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Environmental benefits of soy-based bio-adhesives as an alternative to formaldehyde-based options

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Abstract:	The restrictions imposed in relation to the use of formaldehyde in the formulation of the synthetic resins used in the manufacture of wood panels have been the driving force in the development of clean production strategies. The use of non-renewable raw materials is desirable to be changed by bio-based options. In this context, the environmental profile of different alternatives of soy-based adhesives is investigated, as possible options for replacing the commonly used synthetic resins. This report includes the modeling of the reaction stage on a large scale as well as the evaluation of life cycle impacts associated with each soy-based alternative. Among the six proposals presented, the one that stands out for its potentiality to replace synthetic resins is the soy protein bio-adhesive with tannin-based resin.
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Philippe Garriges
Editor-in-Chief, Biomass & Bioenergy.

Dear Philippe Garriges:

We are pleased to enclose an original manuscript of our paper entitled *“Environmental benefits of soy-based bio-adhesives as an alternative to formaldehyde-based options”* by Ana Arias, Sara González-García, Gumersindo Feijoo and Maria Teresa Moreira which can hopefully be published in the *Environmental Science and Pollution Research*. This paper has not been previously published, in whole or in part, and is not under consideration by any other journal.
The restrictions imposed in relation to the use of formaldehyde in the formulation of the synthetic resins used in the manufacture of wood panels have been the driving force in the development of bio-based options. In this context, the environmental profile of different alternatives of soy-based adhesives is investigated, as possible options for replacing the commonly used synthetic resins. This report includes the modeling of the reaction stage on a large scale as well as the evaluation of life cycle impacts associated with each soy-based alternative. Among six different options, soy protein bio-adhesive with tannin-based resin has an overall better profile. Sensitivity analysis on the formulation and process conditions lead to identify potential improvements in the environmental profile. This study provides useful information for researchers on where to focus on the development of bio-adhesives.

We hope that this work is appropriate for publication in *Environmental Science and Pollution Research*.

Yours sincerely,

Ana Arias Calvo

Environmental benefits of soy-based bio-adhesives as an alternative to formaldehyde-based options

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Abstract

The restrictions imposed in relation to the use of formaldehyde in the formulation of the synthetic resins used in the manufacture of wood panels have been the driving force in the development of clean production strategies. The use of non-renewable raw materials is desirable to be changed by bio-based options. In this context, the environmental profile of different alternatives of soy-based adhesives is investigated, as possible options for replacing the commonly used synthetic resins. This report includes the modeling of the reaction stage on a large scale as well as the evaluation of life cycle impacts associated with each soy-based alternative. Among the six proposals presented, the one that stands out for its potentiality to replace synthetic resins is the soy protein bio-adhesive with tannin-based resin.

Keywords: Soybean; Soy protein; Wood panel; Sensitivity analysis; Life Cycle assessment; Environmental profile

1. Introduction

The wood-based panel sector has achieved significant positions in the global market, with a size of 144.67 USD billion by 2019, and grow significantly in terms of CAGR of up to 6.9% from 2020 to 2027 (Grand View Research, 2020). Regarding the different types of wood panels, the category that stands out is plywood: a wood product manufactured from the gluing of multiple thin layers of wood with excellent properties in terms of strength, durability, water resistance, among others (Jia et al., 2019). Although this type of product is made from wood and is supposed to be environmental-friendly, the manufacture of boards requires adhesives in the gluing stage (González-García et al., 2009), which are traditionally fossil-based (phenol-formaldehyde, urea-formaldehyde, etc.). Their use has posed some environmental problems related to formaldehyde emissions, not only in the manufacturing process but also diffuse emissions that can affect indoor air quality (Hemmilä et al., 2017).

One of the options being considered to develop adhesives with lower environmental impact is based on the partial or total replacement of formaldehyde by the formulation of vegetable protein-based bio-adhesives (Kajaks et al., 2012). The techno-economic viability of this type of bio-adhesives should ensure that a number of requirements are met; similar performance to synthetic alternatives, adequate strength and stiffness properties according to standards and competitive cost (Frihart et al., 2014). In addition, it is anticipated that the use of bio-adhesives could lead to a number of environmental benefits, especially regarding the depletion of fossil resources and associated CO_2 emissions. Despite the promising benefits of bio-based adhesives, an unbiased holistic perspective must be conducted, so that all environmental aspects related to the process are addressed; especially if the impact categories linked to the energy consumption of a process that has not been optimized or also the land use for biomass cultivation are taken into account.

Soy is a renewable resource with favorable characteristics for use as a raw material in the manufacture of bio-adhesives, namely, abundance, ease of processing and low cost (Vnuĉec et al., 2017). Its derivatives, including soy protein (SPI), defatted soy flour (DSF) and soy flour (SF) are ideal materials for the production of bio-adhesives. Although soy protein has high protein purity, with levels exceeding 90% raw protein (Hojilla-Evangelista 2010), the transition to an industrial-scale process has been significantly limited due to its high cost. For this reason, recent research related to soy-based adhesives has been directed primarily at modifying soy flour fractions. Analysis of the performance of soy-based bio-adhesives shows acceptable but not excellent water resistance, which may be a disadvantage compared to well-established alternatives of petrochemical origin (Lei et al., 2014). In order to solve these limitations, it is considered that the addition of cross-linking agents can improve the performance of the adhesive (Ferdosian et al., 2017). For this reason, the optimization of the use of crosslinking agents and the adaptation of the process operating conditions are key aspects to improve the environmental, technical and economic profiles of the bio-adhesives.

Looking at the most recent reports in literature, the environmental profile of bio-adhesives has been scarcely addressed (McDevitt & Grigsby, 2014; Yang et al., 2020; Arias et al., 2020). Moreover, the comparison between different bio-adhesive options is not simple because it will depend on whether the selection of the functional unit, the system boundaries, and the impact assessment methodology are similar. The available reports on soy-based adhesives focus on the formulation of the adhesive and its validation for use in the wood processing industry (He, Z., 2017; Ferdosian et al. 2017). This study aims to provide essential information on the different alternatives for soy-based adhesives for which no environmental studies have been carried out. For this purpose, due to the lack of production data on an industrial scale, it is necessary to pose mass and energy balances required in the process simulation using the Aspen Hysys® tool. The conceptual design of the process is essential to provide key inventory data on the production process, not only raw materials, products and energy but also emissions from each stage of the manufacturing process. It will be based on the quantitative analysis of global environmental impacts according to the life cycle methodology, when it will be possible to identify real competition opportunities between a widely tested fossil-based adhesive with a biotechnological alternative that aims to stand out as a more sustainable product.

2. Methodology

2.1. Definition of the goal and scope of the study

The aim of this study is to determine the environmental burdens associated with the production of soy-based adhesives on a large scale, in order to evaluate their potential for use in the wood panel industry as substitutes for the fossil-based resins currently in use. Given that the production processes of bio-adhesive are not developed on an industrial scale, a simulation tool has been used with the aim of extrapolating the available data reported at laboratory and pilot scale.

As for soy-based adhesives, the most commonly used crosslinkers for their production, and those that have proven to be effective, are those based on epoxy chemicals and aldehydes, as well as their derivatives (Lei et al., 2014), which has permitted to identify six scenarios of soy-based bio-adhesives, which result of the combination of soy flour and soy protein with the following crosslinkers and additives: dicyandiamide, waterborne polyurethane, Sodium dodecylbenzene sulfonate (SDS) and epoxy resin, polyacrylamide and epoxy resin, maleic anhydride and tannin-based resin.

Thus, although the raw material used to formulate this type of bio-adhesive is based on soy protein, other chemicals used in its production can also have a significant environmental impact. For this reason, this study aims to determine the environmental profiles associated with the production of the different alternatives of soy-based bio-adhesives and to demonstrate (or not) their environmental benefits when compared with their fossil-based counterparts. Thus, a comparison will be made with the most used fossil adhesives in the manufacture of wood panels: urea-formaldehyde (UF), phenol-formaldehyde (PF) and melamine-urea-formaldehyde (MUF). Therefore, the life cycle assessment (LCA) methodology (ISO 14040, 2016) is proposed for the assessment of environmental impacts from a global point of view.

This methodology allows the use of different evaluation methods: ReCiPe 2016 hierarchist Midpoint method V1.03 World (2010), which was the tool for reporting the environmental profile and identifying the environmental hotspots in terms of 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).

Once the environmental profiles of soy-based bio-adhesive alternatives have been determined, their comparison with fossil-based alternatives will be conducted in terms of a single score that integrates three impact categories (human health, ecosystem quality, and resource scarcity). Finally, it is important to mention that, in addition to the need to reduce dependence on fossil resources and improve the quality of the environment, another reason that drives the development of studies and research for alternatives to synthetic adhesives is the interest of consumers to acquire more sustainable products with less potential risk to human health. Therefore, it is important to study the impact of adhesives on human toxicity, for which the USEtox ® V1.01 assessment method has been selected.

Considering that the manufacturing process of bio-adhesives is not developed on a real scale, mass and energy balances will be developed within the framework of a conceptual design based on the data available at laboratory scale. Simulations of the bio-adhesive production systems are then carried out using Aspen Hysys® software. The functional unit used to report the environmental results is 1 kg of adhesive.

2.2. System boundaries and assumptions for the study.

A cradle-to-gate approach has been considered in this study (**Figure 1**), which implies that the stages from the extraction of raw materials, bio-adhesive production and on-site emissions are included within the system boundaries, excluding from the analysis the production and maintenance of infrastructure, as well as all transport activities of inputs (chemicals and raw materials) to the factory gate. The rationale behind this hypothesis is based on the consideration that both activities entail minor environmental loads (Jia et al., 2018; Yang & Rosentrater, 2020).

< **Figure 1** over here>

The manufacturing processes of soy-based bio-adhesives have been developed according to the conditions developed at laboratory scale, with the estimation of production yields, emissions or waste streams.

2.3. Description of soy bio-adhesives formulations

Six scenarios have been considered for the production of soy-based bio-adhesives: soy flour bio-adhesive with dicyandiamide as crosslinker (SF+D), bio-adhesive formulated with soybean flour and waterborne polyurethane (SF+WP), soybean flour bio-adhesive crosslinked with SBDS (Sodium dodecylbenzene sulfonate) and epoxy resin (SF+SE), cross-linked soy protein bio-adhesive with epoxy resin and polyacrylamide (SP+EP), soy protein bio-adhesive with maleic anhydride (SP+MA) and soy protein bio-adhesive with tannin-based resin (SP+TR).

The importance of adding crosslinkers in the formulation of bio-adhesives correlates with the need to improve the physical-chemical characteristics of the final product, especially in relation to those relevant to the manufacture of wood panels. The crosslinkers used for the formulation of the different soy-based bio-adhesives proposed could be mixed during the adhesive production process or just before its application to the wood board (Lei et al., 2014). It is also viable in the manufacture of soy flour and soy protein bio-adhesives (Ghahri S. & Mohebby B., 2017). The addition of crosslinkers not only improves the production stage of the board, but also produces the structural quality of the wood board (Frihart, 2005) and increases its tolerance to humidity (Pizzi A. & Ibeh C., 2014).

Selected feedstocks: soy flour and soy protein

In this study, two feedstocks have been proposed for the formulation of soy-based bio-adhesives, namely soy flour and soy protein. The agricultural activities of soybean cultivation, including fertilization and irrigation are included in the analysis. After harvesting, the soaked soybeans are then pre-cooked and dehulled, before being dried, grinded and sieved. Soybean flour was used for the alternatives of SF+D, SF+WP and SF+SE, while soy protein is used for the alternatives of SP+EP, SP+MA and SP+TR. Therefore, the scenarios based on the use of the latter require an additional step within the system boundaries based on the extraction of the protein from the soy flour. The extraction process is intensive in terms of chemicals and energy requirements and information regarding their performance has been taken from Berardi et al. (2015). Regarding the products, the extraction process yields into three co-products: soy protein, whey and spent flour. Accordingly, a mass-based allocation has been considered to allocate the environmental burdens among these co-products. Concerning the soybean meal, inventory data have been taken from the Ecoinvent® database. According to the data available in the literature on the average production of synthetic resin facilities (Yang and Rosentrater, 2019a), a production capacity of 24 t/d of bio-adhesive has been selected for simulation on a large scale.

2.3.1. Soy flour bio-adhesive with dicyandiamide as crosslinker (SF+D bio-adhesive)

According to the procedure proposed by Solt et al. (2019), the soy flour is mixed with water at 35°C for 60 min and later HCl containing 0.01% (w/w) of ferric chloride is added to the mixture. These three components are mixed for 30 min under constant agitation. Once this time has elapsed, the cross-linking agent: dicyandiamide is added and the mixture is mixed for 60 min. Finally, NaOH is added to adjust the pH of the adhesive. The data for the foreground system has been estimated by a plant-wide modeling in Aspen Hysys® and a summary of the most relevant inventory data is detailed in **Table SM1** and **Figure S1a** of the Supplementary Material, respectively. The inventory data associated with the production of soy flour has been taken from Ecoinvent® data-base v3.5.

2.3.2. Soy bio-adhesive formulated with soybean flour and waterborne polyurethane (SF + WP bio-adhesive)

One of the main problems related to the use of soy protein-based bio-adhesives is the presence of hydrophilic amino acids in their molecular structure, which has led to weak resistance to moisture. This characteristic could be improved by the use of waterborne polyurethane (WPU) as a cross-linking agent, which would lead to the production of soy bio-adhesives with better mechanical properties and higher water/solvent/thermal and abrasion resistance (Hu et al., 2016; Zhang et al., 2012) through the formation of stronger intermolecular interactions with the reactive groups of the soybean flour. One of the most widely used WPUs for the formulation of soy-based bio-adhesives is called poly-(butylene adipate), a polymer formed by the monomers adipic acid and 1,4-butylene glycol.

WPU are reactive compounds that form covalent and hydrogen bonds. The cross-linking chemical is added by dispersion into the soy flour (Liu et al., 2017), that is why it became necessary to mix the soy flour with water and pH is adjusted using ethylene glycol, urea and sodium hydroxide. This first mixing is done by constant stirring for 60 min at 80°C (Wrang et al., 2019). Once this slurry is formed, the WPU is added to the mixture under constant agitation during 160 min at 76°C to promote the formation of covalent bonds.

After this time, the mixture is cooled to room temperature (Wang et al., 2018). The inventory data for the impact assessment and the system boundaries considered for the production are shown in **Table SM2** and **Figure S1b** in the Supplementary Material, respectively.

2.3.3. Soy flour bio-adhesive crosslinked with SBDS (Sodium dodecylbenzene sulfonate) and epoxy resin (SF + SE bio-adhesive)

The adhesion properties of soy protein are improved with the use of sodium dodecyl benzene sulfonate in terms of increase in water resistance and shear strength (Huang, W. & Sun, X., 2000). The addition of epoxy resin also helps to improve these adhesive properties, as a result of the increased formation of covalent bonds with the reactive groups of the soybean flour.

The formulation of this bio-adhesive requires the reaction of an aqueous solution formed by SDBS, CaO and NaOH with the soy flour, maintaining constant agitation at 88-92 ºC for 3 h. Epoxy resin (6.5% weight) is added to this slurry, resulting in a 41% improvement in wet shear strength compared to the adhesive without epoxy resin (Lei et al., 2014). The inventory data for the impact assessment and the system boundaries considered for the production are shown in **Table SM3** and **Figure S1c** in the Supplementary Material, respectively.

2.3.4. Soy protein bio-adhesive cross-linked with epoxy resin and polyacrylamide (SP + EP bio-adhesive)

First of all, it is important to mention that the epoxy resin used as a bio-adhesive crosslinker is a waterborne epoxy resin, considered to be an effective cross-linking agent for soy bio-adhesives (Luo et al., 2016). The production process is developed in two reaction stages, the first of which is the extraction of the protein from the soybean. The second consists of the addition of the soy protein to water containing 0.05% polyacrylamide for 20 min at 40°C under continuous agitation. The epoxy resin is then added to the mixture, maintaining agitation for 30 min and raising the temperature to 45°C (Wang et al., 2019). The inventory data for the impact assessment and the system boundaries considered for the production are shown in **Table SM4** and **Figure S1d** of the Supplementary Material, respectively.

2.3.5. Soy protein bio-adhesive with maleic anhydride (SP + MA bio-adhesive)

The grafting process between soy protein and maleic anhydride is carried out through the formation of amide bonds and the reaction with hydroxyl groups present in the protein (Liu, Y. & Li, K., 2007). Once these bonds are formed, hexamethylenediamine must be added to improve the physicochemical characteristics of the bio-adhesive in terms of strength and water resistance (Liu, Y. & Li, K., 2007). Therefore, the protocol of the bio-adhesive production starts with the mixing of the soy protein with water for 120 min and 60°C before the addition of maleic anhydride as described by Pizzi et al., (2020). Thereafter, hexamethylenediamine is added after raising the temperature to 90°C for a period of 120 min. Finally, with the aim of reducing the water content of the bio-adhesive to 50%, a roto-evaporation is performed at 60°C for 1 h (Xi et al., 2020). The inventory data for the impact assessment and the system boundaries considered for the production are shown in **Table SM5** and **Figure S1e** in the Supplementary Material, respectively.

2.3.6. Soy protein bio-adhesive with tannin-based resin (SP + TR bio-adhesive)

The production of this type of soy bio-adhesive requires two process steps. The first one is based on the production of the tannin resin, in which the tannin slurry is mixed with methanol, resorcinol, glyoxal and sodium hydroxide at 80 °C for 3 h, thus achieving the formation of a self-crossing structure (Zhou et al., 2013). Secondly, the soy protein, previously mixed with water and stirred for 20 min, to obtain a homogeneous slurry, is added to the tannin resin for another 10 min at room temperature (Chen et al., 2017).

The reaction of the tannins with the soy protein leads to the formation of bonds between the polypeptide chains, resulting in an increase in the mechanical properties and moisture resistance of wood boards made with the bio-adhesive (Ghahri et al., 2018). The inventory data for the impact assessment and the system boundaries considered for the production are shown in **Table SM6** and **Figure S1f** in the Supplementary Material, respectively.

2.4 Primary data source.

The inventory data of the background processes involved in the formulation of synthetic PF and UF based resins and the production of energy and chemicals required for the formulation of bio-adhesives have been taken from the Ecoinvent® database version 3.2. In the case of MUF resin, data from Silva et al. (2015) have been selected for the evaluation.

3. Results and discussion

3.1. The environmental profile of soy-based bio-adhesives

In the search for the selection of the best alternatives of soy-based bio-adhesives to replace synthetic ones in the manufacture of wood panels, a comparative study is required. First, the ReCipe 2016 hierarchist Midpoint method V1.03 World (2010) was used to determine the environmental profile of each bio-adhesive based on the impact categories selected for analysis. Although the burdens shown in **Table 1** allow for an overall environmental assessment of the proposed bio-adhesive alternatives, it is important to conduct a separate study for each in order to determine the contribution of the components that conform the production system to the overall environmental profile.<**Table 1** around here>

3.2. Environmental assessment of the SF + D bio-adhesive

The use of soybean meal is one of the main contributors to the environmental impact associated with the bio-adhesive **(Figure 2)**. This contribution is the result of the background processes associated with the soybean cultivation processes, specifically on in situ emissions from agricultural activities and the use of fertilizers in the cultivation stages. The second main contributor to the environmental profile of this bio-adhesive is the dicyandiamide production process, with a contribution share of more than 50% in half of the impact categories studied. On the other hand, this chemical compound has some potential to cause toxicity to aquatic species. Although it is a stable compound under normal conditions, its decomposition results in the release of carbon monoxide, carbon dioxide and nitrogen oxides, recognized as GHG emissions. Due to these possible effects on the environment, the possibility of replacing it with another crosslinking agent should be studied, or considering that this process is not optimized, a reduction of the dose used for the formulation of the bio-adhesive could be proposed.

We should be aware of another relevant aspect in the selection of a cross-linking agent. Beyond its contribution to the environmental profile, its potential adverse effect on human health must also be considered. When developing a new product, it is important not only to study the environmental impact of the process, but also whether the use of the product could pose a risk to human health and natural ecosystems in terms of toxicity. According to the EU classification, dicyandiamide is a compound that, despite not having carcinogenic or mutagenic effects, as was the case with formaldehyde, exposure could entail certain potential health effects such as skin irritation, formation of methemoglobin if absorbed by the body and gastrointestinal discomfort if inhaled. Although the concentration of dicyandiamide used for the formulation of the proposed bio-adhesive is low, which considerably reduces its possible adverse effects, the toxicity potential should be assessed in the weighting for the selection of the different bio-based alternatives.

< **Figure 2** over here>

3.3. *Environmental assessment of the SF + WP bio-adhesive*

Two main hotspots could be identified within this environmental profile (**Figure 3**): the soy flour and waterborne polyurethane as the cross-linking agent. This compound is obtained from non-renewable sources, specifically, produced by a petrochemical route (Yu et al., 2014), which leads to the emission of pollutants into the air, water and soil.

As an alternative to improve the environmental profile of this bio-adhesive, different recent works have been developed on the production of adipic acid, one of the compounds used for the formulation of WPU, through a biological synthesis (Raj et al., 2018; Sun et al., 2018; Kruyer et al., 2020), thus avoiding the use of the petrochemical route.

<**Figure 3** around here>

3.4. Environmental assessment of the SF + EP bio-adhesive

The environmental impact results (**Figure 4**) showed that the background activities related with the production of the soybean flour and the epoxy resin play a key role in the environmental profile in all the Midpoint categories considered for the assessment. As for the contribution derived from the use of chemicals, the only one that stands out in the environmental profile associated with the category of fossil resource scarcity associated with the production of epoxy resin. There is also some implication in the categories of ecotoxicity, eutrophication and acidification. Its release into the aquatic environment causes reproductive effects in aquatic organisms, and in terms of toxicity to soil and plants, the epoxy resin could be rapidly absorbed by the roots and metabolized into glycosidic compounds (Carlisle et al., 2009), causing potential harmful effects on seeds, leaves, stems, etc. (Plumlee, K.H., 2004).

< **Figure 4** over here>

3.5. Environmental assessment of the SP + EP bio-adhesive

Bearing in mind the results depicted in **Figure 5**, the environmental profile is dominated by the impacts derived from the background processes involved in the production of soybean meal. Therefore, the main cause of the contribution of the soy protein is the result of the combination of energy consumption associated to the extraction process and the background processes required in the agricultural cultivation. The reason for the environmental contribution of epoxy resin is analogous to the one mentioned in the previous section.

Although the contribution of polyacrylamide is not significant in the environmental profile of this bio-adhesive, its hazard classification should be taken into account. According to the EU classification labels, exposure to this substance can have a number of adverse effects due to its potential for carcinogenesis and mutagenesis, acute toxicity if ingested, inhaled or in contact with the eyes, and it is also suspected to contribute to impaired fertility. Due to the classification of this substance, although its content in the bio-adhesive formulation is relatively low, which reduces its possible risk to human health, it is an aspect that should be taken into account, as it can be a cause of exclusion in the selection of the best bio-adhesive alternative.

<**Figure 5** around here>

3.6. Environmental assessment of the SP + MA bio-adhesive

The distribution of burdens shown in **Figure 6** showed that HDMA (Hexamethylenediamine) production is the main hotspot in the global system profile, with a share of more than 40% in categories such as GW, TA, ME, TET and FRS. The reason for the environmental contribution of maleic anhydride is the result of the energy needs of its production based on fossil resources. Once again, as in the previous environmental profiles, the contribution of soy is also significant, mainly related to the extraction of the soy protein.

< **Figure 6** over here>

3.7. Environmental assessment of the SP + TR bio-adhesive

The results obtained for the environmental profile of the adhesive showed that the use of tannin-based resin is a good choice of cross-linking agent, since its environmental contribution on the entire system is not very significant (**Figure 7**). As for the previous scenarios, the contribution of the soy protein extraction is remarkable. Therefore, optimizing the protein extraction process, or even reducing the amount of soy protein used for the formulation, becomes a key aspect of improvement to achieve a bio-adhesive option with the potential to replace synthetic adhesives.

< **Figure 7** over here>

3.8. Benchmarking the soy-based bio-adhesives and fossil-based counterparts.

In order to develop a comparison with synthetic-based resins, the ReCipe 2016 hierarchist Endpoint V1.03 (2010) World H/H methodology has been applied (**Table 2**). The damage categories considered were human health (HH), ecosystem quality (EQ) and fossil resource scarcity (FRS). In this way, by applying the normalization and weighting values of each of the categories, a single environmental score is estimated, which allows for an overall value that includes three key elements to be considered for sustainable development.

< **Table 2** over here>

Considering soybean as a raw material, the soy-based bio-adhesives show better environmental profiles than those for UF and PF. Even if compared with MUF, which is the fossil resin with the best profile but the exception of the soy flour enhanced by the WPU-based adhesive, where a greater contribution is observed in the HH category compared to the value obtained by MUF (30 mPt versus 24 mPt, respectively), which affects the overall environmental score (36 mPt versus 30 mPt, respectively).

Finally, the method of calculating USEtox has been used to carry out a more comprehensive assessment of the human health category, in which two impact categories are developed: human toxicity, cancer (HT, c) and human toxicity, non-cancer

(HT, nc) (**Table 3**). The values obtained showed that bio-adhesive alternatives have a lower potential risk on human health, since their toxicity levels (both carcinogenic and non-carcinogenic) are significantly lower than those obtained for UF and PF resins, which are the most widely used in the field of wood panel production.

< **Table 3** over here>

3.9. Sensitivity analysis and alternative formulations of the bio-adhesives

Although the results obtained for the soy bio-adhesives are promising, their environmental profiles could be even better if the main critical points previously identified for each bio-adhesive were improved and optimized. In the case of adhesives that required the protein extraction stage, it was observed that the energy requirements of the process contributed significantly to the environmental profile, and therefore, by developing an energy optimization of the production process, they could be reduced by up to 25%. On the other hand, it has also been found that the use of certain chemicals as cross-linking agents has an important contribution to the environmental profile obtained, as has been reported in the case of the use of dicyandiamide, WPU, epoxy resin and, above all, hexamethylenediamine. Therefore, a sensitivity analysis has been carried out focusing on the reduction of the amount of chemical dosage, assuming that the reduction of the amount of cross-linking agent by 20% in the bio-adhesive formulation. The first case considered for study (**Figure 8a**) refers to the reduction of the amount of dicyandiamide used for the formulation of the SF+D bio-adhesive. The greatest environmental improvement is observed in the categories of TA, FE and FRS, where impact reductions of 13%, 14% and 12%, respectively, has been obtained. It should be noted that in the categories of GW, SOD and ME no significant changes are observed in comparison with the base case. The reason for this low level of improvement stems from the fact that the main contributor in these impact categories is not the use of dicyandiamide, but rather the background activities associated with the production of soybean.

The second object of study is based on the SF + WP bio-adhesive, in which it has been proposed to reduce the dose of WPU used for the formulation of the adhesive. The results obtained (**Figure 8b**) showed a significant improvement in the environmental profile, reaching reduction percentages of up to 20%. The only impact category where the improvement obtained is negligible in the ME. As in the previous analysis, the environmental contribution in this category of impact is the result of land management and agricultural activities associated with soybean cultivation.

In the case of reducing the dose of HDMA used for the production of the SP + MA bio-adhesive, improvement percentages of between 13% and 14% have been achieved for the impact categories of ME, TET and FRS (**Figure 8c**). The formulation of this adhesive requires the process of extracting the protein from the soybean, a stage that requires significant energy consumption, resulting in a high contribution to the environmental profile obtained for the adhesive. The fact that two hotspots of the bio-adhesive production process are identified means that, although an improvement in the environmental profile has been obtained by reducing the dosage of crosslinker used, the impact reduction values are not as high as desired. A joint reduction of the two main contributors to the system would lead to a more significant improvement in the profile.

The last alternative focused on reducing the amount of cross-linking agent used for the formulation is that of SF+SE bio-adhesive (**Figure 8d**). The use of epoxy resin for the formulation of the bio-adhesive implies the use of non-renewable fossil resources. Therefore, a reduction in the dose of this chemical used leads to a significant improvement in the impact category of SF+SE, reaching a percentage of reduction of 15%. However, in the SOD and ME categories, the percentages of impact reduction are less than 1%. The reason for this low value is that in these categories the process of extraction of the protein from the soybean, together with the background activities associated with its cultivation, are the main contributors.

The most significant improvements in the environmental profiles have been achieved by reducing the electrical requirements of the soy protein extraction process by 25% (**Figure 8e**). The highest percentages of improvement were obtained for the FE (22%), MET (21%) and FRS (20%) categories. The only impact categories in which the improvement has not been as high as expected are SOD and ME, in which the percentages of impact reduction were 3% and 2%, respectively. In these categories, the influence of the background activities of soybean cultivation is highly significant, reaching practically the entire impact generated.

< **Figure 8** over here>

Conclusions

In this study LCA methodology was used for the environmental assessment of different soy-based bio-adhesives as alternatives to fossil-based ones. Certain improvements in production processes can be identified that would considerably reduce the environmental impacts of the bio-adhesives production. The optimization of the protein extraction process, in terms of the use of energy resources, is considered a key aspect to be improved in those bio-adhesives that require this initial stage of the process.

In general, once the comparative studies have been carried out, it is considered that the alternatives of SF + D, SP + EP and SF + SE are the best from the environmental point of view, as they are the ones that have less impact on ecosystem quality, fossil resource scarcity, human health categories. With regard to the health effects that may be caused by dicyandiamide and polyacrylamide, toxicity data should be considered as a criterion for selecting the best soy-based bio-adhesive to replace synthetic resins. With this in mind, the SF+SE bio-adhesive is selected as the best alternative considering both environmental and toxicity perspectives.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

All data generated or analysed during this study are included in this published article [and its supplementary information files].

Competing interest

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

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Authors' contribution

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

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Figures

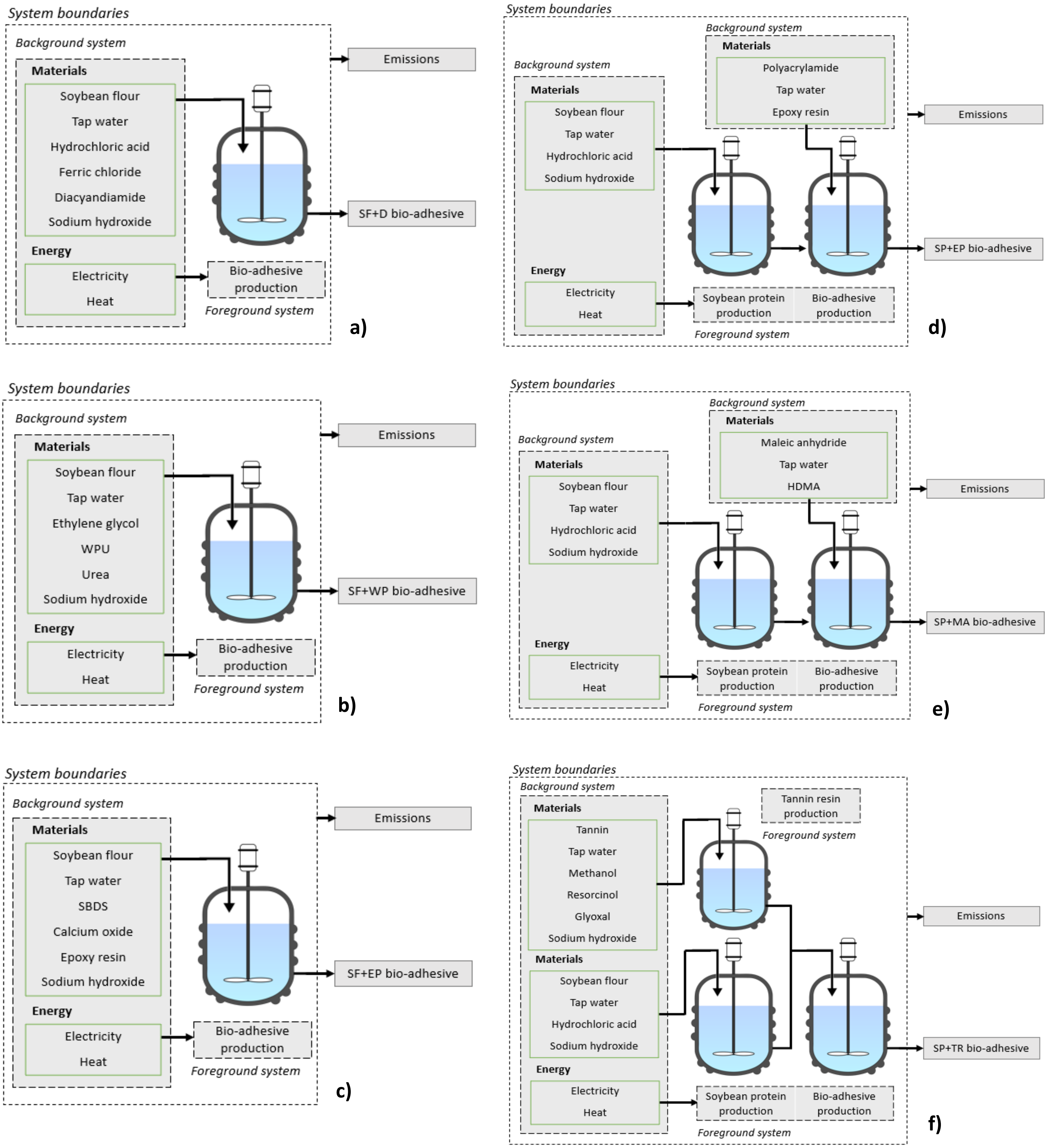

Figure 1. System boundaries of bio-adhesives production processes. SM1a) SF+D bio-adhesive; SM1b) SF+WP bio-adhesive; SM1c) SF+SE bio-adhesive; SM1d) SP+EP bio-adhesive; SM1e) SP+MA bio-adhesive and SM1f) SP+TR bio-adhesive.

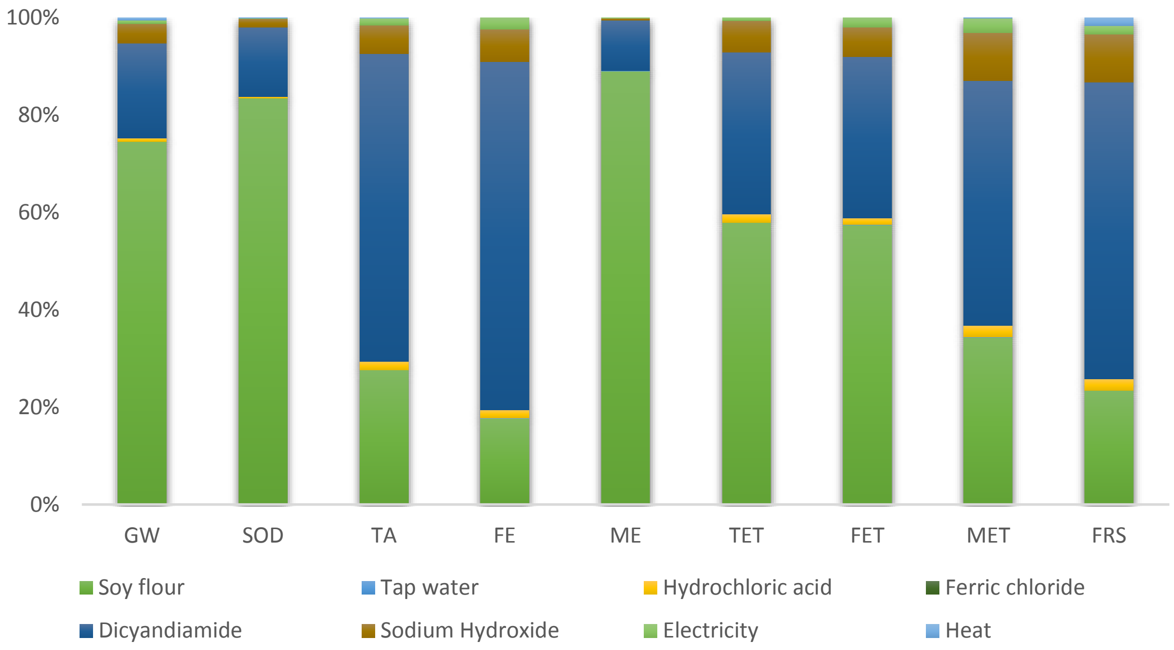

Figure 2. Impact assessment results per impact category for SF+D bio-adhesive production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

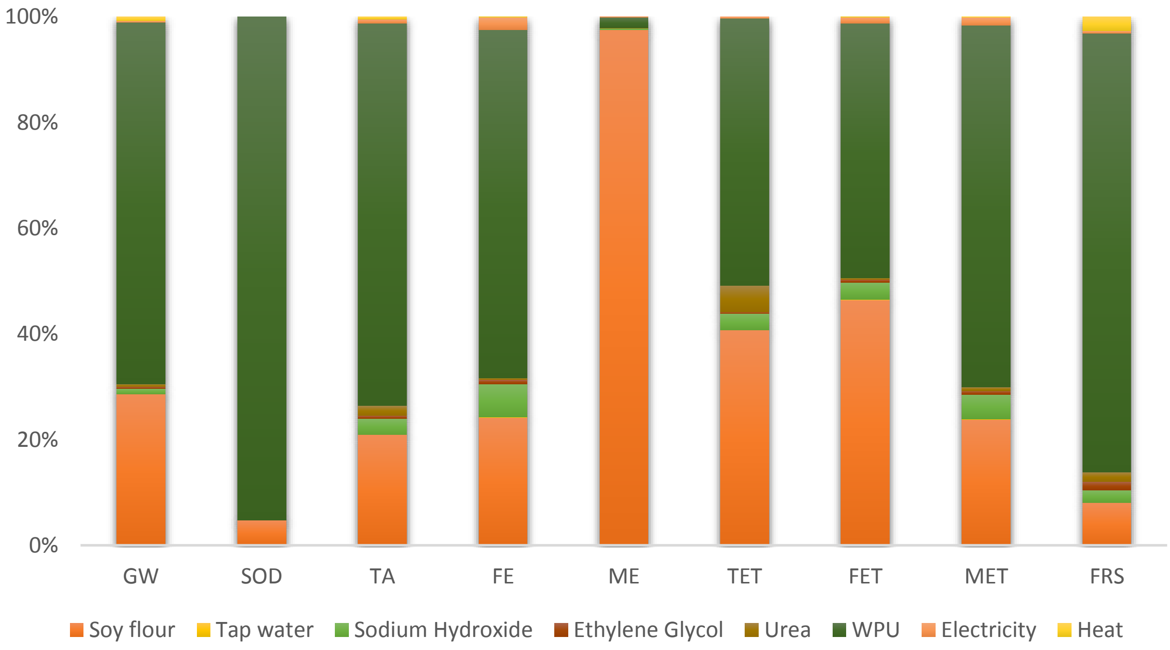

Figure 3. Impact assessment results per impact category for SF+WP bio-adhesive production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

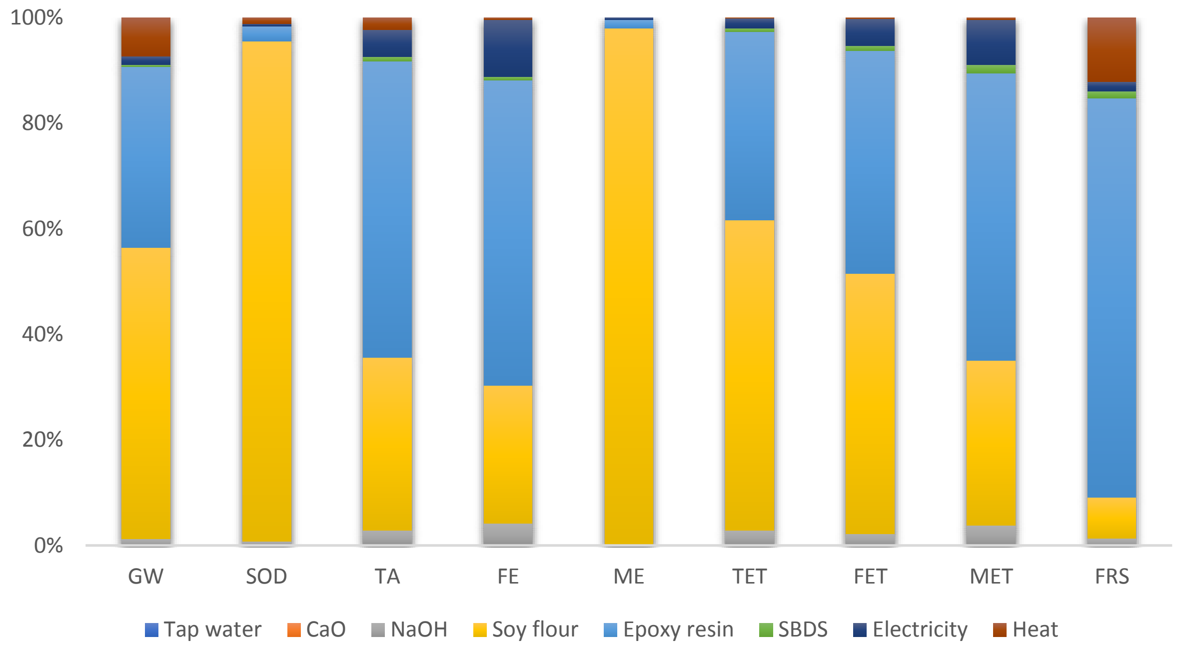

Figure 4. Impact assessment results per impact category for SF+SE bio-adhesive production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

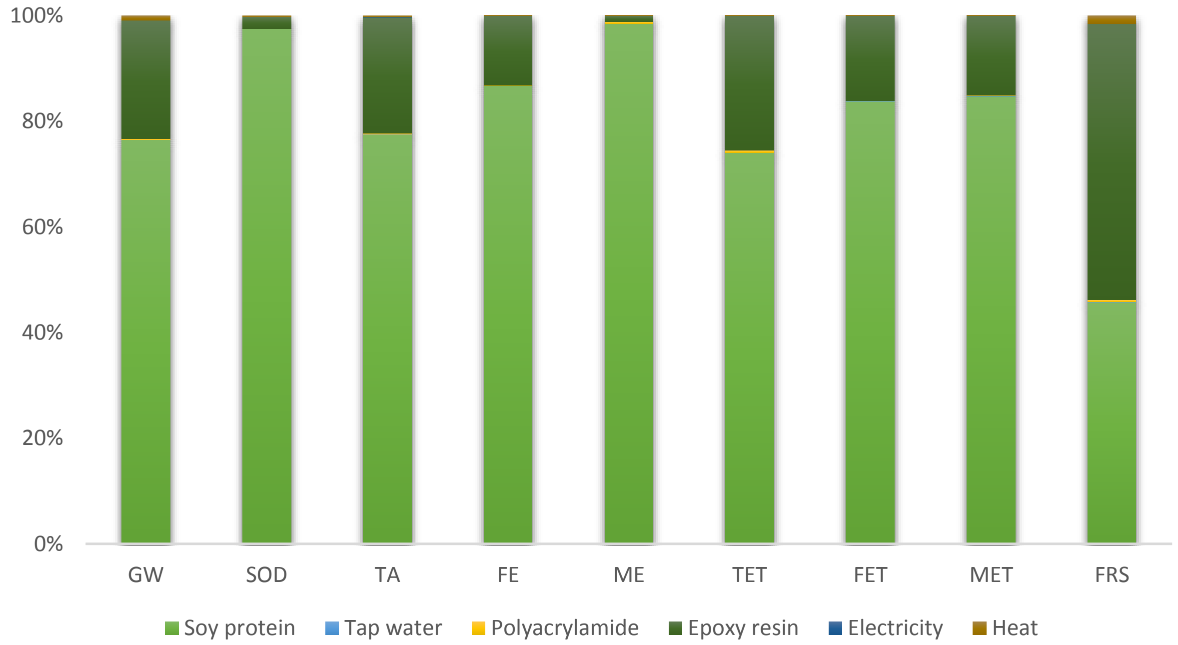

Figure 5. Impact assessment results per impact category for SP+EP bio-adhesive production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

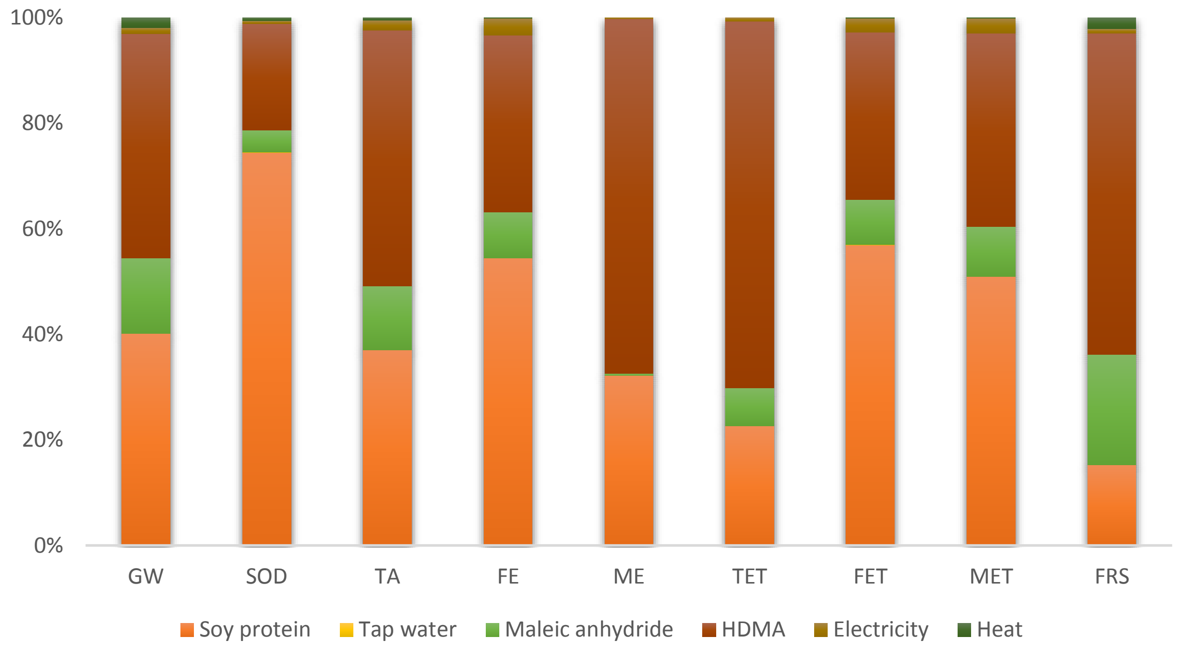

Figure 6. Impact assessment results per impact category for SP+MA bio-adhesive production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

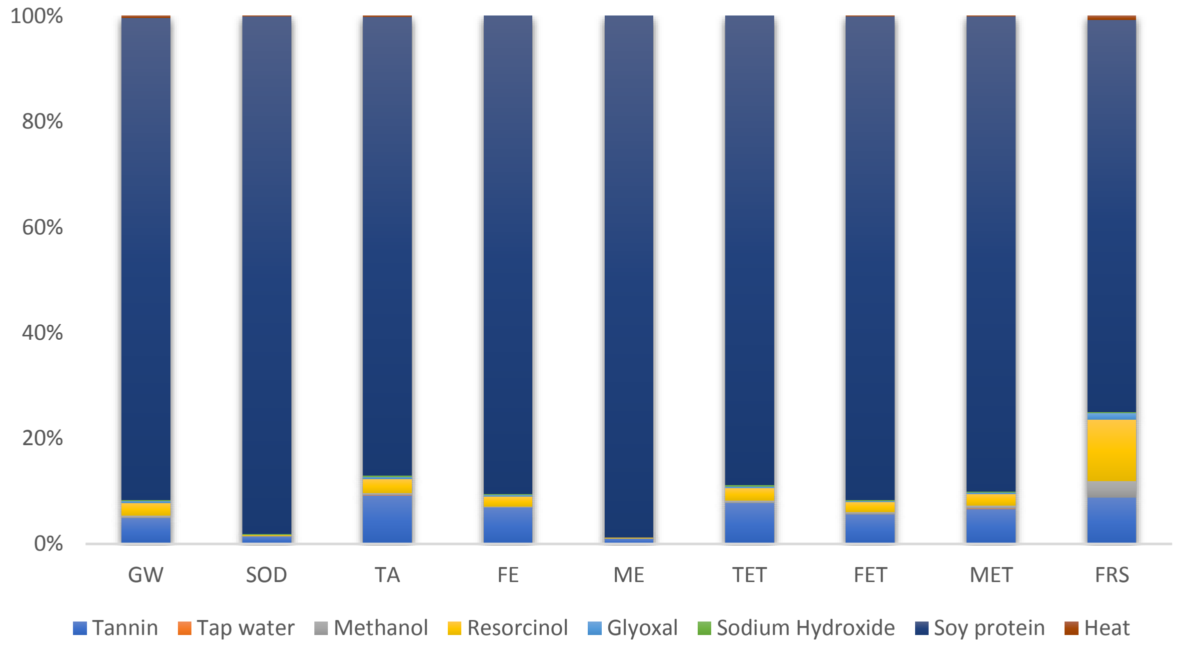

Figure 7. Impact assessment results per impact category for SP+TR bio-adhesive production. Acronyms: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS).

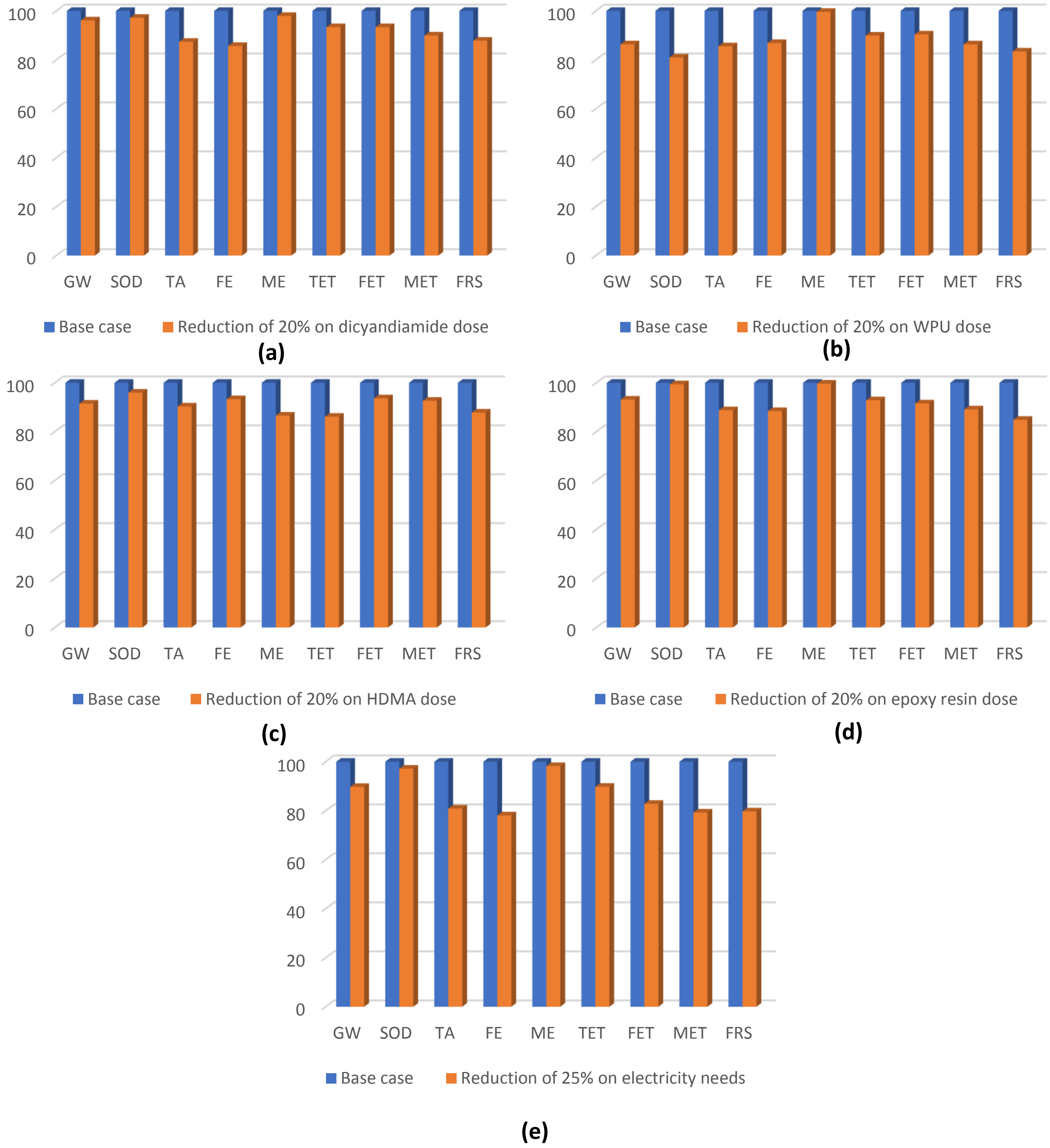

Figure 8. Sensitivity analysis of bio-adhesives production processes under study; a) Reduction of 20% on dicyandiamide dose on SF+D bio-adhesive b) Reduction of 20% on WPU dose on SF+WP bio-adhesive c) Reduction of 20% on HDMA dose on SP+MA bio-adhesive8d) Reduction of 20% on epoxy resin dose on SF+SE bio-adhesive e) Reduction of 25% on electricity needs on soy protein extraction process. 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).

Tables

Table 1. Impact assessment values obtained for the bio-adhesives per impact category. 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), fossil resource scarcity (FRS), soy flour with dicyandiamide crosslinker bio-adhesive (SF+D), soy protein crosslinked with epoxy resin and polyacrylamide bio-adhesive (SP+EP), soy flour enhanced by waterborne polyurethane bio-adhesive (SF+WP), soy protein with maleic anhydride bio-adhesive (SP+MA), soy flour crosslinked with SBDS and epoxy resin bio-adhesive (SF+SE) and soy protein with tannin-based resin bio-adhesive (SF+TR).

Impact Category	**Unit**	**SCENARIOS**					
		SF+ D	SP + EP	SF + WP	SP + MA	SF + SE	SP + TR
GW	kg CO_2 eq	0.61	0.71	2.56	2.14	1.20	0.67
SOD	mg CFC_{11} eq	1.62	1.08	46.07	2.23	2.07	1.22
TA	g SO_2 eq	1.60	1.94	3.41	6.39	1.96	1.95
FE	g P eq	0.19	0.30	0.22	0.76	0.19	0.33
ME	g N eq	0.33	0.23	0.49	1.12	0.43	0.26
TET	kg 1,4-DCB	0.44	0.43	1.00	2.19	0.62	0.40
FET	g 1,4-DCB	6.14	10.42	12.24	24.15	10.35	10.77
MET	g 1,4-DCB	5.30	11.85	12.33	31.06	8.46	12.61
FRS	kg oil eq	0.06	0.15	0.30	0.73	0.28	0.11

Table 2. Single score values obtained for the bio-adhesives per damage category, and its comparison with synthetic resins. Acronyms: Human Health (HH), Ecosystem Quality (EQ), Fossil Resource Scarcity (FRS), soy flour with dicyandiamide crosslinker bio-adhesive (SF+D), soy protein crosslinked with epoxy resin and polyacrylamide bio-adhesive (SP+EP), soy flour enhanced by waterborne polyurethane bio-adhesive (SF+WP), soy protein with maleic anhydride bio-adhesive (SP+MA), soy flour crosslinked with SBDS and epoxy resin bio-adhesive (SF+SE) and soy protein with tannin-based resin bio-adhesive (SF+TR).

Damage Category	**Unit**	**SCENARIOS**					
		SF+ D	SP + EP	SF + WP	SP + MA	SF + SE	SP + TR
HH	mPt	7.16	8.34	30	25	14	7.89
EQ	mPt	1.28	1.55	4.68	4.65	2.31	1.49
FRS	mPt	0.23	0.45	1.00	2.65	1.04	0.25
TOTAL		**8.67**	**10**	**36**	**32**	**17**	**9.64**
Damage Category	**Unit**	**SYNTHETIC RESINS**					
		UF		MUF		PF	
HH	mPt	31		24		41	
EQ	mPt	6		5		7	
FRS	mPt	5		3		7	
TOTAL		**41**		**33**		**56**	

Table 3. Impact assessment values obtained for the bio-adhesives per impact category to evaluate the human impacts caused, referring to the environmental indicator of Human Toxicity (unit values: CTUh) and its comparison with synthetic resins. Acronyms: HT,c (Human Toxicity, cancer), HT,nc (Human Toxicity, non-cancer), soy flour with dicyandiamide crosslinker bio-adhesive (SF+D), soy protein crosslinked with epoxy resin and polyacrylamide bio-adhesive (SP+EP), soy flour enhanced by waterborne polyurethane bio-adhesive (SF+WP), soy protein with maleic anhydride bio-adhesive (SP+MA), soy flour crosslinked with SBDS and epoxy resin bio-adhesive (SF+SE) and soy protein with tannin-based resin bio-adhesive (SF+TR).

Impact Category	Unit	**SOYBEAN AS RAW MATERIAL**					
		SF+ D	SP + EP	SF + WP	SP + MA	SF + SE	SP + TR
HT,c	$\cdot 10^{-10}$ CTUh	3.55	2.46	5.79	36	5.49	2.30
HT,nc		4.41	2.76	5.60	3.87	6.29	2.22
Total		**7.97**	**5.22**	**11**	**40**	**12**	**4.52**

Impact Category	Unit	**SYNTHETIC RESINS**		
		UF	PF	MUF
HT,c	$\cdot 10^{-10}$ CTUh	259	92	1.91
HT,nc		5.38	12	1.77
Total		**264**	**104**	**3.68**

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