Science of the Total Environment 738 (2020) 140357

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Cradle-to-gate Life Cycle Assessment of bio-adhesives for the wood panel industry. A comparison with petrochemical alternatives

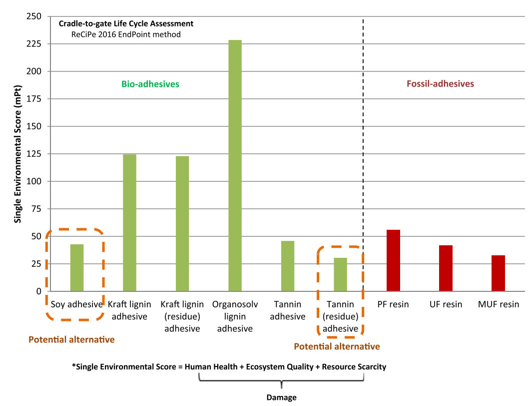
Ana Arias [1], Sara González-García *,[1], Sandra González-Rodríguez, Gumersindo Feijoo, María Teresa Moreira

CRETUS Institute, Department of Chemical Engineering, School of Engineering, Universidade de Santiago de Compostela, Spain

HIGHLIGHTS

- Four different bio-adhesives have been modelled and compared from an environmental approach
- Soy and tannin based bio-adhesive have an overall better profile than fossil resins
- Lignin-based adhesives are less environmentally friendly
- The glyoxalation step in lignin-based adhesives ranks first in terms of environmental burdens
- Optimization of production systems is required to be technically and environmentally competitive

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 5 May 2020
Received in revised form 16 June 2020
Accepted 17 June 2020
Available online 18 June 2020

Editor: Damia Barcelo

Keywords:
Bio-based adhesives
Environmental impacts
Life cycle assessment
Petrol-based adhesives
Sustainability

ABSTRACT

The wood panel industry requires the introduction of more environmental-friendly adhesives due to the strict current regulations on formaldehyde-based emissions. The purpose of this study was to environmentally analyse the production of four different bio-adhesives as alternatives to the most conventional fossil resins used in the production of wood panels. The bio-adhesives proposed for analysis derived from different available renewable biopolymers such as protein (soy) and lignin (Kraft and Organosolv), as well as tannin.

The production systems were evaluated from a cradle-to-gate perspective using the Life Cycle Assessment methodology, with the aim of identifying critical parameters and comparing them with fossil substitutes. Inventory data of bio-adhesives were modelled at large scale from lab scale experiments and completed with literature reports. Our results showed that the soy-based and tannin based bio-adhesive had an overall better profile than fossil resins, identifying the production of polyacrylamide for the former, and the production of condensed tannin and glyoxal for the latter, as the main environmental hotspots. In contrast, further research is required on the use of lignins, specifically because of the electricity requirements in the lignin glyoxalation stage (a process required for the functionalization of lignin). Sensitivity analyses were conduced on these key parameters suggesting that there is room for improvement.This study provides useful information for researchers and policy-makers on where to focus their activities with the aim of making the future of bio-adhesives more technically and environmentally favourable.

* Corresponding author.
E-mail address: sara.gonzalez@usc.es (S. González-García).
[1]Both authors equally contributed to the work

https://doi.org/10.1016/j.scitotenv.2020.140357

1. Introduction

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

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

Traditional adhesives (known as petroleum-based adhesives) are derived from the co-products of petroleum processing and formaldehyde-based adhesives are currently used for the production of wooden flooring, mainly urea-formaldehyde (UF), phenol-formaldehyde (PF) and melamine-urea formaldehyde (MUF). The use of these adhesives (also known as resins) is supported by several features such as low curing temperatures, excellent adhesion properties, water resistance and low price (Kim, 2009; Yang and Rosentrater, 2019a, 2019b). On the contrary, the main drawback of this type of adhesives is the possibility of releasing volatile organic compounds (VOCs) and formaldehyde vapors that harm human health, causing nasal carcinomas as well as eyes and throat irritation, among other adverse effects (Kim, 2009; Li et al., 2009). As a result, and due to the growing demand for adhesives, there is a worldwide effort to develop more environmentally and health –friendly adhesives. In this regard, studies are focused on alternatives to formaldehyde-based adhesives, by partially or completely eliminating formaldehyde from the composition (Pizzi, 2006; McDevitt and Grigsby, 2014; Stefani et al., 2008). Bio-based resins are natural polymers that act as adhesives. In this sense, biomass sources such as soy and corn proteins, tannins, starches, bio-glycerol and Kraft/Organosolv lignin are examples of feedstocks that have been assessed as potential for the production of these chemicals (Van Langenberg et al., 2010; Moubarik et al., 2009; McDevitt and Grigsby, 2014). Global demand for bio-adhesives is emerging because of their potential environmental and health benefits (Mathias et al., 2016) although the technologies are in their infancy.

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

Soy protein is one of the first feedstocks in the commercial production of bio-adhesives used in the wood panel sector as an alternative to PF and UF (Hemmilä et al., 2017). Soy protein is obtained by mechanical or solvent extraction for soybean oil (Li et al., 2004). Soy protein-based adhesive has several advantages, such as low cost, ease of handling, low pressing temperatures, in addition to being used for wood with high moisture content. Nevertheless, this type of bio-adhesive reports poor water resistance and is quite sensitive to biological degradation, among other drawbacks. On the contrary, this feedstock has been considered in recent years as a potential raw material due to its availability and interest worldwide (Li et al., 2004).Lignin-based adhesives are of potential interest as promising strategies for integrating bio-refineries into the wood sector, especially in pulp mills. Lignin is a side-stream from pulp manufacturing processes, which has been commonly used for energy purposes while recovering pulping chemicals (Geng and Li, 2006). Nevertheless, lignin streams are very different from each other in terms of their composition (i.e., sulphur-containing lignin -Kraft and sulphur-free lignin -Organosolv), which opens up a wide range of possibilities. Due to its composition, lignin is structurally similar to phenol (both contain phenolic hydroxyl groups), which makes it possible for lignin to be used as a substitute for phenol in the synthesis of PF (Sellers, 2001; Jin et al., 2010). Nevertheless, further research is required due to its lower reactivity to formaldehyde, which is a major disadvantage in applications where fast curing times are required (Hemmilä et al., 2017).Thus, lignin streams require some chemical modifications to increase their reactivity for the synthesis of bio-resins.

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

Life Cycle Assessment (LCA) is an environmental methodology that can be used to estimate the environmental burdens associated with a product, process or activity throughout its life cycle. This tool has been considered to demonstrate the environmental benefits associated with green products and novel technologies compared to petrol-based ones (Cherubini and Strømman, 2011; Rajagopalan et al., 2012; Collingea et al., 2015). In the context of adhesives and wooden floorings, several studies are also available in the literature that assess the environmental benefits of introducing green processes (González-García et al., 2011a; McDevitt and Grigsby, 2014; Yang and Rosentrater, 2019a, 2019b, 2020).

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

2. Methodology

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

2.1. Goal and scope definition

The goal of this LCA study is to provide an overview of the large-scale production of four different bio-adhesives, as well as to determine the yields and environmental improvements compared to petroleum-based adhesives. To this end, bio-adhesives production systems have been modelled at full-scale at 40 t/d based on laboratory-scale data (Luo et al., 2015; Navarrete et al., 2010; Geng and Li, 2006; Dongre et al., 2015; Li et al., 2004). The selection of this capacity is based on the fact that commercial formaldehyde-based resin plants have a capacity close to 15,000 t/year (Yang and Rosentrater, 2019a). An attributional cradle-to-factory gate approach has been considered, with soybean meal, Kraft lignin, Organosolv lignin and pine tannin as raw materials. Accordingly, the estimated environmental profiles will be those derived from the processes and the mass and energy flows involved throughout their life cycles. Therefore, energy and mass balances have been performed for the modelling of full-scale production systems in order to gather all the required data for the Life Cycle Inventory stage. Plant-wide modelling was conducted using Aspen Plus® software not only to identify the inventory data, but also to design the equipment involved in each system according to the required conditions.

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

2.2. Description of bio-adhesives production systems and inventory analysis

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

2.2.1. Soy-based bio-adhesive

The production system considered for the analysis is based on the use of a cross-linker in order to improve water resistance. Among the cross-linkers, 5,5-Dimethyl hydantoin polyepoxide (DMHP) reports multiples useful properties (Luo et al., 2015) and has therefore been taken into consideration. The overall foreground production system is divided into two main stages: DMHP production and Bio-adhesive production as detailed in Fig. 1a. Accordingly, the production chain starts with the soybean cultivation and processing in order to obtain the soybean meal. In this case, the production of the cross-linker takes place in the plant itself following the procedure detailed by Luo et al. (2015). Epichlorohydrin (ECH), sodium hydroxide solution (NaOH, 50% wt) and 5,5-Dimethyl hydantoin (DMH) are mixed at 80 °C for 1 h at a molar ratio of 1:8:1 (DMH/ECH/NaOH). The bio-adhesive production process is carried out at 20 °C for 10 min in a stirred that incorporates soy protein, DMHP, polyacrylamide (PAM) and sodium dodecyl sulfate

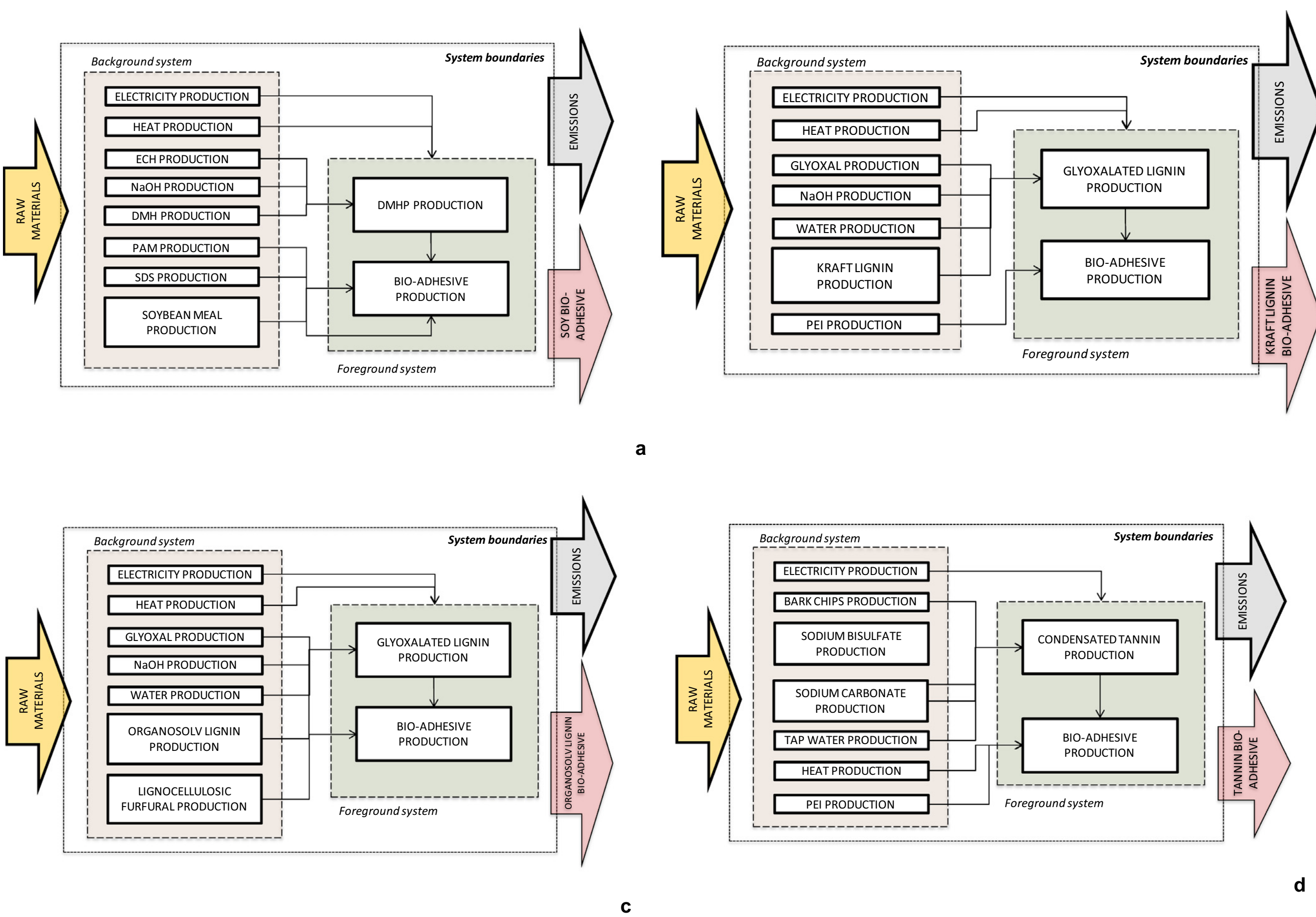


Fig. 1. System boundaries of the bio-adhesives production on a real scale. a) Soy-based bio-adhesive; b) Kraft lignin- based bio-adhesive; c) Organosolv lignin-based bio-adhesive; d) maritime pine tannin-based bio-adhesive.

(SDS). The management of residual streams produced in both stages in which the production process is classified have been computed within the system boundaries, that are wastewater treatment, landfilling and incineration. The data corresponding to the foreground system have been determined by means of plant-wide modelling in Aspen Plus® and a summary of the most relevant mass and energy flows is detailed in Table 1.

2.2.2. Kraft lignin based bio-adhesive

Among lignins, Kraft lignin is the most abundant in pulp mills. This type of lignin is insoluble in water and most solvents, except in highly alkaline environments (Hemmilä et al., 2017). Research is being conducted with the aim of totally replacing PF in board manufacture with a combination with glyoxal (El-Mansouri et al., 2007; Navarrete et al., 2010). Thus and bearing in mind that lignin streams require a chemical modification, the use of glyoxalated lignin should allow the production of boards with good internal bond strength characteristics (Hemmilä et al., 2017).Therefore, this strategy has been taken into consideration in the modelling of the Kraft lignin based bio-adhesive. The general production system in the foreground is divided into two main steps which are the glyoxalation of Kraft lignin and the production of bio-adhesives as detailed in Fig. 1b. Accordingly, the production chain starts with the production of Kraft hardwood lignin at the pulp mill, where the bio-adhesive-based biorefinery strategy should be incorporated. It is considered that lignin is precipitated from the black liquor of the pulp mill under acidic conditions (mainly, with hydrochloric acid - HCl), washed and dried. An additional alkaline treatment is then required considering an aqueous solution of NaOH at 180 °C. The lignin suspension after pulping is filtered and the liquid phase obtained is subjected to precipitation by acidification with HCl. The procedure reported by Radoykova et al. (2013) has been considered. The lignin powder is then glyoxalated in a reaction with water, sodium hydroxide (30%) and glyoxal (40%) solutions in a stirrer for 8 h at 58 °C (Navarrete et al., 2010). The cost of this step could be reduced by understanding how to optimize the amounts of glyoxal and NaOH. Glyoxal lignin is further mixed with polyethylenimine (PEI) in a stirrer for 40 min at 20 °C and a lignin/PEI weight ratio of 5:1 (Geng and Li, 2006). Waste streams produced in both stages and their management in wastewater treatement plants and incineration facilities have been computed within the system boundaries. Table 2 summarizes data corresponding to the foreground system at full-scale.

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

Inputs from technophere		Outputs to technosphere	
DMHP production			
Materials		Products	
NaOH solution 50%wt	3.10 g	DMHP to Bio-adhesive production	0.141 kg
ECH	18.06 g	Waste to treatment	
DMH	0.988 g	Wastewater	1.69 mL
Energy		Landfill	0.31 g
Electricity	4.23 Wh		
Heat	1.86 kJ		
Bio-adhesive production			
Materials		Products	
Soybean meal	0.268 kg	Bio-adhesive	1.00 kg
DMHP from DMHP production	0.141 kg	Waste to treatment	
PAM	0.690 kg	Incineration	97.11 g
SDS	12.90 g	Landfill	14.11 g
Energy			
Electricity	30.30 Wh		
Heat	12.04 kJ		

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

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

Inputs from technophere		Outputs to technosphere	
Glyoxalated lignin production			
Materials		Products	
NaOH	40.90 g	Glyoxalated lignin to Bio-adhesive production	0.741 kg
Water	0.469 kg	Waste to treatment	
Glyoxal	33.90 g	Wastewater	48.9 mL
Kraft lignin	0.279 kg	Incineration	27.90 g
Energy		Emissions into air	
Electricity	12.52 kWh	Glyoxal	3.40 g
Heat	102.9 kJ		
Bio-adhesive production			
Materials		Products	
Glyoxalated lignin from Bio-adhesive production	0.741 kg	Bio-adhesive	1.00 kg
PEI	0.370 kg	Waste to treatment	
Energy		Incineration	0.11 kg
Electricity	740 Wh		
Heat	102.9 kJ		

NaOH - Sodium Hydroxide; PEI - Polyethylenimine.

2.2.3. Organosolv lignin based bio-adhesive

As detailed above, there are several types of lignins depending on the pulping process. Organosolv lignin is essentially water-insoluble and is obtained in non-sulphur biorefineries, closely related to the cellulosic biofuel industry (Hemmilä et al., 2017). Following the structure and design proposed for the analysis for the Kraft-lignin based bio-adhesive, the production scenario has been classified into two steps that are the production of glyoxalated lignin and the production of bio-adhesive. Navarrete et al. (2010) and El-Mansouri et al. (2007) developed the glyoxalation of organosolv lignin to obtain synthetic resin-free adhesives. The glyoxalation process described by Navarrete et al. (2010) has been followed in detail according to the Kraft lignin based case study. The system under study starts with the production of Organosolv softwood lignin in a biorefinery, following the approach reported by Kautto et al. (2013). Secondy, lignin is mixed with water, NaOH (30%) and glyoxal (40%) to be glyoxalated. Glyoxalated lignin is

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

Inputs from technophere		**Outputs to technosphere**	
Glyoxalated lignin production			
Materials		Products	
NaOH	58.30 g	Glyoxalated lignin to Bio-adhesive production	1.06 kg
Water	0.668 kg	Waste to treatment	
Glyoxal	48.30 g	Wastewater	48.9 mL
Organosolv lignin	0.398 kg	Incineration	39.8 g
Energy		Emissions into air	
Electricity	30.35 kWh	Glyoxal	4.80 g
Heat	146.6 kJ		
Bio-adhesive production			
Materials		Products	
Glyoxalated lignin from Glyoxalated lignin production	1.06 kg	Bio-adhesive	1.00 kg
Furfural	56.00 g	Waste to treatment	
Energy		Incineration	0.112 kg
Electricity	2.31 kWh		
Heat	10.47 kJ		

NaOH - Sodium Hydroxide.

finally mixed with furfural (as a substitute for formaldehyde) under acidic conditions and at 90 °C for 1 h. Furfural has been identified by the US Department of Energy as one of the top value-added products (Luo et al., 2019). The production of lignocellulosic furfural derived from biomass under a biorefinery approach has been taken into account following the production procedure reported by Bello et al. (2018). Fig. 1c depicts the system boundary corresponding to this scenario and Table 3 summarizes the inventory data of the foreground system.

2.2.4. Pine tannin based bio-adhesive

In this scenario, the production system comprises the processes of condensated tannin production and tannin-based bio-adhesive production as detailed in Fig. 1d. Condensed tannin (flavonoid polymer) commercially produced from maritime pine bark has been considered as the main raw material and information regarding its production has been taken from González-García et al. (2016). Concerning the bio-adhesive, the production process has been modelled following the lab experiment developed by Zhang et al. (2019). The background processes involved in the production of the condensed tannin, including the forest activities and the extraction stage, have been computed, as well as the management of derived waste produced in the bio-adhesive plant. Fig. 1d depicts the system boundary and Table 4 summarizes the most representative inventory data for the foreground system.

Once the scenarios have been modelled, several assumptions have been considered to complete the inventory data. As detailed above, information from different experiments performed at lab scale (procedures, ecuations, equipment) has been considered to design full-scale bio-adhesives production systems. Thus, the stoichiometric amounts of each chemical considering the lab protocols have been computed in the inventory data assuming a 90% production yield in all experiments. Whenever possible, primary data were considered in the inventory data collection. However, it has been necessary to handle secondary data specifically for the background system (see Fig. 1). The background system involves the activities required to produce the utilities (electricity and heat) and all the inputs (i.e. chemicals, tap water and biomass sources) required in the foreground system. Thus, the Ecoinvent® database version 3.5 (Wernet et al., 2016) has been considered as the main secondary data source. This database has been also considered to inventory the processes involved in solid waste and wastewater treatment linked to each scenario.

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

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

As regards the cleaning activities involved in each production system, they have been excluded from the system boundaries. The rationale behind their exclusion is twofold: a) the lack of information on this step and b) the final goal of our study is to compare different bio-adhesive production scenarios and the effect on their profiles of the cleaning stage could be considered the same. As for the infrastructure process, it has not been included within the system boundaries considering that the impacts from construction, decommissioning and equipment maintenance is negligible over the environmental profiles derived from each biorefinery plants under study according to Bello et al. (2018), González-García et al. (2011b) and Uihlein and Schebek (2009). Finally, the production of PF, UF and MUF has been taken into

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

Inputs from technophere		Outputs to technosphere	
Condensed-tannin production			
Materials		Products	
Bark chips	0.024 m^3	Condensed tannin to Condensed-tannin production	0.788 kg
Sodium bisulfate	0.180 kg		
Sodium carbonate	45.03 g		
Tap water	3.95 g		
Energy			
Electricity	1.97 kWh		
Bio-adhesive production			
Materials		Products	
Condensed tannin from Condensed-tannin production	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
Energy			
Electricity	0.001 kWh		
Heat	463.05 kJ		

PEI - Polyethylenimine.

Table 5
Description of the main Ecoinvent ® database version 3.2 processes considered in this study for the 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

account in order to compare the profiles between bio-adhesives and petrochemical ones, the inventory data for the background processes of the petrochemical adhesives have taken from the Ecoinvent® database version 3.5 (PF and UF) and Silva et al. (2015) (MUF).

2.3. Environmental assessment methods

Two impact assessment methods will be considered in the analysis. Firstly, the ReCiPe 2016 hierarchist Midpoint method V1.03 World (2010) (Huijbregts et al., 2017) has been used for the selection of characterization factors required to estimate the environmental burdens and a set of impact categories at midpoint level has been considered to report the environmental profiles. Regarding the set of impact categories, the following impacts have been considered: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS). Secondly, the ReCiPe 2016 hierarchist Endpoint method V1.03 World (2010) H/H (Huijbregts et al., 2017) has been considered to compare the profiles of bio-adhesives with those of fossil-adhesives. The choice of this endpoint approach is based on the fact of having only one single score for the overall impacts that can help disseminate the message to stakeholders. The SimaPro software v9.0 (PRé Consultants, 2020) has been used for the computational implementation of the life cycle inventories.

3. Results and discussion

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

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

3.1. Environmental performance of Soy based bio-adhesive

The characterization results corresponding to soy-based adhesive are shown in Fig. 2a. Taking into account this profile, the PAM production required in the formulation can be appointed as the most burdensome input with shares ranging from 72% to 95%, except for SOD, where the production of soybean meal is the environmental hotspot

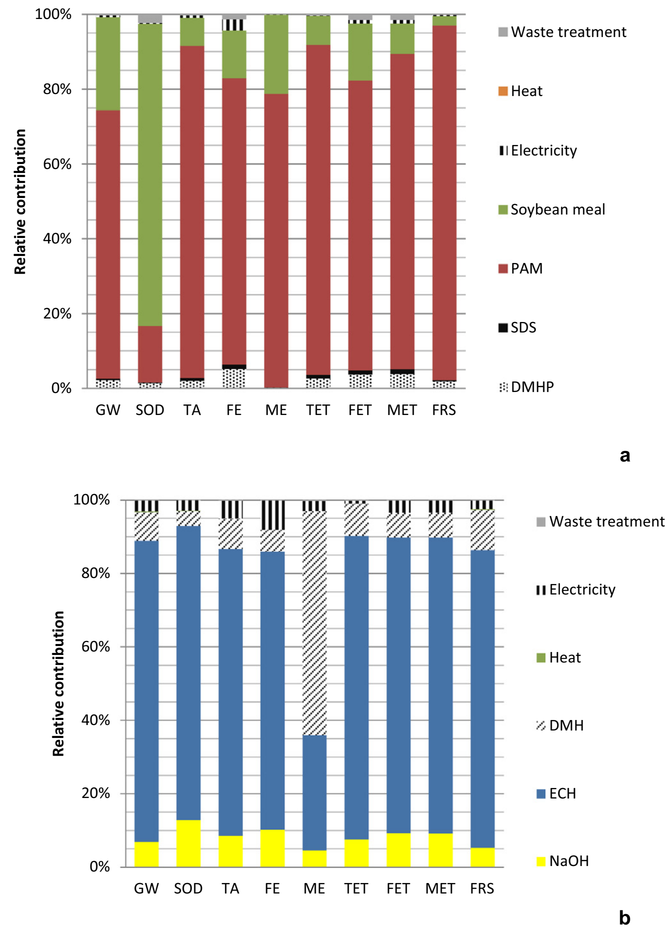


Fig. 2. Distribution of burdens between contributing inputs/outputs involved. a) Soy-based bio-adhesive production; b) DMHP production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial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

(81% of the total). The contributions from PAM is really outstanding and therefore, further research should be undertaken to propose an alternative chemical in the bio-adhesive formulation (if possible) or to optimize the system, as a reduction of the PAM dosis should have a direct effect on the global profile.

Soybean meal production involves agricultural activities in the field to produce the soybeans and subsequent processing, in which beans are cleaned to remove foreign material and pre-treated with heat to remove the hulls from the surface (i.e. to increase the protein content) and then, dried and flaked. Finally, oil is extracted from the soy flakes with a solvent to produce the soybean meal (Soymeal.org 2020). The production of the raw material also reports outstanding contributions (in addition to SOD) to GW (25%), FE (13%), ME (21%) and FET (15%). The rationale

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

		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

behind these burdens is associated with the requirement of fertilizers in agricultural activities and the corresponding on-field emissions (effect on FE and ME), as well as with the energy requirements in the processing plant (effect on GW, SOD and FET).

The contributions of the other processes involved (SDS, electricity, heat and waste treatment) are always below 2%. The effect on the overall production profile of the cross-linker required to improve the water resistance of the adhesive, i.e. DMHP, is also not remarkable, with contributions lower than 5%. Nevertheless, and given that it is an activity that takes place within the factory, as detailed in Fig. 1a, it has been analysed in detail in order to identify the responsible inputs to its derived environmental burdens. Fig. 2b details the distribution of burdens per impact category among the contributing factors. Having in mind this figure, the impacts can be mainly assigned to one specific input that is, ECH. The effect of the production of ECH requirements ranges around 80% in all categories, except in terms of ME (31%), where the burdens are dominated by the production of DMH (61%) due to ammonium and nitrate emissions into water. Once again, the production of energy requirements (heat and electricity) as well as management of waste do not report an outstanding effect on the global profile.

3.2. Environmental performance of Kraft lignin based bio-adhesive

Lignin is one of the most abundant renewable natural polymers, with Kraft lignin being the most abundant industrial lignin (Geng and Li, 2006). Altough it is usually burned for energy purposes, it has multiple applications, with the bio-adhesive production being one of the most promising options. In this system, the production of Kraft lignin based bio-adhesive has been environmentally evaluated and Fig. 3a depicts the distribution of environmental burdens per contributing parameters. Having in mind this figure, the production of the glyoxalated lignin is the largest responsible for environmental impacts, regardless of the category assessed, which implies contributing ratios ranging from 54% to 86% (except in ME, where the ratio decreases to 24%). This material involves the glyoxalation step of Kraft lignin within the factory and therefore requires specific analysis. The lignin glyoxalation is required as a pre-treatment step so that it can be chemically modified to enhance its further reactivity in the synthesis of bio-based resins (Ang et al. 2015). In this case study, glyoxal – a non-toxic and non-volatile dialdehyde – in combination with NaOH has been considered under the conditions outlined by Navarrete et al. (2010). This step demands large amount of electricity, since the required stirring is 8 h. As a consequence, the production of this electricity requirement, which is taken directly from the grid, is by far the main parameter responsible for the environmental burdens derived from lignin glyoxalation, as shown in Fig. 3b.

The effect of this parameter on the glyoxalated lignin profile is remarkable in all the categories with ratios above 72%. Nevertheless, the effect of the chemicals, heat to maintain temperature (around 58 °C) and waste treatment is negligible, as depicted in Fig. 3b. Conversely, attention must be also paid to on-site emissions from glyoxalation. These are uncontrolled glyoxal emissions into air which considerably affect TET with a contributing ratio of 18%. Finally, Kraft lignin used as raw material presents a considerable low uniform distribution in the overall profile (contributing ratios up to 10%).

To conclude, the effect on the profile derived from Kraft lignin based bio-adhesive from heat and electricity requirements as well as from waste treatment is negligible (see Fig. 3a). On the contrary, the production of PEI required in the formulation of the bio-adhesive plays a key role with ratios close to 20% in several categories except in ME (due to ammonium and nitrate emissions inwater), TET (due to copper emissions into air) and FRS (crude oil and natural gas demand) where they amount to 75%, 44% and 33%, respectively, due to the background processes involved in the manufacture. PEI is a synthetic organic chemical derived from aziridine (derived from petroleum). However, as research activities focused on the development of platform chemicals from

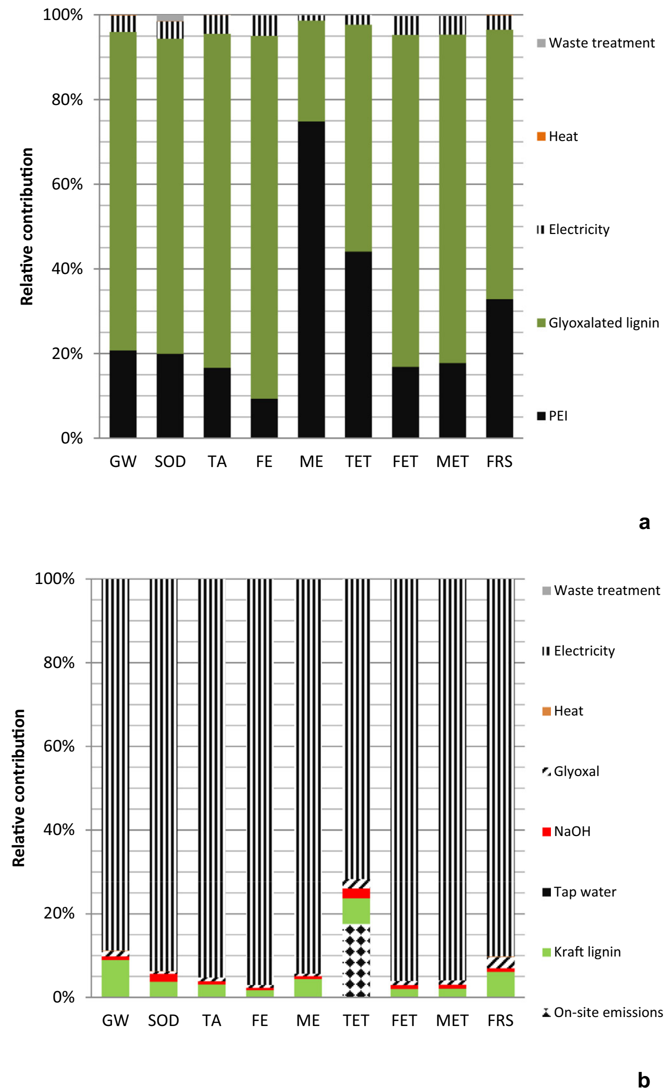


Fig. 3. Distribution of burdens between contributing inputs/outputs involved. a) Kraft lignin-based bio-adhesive production; b) Glyoxalated lignin production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

renewable sources, it could be expected a future synthesis of aziridine from non-petroleum feedstocks (Williams and Hillmyer, 2008). Thus, further improvements in reducing uncontrolled on-site emissions should be made to improve the profile in terms of ME by optimizing the dose of PEI required and the electricity consumption in the lignin glyoxalation step. These issues will be discussed in detail below.

3.3. Environmental performance of Organosolv lignin based bio-adhesive

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

Fig. 4a depicts the distribution of burdens per impact category for the Organosolv lignin bio-adhesive. The production stage focusing on

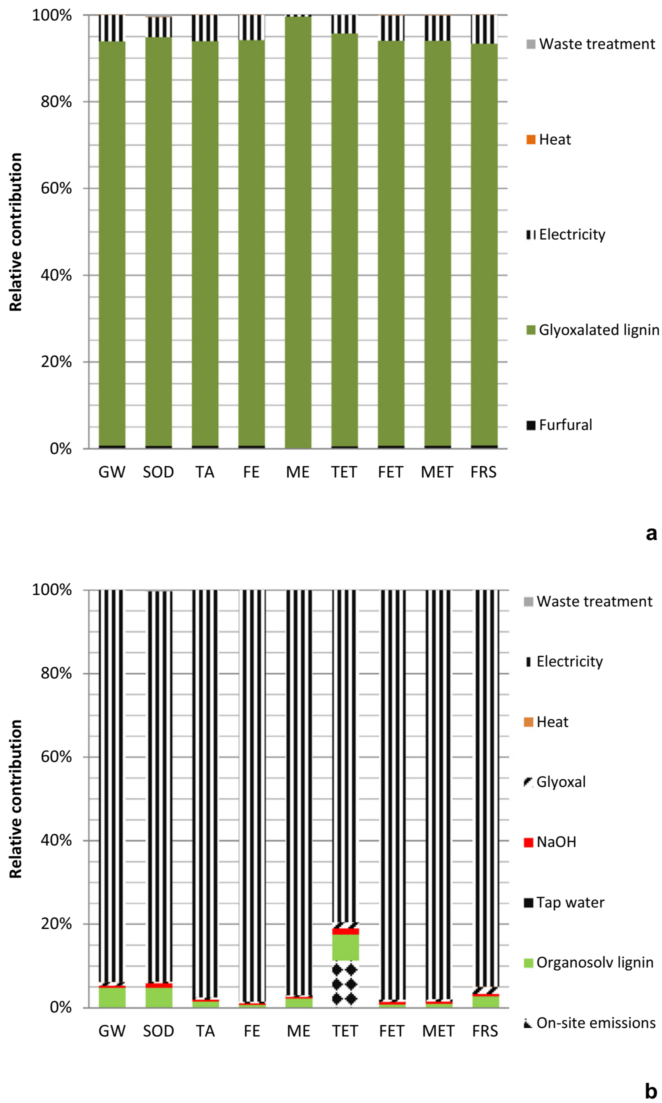


Fig. 4. Distribution of burdens between contributing inputs/outputs involved. a) Organosolv lignin-based bio-adhesive production; b) Glyoxalated lignin production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

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

3.4. Environmental performance of Tannin based bio-adhesive

Fig. 5a depicts the distribution of burdens between the impact categories for this resin, where condensed tannin is used as raw material in the production of bio-adhesive. The results show that two parameters are by far responsible for the largest contributions to the environmental profile, regardless of the impact category, i.e. the production of condensed tannin and glyoxal. The background activities involved in the manufacture of tannin are responsible for environmental contributions ranging from 58% to 83%, depending on the category, except in ME (11%) and TET (15%). Regarding FE, the production of glyoxal required for the bio-adhesive formulation plays a key role with a contribution of 39% (higher than the one for the production of the condensed tannin, 27%) due to the emissions of glyoxal into water from the background processes involved. Concerning TET, air emissions derived from the manufacture of the bio-adhesive are responsible for 33% of the

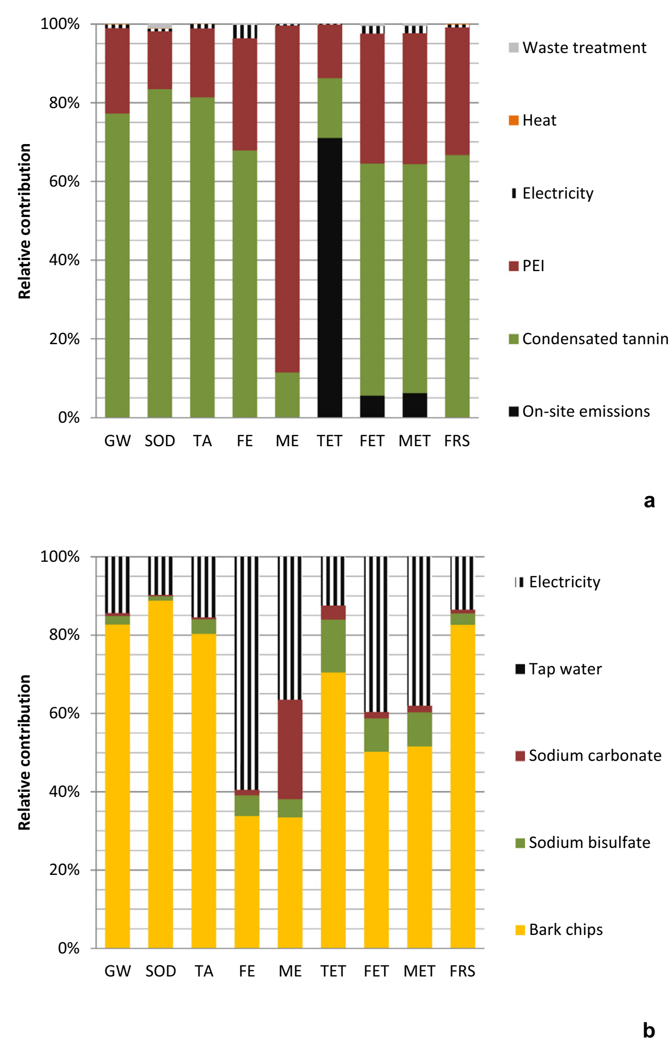


Fig. 5. Distribution of burdens between contributing inputs/outputs involved. a) Tannin-based bio-adhesive production; b) Condensated tannin production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication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

contributions to this impact category, which are associated to the realease of glyoxal (the effect of these emissions in FET category is around 1% of the total contributions). Accordingly, attention should be paid to the control of on-site emissions produced in the factory. The action on the production of glyoxal is not proposed for assessment as it is outside the performance of the plant.

Nevertheless, an optimization of the glyoxal dose could be interesting to reduce the burdens. As far as the production of the raw material: tannin is concerned, it is one of the main contributors in numerous environmental impact categories (see Fig. 5a). Therefore, a detailed analysis of this process is required with the aim not only of identifying the hotspots responsible but also of proposing further improvements (if possible). The distribution of burdens between the processes involved in the production of condensated tannin is shown in Fig. 5b. Consequently, one process involved in the production of the condensed tannin presents the major contributions to all impact categories and that is, the production of the bark chips. It is important to bear in mind that bark chips have not been managed as a waste from sawnmilling activities or from other wood processing facilities. The rationale behind this approach is based on the fact that bark chips have multiple potential applications, such as energy, biofoams or chemicals production (González-García et al. 2016; Feng et al. 2013). In this study, bark chips from a representative sawmill have been taken into consideration and a volume-allocation approach (14.4%) has been assumed to assign the impacts of sawnmilling activities between bark and other co-products (sawn timber and residual wood). According to Fig. 5b, bark chips production is the greatest contributor to GW (83%), SOD (89%), TA (80%), TET (71%), FET (50%), MET (52%) and FRS (83%). In these categories, the forestry activities involved in the production of the required roundwood are behind these high ratios, specifically due to the diesel requirements in the forest machinery (mainly the harvester). Tannin production includes the mixing of the bark chips with hot water and extractive chemicals in a vessel, the subsequent evaporation step to concentrate the extract and a final spray-drying. As a result, there is a significant electricity demand and its production (which is taken directly from the grid) greatly contributes to FE (59%), ME (36%), FET (40%) and MET (38%). One of the extractive chemicals required to extract tannin from the bark, i.e., sodium carbonate, plays a key role in terms of ME (25%). Sodium carbonate is co-produced in a factory together with ammonium chloride using the Solvay process. Therefore, there is a consumption of liquid ammonia and as consequence, there are indirect ammonium emissions into water derived from this industrial process, which are behind the effect from sodium carbonate on ME.

3.5. Sensitivity analysis and improvement options

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

3.5.1. Soy-based bio-adhesive

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

3.5.2. Kraft lignin based bio-adhesive

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

1. Firstly, a sensitivity assessment has been conducted focused on the reduction of these parameters in the glyoxalated lignin taking into account that they could be optimized under industrial conditions. A reduction of 10% and 20% in electricity requirements combined with reductions of 10% and 25%, respectively, in on-site emissions. Fig. 6b depicts the comparative profile under the introduction of both improvement considerations. Accordingly, a 10% reduction in both parameters has associated slight improvements in the global profile, ranging from 2% to 8%. Nevertheless, a reduction of both 20% in electricity demand and 25% in on-site emissions involves improvements of up to 17% in categories such as FE, TA, FET and MET.
2. Secondly, an optimization in the dose of PEI required in the formulation of the bio-adhesive could be really attractive from an environmental perspective, since this parameter plays a key role in the global profile as depicted in Fig. 3a. Therefore, a sensitivity assessment has been conducted considering potential reductions of 10% and 20% on the PEI consumption. Improvements should be achieved as depicted in Fig. 6c specifically in this category where PEI is considered as the hotspot that is ME. In this category, the burdens should be reduced around 7% and 15%, respectively for 10% and 20% of reductions.

3.5.3. Organosolv lignin based bio-adhesive

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

3.5.4. Condensed tannin-based bio-adhesive

In this scenario, a double sensitivity analysis has been proposed based on improvements on on-site emissions and chemical dose.

1. Reduction of on-site emissions from tannin-based bio-adhesive. As detailed above, on-site emissions considerably contributes to TET. These are uncontrolled emissions, which could be reduced by means of the installation of an specific control system. Two reduction ratios have been analysed, 10% and 25%. Accordingly, improvements of 3% and 8% could be achieved in TET, respectively for the reduction proposed of 10% and 25%. As expected, there is no effect on the remaining categories as detailed in Fig. 6d.
2. Reduction of glyoxal dose in the production of tannin based bio-adhesive. As detailed in Fig. 5a, the consumption of glyoxal is an environmental hotspot because of the effect on the profile from the background activities involved. An optimization of its consumption dose should be associated with an environmental improvement in the profile of tannin-based bio-adhesives. Reductions on this parameter have been combined with the proposal discussed above, as they should be directly related. It can be seen that the environmental profile is considerably improved when actions are proposed on glyoxal consumption. A reduction of 10% should lead to a reduction of burdens in all impact

categories but especially, in terms of FET, MET and FRS (around 4%). When the reduction ratio is increased to 20%, the effect is more noticeable achieving improvement ratios of around 9% in the mentioned categories as detailed in Fig. 6f.

3.6. Comparison with conventional adhesives

Conventional adhesives or resins used for the production of wooden flooring are UF, PF and MUF. These resins derive from fossil resources

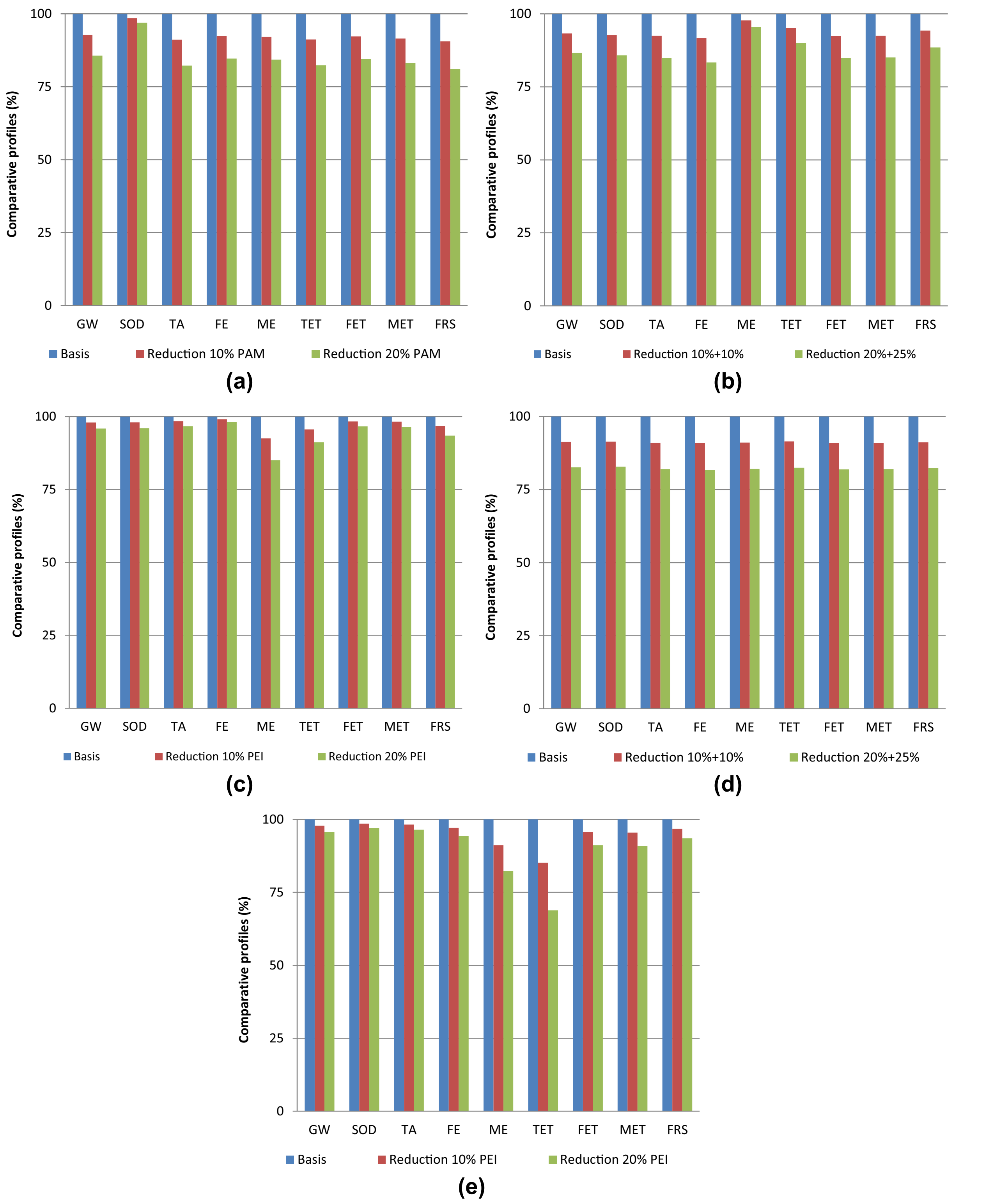


Fig. 6. Sensitivity analysis over the global profiles of bio-adhesives production under study, conducted on a) PAM dose (−10% and −20%) in soy-based bio-adhesive; b) Electricity consumption (−10% and −20%) and on-site emissions (−10% and −25%) in glyoxalation step in Kraft lignin based bio-adhesive;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

and have associated negative environmental problems, mainly due to the formaldehyde emissions produced during their entire life cycle. Nevertheless, and although the interest on developing technologies to produce renewable adhesives as fossil substitutes is the order of the day, the production of bio-adhesives is still under development and the commercional bio-adhesives have a comparatively higher cost than conventional ones (Hemmilä et al. 2017). In this regard, an environmental comparison between conventional resins and bio-adhesives proposed for analysis based on an end-point method has been proposed in order to have a single environmental score per adhesive (in millipoints –mPt). For this purpose, the normalization and weighing factors taken from ReCiPe 2016 Endpoint method have been considered. The analysis at endpoint level reports the results in terms of three indicators to express the relative severity of damage according to the following: Human Health (HH), Ecosystem Quality (EQ) and Resource Scarcity (RS). Those three endpoint indicators are the result of an aggregation process from midpoint categories by means of specific endpoint characterization factors. In this analysis, only those impact categories previously selected for the environmental assessment at midpoint level (GW, SOD, TA, FE, ME, TET, FET, MET and FRS) will be considered for the estimation of the single environmental score, as depicted in Fig. 7a, taking into account the corresponding normalization and weighthing factors established by the method.

The result of the comparison is illustrated in Fig. 7b and the size of the circles corresponds to the magnitude of the global impact. In general, the impact trend revealed by the bio-adhesives does not follow that of the fossil-ones, except in the case of soybean and tannin based bio-adhesives. The single score corresponding to the soy-based adhesive (43 mPt) and tannin-based adhesive (46 mPt) are in line with the one for UF resin (41 mPt) and around 25% lower and 30% higher than the scores for PF resin (56 mPt) and MUF resin (33 mPt), respectively. Thus, soy based bio-adhesive can be considered as an environmentally potential substitute to the fossil resins. On the contrary, the single scores estimated for the other three bio-adhesives are considerably higher than those for fossil ones as depicted in Fig. 7b – Organosolv lignin-based (228 mPt) reports the worst profile followed by tannin-based (122 mPt) and Kraft lignin-based (123 mPt) adhesives. Accordingly, further research on the development of these three bio-adhesives is required in order to be environmentally competitive, taking into account the assumptions established in our systems (see Section 2.3). Given the fact that bio-adhesives have a significant contribution from renewable feedstocks, it could be expected a more significant difference between fossil and renewable adhesives. However, it must be noticed that in no case feedstocks have been considered as waste from other activites. Thus, it must be remarked the effect over the overall profile of the demand of fossil resources in the background processes involved in the production of the feedstocks. This is the case, for example, of diesel in agricultural and forestry activities, as well as natural gas and electricity in pulp manufacturing activities. If a detailed analysis of the scores and the distribution between the level of damage (HH, EQ and RS) is carried out, the scores, regardless of the adhesive, are mainly affected by the damage to human health (on average, 78%). In all adhesives, the use of fossil resources in their background systems is behind the contributions to HH, mainly due to GHG emissions (99% of total contributions). This effect is considerable outstanding in the Organosolv lignin based bio-adhesive (6 and 7 times higher than soy and MUF-based adhesives, respectively). The rationale behind that results is the effect over the profile from the high electricity requirements (and thus, corresponding production effect) to perform the glyoxalation of the lignin, a process required for the functionalization of lignin.

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

A direct comparison of the results of our study with those of others already reported has been quite complicated (McDevitt and Grigsby 2014; Yang and Rosentrater 2020), mainly due to the constrains of using different databases and environmental assessment methods. McDevitt and Grigsby (2014) considered a protein-lignin adhesive formulated with corn protein, soy protein, lignin and tannin. When comparing our results with those of McDevitt and Grigsby (2014), both studies highlight the higher impact on ecosystem damage associated to bio-adhesives over fossil ones, mainly due to the effect of the renewable feedstock. Yang and Rosentrater (2020) analysed the production of bio-glycerol based adhesive (from soybean) and fossil glycerol based adhesive but considering alternative assessment methods. Our study agrees with some of the findings reported in that study in which soy-based adhesive had a lower impact than fossil one. Moreover, the impact of resource scarcity damage is higher in fossil adhesive than in the bio one (around 50%), while in our study the PF and UF reported an impact on resource scarcity damage 59% and 10% higher than the soy-based adhesive. The same trend is observed in human health damage, indicating that soy-based adhesive is less harmfull to humans. Nevertheless, this trend is not observed in damage to EQ since Yang and Rosentrater (2020) also identified major impact on fossil adhesives, opposed to our study and McDevitt and Grigsby (2014).

As detailed above, the bio-adhesives analysed are based on raw materials that are not considered to be waste from other industrial activities and therefore the background activities involved in their production are included within the system boundaries as well as derived burdens are computed. In the case of tannin-based adhesive, bark chips from sawmilling activities have been managed as raw material. Nevertheless, bark could be alternatively taken from wood-based industrial processes, where it is co-produced as a residue. This could be the case for bark from forestry activities or even from panel factories, where the accumulation of tons of bark-based waste can be identified. As for Kraft lignin, it is derived from the black liquor obtained in the pulp stage that is commonly used for energy purposes in kraft pulp mills, combined with other wood residues.

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

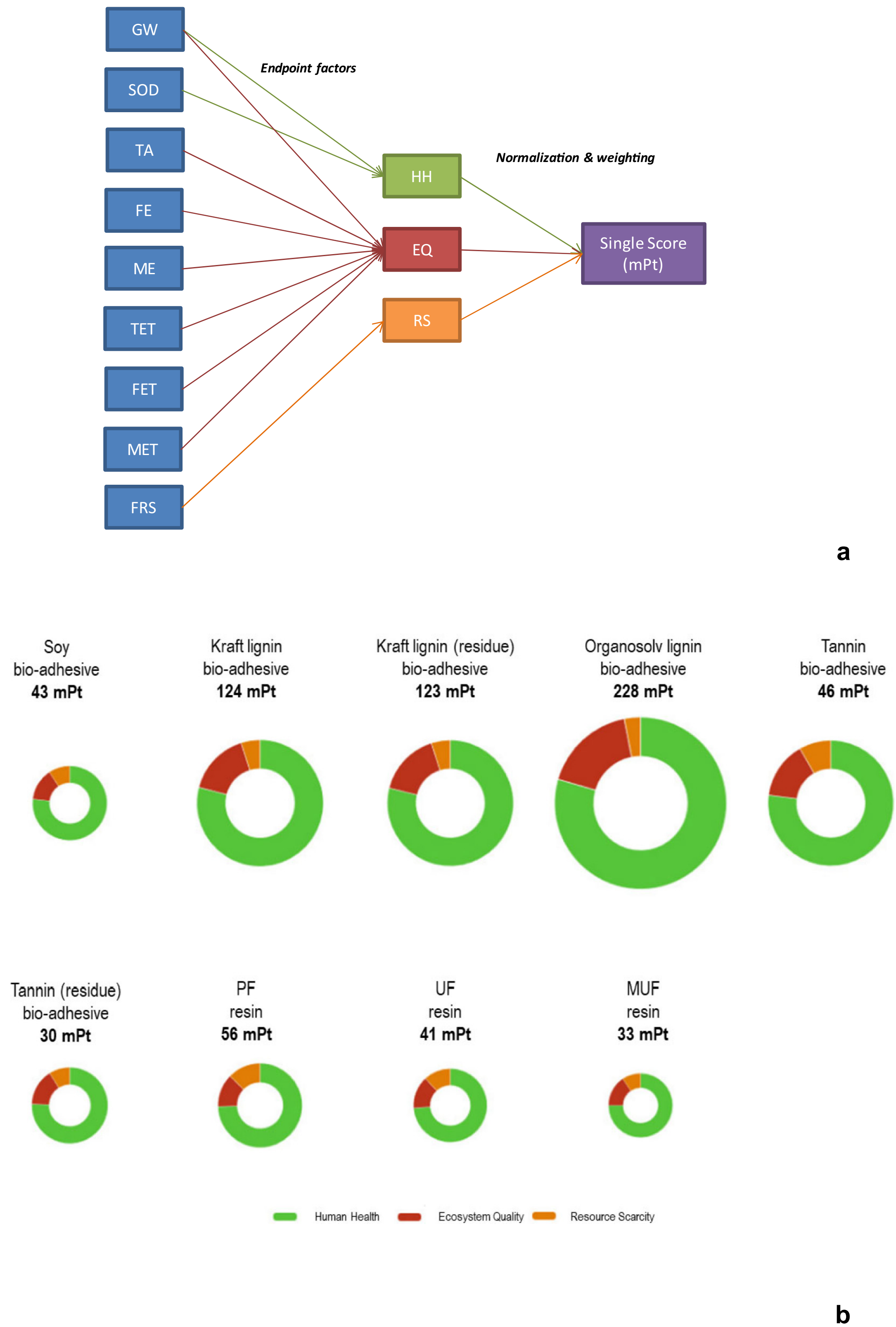


Fig. 7. a) Schematic procedure for estimating single environmental scores; b) Comparison of Life cycle impacts and damage contributions on human health, ecosystem and resources.

4. Conclusions

Given the variety of feedstocks that can be used as such to produce substitutes for alternative fossil resins, it can be concluded that there is a wide spectrum of possibilities for producing more environmental-friendly adhesives, which will reduce dependence on crude oil. However, there are only a few industrial bio-adhesive plants that supply products at high costs. According to this study, it is confirmed that LCA can be a useful tool to perform a gross evaluation and comparison between adhesives in order to identify the advantages and disadvantages, as well as with the aim of identifying parameters on which to act in the short and medium term. Nevertheless, attention needs to be paid on the current constraints that considerably affect the outcomes and thus, decision making related with the early development in which are the bio-adhesives production systems. This situation makes it difficult to compare with the conventional fossil resins, which are totally optimized and developed, supplying products at low price but involving controversial impacts. Our results show that soy-based bio-adhesive can be considered as a potential fossil substitute.

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

CRediT authorship contribution statement

Ana Arias: Methodology. **Sara González-García:** Writing - original draft, Formal analysis, Writing - review & editing, Validation. **Sandra González-Rodríguez:** Methodology. **Gumersindo Feijoo:** Validation. **María Teresa Moreira:** Conceptualization, Writing - original draft, Writing - review & editing, Validation.

Declaration of competing interest

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

Acknowledgements

This research has been financially supported by an ERA-CoBIOTECH project (PCI2018-092866) Programación Conjunta Internacional 2018 – WOOBADH project. Dr. S. González-García would like to express their gratitude to the Spanish Ministry of Economy and Competitiveness for financial support (Grant reference RYC-2014-14984). S. González-Rodríguez thanks to the Spanish Ministry of Science, Innovation and Universities for financial support (Grant reference BES-2017-081677). The authors belong to the Galician Competitive Research Group GRC 2013-032 and to CRETUS Strategic Partnership (AGRUP2015/02). All these programmes are co-funded by Xunta de Galicia and FEDER (EU).

References

Abdalla, S., Pizzi, A., Ayed, N., Charrier-El Bouthoury, F., Charrier, B., Bahabri, F., Ganash, A., 2014. Maldi-Tof analysis of Aleppo pine (Pinus halepensis) bark tannin. BioResoures 9, 3396–3406.

Ang, A., Ashaari, Z., Suhaimi Bakar, E., Ibrahim, N.A., 2015. Characterization and optimization of the glyoxalation of a methanol-fractionated alkali lignin using response surface methodology. BioResources, 10. https://ojs.cnr.ncsu.edu/index.php/BioRes/article/view/BioRes_10_3_4795_Ang_Optimization_Glyoxalation_Alkali_Lignin.

Bello, S., Ríos, C., Feijoo, G., Moreira, M.T., 2018. Comparative evaluation of lignocellulosic biorefinery scenarios under a life-cycle assessment approach. Biofuels, Bioprod. Bioref. 12, 1047–1064.

Cherubini, F., Strømman, A.H., 2011. Life cycle assessment of bioenergy systems: state of the art and future challenges. Bioresour. Technol. 102, 437–451.

Collingea, W.O., Thiela, C.L., Campiona, N.A., Al-Ghamdia, S.G., Woloschina, C.L., Soratanab, K., Landisb, A.E., Bilec, M.M., 2015. Integrating life cycle assessment with green building and product rating systems: north American perspective. Procedia Eng 118, 662–669.

Dababi, I., Gimello, O., Elaloui, E., Quignard, F., Brosse, N., 2016. Organosolv lignin-BasedWood adhesive. Influence of the lignin extraction conditions on the adhesive performance. Polymers 8, 340.

Dongre, P., Driscoll, M., Amidon, T., Bujanovic, B., 2015. Lignin-furfural based adhesives. Energies 8, 7897–7914.

Ebnesajjad, S., 2009. Chapter 4 classification of adhesives and compounds. Adhesives Technology Handbook. Elsevier, pp. 47–62. https://doi.org/10.1016/B978-0-8155-1533-3.50007-4.

El-Mansouri, N., Pizzi, A., Salvadó, J., 2007. Lignin-based wood panel adhesives without formaldehyde. Holz Roh Werkst. 65, 65–70.

Feng, S., Cheng, S., Yuan, Z., Leitch, M., Xu, C., 2013. Valorization of bark for chemicals and materials: a review. Renew. Sust. Energ. Rev. 26, 560–578.

Geng, X., Li, K., 2006. Investigation of wood adhesives from kraft lignin and polyethylenimine. J. Adhes. Sci. Tecnol. 20 (8), 847–858. https://doi.org/10.1163/156856106777638699.

Globenewswire, 2017. https://www.globenewswire.com/news-release/2017/10/17/1148538/0/en/Global-Adhesives-Market-2017-2022-53-5-Billion-Opportunity-Analysis-and-Industry-Forecasts.html, Accessed date: 24 March 2020.

González-García, S., Feijoo, G., Heathcote, C., Kandelbauer, A., Moreira, M.T., 2011a. Environmental assessment of green hardboard production coupled with a laccase activated system. J. Clean. Prod. 19, 445–453.

González-García, S., Hospido, A., Agnemo, R., Svensson, P., Selling, E., Moreira, M.T., Feijoo, G., 2011b. Environmental life cycle assess¬ment of a Swedish dissolving Pulp Mill integrated biorefinery. J. Ind. Ecol. 15, 568–583.

González-García, S., Lacoste, C., Aicher, T., Feijoo, G., Lijó, L., Moreira, M.T., 2016. Environmental sustainability of bark valorisation into biofoam and syngas. J. Clean.Prod. 125, 33–43.

Hemmilä, V., Adamopoulos, S., Karlssonb, O., Kumar, A., 2017. Development of sustainable bio-adhesives for engineered wood panels – a review. RSC Adv. 7, 38604.

Huijbregts, M.A.J., Steinmann, Z.J.N., Elshout, P.M.F., Stam, G., Verones, F., Vieira, M.D.M., Hollander, A., Zijp, M., van Zelm, R., 2017. ReCiPe 2016 v1.1.

ISO 14040, 2006. Environmental Management - Life Cycle Assessment - Principles and Framework. 2nd edition. (Geneva, Switzerland).

Jin, Y., Cheng, X., Zheng, Z., 2010. Preparation and characterization of phenol–formaldehyde adhesives modified with enzymatic hydrolysis lignin. Bioresour. Technol. 101, 2046–2048.

Kautto, J., Realff, M., Ragauskas, A.J., 2013. Design and simulation of an organosolv process for bioethanol production. Biomass Conversion and Biorefinery 3, 199–212.

Kim, K., 2009. Environment-friendly adhesives for surface bonding of wood-based flooring using natural tannin to reduce formaldehyde and TVOC emission. Bioresour. Technol. 100, 744–748.

Kolfschoten, R.C., Bruins, M.E., Sanders, J.P.M., 2014. Opportunities for small-scale biorefinery for production of sugar and ethanol in the Netherlands. Biofuels Bioprod. Biorefin. 8, 475–486.

Li, K., Geng, X., Simonsen, J., Karchesy, J., 2004. Novel wood adhesives from condensed tannins and polyethylenimine. Int. J. Adhes. Adhes. 24, 327–333.

Li, X., Li, Y., Zhong, Z., Wangb, D., Ratto, J.A., Sheng, K., Sun, X.S., 2009. Mechanical and water soaking properties of medium density fiberboard with wood fiber and soybean protein adhesive. Bioresour. Technol. 100, 3556–3562.

Liu, S., Lu, H., Hu, R., Shupe, A., Line, L., Liang, B., 2012. A sustainable woody biomass biorefinery. Biotechnol. Adv. 30, 785–810.

Luo, J., Li, G., Li, X., Luo, J., Gao, Q., Li, J., 2015. A new soybean meal-based bio-adhesive enhanced with 5, 5-dimethyl hydantoin polyepoxide for the improved water resistance of plywood. RSC Adv. 5, 62957–62965.

Luo, Y., Li, Z., Li, X., Liu, X., Fanc, J., 2019. The production of furfural directly from hemicellulose in lignocellulosic biomass: a review. Catal. Today 319, 14–24.

MarketsandMarkets, 2019. https://www.marketsandmarkets.com/PressReleases/adhesive-and-sealant.asp, Accessed date: April 2020.

Mathias, J.-D., Grédiac, M., Michaud, P., 2016. Bio-based adhesives. Biopolymers and Biotech Admixtures for Eco-Efficient Construction Materials, 1st Edition Woodhead Publishing, p. 464. https://doi.org/10.1016/B978-0-08-100214-8.00016-6 Chapter 16.

McDevitt, J.E., Grigsby, W.J., 2014. Life cycle assessment of bio- and petro-chemical adhesives used in fiberboard production. J. Polym. Environ. 22, 537–544.

Moubarik, A., Pizzi, A., Allal, A., Charrier, F., Charrier, B., 2009. Cornstarch and tannin in phenol-formaldehyde resins for plywood production. Ind. Crop. Prod. 30, 188–193.

Navarrete, P., Mansouri, H.R., Pizzi, A., Tapin-Lingua, S., Benejlloun-Mlayah, B., Pasch, H., et al., 2010. Wood panel adhesives from low molecular mass lignin and tannin without synthetic resins. J. Adhes. Sci. Tecnol. 24, 1597–1610.

Nitzsche, R., Budzinski, M., Gröngröft, A., 2016. Techno-economic assessment of a wood-based biorefinery concept for the production of polymer-grade ethylene, organosolv lignin and fuel. Bioresour. Technol. 200, 928–939.

Pizzi, A., 2006. Recent developments in eco-efficient bio-based adhesives for wood bonding: opportunities and issues. J. Adhes. Sci. Technol. 20, 829–846.

Posada, J.A., Patel, A.D., Roes, A., Blok, K., Faaij, A.P.C., Patel, M.K., 2013. Potential of bioethanol as a chemical building block for biorefineries: preliminary sustainability assessment of 12 bioethanol-based products. Bioresour. Technol. 135, 490–499.

PRé Consultants, 2020. SimaPro Database Manual. Methods library, The Netherlands.

Radoykova, T., Nenkova, S., Valchev, I., 2013. Balck liquor lignin products, isolation and characterization. J. Chem. Technol. Metall. 48, 524–529.

Rajagopalan, N., Bilec, M.M., Landis, A.E., 2012. Life cycle assessment evaluation of green product labeling systems for residential construction. Int. J. Life Cycle Assess. 17, 753–763.

Saad, H., Khoukh, A., Ayed, N., Charrier, B., Charrier-El Bouhtoury, F., 2014. Characterization of Tunisian Aleppo pine tannins for a potential use in wood adhesive formulation. Ind. Crop. Prod. 61, 517–525.

Sanders, J., Scott, E., Weusthuis, R., Mooibroek, H., 2007. Biorefinery as the bio-inspired process to bulk chemicals. Macromol. Biosci. 7, 105–117.

Sanders, J.P.M., Clark, J.H., Harmsen, G.J., Heeres, H.J., Heijnen, J.J., Kersten, S.R.A., van Swaaij, W.P.M., Moulijn, J.A., 2012. Process intensification in the future production of base chemicals from biomass. Chem. Eng. Process. 51, 117–136.

Sellers, T., 2001. Wood adhesive innovations and applications in north America. Forest prod. J. 51, 11–22.

Silva, D.A.L., Lahr, F.A.R., Varanda, L.D., Christoforo, A.L., Ometto, A.R., 2015. Environmental performance assessment of the melamine-urea formaldehyde (MUF) resin manufacture: a case study in Brazil. J. Clean. Prod. 96, 299–307.

Soymeal.org, 2020. https://www.soymeal.org/wp-content/uploads/2018/04/soybean_processing.pdf (accessed April, 2020).

Stefani, P.M., Peña, C., Ruseckaite, R.A., Piter, J.C., Mondragon, I., 2008. Processing conditions analysis of Eucalyptus globulus plywood bonded with resol-tannin adhesives. Bioresour. Technol. 99, 5977–5980.

de la Torrea, M.J., Moral, A., Hernández, M.D., Cabeza, E., Tijero, A., 2013. Organosolv lignin for biofuel. Ind. Crop. Prod. 45, 58–63.

Uihlein, A., Schebek, L., 2009. Environmental impacts of a lignocellulose feedstock biorefinery system: an assessment. Biomass Bioenergy 33, 793–802.

Van Langenberg, K., Grigsby, W., Ryan, G., 2010. Green Adhesives: Options for the Australian Industry—Summary of Recent Research into Green Adhesives from Renewable Materials and Identification of those that Are Closest to Commercial Uptake. Project No. PNB158-0910. FWPA.

Vargas, F., Domínguez, E., Vila, C., Rodríguez, A., Garrote, G., 2016. Biorefinery scheme for residual biomass using autohydrolysis and organosolv stages for oligomers and bioethanol production. Energy Fuel 30, 8236–8245.

Wernet, G., Bauer, C., Steubing, B., Reinhard, J., Moreno-Ruiz, E., Weidema, B., 2016. The ecoinvent database version 3 (part I): overview and methodology. Int. J. Life Cycle Assess. 21, 1218–1230.

Williams, C.K., Hillmyer, M.A., 2008. Polymers from renewable resources: a perspective for a special issue of polymer reviews. Polym. Rev. 48, 1–10.

Yang, M., Rosentrater, K.A., 2019a. Techno-economic analysis of the production process of structural bio-adhesive derived from glycerol. J. Clean. Prod. 228, 388–398.

Yang, M., Rosentrater, K.A., 2019b. Life cycle assessment and techno-economic analysis of pressure sensitive bio-adhesive production. Energies 12, 4502.

Yang, M., Rosentrater, K.A., 2020. Cradle-to-gate life cycle assessment of structural bio-adhesives derived from glycerol. Int. J. Life Cycle Assess. https://doi.org/10.1007/s11367-020-01733-9.

Zhang, J., Xi, X., Liang, J., Pizzi, A., Du, G., Deng, S., 2019. Tannin-based adhesive cross-linked by furfuryl alcohol-glyoxal and epoxy resins. Int. J. Adhes. Adhes. 94, 47–52.