxygen avoids nitrogen dilution, thereby substantially increasing the energy content of the producer gas (LHV = 10–20 MJ/Nm^3) [22], an air separation unit was not considered, because of the extremely high capital and operation costs offsetting any advantage [16, 21]. The downdraft gasifier is operated under autothermal conditions, which means that the feedstock reacts with the gasifying agent and produces enough heat to sustain the high temperatures required for the endothermic reactions to occur. Two product streams are generated in the gasifier, namely, hot producer gas and biochar, which is discharged from the bottom. The direct use of the producer gas from the downdraft gasifier with traces of tar, ash, moisture and other inorganic impurities is not feasible in internal combustion engines, since it compromises their structural integrity [17, 18, 44]. For this reason, a producer gas cooling and cleaning unit must be included downstream of the gasifier, consisting of a cyclone, a Venturi scrubber and a series of filters. After a mild cleaning stage, the producer gas is ready to be used to drive a conventional spark-ignition engine coupled to an electric generator.

Apart from the renewable electricity generation for self-consumption by the oil mill or sale of surpluses, the integrated gasification plant additionally produces two waste heat streams from the gas engine: hot water and hot exhaust gases [18, 20, 45]. The return temperature of the cooling water from the combustion engine is close to 90 °C, so it can be used to supply the hot water demanded by the malaxing stage of the olive oil extraction process [45]. Recycling this residual hot water allows abandoning the current practice of burning a significant part of the olive pit production, which can later be sold at about 0.06–0.08 €/kg [40, 45]. In addition, the required heat for the drying process of WOP is partially supplied by means of the hot exhaust gases leaving the internal combustion engine running on producer gas from gasification, while the remaining thermal requirements are met by burning DOP in an auxiliary furnace. The electrical efficiency (η_e) and CHP efficiency (η_{CHP}) of the integrated gasification plant are respectively given by the following equations:

$$\eta_e = \frac{P_e}{\dot{m}_{dop}\text{LHV}_{dop}} \tag{1}$$

$$\eta_{\text{CHP}} = \frac{P_e + \dot{m}_w c_w \Delta T + \dot{m}_{eg} c_{p,eg} \Delta T}{\dot{m}_{dop}\text{LHV}_{dop}} \tag{2}$$

where P_e represents the electric power; $\dot{m}_{dop}$, $\dot{m}_w$ and $\dot{m}_{eg}$ are respectively the mass flow rates of DOP pellets, cooling water and exhaust gases; c_w is the specific heat capacity of water; $c_{p,eg}$ is the specific heat capacity of the exhaust gases at constant pressure; ΔT stands for the temperature increases and LHV_{dop} is the lower heating value of DOP pellets [18].

Another typical by-product of the downdraft gasification technology is biochar, a carbonaceous solid material with an attractive potential for the olive oil sector. Biochar production accounts for roughly 15–20% by weight of the feedstock to the gasifier and can provide numerous benefits to the olive sector. Due to its extremely high porosity, biochar can retain nutrients and absorb water up to 5 times its own weight [46]. Furthermore, it supports the growth of plants and roots and prevents soil erosion and leaching [47, 48]. Under an environmental perspective, biochar constitutes a long-term carbon sequestration in the olive grove [49]. This is because a considerable fraction of the carbon content that was initially captured by the olive grove is eventually returned to the soil, avoiding its emission back into the atmosphere in the form of carbon dioxide [24]. Under an economic perspective, the markets for biochar products have grown very fast within the recent years and achieved high market volumes. Therefore, biochar also opens many opportunities for new business models [50].

The mass and energy balances of the integrated gasification plant were performed in Aspen Plus® process simulator with the aid of manufacturers specification data. Accordingly, the model of the olive pomace oil extraction plant described in the previous subsection was slightly modified in order to include an additional source of thermal energy to that of the existing furnace. A significant reduction of the amount of DOP to be burned in the auxiliary furnace can be achieved by using the hot exhaust gases from the gas engine fueled with the producer gas from gasification for partially drying the WOP feedstock in a co-current flow rotary drum dryer. The electrical and CHP efficiencies of the integrated gasification plant were estimated at 13.5% and 32%, respectively. A medium-size gasification plant with a nominal electric power of 500 $\mathrm{kW_e}$ was considered as basis for the process simulation, allowing to manage 2.305 kg/h of WOP for cogeneration in addition to biochar production. It is worth noting that olive oil mills with an average production capacity above 1.000–5.000 tonnes/year produce most (55%) of the VOOs in Spain. The size of the gasification plant was deliberately chosen in order to ensure sufficient representativeness, because just above two fifths (42%) of the Spanish olive oil mills are within this range of olive oil production capacity [36]. Table 3 summarizes the main features of the integrated gasification plant.

Table 3: Features of the integrated gasification plant.

Subsystem	**Parameter**	**Value**
Gasifier	Type	Fixed bed, downdraft
	Fuel type	DOP pellets
	Fuel consumption	605 kg/h
	Gasifying agent	Air
Engine	Type	Four strokes, spark-ignited
	Flow rate of cooling water	31.6 m^3/h
	Temperature of cooling water return	~90 ºC
	Flow rate of exhaust gases	2128 Nm^3/h
	Temperature of exhaust gases	~500 ºC
Electric generator	Type	Synchronous, brushless
	Nominal electric power	500 kW
	Electricity generation efficiency	90%
	AC/DC/AC conversion efficiency	97%
Furnace	Fuel type	DOP pellets
	Fuel consumption	180 kg/h
	Excess air	5%
	Thermal efficiency	80%
Dryer	Type	Rotary drum, co-current flow
	Inlet temperature of exhaust gases	~600 ºC
	Outlet temperature of exhaust gases	~100 ºC
	Inlet moisture content of WOP	70% (wb)
	Outlet moisture content of DOP	12% (wb)
	Mass flow rate of WOP	2305 kg/h
	Drying thermal efficiency	65%

2.2. *Goal and scope definition*

Pursuant to the main goal of the LCA, this work aims to characterize and compare the most representative current scenario for the value chain of VOOs production in Spain and a proposed novel scenario incorporating an integrated gasification plant in the olive oil mills. The functional unit chosen for comparison is 1 kg of unpacked VOOs at the oil mill, in accordance with other works [10, 11, 51].

The scope of this work covers the farming and industrial phases of VOOs production under a “cradle to gate” approach. These phases take into account the materials, activities, energy consumption and transport involved in the productive processes, as well as waste management and air, soil and water pollutants. While the upstream processes are related to the inputs and outputs of olive harvesting (farming phase), the core processes include olive oil extraction, generation of by-products and waste management processes (industrial phase).

Two different scenarios are evaluated in this work: A) “Traditional” and B) “Gasification”, the main processes and relations of which are described in Fig. 2. Both scenarios share the main productive processes of the farming phase, with only one difference: Scenario B includes the use of biochar from the gasification process as soil enhancer, instead of an equivalent quantity of products with another origin. The largest differences can be found in the industrial phase: in Scenario A, WOP is transported and treated in a pomace oil extraction plant and, subsequently, the resultant EOP is also transported to a thermal power plant for bulk electricity generation; while in Scenario B, WOP is managed entirely through the integrated gasification plant installed near the oil mill. The by-products generated in the industrial phase of Scenario A are olive pits, crude pomace olive oil and electricity. By contrast, the by-products in Scenario B are olive pits, electricity and biochar. It is important to highlight that different intermediate products are also generated and consumed in Scenario B, producing an additional improvement of the productive process in the oil mill. For example, waste heat in the form of jacket cooling water and exhaust gases from the engine are used in the olive oil extraction process and the pomace management process, respectively. It is also remarkable that, as opposed to Scenario A, Scenario B involves electricity generation at the point of consumption by the oil mill, thereby avoiding power losses in the transport and distribution network. In addition, the net production of olive pits is also slightly higher in Scenario B, as the hot water for the olive oil extraction process is entirely supplied by means of the jacket cooling water from the engine, in contrast to the traditional practice of burning a fraction of the olive pit production, represented by Scenario A.

Two specific questionnaires were elaborated following the PEFCR guidelines to obtain the main primary data for both phases of Scenario A. Qualitative and quantitative information was collected in order to build the Life Cycle Inventory (LCI). The collected data include the surface

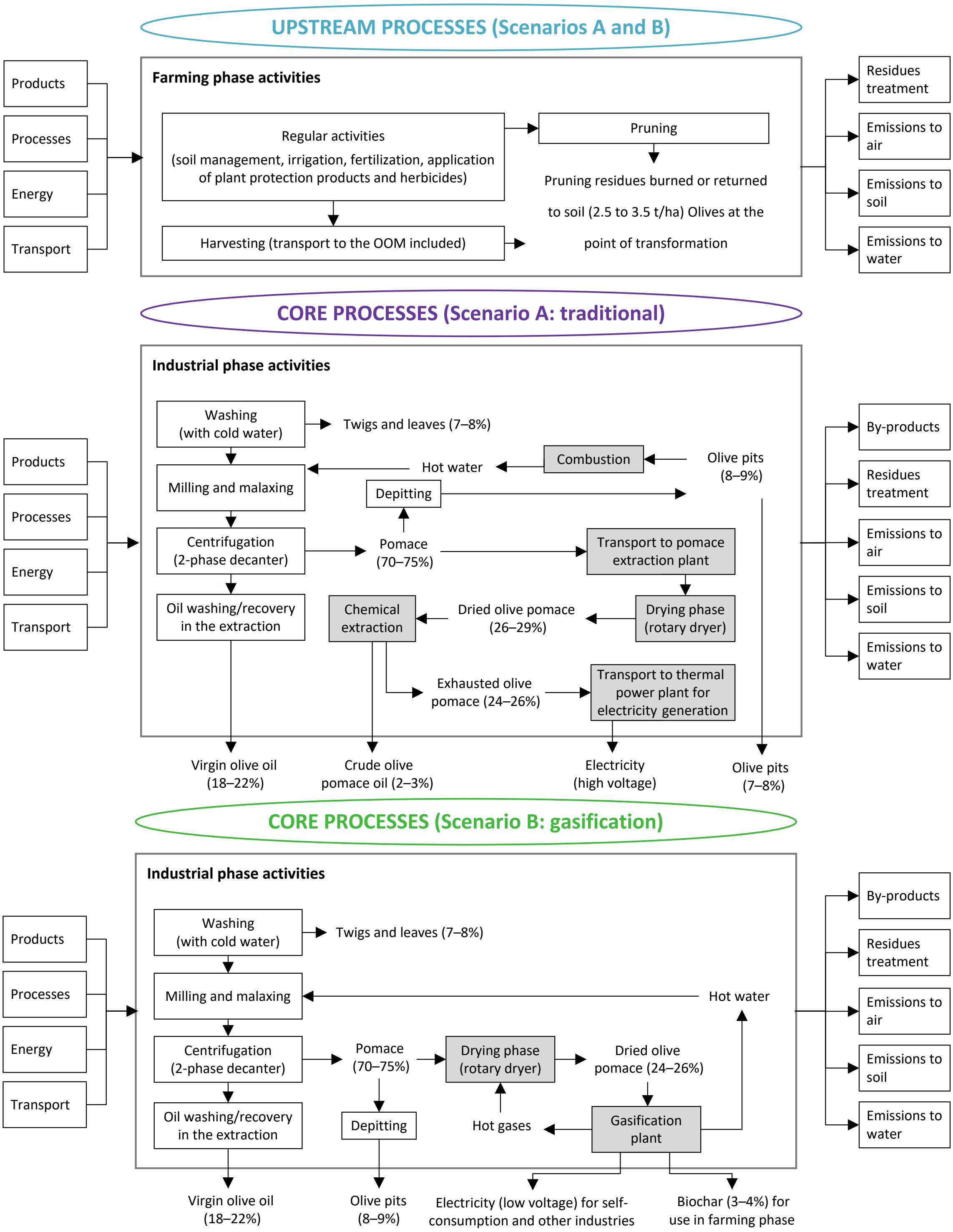

Figure 2: Scope of the scenarios considered for the present work (in brackets, the mass distribution of products in relation to the input mass of olives).

and yearly olive yield according to the EU Regulation (EC) 543/2009. Data from surveys were classified, analyzed and processed following the Data Quality Rating (DQR) contained in the PEFCR, with the purpose of having an "excellent quality" in the PEFCR scale of quality. Values out of range and outliers were discarded. Finally, the primary data were tested before the final version to validate their content with farming experts and experienced researchers in this field.

The targets of the surveys were olive farmers with traditional type plantations older than 25 years and managers of two-phase oil mills with different structures and sizes in Andalusia. As mentioned before, the period of study was 3 consecutive harvests, from 17–18 to 19–20, covering a sufficient time span to be considered representative and in a high quality rating as stated in the PEFCR. Data from the questionnaires were taken similarly to other works [10, 11, 52, 53]: in person or phone call, visiting the corresponding plantation or facilities when it was necessary to oversee the real conditions. The data source for the LCI was essentially the survey; however, official documents and scientific publications served as basis of secondary data, which were used to complete the primary data gaps. Table A.1 shows the sources for all the items considered in the present model.

The EI of VOOs and other by-products generated in the different scenarios were subject to an economic allocation, as reported in the methodological guidelines of the work by Notarnicola et al. [54]. Olive residues and their respective management processes were also considered in the model. The EIs of each process were allocated to the main product in accordance with their total economic value as shown in the next expression:

$$EA = \frac{EV \times M}{\sum_n (EV_n \times M_n)} \times 100 \tag{3}$$

where EA is the economic allocation (%), EV indicates the economic value in euros (EUR) per kg and M refers to the mass of each output (kg) [54].

The products of the olive oil supply chain have been following a downward trend since the

2016–2017 harvest. The prices for VOOs and crude pomace olive oil were reduced from 3.00 to 1.94 €/kg and from 1.41 to 0.68 €/kg, respectively, throughout this period. Prices for olive pits and EOP ranged from 0.056 to 0.073 €/kg and from 0.014 to 0.015 €/kg, respectively [55, 56]. Table 4 shows the main features of the olive farms and oil mills surveyed.

Table 4: Main characteristics of the surveyed olive groves for the farming phase and oil mill for the industrial phase in Andalusia

Farming phase		**Source**
Type of olive farming	Traditional	Survey
Area under irrigation (%)	28.67	[36]
Olive tree density (trees/ha)	80–150	Survey
Total surveyed area (ha)	2,093	Survey
Olive yield range (t/ha)	0.7–9.5	Survey
Average olive yield (t/ha)	3.64	Survey
Industrial phase		**Source**
Average production per oil mill (t)	3,091	Survey
Yearly yield range of VOOs (%)	18.1–22.3	Survey
Average yield of VOOs (%)	20.8	Survey
VOOs price (€/kg)	2.655	[55]
Crude pomace price (€/kg)	1.249	[55]
EOP price (€/kg)	0.017	[56]
Olive pit price (€/kg)	0.063	[56]
Extraction process	Two-phase	Survey

Three recent harvests with their average values of inputs and outputs were considered in the LCI of Scenario A. Generally, only minor variations exist in the values obtained per kg of VOOs between seasons in both phases. Despite this, the agricultural yield shows a greater fluctuation between years mostly attributable to the different weather circumstances of each year and the biological conditions inherent to the olive tree crops. This fact determines the average result of the LCA in the mentioned scenario for the different EIs [5, 52, 57]. Additionally, there is another variation in the industrial phase due to the different productions of VOOs between seasons. As opposed to the farming phase, these characteristics in the industrial phase did not significantly influence the results of the LCA.

Scenario B was elaborated according to the model of the integrated gasification plant described in the previous section with the Scenario A as baseline. Both phases were studied separately and linked by the use of the described functional unit . The data gaps and non-representative values were superseded by the assumptions given in the next section.

2.3. Life cycle inventory

This section provides the qualitative and quantitative information related to the LCI of the VOOs production considering the different activities included in the cycle (shown in Fig. 2). Inputs such as different types of fuel, active ingredients of the main fertilizers and plant protection products, electric supplies and water consumption were considered the most important. The complete list of inputs to the farming phase for both scenarios is shown in Table A.1.

The main activities considered in the farming and industrial phases are those proposed by the PEFCR and reported in [10]. They are composed of harvesting, cutting, PPP & herbicides, soil management, pruning and fertilizing for the farming phase, as shown in Table A.2 with their respective inputs. They were evaluated for the different types of tree crops that belong to the traditional group, while the traditional group was built with the weighted average of them in accordance to their representativeness.

In order to develop the LCA, the following assumptions were made for the farming phase:

- The olive trees that are part of the present study are older than 25 years, and thus, the plantation phase is considered negligible in terms of EI [1, 3].

- The irrigation activity requires water and a substantial electricity consumption that is normally provided by the Spanish electricity grid. The EIs of diesel-powered and photovoltaic systems are not included in the LCI of this activity because their presence is not representative in the current value chain [10].

- The vast majority of the land under cultivation has been occupied by the traditional type of tree crop for over 100 years without changes. Therefore, the land use has remained unchanged in the present work.

- The farming phase activities are responsible for emissions of pollutants to the air, water and soil. These EIs were considered as recommended by the Intergovernmental Panel on Climate Change (IPCC) Guidelines for National Greenhouse Gas Inventories [58].

- Biogenic carbon. Due to the action of the natural carbon cycle, part of the carbon absorbed by the tree is reintroduced into the atmosphere, while another part (mostly leaves, light pruning wood and chopped hard wood) remains stored in the soil under soft techniques of soil management [59–61]. This LCI does not consider the biogenic carbon sequestration originated in the tree, because the soil regime is normally high and it is possible to assume that the largest part of the biogenic carbon generated returns to the atmosphere [62].

- The pruning of the tree crops is regulated and only allowed under authorization. Nonetheless, burning of light biomass residues is still performed in some cases. Most of the pollutants emitted in the pruning process are not representative in the LCA [63]. The carbon dioxide and monoxide emissions from this activity belong to the short carbon cycle, and hence, they are not considered in the LCA either [62].

- The inputs to the crops are transported from 300 km far away, which is an average distance between the main ports of Spain and their destination in the province of Jaén. The average distance between the olive tree crops and oil mills is considered to be 10 km.

- The infrastructure and related inputs and emissions are not considered in the LCI, due to their low impact in the LCA of VOOs production [3].

The activities in the industrial phase of Scenario A are grouped into olive oil extraction and WOP management. The first take place in the oil mill, whereas the second are carried out in the pomace extraction plant and in the thermal power plant. They are described below and their inputs are quantified in Table A.3.

i) Washing of olives with water when they arrive at the oil mill. Olives are separated from the rest of biomass and dust, which can be used as livestock feed or soil amendment, respectively.

ii) Milling and malaxing of olives to obtain an olive paste previously to the oil extraction.

iii) Centrifugation of the olive paste through a two-phase decanter, which requires a high electricity consumption, water and heat to extract the VOOs. The oil extraction process ends with the washing of the VOOs. The remaining mixture of biomass and water becomes WOP, a non-profitable by-product in a first instance, reason why it requires further treatments.

iv) Extraction and depitting to separate the pits from the two-phase WOP by means of a depitting machine. Olive pits have a higher price than that of other biomass by-products of the olive oil value chain.

v) Transport of WOP to the olive pomace oil extraction plant in a cargo truck.

vi) Olive pomace oil extraction and WOP drying in the pomace extraction plant to obtain crude pomace oil and EOP.

vii) Transport of EOP to the thermal power plant in a cargo truck.

viii) Combustion of EOP in the thermal power plant to obtain electricity as definitive by-product.

A series of assumptions were also made in the LCI of the industrial phase in Scenario A. These include:

- The remaining biomass from the washing of the olives is used by farmers as livestock feed or soil amendment.
- About 15% of the olive pits are burned to produce the hot water required for the centrifugation process [40]. These CO_2 emissions only account as biogenic carbon, as olive pits are a renewable biomass source.

- Despite the high amount of WOP produced per kg of VOOs, its moisture content was established as 70% (wet basis) [40].

- Emissions of greenhouse gases during WOP drying management were calculated as reported in the guidelines of Amstel [58], similarly to Figueiredo et al. [64] and Paraskeva and Diamadopoulos [65]. A chemical oxygen demand of 82.5% (90% for phenols and 61% for biochemical oxygen demand) was considered for the evaporation of WOP. Methane (CH_4) emissions are the most harmful in climate change category, they can be generated as a result of natural evaporation in lagoons due to prolonged ambient temperatures above 15 °C, but these circumstances are not usual. The emissions of nitrous oxide (N_2O), another impactful element generated in this treatment, were calculated considering a concentration of nitrogen in WOP of 0.88% and an emission factor of 0.25 [66].

- As for the agricultural inputs, the material inputs consumed in the oil mill are equally transported from a distance of 300 km. Regarding the transportation of intermediate products, the distance between the oil mill and the pomace extraction plant is considered to be 20 km, and from this point to the thermal power plant it has been considered an average of 150 km.

- The only infrastructure considered in the LCI are the oil mill, the pomace oil extraction plants and the thermal power plant, all of them with 50 years of lifetime, as indicated in the PEFCR.

- The amount of electricity generated in the thermal power plant substitutes the same amount of electricity in the Spanish power generation mix. As a result, the renewable electricity generated from the EOP combustion substitutes also the EIs of the current power generation mix in Spain.

The processes and assumptions shown above are used currently in the most representative olive oil value chain of Spain. However, Scenario B is based on additional databases, thereby modifying the model of Scenario A. These new databases correspond to the activities of the gasification plant (Table A.4), which replace activities v) to viii) of Scenario A, while the biochar obtained as by-product is deposited in the agricultural soil of the olive grove (Table 5). Thus, the new LCIs of Scenario B modify the industrial and farming phases, respectively.

In Scenario B, the integrated gasification plant substitutes the current WOP and EOP management in the industrial phase. Accordingly, the new LCI includes all the inputs, outputs and emissions considered in the gasification system. The WOP is entirely managed in the integrated gasification plant, where most of the DOP is consumed by the gasifier and the rest is burned in the auxiliary furnace, contributing to the additional thermal energy requirements for drying the WOP from 70% to 12%. A substantial part of the electricity generated in the integrated gasification plant is consumed by the olive oil mills, avoiding about 85% of the original electric consumption from the Spanish power generation mix during the milling period (from October to March). The excess electricity is injected into the Spanish power grid out of the milling period. An average electricity sale price of 0.05 €/kWh was considered in this work. The cooling water from the gas engine is able to provide about 2,190 MJ of heat per tonne of DOP, which represents a surplus on the thermal energy required in the olive oil extraction process (roughly 1,000 MJ per tonne of VOOs), avoiding the burning of 61.18 kg of olive pits per tonne of VOOs. Additionally, as a by-product of the gasification process, about 164 kg of biochar are produced per every tonne of DOP supplied to the gasification plant. Biochar can be used as a soil amendment to return their carbon and mineral content to the agricultural soil, thereby enhancing or restoring soil functions and fertility [26, 27, 67]. The carbon content of biochar from gasification is around 59% by weight (dry basis). Unlike combustion processes, in gasification processes the carbonaceous feedstock is only partially oxidized, and hence the higher carbon content of biochar. As a result, the CO_2 emissions from gasification are approximately 15% lower than those from combustion of the same amount of DOP [22].

A series of biochar samples from the gasification process were physicochemically characterized in order to determine their proximate and ultimate analyses. About 59% of the sample is made up of carbon, about 1% is nitrogen and another 1% is hydrogen, the rest is composed by about 31% of ashes rich in oxygen, potassium, sodium, calcium, magnesium and silicon. As determined by their proximate analysis, approximately 8% by weight are volatile compounds (H_2O, H, C, CO, CO_2, H_2 and CH_4). The content of soil enhancing elements of biochar is returned to the agricultural soil, avoiding the equivalent amounts of fertilizing products reported in Table 5. Therefore, the inputs are discounted in the same amount as those of other products with a similar purpose and a farther origin. Consequently, transportation of such products in cargo trucks is significantly reduced in terms of tkm, while the use of tractors and trailers is slightly increased according to the values reported in Table 5.

Table 5: Impact of biochar returned to the soil on the LCI of Scenario B (units per hectare).

Activity / Product	
Nitrogen fertilizer (kg)	−1.458
Potassium fertilizer (kg)	−4.785
Calcium carbonate (kg)	−1.120
Magnesium (kg)	−0.781
Transportation, tractor and trailer (tkm)	1.326
Transportation, freight (tkm)	−39.768

The main additional assumptions considered in Scenario B are described below:

- Despite the proved positive effects of biochar in different types of crops such as the increment in agricultural yield and capacity of water retention among other [25, 67–69], these improvements were not considered in this work, due to the lack of quantitative data on its application to the agricultural soil where olive trees grow.
- Biogenic carbon sequestered from biochar into the soil is considered to be removed from the system for over 500 years and therefore, it is not taken into account in the LCA [24, 49, 70].
- The machinery and tools of the gasification plant are included in this work with technical

data from their data sheets, but not the materials, installation and maintenance due to its negligible significance [3, 32].

- According to the assumption in scenario A, the surplus of electricity generated in the integrated gasification plant at the olive oil mill substitutes the same amount of electricity in the Spanish power generation mix, replacing their EIs.

2.4. LCA applied to virgin olive oils production

The LCA application tool used to build the environmental model was SimaPro 9.0 ("System for Integrated Environmental Assessment of Products"), which takes representative values of EI from different databases (ecoinvent 3.5, Agri-footprint 4.0, ELCD, Industry data 2.0, Methods) and provides values for the emissions and effluents of processes. This software was used to manage the LCI databases and analyze the different scenarios through the 2011 ILCD Mid-point+ method [71, 72] to obtain the EIs in 16 different environmental categories. Results are based in the environmental model built with inputs and outputs, which is quantitative and systematic [73, 74]. They were monitored showing the percentage and quantity of EI for every process, inputs and outputs of the functional unit considered in the different EI categories.

The most relevant EI categories in the VOOs production process according to the PEFCR are the following: climate change (without biogenic carbon), water resource depletion, fresh water ecotoxicity, eutrophication, acidification, land use, resource depletion, ozone depletion and photochemical ozone formation. The rest of impact categories obtained in the analysis were included in the results as additional information, with special consideration to the climate change category with biogenic carbon. This category represents the EI including the combustion of different biomass products in the industrial phase.

3. Results and discussion

This section is structured in three parts. The first two parts include the EI results of Scenarios A and B, respectively. The third part presents a comparative discussion on the environmental performance of both scenarios.

3.1. Environmental impacts of Scenario A

The EIs of the different categories in Scenario A (the current most representative value chain in Spain) are detailed in Table 6. The EIs allocated to the most profitable product (VOOs) represents 91.38% of the net profit. The total EIs of each category were distributed between the farming phase and the industrial phase of the supply chain. The EIs of the different activities that make up the farming phase were systematically analyzed in previous works [10, 11], whereas the EIs of the industrial phase were conveniently subdivided into three main activities: olive oil extraction, pomace oil extraction for WOP management and electricity generation in a thermal power plant. It is noteworthy that the last two activities include transport of WOP and EOP for use as feedstock in pomace oil production and electricity generation, respectively.

Table 6: EIs for different impact categories in Scenario A “Traditional".

EI category	Unit	Total	Farming phase	Industrial phase	Olive oil extraction	Pomace oil extraction	Thermal power plant
Climate change (without biogenic C)	kg CO_2 eq.	2.21	2.28	−0.07	0.08	0.03	−0.18
Climate change (with biogenic C)	kg CO_2 eq.	4.48	2.28	2.20	0.18	0.83	1.19
Ozone depletion	kg CFC-11 eq.	$2.07{\cdot}10^{-7}$	$2.20{\cdot}10^{-7}$	$-1.32{\cdot}10^{-8}$	$9.66{\cdot}10^{-9}$	$5.11{\cdot}10^{-9}$	$-2.80{\cdot}10^{-8}$
Human toxicity, non-cancer effects	CTUh	$9.76{\cdot}10^{-7}$	$9.82{\cdot}10^{-7}$	$-6.71{\cdot}10^{-9}$	$6.30{\cdot}10^{-8}$	$1.05{\cdot}10^{-8}$	$-8.02{\cdot}10^{-8}$
Human toxicity, cancer effects	CTUh	$8.42{\cdot}10^{-8}$	$8.32{\cdot}10^{-8}$	$1.02{\cdot}10^{-9}$	$1.33{\cdot}10^{-8}$	$2.81{\cdot}10^{-9}$	$-1.51{\cdot}10^{-8}$
Particulate matter	kg PM2.5 eq.	$1.76{\cdot}10^{-3}$	$1.80{\cdot}10^{-3}$	$-4.46{\cdot}10^{-5}$	$6.65{\cdot}10^{-5}$	$1.86{\cdot}10^{-5}$	$-1.30{\cdot}10^{-4}$
Ionizing radiation HH	kBq ^{235}U eq.	$2.46{\cdot}10^{-1}$	$3.43{\cdot}10^{-1}$	$-9.72{\cdot}10^{-2}$	$4.64{\cdot}10^{-2}$	$6.52{\cdot}10^{-3}$	$-1.50{\cdot}10^{-1}$
Photochemical ozone formation	kg NMVOC eq.	$4.90{\cdot}10^{-2}$	$9.51{\cdot}10^{-3}$	$3.94{\cdot}10^{-2}$	$4.57{\cdot}10^{-4}$	$6.25{\cdot}10^{-3}$	$3.27{\cdot}10^{-2}$
Acidification	molc H^+ eq.	$1.54{\cdot}10^{-2}$	$1.57{\cdot}10^{-2}$	$-3.51{\cdot}10^{-4}$	$7.56{\cdot}10^{-4}$	$2.10{\cdot}10^{-4}$	$-1.32{\cdot}10^{-3}$
Terrestrial eutrophication	molc N eq.	$6.16{\cdot}10^{-2}$	$5.97{\cdot}10^{-2}$	$1.89{\cdot}10^{-3}$	$1.58{\cdot}10^{-3}$	$6.15{\cdot}10^{-4}$	$-3.01{\cdot}10^{-4}$
Freshwater eutrophication	kg P eq.	$4.17{\cdot}10^{-4}$	$4.83{\cdot}10^{-4}$	$-6.59{\cdot}10^{-5}$	$4.03{\cdot}10^{-5}$	$7.04{\cdot}10^{-6}$	$-1.13{\cdot}10^{-4}$
Marine eutrophication	kg N eq.	$4.18{\cdot}10^{-3}$	$4.03{\cdot}10^{-3}$	$1.51{\cdot}10^{-4}$	$1.69{\cdot}10^{-4}$	$5.25{\cdot}10^{-5}$	$-7.10{\cdot}10^{-5}$
Freshwater ecotoxicity	CTUe	147	151	−4	2	1	−7
Land use	kg C deficit	57.9	57.4	0.5	0.6	0.2	−0.3
Water resource depletion	m^3 water eq.	$6.04{\cdot}10^{-2}$	$6.28{\cdot}10^{-2}$	$-2.43{\cdot}10^{-3}$	$-7.78{\cdot}10^{-4}$	$-9.01{\cdot}10^{-5}$	$-1.56{\cdot}10^{-3}$
Mineral, fossil & ren. resource depletion	kg Sb eq.	$2.75{\cdot}10^{-4}$	$2.29{\cdot}10^{-4}$	$4.56{\cdot}10^{-5}$	$4.36{\cdot}10^{-5}$	$7.10{\cdot}10^{-6}$	$-5.12{\cdot}10^{-6}$

Rows shaded in light gray indicate the most relevant EI categories in olive oil production according to PEFCR [32].

The total EI of the climate change impact category is 2.21 kg CO_2 eq. (without biogenic carbon). The farming phase contributes to the emission of 2.28 kg CO_2 eq., while the industrial phase is overall a weak carbon sink with approximately −0.07 kg CO_2 eq., as a result of centralized renewable electricity generation in a conventional thermal power plant fueled with EOP. By contrast, if biogenic carbon is taken into account, the climate change EI substantially increases up to 4.48 kg CO_2 eq., mostly due to the burning of olive pits and EOP in the industrial phase, which is responsible for the emission of 2.20 kg CO_2 eq. The climate change EI of the oil extraction process is 0.08 kg CO_2 (without biogenic carbon), produced mainly by electricity consumption. By contrast, considering the biogenic carbon emissions from the burning of olive pits to supply the hot water required by the malaxing stage of the olive oil extraction process, the climate change EI increases to 0.18 kg CO_2 eq. The climate change EI of the olive pomace oil extraction plant is only 0.03 kg CO_2 eq. (without biogenic carbon). However, if the biogenic carbon is considered, this EI increases drastically up to 0.83 kg CO_2 eq. due to the CO_2 emissions derived from combustion of large amounts of EOP in furnaces in order to supply the heat required for the drying process of WOP. Regarding the thermal power plant, the contribution of only fossil sources of EI on CC is -0.18 kg CO_2 eq. due to the generation of renewable electricity that substitutes the fossil electricity in the Spanish power generation mix. However, if biogenic carbon is considered, the climate change EI rises up to 1.19 kg CO_2 eq.

3.2. Environmental impacts of Scenario B

The EIs of the different impact categories in Scenario B (the proposed novel scenario with an integrated gasification plant) are reported in Table 7. The EI allocated to the most profitable product (VOOs) in this case has increased its share up to 97.24%, due to the absence of olive pomace oil as a by-product. The total EIs of each category were distributed between the farming phase and the industrial phase of the supply chain.

The difference in the EI of the farming phase is mainly due to the reincorporation of biochar into the agricultural soil, providing a series of nutrients and avoiding the use of a same proportion of soil enhancers manufactured and transported from elsewhere. The application of biochar into the agricultural soil, in addition to promoting plant growth, constitutes a long-term carbon sequestration [49, 68, 70]. The high carbon content in biochar is equivalent to the difference in CO_2 emissions between gasification and combustion.

Table 7: EIs for different impact categories in Scenario B “Gasification".

EI category	Unit	Total	Farming phase	Industrial phase	Olive oil extraction	Gasification plant
Climate change (without biogenic C)	kg CO_2 eq.	1.74	2.25	−0.51	0.08	−0.59
Climate change (with biogenic C)	kg CO_2 eq.	3.92	2.25	1.67	0.08	1.59
Ozone depletion	kg CFC-11 eq.	$1.86\cdot10^{-7}$	$2.16\cdot10^{-7}$	$-3.05\cdot10^{-8}$	$9.66\cdot10^{-9}$	$-4.01\cdot10^{-8}$
Human toxicity, non-cancer effects	CTUh	$9.24\cdot10^{-7}$	$9.68\cdot10^{-7}$	$-4.40\cdot10^{-8}$	$6.30\cdot10^{-8}$	$-1.07\cdot10^{-7}$
Human toxicity, cancer effects	CTUh	$7.32\cdot10^{-8}$	$8.12\cdot10^{-8}$	$-7.98\cdot10^{-9}$	$1.33\cdot10^{-8}$	$-2.13\cdot10^{-8}$
Particulate matter	kg PM2.5 eq.	$1.64\cdot10^{-3}$	$1.77\cdot10^{-3}$	$-1.30\cdot10^{-4}$	$6.65\cdot10^{-5}$	$-1.96\cdot10^{-4}$
Ionizing radiation HH	kBq ^{235}U eq.	$1.94\cdot10^{-1}$	$3.38\cdot10^{-1}$	$-1.44\cdot10^{-1}$	$4.64\cdot10^{-2}$	$-1.90\cdot10^{-1}$
Photochemical ozone formation	kg NMVOC eq.	$9.83\cdot10^{-3}$	$9.39\cdot10^{-3}$	$4.41\cdot10^{-4}$	$3.65\cdot10^{-4}$	$7.61\cdot10^{-5}$
Acidification	mole H^+ eq.	$1.39\cdot10^{-2}$	$1.53\cdot10^{-2}$	$-1.48\cdot10^{-3}$	$7.56\cdot10^{-4}$	$-2.24\cdot10^{-3}$
Terrestrial eutrophication	mole N eq.	$5.90\cdot10^{-2}$	$5.84\cdot10^{-2}$	$5.84\cdot10^{-4}$	$1.58\cdot10^{-3}$	$-9.94\cdot10^{-4}$
Freshwater eutrophication	kg P eq.	$3.70\cdot10^{-4}$	$4.74\cdot10^{-4}$	$-1.04\cdot10^{-4}$	$4.03\cdot10^{-5}$	$-1.44\cdot10^{-4}$
Marine eutrophication	kg N eq.	$3.97\cdot10^{-3}$	$3.91\cdot10^{-3}$	$5.91\cdot10^{-5}$	$1.69\cdot10^{-4}$	$-1.10\cdot10^{-4}$
Freshwater ecotoxicity	CTUe	144	151	−7	2	−9
Land use	kg C deficit	57.3	57.2	0.1	0.6	−0.5
Water resource depletion	m^3 water eq.	$5.92\cdot10^{-2}$	$6.19\cdot10^{-2}$	$-2.68\cdot10^{-3}$	$-7.78\cdot10^{-4}$	$-1.90\cdot10^{-3}$
Mineral, fossil & ren. resource depletion	kg Sb eq.	$2.54\cdot10^{-4}$	$2.18\cdot10^{-4}$	$3.53\cdot10^{-5}$	$4.36\cdot10^{-5}$	$-8.26\cdot10^{-6}$

Rows shaded in light gray indicate the most relevant EI categories in olive oil production according to PEFCR [32].

The EIs of the industrial phase in Scenario B were divided into three main sub-processes: olive oil extraction, gasification of DOP pellets and burning of DOP pellets in an auxiliary furnace. The climate change EIs with biogenic carbon significantly increase with respect to those of Scenario A, due to the increase in the total consumption of DOP pellets, despite the substantially lower emissions of CO_2 eq. per kg of DOP pellets. In the industrial phase, the extraction process no longer requires the burning of olive pits for supplying the hot water and thus, the climate change EI with and without biogenic carbon share the same value. In the gasification process, the electric power generation is used for self-consumption of the whole industrial phase with surpluses of electricity discharged into the electric grid, thereby reducing the use of the same amount of electricity from the Spanish power generation mix. Another important point to be raised here is the effect of future changes to the current power generation mix in Spain; for example, a shift toward renewable energy sources may reduce the environmental benefits from the utilization of biomass by-products as energy sources in the future. An increase in emissions due to the gasification of DOP pellets is observed if the climate change EI with biogenic carbon is considered. The furnace aims to supply the thermal energy required for the drying process of WOP by combustion of a significant part of the DOP. The biogenic CO_2 emissions derived from the combustion process, among other EIs, are also included in Table 7.

3.3. Improvement of the environmental performance

As the environmental indicators of each impact category have diverse units and orders of magnitude, a convenient comparison of the environmental performances between Scenario A and Scenario B can be performed by means of a radar chart. Fig. 3 displays the EIs of each impact category on a radar chart where the axes for each EI category are represented in decimal logarithmic scales. It can be observed that the EIs of nearly all impact categories are improved in Scenario B with respect to Scenario A. In particular, the EIs of climate change, ionizing radiation and photochemical ozone formation present the largest differences in terms of environmental performance.

Fig. 4 shows the relative changes in environmental performance of Scenario B with respect to Scenario A for each impact category. As clearly observed, an integrated gasification plant for cogeneration and biochar production reduces most of the EIs derived from olive oil production. The total reduction in climate change reaches 0.47 kg CO_2 eq. per kg of VOOs (−21%). This means that the gasification plant of Scenario B applied to the target oil mills with an average production capacity between 1.000–5.000 t/year, which represents the majority (55%) of the VOOs production in Spain [36], allows achieving a reduction of about 360,000 tonnes of CO_2 eq. (considering the average VOOs production of the three consecutive harvests under study, namely, 2017–18, 2018–19 and 2019–20 [75]). In other words, the EI reduction in the climate change impact category is equivalent to the CO_2 emissions of 220,000 vehicles according to the latest survey of the Spanish National Institute of Statistics (INE) [76] or the CO_2 emissions of 65,200 inhabitants in Spain, calculated from the per capita CO_2 emissions in 2018 [77].

Apart from that, the economic product allocation is affected by the loss of a valuable by-product such as olive pomace oil, which curbs the EI reduction. Although most EIs are improved in Scenario B, considering the new economic product distribution of VOOs, the EI reductions of all the impact categories under consideration are partially offset by the loss of olive pomace oil as a

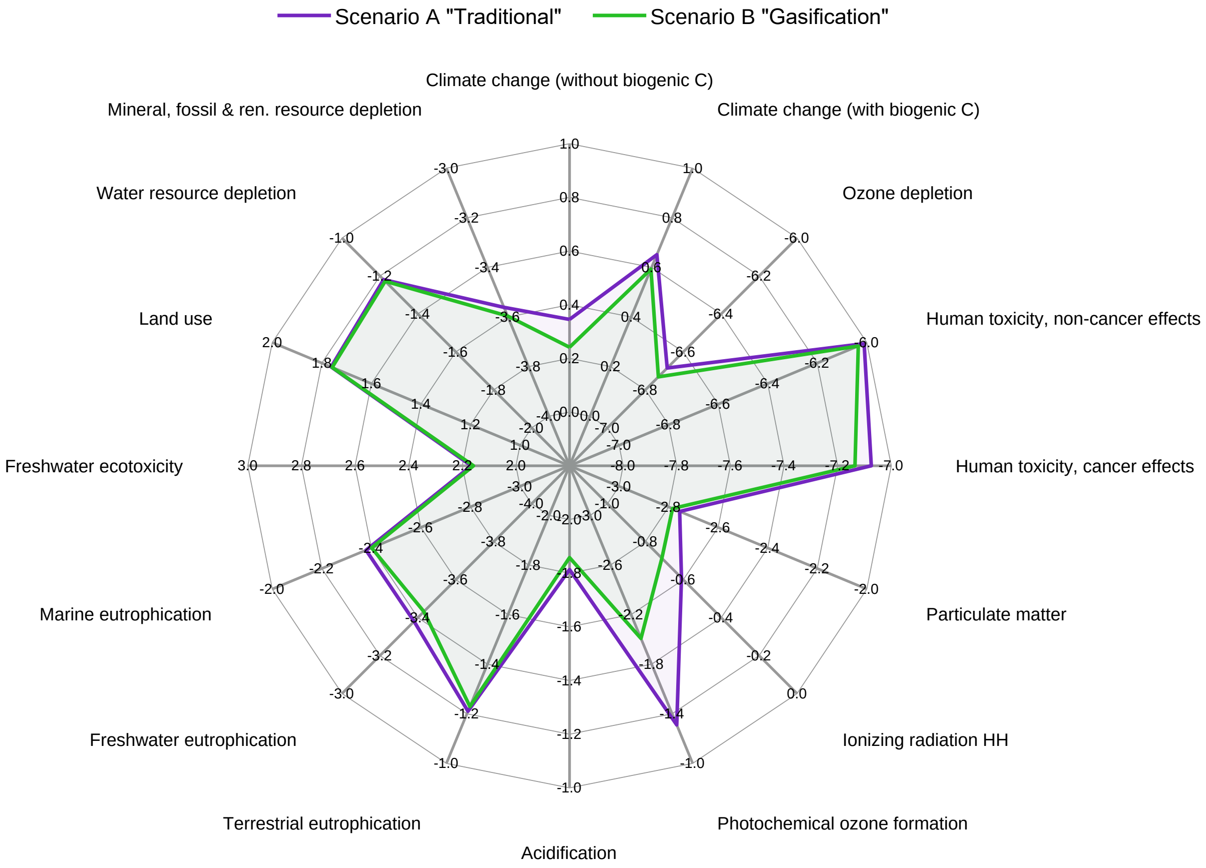

Figure 3: Radar chart of the environmental performances of Scenario A and Scenario B. The EIs of each impact category are represented in decimal logarithmic scales.

profitable by-product. As displayed in Fig. 4, the total EIs of all impact categories have improved in Scenario B with respect to Scenario A. However, with the new economic product distribution in Scenario B, only four impact categories present minor increases in the EIs allocated to VOOs, namely, freshwater ecotoxicity, terrestrial eutrophication, land use and water resource depletion.

In order to compare the EIs of different impact categories with each other in terms of their relative importance, they must be normalized in advance. Normalization provides the benefits of placing the EIs in a broader context [78]. The normalized EIs are calculated as the EIs of each impact category divided by a reference value according to Eq. (4).

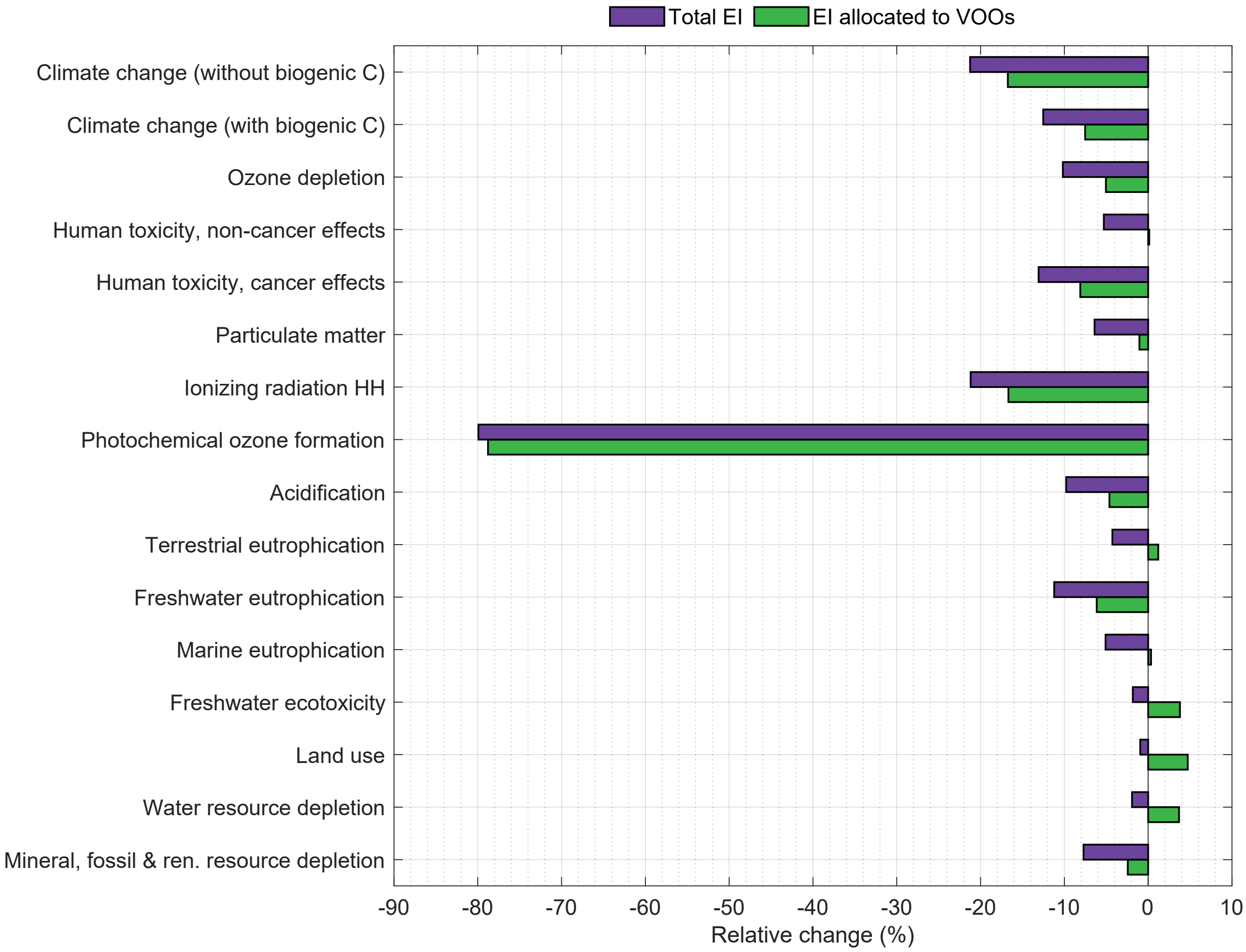

Figure 4: Improvement of environmental performance.

$$N_k = \frac{S_k}{R_k} \tag{4}$$

where k denotes the impact category, N is the normalized EI, S is the impact category indicator from the characterization phase and R is the reference value or normalization factor. As impact categories and their corresponding reference values have the same units, normalized EIs are dimensionless.

The normalized EIs of each impact category for both scenarios (A and B) are displayed in Fig. 5. The most affected impact category in both scenarios is freshwater ecotoxicity, distantly followed by resource depletion and human toxicity. Again, all impact categories present a smaller

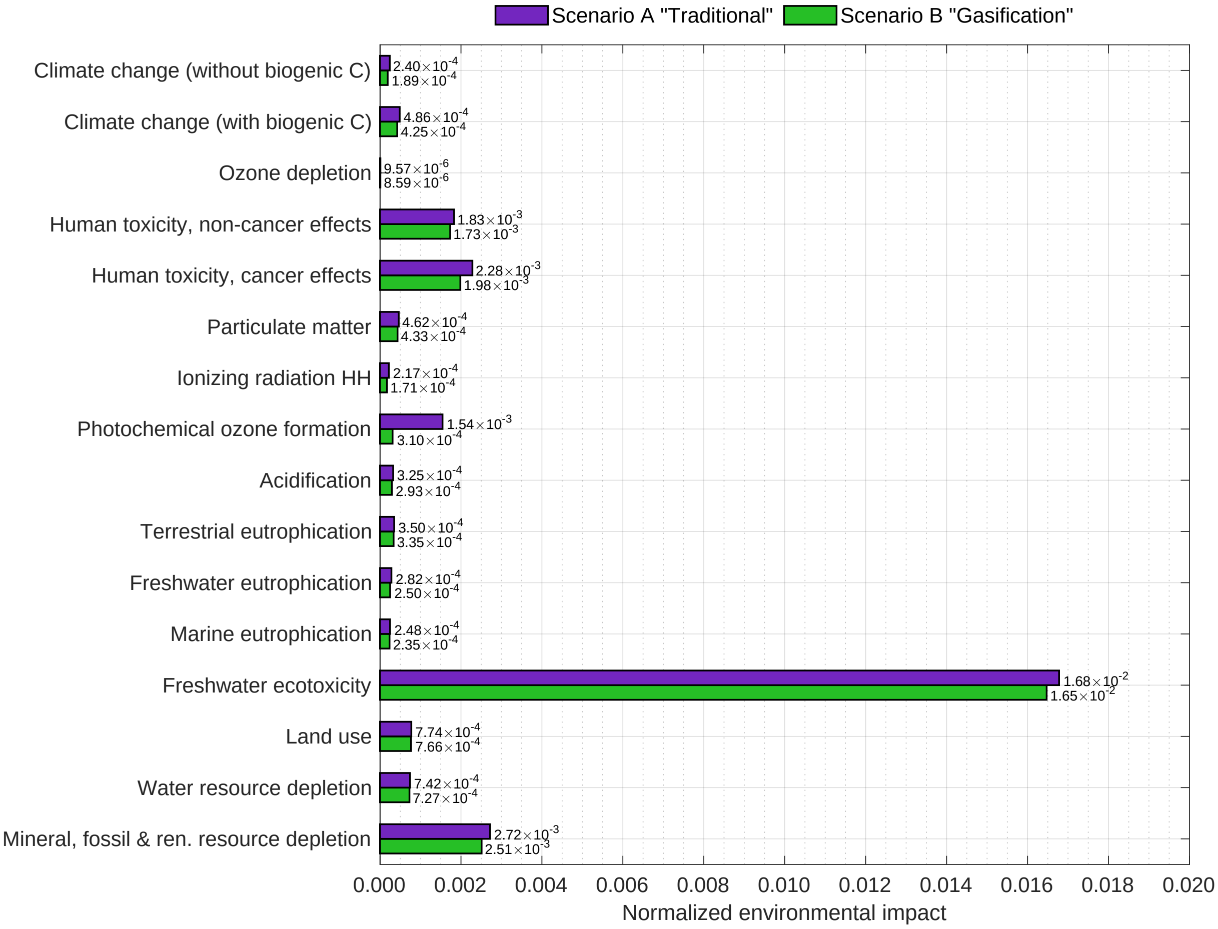

Figure 5: Normalized EIs of each impact category in Scenario A and Scenario B.

normalized EI in Scenario B with respect to Scenario A. Considering all the impact categories except climate change with biogenic carbon, the overall normalized EI is calculated as the sum of the EIs of each category. The overall normalized EI of Scenario A is 0.0290, whereas that of Scenario B is 0.0267. Therefore, the integrated gasification plant in Scenario B allows achieving an overall EI reduction of 8.25% with respect to the baseline Scenario A. After normalization, freshwater ecotoxicity stands out as the most impactful category in the olive oil value chain, the EI of which accounts for 57.8% and 61.8% of the total EI in Scenarios A and B, respectively. Therefore, special attention should be paid to reducing the EI of this impact category.

4. Conclusions

This research work proposes a new approach for the olive oil supply chain with an integrated gasification plant that can be installed directly at oil mills. The gasification plant is aimed at combined heat and power (CHP) generation and biochar production from the wet olive pomace produced at massive rates in oil mills. The proposed plant consists of a dried pomace pelletizing machine or pelletizer, a downdraft fixed bed gasifier fueled with dried olive pomace pellets, a producer gas cooling and cleaning unit, a spark-ignition engine–generator set for electric power generation, an auxiliary furnace and a co-current flow rotary drum dryer for drying the wet olive pomace with the hot exhaust gases leaving the gas engine. The electricity generation and CHP efficiencies were estimated at 13.5% and 32%, respectively. The gasification technology applied to the olive oil industry is able to manage all the pomace from the oil extraction process on site, avoiding transportation to pomace oil extraction plants and their waste management processes. The gasification plant allows generating 0.88 kWh of renewable electricity per kg of olive oil and enough heat to stop olive pits consumption in the VOO extraction process. Additionally, the gasifier produces biochar, a valuable by-product in agriculture, which is very useful as soil amendment.

In terms of environmental performance, the EIs of producing 1 kg of unpacked virgin olive oil were estimated by following the Life Cycle Assessment (LCA) methodology under a "cradle-to-gate" approach. The results from the LCA show that the integrated gasification plant leads to a 8.25% reduction in the normalized EI of olive oil production with respect to the baseline scenario representing the current situation in most Spanish olive oil mills. All the impact categories show a reduction in their total EI, with values of improvement up to 80% (i.e., photochemical ozone formation). In terms of climate change, the EI of the functional unit is reduced from 2.21 to 1.74 kg CO_2 eq. (−21%) and the industrial phase of olive oil production becomes a carbon sink with −0.51 kg of CO_2 eq. per kg of olive oil. As evidenced from these results, the environmental performance of the traditional approach for olive pomace management shows considerable room for improvement, as GHG emissions could be substantially reduced through almost complete substitution of fossil energy by renewable energy sources such as bioenergy. Accordingly, the integrated gasification plant is regarded as an attractive alternative for most olive oil mills to invest in sustainability through waste management and recovery. Future research works concerning sustainability in the olive oil sector will explore other innovative approaches for olive pomace management, such as an integrated gasification plant with a microturbine as power generation unit or a combustion plant with an Organic Rankine Cycle (ORC) unit. This work can be useful for researchers, professionals, Non-Governmental Organizations (NGOs) as well as for policy decision-making processes on sustainability and energy planning in the olive sector at different geographical scales.

List of abbreviations

CHP	Combined heat and power
DOP	Dried olive pomace
EF	Environmental footprint
EI	Environmental impact
EOP	Exhausted olive pomace
GHG	Greenhouse gas
LCA	Life cycle assessment
LCC	Life cycle costing
LCI	Life cycle inventory
PEF	Product environmental footprint
PEFCR	Product environmental footprint category rules
VOO	Virgin olive oil
WOP	Wet olive pomace

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CRediT authorship contribution statement

L. Fernández-Lobato: Data curation, Formal analysis, Investigation, Methodology, Writing – Original Draft, Writing – Review & Editing. **R. Aguado:** Formal analysis, Investigation, Methodology, Visualization, Writing – Original Draft, Writing – Review & Editing. **F. Jurado:** Resources, Supervision. **D. Vera:** Conceptualization, Investigation, Funding acquisition, Project administration, Supervision, Validation, Writing – Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendices

Table A.1: Data source of the items in the Life Cycle Inventories.

Farming phase		**Industrial phase**	
Activity / Product	Data source	Activity / Product	Data source
Harvesting		Olive oil extraction	
Petrol, two-stroke blend (kg)	Survey	Olives (kg)	Survey
Transportation, tractor and trailer (tkm)	Survey	Electricity, low voltage (kWh)	Survey
Polyethylene, linear low density (kg)	Survey	Water (m^3)	Survey
Cutting		Olive pits (kg)	Survey
Petrol, two-stroke blend (kg)	Survey	Transportation, tractor and trailer (tkm)	Survey
Lubricating oil (kg)	Survey	Petrol, two-stroke blend (kg)	Survey
Irrigating		Lubricating oil (kg)	Survey
Electricity, low voltage (kWh)	Survey	Soap (kg)	Survey
Water (m^3)	Survey	Sodium perborate, powder (kg)	Survey
Polyethylene, linear low density (kg)	Survey	Area of oil mill dedicated with an expected with an expected lifetime of 50 years (m^2)	Survey
Polyethylene, high density (kg)	Survey	Pomace treated (kg)	Survey
Polyvinyl chloride (kg)	Survey	Crude pomace olive oil extraction	
PPP & Herbicides		Exhausted pomace (kg)	Aspen Plus model
Application of PPP (ha)	Survey / PEFCR	Electricity, low voltage (kWh)	PEFCR
Water (m^3)	Survey / PEFCR	Water (kg)	PEFCR
Insecticide (kg)	Survey	Transport, freight, lorry (tkm)	PEFCR
Fungicide (kg)	Survey	Hexane (kg)	PEFCR
Herbicide (kg)	Survey	Dedicated portion of pomace oil mill (u)	PEFCR
Polypropylene (kg)	Survey	By-products generation	
Polyethylene (kg)	Survey	Olive pits (kg)	Survey
Transportation, truck 7.5–16 t (tkm)	Survey	Exhausted pomace (kg)	Aspen Plus model
Transportation, tractor and trailer (tkm)	Survey	Crude pomace olive oil (kg)	Survey / PEFCR
Soil management		Residues generation	
Harrowing (ha)	Survey	Water evaporated from pomace (kg)	Vera et al. [40]
Tillage (ha)	Survey	Wastewater from cleaning (kg)	Survey
Ploughing (ha)	Survey		
Mowing, by rotary mower (ha)	Survey		
Transportation, truck 7.5–16 t (tkm)	PEFCR		
Occupation, permanent crop (ha)	PEFCR		
Pruning			
Transportation, tractor and trailer (tkm)	Survey		
Agricultural machinery (kg)	PEFCR		
Fertilizing			
Fertilizing, by broadcaster (ha)	Survey		
Nitrogen fertilizer (kg)	Survey		
Potassium fertilizer (kg)	Survey		
Phosphate fertilizer (kg)	Survey		
Borax (kg)	Survey		

Table A.2: Farming phase LCI for the traditional tree crop types in Spain.

Type	**High slope**		**Extensive**		**Traditional**
Subtype	Rainfed	Irrigated	Rainfed	Irrigated	Mix
Representativeness	19.5%	3.8%	31.0%	16.5%	70.8%
Olive yield (kg olives/ha)	2,677	6,000	2,786	5,858	3,644
Activity / Product	**Quantity (units per ha)**				
Harvesting					
Petrol, two-stroke blend (kg)	36.5	28.7	14.3	13	20.9
Transportation, tractor and trailer (tkm)	14.7	40.3	16.1	43.5	23.4
Polyethylene, linear low density (kg)	1.5	0.5	2.4	1.7	1.9
Cutting					
Petrol, two-stroke blend (kg)	9.3	3.4	1.9	3.2	4.3
Lubricating oil (kg)	3.5	5.1	1.9	0.9	2.3
Irrigating	-	1	-	1	0.3
Electricity, low voltage (kwh)	-	450.8	-	1,670.8	413.6
Water (m^3)	-	569.5	-	1,215.0	313.7
Polyethylene, linear low density (kg)	-	4.2	-	4.1	1.2
Polyethylene, high density (kg)	-	1.9 -	1.9	0.5	
Polyvinyl chloride (kg)	-	3.1	-	3	0.9
PPP & Herbicides					
Application of PPP (ha)	1	1.7	2.4	2.2	1.9
Water (m^3)	0.6	1.7	2	2.4	1.7
Insecticide (kg)	17.1	0.1	1.1	2	5.6
Fungicide (kg)	6.2	5.2	3.8	7.4	5.4
Herbicide (kg)	0.6	0.8	1.2	27	7
Polypropylene (kg)	0.2	0.2	0.1	0.3	0.2
Polyethylene (kg)	7.5	8.7	5.7	12.1	7.9
Transportation, truck 7.5–16 t (tkm)	6.5	7.6	5	10.5	6.8
Transportation, tractor and trailer (tkm)	0	0.7	0.2	0.6	0.3
Soil Management					
Harrowing, by spring tine harrow (ha)	-	-	0.2	1	0.3
Tillage, rotary cultivator (ha)	1	1	0.2	0.3	0.5
Mowing, by rotary mower (ha)	-	-	-	1	0.2
Transportation, truck 7.5–16 t (tkm)	0.1	0.1	0.1	0.1	0.1
Occupation, permanent crop (ha)	1	1	1	1	1
Pruning					
Transportation, tractor and trailer (tkm)	388.3	269.8	105.5	265.4	229.5
Agricultural machinery (kg)	0.1	0.1	0.1	0.1	0.1
Fertilizing					
Fertilizing, by broadcaster (ha)	1	0.8	1.6	0.2	1.1
Nitrogen fertilizer (kg)	28.6	-	15.5	56.7	27.9
Potassium fertilizer (kg)	22	0.2	9.6	49.6	21.8
Phosphate fertilizer (kg)	17.9	-	7.1	33.4	15.8
Borax (kg)	0.1	-	0	0	0
Ammonium sulfate (kg)	115.7	33.5	30.8	-	47.2
Potassium nitrate (kg)	-	18.8	6.1	0.9	3.9
Urea (kg)	0.1	10.7	14.2	9.4	9
Ammonium phosphate (kg)	-	7.8	3	0.4	1.8
Polypropylene (kg)	0.5	0.1	0.2	0.5	0.3
Polyethylene, high density (kg)	18.8	4	6.2	20.8	12.9
Transportation, truck 7.5–16 t (tkm)	163.1	35	53.8	180.6	112.5
Transportation, tractor and trailer (tkm)	0.1	0.1	0.2	0.6	0.2

Table A.3: Industrial phase LCI for the 2-phase extraction system in Spain.

Industrial phase LCI (units per tonne of VOOs)	
Activity / Product	Value
Olive oil extraction	
Olives (kg)	4,820
Electricity, low voltage (kWh)	156
Gas (kg)	-
Water (m^3)	2.02
Olive pits (kg)	61.18
Transportation, tractor and trailer (tkm)	19.57
Petrol, two-stroke blend (kg)	0.01
Lubricating oil (kg)	0.07
Soap (kg)	0.36
Sodium perborate, powder (kg)	0.12
Area of oil mill dedicated with an expected lifetime of 50 years (m^2)	0.03
Pomace treated (kg)	4,065
EOP transported to the thermal power plant (kg)	870
By-products generation	
Olive pits (kg)	347
Electricity generated in thermal power plant (kWh)	675
Crude pomace olive oil (kg)	156
Residues generation	
Water evaporated from pomace (kg)	2,679
Wastewater (kg)	1,818

Table A.4: LCI of Scenario B (units per tonne of VOOs).

Inputs	
DOP for gasification (kg)	1,069
DOP for combustion (kg)	315
Outputs	
Renewable electricity, low voltage (kWh)	820
Heat in engine cooling water (MJ)	2,341
Biochar (kg)	175
Additional olive pits for sale (kg)	61
Emissions	
Biogenic CO_2 from gasification process (kg)	1,358
Biogenic CO_2 from combustion process (kg)	398
Water vapor (kg)	77.75
Oxygen (kg)	80.27
Carbon monoxide (kg)	6.37
Carbon (kg)	1.92
Methane (kg)	1.21
Nitrogen oxides (kg)	0.56
Other hydrocarbons (kg)	0.47
Hydrogen (kg)	0.41