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Environmental assessment of green hardboard production coupled with a laccase activated system

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ABSTRACT

European consumption of wood-based panels reached record levels in recent years driven mostly by demand from end-use sectors: residential construction, furniture, cabinets, flooring and mouldings. The main panel types are composite boards such as particleboard, high density fiberboard (HDF), medium-density fiberboard (MDF) and other adhesively bonded composites such as plywood and wet-process fiberboard (hardboard). The synthetic resins used in their manufacture come from non-renewable resources, such as oil and gas. Several consequences are associated to this type of adhesives: variation in the availability and cost of these wood adhesives depends on raw materials, the formaldehyde emissions as well as the limited recyclability of the final product. Hence, in the search for alternatives to petroleum-based wood adhesives, efforts are being devoted to develop adhesives by using phenolic substitutes based on lignin, tannin or starch. In this context, the forest industry is increasingly approaching to enzyme technology in the search of solutions. The main goal of this study was to assess the environmental impacts during the life cycle of a new process for the manufacture of hardboards manufacture, considering the use of a two-component bio-adhesive formulated with a wood-based phenolic material and a phenol-oxidizing enzyme. This new product was compared to the one manufactured with the conventional phenol-formaldehyde resin. The study covers the life cycle of green hardboards production from a cradle-to-gate perspective, analysing in detail the hardboard plant and dividing the process chain in three subsystems: Fibers Preparation, Board Forming and Board Finishing.

Auxiliary activities such as chemicals, bio-adhesive, wood chips, thermal energy and electricity production and transport were included within the system boundaries.

Global warming (GW), photochemical oxidant formation (PO), acidification (AC) and eutrophication (EP) were the impact categories analysed in this study. Additionally, the cumulative energy demand was evaluated as another impact category. According to the results, four stages significantly influenced the environmental burdens of the production system: laccase production, on-site thermal energy and electricity production as well as wood chipping stage. Due to the environmental impact associated to the production of green bonding agents, a sensitivity analysis with special focus on the eutrophying emissions was carried out by evaluating the amount of laccase and lignin based phenolic material used. The combined reduction in both bonding agents may slightly reduce the contributions to this impact category. In addition, a hypothetical scenario with no laccase and with a higher concentration of the lignin based material (25% more) could improve the environmental profile in all impact categories with a reduction of 1.5% in EP.

Further research should focus mainly on laccase production, in order to reduce its energy demand as well as on the amount of green adhesive required to obtain mechanical and swelling properties similar to those of conventional hardboard.

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1. Background, aim, and scope

The European Union (EU) is one of the largest producers, traders and consumers of forest products worldwide (European Commission, 2010). Forest-based industries and other related industries make up

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one of the most important and dynamic industrial sectors in the EU, representing up to 10% of the total European manufacturing industries. In 2005, these industries employed roughly 3 million people in 350,000 companies with an economic turnover of about 380 billion €, producing an added value of around 116 billion € (European Commission, 2010).

The forest sector (forestry, forest-based and related industries) comprises the following industrial sectors: i) woodworking; ii) cork and other forest-based materials; iii) pulp, paper and paper-board manufacturing; iv) paper and paper-board converting and; v) printing industries. Specifically, wood processing involves the conversion of trees into useful consumer products and/or building materials such as wood boards. The woodworking industries supply basic products such as sawn goods, wood panels and builders' carpentry for construction, internal decoration and packaging (pallets) (European Commission, 2010). In general, the European consumption of panels reached record levels in 2006 (~ 64.7 million m^3), driven mainly by demand from end-use sectors: residential construction, furniture, cabinets, flooring and mouldings (UNECE, 2006/FAO).

Wood panels are characterized by their variable physical and mechanical properties. The main panel types in Europe are composite boards such as particleboard, high density fiberboard (HDF) and medium-density fiberboard (MDF), and other adhesively bonded composites such as plywood and wet-process fiberboard (hardboard). Fiberboard is an engineered product made from compressed wood or non-wood lignocellulosic fibers. Because of their high resistance and strength, fiberboards can be used as a raw material for laminate flooring, exterior siding and trim, garage doors, furniture, wall panelling, interior trim and perforated boards. Nowadays, the consumer market is conscious of the environmental problems derived from the industrial sector. Important amounts of petroleum based adhesives (such as urea or phenol formaldehyde) are required for the manufacture of panels. Therefore, formaldehyde emissions during production and end-use are a relevant consequence with negative environmental impacts on ecosystem quality (Imam et al., 1999; US. EPA, 2002). Therefore, special attention is focused on the reduction of this type of adhesives as well as on their replacement by more environmentally-friendly, natural and safer alternatives such as lignin based materials (Moubarik et al., 2009): lignosulfonates (a lignin co-product of sulfite pulping), organosolved lignin, kraft lignin, flavonoid-based tannins from certain trees (Widsten et al., 2009), starch from renewable sources or glues derived from animal tissues casein (Imam et al., 1999). In this context, non-conventional processes based on the treatment of lignin with enzymes have been investigated for fiberboards production at laboratory and pilot scale (Widsten and Kandelbauer, 2008). Moubarik et al. (2009) demonstrated the performance of cornstarch–quebracho tannin-based resins used as adhesives in the plywood production to partially substitute phenol-formaldehyde resin (PF). The new plywood panels showed better mechanical properties and water resistance when compared to conventional PF panels, as well as lower formaldehyde emissions. Moreover, starch has been used as a wood adhesive not only for interior but also for external applications with other polymers such as PF and urea-formaldehyde (UF) (Imam et al., 1999).

Life Cycle Assessment (LCA) has proved to be a valuable methodology for evaluating the environmental impacts of products and service systems, and should be part of the decision-making process toward sustainability (Baumann and Tillman, 2004). Several studies have focused on the production of conventional wood-based products such as load boxes (Echevenguá Teixeira et al., 2010), wood floor coverings (Nebel et al., 2006; Petersen and Solberg, 2003), particleboards (Rivela et al., 2006), MDF (Rivela et al., 2007), hardboards (González-García et al., 2009a) and related wood items such as window frames (Asif et al., 2002; Richter and Gugerli, 1996), walls (Werner, 2001) and furniture (Taylor and van Langenberg, 2003). Furthermore, wood products tend to have a more favourable environmental profile compared to functionally equivalent products obtained from other materials such as plastics, aluminium or steel (Werner and Richter, 2007). To date, LCA studies for green production of boards are not available. The objective of this paper is to environmentally analyse the industrial process of green hardboard manufacture considering the substitution of the phenol-formaldehyde resin by a two-component adhesive with a wood-based phenolic material and a phenol-oxidizing enzyme (i.e. laccase). Additionally, this new product will be compared to the one manufactured with the conventional PF resin used as the main bonding agent (González-García et al., 2009a).

2. Goal and scope definition

2.1. Objectives

This study aimed at analysing the manufacture of green hardboard from an LCA perspective in order to detect the environmental 'hot spots' throughout the production life cycle. Furthermore, a comparison of these environmental results with the conventional production of hardboard was also discussed. An Austrian hardboard plant, which has implemented the biotechnological process of green hardboards production, was selected to study the process in detail. The study covers the whole life cycle of green hardboard manufacture from raw material production to plant gate.

2.2. Functional unit

The functional unit provides a reference point for inputs and outputs (ISO 14040, 2006). In this paper, it is defined as 1 m^3 of finished green hardboard (for interior applications). The board density is approximately 900 kg m^{-3} and its moisture content ~7%.

2.3. Description of the system under study

Conventional hardboards (HB) are composite panel products consisting of lignocellulosic fibers manufactured under heat and pressure in a wet process with a small dose of phenolic binder (Widsten et al., 2009). Additives such as paraffin wax can be used to improve certain characteristics such as abrasion and moisture resistance. A panel of this kind has homogeneous thickness, density, appearance and no grain. Phenol-formaldehyde (PF) resin is commonly used in the production of these panels but the presence of synthetic resins limits the recycling and final disposal of used hardboards (Smith, 2004). An alternative process is the substitution of these synthetic resins by bio-based phenolic materials in combination with phenol-oxidizing enzymes. This process has been named as a "green strategy". Examples of bio-based phenolic materials could be lignosulfonates (co-product from dissolving pulp mills) or flavonoid-based tannins (chestnut tannin, tara tannin, mimosa tannin or quebracho tannin). The main phenol-oxidizing enzymes is laccase, which is already commercially produced by a genetically modified fungus in submerged fermentation. In the presence of oxygen, laccase catalyzes the oxidation of phenolic substrates (e.g. certain lignin phenylpropane units) to phenoxy radicals.

The studied HB plant uses a smooth-one-side type production process, which renders good natural fiber to fiber interfelting and bonding with minimum added binder required and provides a moist surface of high plasticity giving the desired embossing sensitivity. The hardboards were produced according to a large

Table 1
Main features of green HB.

Nominal width	1220–1524 m
Moisture content	2–9%
Board thickness	3.0–3.6 mm
MoR	38–60 N mm^{-2}
MoE	3200–3900 N mm^{-2}
Internal bond (IB)[a]	0.8–2.2 N mm^{-2}

[a] with a 24 h water swelling (20 °C) of 29–57%.

range of properties. The boards are produced with a two-component adhesive based on laccase activated tannin system instead of PF resin. The main features of green HB vary according to the brand but standard quality boards are shown in Table 1.

According to our previous LCA study of conventional HB production (González-García et al., 2009a), the process chain was divided into three main subsystems: Fibers Preparation, Board Forming and Board Finishing. Auxiliary activities included chemicals, laccase, lignin based phenolic material, thermal energy and electricity production, transport activities and wood chips production. Concerning the production of the lignin based phenolic component, several materials can be used. The innovative nature of this process gives rise to some problems when collecting data for an LCA study, since it is difficult to find quality data, as well as detailed descriptions of the production processes of these alternative materials. Fortunately, the production of lignosulfonates has been reported in literature (Widsten et al., 2009). Therefore, the use of lignosulfonates derived from dissolving pulping as a bio-based phenolic component was considered and included within the system boundaries. The system investigated is illustrated in Fig. 1.

2.3.1. Subsystem of fibers preparation

The main raw materials are green wood chips obtained from Norway spruce (*Picea abies*) and European beech (*Fagus sylvatica*). This material is delivered by truck from Austrian wood-based industries such as sawmills, satellite chip mills, etc. Initially, wood chips are washed to remove dirt and other debris. Clean chips and additional raw material from the plant (sanding dust, sawdust, trimmings, rejected hardboard etc.) are pre-heated to soften lignin and enable subsequent fiber separation. The following step consists of their reduction into fibers, followed by an enzymatic treatment and placement in a storage bin. The enzymatic treatment is carried out by immersing the fibers in the aqueous laccase containing solution for a 20 min incubation time. Enzyme dose may be reduced by applying longer incubation time and/or recirculation. Spectrophotometric assays of laccase activity were carried out with 10 mM 2,2′-azino-bis-(3-ethylbenzthiazolinesulphonate) (ABTS) as substrate in 100 mM sodium acetate buffer (pH 5.0). The absorbance was monitored at 436 nm (extinction coefficient = 29,300 M^{-1} cm^{-1}). One U of enzyme activity is defined as the amount of enzyme releasing 1 μmol min^{-1} oxidized product at 25 °C. Regarding the dose of the green bonding agent, 10.5 kg of laccase and 40 kg of lignin based phenolic material are used in the green HB production process, instead of 34 kg of PF used in the conventional process. A fraction of the wood material is burnt in biomass boilers to produce thermal energy for plant activities.

2.3.2. Subsystem of board forming

Fibers with the two-component adhesive are transported from the storage bin to the forming machine where they are placed onto a moving conveyor belt to form a mat. The mat is pre-pressed and trimmed before being loaded into the hot press. This press applies heat and pressure to cure the adhesive and bond the fibers for 4 min. The press uses a multi-opening batch system and is heated (~170–180 °C, above the normal industrial level) by the steam produced at the thermal energy plant.

2.3.3. Subsystem of board finishing

After the pressing process, the boards are placed in a conditioning room and then sanded and sawed into final size. Finally, the boards are packaged and sent to the warehouse. Trimming residues are recycled back to board production or used for on-site energy production.

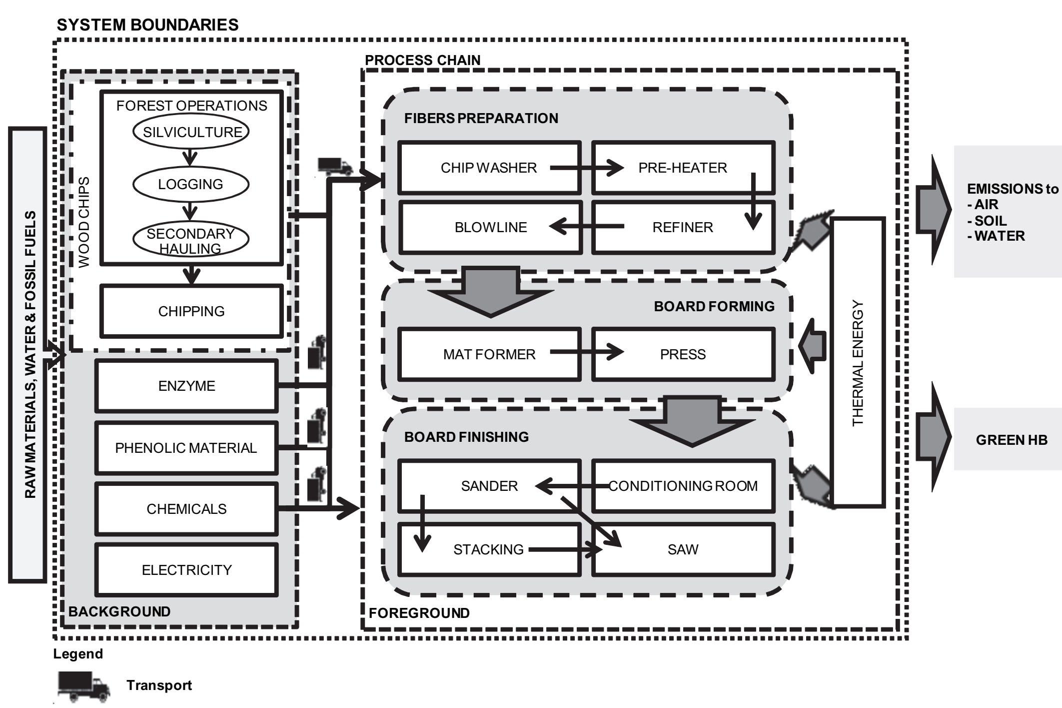

Fig. 1. System boundaries and process chain under study.

2.3.4. Ancillary activities

Several auxiliary activities were included within the system boundaries. Laccase, paraffin emulsion and other chemicals were part of the Fibers Preparation subsystem. In addition, their transport by truck from wholesaler to plant gate (roughly 300 km) was included in the study.

Softwood chips production (the main raw material) was included, considering from softwood plantation to roundwood chipping and delivery to fiberboard plant (Fig. 1). Although not specifically present in the figure, both silviculture, logging operations and transport of roundwood to sawmill (100 km by truck) were considered. All the wood produced in the plantations is assumed to be used in chips manufacture since this kind of wood can be considered as raw material for other forest-based industries (e.g. paper pulp production). Moreover, transport of workers, machinery and materials (fertilizers, pesticides and fuels) to and from forest plantations were also included. Seedling production was excluded due to lack of data. A more detailed description of these activities can be found in González-García et al. (2009b). Roundwood is then processed into green chips, which will be delivered to the plant by truck (an estimated distance of 100 km).

Austrian electricity generation was taken into account with the following profile: 79.7% from hydroelectric plants, 20.2% from fossil fuels, mainly hard coal and lignite, and 0.1% from renewable resources. Finished HB delivery was not included within the system boundaries because its production is carried out at pilot scale at present and green HBs are mainly used for R&D activities.

Finally, all the thermal energy consumed in the HB manufacturing process (steam, hot oil and hot gas) is obtained from the biomass and is produced at the plant as described above. Wood waste from wood-based mills is used as fuel and its transport to plant gate (100 km) was included within the system boundaries.

2.4. Data quality and simplifications

Inventory data for the foreground system (green HB manufacturing process) consisted of average data obtained by on-site measurements. When possible, typical process-specific data were collected to avoid anomalous conditions.

The key emission sources identified were dryers, presses, mat formers, biomass boilers and finishing operations (sanding and sawing). All the emission data corresponded to field data. Other inventory data for the background system such as electricity, paraffin and aluminium sulphate production were obtained from the Ecoinvent database (Althaus et al., 2007; Dones et al., 2007). Regarding chemicals and green chips requirements and wood waste transportation routes were supplied by plant workers, while emission factors were obtained from the Ecoinvent database (Spielmann et al., 2007). Inventory data for forest operations related to softwood production were taken from González-García et al. (2009b), where softwood plantations were assessed. In addition, inventory data for the wood chipping stage were taken from the literature (ETH-ESU 96, 2004). With regard to laccase production, life cycle data for a representative industrial process were found in Nielsen et al. (2007).

Inventory data for the lignin based phenolic material production were taken from González-García et al. (Submitted for publication). In this study, an LCA analysis was performed in order to analyse the environmental burdens in a Swedish biorefinery where dissolving cellulose is produced as the main product, together with ethanol and lignosulfonates as co-products. In addition, extra steam is produced from the biorefinery waste which avoids fossil fuel consumption. A summarized inventory table for the lignin based phenolic material considered is shown in Table 2.

Table 2
Global inventory data per 1 kg of lignin based phenolic material.

INPUTS from TECHNOSPHERE			
Materials		Energy	
Biomass		Electricity from grid	40.2 kWh
Green logs (50% moisture)	0.26 m^3	Steam from biomass[a]	208.4 kWh
Chemicals		*Transport*	
Hydrogen Peroxide	2.7 kg	20-28 t truck	0.77 t km
Sodium hydroxide	4.5 kg	16 t truck	0.07 t km
Sulfurous acid	0.14 kg		
Chelant	0.10 kg		
OUTPUTS			
To TECHNOSPHERE		To ENVIRONMENT	
Materials		Emissions to air	
Lignosulfonate	1 kg	CO_2 biogenic	0.11 kg
Ethanol	2.5 kg	CO_2 fossil	4.2 kg
Dissolving pulp	42.0 kg	*Emissions to water*	
		COD	1.8 kg
		BOD_5	0.40 kg

In contrast to the conventional process where fiber drying and mat forming contribute to the environmental impact due to the uncontrolled formaldehyde emissions derived from the PF adhesive, the use of this green adhesive avoids these harmful emissions.

The production and maintenance of capital goods in both conventional and green HB systems were excluded from the analysis, as environmental data were assumed to be comparable in both scenarios (González-García et al., 2009a). Additionally, several industrial LCA studies have shown that the environmental load from the production of capital goods is insignificant when compared to their operation stage (Rivela et al., 2006,2007). Hence, the exclusion of such processes is also justified. Table 3 summarises the data sources handled in this paper.

2.5. Allocation procedure

An important feature of the wood-based industry is the simultaneous production of several products. For the panel industry, the main product is the panel, while residual wood is obtained as a by-product. An allocation procedure is only necessary for the panels, since the residual wood is used for on-site generation of thermal energy. Forest waste was taken into account to complete the energy balance of the biomass plant. No environmental burden allocation was assumed to forest waste from previous processes and only their transport and later processing were computed.

3. Environmental impact assessment

A retrospective LCA for green HB manufacture was carried out according to the CML 2 baseline 2000 V2.1 biogenic method to quantify the environmental impact (Guinée et al., 2001). This method results in the definition of an environmental profile for the

Table 3
Summary of data sources.

Energy	Electricity	Ecoinvent database (Dones et al., 2007)
Transport	Truck 16 t and 40 t	Ecoinvent database (Spielmann et al., 2007)
Chemicals	Paraffin and aluminium sulphate	Ecoinvent database (Althaus et al., 2007)
Enzyme	Laccase	Nielsen et al. (2007)
Phenolic compound	Lignin based material	González-García et al. (Submitted for publication)
Raw materials	Wood chips	ETH-ESU 96 database (ETH-ESU 96, 2004); González-García et al. (2009a)

Table 4
Impact assessment results (Characterization step) of green hardboard manufacture for 1 m^3 of finished green hardboard.

Impact category	Unit	Value
Global Warming (GW)	kg $CO_{2\ eq}$	346.8
Photochemical Oxidation (PO)	kg $C_2H_{2\ eq}$	0.126
Acidification (AC)	kg $SO_{2\ eq}$	3.93
Eutrophication (EP)	kg $PO_4^{-3}{}_{eq}$	0.849
Cumulative Energy Demand (CED)	MJ low heat value	6233.4

assessed production by quantifying the environmental effects on different categories, while only indirect or intermediate effects on humans can be assessed. The impact categories analysed in this study were: global warming (GW), photochemical oxidants formation (PO), acidification (AC) and eutrophication (EP). In addition, the application of the cumulative energy demand method (CED) was also included, in order to evaluate this indicator (VDI-Richtlinien, 1997) as another impact category (in terms of MJ equivalent) as the production of enzymes is likely to be energy intensive (Nielsen et al., 2007). The CED states the entire demand, valued as primary energy, which arises in connection with production, use and disposal of an economic good. LCA software SimaPro 7.10 developed by PRé Consultants (PRé Consultants, 2008) was used for the impact assessment. The results for the characterisation step are shown in Table 4.

Fig. 2 shows the relative contributions of the green HB production process to each impact category under study. The Fibers Preparation subsystem presented the highest contribution (more than 54%) to all categories, followed by Board Forming and Board Finishing. This result was mainly due to the higher electricity consumption compared to the remaining subsystems and also to the laccase requirement. Fig. 3 shows a more detailed study with the relative contribution of the main processes to each impact category.

3.1. Global warming potential

The Fibers Preparation subsystem was responsible for most of the GW contributions (83%). Electricity and laccase production were responsible for 39% and 31% of total CO_2 equivalent emissions (Fig. 3). Fossil fuel consumption (mainly natural gas and hard coal) for electricity production accounted for more than 39% of the total contribution followed by the diesel requirement in wood chipping stage (17%). It is important to highlight that the combustion of biomass (wood waste) in biomass boilers to produce thermal energy requirements, gave rise to biogenic CO_2 emissions. This

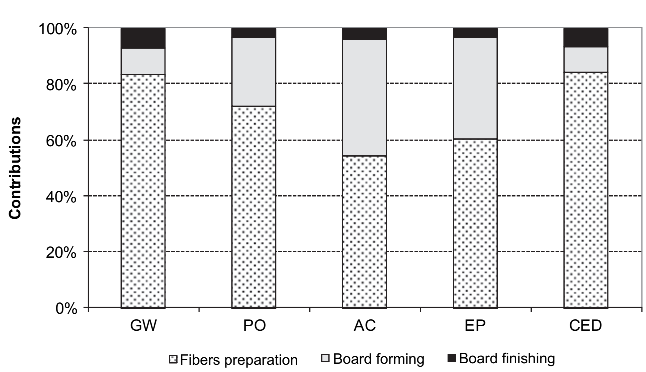

Fig. 2. Relative contributions per subsystems (in %) to each impact category. Impact category acronyms: GW = Global Warming, PO = Photo-Oxidant formation, AC = Acidification, EP = Eutrophication and CED = Cumulative Energy Demand.

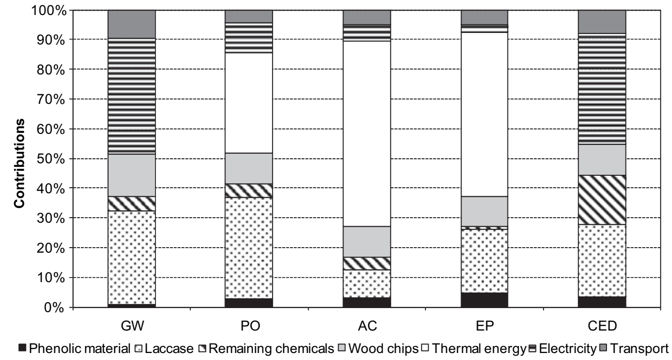

Fig. 3. Relative contributions per processes (in %) to each impact category. "Wood chips" includes not only the roundwood chipping step but also, all forest activities focused on roundwood production.

amount of CO_2 could be considered equal to that uptaken by photosynthesis during the biomass growth. Consequently, biomass burning is CO_2-neutral but not CO_2-free. Fossil CO_2 emissions gave the greatest contribution (~97%) to this impact category, followed by N_2O and CH_4.

3.2. Photochemical oxidants formation potential

The subsystem of Fibers Preparation had the largest contribution to the potential impact of photochemical oxidant formation (PO): 72%. Board forming and board finishing are responsible for 24% and 3%, respectively (Fig. 2). Both laccase and on-site thermal energy production showed the highest contributions to this impact category (34% each). The PO of the system studied was mainly caused by SO_2 and CO emissions which were strongly related to energy use.

3.3. Acidification potential

The Fibers Preparation and Board Forming subsystems were the most important contributors to AC: 54% and 42%, respectively, followed by the subsystem of Board Finishing (Fig. 2). This result agrees with those of related studies (González-García et al., 2009a; Rivela et al., 2006,2007). Specifically, on-site thermal energy production was the main hot spot (63% of total contributions) due to the emissions of NO_x and SO_2 from the biomass boilers. It is important to mention the contributing emissions from fossil fuel combustion during green chips production as well as the production of energy requirements in the laccase production process (Fig. 3).

Lignin based phenolic material production shows a small contribution (~3%) caused by the cogeneration unit where black liquor and fuel oil (used in the start-up of the boilers) are burnt to recover the cooking chemicals and to fulfill the steam requirements in the biorefinery.

3.4. Eutrophication potential

Once again, the Fibers Preparation subsystem had the largest contribution to this impact category (~61%) followed by Board Forming (36%) and Board Finishing (3%) (Fig. 2). The thermal energy plant was the main contributor to this impact category (55% of total) according to Fig. 3, followed by laccase production (22%) and chipping stage (10%). The production of the phenolic compound contributed to 5% of total eutrophying emissions mainly due to COD emissions derived from the wastewater treatment plant in the biorefinery. Airborne NO_x emissions showed the greatest contribution to EP (~95%), followed by those of NH_4^+ and COD (~3%). Specifically, it is important to point out the NO_x emissions from the biomass energy converters and electricity production (Fig. 3).

3.5. Cumulative energy demand

According to the results, the Fibers Preparation subsystem shows the largest contribution to the environmental profile (~84%). If it is analysed in detail, electricity production was the main hot spot in terms of MJ (low heat value) equivalent (37%) followed by the laccase production (24%) and the production of the remaining chemicals (17%). Once again, phenolic material production shows a small contribution to this indicator (3% of total). Specifically, it is interesting to remark two aspects: 1) the contribution derived from fossil fuel consumption (non-renewable energy) in processes such as chemicals and electricity production and 2) the production of a green adhesive (laccase and lignin based material) requires up to 58% less energy in comparison with the production of PF.

4. Discussion

In this study, the green HB production process was analysed in detail with the purpose of identifying the environmental burdens and hot spots of a wood-based product: green HB. This production process is an example of the industrial use of the enzyme laccase for the production of green adhesives in substitution of conventional petrochemical adhesives. According to Fig. 3, four stages considerably influenced the environmental burdens of the production system: laccase production, on-site thermal energy production from wood waste, electricity production and finally, to a lesser extent, the wood chipping stage. Previous studies on conventional fiberboards manufacture (MDF, particleboards and HB) have also identified that some of these processes such as electricity and chemicals production are the main contributors to the environmental impact (González-García et al., 2009a; Rivela et al., 2006,2007).

The HB plant presented an important use of renewable energy since all heat requirements (steam for defibrator, hot oil for pressing and hot gas for drying) were fulfilled by on-site biomass waste burning. Approximately 98% of the energy required was obtained from internal recycling (e.g. rejected HBs, sanding dust, sawdust or trimmings) and barely 2% from external biomass (wood waste from other factories and forest operations). For this reason, the biomass boilers showed high contribution in impact categories such as AC, EP and PO due to the emission of NO_x and SO_2 (Jawjit et al., 2007; González-García et al., 2009c).

In contrast with other studies on conventional fiberboards production where petrochemical resins (UF and PF) are used as adhesives, a two-component adhesive based on laccase activated lignin was considered instead of PF resin. Laccase acts as a catalyst for the oxidation of phenolic hydroxyls to phenoxy radicals by molecular oxygen, which is reduced to water (Widsten et al., 2004). If phenoxy radicals located on different fibers are brought into close contact, the formation of covalent interfiber bonds could occur by radical coupling. An adhesive effect equal to that obtained with synthetic resins could be obtained if the frequency of interfiber linkages is high enough (Widsten et al., 2004). The effect from the lignin based phenolic production was really small in all impact categories under study (less than 5%). Furthermore, in the HB production process, it was reported that some phenolic compounds, such as condensed tannins (e.g. mimosa tannin), do not improve mechanical properties of the HB more than the laccase treatment alone (Widsten et al., 2009). Therefore, a sensibility analysis was carried out and several alternative scenarios were proposed in order to assess the influence of the two-component bio-adhesive production on the environmental profile:

- **Scenario A** is the current conventional HB production process, considering the use of 34 kg of PF as bonding agent (González-García et al., 2009a).
- **Scenario B** is the current green process of the HB plant under study considering as bonding agent 10.5 kg of laccase and 40 kg of lignin based phenolic material. It is interesting to underline that the cost of this scenario is 12 fold the value corresponded to the conventional process due to the huge dose of enzyme.
- **Scenario C** is characterized by the use of 10.5 kg of laccase (hypothetical alternative)
- **Scenario D** is characterized by the reduction of 10% in the two-component bio-adhesive dose required (9.45 kg of laccase and 36 kg of lignin based material).
- **Scenario E** is characterized by the exclusion of the laccase (the enzyme production is a hot spot in the current process) and increasing by 25% the dose of lignin based material, that is 50 kg. In fact, this scenario was carried out at pilot scale and HBs with similar properties were obtained. From an economic point of view, this scenario may be an interesting alternative, since the related costs are considerably lower as it did not require the the enzyme.

Fig. 4 shows the comparative environmental profile between conventional HB using PF as a resin (scenario A) and alternative scenarios of green HB production (scenarios B, C, D and E). The values were indexed using Scenario A as the baseline (index = 100 for each impact category under study). As expected, the highest reduction in the environmental profile was achieved with a reduction in the laccase dose (Scenario E, removal of laccase dose from the adhesive). More research should be carried out in this issue as the use of this enzyme as biocatalyst in board production is very recent (Widsten et al., 2009) and the definition of optimum dose should require further study.

The exclusion or dose reduction of the lignin based component also causes an environmental benefit in some impact categories. Scenario C, where only laccase is considered, entails a reduction up to 55% of contributions to PO, and up to 30% the consumption of primary energy in comparison with the conventional HB production process. In addition, the mechanical properties of the wood composite board could be similar to the current green HB properties (Widsten et al., 2009).

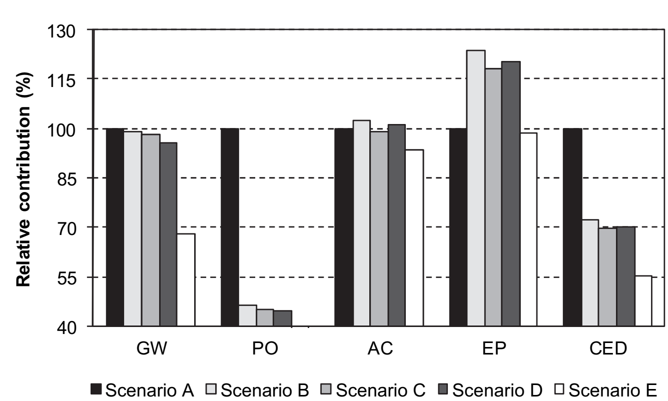

Fig. 4. Relative environmental profile of the different hardboard production processes according to the blending agents, the conventional process serving as the baseline (Index = 100).

In Scenario D (with a reduction of only 10% in the dose of green adhesive) it was possible to obtain reductions up to 4%, 55% and 30% in GW, PO and CED, respectively. In contrast, for the remaining impact categories the contributions increased.

On the contrary, Scenario E showed reductions in all impact categories: 32% for GW, 69% for PO, 6% for AC, 1.5% for EP and 45% for CED. This potential scenario is the only one which achieved improvement in the environmental profile in terms of AC and EP in comparison with the conventional one, specifically due to the non-utilization of laccase and the subsequent reduction of associated energy.

Therefore, and according to these results (Fig. 4), the change from conventional HB to green HBs can reduce the contributions to almost all impact categories under study excluding EP, where the enzyme manufacture shows an important role. The large use of energy, as well as the carbohydrates (sugar and starch) and protein consumption in the laccase production process (Nielsen et al., 2007) and the emissions of COD in the lignin based material production process (González-García et al., Submitted for publication) are the main responsible for this increase of nutrient enrichment in comparison with the conventional process. The removal of the laccase from the adhesive is the only possible option to reduce the eutrophying emissions.

With regard to CED, the entire energy demand could be reduced by 45% since almost 30% of energy required in the subsystem of Wood preparation is associated to the production of laccase.

Important reductions can be achieved regarding PO (it is possible to reduce the environmental profile up to 55% changing conventional HB to green HBs), in particular due to the avoidance of the emission of formaldehyde by means of the production and use of green bonding agents instead of PF.

4.1. Sensitivity analysis

In order to make substantial improvements in the environmental performance of green HB production process, it should be necessary to address the scenarios of major contribution to environmental impact. The normalization phase allows us to compare all environmental impacts using the same scale as well as to add the normalization values of impact categories in order to obtain a unique value per scenario. The dose of green bonding agent was analysed in more detail and reductions of 10%, 20% and 25% in the dose of laccase, lignin based material and both were proposed and analysed. Fig. 5 shows the normalization index per scenario only taking into account the normalization values of the four impact categories under analysis: GW, PO, AC and EP. The situation in Western Europe has been taken as the reference for all impact categories (data from year 1995) as this is the most complete list available (Guinèe et al., 2001). Regarding the energy requirements (CED), the CML methodology does not include it, therefore this flow indicator has been only considered at the characterisation step. According to these outcomes, the categories can be arranged in the following order:

1. Hightly significant: AC
2. Significant: EP
3. Lightly significant: GW and PO

According to the results shown in Fig. 5, the current green HB production process (0% of reduction) improves the environmental profile up to 18% in comparison with the conventional process using PF. The reduction in the dose of green adhesive (up to 25%) only improves the environmental profile by 19%. However, both reductions of 100% in the laccase and increases of 25% in the lignin material dose were also performed, taking into account that this scenario was just assessed at pilot scale and there is no large difference on the HBs properties. According to its results, the normalization index could be reduced by 93% in comparison with the conventional process.

If we assess the results in more detail, two impact categories were identified as the most significant in terms of normalization values: AC and EP. According to our previous results (Fig. 4), the EP is the impact category with greatest increase (up to 25%) when green HBs are produced instead of conventional HBs with PF as bonding agent. Moreover, this result is already known since the application of enzymes to other industrial processes such as paper pulp bleaching also gives off more eutrophication in comparison with the conventional process (Fu et al., 2003) as the industrial production of enzymes requires energy and raw materials (Nielsen et al., 2007) and there may be processes in which the enzyme application is environmentally advantageous and there may be others, where they are not (Nielsen and Wenzel, 2007). The other impact category which is influenced by the laccase production (due to the energy requirement) is AC (Fu et al., 2003). Hence, a more detailed assessment is shown for both impact categories in Fig. 6 and Fig. 7.

4.1.1. Sensitivity analysis of EP and AC

As expected, the only reduction in the lignin based material dose (Fig. 6, Green HB-Li) supposes minimum influence on the

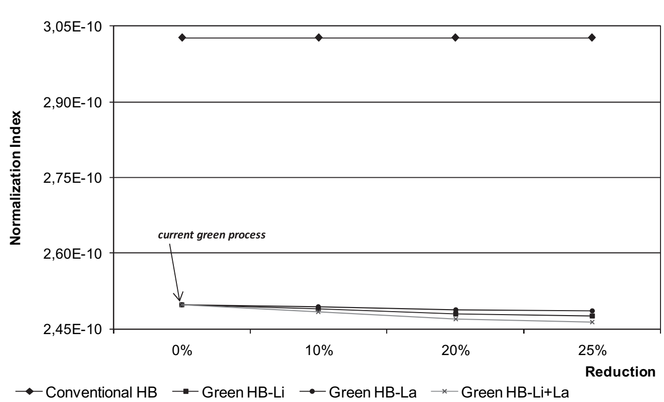

Fig. 5. Sensitivity analysis of the normalization index (per functional unit) taking into account normalization values of GW, AC, EP and PO for the scenarios under study. Acronyms: HB-Li = reductions of lignin based material, HB-La = reductions of laccase and HB-Li+La = reductions of lignin and laccase amount.

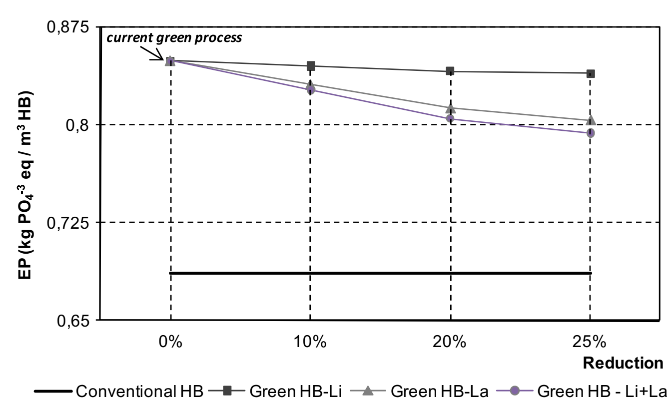

Fig. 6. Sensitivity analysis of equivalent PO_4^{-3} emissions as a result of the reduction on the dose of green blending agents. Acronyms: HB-Li = reductions of lignin based material, HB-La = reductions of laccase and HB-Li+La = reductions of lignin and laccase amount.

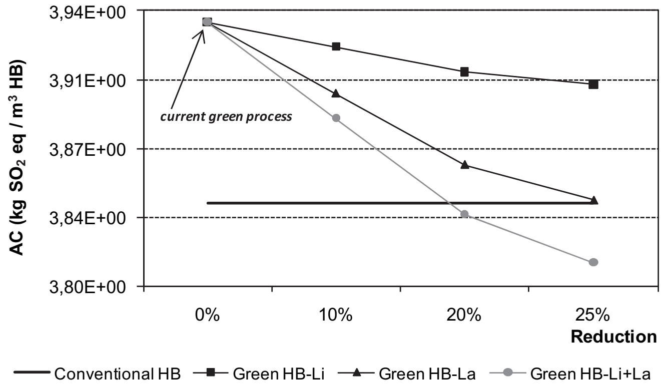

Fig. 7. Sensitivity analysis of equivalent SO_2 emissions as a result of the reduction on the dose of green blending agents. Acronyms: HB-Li = reductions of lignin based material, HB-La = reductions of laccase and HB-Li + La = reductions of lignin and laccase amount.

contributions to EP in comparison with the current green process (up to 1.2% with reduction of 25% in the dose) since in the current green production, it represents 5% of total contributions. However, the reduction of the laccase dose should produce increased effects on the eutrophying emissions. If the dose of lignin based material is maintained but the dose of laccase is modified and reduced by 25% (Fig. 6, Green HB-La), it is possible to achieve a reduction of 5.6% in the eutrophying emissions. Moreover, the combined reduction in both bonding agent means the highest reductions (Fig. 6, Green HB-Li + La): up to 7% with a decrease in the amount of both bonding agents. Although it is not presented in the figure, the removal of laccase (100%) with an increase in the lignin material by 25% shows the best environmental profile in terms of eutrophying emissions given that it is possible to reduce the equivalent PO_4^{-3} emissions up to 1.5% in comparison with the conventional process with PF.

As regards the results in terms of AC, the reduction of 20% and 25% in the dose of both bonding agents (Fig. 7, Green HB-Li + La) would allow reducing the equivalent SO_2 emissions below the result obtained for the conventional process. Reductions in lignin based material dose lightly reduce the acidifying emissions.

Furthermore, other aspects should be taken into account when conventional and green HBs are compared such as i) preventing the health-hazardous formaldehyde emissions during board production, ii) the non-toxicity application conditions of laccase technology, iii) biodegradability of generated wastewaters, iv) the possibility to increase the options of reusing of discarded boards, v) boards burning for energy without the generation of harmful emissions and vi) generating and usage of residuals from composite panel products, not only environmental but also economical and health benefits could thus be obtained from binderless (synthetic resin-free) boards production processes.

5. Conclusions

This work focused on the assessment of a wet-process green hardboard manufacture by means of a two-component adhesive based on laccase activated phenolic system was considered instead of PF resin. The relative environmental improvement potential of the green hardboard production in terms of several impact categories (GW, AC, EP, PO and FF) was assessed in detail in this paper in order to provide valuable information that can assist the forest-based industry, specifically the wood panel industry, to incorporate more environmentally-friendly adhesives and to improve their environmental performance and sustainability. Laccase, on-site thermal energy and electricity production as well as chipping stage considerably influenced the environmental impacts.

6. Recommendations and perspectives

The results indicate the production of green hardboards using a two-component bio-adhesive based on both a wood phenolic material and a phenol-oxidizing enzyme is industrially viable, meeting the specification of hardboards produced with the conventional phenolic resin. Further research should focus on the laccase production process in order to reduce its energy demand, as well as on the amount of green adhesive required and application conditions to obtain the same mechanical properties as the conventional hardboard.

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