

26 focus their activities with the aim of making the future of bio-adhesives more technically and
27 environmentally favourable.

28

29 **Keywords:** Bio-based adhesives; Environmental impacts; Life Cycle Assessment; Petrol-based
30 adhesives; Sustainability

31

32 **1. Introduction**

33 The depletion of fossil fuels, the growing concern about the global warming potential and its
34 consequences on environment and human health are driving the development of bioeconomy. In this
35 regard, the production of different chemicals from renewable sources as an alternative to their
36 petrochemical counterparts receives special attention (Sanders et al., 2007; Nitzsche et al., 2016; Yang
37 and Rosentrater, 2019). As consequence, biomass plays a fundamental role, not only in food and feed
38 systems (Sanders et al., 2012; Posada et al., 2013), but also because it opens up a range of production
39 processes of a markedly biotechnological nature. Thus, valorising biomass-based flows in a wide scale
40 of applications that guarantee non-competition with food/feed (Liu et al., 2012; Kolfshoten et al., 2014;
41 Nitzsche et al., 2016; Vargas et al., 2015).

42 Adhesives are chemicals that have been used for many years as binding agents in multiple sectors,
43 such as the automotive, aerospace and wood industries (Yang and Rosentrater, 2019). Regarding the
44 latter, adhesives constitute an important constituent in the manufacture of wood-based panels (Kim,
45 2009). According to GlobeNewswire (2017), the global market for adhesives will reach an annual growth
46 rate of 5% in the period 2017-2022, reaching a market size of \$53.5 billion by 2022, supported by their
47 growing demand for consumer goods. The adhesives market can be segmented by adhesive
48 formulating technology, type of resin, flexibility and mode of application, among others (Ebnesajjad,
49 2009; Yang and Rosentrater, 2020).

50 Traditional adhesives (known as petroleum-based adhesives) are derived from the co-products of
51 petroleum processing and formaldehyde-based adhesives are currently used for the production of
52 wooden flooring, mainly urea-formaldehyde (UF), phenol-formaldehyde (PF) and melamine-urea
53 formaldehyde (MUF). The use of these adhesives (also known as resins) is supported by several
54 features such as low curing temperatures, excellent adhesion properties, water resistance and low price
55 (Kim, 2009; Yang and Rosentrater, 2019). On the contrary, the main drawback of this type of adhesives
56 is the possibility of releasing volatile organic compounds (VOCs) and formaldehyde vapors that harm

57 human health, causing nasal carcinomas as well as eyes and throat irritation, among other adverse
58 effects (Kim, 2009; Li et al., 2009). As a result, and due to the growing demand for adhesives, there is a
59 worldwide effort to develop more environmentally and health –friendly adhesives. In this regard, studies
60 are focused on alternatives to formaldehyde-based adhesives, by partially or completely eliminating
61 formaldehyde from the composition (Pizzi, 2006; McDevitt and Grigsby, 2014; Stefani et al., 2008). Bio-
62 based resins are natural polymers that act as adhesives. In this sense, biomass sources such as soy
63 and corn proteins, tannins, starches, bio-glycerol and Kraft/Organosolv lignin are examples of
64 feedstocks that have been assessed as potential for the production of these chemicals (Langenberg et
65 al., 2010; Moubarik et al., 2009; McDevitt and Grigsby, 2014). Global demand for bio-adhesives is
66 emerging because of their potential environmental and health benefits (Mathias et al., 2016) although
67 the technologies are in their infancy.

68 McDevitt and Grigsby (2014) reported that bio-based adhesives have a 22% lower life cycle impact than
69 those based on petrochemical feedstocks. González-García et al. (2011a) detailed reductions in the
70 global environmental profile of wood-based panel production when PF is substituted by a two-
71 component bio-adhesive formulated with liginosulfonate and phenol-oxidizing enzymes. Regarding
72 health, formaldehyde and VOC emissions should be significantly reduced (Hemmilä et al., 2017).
73 According to Markets and Markets estimates (MarketsandMarkets, 2019), the global market for bio-
74 adhesives is expected to grow at a compound annual growth rate of 10% over the period 2019-2024,
75 reaching a market size of \$9.1 billion, opening an interesting market opportunity.

76 Soy protein is one of the first feedstocks in the commercial production of bio-adhesives used in the
77 wood panel sector as an alternative to PF and UF (Hemmilä et al., 2017). Soy protein is obtained by
78 mechanical or solvent extraction for soybean oil (Li et al., 2004). Soy protein-based adhesive has
79 several advantages, such as low cost, ease of handling, low pressing temperatures, in addition to being
80 used for wood with high moisture content. Nevertheless, this type of bio-adhesive reports poor water
81 resistance and is quite sensitive to biological degradation, among other drawbacks. On the contrary, this

82 feedstock has been considered in recent years as a potential raw material due to its availability and
83 interest worldwide (Li et al., 2004). Lignin-based adhesives are of potential interest as promising
84 strategies for integrating bio-refineries into the wood sector, especially in pulp mills. Lignin is a side-
85 stream from pulp manufacturing processes, which has been commonly used for energy purposes while
86 recovering pulping chemicals (Geng and Li, 2006). Nevertheless, lignin streams are very different from
87 each other in terms of their composition (i.e., sulphur-containing lignin -Kraft and sulphur-free lignin -
88 Organosolv), which opens up a wide range of possibilities. Due to its composition, lignin is structurally
89 similar to phenol (both contain phenolic hydroxyl groups), which makes it possible for lignin to be used
90 as a substitute for phenol in the synthesis of PF (Sellers, 2001; Jin et al., 2010). Nevertheless, further
91 research is required due to its lower reactivity to formaldehyde, which is a major disadvantage in
92 applications where fast curing times are required (Hemmilä et al., 2017). Thus, lignin streams require
93 some chemical modifications to increase their reactivity for the synthesis of bio-resins.

94 Currently, there are industrial applications for tannin-based adhesives, although with limited capacity
95 (Hemmilä et al., 2017) due to the relatively limited supply of tannins (Dababi et al., 2016). Tannins are
96 naturally found in the bark (present at high concentrations in pine, quebracho, oak and chesnut), leaves
97 and fruits of plants and have been widely used in the manufacture of inks and dyes, among other uses
98 (Hemmilä et al., 2017). Tannin extraction can be performed under different extraction methods and
99 influences the adhesive properties of tannin extracts (Saad et al., 2014; Abdalla et al., 2014).

100 Life Cycle Assessment (LCA) is an environmental methodology that can be used to estimate the
101 environmental burdens associated with a product, process or activity throughout its life cycle. This tool
102 has been considered to demonstrate the environmental benefits associated with green products and
103 novel technologies compared to petrol-based ones (Cherubini et al., 2011; Rajagopalan et al., 2012;
104 Collinge et al., 2015). In the context of adhesives and wooden floorings, several studies are also
105 available in the literature that assess the environmental benefits of introducing green processes

106 (González-García et al., 2011a; McDevitt and Grigsby, 2014; Yang and Rosentrater, 2019a; 2019b;
107 2020).

108 The aim of this study is to use LCA to quantify the environmental profiles associated with the production
109 of different bio-adhesives considering a cradle-to-factory gate approach. Among the bio-based
110 alternatives, tannin, lignin (Kraft and Organosolv) and soybean meal have been considered as
111 feedstocks for the bio-adhesives production. In addition, the main responsible factors will be identified
112 and the environmental benefits and/or disadvantages linked to bio-adhesives in comparison with
113 conventional fossil-adhesives will be highlighted. The four production systems were modelled from
114 laboratory experiments to large-scale. Regarding the conventional fossil resins, UF, PF and MUF were
115 selected for comparison.

116

117 **2. Methodology**

118 Life Cycle Assessment (LCA) is an environmental and management approach that takes into account all
119 the aspects of resource use and environmental emissions associated with the manufacture of a product
120 under study throughout its life cycle. Accordingly, this tool can be used to support decision-making for
121 sustainable environmental development (ISO 14040, 2006).

122

123 **2.1. Goal and scope definition**

124 The goal of this LCA study is to provide an overview of the large-scale production of four different bio-
125 adhesives, as well as to determine the yields and environmental improvements compared to petroleum-
126 based adhesives. To this end, bio-adhesives production systems have been modelled at full-scale at 40
127 t/d based on laboratory-scale data (Luo et al., 2015; Navarrete et al., 2010; Geng et al., 2006; Dongre et
128 al., 2015; Li et al., 2004). The selection of this capacity is based on the fact that commercial
129 formaldehyde-based resin plants have a capacity close to 15,000 t/year (Yang and Rosentrater, 2019a).
130 An attributional cradle-to-factory gate approach has been considered, with soybean meal, Kraft lignin,

131 Organosolv lignin and pine tannin as raw materials. Accordingly, the estimated environmental profiles
132 will be those derived from the processes and the mass and energy flows involved throughout their life
133 cycles. Therefore, energy and mass balances have been performed for the modeling of full-scale
134 production systems in order to gather all the required data for the Life Cycle Inventory stage. Plant-wide
135 modelling was conducted using Aspen Plus® software not only to identify the inventory data, but also to
136 design the equipment involved in each system according to the required conditions.

137 The functional unit used to report and compare the environmental results is 1 kg of adhesive. This
138 choice is also considered in other related studies and allows the corresponding comparison of
139 environmental profiles (Yang and Rosentrater, 2019b). As far as the system boundaries are concerned,
140 all the stages from raw materials extraction, adhesives manufacture and waste management have been
141 included, excluding from the analysis the transport activities involved in the distribution of the chemicals
142 and feedstock up to the factory gate. The rationale behind this assumption is twofold: the lack of
143 valuable data and the findings of other related studies (Yang and Rosentrater, 2019a; 2019b; 2020)
144 where the effect of transportation over the life cycle environmental impacts was not significant.
145 Moreover, it is expected that waste treatment facilities will be located in the vicinity of the plants.

146

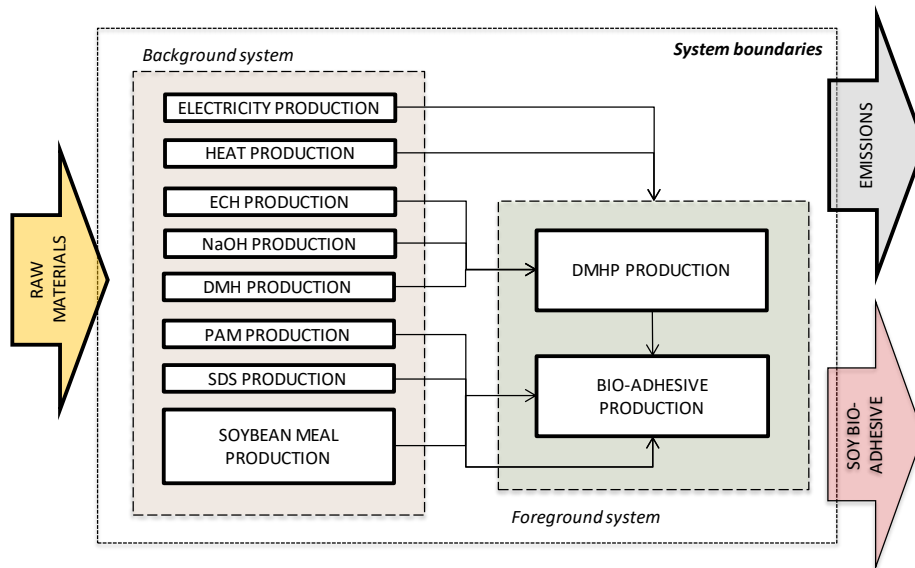
147 **2.2. Description of Bio-adhesives production systems and inventory analysis**

148 As detailed above, four different scenarios have been considered for the analysis based on different
149 biomass sources and production conditions.

150 *Soy-based bio-adhesive*

151 The production system considered for the analysis is based on the use of a cross-linker in order to
152 improve water resistance. Among the cross-linkers, 5,5-Dimethyl hydantoin polyepoxide (DMHP) reports
153 multiples useful properties (Luo et al., 2015) and has therefore been taken into consideration. The
154 overall foreground production system is divided into two main stages: DMHP production and Bio-
155 adhesive production as detailed in **Figure 1a**. Accordingly, the production chain starts with the soybean

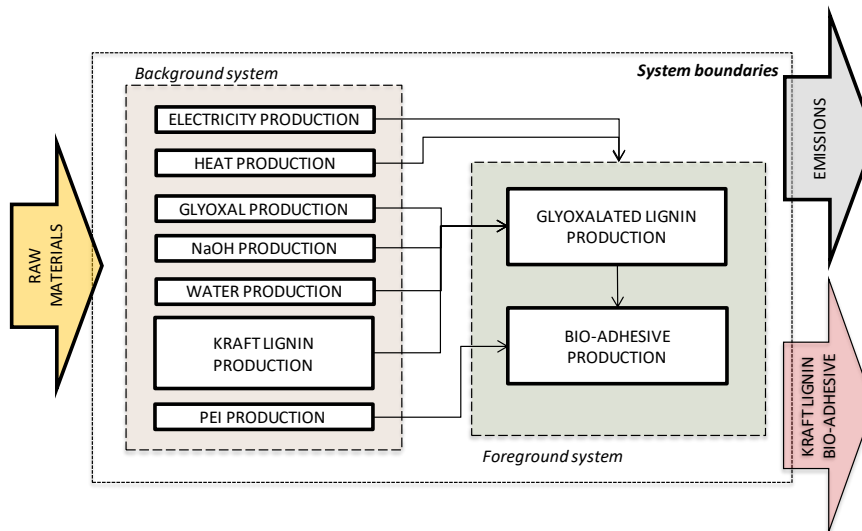
156 cultivation and processing in order to obtain the soybean meal. In this case, the production of the cross-
 157 linker takes place in the plant itself following the procedure detailed by Luo et al. (2015).



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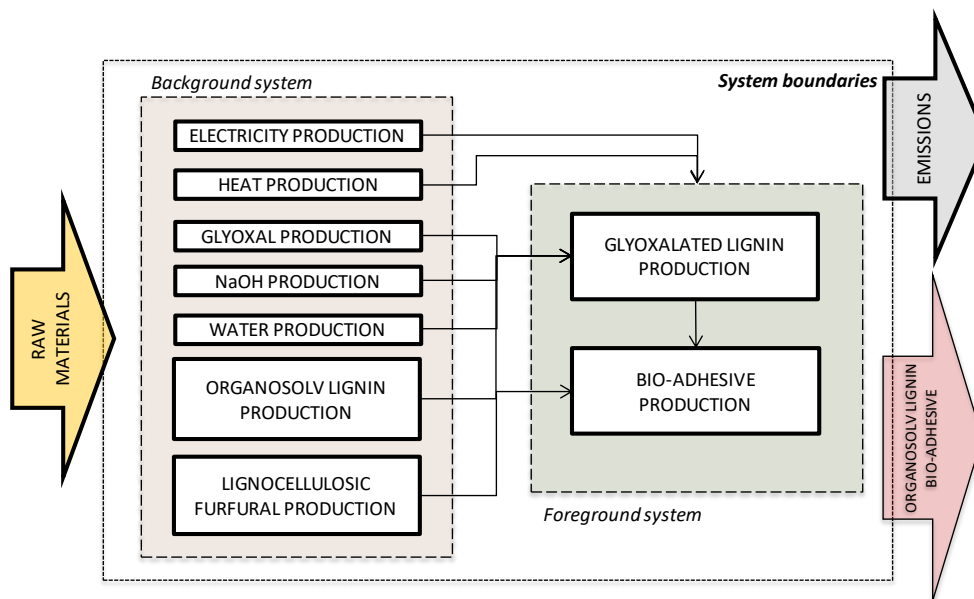
Figure 1a



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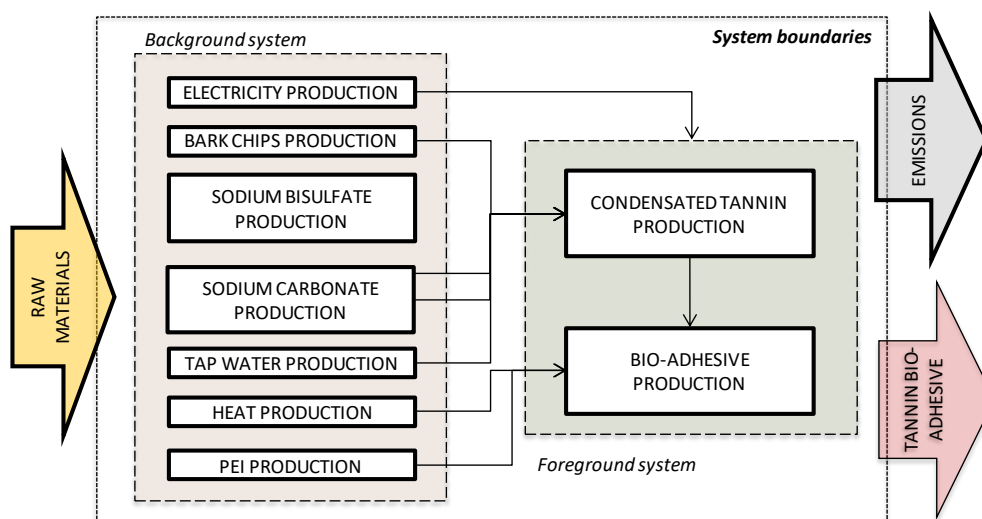
Figure 1b



162

163

Figure 1c



164

165

Figure 1d

166 **Figure 1.** System boundaries of the bio-adhesives production on a real scale. a) Soy-based bio-adhesive; b) Kraft lignin-
 167 based bio-adhesive; c) Organosolv lignin-based bio-adhesive; d) maritime pine tannin-based bio-adhesive.

168 Epichlorohydrin (ECH), sodium hydroxide solution (NaOH, 50% wt) and 5,5-Dimethyl hydantoin (DMH)
 169 are mixed at 80°C for 1 h at a molar ratio of 1:8:1 (DMH/ECH/NaOH). The bio-adhesive production
 170 process is carried out at 20°C for 10 min in a stirred that incorporates soy protein, DMHP,
 171 polyacrylamide (PAM) and sodium dodecyl sulfate (SDS). The management of residual streams
 172 produced in both stages in which the production process is classified have been computed within the

173 system boundaries, that are wastewater treatment, landfilling and incineration. The data corresponding
 174 to the foreground system have been determined by means of plant-wide modelling in Aspen Plus® and
 175 a summary of the most relevant mass and energy flows is detailed in **Table 1**.

176 **Table 1.** Global Life Cycle Inventory data corresponding to the foreground system for the production of soy-based
 177 bio-adhesive. Data are reported per functional unit (1 kg bio-adhesive).
 178

INPUTS FROM TECHNOSPHERE		OUTPUTS TO TECHNOSPHERE	
DMHP production			
<i>Materials</i>		<i>Products</i>	
NaOH solution 50%wt	3.10 g	DMHP to Bio-adhesive production	0.141 kg
ECH	18.06 g	<i>Waste to treatment</i>	
DMH	0.988 g	Wastewater	1.69 mL
<i>Energy</i>		Landfill	0.31 g
Electricity	4.23 Wh		
Heat	1.86 kJ		
Bio-adhesive production			
<i>Materials</i>		<i>Products</i>	
Soybean meal	0.268 kg	Bio-adhesive	1.00 kg
DMHP from DMHP production	0.141 kg	<i>Waste to treatment</i>	
PAM	0.690 kg	Incineration	97.11 g
SDS	12.90 g	Landfill	14.11 g
<i>Energy</i>			
Electricity	30.30 Wh		
Heat	12.04 kJ		

179 NaOH - Sodium Hydroxide; ECH – Epichlorohydrin; DMH - 5,5-Dimethyl hydantoin; DMHP - 5,5-Dimethyl hydantoin polyepoxide; PAM –
 180 Polyacrylamide; SDS - Sodium dodecyl sulfate.

181
 182 *Kraft lignin based bio-adhesive*

183 Among lignins, Kraft lignin is the most abundant in pulp mills. This type of lignin is insoluble in water and
 184 most solvents, except in highly alkaline environments (Hemmilä et al., 2017). Research is being
 185 conducted with the aim of totally replacing PF in board manufacture with a combination with glyoxal
 186 (Mansouri et al., 2007; Navarrete et al., 2012). Thus and bearing in mind that lignin streams require a
 187 chemical modification, the use of glyoxalated lignin should allow the production of boards with good
 188 internal bond strength characteristics (Hemmilä et al., 2017). Therefore, this strategy has been taken into
 189 consideration in the modelling of the Kraft lignin based bio-adhesive. The general production system in
 190 the foreground is divided into two main steps which are the glyoxalation of Kraft lignin and the

191 production of bio-adhesives as detailed in **Figure 1b**. Accordingly, the production chain starts with the
 192 production of Kraft hardwood lignin at the pulp mill, where the bio-adhesive-based biorefinery strategy
 193 should be incorporated. It is considered that lignin is precipitated from the black liquor of the pulp mill
 194 under acidic conditions (mainly, with hydrochloric acid - HCl), washed and dried. An additional alkaline
 195 treatment is then required considering an aqueous solution of NaOH at 180°C. The lignin suspension
 196 after pulping is filtered and the liquid phase obtained is subjected to precipitation by acidification with
 197 HCl. The procedure reported by Radoykova et al. (2013) has been considered. The lignin powder is
 198 then glyoxalated in a reaction with water, sodium hydroxide (30%) and glyoxal (40%) solutions in a
 199 stirrer for 8 h at 58°C (Navarrete et al., 2010). The cost of this step could be reduced by understanding
 200 how to optimize the amounts of glyoxal and NaOH. Glyoxal lignin is further mixed with polyethylenimine
 201 (PEI) in a stirrer for 40 min at 20°C and a lignin/PEI weight ratio of 5:1 (Geng and Li, 2006).

202 **Table 2.** Global Life Cycle Inventory data corresponding to the foreground system for the production of Kraft
 203 lignin-based bio-adhesive. Data are reported per functional unit (1 kg bio-adhesive).
 204

INPUTS FROM TECHNOSPHERE		OUTPUTS TO TECHNOSPHERE	
Glyoxalated lignin production			
<i>Materials</i>		<i>Products</i>	
NaOH	40.90 g	Glyoxalated lignin to <i>Bio-adhesive production</i>	0.741 kg
Water	0.469 kg	<i>Waste to treatment</i>	
Glyoxal	33.90 g	Wastewater	48.9 mL
Kraft lignin	0.279 kg	Incineration	27.90 g
<i>Energy</i>		<i>Emissions into air</i>	
Electricity	12.52 kWh	Glyoxal	3.40 g
Heat	102.9 kJ		
Bio-adhesive production			
<i>Materials</i>		<i>Products</i>	
Glyoxalated lignin from <i>Bio-adhesive production</i>	0.741 kg	Bio-adhesive	1.00 kg
PEI	0.370 kg	<i>Waste to treatment</i>	
<i>Energy</i>		Incineration	0.11 kg
Electricity	740 Wh		
Heat	102.9 kJ		

205 NaOH - Sodium Hydroxide; PEI - Polyethylenimine.

206 *Organosolv lignin based bio-adhesive*

207

208 Waste streams produced in both stages and their management in wastewater treatment plants and
209 incineration facilities have been computed within the system boundaries. **Table 2** summarizes data
210 corresponding to the foreground system at full-scale.

211 As detailed above, there are several types of lignins depending on the pulping process. Organosolv
212 lignin is essentially water-insoluble and is obtained in non-sulphur biorefineries, closely related to the
213 cellulosic biofuel industry (Hemmilä et al., 2017). Following the structure and design proposed for the
214 analysis for the Kraft-lignin based bio-adhesive, the production scenario has been classified into two
215 steps that are the production of glyoxalated lignin and the production of bio-adhesive. Navarrete et al.
216 (2010), Mansouri et al. (2007) and Lei et al. (2008) developed the glyoxalation of organosolv lignin to
217 obtain synthetic resin-free adhesives. The glyoxalation process described by Navarrete et al. (2010) has
218 been followed in detail according to the Kraft lignin based case study. The system under study starts
219 with the production of Organosolv softwood lignin in a biorefinery, following the approach reported by
220 Kautto et al. (2013). Secondly, lignin is mixed with water, NaOH (30%) and glyoxal (40%) to be
221 glyoxalated. Glyoxalated lignin is finally mixed with furfural (as a substitute for formaldehyde) under
222 acidic conditions and at 90°C for 1 h. Furfural has been identified by the US Department of Energy as
223 one of the top value-added products (Luo et al., 2019). The production of lignocellulosic furfural derived
224 from biomass under a biorefinery approach has been taken into account following the production
225 procedure reported by Bello et al. (2018). **Figure 1c** depicts the system boundary corresponding to this
226 scenario and **Table 3** summarizes the inventory data of the foreground system.

227

228 *Pine tannin based bio-adhesive*

229 In this scenario, the production system comprises the processes of condensed tannin production and
230 tannin-based bio-adhesive production as detailed in **Figure 1d**. Condensed tannin (flavonoid polymer)
231 commercially produced from maritime pine bark has been considered as the main raw material and
232 information regarding its production has been taken from González-García et al. (2016). Concerning the

233 bio-adhesive, the production process has been modelled following the lab experiment developed by
 234 Zhang et al. (2019). The background processes involved in the production of the condensed tannin,
 235 including the forest activities and the extraction stage, have been computed, as well as the
 236 management of derived waste produced in the bio-adhesive plant. **Figure 1d** depicts the system
 237 boundary and **Table 4** summarizes the most representative inventory data for the foreground system.

238 **Table 3.** Global Life Cycle Inventory data corresponding to the foreground system for the production of
 239 organosolv lignin-based bio-adhesive. Data are reported per functional unit (1 kg bio-adhesive).
 240

INPUTS FROM TECHNOSPHERE		OUTPUTS TO TECHNOSPHERE	
Glyoxalated lignin production			
<i>Materials</i>		<i>Products</i>	
NaOH	58.30 g	Glyoxalated lignin to <i>Bio-adhesive production</i>	1.06 kg
Water	0.668 kg	<i>Waste to treatment</i>	
Glyoxal	48.30 g	Wastewater	48.9 mL
Organosolv lignin	0.398 kg	Incineration	39.8 g
<i>Energy</i>		<i>Emissions into air</i>	
Electricity	30.35 kWh	Glyoxal	4.80 g
Heat	146.6 kJ		
Bio-adhesive production			
<i>Materials</i>		<i>Products</i>	
Glyoxalated lignin from <i>Glyoxalated lignin production</i>	1.06 kg	Bio-adhesive	1.00 kg
Furfural	56.00 g	<i>Waste to treatment</i>	
<i>Energy</i>		Incineration	0.112 kg
Electricity	2.31 kWh		
Heat	10.47 kJ		

241 NaOH - Sodium Hydroxide.

242

243 Once the scenarios have been modelled, several assumptions have been considered to complete the
 244 inventory data. As detailed above, information from different experiments performed at lab scale
 245 (procedures, equations, equipment) has been considered to design full-scale bio-adhesives production
 246 systems. Thus, the stoichiometric amounts of each chemical considering the lab protocols have been
 247 computed in the inventory data assuming a 90% production yield in all experiments. Whenever possible,
 248 primary data were considered in the inventory data collection. However, it has been necessary to handle
 249 secondary data specifically for the background system (see **Figure 1**). The background system involves

250 the activities required to produce the utilities (electricity and heat) and all the inputs (i.e. chemicals, tap
 251 water and biomass sources) required in the foreground system. Thus, the Ecoinvent® database version
 252 3.5 (Wernet et al., 2016) has been considered as the main secondary data source. This database has
 253 been also considered to inventory the processes involved in solid waste and wastewater treatment
 254 linked to each scenario.

255 **Table 4.** Global Life Cycle Inventory data corresponding to the foreground system for the production of pine
 256 tannin-based bio-adhesive. Data are reported per functional unit (1 kg bio-adhesive).
 257

INPUTS FROM TECHNOSPHERE		OUTPUTS TO TECHNOSPHERE	
Condensed-tannin production			
<i>Materials</i>		<i>Products</i>	
Bark chips	0.024 m ³	Condensed tannin to <i>Condensed-tannin production</i>	0.788 kg
Sodium bisulfate	0.180 kg		
Sodium carbonate	45.03 g		
Tap water	3.95 g		
<i>Energy</i>			
Electricity	1.97 kWh		
Bio-adhesive production			
<i>Materials</i>		<i>Products</i>	
Condensed tannin from <i>Condensed-tannin production</i>	0.157 kg	Bio-adhesive	1.00 kg
Furfuryl alcohol	0.088 kg	Emissions into air	
Glyoxal	0.394 kg	Furfuryl alcohol	0.047 g
Tap Water	0.216 kg	Glyoxal	10.63 g
Acetic acid	0.036 kg	Tap Water	1.13 g
Epoxy resin, E-44	0.121 kg	Acetic acid	0.15 g
<i>Energy</i>			
Electricity	0.001 kWh		
Heat	463.05 kJ		

258 PEI - Polyethylenimine.
 259

260 Finally, the production of FP, UF and MUF has been taken into account to compare the profiles between
 261 bio-adhesives and petrol-based ones. Once again, the inventory data corresponding to the background
 262 processes involved in the petroleum-based adhesive production system have been taken from the
 263 Ecoinvent® version 3.5 database. A summary of the Ecoinvent® processes considered in the
 264 formulation of the scenarios under evaluation is reported in **Table 5**.

265 Regarding Kraft lignin, Organosolv lignin and condensed tannin production, the data corresponding to
 266 their foreground systems have been taken from bibliographic sources as detailed in Section 2.3.
 267 Nevertheless, the background processes associated with the production of, for example, the required
 268 chemicals and energy have been taken from the Ecoinvent® database.

269 **Table 5.** Description of the main Ecoinvent® database version 3.2 processes considered in this study for the
 270 background processes.

Input	Process
Energy	
Electricity	Electricity, medium voltage {ES} market for Alloc Rec, U
Heat	Heat, district or industrial, natural gas {RER} market group for Cut-off, U
Chemicals	
Sodium hydroxide	Sodium hydroxide, without water, in 50% solution state {GLO} market for Cut-off, U
Epichlorohydrin	Epichlorohydrin {GLO} market for Cut-off, U
5,5-Dimethyl hydantoin polyepoxide	Imidazole {GLO} market for Cut-off, U
Sodium dodecyl sulfate	Sodium sulfate, anhydrite {RER} market for Cut-off, U
Polyacrylamide	Polyacrylamide {GLO} market for Cut-off, U
Tap water	Tap water {Europe without Switzerland} market for Cut-off, U
Sodium bisulfate	Sodium sulfate, anhydrite {RER} market for Cut-off, U
Sodium carbonate	Sodium carbonate from ammonium chloride production, at plant/GLO U
Polyethylenimine	Ethylenediamine {RER} market for ethylenediamine Cut-off, U
Glyoxal	Glyoxal {RER} market for glyoxal Cut-off, U
Acetic acid	Acetic acid, without water, in 98% solution state {GLO} market for Cut-off, U
Bisphenol A	Bisphenol A, powder {GLO} market for Cut-off, U
Feedstocks	
Soybean meal	Soybean meal {GLO} market for Cut-off, U
Roundwood	Roundwood, pine from sustainable forest management, under bark {GLO} market for Cut-off, U
Waste management	
Solid waste treatment	Biowaste {GLO} treatment of biowaste, municipal incineration Cut-off, U
	Hazardous waste, for underground deposit {GLO} market for Cut-off, U
Wastewater treatment	Wastewater, average {Europe without Switzerland} market for wastewater, average Cut-off, U

272 As regards the cleaning activities involved in each production system, they have been excluded from
273 the system boundaries. The rationale behind their exclusion is twofold: a) the lack of information on this
274 step and b) the final goal of our study is to compare different bio-adhesive production scenarios and the
275 effect on their profiles of the cleaning stage could be considered the same. As for the infrastructure
276 process, it has not been included within the system boundaries considering that the impacts from
277 construction, decommissioning and equipment maintenance is negligible over the environmental profiles
278 derived from each biorefinery plants under study according to Bello et al. (2018), González-García et al.
279 (2011b) and Uihlein and Schebek (2009). Finally, the production of PF, UF and MUF has been taken
280 into account in order to compare the profiles between bio-adhesives and petrochemical ones, the
281 inventory data for the background processes of the petrochemical adhesives have taken from the
282 Ecoinvent® database version 3.5 (PF and UF) and Silva et al. (2015) (MUF).

283

284 **2.3. Environmental Assessment methods**

285 Two impact assessment methods will be considered in the analysis. Firstly, the ReCiPe 2016 hierarchist
286 Midpoint method V1.03 World (2010) (Huijbregts et al., 2017) has been used for the selection of
287 characterization factors required to estimate the environmental burdens and a set of impact categories
288 at midpoint level has been considered to report the environmental profiles. Regarding the set of impact
289 categories, the following impacts have been considered: global warming (GW), stratospheric ozone
290 depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication
291 (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil
292 resource scarcity (FRS). Secondly, the ReCiPe 2016 hierarchist Endpoint method V1.03 World (2010)
293 H/H (Huijbregts et al., 2017) has been considered to compare the profiles of bio-adhesives with those of
294 fossil-adhesives. The choice of this end-point approach is based on the fact of having only one single
295 score for the overall impacts that can help disseminate the message to stakeholders. The SimaPro

296 software v9.0 (PRéConsultants, 2020) has been used for the computational implementation of the life
 297 cycle inventories.

298

299 3. Results and discussion

300 The results of the life cycle impact assessment using ReCiPe 2016 hierarchist method V1.03 method
 301 are shown in **Table 6**. Bearing in mind the results, there is a remarkable difference on the profiles of bio-
 302 adhesive production depending on the feedstock considered. Consequently, the bio-adhesive from
 303 Organosolv lignin should be the worst choice from an environmental perspective, since the burdens for
 304 all the categories (except in ME) are higher than those of the other scenarios. In terms of ME, soy-
 305 based bio-adhesives would be the worst alternative. In general, the soybean-based and tannin-based
 306 bio-adhesives have the best profiles, as they have the lowest loads in all impact categories.

307 **Table 6.** Environmental characterization results estimated per 1kg of bio-adhesives considered in the study.

308

		Soy	Kraft lignin	Organosolv lignin	Tannin
Global Warming (GW)	kg CO ₂ eq	2.79	8.34	15.54	2.97
Stratospheric Ozone Depletion (SOD)	mg CFC11 eq	2.50	4.33	8.49	1.45
Terrestrial Acidification (TA)	g SO ₂ eq	9.78	38.17	77.16	11.62
Freshwater Eutrophication (FE)	g P eq	0.47	6.89	15.26	1.11
Marine Eutrophication (ME)	g N eq	2.07	1.79	1.08	0.09
Terrestrial Ecotoxicity (TET)	kg 1,4-DCB	6.36	13.20	16.52	6.59
Freshwater Ecotoxicity (FET)	g 1,4-DCB	44.3	277.4	560.2	63.4
Marine Ecotoxicity (MET)	g 1,4-DCB	59.7	380.2	759.0	86.0
Fossil Resource Scarcity (FRS)	kg oil eq	1.07	2.49	3.94	1.18

309

310 Thus, the contributions to the environmental burdens per scenario in each of the selected impact
 311 categories are presented below with the aim of identifying the corresponding environmental hotspots
 312 and proposing further improvements. Once described the analysis per bio-adhesive, the comparison
 313 with the conventional alternatives will be performed.

314

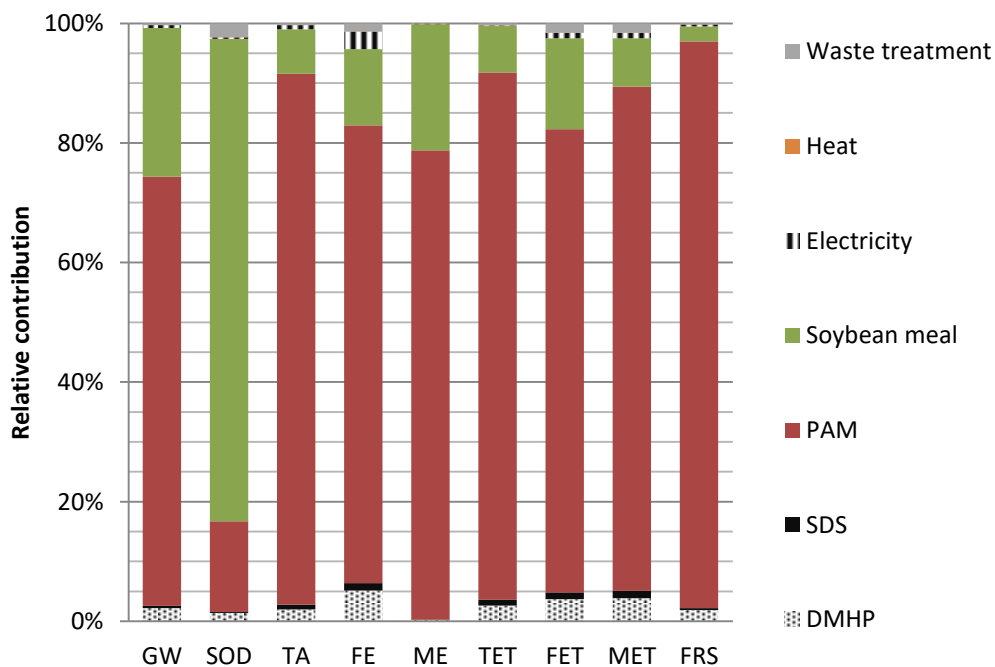
315 **3.1. Environmental performance of Soy based bio-adhesive**

316 The characterization results corresponding to soy-based adhesive are shown in **Figure 2a**. Taking into
317 account this profile, the PAM production required in the formulation can be appointed as the most
318 burdensome input with shares ranging from 72% to 95%, except for SOD, where the production of
319 soybean meal is the environmental hotspot (81% of the total). The contributions from PAM is really
320 outstanding and therefore, further research should be undertaken to propose an alternative chemical in
321 the bio-adhesive formulation (if possible) or to optimize the system, as a reduction of the PAM dosis
322 should have a direct effect on the global profile.

323 Soybean meal production involves agricultural activities in the field to produce the soybeans and
324 subsequent processing, in which beans are cleaned to remove foreign material and pre-treated with
325 heat to remove the hulls from the surface (i.e. to increase the protein content) and then, dried and
326 flaked. Finally, oil is extracted from the soy flakes with a solvent to produce the soybean meal
327 (Soymeal.org, 2020). The production of the raw material also reports outstanding contributions (in
328 addition to SOD) to GW (25%), FE (13%), ME (21%) and FET (15%). The rationale behind these
329 burdens is associated with the requirement of fertilizers in agricultural activities and the corresponding
330 on-field emissions (effect on FE and ME), as well as with the energy requirements in the processing
331 plant (effect on GW, SOD and FET).

332 The contributions of the other processes involved (SDS, electricity, heat and waste treatment) are
333 always below 2%. The effect on the overall production profile of the cross-linker required to improve the
334 water resistance of the adhesive, i.e. DMHP, is also not remarkable, with contributions lower than 5%.
335 Nevertheless, and given that it is an activity that takes place within the factory, as detailed in **Figure 1a**,
336 it has been analysed in detail in order to identify the responsible inputs to its derived environmental
337 burdens. **Figure 2b** details the distribution of burdens per impact category among the contributing
338 factors. Having in mind this figure, the impacts can be mainly assigned to one specific input that is,
339 ECH. The effect of the production of ECH requirements ranges around 80% in all categories, except in

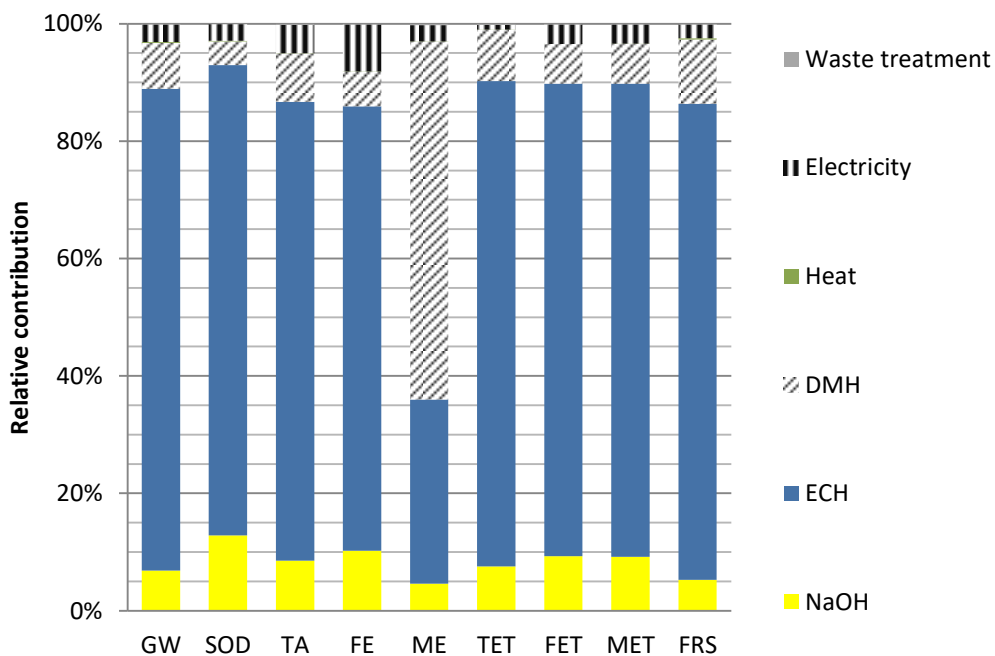
340 terms of ME (31%), where the burdens are dominated by the production of DMH (61%) due to
 341 ammonium and nitrate emissions into water. Once again, the production of energy requirements (heat
 342 and electricity) as well as management of waste do not report an outstanding effect on the global profile.



343

344

Figure 2a



345

346

347

Figure 2b

348 **Figure 2.** Distribution of burdens between contributing inputs/outputs involved. a) Soy-based bio-adhesive production; b)
349 DMHP production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA),
350 freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine
351 ecotoxicity (MET) and fossil resource scarcity (FRS).

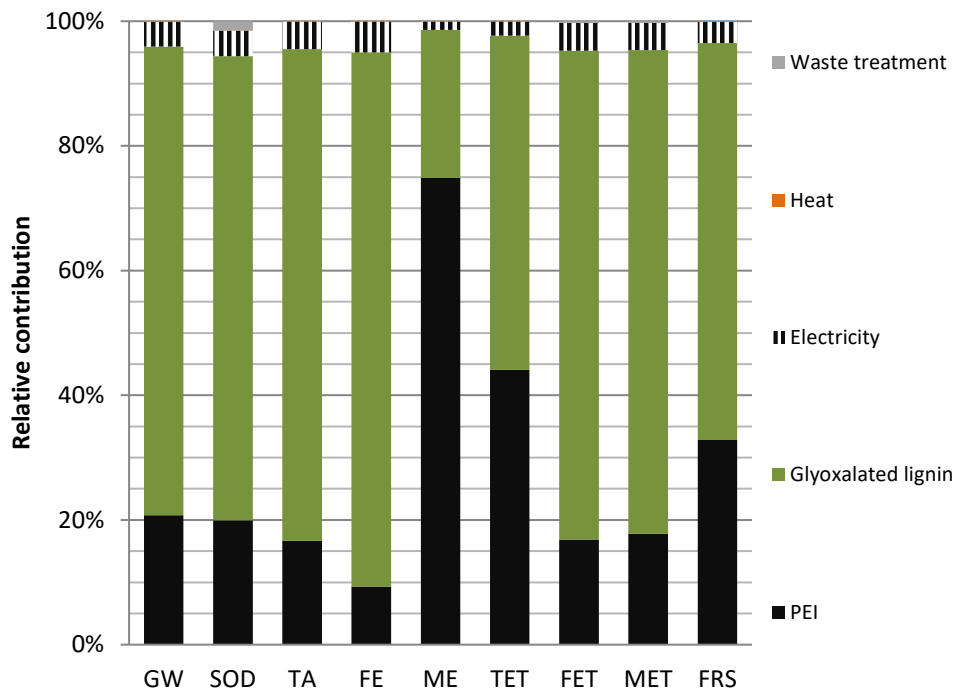
352 **3.2. Environmental performance of Kraft lignin based bio-adhesive**

354 Lignin is one of the most abundant renewable natural polymers, with Kraft lignin being the most
355 abundant industrial lignin (Geng and Li, 2015). Although it is usually burned for energy purposes, it has
356 multiple applications, with the bio-adhesive production being one of the most promising options. In this
357 system, the production of Kraft lignin based bio-adhesive has been environmentally evaluated and
358 **Figure 3a** depicts the distribution of environmental burdens per contributing parameters. Having in mind
359 this figure, the production of the glyoxalated lignin is the largest responsible for environmental impacts,
360 regardless of the category assessed, which implies contributing ratios ranging from 54% to 86% (except
361 in ME, where the ratio decreases to 24%). This material involves the glyoxalation step of Kraft lignin
362 within the factory and therefore requires specific analysis. The lignin glyoxalation is required as a pre-
363 treatment step so that it can be chemically modified to enhance its further reactivity in the synthesis of
364 bio-based resins (Ang et al., 2015). In this case study, glyoxal – a non-toxic and non-volatile dialdehyde
365 – in combination with NaOH has been considered under the conditions outlined by Navarrete et al.
366 (2010). This step demands large amount of electricity, since the required stirring is 8 h. As a
367 consequence, the production of this electricity requirement, which is taken directly from the grid, is by far
368 the main parameter responsible for the environmental burdens derived from lignin glyoxalation, as
369 shown in **Figure 3b**.

370 The effect of this parameter on the glyoxalated lignin profile is remarkable in all the categories with
371 ratios above 72%. Nevertheless, the effect of the chemicals, heat to maintain temperature (around

372

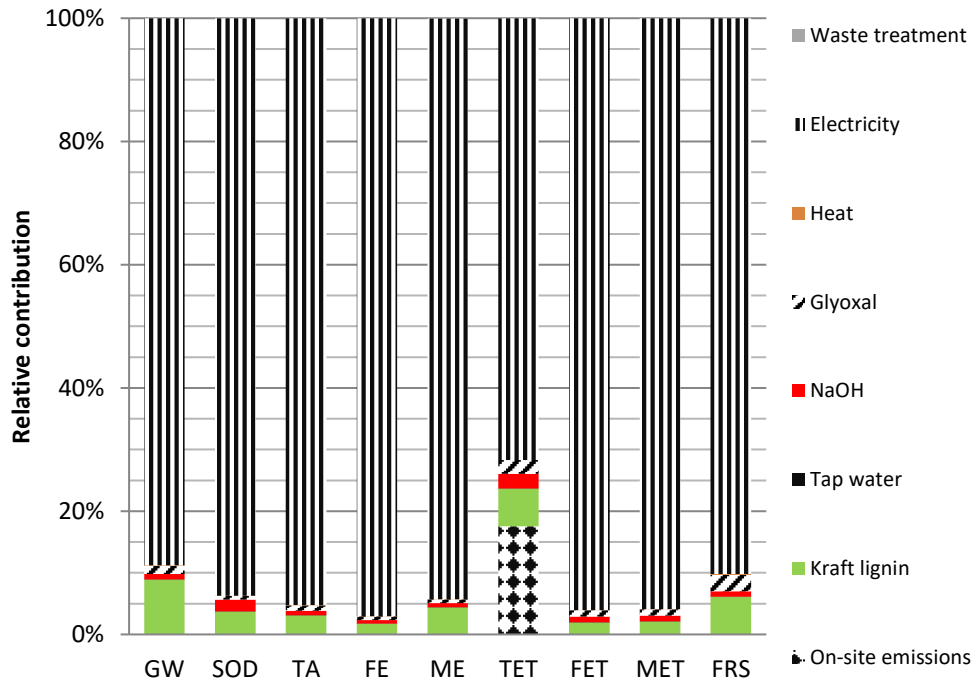
58°C) and waste treatment is negligible, as depicted in **Figure 3b**.



373

374

Figure 3a



375

376

Figure 3b

377 **Figure 3.** Distribution of burdens between contributing inputs/outputs involved. a) Kraft lignin-based bio-adhesive production;

378 b) Glyoxalated lignin production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial

379 acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater
380 ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

381 Conversely, attention must be also paid to on-site emissions from glyoxalation. These are uncontrolled
382 glyoxal emissions into air which considerably affect TET with a contributing ratio of 18%. Finally, Kraft
383 lignin used as raw material presents a considerable low uniform distribution in the overall profile
384 (contributing ratios up to 10%).

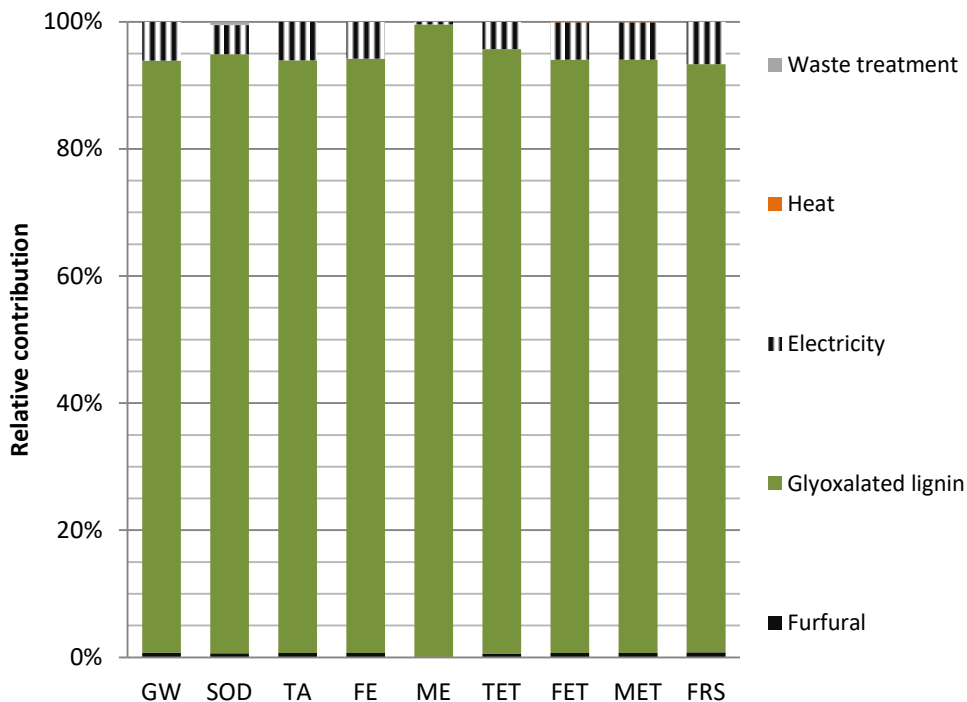
385 To conclude, the effect on the profile derived from Kraft lignin based bio-adhesive from heat and
386 electricity requirements as well as from waste treatment is negligible (see **Figure 3a**). On the contrary,
387 the production of PEI required in the formulation of the bio-adhesive plays a key role with ratios close to
388 20% in several categories except in ME (due to ammonium and nitrate emissions in water), TET (due to
389 copper emissions into air) and FRS (crude oil and natural gas demand) where they amount to 75%,
390 44% and 33%, respectively, due to the background processes involved in the manufacture. PEI is a
391 synthetic organic chemical derived from aziridine (derived from petroleum). However, as research
392 activities focused on the development of platform chemicals from renewable sources, it could be
393 expected a future synthesis of aziridine from non-petroleum feedstocks (Williams and Hillmyer, 2007).
394 Thus, further improvements in reducing uncontrolled on-site emissions should be made to improve the
395 profile in terms of ME by optimizing the dose of PEI required and the electricity consumption in the lignin
396 glyoxalation step. These issues will be discussed in detail below.

397

398 **3.3. Environmental performance of Organosolv lignin based bio-adhesive**

399 Organosolv is an interesting and promising chemical pretreatment in pulp manufacturing technologies
400 that consists of contacting the feedstock with an aqueous solution of an organic compound (mainly,
401 ethanol or methanol because their lower costs) to make lignin and hemicellulosic fractions more soluble.
402 This lignin is obtained through a less aggressive process (de la Torre et al., 2013) and is proposed to be
403 used as main raw material in this scenario to produce the bio-adhesive. According to the kraft lignin-
404 based system, lignin is required to be glyoxalated.

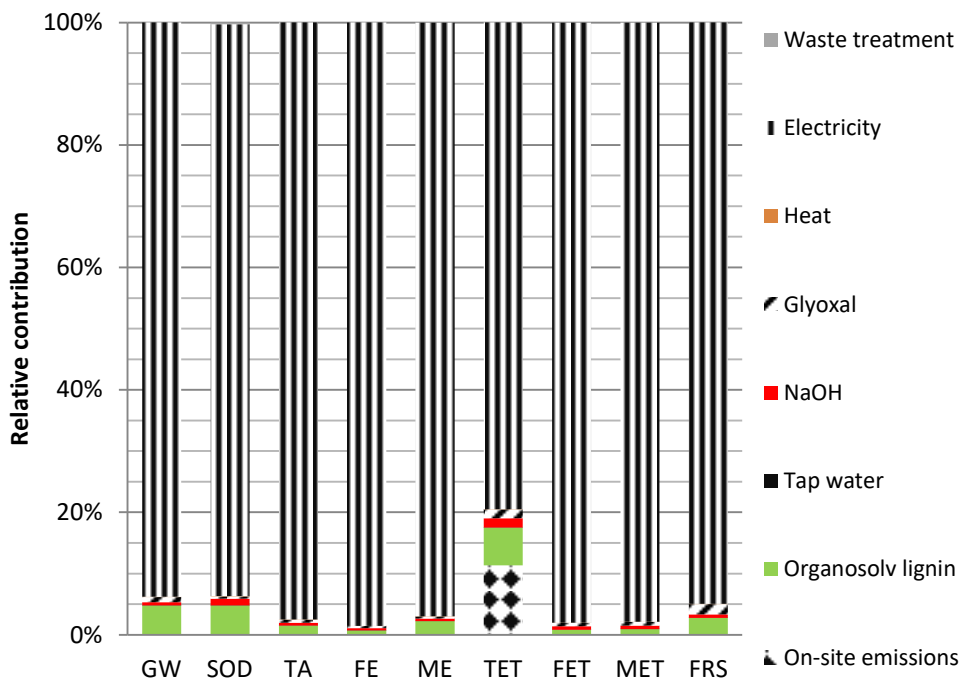
405 **Figure 4a** depicts the distribution of burdens per impact category for the Organosolv lignin bio-
406 adhesive. The production stage focusing on the lignin glyoxalation is the largest contributor to all impact
407 categories, with contributing ratios of 85% on average, followed by the electricity requirements (ratios
408 lower than 6% in all impacts). Thus, the effect on the environmental profile of furfural consumption, heat
409 requirement and waste treatment is negligible. Accordingly, it is necessary to analyse in detail the
410 burdens derived from the production of glyoxalated lignin with the aim of identifying the hotspot and
411 proposing further improvements (if possible). **Figure 4b** shows the distribution of burdens among the
412 processes involved in the production of the glyoxalated lignin and, bearing in mind these results, the
413 production of the organosolv lignin from a bioethanol-based biorefinery (Kautto et al., 2013) does not
414 contribute much to the overall profile, as it involves contributing ratios ranging from 1% to 6% depending
415 on the category. Nevertheless, the production of electricity requirements to perform the glyoxalation can
416 be considered as the environmental hotspot, with an outstanding effect on the profile. The effect of other
417 processes involved is also negligible, as depicted in **Figure 4b**. It is also interesting to highlight the
418 effect that glyoxal air-emissions produced on the glyoxalation have on TET (11% of total contributions to
419 this impact category). Thus, attention should be paid to the optimization of electricity requirements in the
420 equipment involved in the production of glyoxalated lignin and, the reduction of on-site glyoxal
421 emissions. Both issues will be considered under a sensitivity analysis in order to identify potential
422 improvements on the profile.



423

424

Figure 4a



425

426

Figure 4b

427 **Figure 4.** Distribution of burdens between contributing inputs/outputs involved. a) Organosolv lignin-based bio-adhesive
 428 production; b) Glyoxalated lignin production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD),

429 terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET),
430 freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

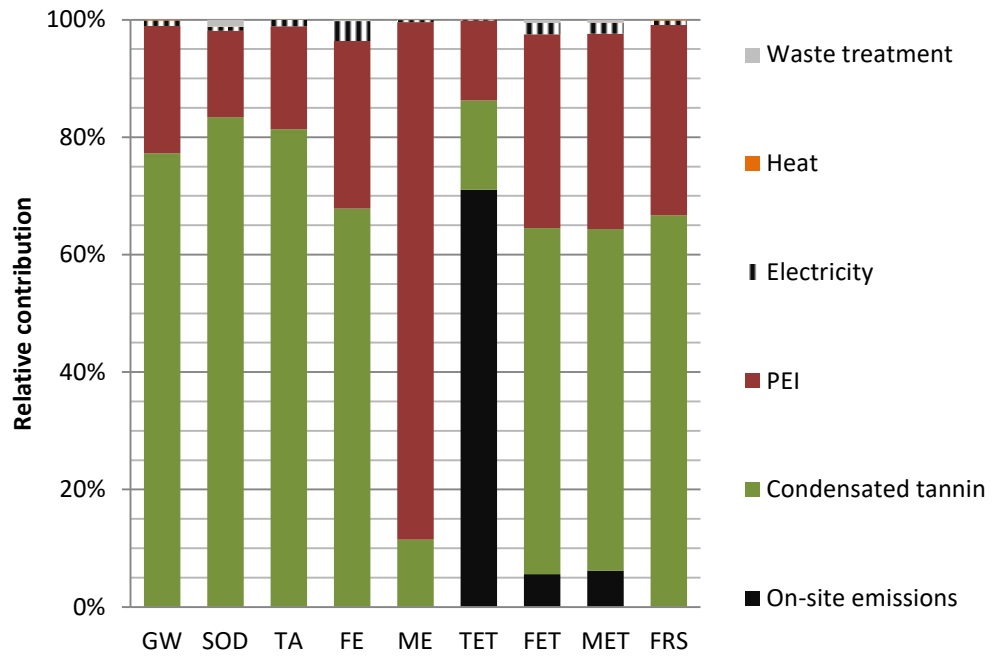
431

432 **3.4. Environmental performance of Tannin based bio-adhesive**

433 **Figure 5a** depicts the distribution of burdens between the impact categories for this resin, where
434 condensed tannin is used as raw material in the production of bio-adhesive. The results show that two
435 parameters are by far responsible for the largest contributions to the environmental profile, regardless of
436 the impact category, i.e. the production of condensed tannin and glyoxal. The background activities
437 involved in the manufacture of tannin are responsible for environmental contributions ranging from 58%
438 to 83%, depending on the category, except in ME (11%) and TET (15%). Regarding FE, the production
439 of glyoxal required for the bio-adhesive formulation plays a key role with a contribution of 39% (higher
440 than the one for the production of the condensed tannin, 27%) due to the emissions of glyoxal into
441 water from the background processes involved. Concerning TET, air emissions derived from the
442 manufacture of the bio-adhesive are responsible for 33% of the contributions to this impact category,
443 which are associated to the release of glyoxal (the effect of these emissions in FET category is around
444 1% of the total contributions). Accordingly, attention should be paid to the control of on-site emissions
445 produced in the factory. The action on the production of glyoxal is not proposed for assessment as it is
446 outside the performance of the plant.

447 Nevertheless, an optimization of the glyoxal dose could be interesting to reduce the burdens. As far as
448 the production of the raw material: tannin is concerned, it is one of the main contributors in numerous
449 environmental impact categories (see **Figure 5a**). Therefore, a detailed analysis of this process is
450 required with the aim not only of identifying the hotspots responsible but also of proposing further
451 improvements (if possible). The distribution of burdens between the processes involved in the
452 production of condensed tannin is shown in **Figure 5b**. Consequently, one process involved in the
453 production of the condensed tannin presents the major contributions to all impact categories and that is,
454 the production of the bark chips. It is important to bear in mind that bark chips have not been managed

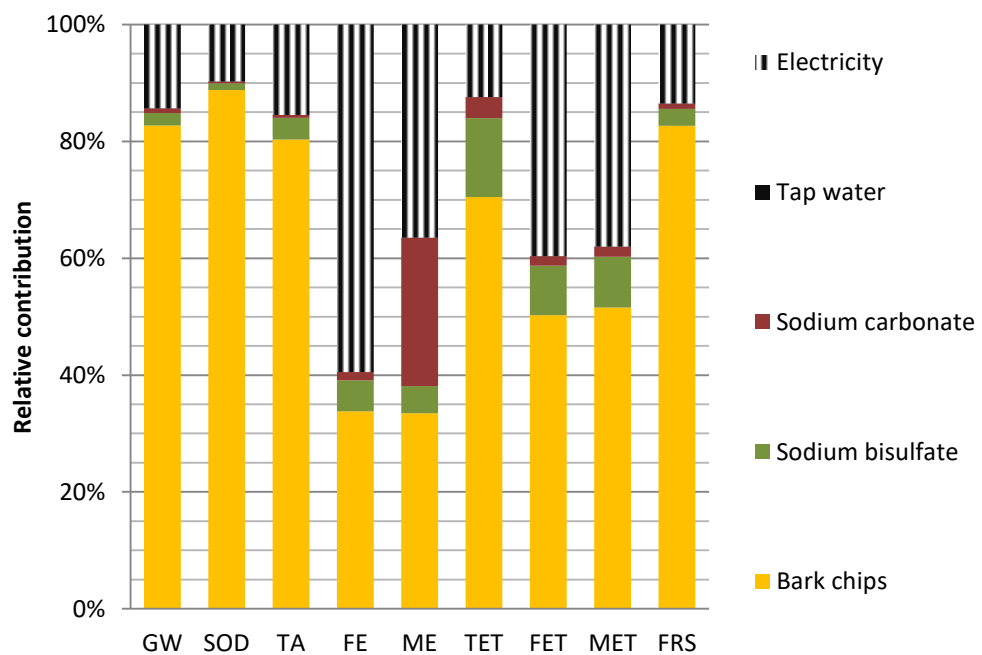
455 as a waste from sawmilling activities or from other wood processing facilities. The rationale behind this
456 approach is based on the fact that bark chips have multiple potential applications, such as energy,
457 biofoams or chemicals production (González-García et al., 2016; Feng et al., 2013). In this study, bark
458 chips from a representative sawmill have been taken into consideration and a volume-allocation
459 approach (14.4%) has been assumed to assign the impacts of sawmilling activities between bark and
460 other co-products (sawn timber and residual wood). According to **Figure 5b**, bark chips production is
461 the greatest contributor to GW (83%), SOD (89%), TA (80%), TET (71%), FET (50%), MET (52%) and
462 FRS (83%). In these categories, the forestry activities involved in the production of the required
463 roundwood are behind these high ratios, specifically due to the diesel requirements in the forest
464 machinery (mainly the harvester). Tannin production includes the mixing of the bark chips with hot water
465 and extractive chemicals in a vessel, the subsequent evaporation step to concentrate the extract and a
466 final spray-drying. As a result, there is a significant electricity demand and its production (which is taken
467 directly from the grid) greatly contributes to FE (59%), ME (36%), FET (40%) and MET (38%). One of
468 the extractive chemicals required to extract tannin from the bark, i.e., sodium carbonate, plays a key role
469 in terms of ME (25%). Sodium carbonate is co-produced in a factory together with ammonium chloride
470 using the Solvay process. Therefore, there is a consumption of liquid ammonia and as consequence,
471 there are indirect ammonium emissions into water derived from this industrial process, which are behind
472 the effect from sodium carbonate on ME.



473

474

Figure 5a



475

476

Figure 5b

477 **Figure 5.** Distribution of burdens between contributing inputs/outputs involved. a) Tannin-based bio-adhesive production; b)
 478 Condensated tannin production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial
 479 acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater
 480 ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

481 3.5. Sensitivity analysis and improvement options

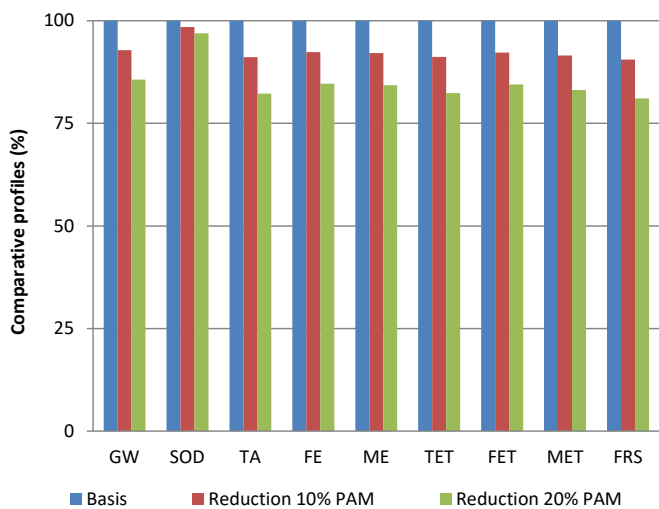
482 As detailed above, environmental profiles for the proposed bio-adhesives have been identified for
483 analysis that identifies the key processes, which are responsible for the highest contributions to the
484 environmental burdens. Thus, further research activities should be conducted in order to reduce their
485 environmental impacts by introducing improvement measures. To this end, a sensitivity analysis has
486 been carried out per scenario to determine how the profiles could be improved. Thus, attention has
487 been paid to some parameters, such as chemical doses, on-site emissions and electricity consumption.

488
489 *Soy-based bio-adhesive:* The sensitivity analysis has been focused on reducing the PAM dose in the
490 production of the adhesive due to the prominent effect that this chemical has on the global profile (see
491 **Figure 2a**). In this analysis, a reduction of 10% and 20% has been proposed, bearing in mind that when
492 the system is performed at commercial level, the chemical doses should be optimized in order to reduce
493 costs. Thus, improvements could be achieved in all impact categories, as depicted in **Figure 6a**. The
494 highest improvement ratios should be identified in toxicity related categories as well as in TA.

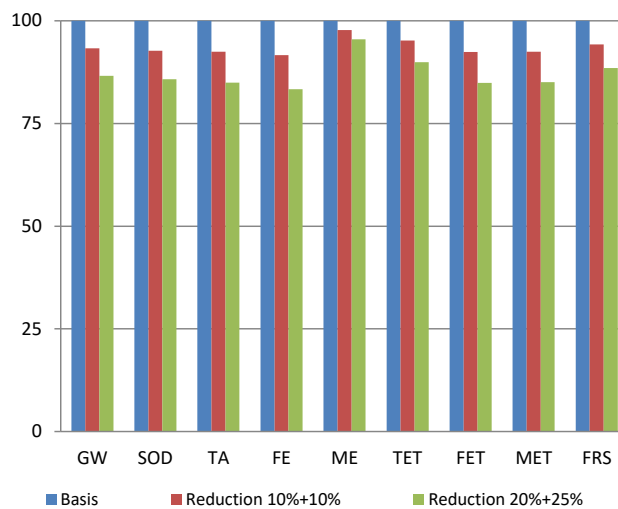
495
496 *Kraft lignin based bio-adhesive:* The environmental profile associated with the production of this bio-
497 adhesive is clearly dominated by the impacts from the lignin glyoxalation step as identified in **Figure 3a**,
498 mainly due to the high electricity requirements and uncontrolled glyoxal emissions into air (see **Figure**
499 **3b**).

500 1. Firstly, a sensitivity assessment has been conducted focused on the reduction of these
501 parameters in the glyoxalated lignin taking into account that they could be optimized under industrial
502 conditions. A reduction of 10% and 20% in electricity requirements combined with reductions of 10%
503 and 25%, respectively, in on-site emissions. **Figure 6b** depicts the comparative profile under the
504 introduction of both improvement considerations. Accordingly, a 10% reduction in both parameters has
505 associated slight improvements in the global profile, ranging from 2% to 8%. Nevertheless, a reduction

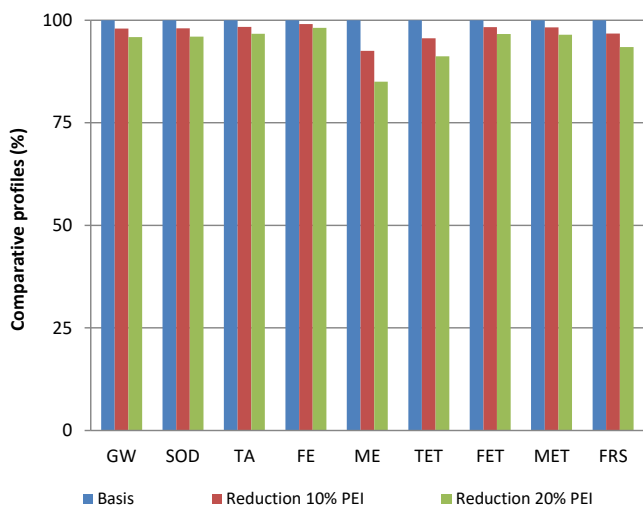
506 of both 20% in electricity demand and 25% in on-site emissions involves improvements of up to 17% in
 507 categories such as FE, TA, FET and MET.



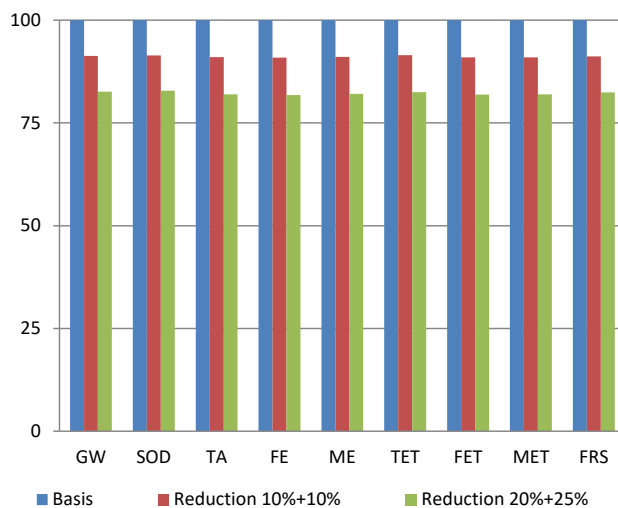
(a)



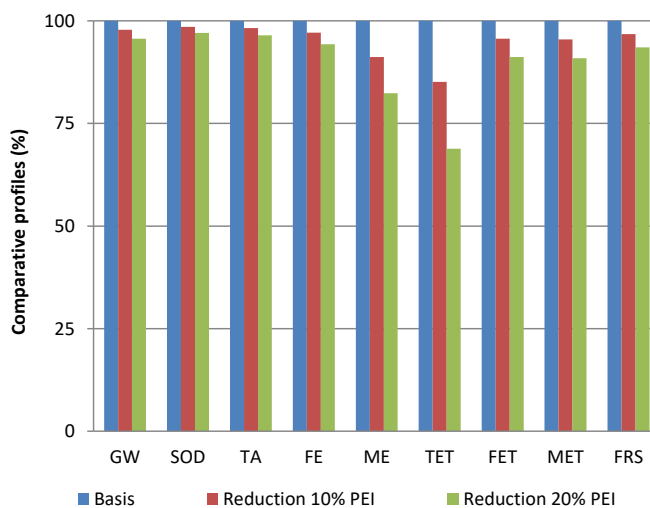
(b)



(c)



(d)



(e)

Figure 6

508 **Figure 6.** Sensitivity analysis over the global profiles of bio-adhesives production under study, conducted on a) PAM dose (-
 509 10% and -20%) in soy-based bio-adhesive; b) Electricity consumption (-10% and -20%) and on-site emissions (-10% and -
 510 25%) in glyoxalation step in Kraft lignin based bio-adhesive; c) PEI dose (-10% and -20%) in Kraft lignin based bio-adhesive;
 511 d) Electricity consumption (-10% and -20%) and on-site emissions (-10% and -25%) in glyoxalation step in Organosolv lignin
 512 based bio-adhesive; e) PEI dose (-10% and -20%) and on-site emissions (-10% and -25%) in tannin based bio-adhesive.
 513 Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater
 514 eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity
 515 (MET) and fossil resource scarcity (FRS).

516 2. Secondly, an optimization in the dose of PEI required in the formulation of the bio-adhesive
 517 could be really attractive from an environmental perspective, since this parameter plays a key role in the
 518 global profile as depicted in **Figure 3a**. Therefore, a sensitivity assessment has been conducted
 519 considering potential reductions of 10% and 20% on the PEI consumption. Improvements should be
 520 achieved as depicted in **Figure 6c** specifically in this category where PEI is considered as the hotspot
 521 that is ME. In this category, the burdens should be reduced around 7% and 15%, respectively for 10%
 522 and 20% of reductions.

523
 524 *Organosolv lignin based bio-adhesive:* The production of glyoxalated organosolv lignin is by far the main
 525 parameter responsible for environmental loads, as detailed in **Figure 4a**, mainly due to the production of
 526 the corresponding electricity requirements although uncontrolled glyoxal emission into air is also relevant
 527 in TET (see **Figure 4b**). Thus, an optimization of these electricity requirements (i.e., reductions of 10%
 528 and 20%) in the equipment involved in the production of glyoxalated lignin has been considered as key
 529 issue to improve the profile combined with a reduction of uncontrolled glyoxal emission (10% and 25%,
 530 respectively). Accordingly, improvements in all impact categories could be achieved, as depicted in
 531 **Figure 6d**. A reduction of 10% in both parameters should lead to improvements of 9% in all impacts.
 532 The scenario with the highest reduction ratios should result in improvements of 17-18% in all categories.

533

534 *Condensed tannin-based bio-adhesive*: in this scenario, a double sensitivity analysis has been
535 proposed based on improvements on on-site emissions and chemical dose.

536 1. Reduction of on-site emissions from tannin-based bio-adhesive. As detailed above, on-site
537 emissions considerably contributes to TET. These are uncontrolled emissions, which could be reduced
538 by means of the installation of an specific control system. Two reduction ratios have been analysed,
539 10% and 25%. Accordingly, improvements of 3% and 8% could be achieved in TET, respectively for the
540 reduction proposed of 10% and 25%. As expected, there is no effect on the remaining categories as
541 detailed in **Figure 6d**.

542 2. Reduction of glyoxal dose in the production of tannin based bio-adhesive. As detailed in **Figure**
543 **5a**, the consumption of glyoxal is an environmental hotspot because of the effect on the profile from the
544 background activities involved. An optimization of its consumption dose should be associated with an
545 environmental improvement in the profile of tannin-based bio-adhesives. Reductions on this parameter
546 have been combined with the proposal discussed above, as they should be directly related. It can be
547 seen that the environmental profile is considerably improved when actions are proposed on glyoxal
548 consumption. A reduction of 10% should lead to a reduction of burdens in all impact categories but
549 especially, in terms of FET, MET and FRS (around 4%). When the reduction ratio is increased to 20%,
550 the effect is more noticeable achieving improvement ratios of around 9% in the mentioned categories as
551 detailed in **Figure 6f**.

552

553 **3.6. Comparison with conventional adhesives**

554 Conventional adhesives or resins used for the production of wooden flooring are UF, PF and MUF.
555 These resins derive from fossil resources and have associated negative environmental problems,
556 mainly due to the formaldehyde emissions produced during their entire life cycle. Nevertheless, and
557 although the interest on developing technologies to produce renewable adhesives as fossil substitutes
558 is the order of the day, the production of bio-adhesives is still under development and the commercial

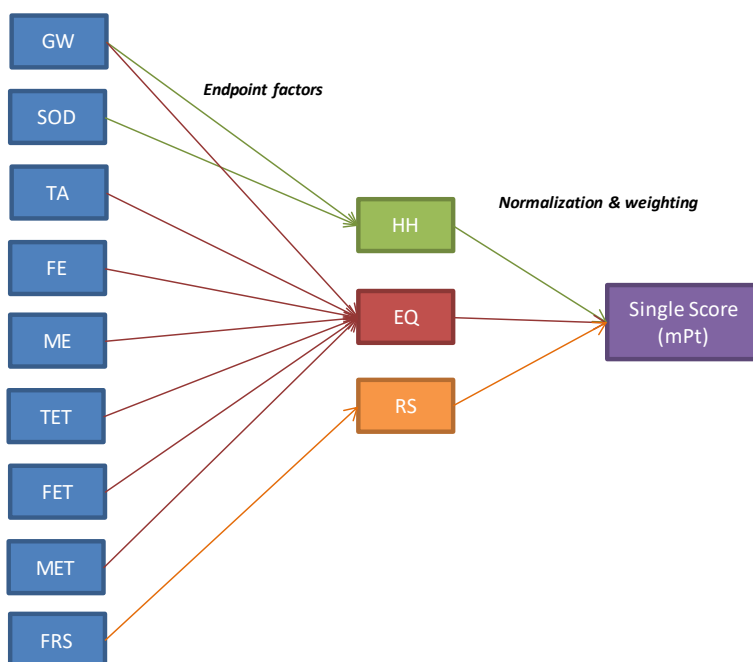
559 bio-adhesives have a comparatively higher cost than conventional ones (Hemmilä et al., 2017). In this
560 regard, an environmental comparison between conventional resins and bio-adhesives proposed for
561 analysis based on an end-point method has been proposed in order to have a single environmental
562 score per adhesive (in millipoints –mPt). For this purpose, the normalization and weighing factors taken
563 from ReCiPe 2016 Endpoint method have been considered. The analysis at endpoint level reports the
564 results in terms of three indicators to express the relative severity of damage according to the following:
565 Human Health (HH), Ecosystem Quality (EQ) and Resource Scarcity (RS). Those three endpoint
566 indicators are the result of an aggregation process from midpoint categories by means of specific
567 endpoint characterization factors. In this analysis, only those impact categories previously selected for
568 the environmental assessment at midpoint level (GW, SOD, TA, FE, ME, TET, FET, MET and FRS) will
569 be considered for the estimation of the single environmental score, as depicted in **Figure 7a**, taking into
570 account the corresponding normalization and weighing factors established by the method.

571 The result of the comparison is illustrated in **Figure 7b** and the size of the circles corresponds to the
572 magnitude of the global impact. In general, the impact trend revealed by the bio-adhesives does not
573 follow that of the fossil-ones, except in the case of soybean and tannin based bio-adhesives. The single
574 score corresponding to the soy-based adhesive (43 mPt) and tannin-based adhesive (46 mPt) are in
575 line with the one for UF resin (41 mPt) and around 25% lower and 30% higher than the scores for PF
576 resin (56 mPt) and MUF resin (33 mPt), respectively. Thus, soy based bio-adhesive can be considered
577 as an environmentally potential substitute to the fossil resins. On the contrary, the single scores
578 estimated for the other three bio-adhesives are considerably higher than those for fossil ones as
579 depicted in **Figure 7b** – Organosolv lignin-based (228 mPt) reports the worst profile followed by tannin-
580 based (122 mPt) and Kraft lignin-based (123 mPt) adhesives. Accordingly, further research on the
581 development of these three bio-adhesives is required in order to be environmentally competitive, taking
582 into account the assumptions established in our systems (see *Section 2.3*). Given the fact that bio-
583 adhesives have a significant contribution from renewable feedstocks, it could be expected a more

584 significant difference between fossil and renewable adhesives. However, it must be noticed that in no
585 case feedstocks have been considered as waste from other activities. Thus, it must be remarked the
586 effect over the overall profile of the demand of fossil resources in the background processes involved in
587 the production of the feedstocks. This is the case, for example, of diesel in agricultural and forestry
588 activities, as well as natural gas and electricity in pulp manufacturing activities. If a detailed analysis of
589 the scores and the distribution between the level of damage (HH, EQ and RS) is carried out, the scores,
590 regardless of the adhesive, are mainly affected by the damage to human health (on average, 78%). In
591 all adhesives, the use of fossil resources in their background systems is behind the contributions to HH,
592 mainly due to GHG emissions (99% of total contributions). This effect is considerable outstanding in the
593 Organosolv lignin based bio-adhesive (6 and 7 times higher than soy and MUF-based adhesives,
594 respectively). The rationale behind that results is the effect over the profile from the high electricity
595 requirements (and thus, corresponding production effect) to perform the glyoxalation of the lignin, a
596 process required for the functionalization of lignin .

597 Damage to EQ ranks second in all scores, as shown in **Figure 7b** although with ratios ranging from 1%
598 (PF resin and Soy based bio-adhesive) to 17% (Organosolv lignin based bio-adhesive). The contribution
599 from RS to the global environmental single score is really low, with ratios ranging from 3% (Organosolv
600 lignin based bio-adhesive) to 10% (fossil adhesives). Regarding the former, damage to ecosystem
601 quality is mainly associated with GHG emissions from background processes in all scenarios (70% on
602 average of total contributions from impact categories). Moreover, damage to EQ is higher for bio-
603 adhesives than for fossil ones (except for Soy based and tannin based bio-adhesives, which are in line
604 with those for fossil resins), which is linked to the use of renewable feedstocks and corresponding
605 ecosystem degradation. Given the variety of feedstocks that have been considered to produce the bio-
606 adhesives, it can be concluded from this study that soy-based bio-adhesive can be selected as the best
607 fossil-adhesive substitute, closely followed by the tannin-based one, since it has a global impact about
608 25% lower than PF and practically equal to UF.

609 A direct comparison of the results of our study with those of others already reported has been quite
610 complicated (McDevitt and Grigsby, 2014; Yang and Rosentrater, 2020), mainly due to the constraints of
611 using different databases and environmental assessment methods. McDevitt and Grigsby (2014)
612 considered a protein-lignin adhesive formulated with corn protein, soy protein, lignin and tannin. When
613 comparing our results with those of McDevitt and Grigsby (2014), both studies highlight the higher
614 impact on ecosystem damage associated to bio-adhesives over fossil ones, mainly due to the effect of
615 the renewable feedstock. Yang and Rosentrater (2020) analysed the production of bio-glycerol based
616 adhesive (from soybean) and fossil glycerol based adhesive but considering alternative assessment
617 methods. Our study agrees with some of the findings reported in that study in which soy-based
618 adhesive had a lower impact than fossil one. Moreover, the impact of resource scarcity damage is
619 higher in fossil adhesive than in the bio one (around 50%), while in our study the PF and UF reported an
620 impact on resource scarcity damage 59% and 10% higher than the soy-based adhesive. The same
621 trend is observed in human health damage, indicating that soy-based adhesive is less harmful to
622 humans. Nevertheless, this trend is not observed in damage to EQ since Yang and Rosentrater (2020)
623 also identified major impact on fossil adhesives, opposed to our study and McDevitt and Grigsby (2014).
624 As detailed above, the bio-adhesives analysed are based on raw materials that are not considered to be
625 waste from other industrial activities and therefore the background activities involved in their production
626 are included within the system boundaries as well as derived burdens are computed. In the case of
627 tannin-based adhesive, bark chips from sawmilling activities have been managed as raw material.
628 Nevertheless, bark could be alternatively taken from wood-based industrial processes, where it is co-
629 produced as a residue. This could be the case for bark from forestry activities or even from panel
630 factories, where the accumulation of tons of bark-based waste can be identified. As for Kraft lignin, it is
631 derived from the black liquor obtained in the pulp stage that is commonly used for energy purposes in
632 kraft pulp mills, combined with other wood residues.



633

Figure 7a



634
635
636

Figure 7b

637 **Figure 7.** a) Schematic procedure for estimating single environmental scores; b) Comparison of Life cycle impacts and
638 damage contributions on human health, ecosystem and resources.

639 However, it has no direct economic value on the markets. Therefore, a sensitivity analysis has been
640 carried out to determine how the environmental profile of tannin- and Kraft lignin-based bio-adhesives
641 could change if the corresponding feedstocks (bark chips and black liquor) were assumed as residues
642 from industrial activities. Thus, burdens derived from the production of both bark chips and black liquor
643 have been assumed as zero, allocating them to the corresponding main products of their production
644 systems (e.g., boards and pulp). With this consideration, improvements of the environmental scores
645 could be identified specifically concerning tannin-based adhesive. Accordingly, **Figure 7b** includes the
646 profiles corresponding to Kraft lignin (residue) and Tannin (residue) adhesives and reductions on the
647 scores of 1% and 53% (123 mPt and 30 mPt, respectively) should be identified in comparison with the
648 corresponding adhesives where the feedstock is not managed as waste. In this regard, it must be
649 noticed the low effect over the global profile from lignin production itself. Regarding the use of residual
650 bark as raw material brings benefits to the point of making the adhesive competitive with the fossil ones
651 as depicted in **Figure 7b**, resulting in an environmental single score even better in comparison with that
652 of soy based bio-adhesive.

653

654 **4. Conclusions**

655 Given the variety of feedstocks that can be used as such to produce substitutes for alternative fossil
656 resins, it can be concluded that there is a wide spectrum of possibilities for producing more
657 environmental-friendly adhesives, which will reduce dependence on crude oil. However, there are only a
658 few industrial bio-adhesive plants that supply products at high costs. According to this study, it is
659 confirmed that LCA can be a useful tool to perform a gross evaluation and comparison between
660 adhesives in order to identify the advantages and disadvantages, as well as with the aim of identifying
661 parameters on which to act in the short and medium term. Nevertheless, attention needs to be paid on
662 the current constraints that considerably affect the outcomes and thus, decision making related with the
663 early development in which are the bio-adhesives production systems. This situation makes it difficult to

664 compare with the conventional fossil resins, which are totally optimized and developed, supplying
665 products at low price but involving controversial impacts. Our results show that soy-based bio-adhesive
666 can be considered as a potential fossil substitute.

667 In this sense, our results can help policy makers and researchers to move towards these parameters and
668 measures that require more attention (e.g., electricity and chemicals consumption, as well as uncrotrolled
669 emissions) from the environmental point of view. In addition, more research is needed not only on improving
670 adhesive-related characteristics (e.g. water resistance, reactivity) but also on developing renewable cross-linkers
671 required to achieve the required properties of adhesives, as synthetic ones are still used. The results of this
672 analysis also recommend re-evaluating lignin-based processes as alternatives in the production of
673 bioadhesives. In this regard, production alternatives involving significant environmental improvements
674 will be further investigated in the framework of formaldehyde-free alternatives.

675

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