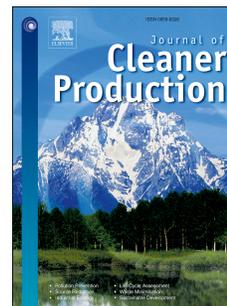


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1 **Environmental assessment of olive pomace valorization through**
2 **two different thermochemical processes for energy production**

3
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11
12
13

14 **Abstract**

15

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

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

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

39 1. Introduction

40 The environmental issues of the contemporary world are mainly caused by the direct
41 and indirect action of anthropological factors. In addition, overpopulation is a current
42 problem in the world due to its effect on the environment (Harte, 2007). The
43 demographic growth is one of the most important issues that lead to increased resources
44 consumption (materials and energy) and the degradation of the environment (Patten,
45 2014). Pollutants, such as gases and solid particles, from industries and domestic
46 activities, have a negative impact on the environment (Patten, 2014). On the other hand,
47 the consumption of the fossil fuels produces greenhouse gases emissions, which are
48 responsible for the global warming and climate change (Rahman and Miah, 2017).

49 In recent years, biomass has been ranked among the most important renewable
50 energy sources, with the greatest growth potential in the future. The use of biomass as a
51 source of renewable energy has many advantages, including that biomass is considered
52 as "carbon neutral", being abundant and available in many regions (Abbasi and Abbasi,
53 2010; Field et al., 2008). It could also reduce the environmental stress by diminishing
54 the dependence of the traditional energy sources and the amount of wastes deposited in
55 landfills and is a raw material for the synthesis of different products as ethanol and
56 similar fuels (Abbasi and Abbasi, 2010; Field et al., 2008).

57 Olive is mainly cultivated in Mediterranean countries (Spain, Italy, Greece, etc.) but
58 also in other countries from America, Africa, and Australia, where the olive oil is a
59 product of great economic importance (Salomone and Ioppolo, 2012; Tsarouhas et al.,
60 2015). According to the ESYRC of 2016 (ESYRC, 2016), Spain has 2,623,156 ha of
61 olive groves; 152,345 (5.81%) of these are destined to table olives. The region of
62 Castilla-La Mancha in Spain has an area of 415,300 ha of olive groves (ESYRC, 2016),

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

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

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

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

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

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

105

106 **2. Material and methods**

107 **2.1. Goal and scopes**

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

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

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

119

120 **2.2. System boundaries and assumptions**

121 An important component in the biomass-to-energy chain is to include the previous
122 stages to the production of energy in the limit of the system, such as the production, the
123 pre-processing and the biomass transport (Gold and Seuring, 2011; Nguyen et al., 2013;
124 Patel et al., 2016; Raynolds et al., 2000). This is mainly due to the fact that the
125 environmental impacts of the previous stages affect the results, and, implicitly, the final
126 decision making. The biomass valorisation considered in this work was analyzed from
127 the raw material production (olive production) to the conversion of the olive pomace via
128 the two thermochemical processes pathways to obtain energy as the final product.
129 Therefore, the system boundary selected to perform the LCA of energy production from
130 olive pomace through the combustion and gasification processes is presented in Figure
131 1.

132 Several hypotheses are to be considered in the actual approach in order to avoid
133 overlapping in the making-decision process. Planting and tree growth have been omitted
134 due to the long time in which there is no production. Input and output data for one year
135 (2015) has been considered. The transportation of the olive to the olive oil extraction
136 plant and the transportation of the fertilizers has been also considered. It was assumed

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

141

142 **2.3. Life cycle inventory analysis**

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

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

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

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

164

165 **2.3.1. Biomass processing (block I and block II)**

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

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

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

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

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

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

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

198

199 **2.3.2. Olive pomace conversion processes**

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

211

212 **2.3.2.1.Olive pomace combustion process (block IIIA)**

213 The combustion process was simulated and the flowsheet diagram is shown in Figure
214 2a. Table 5 shows a brief explanation of each block used for the simulation of the
215 biomass combustion.

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

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

238

239 **2.3.2.2.Olive pomace gasification process (block IIIB)**

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

243 Taking into consideration the particularities of the gasification process, several
244 assumptions were established. H_2 , CH_4 , CO , CO_2 , H_2O , NH_3 , HCl and H_2S were
245 considered as gases evolved during the gasification process. Char is composed of
246 carbon and ash, where ash is considered to be inert. 100 % of ash conversion was
247 obtained during gasification. In addition, it was assumed that all the reactions involved
248 in the gasification process reached the equilibrium (Formica et al., 2016; Pala et al.,
249 2017).

250 The pre-treatment of the biomass in the gasification process is the same as that in the
251 combustion process. Therefore, the biomass was allowed to air dry for 48 hours and
252 then crushed (particle size of 5 mm) to obtain optimum conditions for the gasification
253 process. The resulting stream was fed to reactor DECOMP (RYield), which was used to
254 simulate the release of the volatiles through pyrolysis process based on the conversion
255 of the nonconventional biomass into conventional components. The obtained char (100
256 % carbon) was split in CHARSEP (Sep 2) and then it was fed to the combustion reactor
257 BURN (RStoic). The aim of the char combustion was to increase the temperature of the
258 bed particles, providing the heat necessary for the reaction in the gasification chamber.
259 In addition, the airflow required for the combustion was determined by either using a
260 calculating block that takes the char burnt in the combustion chamber as a reference and

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

273

274 **2.4. Impact assessment methodology**

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

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

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

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

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

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

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

320 The economical allocation for the olive pomace as a co-product must be used,
321 because it is intended to obtaining energy through a subsequent process (PCR, 2014)
322 and considered to reflect better the value of the products by granting most of the
323 impacts to virgin olive oil, which also has the highest unit price (Schau et al., 2016). In
324 this way, the economic allocation factor for the olive oil is 97.2 % (the price for 1 kg of
325 extra olive oil is € 3.65 / kg); for the olive pomace, it is 1.7 % (the average price for
326 olive pomace is € 15 / ton); and, for the olive stone, it is 1.1% (the price of olive stone is
327 € 90 / ton).

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

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

340

341 **3. Results and discussion**

342 **3.1. Environmental assessment of the biomass valorization**

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

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

354 Figure 3a shows that, in the case of the combustion scenario, the extraction stage of
355 olive oil has the highest impact in almost all the assessed categories, followed by olive
356 production and the combustion process. This fact is mainly related to the consumption
357 of energy and diesel and the emissions released. A different trend is observed in the case
358 of the HT and POF categories. In this case, the combustion process is the one that more
359 affects them, mainly due to the ash and emissions generated. On the other hand, Figure
360 3b shows that the gasification process and the olive oil extraction play the leading role

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

365 The emissions (Table 1, 2 and 3) could play an important role in different impact
366 categories (Foteinis and Chatzisymeon, 2016; Wagner and Lewandowski, 2017).
367 Human activities are the main factors that contribute to greenhouse gas emissions,
368 carbon dioxide, methane, chlorofluorocarbons and nitrous oxide and are well known
369 for their global warming potential (GWP) (Houghton et al., 1992; Zhang et al., 2014),
370 which is closely related with the CC impact observed for the olive production stage.
371 The higher GHG emission (CH_4 and CO_2) observed for the gasification scenario if
372 compared to that of the combustion one (Table 6) could explain the higher CC impact of
373 the first one (Figure 3). Although N_2O is a greenhouse gas which is about 300 times
374 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,
375 2006), its presence in traces contributed to a small GWP in comparison with the larger
376 CO_2 emissions. Furthermore, the CO_2 and CH_4 are also responsible for the POF impact
377 category. This impact category and PMF can be also affected by SO_2 and NO_x
378 emissions (Derwent et al., 1996). On the other hand, CH_4 is mainly involved in the OD
379 impact category.

380 In this study, the nitrogen-based emissions released (NH_3 , NO_x and NO_3) during
381 olive production stage (Table 1) were indirectly responsible for the TA, FE, ME and HT
382 impacts (Brentrup et al., 2004; Goedkoop et al., 2009). In this stage, the phosphorus
383 emissions in water are the main contributors in the FE impact category (Brentrup et al.,
384 2004). Moreover, the SO_2 and the NO_x emissions (Table 2) released during the olive oil
385 extraction stage influence mainly the TA and ME impacts (Goedkoop et al., 2009). In

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

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

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

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

402 The application of fertilizers during the olive production stage is responsible to the
403 value of the HT category (Iribarren et al., 2014; Kleinman et al., 2011; Nayal et al.,
404 2016; Peters et al., 2015b; Sharpley et al., 1990; Uzoma et al., 2015) (Table 1).
405 Furthermore, the high HT value obtained in the olive oil extraction stage could be
406 attributed to the high values of waste and wastewater generated (Table 2).

407

408 **3.2. Environmental assessment of olive pomace thermochemical conversion**

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

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

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

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

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

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

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

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

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

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

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

454 As explained above, all the impact values are attributable primarily to the energy
455 consumption needed in the combustion and gasification processes (Susmozas et al.,
456 2016), but it is also partly due to the environmental impacts of the previous stages
457 (production of olives and extraction of olive oil) (PRéConsultants, 2016).

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

460 generated by the energy production process. Ammonia (0.0014 kg) is the main emission
461 that contributes to this impact category, but the generation of SO₂ (0.002 kg) should not
462 be dismissed (Table 6). Regarding the combustion process, the value observed for this
463 impact category (6.14E-5) is given by the generation of NO₂, NO and SO₂ (6.09E-7 kg,
464 0.003 kg and 0.002 kg, respectively). On the other hand, Figure 4 also shows that for the
465 combustion process the impact category with the highest value was HT impact, whereas
466 for the gasification process it was third highest in value. One of the contributing factors
467 in the HT category is the amount of ash (Table 3) (Oreggioni et al., 2017); other is the
468 gases released (Table 6).

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

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

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

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

486

487 **3.3. Overview of the combustion and gasification scenarios**

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

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

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

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

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

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

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

531

532 **Conclusions**

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

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

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

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

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

550

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554

555

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725

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		Gasifying agent	MJ	0.54	
CYCLONE	<i>Input</i>	Gas + ash	kg	2.65	CYCLONE	<i>Output</i>	Gas + ash	kg	3.98
		Energy	MJ	0.02			Gas + ash	kg	3.98
	<i>Output</i>	Gas	kg	2.62		<i>Input</i>	Gas + ash	kg	3.98
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 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)

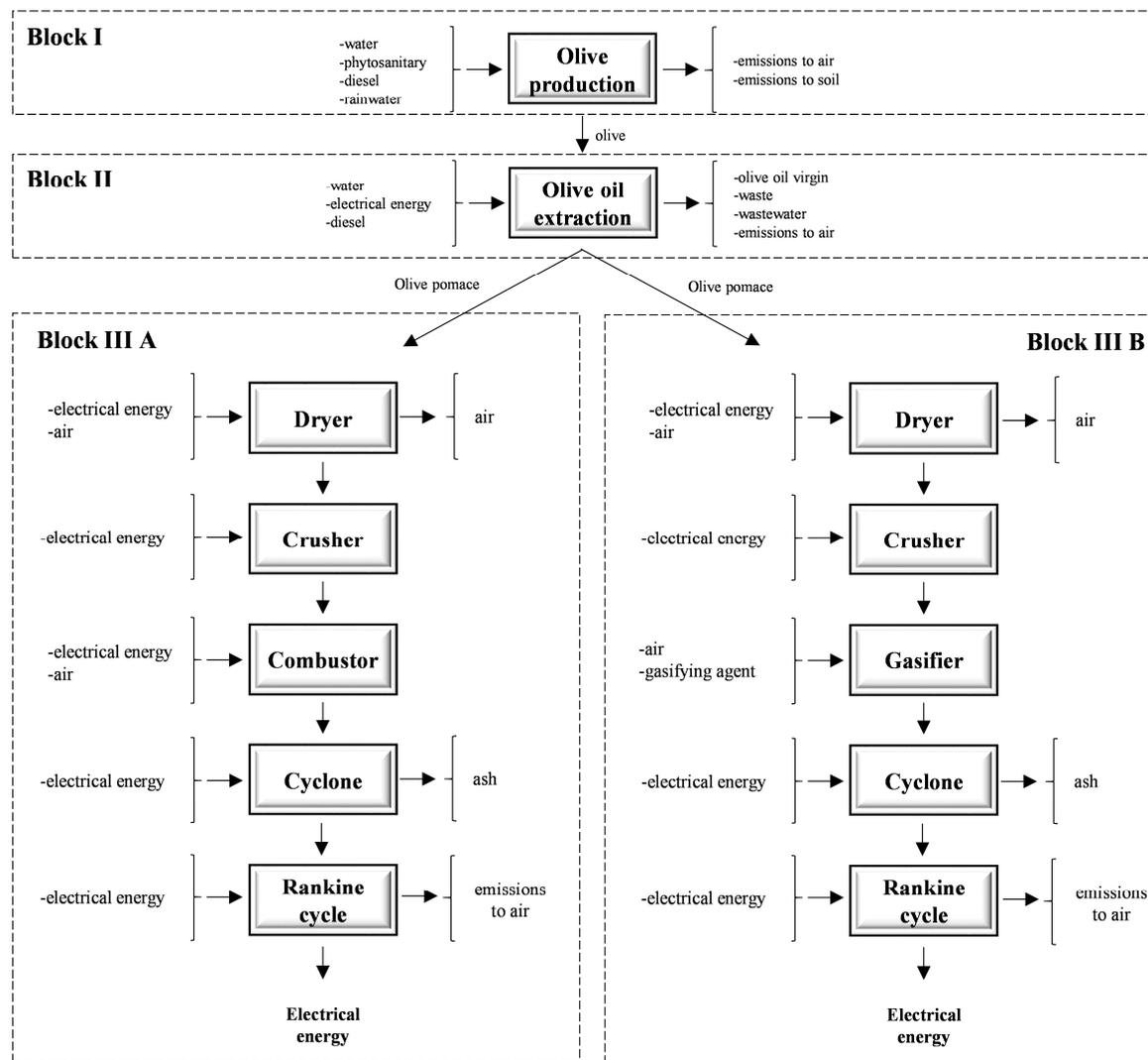
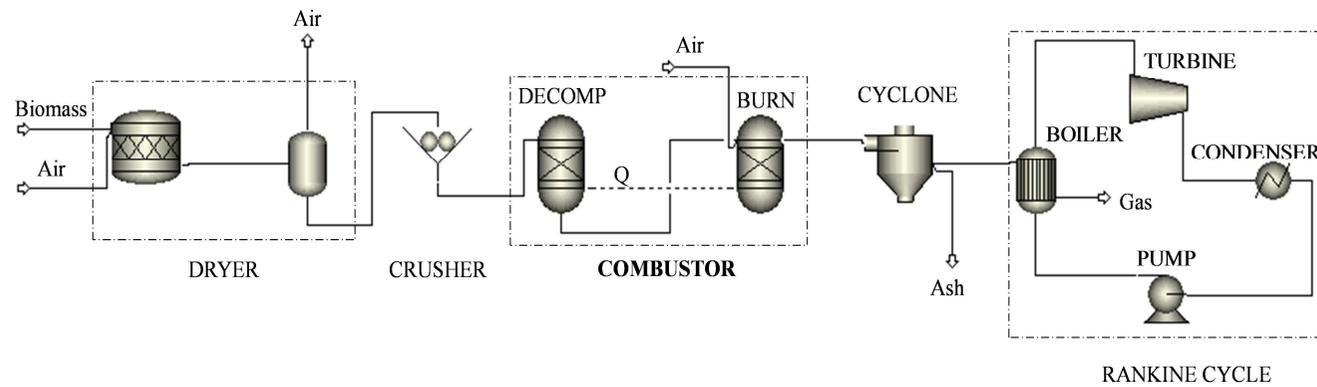


Figure 1

a.



b.

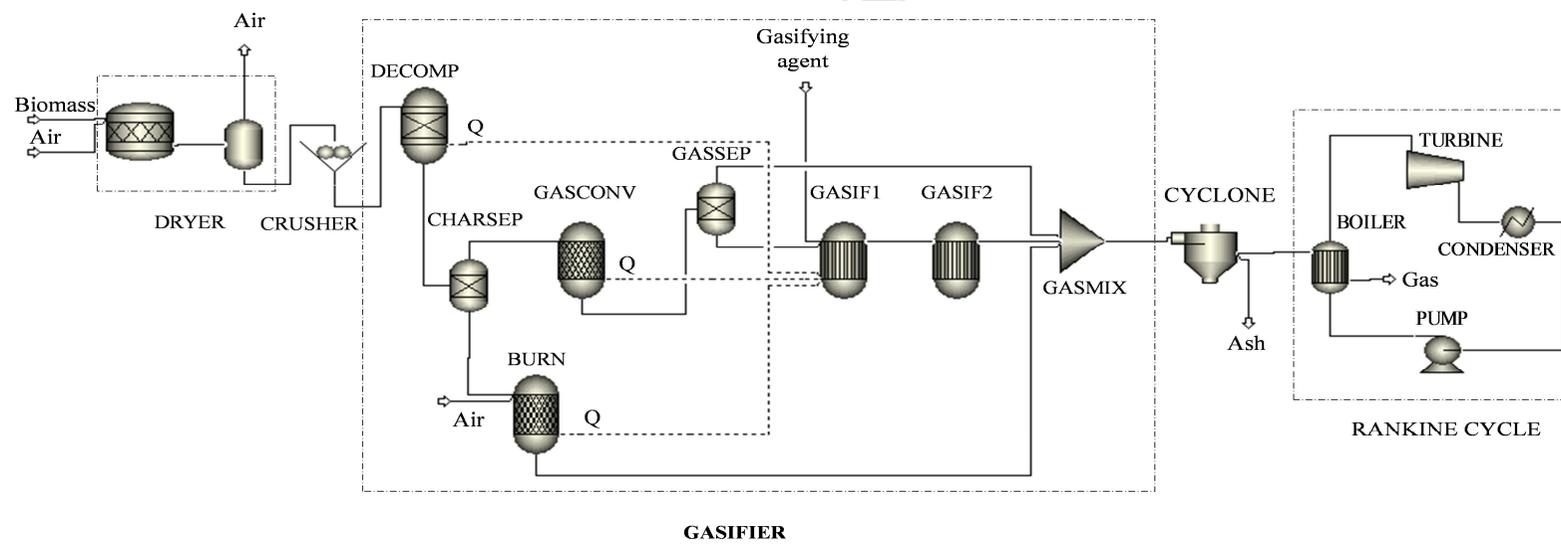


Figure 2

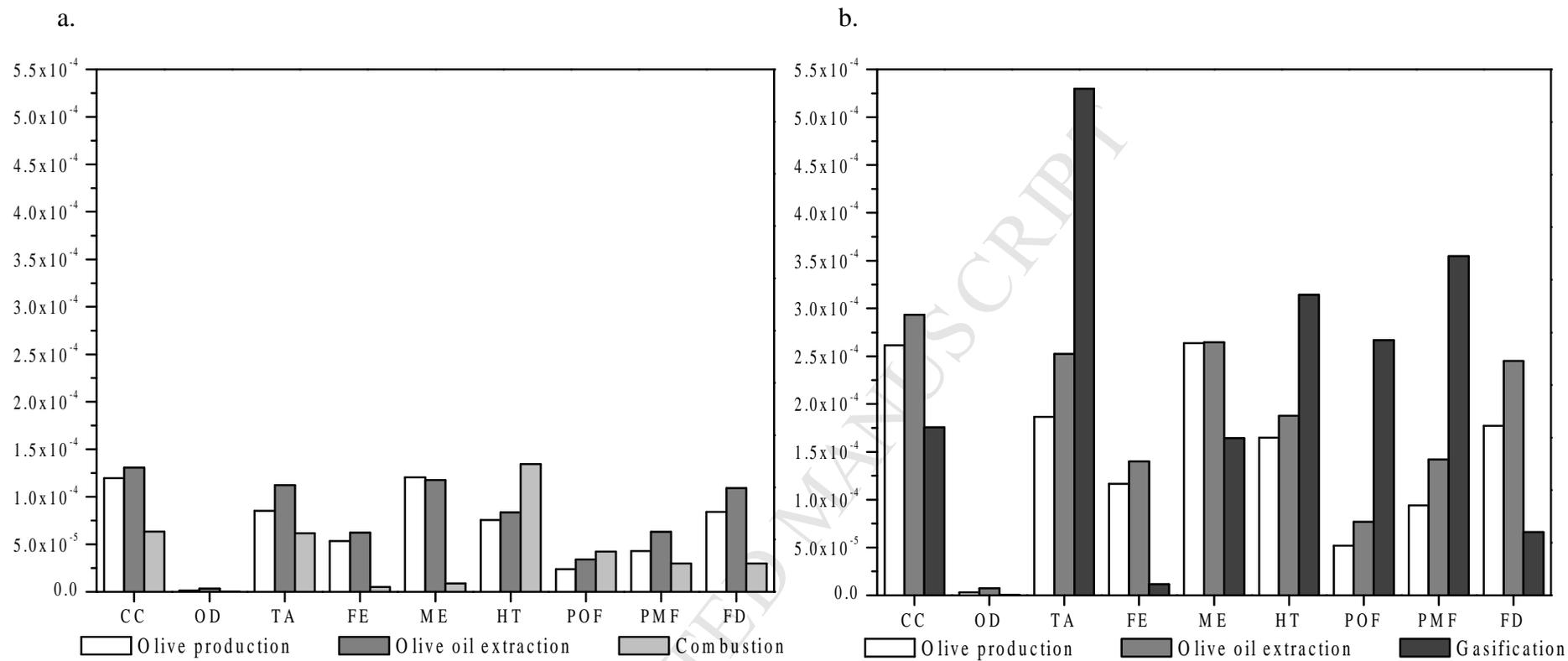


Figure 3

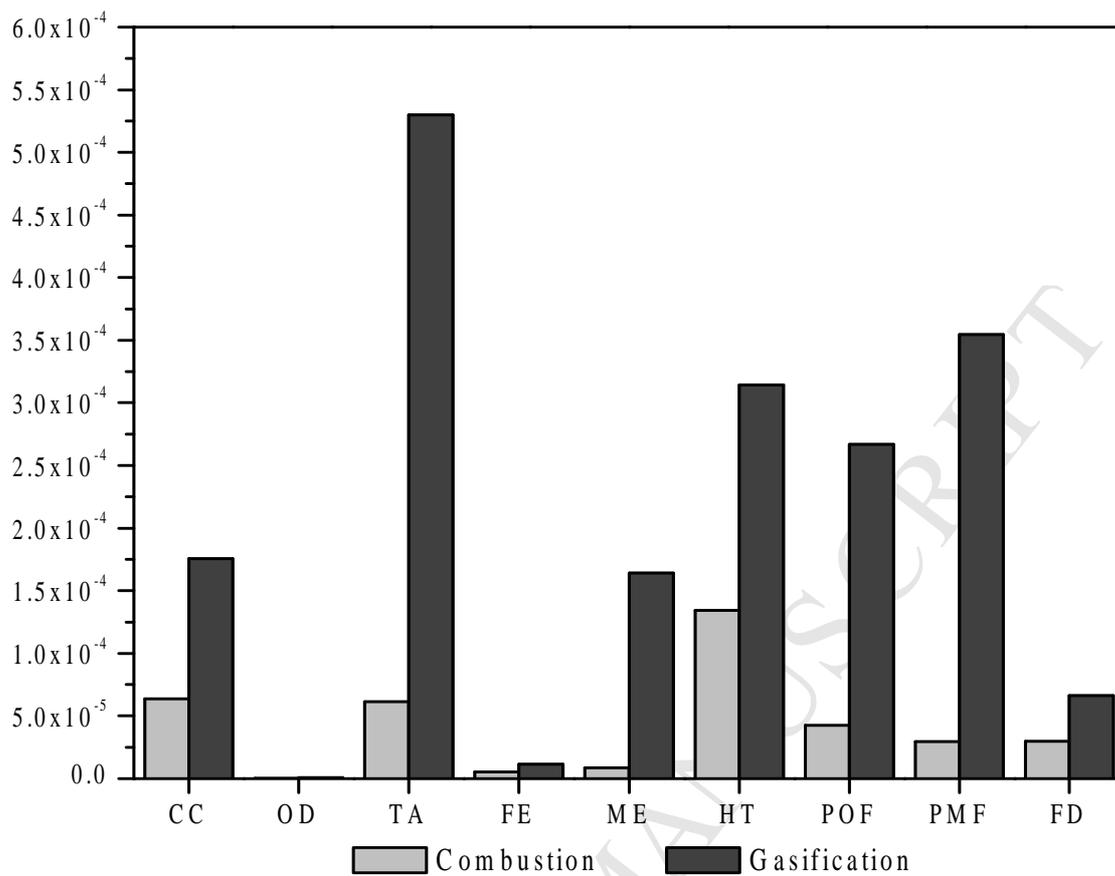


Figure 4

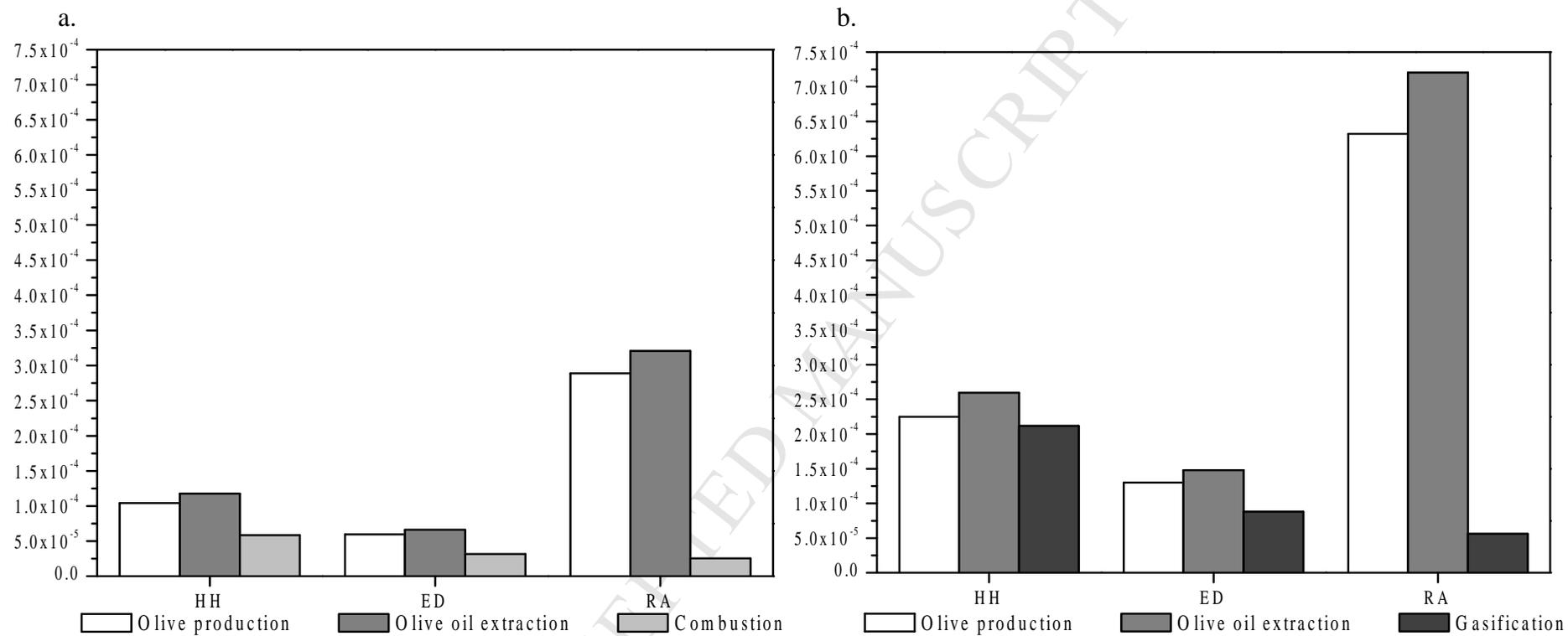


Figure 5

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.