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# 1 **Integrated evaluation of wine lees valorization to produce value-added products**

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## 8 **Abstract**

9 The integrated evaluation of the valorization of wine lees to produce value-added  
10 products was carried out in this study from a life-cycle perspective. The consumption of  
11 steam has been demonstrated as the main hot spot, reaching 85.7% of the impact on  
12 Fossil Depletion and 85.3% on Climate Change. Bearing in mind that four different  
13 value-added products are produced, a sensitivity analysis was carried out in order to  
14 ascertain the influence of the functional unit and the allocation method on the  
15 environmental outcomes. The performance of this system was compared to other  
16 processes that produce antioxidants from different raw materials. These processes were  
17 phycocyanin recovery from *Spirulina platensis* cyanobacterium, the production of the  
18 red antioxidant astaxanthin by microalgae and the valorization of the macroalgae  
19 *Sargassum muticum*. Wine lees valorization showed a better environmental profile  
20 throughout the entire life cycle, due to the fact that most of the operations performed are  
21 physical (solid/liquid separations, distillations, evaporations, etc.) and do not involve a  
22 large consumption of electricity or chemicals. However, there is still room for  
23 improvement, and future research should focus on optimizing the extraction of  
24 antioxidants from wine lees using two-stages aqueous systems, ultrasonic or microwave

25 assisted extraction, in the pursuit of better performance and lower environmental  
26 impact.

27 **Keywords:** Wine lees valorization; Biorefinery; Life Cycle Inventory; Environmental  
28 assessment; Value-added products.

## 29 **Abbreviations**

LCA Life Cycle Assessment

LCI Life Cycle Inventory

FU Functional Unit

SS Subsystem

ISO International Organization for Standardization

CC Climate Change

OD Ozone Layer Depletion

TA Terrestrial Acidification

FE Freshwater Eutrophication

ME Marine Eutrophication

HT Human Toxicity

POF Photochemical Oxidant Formation

TET Terrestrial Ecotoxicity

FET Freshwater Ecotoxicity

MET Marine Ecotoxicity

FD Fossil Depletion

30

31

## 32 **1. Introduction**

33 The primary sector is one of the largest industrial sectors in terms of resources and  
34 energy consumption (Roy et al., 2009) and has therefore been identified as one of the  
35 main actors in climate change, with 30% of total greenhouse gas (GHG) emissions.

36 With the aim of developing a competitive low-carbon economy (European Commission,  
37 2011), action plans and measures have been put forward to reduce the current level of  
38 GHG emissions throughout the food supply chain.

39 Beyond environmental indicators related to climate change, the low efficiency in food  
40 production and processing has been recognized as a major issue. According to a recent  
41 FAO report (FAO, 2011), up to 33% of the food produced for human consumption is  
42 lost or wasted along the supply chain. Not only is food wasted, but the resources needed  
43 to produce it, such as water, energy, chemicals or fuels used, are also misused,  
44 representing a value of nearly 1.3 billion tons per year. In addition, according to data  
45 estimated by United Nations (2017), in relation to population growth, world's  
46 population is expected to increase to 8.6 billion by 2030, which will probably aggravate  
47 the problem of food waste.

48 Biorefinery is a clear example of the change of paradigm in the framework of  
49 sustainable development. Biorefineries represent the transition from oil refineries to  
50 sustainable systems based on the valorisation of waste flows with the aim of producing  
51 value-added compounds such as biogas, electricity, chemical products or biomaterials  
52 (Cherubini, 2010). Following this principle, food waste can be valorised through  
53 different technologies, such as anaerobic digestion to generate bio-hydrogen and bio-  
54 methane (Algapani et al., 2019), co-composting with other types of organic waste for  
55 the production of bioenergy and fertilizers (Vico et al., 2018), conversion into animal

56 feed (Makkar, 2018) or recovery of sugars, organic acids, pigments, fibre, proteins, oils,  
57 antioxidants and vitamins from food waste (García-Herrera et al., 2010).

58 Within the primary sector, wine production is becoming increasingly important as a  
59 symbol of a quality product, with a growing influence on exports from producing  
60 countries. World wine production is dominated by Italy, France and Spain, which  
61 together account for 48% of total production (OIV, 2018). From this perspective, the  
62 wine sector can be considered a reference point in the EU strategy within the primary  
63 production sector (Christ and Burritt, 2013). The winemaking process comprises a  
64 numerous sequence of activities (Escribano-Viana et al., 2018), from the cultivation of  
65 the vine, the harvest, the process of fermentation and maturation of the wine in the  
66 winery to the management of waste generated at each stage of the process. The main  
67 effluents from the wine sector are wastewater and organic solid waste (Ruggieri et al.,  
68 2009). Solid organic waste includes wine lees and grape pomace, which is composed of  
69 stalks, skins and seeds. In general, the volume of waste generated is around 20-30% of  
70 total wine production, which can be considered a meaningful percentage (Zabaniotou et  
71 al., 2018).

72 In order to improve the overall efficiency and environmental impact of the winemaking  
73 process, actions have been identified that allow for the minimization, management and  
74 effective recovery of waste streams from a circular economy perspective (Musee et al.,  
75 2007). The approach of circular economy grants and consolidates the value of each  
76 element of the productive chain and deepens the awareness of action, essential to  
77 achieve a real change towards sustainability, with efficient use of resources and  
78 valorization of by-products and wastes.

79 In this framework, Gullón et al. (2018) analysed the environmental impacts of different  
80 routes for the valorisation of vine shoots. Poveda et al. (2018) proposed revaluing the  
81 by-products of winemaking, grape pomace and stems as a source of natural  
82 preservatives. Nayak et al. (2018) developed a method for recovering polyphenols from  
83 exhausted grape pomace through activated carbon. Zhang et al. (2017) compared two  
84 methods for the valorisation of grape pomace, which is the major component of wine  
85 production waste, to add value to economic and environmental balance of the overall  
86 process. These processes were combustion to produce electricity, and pyrolysis to  
87 produce bio-gas, bio-oil and bio-char. The detailed analysis of the wine lees fraction  
88 presents high concentrations of macronutrients and polyphenols and low concentrations  
89 of micronutrients and heavy metals (Devesa-Rey et al., 2011). Moreover, the presence  
90 of other compounds of potential interest such as polyphenols and antioxidants identifies  
91 this stream as an ideal candidate to be valorized (Dimou et al., 2015; Kopsahelis et al.,  
92 2018; Martinez et al., 2016).

93 These valorization options can be evaluated according to their environmental  
94 performance. Several environmental assessment methods can be found in literature, as  
95 material flow analysis, energy balance, exergy analysis and life cycle assessment  
96 (Vandermeersch et al., 2014). The latter seems to be the best choice since it can  
97 consider the full life cycle (cradle-to-gate or cradle-to-grave).

98 The objective of this work seeks to delve into the different strategies for valuing  
99 winemaking-derived waste, proposing the identification of critical points in the  
100 environmental profile of the process under study, prior to its development and  
101 marketing of the products obtained. The function of the system under study is the use of  
102 wine lees to produce some bio-based products with marketable added value. It is  
103 therefore a question of identifying the most suitable process alternatives in a system

104 under development, which may suffer from limitations in terms of data availability, but  
105 it may also make it possible to establish a roadmap in the search for viable options from  
106 a techno-economic and environmental point of view.

## 107 **2. Materials and methods**

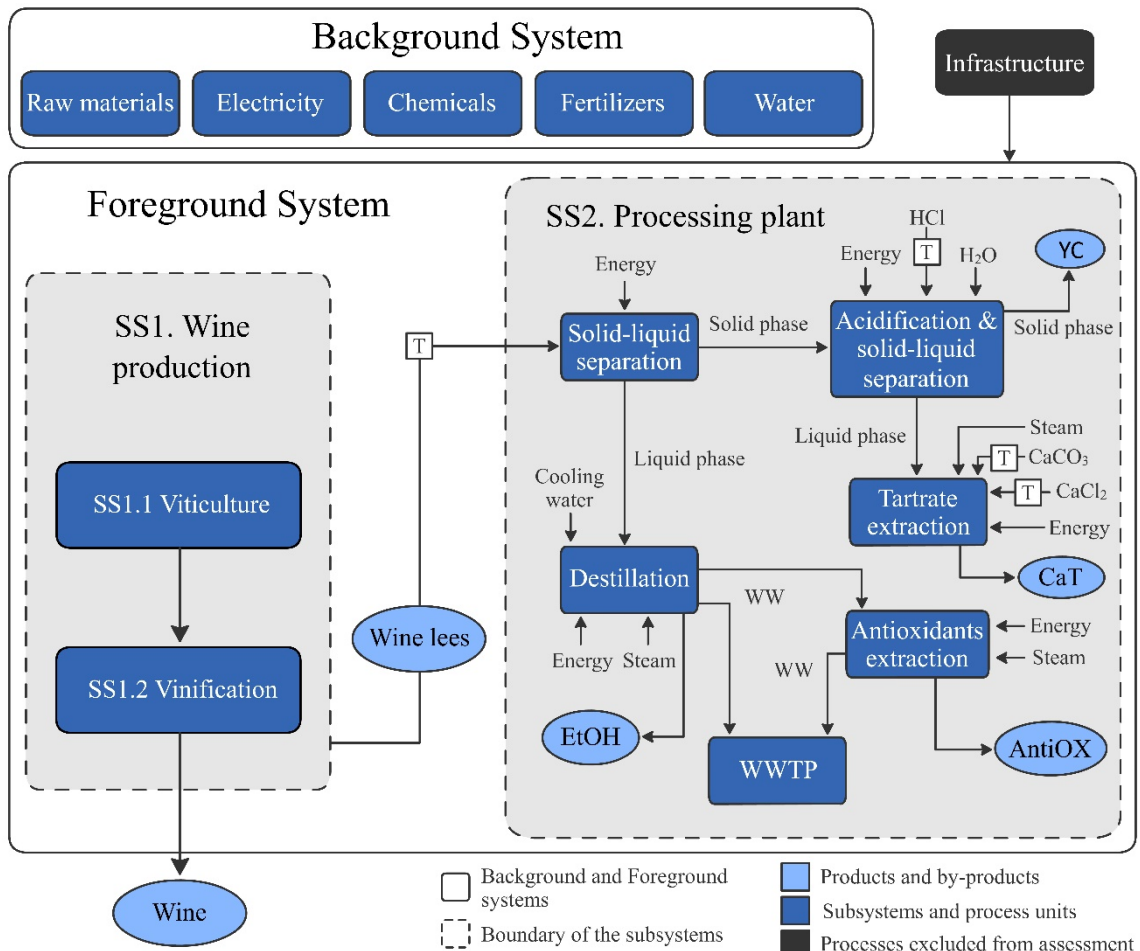
### 108 *2.1. Definition of scope and system boundaries*

109 The functional unit is a measure of the function of the system studied and provides a  
110 reference to which the inputs and outputs can be related (ISO 14040; 14044). The  
111 selection of the functional unit in biorefinery studies is made on the basis of three  
112 possible options, i.e. total flow of raw materials, quantity of a single target product or  
113 the combination of products (Khoshnevisan et al., 2018). The FU considered in this  
114 study was 1 tonne of wine lees processed in the winery facility. This feedstock-based  
115 FU is consistent with the choice of other similar studies, in which a similar FU was  
116 chosen because of their multiple-output nature (Lam et al., 2018; Vaskan et al., 2018).  
117 The production plan was evaluated considering all the processes from the production of  
118 raw materials to the final products obtained from the wine lees, in such a way that the  
119 processes in the winery are analyzed, mainly those associated with the production of  
120 raw materials, electricity, fertilizers, chemical products and water, as well as the  
121 consumption of fuel used in the transport of materials.

122 The foreground system includes process units that are the direct object of the study.  
123 This system is divided into two subsystems represented in Figure 1: SS1. Wine  
124 production and SS2. Processing plant. Figure 1 shows the block diagram of the process,  
125 identifying the system boundaries, the subsystems considered, and the main inputs and  
126 outputs. No infrastructure process was considered in the study since the environmental  
127 impacts of construction, installation, etc. have been considered negligible during the

128 useful life of the facility. This assumption is a common practice in other life cycle  
 129 assessment studies of different biorefineries (Jeswani et al., 2015; Karlsson et al., 2014).  
 130 As far as storage processes, it was considered unnecessary in this study, since only a  
 131 small warehouse within the facilities is needed to store the wine lees at room  
 132 temperature, so it is included within the infrastructure processes.

133 Subsystem 1 is divided into two sections: SS1.1. Viticulture comprises different  
 134 activities considered in the agricultural phase of wine production, including fertilization,  
 135 field operations or soil management and SS1.2. Vinification includes the processing of  
 136 grapes in the winery: wine production, bottling and packaging (Vázquez-Rowe et al.,  
 137 2012). Wine lees are a residue generated during this process, which is further processed  
 138 in Subsystem 2.



139



140 **Figure 1.** System boundaries of the wine lees biorefinery to produce value-added  
141 products. Caption: T: Transport; EtOH: Bioethanol; CaT: Calcium tartrate; YC: Yeast  
142 cells; AntiOX: Antioxidants; WW: Wastewater; WWTP: Wastewater Treatment Plant  
143 Subsystem 2 is the industrial process in which wine lees are valorized to produce four  
144 end-products, in this case bioethanol, calcium tartrate, an extract rich in antioxidants  
145 and solid fraction rich in protein, which can be marketed with an economic return. The  
146 description of the stages of the valorization process is described below. The first step is  
147 the separation of the liquid and solid fractions by centrifugation of the wine lees in a  
148 disc centrifuge. Wine lees fed the facilities at room temperature (25°C) and with a  
149 content of 62.9% (w/w) water, 5.7% (w/w) ethanol and 31.4% (w/w) solids (Dimou et  
150 al., 2016). It is possible to recover the residual fraction of ethanol present in the liquid  
151 fraction by distillation at 100°C. In this step, the product stream of the bottom contains  
152 mainly water and is sent to treatment. The solid fraction is then sent to an extraction  
153 tank in which it is mixed with the ethanol recovered in the previous stage. The recovery  
154 of ethanol implies a significant reduction in the cost associated with this stage since it is  
155 available in the original wine lees (Dimou et al., 2016). A new solid-liquid separation is  
156 then carried out in a disc centrifuge so that the liquid fraction is fed to another  
157 distillation column in which the antioxidant-rich extract is separated as a bottom  
158 product. The removal of water from this stream takes place in an evaporator and in a  
159 spray dryer which works with high pressure steam. The solid stream from the second  
160 solid-liquid separation is mixed with HCl and the tartrate salt (insoluble in water) is  
161 transformed into tartaric acid (water-soluble). This compound is mixed with calcium  
162 carbonate (CaCO<sub>3</sub>) and calcium chloride (CaCl<sub>2</sub>), transforming water-soluble tartaric  
163 acid into water-insoluble calcium tartrate. Calcium tartrate is obtained as a solid product

164 after the drying stage. Yeast cells are also obtained from the solid fraction, which can be  
165 marketable as animal feed to obtain an economic return.

## 166 *2.2. Data acquisition*

167 The data used in this study come from complementary studies that consider all stages of  
168 the life cycle of wine production, which includes the vine cultivation and the winery  
169 processing stage (Subsystem 1) to the valorization of the different fractions of wine and  
170 its by-products within the biorefinery (Subsystem 2) with a processing capacity of 500  
171 kg/h of wine lees developed in Dimou et al. (2016). An environmental assessment was  
172 carried out with a common perspective defined on the basis of an identical functional  
173 unit for both subsystems: 1 tonne of wine lees. The inventory data for the production of  
174 all system inputs from background processes were taken from the Ecoinvent® database.  
175 These inputs include the production of the different chemical products necessary for the  
176 extraction of calcium tartrate, the electricity consumed in the different stages, the  
177 fertilizers for the vine cultivation and any other type of raw material or resource. An  
178 average transport distance of 800 km was considered within continental Europe for  
179 chemical products (Pérez-López et al., 2014b) and 100 km of average distance for the  
180 transport of the wine lees from the winery to the processing plant were assumed (Hajjaji  
181 et al., 2013).

## 182 *2.3. Life cycle inventory*

183 The life cycle inventory is the compilation of the data set for evaluation and involves  
184 the collection of quantitative input/output data for the system. In this study, a cradle-to-  
185 gate approach has been proposed, taking into account all stages, from the cultivation of  
186 the vine to the activities of the winery and the valorisation of the wine lees. Considering  
187 that the economic allocation reflects the function and objective of the production

188 process, which is obviously the marketing of a main product: wine and the associated  
189 economic revenues, mass or energy-based allocations were discarded (Rugani et al.,  
190 2013). Since wine lees are a residue derived from the production of wine, it is necessary  
191 to make a cost estimate as a requirement for an economic allocation that allows the  
192 environmental impacts corresponding to each fraction to be assessed. Thus, the selling  
193 price of a bottle of wine produced after SS1 was compared with the potential selling  
194 price of the various products obtained from the valorisation route (extract rich in  
195 antioxidants, ethanol, yeast cells and calcium tartrate). A market prize of €4 for a 750  
196 mL bottle of wine with designation of origin was considered. Considering that the  
197 production of a bottle of this wine generates 11.48 ml of wine lees, the sale of these  
198 products generates a profit of €0.022 and when comparing the contributions of both  
199 products in terms of their market value, the economic allocation factors are 99.45% for  
200 wine and 0.55% for wine lees.

#### 201 *2.4. Life cycle impact assessment: methodology*

202 SimaPro 8.5.2 (PRé Consultants, 2017) has been the software used for the  
203 implementation of the Life Cycle Inventory. To analyze the inputs and outputs of the  
204 LCI, the Classification and Characterization guidelines defined by ISO were followed.  
205 In this phase, in order to translate the long list of life cycle inventory results into a small  
206 number of environmental impact indicators, the ReCiPe Midpoint methodology was  
207 used. This method provides a common framework in which both mid-point and end-  
208 point indicators can be used (Goedkoop et al., 2009). The characterization factors  
209 reported by the ReCiPe Midpoint methodology were applied, and the potential impact  
210 categories considered were: Climate Change, Ozone Layer Depletion, Terrestrial  
211 Acidification, Freshwater Eutrophication, Marine Eutrophication, Human Toxicity,

212 Photochemical Oxidant Formation, Terrestrial Ecotoxicity, Freshwater Ecotoxicity,  
213 Marine Ecotoxicity and Fossil Fuel Depletion.

### 214 **3. Results and discussion**

#### 215 *3.1. Quantitative analysis of inputs and outputs to the overall system*

216 Life Cycle Assessment is a structured and comprehensive method for quantifying  
217 material and energy flows and associated impacts on the life cycle of products (i.e.  
218 goods and services). Within the LCA methodology, Life Cycle Inventory is a  
219 mandatory stage and the availability of LCI data is often the greatest barrier to  
220 completing an LCA study.

221 LCI shows that the consumption of different pesticides is very high in the viticulture  
222 stage, which is in line with other studies on different crops (Caldeira et al., 2018; Liang  
223 et al., 2019). In addition, the consumption of organic fertilizers is also remarkable,  
224 leading to relevant nitrate emissions, so the impact on the eutrophication categories is  
225 expected to be considerable. As for the winemaking phase, the main inputs are  
226 electricity and chemicals such as NaOH and SO<sub>2</sub>. In this system, CO<sub>2</sub> emissions due to  
227 wine fermentation were calculated, but excluded from the environmental assessment, as  
228 biogenic CO<sub>2</sub> at this stage was not taken into account. LCI has also allowed to quantify  
229 the main inputs and outputs of the wine lees valorization system, highlighting the  
230 consumption of High-Pressure Steam and the production of four value-added products.

231 The main product obtained in this process is the extract rich in antioxidants. This  
232 extract, as calculated in Dimou et al. (2016), has a lower total polyphenol content than  
233 other studies (26.1 mg of gallic acid equivalent per g of wine lees). Therefore, this  
234 extraction is not optimized, and the product could be purer. However, wine lees

235 valorization can be considered as a sustainable and environmentally friendly process  
236 due to the fact that most of the operations carried out are physical, such as solid-liquid  
237 extraction or centrifugation, while other studies involve complex operations, such as  
238 ultrasound or microwave assisted extraction (Castro-López et al., 2017;  
239 Mohammadpour et al., 2019; Pereira et al., 2017). Since most of the treatments carried  
240 out at this stage are physical, the use of chemical products is low, as only 174 kg per ton  
241 of treated wine lees are consumed. Further details of Life Cycle Inventory are provided  
242 in electronic Supplementary data.

### 243 *3.2. Analysis of processing plant energy requirements*

244 Electricity and steam consumption reported in supplementary material correspond to the  
245 operation of the plant and is broken down by equipment in Table 1. Although the total  
246 electricity consumption is not too high (125 kWh per FU), more than 95% of this  
247 consumption corresponds to disc centrifuges, which are used to separate the solid phase  
248 from the liquid phase. This separation process is essential to obtain value-added  
249 products, as most of the treatments carried out are physical, such as stages of  
250 distillation, separation, evaporation and spray drying with compressed hot air.

251 Steam consumption is very high, more than 5 tons of steam per ton of wine lees treated.

252 In the process, steam is used in distillation columns (units E-102 and E-104) while in  
253 the unit E-105, steam is used to evaporate water from the antioxidant-rich stream.

254 Finally, in the E-106 and E-107 exchangers, steam is used to heat the compressed air  
255 that will be used in the spray dryers to remove the remaining water from the calcium  
256 tartrate and the antioxidant-rich extract. This consumption, together with the fact that  
257 obtaining steam is an activity with high energy requirements (Nieuwlaar et al., 2015),

258 makes it possible to anticipate that the environmental impact derived from the use of  
259 steam will be high.

260

261 **Table 1.** Analysis of the electricity, steam and cooling water consumption of Subsystem  
 262 2. Processing plant per FU (1 tonne of wine lees)

<b>Operation</b>	<b>Electricity consumption (kWh)</b>	<b>Steam consumption (kg)</b>	<b>Cooling water (m<sup>3</sup>)</b>
<b>Disc centrifuges</b>	<b>119.19</b>		
CF-101	29.80		
CF-102	29.80		
CF-103	29.80		
CF-104	29.79		
<b>Mixing tanks</b>	<b>4.68</b>		
V-101	2.60		
V-102	1.20		
V-103	0.88		
<b>Blower</b>	<b>6.06</b>		
C-101	4.06		
C-102	2.00		
<b>Heat exchangers</b>		<b>5104.84</b>	<b>224.46</b>
E-102		819.44	
E-104		3994.44	
E-105		238.19	
E-106		39.58	
E-107		13.19	
E-101			33.61
E-103			190.85

263  
 264 *3.3. Environmental characterization of wine lees valorization process*

265 The environmental impacts expressed through different impact categories are presented  
 266 in the characterization stage, these results are summarized in Table 2. Figure 2 presents  
 267 the contributions relative to the environmental burdens of each subsystem considered,  
 268 identifying the most problematic ones with the greatest environmental impacts in the  
 269 process of valorisation of wine lees. Specifically, in Subsystem 1, wine production is  
 270 the main contributor to the categories of MET, FET, POF, HT, ME and TA. In MET  
 271 and TET, SS1 represents almost 90% of the total environmental impact, which is related  
 272 to the treatment of solid waste produced during winemaking. SS1.1 includes different

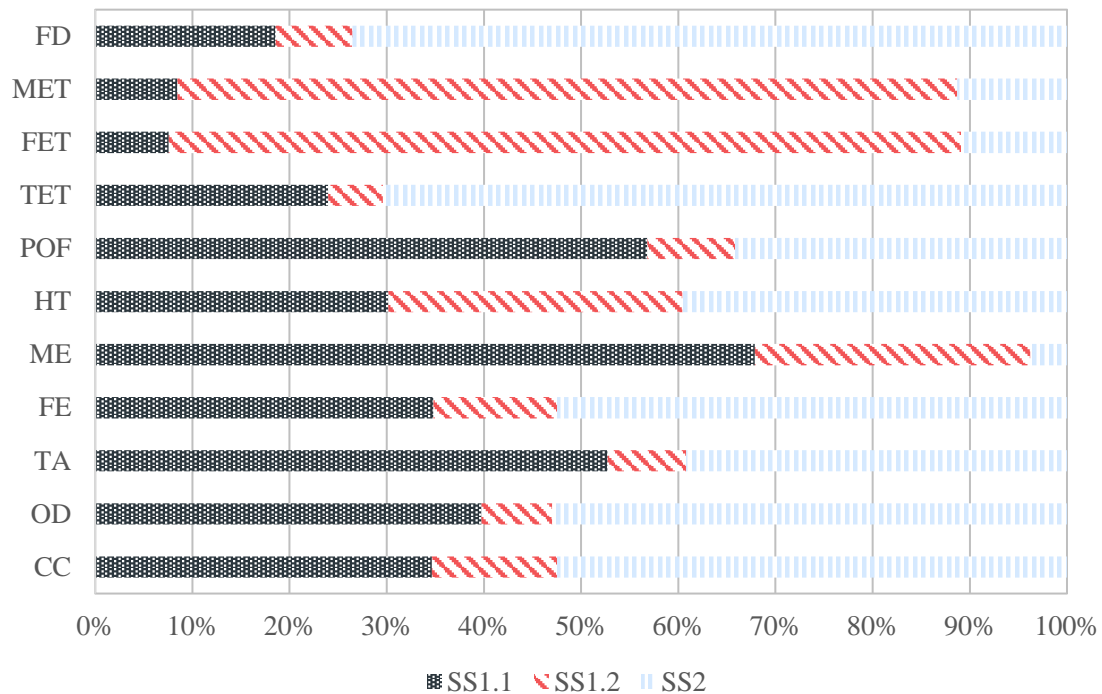
273 activities of the agricultural phase of wine production such as soil management, field  
 274 operations or fertilisation. This phase is clearly the hotspot in ME, POF and TA  
 275 categories with remarkable contributions of 67.9%, 56.8% and 52.7% respectively. It  
 276 should be noted here that the use of fossil fuels for the operation of machinery such as  
 277 broadcasters and rotary cultivators and the use of compost for fertilization cause  
 278 emissions of nitrogen oxides that affect POF category. As for the environmental impacts  
 279 of ME, the application of fertilizers in the agricultural phase of wine production  
 280 involved the greatest relative impact of this category. These results are in accordance  
 281 with the results obtained in another study in which an LCA of red wine production from  
 282 Catalonia was performed (Meneses et al., 2016). This study showed that the vinification  
 283 process has a better environmental profile than the viticulture stage. Regarding HT  
 284 category, there are no major differences between the two considered subsystems  
 285 because the main contributors to the environmental impact in this category are the  
 286 emissions of heavy metals into the atmosphere derived from the consumption of fossil  
 287 fuels for the operation of equipment, transport and steam production.

288 **Table 2.** Impact assessment results associated with the valorization of 1 tonne of wine  
 289 lees

<b>Impact category</b>	<b>Unit</b>	<b>SS1.1</b>	<b>SS1.2</b>	<b>SS2</b>	<b>Total</b>
<b>CC</b>	kg CO <sub>2</sub> eq	8.79·10 <sup>2</sup>	3.26·10 <sup>2</sup>	1.33·10 <sup>3</sup>	2.54·10 <sup>3</sup>
<b>OD</b>	kg CFC-11 eq	1.14·10 <sup>-4</sup>	2.09·10 <sup>-5</sup>	1.52·10 <sup>-4</sup>	2.87·10 <sup>-4</sup>
<b>TA</b>	kg SO <sub>2</sub> eq	6.52	1.00	4.85	12.37
<b>FE</b>	kg P eq	0.15	0.05	0.22	0.42
<b>ME</b>	kg N eq	2.52	1.05	0.14	3.72
<b>HT</b>	kg 1,4-DB eq	1.85·10 <sup>2</sup>	1.86·10 <sup>2</sup>	2.43·10 <sup>2</sup>	6.14·10 <sup>2</sup>
<b>POF</b>	kg NMVOC	4.25	0.68	2.56	7.48
<b>TET</b>	kg 1,4-DB eq	3.67·10 <sup>-2</sup>	8.61·10 <sup>-3</sup>	1.08·10 <sup>-1</sup>	1.53·10 <sup>-1</sup>
<b>FET</b>	kg 1,4-DB eq	5.36	57.81	7.73	70.88
<b>MET</b>	kg 1,4-DB eq	5.25	49.77	7.01	62.03
<b>FD</b>	kg oil eq	1.10·10 <sup>2</sup>	46.85	4.35·10 <sup>2</sup>	5.92·10 <sup>2</sup>



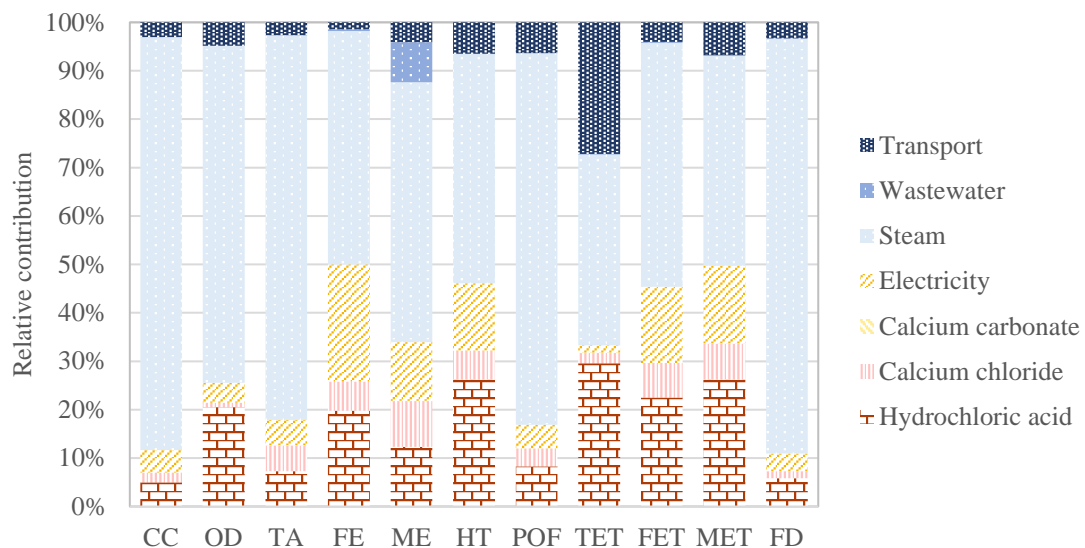
290  
 291 In relation to subsystem 2, the processing plant is the main contributor in FD and TET  
 292 impact categories. In the FD category it reaches a maximum contribution of 75%  
 293 associated with the use of natural gas for steam production. In the TET category, it  
 294 accounts for 70% of the impact due to emissions of heavy metals into the atmosphere  
 295 linked to the use of fossil fuels, either in transport or in the production of high  
 296 temperature steam. However, regarding FE, OD and CC categories, the difference  
 297 between the two subsystems is minimal. Focusing on CC, direct emissions into the  
 298 atmosphere associated with fermentation process are quantified in the winemaking  
 299 process. However, direct CO<sub>2</sub> emissions from SS1 should not be considered as fossil  
 300 carbon, but as biogenic CO<sub>2</sub>. If these emissions were considered as fossil carbon, SS1  
 301 contribution to the CC category would increase to 48.3%, which would mean a global  
 302 value of 2570 kg of CO<sub>2</sub> eq per ton of wine lees.



303

304 **Figure 2.** Relative contributions (in %) by subsystems in the valorization of wine lees

305 Taking into account the economic allocation factors calculated in Section 2.3, the  
 306 characterisation results of the wine lees biorefinery were calculated (Figure 3).  
 307 According to the results obtained, wine lees can be classified as a winery waste that is  
 308 not assigned an associated environmental impact. On the contrary, the use of high  
 309 temperature steam can be considered as the most burdensome component. In this sense,  
 310 the other categories include FD (85.7%), CC (85.3%), TA (79.4%) and POF (76.7%),  
 311 all of which are associated with the high consumption of fossil fuels for the production  
 312 of steam as well as the associated emissions of GHG, SO<sub>2</sub> and NO<sub>x</sub>.



313

314 **Figure 3.** Relative contributions (in %) by component in the overall biorefinery  
 315 production process

316 Electricity has a low impact in almost all impact categories, which is attributed to a low-  
 317 moderate consumption (Table 1). Only FET shows a remarkable value of 24.2%. HCl  
 318 becomes the second most contributing element with an average of 16.7%, standing out  
 319 in the categories of toxicity, either Human (HT) or Ecosystem (TET, MET and FET);  
 320 due to emissions of heavy metals associated with the production of HCl, H<sub>2</sub>SO<sub>4</sub> and Cl<sub>2</sub>  
 321 compounds. In contrast, the impact of CaCl<sub>2</sub> and CaCO<sub>3</sub> are of much lesser importance

322 with average contributions of 4.7% and 0.03% respectively. Transport presents a  
323 considerably uniform distribution of environmental impacts in all categories, with  
324 contributions almost always lower than 7%, except in the case of the TET category, in  
325 which a relative contribution of around 27.2% is reached. This is because this category  
326 gives more weight to emissions to air and soil (which are abundant in transport  
327 processes); in contrast, other toxicity categories such as MET and FET are more  
328 dependent on water emissions, with negligible impacts. Wastewater treatment  
329 contributions are practically negligible except for a percentage of 8.2% in ME category,  
330 due to nitrogen compounds directly discharged into the water.

331 *3.4. Sensitivity analysis. Effect of the selection of the Functional Unit and allocation*  
332 *factors on the environmental profile*

333 The results obtained are based on a FU that focuses on the amount of biomass valorized  
334 rather than on the products obtained. In fact, the choice of FU is a critical point in any  
335 LCA study as it is a subjective action that must be consistent with the objectives of the  
336 study. In this case the function of the system is the treatment of a waste, but this system  
337 allows to obtain four different products. The extract rich in antioxidants is the product  
338 of greatest interest for its potential applications in the food industry, cosmetics and  
339 pharmaceutical industry (Szabo et al., 2018). Therefore, the new FU was selected as 1  
340 kg of antioxidant-rich extract obtained.

341 In addition to the choice of the FU, the allocation of impacts is fundamental, especially  
342 in multi-production processes such as the one studied here. If a mass or economic  
343 allocation is followed, the impacts assigned to each product are different. Therefore, the  
344 factors of mass and economic allocation have been calculated by the quantity produced

345 of each element and its potential market price obtained from various scientific  
 346 publications. A summary of the considered allocation factors is given in Table 3.

347 **Table 3.** Computation of the mass and economic allocation factors for SS2. Processing  
 348 plant

<b>Product</b>	<b>Production<sup>a</sup></b>	<b>Market price</b>	<b>Mass allocation</b>	<b>Economic allocation</b>
Bioethanol	28.22 kg	0.67 €/kg <sup>b</sup>	8.4%	1.3%
Calcium tartrate	58.50 kg	4.41 €/kg <sup>c</sup>	17.5%	17.3%
Yeast cells	241.00 kg	0.88 €/kg <sup>c</sup>	72.0%	14.2%
Antioxidants	6.78 kg	147.67 €/kg <sup>d</sup>	2.0%	67.6%

349 <sup>a</sup> Results per Functional unit (1 tonne of wine lees)

350 <sup>b</sup> (Joelsson et al., 2016)

351 <sup>c</sup> (Dimou et al., 2016)

352 <sup>d</sup> (Vieira et al., 2013)

353

354 These studies show how the market prices of each of the products obtained in the  
 355 biorefinery varies and how the mass and economic allocations factors vary accordingly.  
 356 Joelsson et al. (2016) conducted a research on the production of biogas and bioethanol  
 357 from wheat straw impregnated with acetic acid. In addition to the experimental study,  
 358 they also performed a techno-economic evaluation. In this paper a bioethanol price of  
 359 0.57-0.68 €/kg is estimated. In Dimou et al. (2016) a techno-economic evaluation of the  
 360 biorefinery which is environmentally evaluated in the present study was carried out and  
 361 a market cost for yeast cells of 1 \$/kg was estimated. This price can be converted to  
 362 euros and is equivalent to 0.88 €/kg. Finally, in Vieira et al (2013) a chemical and  
 363 economic evaluation of antioxidants extracted from pulp of *Euterpe edulis* was carried  
 364 out. The manufacturing costs of the crude extracts obtained in this paper were 165.34  
 365 \$/kg, which is equivalent to 147.67 €/kg. These manufacturing costs were assumed as  
 366 the market cost of this extract in order to obtain an economic return.

367 The effect of the alternative FU and allocation factors on the environmental profiles is  
 368 shown in Table 4. When allocation factors (mass or economic) are used, a decrease in

369 environmental impact is always observed, as the environmental impact is distributed  
 370 among the different products. As the amount of the extract rich in antioxidants is low, in  
 371 the case of mass allocation its environmental impact is small. However, this is not  
 372 realistic, as it is the product of greatest interest, so, as shown in Table 4, the economic  
 373 allocation distributes the environmental impacts among the products more accurately.  
 374 To the production of 1 kg of extract rich in antioxidants is assigned, among others,  
 375 251.3 kg of CO<sub>2</sub> eq in CC category and 58.6 kg of oil eq in FD category.

376 **Table 4.** Environmental results for each impact category considering only the  
 377 production of 1 kg of antioxidants-rich extract with no allocation, mass and economic  
 378 allocation factors

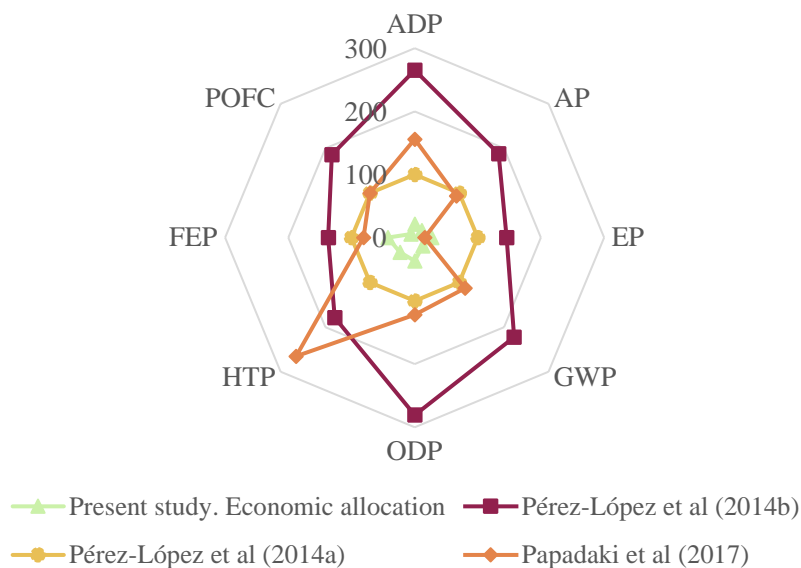
<b>Impact category</b>	<b>Units</b>	<b>No allocation</b>	<b>Mass allocation</b>	<b>Economic allocation</b>
CC	kg CO <sub>2</sub> eq	374.1	7.6	251.3
OD	kg CFC-11 eq	4.2·10 <sup>-5</sup>	8.6·10 <sup>-7</sup>	2.8·10 <sup>-5</sup>
TA	kg SO <sub>2</sub> eq	1.8	3.7·10 <sup>-2</sup>	1.2
FE	kg P eq	6.2·10 <sup>-2</sup>	1.3·10 <sup>-3</sup>	4.2·10 <sup>-2</sup>
ME	kg N eq	5.5·10 <sup>-1</sup>	1.1·10 <sup>-2</sup>	3.7·10 <sup>-1</sup>
HT	kg 1,4-DB eq	90.6	1.8	60.9
POF	kg NMVOC	1.1	2.2·10 <sup>-2</sup>	7.4·10 <sup>-1</sup>
TET	kg 1,4-DB eq	2.3·10 <sup>-2</sup>	4.6·10 <sup>-4</sup>	1.5·10 <sup>-2</sup>
FET	kg 1,4-DB eq	10.4	2.1·10 <sup>-1</sup>	7.0
MET	kg 1,4-DB eq	9.1	1.9·10 <sup>-1</sup>	6,15
FD	kg oil eq	87.3	1.8	58.6

379

380 *3.5. Comparative environmental performance of different methods to obtain*  
 381 *antioxidant-rich extracts*

382 A comparison was made with some processes published in the scientific literature on  
 383 the basis of an identical FU: 1 kg of extract rich in antioxidants and evaluation

384 methodology: CLM. Pérez-López et al. (2014a) evaluated the environmental  
 385 performance of several *Sargassum muticum* macroalgae valorization strategies. In this  
 386 study it was considered that the combined extraction of antioxidants and alginates  
 387 stands out as the most sustainable scenario. Pérez-López et al. (2014b) conducted a life  
 388 cycle assessment of astaxanthin production on a laboratory and pilot scale. In Papadaki  
 389 et al. (2017) an evaluation of the life cycle of the recovery of phycocyanin from  
 390 *Spirulina platensis* cyanobacterium is performed. This study compares six different  
 391 methods based on Ultrasound-Assisted Extraction to recover phycocyanin. In order to  
 392 simplify the comparative study, the results are scaled to 100 and represented in Figure  
 393 4.



394

395 **Figure 4.** Relative environmental profile of the compared valorization scenarios with  
 396 the process published in (Pérez-López et al., 2014a) as baseline (index = 100)

397 According to the results depicted in Figure 4, the production of antioxidants through the  
 398 valorization of wine lees would be the most appropriate route in all the impact  
 399 categories studied. Except in the case of EP, where the recovery of phycocyanin from a  
 400 cyanobacterium is the process with the best environmental profile, since the impacts of

401 EP are directly related to emissions linked to fertilization in the viticulture phase. The  
402 production of astaxanthin from microalgae cultivated in photobioreactors on a pilot  
403 scale reported the worst environmental profile, with results ranging from 37% worse  
404 than the reference (index = 100) in FEP category up to 180% worse in ODP impact  
405 category. If the present study is compared with the process published by Pérez-López et  
406 al. (2014a), the production of antioxidants from the treatment of wine lees presents an  
407 environmental profile that is, on average, 75% better in all impact categories. A  
408 maximum improvement rate of 92% is reached in the FEP impact category. However, in  
409 order to obtain 1 kg of extract rich in antioxidants, it is necessary to treat almost 148 kg  
410 of wine lees, while in order to obtain this same amount of extract from the valorization  
411 of *Sargassum muticum*, it is only necessary to process 1.5 kg of biomass. In particular,  
412 the treatment of wine lees has better environmental results because most of the  
413 operations performed are physical (solid/liquid separations, distillations, evaporations,  
414 etc.) and do not involve the large consumption of electricity or chemicals. There is only  
415 one determinant consumption in the system, the high temperature steam, while in the  
416 rest of the comparative studies, the consumption of electricity and chemicals is  
417 relatively high. Comparative analysis has allowed us to compare the valorization of  
418 wine lees with others related to the production of antioxidants from different raw  
419 materials published in the scientific literature. The valorization of wine lees presents the  
420 best environmental profile in almost all compared categories.

#### 421 **4. Conclusions**

422 Nowadays special interest is being paid into the valorization of different wastes in order  
423 to reduce raw materials consumption. Thus, it has been shown that the integral  
424 valorization of wine lees is a very attractive process to produce value-added products.  
425 Winemaking is a process with a high demand for chemicals, which leads to a high

426 environmental impact. Therefore, this system causes 50% of the total impact of wine  
427 lees valorization. However, on a comparative level with other processes in which an  
428 antioxidant-rich extract can be obtained, it has been demonstrated that the valorization  
429 of wine lees presents the best environmental profile throughout the entire life cycle in  
430 almost all the impact categories studied. However, steam consumption has proven to be  
431 an important hotspot in the process, so it will be necessary to reduce this consumption  
432 in the future. In order to achieve this objective, other residues from the winery, such as  
433 grape stalks, could be used as raw material to obtain high temperature steam in a boiler.

434 Wine waste biorefining is an appropriate way of obtaining products from waste  
435 according to the principles of the Circular Economy, where waste is converted into new  
436 raw materials. This work shows that the LCA methodology is a useful tool for assessing  
437 the environmental impact of wine lees treatment in order to obtain value-added  
438 products. The results of this work should be considered in order to develop a more  
439 sustainable way of obtaining an antioxidant-rich extract from agricultural residues.

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