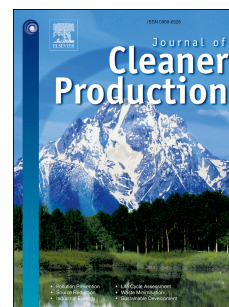


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**Environmental assessment of olive pomace valorization through
two different thermochemical processes for energy production**

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Abstract

In this study, a comparison of olive pomace combustion and gasification through LCA is carried out in order to point out the environmental performance of these processes of these processes to the electrical energy production.

Olive pomace is a by-product from the olive oil industry. The following blocks have been assessed: olive production, olive oil extraction (olive pomace generation) and olive pomace conversion by combustion and gasification processes, respectively. The environmental impacts associated with these stages at mid-point were assessed. In order to obtain a complete profile for the compared scenarios, an end-point level analysis was performed as well. Same data for olive production and olive oil extraction were collected from a Spanish olive mill plant. Thermochemical processes were simulated using Aspen Plus[®] 8.8 software. For a complete perspective, the environmental impact of each equipment involved in the thermochemical processes such as a crusher, combustor / gasifier, cyclone and Rankine cycle were analyzed. Rankine cycle was the major contributor to all impact categories. From environmental and energy point of view, the combustion scenario is the most viable option, considering 1 MJ of energy production as a functional unit.

Keywords: Life cycle assessment, Olive pomace, Gasification, Combustion

1. Introduction

The environmental issues of the contemporary world are mainly caused by the direct and indirect action of anthropological factors. In addition, overpopulation is a current problem in the world due to its effect on the environment (Harte, 2007). The demographic growth is one of the most important issues that lead to increased resources consumption (materials and energy) and the degradation of the environment (Patten, 2014). Pollutants, such as gases and solid particles, from industries and domestic activities, have a negative impact on the environment (Patten, 2014). On the other hand, the consumption of the fossil fuels produces greenhouse gases emissions, which are responsible for the global warming and climate change (Rahman and Miah, 2017).

In recent years, biomass has been ranked among the most important renewable energy sources, with the greatest growth potential in the future. The use of biomass as a source of renewable energy has many advantages, including that biomass is considered as "carbon neutral", being abundant and available in many regions (Abbasi and Abbasi, 2010; Field et al., 2008). It could also reduce the environmental stress by diminishing the dependence of the traditional energy sources and the amount of wastes deposited in landfills and is a raw material for the synthesis of different products as ethanol and similar fuels (Abbasi and Abbasi, 2010; Field et al., 2008).

Olive is mainly cultivated in Mediterranean countries (Spain, Italy, Greece, etc.) but also in other countries from America, Africa, and Australia, where the olive oil is a product of great economic importance (Salomone and Ioppolo, 2012; Tsarouhas et al., 2015). According to the ESYRC of 2016 (ESYRC, 2016), Spain has 2,623,156 ha of olive groves; 152,345 (5.81%) of these are destined to table olives. The region of Castilla-La Mancha in Spain has an area of 415,300 ha of olive groves (ESYRC, 2016),

producing around 111,392 tons of oil in 2015 / 2016, which represents 8.2 % of the national production (1,359,983 tons) (MAPAMA, 2016).

The olive oil industry is a contributor to many environmental problems. The environmental burdens associated with the olive production and the extraction of olive oil are mainly due to the use of resources, the emissions and waste generation (Niaounakis and Halvadakis, 2006; Roig et al., 2006). The olive pomace is the main by-product resulting in the olive oil extraction stage, being an important negative impact in the environment if is not properly disposed or used for fuel production, composting or olive pomace oil extraction.

The main thermochemical conversion processes of the biomass are pyrolysis, combustion and gasification. Energy from biomass can be directly obtained by combustion or indirectly obtained through products that can be assimilated to fuels derived from coal and oil (Arena et al., 2015). In fact, the combustion is the earliest and the most elementary option for energy recovery from biomass. It involves the complete matter oxidation, under an excess of oxygen, to obtain heat for different purposes (Patel et al., 2016). Gasification is a partial oxidation process, which is carried out under oxygen-deficient conditions or in the presence of other gasifying agents such as air or steam allowing the transformation of the biomass into gaseous products, mainly hydrogen and carbon monoxide, but also some carbon dioxide and light hydrocarbons (Patel et al., 2016; Syed-Hassan et al., 2017).

Life Cycle Assessment (LCA) refers to the complete cycle of the product, process or activity, including the extraction and the processing of the raw materials, production, transportation and distribution, use, reuse and maintenance, recycling and final disposal of the product (ISO14040, 2006; Tibor and Feldman, 1996).

Several studies on the LCA associated with the thermochemical conversion of waste generated in the olive industry have been recently reported. Intini F. et al. (2011) investigated the environmental advantages derived from the co-use of the de-oiled pomace (60 %) and waste wood (40 %) in a real combustion plant (located in Italy). El Hanandeh (2015) evaluated the environmental performance of five valorization alternatives: manufacturing briquettes as a solid fuel for house heating, pallets for domestic water heating, pallets for industrial boiler, pyrolysis and composting. Christoforou and Fokaides (2016) evaluated the environmental impact of the olive husk torrefaction process. Rajaeifar et al. (2016) compared the environmental impacts of the olive pomace oil diesel and conventional petroleum diesel taking into account the main stages in the life cycle of fuel (feedstock production, fuel processing and combustion of the fuel).

The main goal of this study is to evaluate the performance associated with the energy production through thermochemical conversion of a specific biomass, which in this study is the olive pomace. In this regard, the combustion and the gasification processes are compared to determine the best alternative for managing the olive pomace. Additionally, the environmental impact analysis performed for each equipment involved in thermochemical processes were also evaluated.

2. Material and methods

2.1. Goal and scopes

The aim of this study is to compare two thermochemical processes (combustion and gasification) in terms of environmental performance, using olive pomace as feed to obtain energy. In this regard, an LCA methodology is used to identify the environmental impact associated with each studied thermochemical conversion process. The life cycle

assessment was carried out in accordance with the cradle-to-gate approach, taking into account the main involved stages: olive production, olive oil extraction, olive pomace combustion process and olive pomace gasification process.

In this study, the LCA was carried out using the SimaPro 8 software (PRéConsultants, 2016). The objective of the olive pomace valorisation through combustion or gasification processes is to produce energy. For this reason, 1 MJ of energy produced was defined as a functional unit (FU).

2.2. System boundaries and assumptions

An important component in the biomass-to-energy chain is to include the previous stages to the production of energy in the limit of the system, such as the production, the pre-processing and the biomass transport (Gold and Seuring, 2011; Nguyen et al., 2013; Patel et al., 2016; Raynolds et al., 2000). This is mainly due to the fact that the environmental impacts of the previous stages affect the results, and, implicitly, the final decision making. The biomass valorisation considered in this work was analyzed from the raw material production (olive production) to the conversion of the olive pomace via the two thermochemical processes pathways to obtain energy as the final product. Therefore, the system boundary selected to perform the LCA of energy production from olive pomace through the combustion and gasification processes is presented in Figure 1.

Several hypotheses are to be considered in the actual approach in order to avoid overlapping in the making-decision process. Planting and tree growth have been omitted due to the long time in which there is no production. Input and output data for one year (2015) has been considered. The transportation of the olive to the olive oil extraction plant and the transportation of the fertilizers has been also considered. It was assumed

that the olive oil extraction plant and the combustion and gasification plants are located in the same place. For this reason, the biomass transportation in this last case is not taken into account. The capital goods such as machinery, equipment, and buildings involved in this analysis, are excluded from the assessment.

2.3. Life cycle inventory analysis

To perform the environmental assessment, a data collection from the inputs and products related to the analyzed processes is required. The Life Cycle Inventory (LCI) is the compilation and quantification phase of all flows (raw materials, energy and others goods and services, emissions, waste and products) related to the production system during its entire life cycle (ISO14040, 2006; ISO14041, 1998). The inventory data, such as the direct inputs and outputs of each stage considered in the boundary system, were collected from a real olive mill plant, the Aspen Plus[®] software and the Ecoinvent database.

In this study, the collection of the main input and output data (raw material, supplement material, final products, waste and some of the emissions) for the olive production and the olive oil extraction were obtained from an olive mill plant *Aceites García de la Cruz* located in Castilla-La Mancha (Toledo, Spain). On the other hand, the air, water and soil emissions associated with the olives production stage were taken from the Ecoinvent 3.4 database (Ecoinvent, 2017).

The combustion and gasification plants were simulated using Aspen Plus[®] 8.8 software (licensed by Aspen Technology, Inc.) to estimate the mass and the energy balances. Therefore, these simulations provided useful information related to emissions, resources and energy consumption.

The main inputs and outputs of the agricultural olives production (block I), olive oil extraction (block II), olive pomace combustion (block IIIA) and olive pomace gasification (block IIIB) are shown in Tables 1, 2 and 3, respectively.

2.3.1. Biomass processing (block I and block II)

Most of the inventory data used for the first two blocks were provided by the real mill plant. These data were processed in order to determine the corresponding values for the functional unit (1 MJ energy produced) as indicated in Table 1.

Olives were considered as a final product in the first stage (olive production – block I), and as a raw material in the second stage (olive oil extraction – block II). Therefore, the area that is taken into account for this evaluation is a plot of 40 ha with an olive yield of 28,736 kg (as based on a 2015 report data). In this study, irrigation is not taken into account, being the rainwater the only form of irrigation. For the season considered in this study, 2,140 m³ / ha of rainwater has been recorded.

The fertilizers used in the olive production stage are supplied by the company Agrogenia Ltd (Córdoba, Spain). The transportation of the fertilizers (285 km) is carried out by a truck belonging to the same company. Two applications per year using 9,000 L of phytosanitary treatment are performed. In this study, fertilizers data have been introduced into SimaPro by using similar products (containing N, K, P and others) available in the Ecoinvent database. The emissions related to the olive production, such as heavy metals in water and soil, but also emissions in the air have been taken from the Ecoinvent (Ecoinvent, 2017). The amount of diesel used for the application of the fertilizers and their transport from Cordoba to the farm is 24.2 L / ha.

The olives collected from the plot are transported to the oil extraction plant using a truck. The distance from the plot to the oil mill is 19 km. At this stage, the main

obtained product is the extra virgin olive oil, with an average yield of 19.14%. Together with the final product (olive oil), olive pomace, olive stone and solid waste (leaves, dust and stones) are produced. Therefore, the amount of extra virgin olive oil, olive pomace, olive stone and solid residues obtained after the olive oil extraction process, considering the whole plot (40 ha), are 5.060 kg, 21.000 kg, 2,299 kg and 376 kg, respectively. The operating regime for the oil extraction process is 24 hours for 3 months with an installed electrical power of 78 kW and a water consumption of 887 m³.

On the other hand, the amount of atmospheric emissions, such as NO_x, SO₂, CO and particulate matter (PM), and wastewater generated by the olive oil extraction process, were also provided by the oil mill plant.

Table 2 shows all the input and output data for the olive oil extraction stage, calculated for 1 MJ of energy produced.

2.3.2. Olive pomace conversion processes

One of the limitations of the Aspen Plus database is that nonconventional products (such as biomass) are not included. For this reason, the olive pomace used as biomass in this study has been simulated through its composition, such as: moisture, fixed carbon, volatile matter and ash (proximate analysis), carbon, oxygen, sulphur, hydrogen and nitrogen (ultimate analysis), and the higher heating value (HHV) (Table 4). For simulation purposes, the stream biomass was used as the feed in both processes (combustion and gasification), which allowed to convert the non-conventional biomass into conventional components. HCOALGEN was the model selected for the enthalpy calculation. Ideal property method was selected for data processing and determination of the thermodynamic properties in the case of gasification process and Peng-Robinson property method was selected for the combustion one.

2.3.2.1.Olive pomace combustion process (block IIIA)

The combustion process was simulated and the flowsheet diagram is shown in Figure 2a. Table 5 shows a brief explanation of each block used for the simulation of the biomass combustion.

It was assumed that after the extraction process of the olive oil the initial moisture content in the olive pomace (54%) was reduced by air drying for 48 hours. The dried biomass was transferred to CRUSHER (Crusher), where biomass was crushed to obtain a particle size of 5 mm. To simulate the combustion process two different reactors (COMBUSTOR) were used. The first reactor, DECOMP (RYield), was used to simulate the release of volatiles whereas the second reactor, BURN (RGibbs), was used to simulate the combustion of the char formed. The product obtained after the combustion process was separated into gas and ash in CYCLONE (Sep 2). It was assumed that all the char obtained in this reactor was 100 % carbon. Finally, the energy was obtained by simulating a Rankine cycle, which was composed of four blocks: heat exchanger, turbine, condenser and pump. The Rankine cycle is used to obtain electrical energy. This energy is generated when the pressure of the steam is reduced. The traditional Rankine cycle is a thermodynamic cycle that uses water as the working fluid, operating conditions being 500 °C and 20 bar (Srinophakun et al., 2001). The gas obtained from the combustion process was fed to the BOILER (Heat X). The resulting steam, at 20 bar and 500 °C, was fed into the TURBINE (Compr) to reduce the gas pressure to 1 bar. The difference between the inlet and the outlet enthalpies is transformed into the outlet energy from the turbine. The resulted steam is then passed to CONDENSER (Heater), where the steam is condensed to become a saturated liquid and, then, is pumped to 20 bar using PUMP (Pump) before being fed to the boiler. In this study, the isentropic

efficiencies for the turbine and the pump were assumed to be 85 % and 65 %, respectively (Liu et al., 2014; Saleh et al., 2007).

2.3.2.2.Olive pomace gasification process (block IIIB)

The flowsheet diagram of the simulated gasification process is shown in Figure 2b. Table 5 shows a brief explanation of each block used for the simulation of the biomass gasification.

Taking into consideration the particularities of the gasification process, several assumptions were established. H_2 , CH_4 , CO , CO_2 , H_2O , NH_3 , HCl and H_2S were considered as gases evolved during the gasification process. Char is composed of carbon and ash, where ash is considered to be inert. 100 % of ash conversion was obtained during gasification. In addition, it was assumed that all the reactions involved in the gasification process reached the equilibrium (Formica et al., 2016; Pala et al., 2017).

The pre-treatment of the biomass in the gasification process is the same as that in the combustion process. Therefore, the biomass was allowed to air dry for 48 hours and then crushed (particle size of 5 mm) to obtain optimum conditions for the gasification process. The resulting stream was fed to reactor DECOMP (RYield), which was used to simulate the release of the volatiles through pyrolysis process based on the conversion of the nonconventional biomass into conventional components. The obtained char (100 % carbon) was split in CHARSEP (Sep 2) and then it was fed to the combustion reactor BURN (RStoic). The aim of the char combustion was to increase the temperature of the bed particles, providing the heat necessary for the reaction in the gasification chamber. In addition, the airflow required for the combustion was determined by either using a calculating block that takes the char burnt in the combustion chamber as a reference and

considers an excess of air of 1.12. The remaining stream after the splitting in separator CHARSEP was then fed to reactor GASCONV (RStoic) where the conversions of nitrogen, chlorine and sulfur contained in the biomass into NH_3 , HCl and H_2S , respectively, are simulated. Separator GASSEP (Sep 2) was used to separate these gaseous compounds from the mainstream, which was fed into reactor GASIF1 (RGibbs). The aim of this block was to simulate the reaction between the biomass char and the gasifying agent which was introduced into the reactor at 1 bar and 150 °C. Reactor GASIF2 (RGibbs) was used to adjust the gas composition. The stream coming from GASIF2 was mixed with that of GASSEP and GASCOMB in mixer GASMIX (Mixer) to obtain a gas, which was separated in cyclone CHARSEP (Sep 2) into ash and syngas. Finally, the energy was computed by simulating a Rankine cycle, which was similar to that of the combustion process.

2.4. Impact assessment methodology

SimaPro software is a professional tool to evaluate the environmental impacts of products, processes and services through their life cycle. It allows to model and analyze the life cycle of a product or service in a systematic and transparent way, following the recommendations of the ISO 14040 series (ISO14040, 2006). The mid-points impacts are considered a point in the chain of cause and effect, focusing on unique environmental problems (e.g.. climate change). The end-point method analyses the environmental impact at the end of this chain of cause and effect (Bare et al., 2000). In the ReCiPe methodology, eighteen mid-point indicators and three more uncertain end-point indicators are calculated (Goedkoop et al., 2009). The conversion of mid-points into end-points simplifies the interpretation of the LCIA results, partly because they are too many impact categories and have a very abstract meaning. In this way, the end-point

approach provides results with a higher degree of interpretation but greater uncertainty. On the other hand, the mid-point approach is more reliable but does not provide damage information (Dong and Ng, 2014).

Due to the advantages and disadvantages of the mid-point and end-point indicators, mid-point and end-point methodologies have been combined in this study. In this way, on the one hand, decisions can be made using mid-point indicators, which are more certain but, in some cases, may have less relevance for decision support. On the other hand, end-point indicators are used, which have been shown to be more relevant and decisions can be made more easily, but have less certainty.

In this study, the ReCiPe Mid-point and End-point were used to determine the environmental performance associated with the energy production. Therefore, mid-point indicators were used to analyze each of the three subsystems presented above, which are based on different impact categories. Thus, the following mid-point impacts were screened: climate change (CC), ozone depletion (OD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), human toxicity (HT), photochemical oxidant formation (POF), particulate matter formation (PMF) and fossil depletion (FD) (Goedkoop et al., 2009).

In addition, for a better understanding, the final point indicators were addressed, analyzing both scenarios (combustion and gasification), taking into account the three stages considered. The following end-point impacts were examined: damage to human health (HH), damage to ecosystem diversity (ED) and damage to resource availability (RA).

The main factors that negatively affect the environment, due to the production of energy from biomass, are the emissions generated along the biomass-to-energy chain. In this way, the CC are influenced mainly by the GHG such as CH₄, N₂O and CO₂ (Patel

et al., 2016). The impact of eutrophication (marine and freshwater) accounts for the environmental persistence of the emission of N and P containing nutrients. The impact values for PMF are due to the particulate matter and different gases generated. Emissions such as SO₂, NO_x and NH₃ (Oreggioni et al., 2017) affect the impact categories of TA. Air emissions and heavy metals, which have direct toxic effects, are responsible for the HT impact (Goedkoop et al., 2009). The values for the FD impact category are determined by the amount of fossil fuel consumed (diesel). On the other hand, transport and energy consumed are responsible for the impact values of almost all the selected categories.

The economical allocation for the olive pomace as a co-product must be used, because it is intended to obtaining energy through a subsequent process (PCR, 2014) and considered to reflect better the value of the products by granting most of the impacts to virgin olive oil, which also has the highest unit price (Schau et al., 2016). In this way, the economic allocation factor for the olive oil is 97.2 % (the price for 1 kg of extra olive oil is € 3.65 / kg); for the olive pomace, it is 1.7 % (the average price for olive pomace is € 15 / ton); and, for the olive stone, it is 1.1% (the price of olive stone is € 90 / ton).

The fact that the normalization results have the same unit for each category of impact facilitates the comparison between the impact scores of different impact categories (Norris, 2001; Sleeswijk et al., 2008). On the other hand, using the normalization value, it is possible to identify easily and faster the impact categories with the highest and lowest contributions that affect the environment, simplifying the final decision making (Mayo et al., 2018; Rowley et al., 2012). Due to these advantages, in this study, the results have been normalized. As defined in ISO 14044, the normalization is a process to calculate the magnitude of the results of impact category indicators, in relation to

certain reference information (ISO14044, 2006). The results for each category of impact are normalized with respect to average European emissions. For this purpose, the characterized results of each impact category are divided by a selected reference value (Goedkoop et al., 2009; PRéConsultants, 2016).

3. Results and discussion

3.1. Environmental assessment of the biomass valorization

The assessed environmental performance of the biomass valorization, taking into account all the evaluated stages and considering the normalization of the data at the mid-point level, is shown in Figure 3. Figure 3a displays the results of the main environmental impacts for the combustion scenario, analyzing the stages of olive production, olive oil extraction and the combustion process. In the same way, Figure 3b shows the main results of the environmental impacts of the gasification scenario.

The results obtained showed that for all the analyzed impact categories the gasification scenario exhibited higher impact values, at mid-point level, than the combustion scenario. This fact is mainly due to the higher amount of olive pomace used by the gasification process to obtain 1 MJ of energy if compared to that required by the combustion one.

Figure 3a shows that, in the case of the combustion scenario, the extraction stage of olive oil has the highest impact in almost all the assessed categories, followed by olive production and the combustion process. This fact is mainly related to the consumption of energy and diesel and the emissions released. A different trend is observed in the case of the HT and POF categories. In this case, the combustion process is the one that more affects them, mainly due to the ash and emissions generated. On the other hand, Figure 3b shows that the gasification process and the olive oil extraction play the leading role

in the impact categories studied for gasification scenario. This fact is related to the consumption of energy and raw material and the generated emissions. Finally, it can be also observed that the impact associated with the gasification process is almost twice as large as that associated with the combustion one.

The emissions (Table 1, 2 and 3) could play an important role in different impact categories (Foteinis and Chatzisyneon, 2016; Wagner and Lewandowski, 2017). Human activities are the main factors that contribute to greenhouse gas emissions, carbon dioxide, methane, chlorofluorocarbons and nitrous oxide and are well known for their global warming potential (GWP) (Houghton et al., 1992; Zhang et al., 2014), which is closely related with the CC impact observed for the olive production stage. The higher GHG emission (CH_4 and CO_2) observed for the gasification scenario if compared to that of the combustion one (Table 6) could explain the higher CC impact of the first one (Figure 3). Although N_2O is a greenhouse gas which is about 300 times worse than CO_2 in terms of the greenhouse effect ($\text{GWP}_{\text{N}_2\text{O}} = 296$; $\text{GWP}_{\text{CO}_2} = 1$) (IPPC, 2006), its presence in traces contributed to a small GWP in comparison with the larger CO_2 emissions. Furthermore, the CO_2 and CH_4 are also responsible for the POF impact category. This impact category and PMF can be also affected by SO_2 and NO_x emissions (Derwent et al., 1996). On the other hand, CH_4 is mainly involved in the OD impact category.

In this study, the nitrogen-based emissions released (NH_3 , NO_x and NO_3) during olive production stage (Table 1) were indirectly responsible for the TA, FE, ME and HT impacts (Brentrup et al., 2004; Goedkoop et al., 2009). In this stage, the phosphorus emissions in water are the main contributors in the FE impact category (Brentrup et al., 2004). Moreover, the SO_2 and the NO_x emissions (Table 2) released during the olive oil extraction stage influence mainly the TA and ME impacts (Goedkoop et al., 2009). In

addition, the HT category of impact includes all the direct toxic effects of human emissions. Therefore, the stage that most affects this impact category is related to thermochemical process (combustion and gasification), due to the ash generated and to the emissions released into the air (Table 3 and 6). The emissions released during the first stage (Table 1), which can potentially have a toxic effect are inorganic air pollutants (NO_x and NH_3), fertilizers and heavy metals (Cd, Pb, Hg, Zn) (Brentrup et al., 2004).

The diesel and energy consumption (Table 1, 2 and 3) are responsible for the FD impact category, but they are also linked to the accumulated demand for natural gas, crude oil and coal, which are necessary for the background processes.

In case of the olive oil extraction and thermochemical processes stages, the high energy demand (Table 2 and 3) contributes significantly to most of the impact categories evaluated (Pattara et al., 2016; Rajaeifar et al., 2014; Rinaldi et al., 2014).

The olives and the fertilizer transport to the oil mill plant contribute to the CC impact category (Koetse and Rietveld, 2009; Zhang et al., 2014). Other impact categories that are affected by transportation are the OD, TA and PMF ones.

The application of fertilizers during the olive production stage is responsible to the value of the HT category (Iribarren et al., 2014; Kleinman et al., 2011; Nayal et al., 2016; Peters et al., 2015b; Sharpley et al., 1990; Uzoma et al., 2015) (Table 1). Furthermore, the high HT value obtained in the olive oil extraction stage could be attributed to the high values of waste and wastewater generated (Table 2).

3.2. Environmental assessment of olive pomace thermochemical conversion

As can be seen in Figures 2a and 2b, the main operations involved in the olive pomace combustion and gasification processes are: biomass crushing, biomass

thermochemical conversion (combustion / gasification), separation of the resulting gas to obtain gas and solid waste (ash) and electricity production through the Rankine Cycle.

Table 7 shows the impact values (normalized) at the mid-point level using the ReCiPe methodology for each equipment involved in the combustion process. In addition, Table 8 shows the aggregated impacts (normalized) according to the ReCiPe Mid-point methodology for each equipment of the gasification process.

RANKINE CYCLE is the major contributor in all impact categories assessed for both processes. This result is mainly attributed to the released gases (Table 6) but it may also be due to the energy required for the water pump operation.

On the other hand, regarding the combustion process, the CRUSHER equipment had impact values quite similar to those of the COMBUSTION and CYCLONE equipment in almost all the impact categories. However, it was observed that, in the case of HT impact, the CYCLONE presented a higher impact which could be due to the generation of ash (0.03 kg). Similar trends were observed for the gasification process. In this case, CRUSHER, GASIFIER and CYCLONE had similar impact values in almost all the impact categories, except for the HT one, where the ash amount generated was 0.05 kg (Table 3).

The results observed for the gasification process in all impact categories analysed and almost for all equipment are higher than twice if compared to those of the combustion one. This fact is due to the higher quantity of olive pomace (0.77 kg for gasification) and, consequently, other inputs used (Table 3) to obtain 1 MJ of energy through the gasification process. To obtain the same energy value, in the case of the combustion process, the required amount of olive pomace was 0.35 kg. On the other

hand, this trend was not observed for the CYCLO RANKINE equipment since, as expected, different emissions released should be considered (Table 6).

The stages of the process with the least environmental impact are CRUSHER, COMBUSTOR and GASIFIER. The impact values were associated to the energy consumption necessary for the operation of CRUSHER, the consumption of the gasifying agent (gasification process) and, in the case of the combustion process, the heat released to the atmosphere (Table 3).

Figure 4 shows for both processes the normalized impact values at the mid-point level using the ReCiPe methodology. In this case, the following order of impact magnitude was observed:

- Combustion process: HT >> CC > TA > POF > FD > PMF > ME > FE > OD.
- Gasification process: TA >> PMF > HT > POF > CC > ME > FD > FE > OD.

Figure 4 shows that the gasification process presented at the mid-point level higher values of all the impact categories than the combustion one, which is related to the higher amount of inputs and outputs required for the former process to obtain 1 MJ. This way, if the combustion process is used for the production of energy, the environmental impacts can be reduced if compared to those of the gasification one in the following percentage: 63.9 % for CC, 52.7 % for OD, 88.4 % for TA, 54.2 % for FE, 94.8 % for ME, 57.2 % for HT, 84.1 % for POF, 91.6 % for PMF and 55 % for FD.

As explained above, all the impact values are attributable primarily to the energy consumption needed in the combustion and gasification processes (Susmozas et al., 2016), but it is also partly due to the environmental impacts of the previous stages (production of olives and extraction of olive oil) (PRéConsultants, 2016).

It can be observed in Figure 4 that for the gasification process the impact category with the highest value ($5.29\text{E-}4$) was TA. This is mainly due to the air emissions

generated by the energy production process. Ammonia (0.0014 kg) is the main emission that contributes to this impact category, but the generation of SO₂ (0.002 kg) should not be dismissed (Table 6). Regarding the combustion process, the value observed for this impact category (6.14E-5) is given by the generation of NO₂, NO and SO₂ (6.09E-7 kg, 0.003 kg and 0.002 kg, respectively). On the other hand, Figure 4 also shows that for the combustion process the impact category with the highest value was HT impact, whereas for the gasification process it was third highest in value. One of the contributing factors in the HT category is the amount of ash (Table 3) (Oreggioni et al., 2017); other is the gases released (Table 6).

In addition, the impact values for POF and PMF were associated with a part of the species found in the gas released such as SO₂, CO, CH₄, NO₂, and NO (Table 6). Moreover, ME and FE were related to the “hidden” emissions coming from the use of the electrical energy (Hsu, 2012; Peters et al., 2015a). On the other hand, NO₂, NO and NH₃ (Table 6), detected in the gas released, affected the eutrophication potential. The GHG emissions, which are related to the use of the electricity, were mainly responsible for the GWP, which is directly linked to the CC impact category (Handler et al., 2014; Zhang et al., 2014; Zhong et al., 2010). Furthermore, the main gases, detected in the gas released after the biomass conversion and associated with the impact value for the CC category, were CO₂ and CH₄ (Table 6).

Moreover, the FD impact category was directly affected by the energy consumption (van Oers and Guinée, 2016), but it could also be related to the utilities consumption (gasifying agent).

This study clearly shows that the combustion process is a more environmentally-friendly process than the gasification one, obtaining lower values in all the impact

categories. This fact is directly correlated to the superior efficiency of the combustion process if compared to the gasification one.

3.3. Overview of the combustion and gasification scenarios

Figure 5 shows the aggregate impact values of the combustion and gasification scenarios, considering all stages (olive production, extraction of olive oil and thermochemical conversion of olive pomace into energy). This type of graphic representation contributes to better understand the comparison between scenarios and facilitates the decision making, taking into account only three categories of global damage (end-point). The global damages incorporate 17 impact categories at the mid-point level. The Tables SS1 and SS2 (supporting information) show the characterized results for combustion and gasification scenario, associated with the functional unit (1 MJ of energy produced), taking into account the olive production, olive oil extraction, and combustion / gasification process (ReCiPe end-point). In this sense, Figure 5 (a, b) confirms that the combustion process affects less to all assessed damages categories than the gasification one. For both evaluated processes, the impact category decreases in the following order: RA > HH > ED (Figure 5b) for the first stages.

It is important to highlight that the first two stages (olive production and olive oil extraction) considered for the two scenarios are exactly the same but the gasification process requires a larger quantity of inputs for generating 1 MJ.

The main factors that affect the three selected impact categories (end-point level) are energy and diesel consumption, the application of fertilizers and the gasifying agent consumption (gasification scenario) (Table 1, 2 and 3). Other factors contributing to the values of the three selected categories of damage are the emissions generated during the whole process, from the production of olives to the conversion of olive pomace into

energy. In this way, the HH category is mainly affected by the following emissions: CO₂, CH₄, SO₂, NH₃, NO_x, N₂O and heavy metals. Furthermore, emissions such as CO₂, CH₄, NO_x, N₂O and SO₂ are responsible for the impact value in the ED category. Nevertheless, the damage category RA is related to the consumption of resources (mineral and fossil). In this study, the resources that contribute to this impact category are: “energy, from gas, natural”, “energy, from oil” “oil, crude”, “gas, natural” and “coal, hard”.

Summarizing, if all impact categories at the mid-point and end-point level are considered the combustion process is a better option than the gasification one. The former process needs less than half of the olive pomace and a lower amount of inputs and outputs, such as raw materials, utilities, emissions and waste.

From the point of view of the energy efficiency, the combustion process overcomes the gasification process. Thus, for the production of 1 MJ of energy, the combustion process requires only 0.039 MJ for keeping the operation of the equipment whereas the gasification one requires 0.054 MJ for the same purpose.

In order to improve the environmental performance, alternatives can be found to reduce the environmental impacts. In the case of the olive production stage, agricultural practices can be modified by using more environmentally friendly fertilizers. In the cases of the thermochemical stage, an improvement of the efficiencies of the equipment, a reduction of the amount of energy necessary for the operation of the equipment and an increase the energy production is required. In addition, ash can be considered as a by-product which could be sold as either an amendment for soil or a fertilizer additive.

Conclusions

In this study, the olive production, olive oil extraction and olive pomace combustion / gasification stages were evaluated through an LCA.

For both assessed scenarios, the highest impact value at the mid-point level was found for the gasification scenario. In the case of the former, the olive oil extraction was the most critical stage in almost all evaluated impact categories, except the HT and POF categories which were affected by the combustion process. In the case of the latter, the gasification process mainly affected to the following impact categories: TA, PMF, HT, and POF.

Rankine Cycle was the major contributor for all impact categories assessed for the combustion and gasification processes, which was to the gases released and to the energy required for the operation of the water pump.

The aggregate impact values of global combustion and gasification scenarios showed similar trends. For both evaluated scenarios, the most remarkable impact category of was RA followed by HH and ED.

From the environmental and the energy generation point of views, the combustion scenario is a better option than the gasification one, due to the combustion process needs less amount of the olive pomace to produce 1 MJ of energy.

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Table 1. Inputs and outputs of the olive production stage, considering the functional unit of 1 MJ energy produced from olive pomace valorisation (Block I)

		Combustion	Gasification
Inputs*			
Water	m ³	9.09E-04	1.98E-03
Phytosanitary (active ingredients)	kg	0.002	0.004
Anhydrous ammonia	kg	0.67	1.46
Diesel	L	0.03	0.06
Rainwater	m ³	1.34	2.9
Outputs*			
Olive	kg	0.48	1.05
Emissions to air**			
NH ₃	kg	6.38E-04	1.39E-03
CO ₂	kg	0.08	0.18
N ₂ O	kg	1.46E-04	3.17E-04
NO _x	kg	2.28E-04	4.96E-04
H ₂ O	m ³	6.62E-04	1.44E-03
Emissions to water**			
Cr	kg	2.17E-06	4.72E-06
Cu	kg	6.77E-07	1.47E-06
Pb	kg	1.68E-07	3.66E-07
Hg	kg	6.09E-10	1.33E-09
Ni	kg	1.85E-07	4.03E-07
NO ₃	kg	4.88E-03	0.01
P	kg	8.51E-06	1.85E-05
H ₂ O	m ³	2.47E-04	5.
Zn	kg	1.69E-06	3.68E-06
Emissions to soil**			
Cd	kg	3.2E-07	6.96E-07
Cr	kg	1.18E-06	2.57E-06
Cu	kg	-5.66E-07	-1.23E-06
Dimethoate	kg	1.45E-07	3.15E-07
Pb	kg	1.23E-06	2.68E-06
Hg	Kg	-2.59E-10	-5.64E-10
Ni	kg	4.36E-07	9.49E-07
Zn	Kg	5.8E-07	1.26E-06

*olive mill plant data; ** Ecoinvent database

Table 2. Inputs and outputs of the olive oil extraction stage, considering the functional unit of 1 MJ energy produced from the olive pomace valorisation (Block I)

		Combustion	Gasification
<i>Inputs*</i>			
Olive	kg	0.48	1.05
Water	m ³	0.002	0.005
Electrical energy	MJ	1.37	3
Diesel	L	0.02	0.05
<i>Outputs*</i>			
Oil virgin	kg	0.08	0.18
Olive pomace	kg	0.35	0.77
Olive stone	kg	0.04	0.08
Waste	kg	0.006	0.014
Wastewater	kg	0.002	0.005
Emissions to air*			
NO _x	kg	2.09E-05	4.54E-05
SO ₂	kg	3.13E-06	6.82E-06
CO	kg	2.74E-04	5.96E-04
PM	kg	1.01E-05	2.19E-05

*olive mill plant data;

Table 3. Inputs and outputs of the combustion and the gasification processes for the olive pomace resulting from the oil extraction from olives (FU = 1 MJ energy production) (Block III A and III B) (from Aspen Plus® software)

Combustion					Gasification				
CRUSHER	<i>Input</i>	Biomass	kg	0.35	CRUSHER	<i>Input</i>	Biomass	kg	0.77
		Energy	MJ	0.013			Energy	MJ	0.03
	<i>Output</i>	Biomass	kg	0.35		<i>Output</i>	Biomass	kg	0.77
COMBUSTOR	<i>Input</i>	Biomass	kg	0.35	GASIFIER	<i>Input</i>	Biomass	kg	0.77
		Air	kg	2.3			Air	kg	2.67
	<i>Output</i>	Heat	MJ	1.68		<i>Output</i>	Gasifying agent	MJ	0.54
		Gas + ash	kg	2.65			Gas + ash	kg	3.98
CYCLONE	<i>Input</i>	Gas + ash	kg	2.65	CYCLONE	<i>Input</i>	Gas + ash	kg	3.98
		Energy	MJ	0.02			Energy	MJ	0.02
	<i>Output</i>	Gas	kg	2.62		<i>Output</i>	Gas	kg	3.93
		Ash	kg	0.03			ash	kg	0.05
RANKINE CYCLE	<i>Input</i>	Gas	kg	2.62	RANKINE CYCLE	<i>Input</i>	Gas	kg	3.93
		Energy	MJ	0.006			Energy	MJ	0.004
	<i>Output</i>	Gas	kg	2.62		<i>Output</i>	Gas	kg	3.93
		Energy	MJ	1			Energy	MJ	1

Table 4. Characterization of the olive pomace (ultimate analysis, proximate analysis and bomb calorimeter)

Ultimate analysis (wt. %)					Proximate analysis (wt. %)				HHV (MJ/kg)
C	H	N	S	O*	Moisture	Volatile matter	Ash	Fixed carbon*	
52.49	6.65	1.51	0.26	31.88	2.12	81.75	7.21	11.04	21.75

O*: obtained by the difference of C, H, N, S and ash; Fixed carbon*: calculated by the difference of ash and volatile matter

Table 5. Blocks description used for the simulation of olive pomace combustion and gasification processes

<i>Combustion</i>			<i>Gasification</i>		
Block name	Aspen Plus® name	Description	Block name	Aspen Plus® name	Description
Dryer	RStoic and Flash 2	To reduce the amount of water.	Dryer	RStoic and Flash 2	To reduce the amount of water.
Crusher	Crusher	To crush biomass to a specific particle size.	Crusher	Crusher	To crush biomass to a specific particle size.
Combustor	RYield (DECOMP)	The first reactor to simulate the volatile matter release.	Gasifier	RYield (DECOMP)	The first reactor to simulate the volatile matter release.
	RGibbs (BURN)	The second reactor based on the stoichiometry of combustion reaction.		Sep2 (CHARSEP)	To separate the amount of char necessary and reach the gasification temperature.
Cyclone	Sep2	To separate ash from the rest of components.		RStoic (BURN)	The second reactor based on the stoichiometry of combustion reaction.
Cycle Rankine	Heat X, Compr, Heater and Pump	The pump delivers liquid water to the boiler where the water is heated. The steam is fed to the turbine to generate power. The condenser is used to cool the steam.		RStoic (GASCONV)	To simulate NH ₃ , HCl and H ₂ O during the gasification process.
				Sep2 (GASSEP)	To separate NH ₃ , HCl and H ₂ O.
				RGibbs (GASIF1)	Biomass char gasifier based on equilibrium models that minimize the free energy Gibbs.
				RGibbs (GASIF2)	Gasifier with the output composition adjusted.
				Mixer (GASMIX)	To mix all the output gas that means during the gasification process
			Cyclone	Sep2	To separate ash from the rest of components.
			Cycle Rankine	Heat X, Compr, Heater, Pump	The pump delivers liquid water to the boiler where the water is heated. The steam is fed to the turbine to generate power. The condenser is used to cool the steam.

Table 6. The composition of the gas obtained through the combustion and the gasification processes (from the Aspen Plus[®] software)

Combustion (kg / h)		Gasification (kg / h)	
Component		Component	
Nitrogen	1.76	Nitrogen	2.04
Water	0.21	Water	0.49
Oxygen	0.006	Oxygen	0.067
Nitrogen dioxide	6.09E-7	Sulphur dioxide	0.002
Nitrogen monoxide	0.003	Hydrogen	0.049
Sulphur	6.0E-8	Carbon monoxide	0.28
Sulphur dioxide	0.002	Carbon dioxide	0.95
Sulphur trioxide	3.65E-07	Methane	0.02
Hydrogen	0.0005	Ammonia	0.014
Carbon monoxide	0.04		
Carbon dioxide	0.6		

Table 7: Impact assessment results (normalised step) of the combustion process, associated with the functional unit, 1 MJ of energy obtained from the valorization of olive pomace, taking into account all the equipment used (ReCiPe mid-point)

	CRUSHER	COMBUSTOR	CYCLONE	RANKINE CYCLE
CC	2.49E-06	2.51E-06	2.73E-06	5.57E-05
OD	6.89E-08	6.95E-08	8.68E-08	9.12E-08
TA	2.27E-06	2.29E-06	2.68E-06	5.42E-05
FE	1.22E-06	1.23E-06	1.38E-06	1.42E-06
ME	2.13E-06	2.15E-06	2.17E-06	2.19E-06
HT	1.58E-06	1.59E-06	6.55E-05	6.57E-05
POF	6.96E-07	7.03E-07	8.36E-07	4.03E-05
PMF	1.31E-06	1.32E-06	1.6E-06	2.54E-05
FD	7.11E-06	7.18E-06	7.67E-06	7.81E-06

Table 8: Impact assessment results (normalised step) of the gasification process, associated with the functional unit, 1 MJ of energy obtained from the valorization of olive pomace, taking into account all the equipment used (ReCiPe mid-point)

	CRUSHER	GASIFIER	CYCLONE	RANKINE CYCLE
CC	5.47E-06	5.47E-06	5.71E-06	1.59E-04
OD	1.52E-07	1.51E-07	1.69E-07	1.96E-07
TA	4.99E-06	4.99E-06	5.39E-06	1.14E-03
FE	2.68E-06	2.68E-06	2.84E-06	3.27E-06
ME	4.68E-06	4.68E-06	4.71E-06	1.50E-04
HT	3.47E-06	3.47E-06	1.43E-04	1.64E-04
POF	1.53E-06	1.53E-06	1.67E-06	2.62E-04
PMF	2.88E-06	2.88E-06	3.16E-06	3.46E-04
FD	1.56E-05	1.56E-05	1.62E-05	1.86E-05

Figure captions

Figure 1. System boundaries. Block: (I) Olive production, (II) Olive oil extraction, (III A) Olive pomace combustion and (III B) Olive pomace gasification

Figure 2. Aspen Plus[®] flowsheet simulation: a. Combustion process and b. Gasification process.

Figure 3. Normalized environmental impact for a. combustion scenario and b. gasification scenario, associated with the functional unit, 1 MJ of energy obtained from the valorization of olive pomace, taking into account the olive production, the olive oil extraction, and the combustion / gasification process (ReCiPe mid-point)

Figure 4. Normalized environmental impact for a. combustion process and b. gasification process, associated with the functional unit, 1 MJ of energy obtained from the valorization of olive pomace (ReCiPe mid-point)

Figure 5. Normalized environmental impact for a. combustion scenario and b. gasification scenario, associated with the functional unit, 1 MJ of energy obtained from the valorization of olive pomace, taking into account the olive production, olive oil extraction, and combustion / gasification process (ReCiPe end-point)

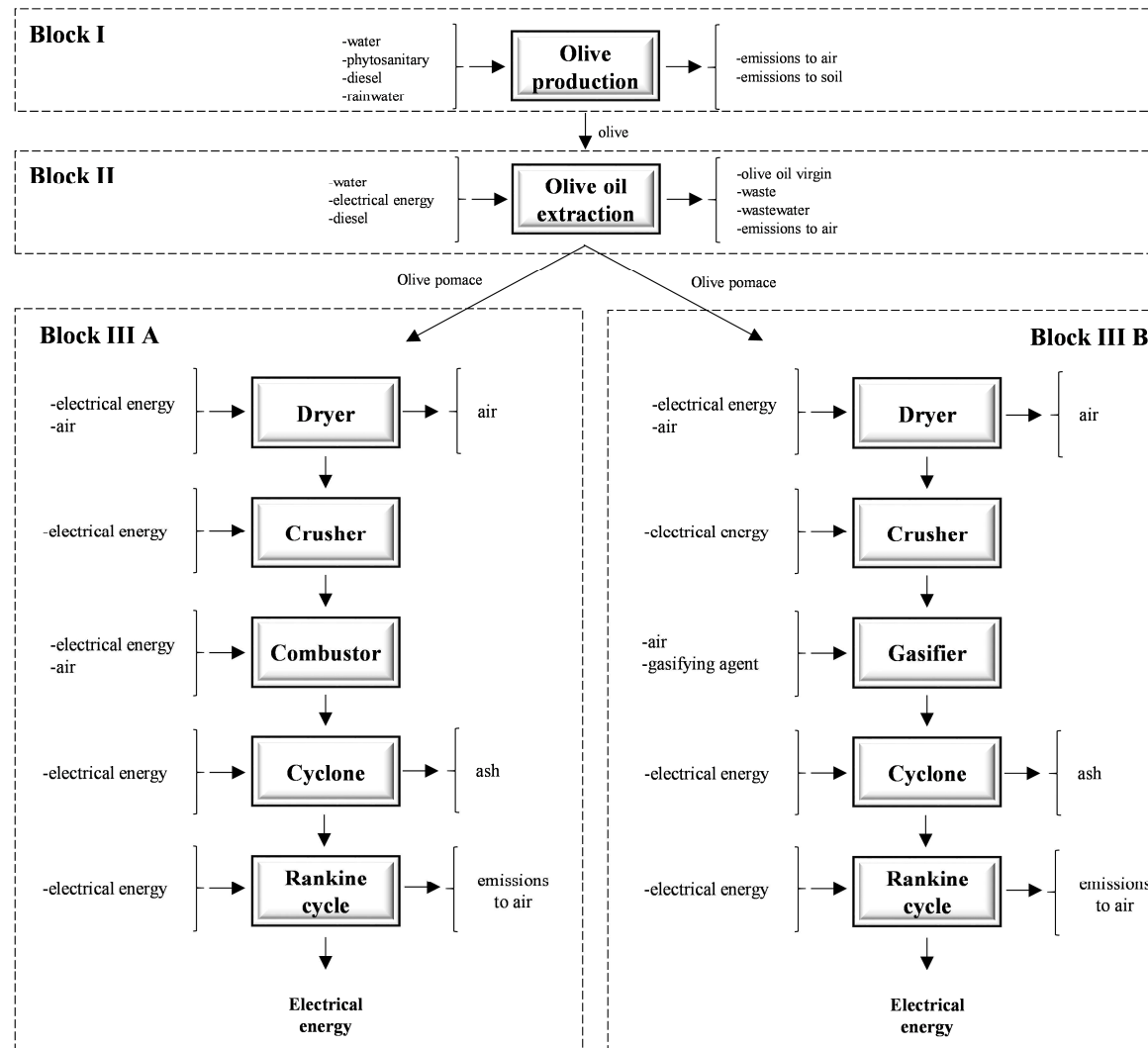
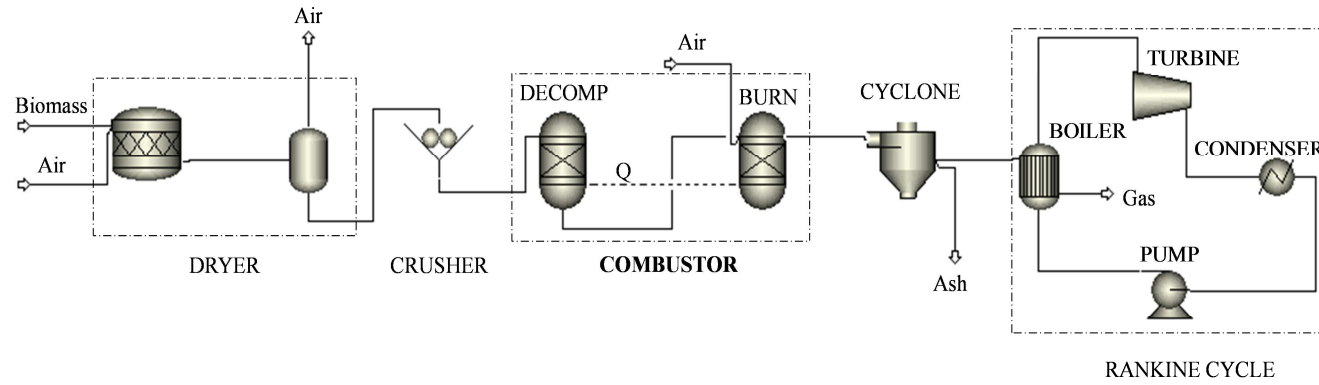


Figure 1

a.



b.

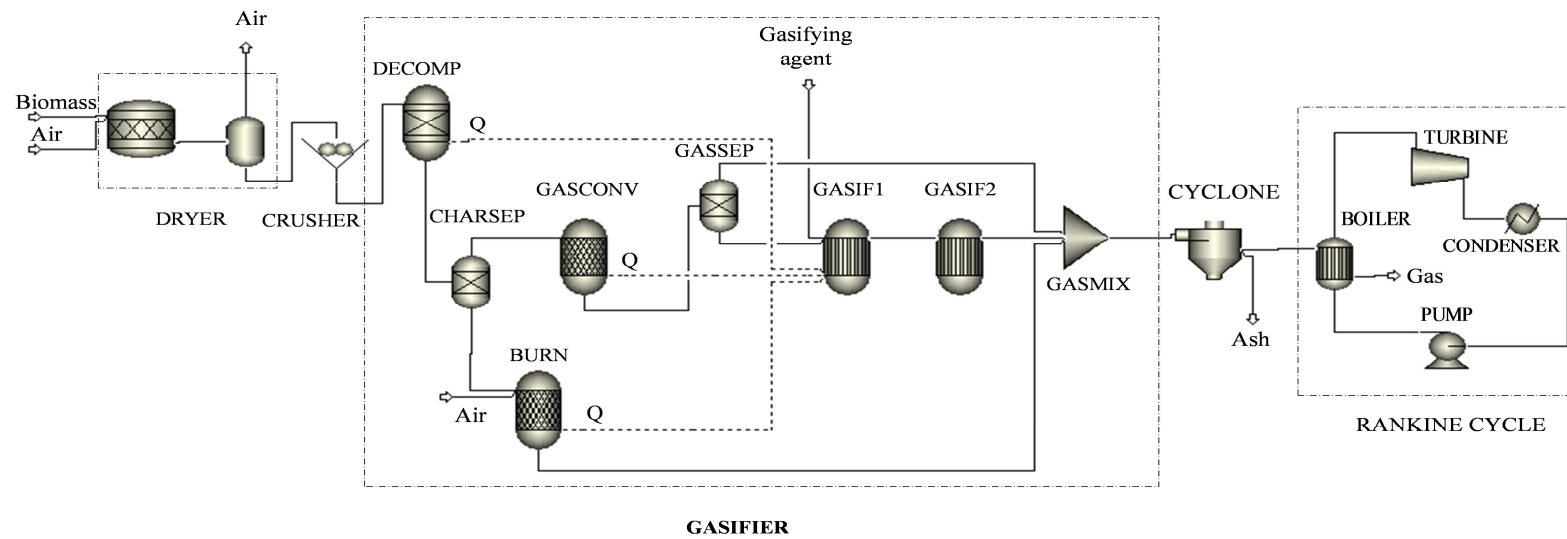


Figure 2

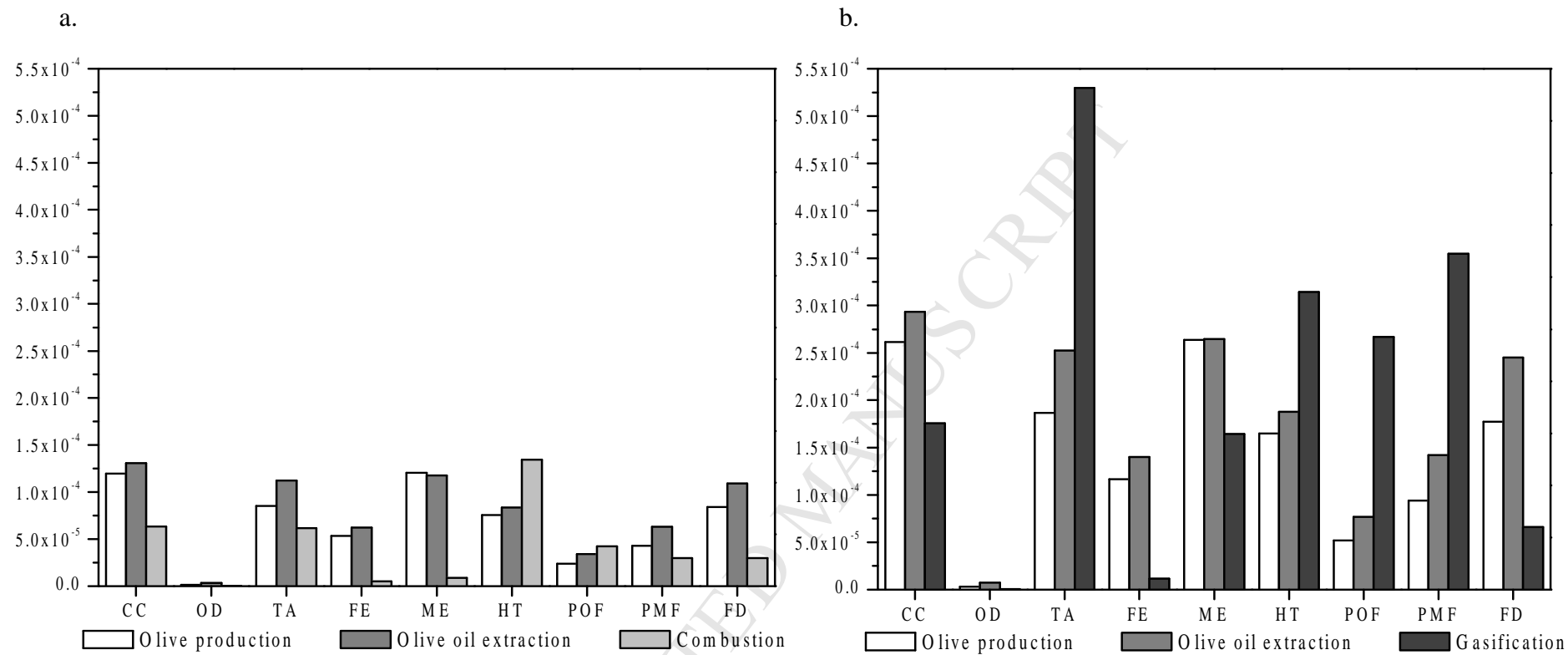


Figure 3

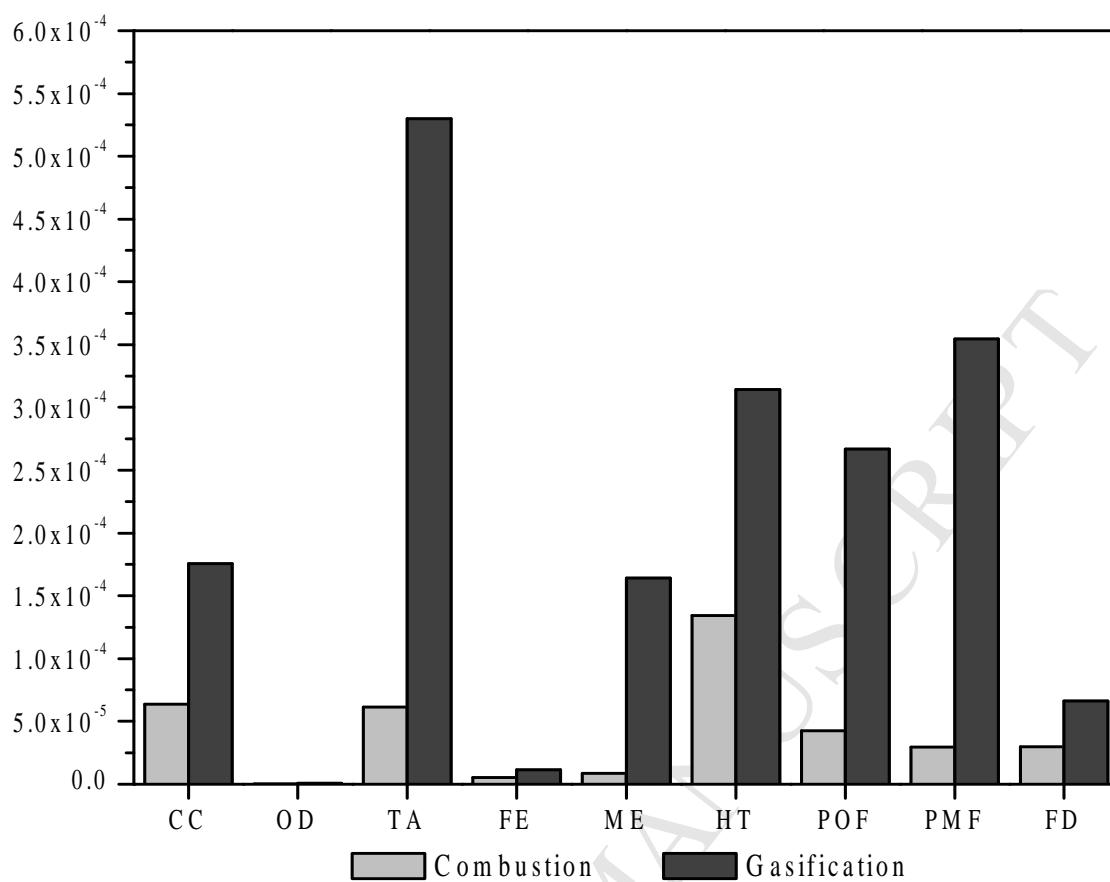


Figure 4

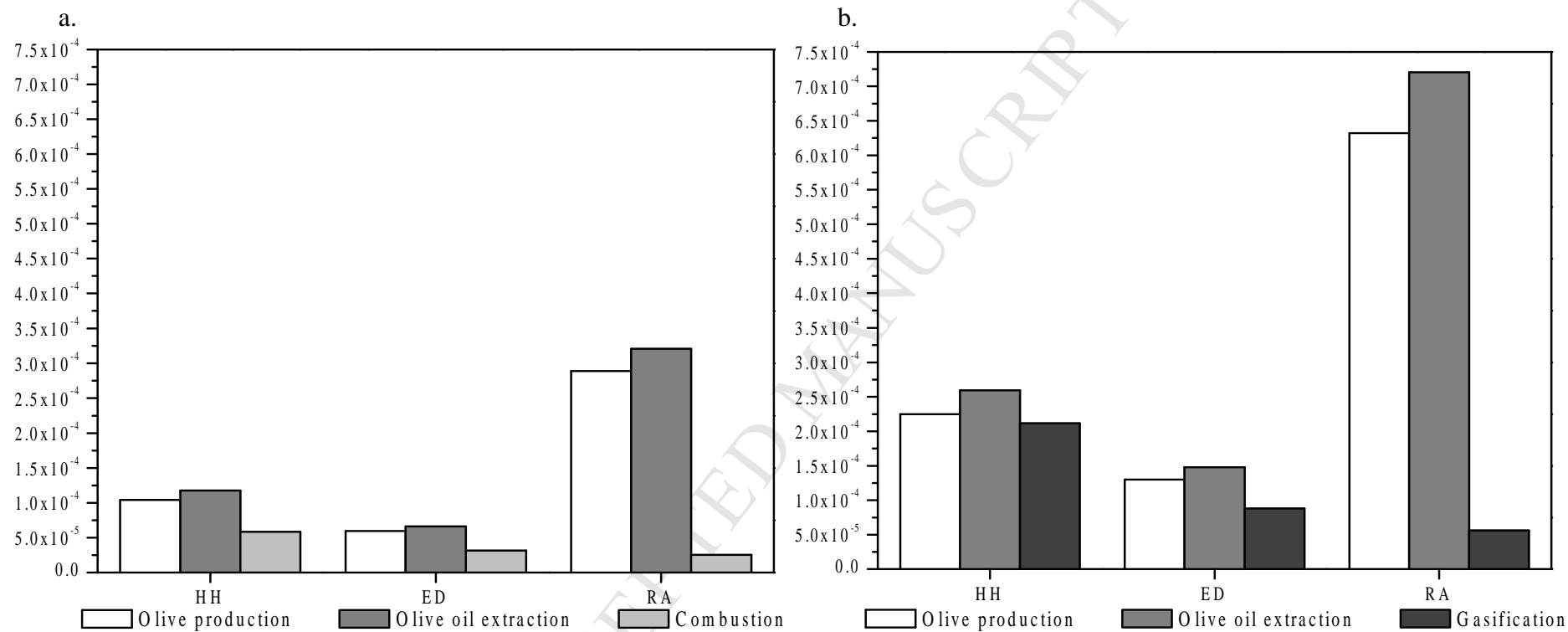


Figure 5

Highlights

- LCA methodology for the olive pomace combustion and gasification processes was performed.
- The olive production and the olive oil extraction stages were evaluated.
- The combustion process is more environmentally friendly than the gasification one.
- Rankine Cycle is the major contributor for all the impact categories assessed.