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Environmental impact and cost assessment of a novel lignin production method

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ABSTRACT

The oil scarcity and the rise in earth temperature have elevated the interest in lignocellulosic biorefineries. Lignin has high potential to be used in various applications including the production of biomaterials and transportation fuels. Among the different sources of lignin, organosolv lignin has the advantage of being sulphur-free and of low ash content compared to other types of industrial lignin. The present study focuses on cradle-to-gate life cycle and cost assessment of a novel organosolv lignin production process from spruce bark. The system boundary included production of tannin, lignin from spruce bark and handling of waste including all the inputs (material and energy) and outputs (emissions) in the process. Baseline scenario and scenarios S1 and S2 were compared to identify the most environmentally and economically suitable scenario. The baseline scenario is lignin production with co-production of tannin and tannin free bark (TFB) from spruce bark; scenario S1 is lignin production from TFB; and scenario S2 is lignin production from TFB with mass allocation. The functional unit was 1 kg lignin produced and ReCiPe 2016 Midpoint (H) method was used for the environmental impact assessment. The results showed that the baseline scenario had higher global warming potential (GWP) (2.14 kg CO_2eq.) and total cost (1.959 €/kg) than S1 (1.39 kg CO_2 eq. and 1.377 €/kg respectively) and S2 (0.23 kg CO_2eq. and 0.998 €/kg respectively) scenarios. The results of sensitivity analysis showed that the use of bioethanol instead of ethanol reduced the burden on GWP but increased the burden on the land use impact category.

1. Introduction

The increasing global population and industrialization leads to an increasing demand for fuels, materials and chemicals, resulting in many social problems, including energy security and environmental distress (Sun et al., 2018). The climate change and the shortage of fossil fuels are driving the inventive utilization of renewable resources on earth (Edenhofer et al., 2011). Nowadays, society needs to move from fossil resources to renewable resources, in which lignocellulosic biomass can play an important role. The lignocellulosic biomass consists mainly of cellulose, hemicellulose and lignin (Matsakas et al., 2019). Whereas some biomass, such as spruce bark, also contain tannins or other type of extractives in significant amounts. Among the different sources of lignocellulosic biomass, the one derived from forest residues is an important renewable resource for European countries such as Sweden, where forests cover 57% of the land and play an important role in the national economy (Sveaskog, 2019).

Lignin is one of the most abundant naturally occurring terrestrial organic material on earth (Mu et al., 2019). Lignin is a large group of aromatic polymers resulting from the oxidative combinatorial coupling of 4-hydroxyphenylpropanoid units (Schlee et al., 2019). It can be used to produce high added-value products, such as carbon fiber, phenolic bioactive compounds, syngas, bio-jet fuel, marine fuel, multifunctional hydrocarbons, building material and various oxidized products (Baral et al., 2019). The worldwide production of lignin is approximately 100 million t/y valued at $732.7

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million in 2015 (Bajwa et al., 2019). It is estimated to reach $913.1 million by 2025 with compound annual growth rate (CAGR) of 2.2% (Bajwa et al., 2019). Nowadays, lignin is mainly generated as a waste product during second generation ethanol production, and pulp production (Cotana et al., 2014). There are three main types of lignin: first is sulphonated lignin which is a by-product of sulphite pulping that covers the highest percentage of lignin production (approximately 88%). Second is kraft lignin (approximately 9%), and third is organosolv lignin (approximately 2%) (Bajwa et al., 2019). Currently, the organosolv process is gaining popularity due to the advantage of producing sulphur free lignin (Nitsos et al., 2016) and that it contains lower ash relative to other types of industrial lignin, such as kraft lignin (De la Torre et al., 2013). Organosolv lignin is expected to present the highest growth, at an estimated CAGR of over 5% from 2016 to 2025 (Kumar et al., 2009).

Organosolv pre-treatment is considered one of the most promising methods for biomass delignification and fractionation, resulting in the production of relatively clean streams of cellulose, hemicellulose, and lignin (Matsakas et al., 2018). It generates three distinct streams, a cellulose-rich solid stream, a liquid stream containing solubilized hemicellulose, and a solid stream of high purity lignin (Raghavendran et al., 2018). Additionally, the lignin produced in this way retains the majority of β-ether bonds, maintaining a structure close to the natural one (Nitsos et al., 2018). As per De la Torre et al. (2013) sulphur free lignin has good potential for use as renewable fuel and high-added value applications.

Apart from being a source of lignocellulosic components, spruce bark is also a good source of tannin (Ding et al., 2017). The global tannin market is predicted to expand from $ 1.64 billion in 2017 to $ 3.05 billion by 2024 (Energias, 2020). Tannins are used in the leather industry, wine production, and pharmaceutical industries (Energias, 2018). In the process of tannin production, tannin free bark (TFB) is a waste material that normally would be used for heat generation. The European Parliament and the Council of the European Union in 2009 targeted the appropriate management and use of residues and waste materials. This is important for minimizing losses and maximizing environmental benefits. In the development of a resource efficient and sustainable society, European Commission (2017) introduced the circular (bio) economy (Zabaniotou, 2018). The recovery and conversion of such waste materials into a new valuable product promotes environmental benefits. The lignin production from TFB by organosolv process is one of them.

Life cycle assessment (LCA) is a tool that evaluates and identifies the most relevant environmental impacts and hotspots during the life cycle of any product or process (ISO, 2006a). Jönsson and Wallberg (2009) conducted LCA on hardwood lignin recovery from the cooking liquor and black liquor streams in a kraft pulp mill. Bernier et al. (2013) conducted LCA on kraft lignin production using the IMPACT 2002+ impact assessment method. They found that the main impacts of kraft lignin were due to the use of the natural gas subsystem for drying, sulphuric acid for washing and sodium hydroxide to make up for sodium losses. LCA studies have been performed on biorefineries for lignin nanoparticle production based on agricultural waste and for integrated lignin-kraft pulp production (Koch et al., 2019; Benali et al., 2016). As per our knowledge, none of previous published studies worked on lignin production from spruce bark using a novel organosolv process.

The aim of the present study was to assess the potential environmental impact and cost of a novel biorefinery (based on organosolv fractionation) process for the production of lignin from spruce bark including co-products: tannin, cellulose and hemicellulose. For identifying, the best process for lignin production with minimum potential environmental impacts and cost, possible changes have been proposed in the main biorefinery process. A prototypical perspective of a spruce bark biorefinery was modelled using process variations. These variations were included within scenarios that investigated specific process parameters and their influence on the environmental impacts as well as the identification of hotspots within the process. These environmental aspects shall be made available for the early stage process development of the sulphur free lignin production.

2. Life Cycle Assessment methodology

2.1. Goal and scope definition

The goal of the present study was to quantify the environmental impact and cost of lignin production from spruce bark. Three different scenarios were proposed (baseline scenario, scenario S1 and scenario S2) and compared.

2.1.1. Functional unit

According to the ISO 14040 standard, the functional unit (FU) provides a reference point for inputs and outputs in an LCA study (ISO, 2006b). In the present study, 1 kg of lignin production from spruce bark was taken as the FU.

2.1.2. System boundary

A cradle-to-gate approach was adopted and thus all inputs (electricity, chemicals, water and spruce bark) and outputs [co-products (tannin, cellulose and hemicellulose) and emissions] were included within the system boundary. The spruce bark used as a feedstock in the lignin production was a waste product of forest industry and thus did not carry any harvesting emissions. It was assumed that the lignin production facility would be established near a sawmill thus no need for transportation of the bark. The schematic presentation of material and energy flows within the process and system boundary are showed in Fig. 1.

3. Life cycle inventory

In the present study, primary data related to lignin production were taken from laboratory experiments that were conducted in Luleå University of Technology, Sweden (Table 1). All mass and energy balances of this study were simulated supported by experimental data. The background data such as production of chemicals: $NaHSO_3$, Na_2CO_3, ethanol, sulphuric acid and Swedish mix electricity were taken from Ecoinvent database 3.5 and literature (Ecoinvent database 3.5, 2019).

3.1. Baseline scenario

The baseline scenario considered the whole biorefinery process that produces lignin together with other co-products (tannin, cellulose and hemicellulose) as showed in Fig. 1.

3.1.1. Tannin extraction and TFB

Norway spruce bark was air-dried at room temperature and milled in a Retsch SM 300 knife mill (Retsch GmbH, Haan, Germany) through a 1 mm screen and stored at room temperature until further use. The moisture content of the bark used for the experiment was 7.9% w/w. Tannin was removed in an air-heated multi-digester apparatus that contains 6 metallic cylinders of 2.5 L each using hot water extraction (Kalogiannis et al., 2018). More specifically, 110 g of dry spruce bark were added in 1.1 L of water containing 2% w/w_{solids} sodium bisulphite and 0.5% w/w_{solids} sodium carbonate to facilitate the tannin extraction (Kemppainen et al., 2014). The solution was placed in the metallic cylinders and treated at 75 °C for 2 h under constant slow mixing through rotating the cylinders. Heating took place with resistances and air

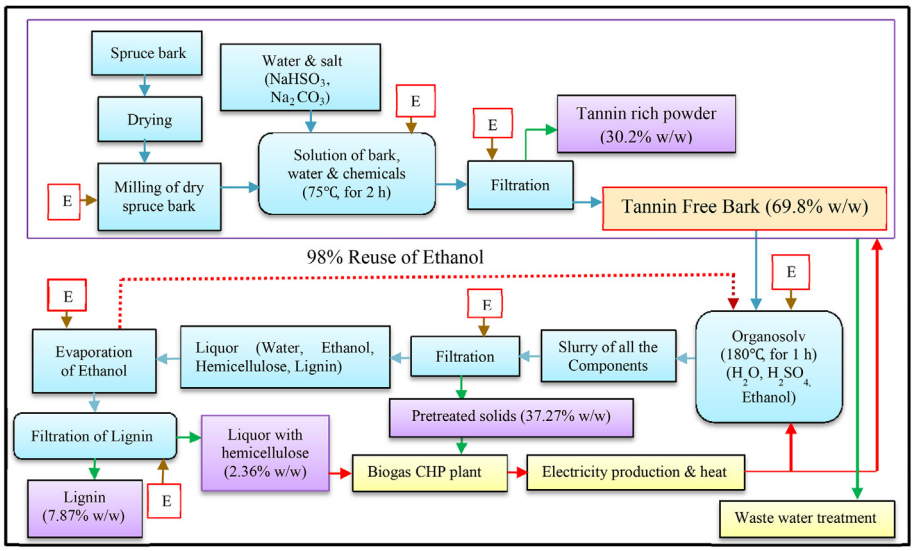

Fig. 1. Schematic outlining of lignin production process (baseline scenario) with material flow and Energy (E) flow in the system.

Table 1
Inputs for 1 kg lignin production in Baseline scenario.

Inputs for hot water extraction of tannin			
Components	Unit	Amount	Reference/Source
Spruce bark	kg	12.71	Experimental laboratory data
Sodium bisulfite ($NaHSO_3$)	g	254.2	
Sodium carbonate (Na_2CO_3)	g	63.55	
Water	L	127.1	
Heating, 75 °C for 2 h	kWh	1.6	Ding et al. (2017)
Milling of bark	kWh	0.18	
Filtration	kWh	0.85	
Inputs for organosolv fractionation of TFB			
TFB	kg	8.87	Experimental laboratory data
Water	L	35.48	
Sulphuric acid	g	88.7	
Ethanol	L	53.22	
Ethanol loss as Air Emissions	L	1.07	Shanmugam et al. (2019)
Heating, 180 °C for 2 h	kWh	2.37	Ding et al. (2017)
Filtration (cellulose recovery)	kWh	0.57	
Filtration (lignin recovery)	kWh	0.26	

flow by using electricity. At the end of the treatment, the solution was vacuum-filtered and two fractions were collected: the tannin solution (containing the 30.2% w/w of the initial spruce bark; roughly 2% w/v solids) from which the tannin rich powder (containing 48.1% w/w tannins) was recovered after water evaporation and the TFB fraction (69.8% w/w of the initial spruce bark). TFB was air-dried at room temperature.

3.1.2. Organosolv fractionation of TFB

TFB was fractionated by organosolv to produce lignin as the main product, and cellulose and hemicellulose as co-products. Specifically, 90 g of dry TFB were added in 0.54 L of ethanol (≥99.8%), 0.36 L of water and 1% w/w_{solids} of concentrated sulphuric acid. Organosolv treatment took place in the same multi-digester apparatus that was used for the tannin extraction, at 180 °C for 1 h. At the end of the treatment, the solution was vacuum-filtrated. Two fractions were collected: a 'cellulose-rich pre-treated solids' fraction and a liquid fraction (containing ethanol, water, hemicellulose, and lignin). The yield of cellulose in pre-treated solids was 53.4% (dry basis). The pre-treated liquor containing lignin and hemicellulose was transferred to solvent distillation unit to remove ethanol and reduce the solubility of lignin. It was assumed that 98% of the ethanol was recovered and re-used (Shanmugam et al., 2020). Then, lignin was recovered from the aqueous solution by vacuum-filtration while the aqueous supernatant still contained the solubilized sugars (hemicellulose). The yielding of lignin and hemicellulose was 11.27% and 4.75% of the TFB (dry basis) respectively. The lignin produced by this process had minimal sugar contamination (2.28% w/w) and ash contamination (0.58% w/w).

After lignin removal, the aqueous solution that contains the solubilized hemicellulose was assumed to be sent to a bio-digester where it was mixed with the pre-treated cellulosic solids and the entire mixture was digested anaerobically. The pre-treated solids were tested for their potential for methane production via anaerobic digestion in the laboratory. For this purpose, biochemical methane potentials tests were done by using an automatic methane potential test system II (Bioprocess Control AB, Lund, Sweden) as described in Matsakas et al. (2015). Digestion took place at thermophilic conditions at 55 °C with an inoculum to substrate ratio of two in terms of volatile solids. The methane yield was found to be 180.79 ml CH_4/g volatile solids, which is equivalent to 172.96 ml CH_4/g total solids (accounting for 4.33% w/w ash content

in the pre-treated solids). According to Himanshu et al. (2018) a sugar rich waste stream generates 314 ml CH_4/g. In the present study, production of 1 kg of lignin generated 4.74 kg cellulose-rich pre-treated solids and 0.30 kg of hemicellulose which in their turn could generate approximately 0.91 m^3 biogas that is equivalent to approximately 9.1 kWh of electricity (FNR, 2009). The biogas generated from the bio-digester was assumed to be combusted in a combined heat and power (CHP) plant to generate electricity and heat which is more than sufficient to meet energy demands of organosolv reaction chamber and solvent recovery units. Excess electricity could be sold to the grid while excess heat could be used in the drying of the spruce bark. In this study, it was assumed that 97% of the process water is recycled because each plant usually has its own onsite closed-circuit wastewater treatment system while there are 3% possibility of water spill due to technical fault in the system (Baral et al., 2019). A schematic overview of the baseline scenario is shown in Fig. 1, whereas in Table 1 the inputs adjusted for the production of 1 kg of lignin.

3.2. Scenario S1

In scenario S1 it was assumed, that the TFB that was used as a feedstock in the lignin production was a by-product of tannin production and thus did not carry any upstream emissions. Waste stream of cellulose-rich pre-treated solids and hemicellulose were treated by anaerobic digestion similarly to baseline scenario. In this scenario production of tannin was not considered under the system boundary (Fig. 2a).

3.3. Scenario S2

In scenario S2, similarly to S1, it was assumed that the TFB that was used as a feedstock in the lignin production was a by-product of tannin production and thus did not carry any upstream emissions. For the analysis of the efficiency of this novel method for lignin production and a comparison of environmental impact with existing lignin production methods. Scenario S2 calculated the environmental impact solely associated to lignin production as showed in Fig. 2b. It was assumed that the co-products cellulose-rich pre-treated solids and hemicellulose were used by a pulpmill or other industry. According to the ISO standard, co-product allocation should be avoided wherever possible in LCA decision making (Mackenzie et al., 2017). However, if needed there are different allocation methods available such as mass, volume based (physical allocation) or economic allocation. In the present study, for assessing the environmental load due to lignin production, mass allocation was used according to Cherubini et al. (2018) and Pré-sustainability (2020). In the latter study, 75% of the emissions and energy consumption was allocated to the 'cellulose-rich pre-treated solids', 20% was allocated (included impurities) to the lignin fraction and 5% to the hemicellulose. Economic allocation would have to be based on assumptions on the market price (Dolezal et al., 2014) of organosolv lignin, which currently is under development and the market price is changing significantly, which will increase the uncertainty of the results. The system boundary of scenario S2 is shown in Fig. 2b. Due to absence of a biogas plant in scenario S2, a Swedish mix electricity grid was used for lignin production.

4. Life cycle impact assessment

The environmental assessment was conducted according to the characterization factors reported in the ReCiPe (World-H) 2016 midpoint method using Simapro 9 LCA software. The impacts categories assessed were global warming potential (GWP) (kg CO_2-eq.), ozone depletion potential (ODP) (kg CFC11 eq.), ionizing radiation potential (IRP) (kBq Co-60 eq.), terrestrial acidification potential (TAP) (kg SO_2 eq.), freshwater eutrophication potential (FWEP) (kg P eq.), freshwater ecotoxicity potential (FEP) (kg 1,4-DCB eq.), marine ecotoxicity potential (MEP) (kg 1,4-DCB eq.), human toxicity potential (HTP) (kg 1,4-DCB eq.), and land use potential (LUP) (m^2a crop eq.).

5. Environmental and production cost

The objective of the environmental and production cost analysis was to estimate and compare the production costs of different scenarios, find out the most economic scenario and provide a first indication of the economic and environmental feasibility of the scenarios. The study focused on the environmental and production cost due to very high uncertainty of the capital investment cost. The production cost was calculated for each scenario based on the consumption of raw material, chemicals and energy consumption using mass and energy calculations as proposed by Daylan and Ciliz (2016) and Thunman et al. (2019). The calculations of production costs require prices for raw materials production as suggested by Özkan et al. (2016). The raw materials prices were: bark at 27 €/t (Swedish Energy agency, 2020), sodium carbonate at 260 €/t (Dong et al., 2017), sulphuric acid at 100 €/t and ethanol at 700 €/t was taken from Mesfun et al. (2019), sodium bisulphite at 487 €/t (TAMU, 2016) and bioethanol at 776 €/t (Chembid, 2020). Electricity price was assumed to be 0.039 €/kWh (Statistics Sweden, 2019). The production cost of all the scenarios was estimated for 1 kg of lignin production, same as the FU of the study. The production cost did not include taxes, subsidies and labour cost. The

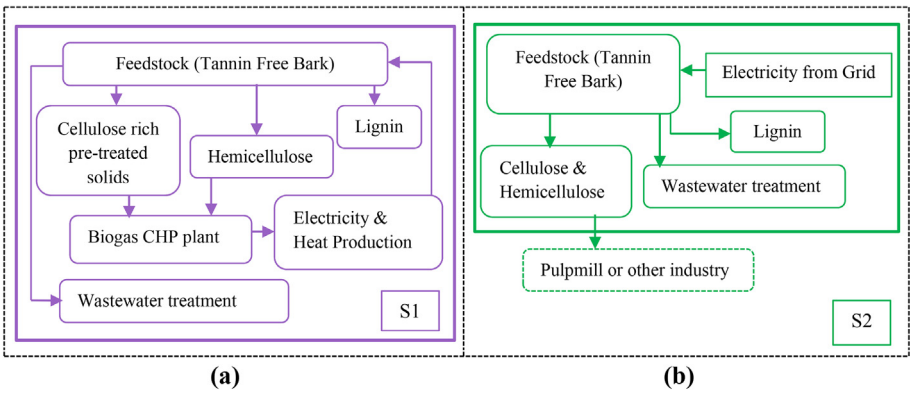

Fig. 2. Graphical representation of scenario S1 and S2.

environmental cost for each impact category was taken at the midpoint level from De Bruyn et al. (2018). Table 2 shows the calculation method of production, environmental and total cost.

6. Results and discussion

Fig. 3 describes the relative contribution on environmental impact categories of all components that were used in the baseline scenario. Use of electricity had the highest contribution on all impact categories except GWP. Use of ethanol and sodium bisulphite contributed considerably on all impact categories. While use of bark, water, sodium carbonate and sulfuric acid had low impact contributions.

Fig. 4 shows the relative contribution on each impact category of tannin and lignin production in the case that tannin would carry all environmental impact for its production as in scenarios S1 and S2 (TFB is considered as a by-product). Tannin production had significant contribution on all the impact categories due to the use of chemicals. This environmental burden could be reduced in the future by avoiding including such chemicals, as it was shown that relative high tannin extraction yields can also be achieved in the absence of chemicals as mentioned by Kemppainen et al. (2014) and Ding et al. (2017). Further research on this field could improve the tannin extraction yields without the need to add such chemicals. Considering TFB as a by-product of tannin production would mean that approximately 30% of the burden could be reduced on GWP, 64% on IRP, 50% on TAP and 45% on HTP impact categories compared to the baseline scenario (Fig. 4). It was found that the main hotspot of the whole process is the source of electricity and use of chemicals.

Table 3 shows the results of environmental impact of different scenarios on the selected impact categories. The baseline scenario had the high environmental impact on all the impact categories. While scenarios S1 and S2 had the low. This is because scenarios S1 and S2 considered TFB as feedstock in lignin production instead of fresh bark, thus reducing the need for chemicals, water and electricity in the process. The baseline scenario had high impact on HTP (3.69 kg 1.4-DCB) impact category due to the use of the chemicals sodium bisulphite and sodium carbonate in the process of tannin extraction and sulphuric acid in the process of cellulose and lignin production. Other reason of high HTP is the use of electricity produced by anaerobic digestion. Electricity generation from biogas may lead to methane release during incomplete combustion of biogas and also there is risks for diffusive emission of nitrous oxide related to biomass storage and digestate management (Paolini et al., 2018). Methane and nitrous oxide can have some adverse effects on human health. Use of electricity, the production process of ethanol and sodium bisulphite that was used in the baseline scenario gave rise to GWP (2.14 kg CO_2 eq.). The use of bark as main feedstock in the process caused a LUP of 0.26 m^2a crop eq. The rest of the other environmental impact categories had very low values as showed in Table 3. If we compare the results of the present study with previous published studies, in scenario S2, GWP (0.23 kg CO_2 eq./kg lignin) and TAP (0.0007 kg SO_2/kg lignin) are lower than the kraft lignin GWP (0.6 kg CO_2 eq./kg lignin) and TAP (0.012 kg SO_2/kg lignin) presented in Bernier et al. (2013). Wells et al. (2015) showed GWP (1.75 kg CO_2 eq./kg lignin) at industrial scale that is higher than the GWP (1.39 kg CO_2 eq./kg lignin) in scenario S1 and S2 (0.23 kg CO_2 eq./kg).

6.1. Sensitivity analysis

The LCA results are sensitive to two key parameters, source of electricity and ethanol used in the process. The sensitivity analysis was performed for all scenarios using (a) bioethanol instead of ethanol and (b) different source of electricity (Swedish, European, Global and biogas based). As shown in results in Table 3, using bioethanol in baseline scenario, scenarios S1 and S2 instead of ethanol had very little difference or same impact on most of the impact categories (ODP, IRP, TAP, FWEP, FEP, MWEP and HTP). However, in baseline scenario the impact on GWP decreased from 2.14 kg CO_2 to 1.54 kg CO_2 and increased on LUP (0.26 m^2a to 2.94 m^2a) impact category due to use of wood in the production process of bioethanol. The same pattern was followed by the scenarios S1 and S2 for GWP and LUP impact categories.

The other parameter that was used for sensitivity analysis was the different source of electricity (biogas generated electricity, Swedish electricity, European electricity and Global electricity). The results are shown in Fig. 5. The highest impact was observed on GWP when global electricity was used. This is due to the high percentage of coal based electricity generation at a global level. The lowest GWP was found when Swedish mix electricity was used. Swedish electricity mix is based on high percentage of nuclear, hydro and renewable power and a low percentage of coal. The impact on IRP was the highest in all the scenarios due to the use of nuclear power in the Swedish grid electricity mix. The use of biogas-based electricity showed the lowest impact on IRP impact category in all scenarios.

6.2. Environmental and production cost

As the novel lignin production process is not yet commercially implemented industrial data are not available and capital investment cost is highly uncertain at experimental level. The production

Table 2
Production and environmental cost of lignin production.

Life cycle cost	Different cost	Components	Subcomponents		
Total cost	Production cost	Materials	Feedstock cost		
			Chemicals cost		
			Electricity cost		
	Environmental cost	Pollution cost	**Impact Indicator**	**Unit**	**Externality Cost**[a] **(€/kg)**
			GWP	kg CO_2 eq.	0.057
			ODP	kg CFC-11 eq.	30.400
			IRP	kBq Co-60 eq.	0.046
			TAP	kg SO_2 eq.	4.970
			FWEP	kg P eq.	1.860
			FEP	kg 1,4-DCB	0.036
			MEP	kg 1,4-D	0.099
			HTP	kg 1,4-DCB	0.085
			LUP	m^2a crop eq.	0.084

[a] Source (De Bruyn et al., 2018).

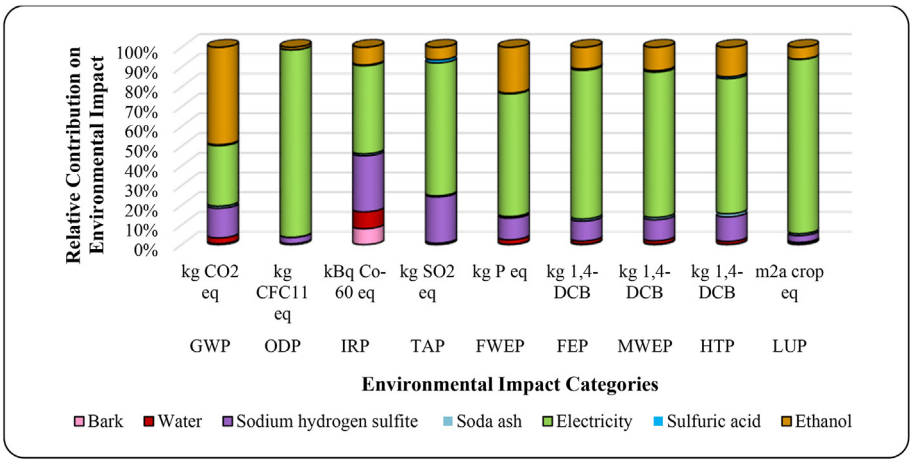

Fig. 3. Relative contribution of each component involved in each impact category in baseline scenario.

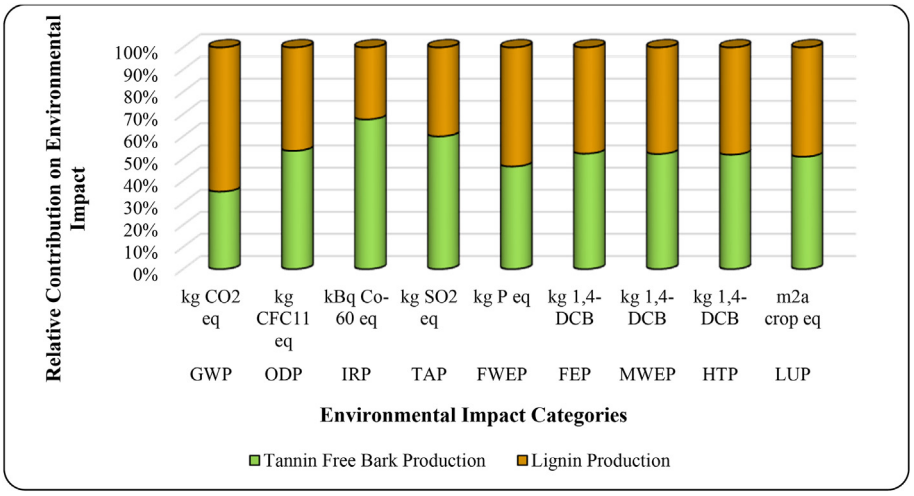

Fig. 4. Relative contribution of tannin & tannin free bark and rest of process (lignin production) on each impact category in baseline scenario.

Table 3
Environmental impact of **baseline** scenario, scenario S1 (**S1**) and scenario S2 (**S2**) using ethanol and bioethanol in lignin production process (per kg lignin).

Impact Category	Unit	Using Ethanol			Using Bioethanol		
		Baseline	S1	S2	Baseline	S1	S2
GWP	kg CO_2 eq.	2.14	1.39	0.23	1.54	0.79	0.11
ODP	kg CFC11 eq.	0.00	0.00	0.00	0.00	0.00	0.00
IRP	kBq Co-60 eq.	0.21	0.07	0.15	0.20	0.06	0.15
TAP	kg SO_2 eq.	0.04	0.02	0.0007	0.04	0.02	0.00
FWEP	kg P eq.	0.00	0.00	0.00	0.00	0.00	0.00
FEP	kg 1,4-DCB eq.	0.15	0.07	0.01	0.14	0.06	0.00
MWEP	kg 1,4-DCB eq.	0.19	0.09	0.01	0.19	0.08	0.01
HTP	kg 1,4-DCB eq.	3.69	1.76	0.15	3.69	1.80	0.16
LUP	m^2a crop eq.	0.26	0.13	0.01	2.94	2.81	0.55

and environmental costs depend on the raw material used in each of the scenarios. Environmental costs were calculated, by multiplying the environmental impact of each category by its externality cost (mentioned in Table 2). Environmental costs are thus proportional to environmental impact. As shown in Table 4, using bioethanol instead of ethanol had a total cost that was higher in all scenarios due to high production cost. It is due to slightly higher price of bioethanol in comparison to ethanol. The manufacturing process of bioethanol from wood had high impact on the LUP impact category that also increased the environmental cost of bioethanol. Scenarios S1 and S2 had lower environmental cost as well as production cost. It is due to using TFB as feedstock that reduced the burden of sodium carbonate, sodium bisulphite and amount of electricity. The highest production cost and total cost (production and environmental cost) were found in baseline scenario with bioethanol 1.296 €/kg and 2.221 €/kg lignin and the lowest was found in scenario S2 with ethanol 0.958 €/kg and 0.998 €/kg lignin, as shown in Table 4.

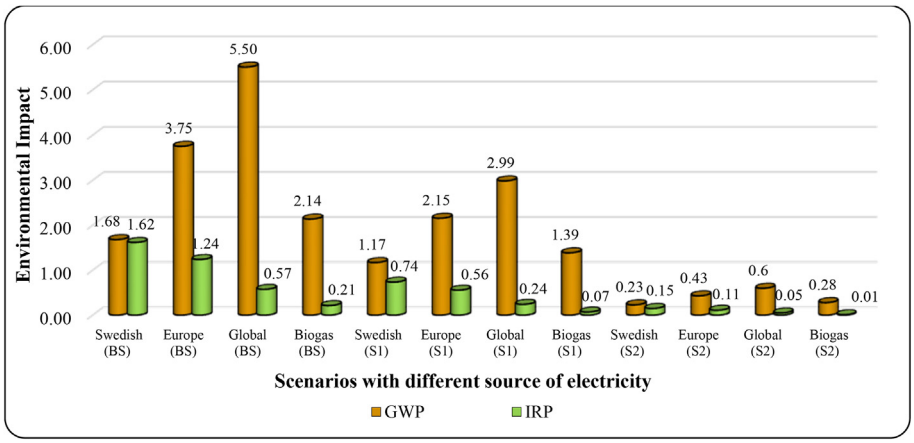

Fig. 5. Environmental impact (GWP and IRP) on baseline scenario (**BS**), scenario S1 (**S1**) and scenario S2 (**S2**) using different source of electricity (per kg lignin production).

Table 4
Production cost, environmental cost and total costs (€/kg lignin) of baseline scenario, scenario S1 and scenario S2 as per functional unit.

Components	Scenarios			Scenarios		
	Baseline	S1	S2	Baseline	S1	S2
	Using ethanol			Using bioethanol		
Spruce bark	0.345	–	–	0.345	–	–
Tannin free bark	–	0.345	0.344	–	0.345	0.344
Sodium carbonate	0.066	–	–	0.066	–	–
Sodium bisulphite	0.031	–	–	0.031	–	–
Sulphuric acid	0.009	0.009	0.009	0.009	0.009	0.009
Ethanol	0.587	0.587	0.587	–	–	–
Bioethanol	–	–	–	0.650	0.650	0.650
Electricity	0.195	0.092	0.019	0.195	0.092	0.019
Sum of production cost (A)	**1.232**	**1.032**	**0.958**	**1.296**	**1.096**	**1.022**
Environmental cost (B)	**0.727**	**0.345**	**0.04**	**0.925**	**0.553**	**0.081**
Total cost (A + B)	**1.959**	**1.377**	**0.998**	**2.221**	**1.649**	**1.103**

7. Conclusion

The study performed LCA and cost analysis of a novel process of lignin production using organosolv biomass fractionation. It was observed that the environmental impact of lignin production depends on subsequent conversion steps, on allocation of impacts between main product and co-products, and on upstream impacts of process components. Lignin produced in scenario S2 had the lowest environmental impact on impact categories. Results of sensitivity analysis showed that use of Swedish mix electricity helps in reducing the global warming potential in all scenarios due to low percentage of fossil fuel in electricity production. Use of biogas based electricity reduced the impact on ionization radiation impact category. Scenario S2 with using ethanol is the best scenario on the basis of environmental impacts as well as total cost. It was observed that the environmental cost depends on ozone depletion impact category due to its high external cost. But in the present study ozone depletion impact is zero in all the scenarios and that is the reason, HTP and TAP impact categories showed significant contribution in environmental cost in all the scenarios. The purity of lignin and other co-products that are produced in the novel process is also very important for the market shift from low value applications of lignin to high value products that generate higher earnings. The process would be more environmentally efficient if placed near the industry such as pulp and paper industry, tannin industry or ethanol industry. The cost of lignin production also depends on the subsidy of bioethanol and other taxes, and labour cost and value of other co-products that make revenue in the cost.

Credit author statement

In this manuscript Pooja Yadav, Dimitris Athanassiadis and Mats Tysklind collected data, conducted life cycle assessment and cost analysis, and manuscript writing. Io Antonopoulou, Ulrika Rova, Paul Christakopoulos, Leonidas Matsakas conducted the laboratory experiment at Luleå University of Technology, and Io Antonopoulou and Leonidas Matsakas helped in writing the manuscript. Finally Pooja Yadav, Dimitris Athanassiadis, Io Antonopoulou and Leonidas Matsakas discussed the results and contributed to the final manuscript.

Declaration of competing interest

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

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References

Bajwa, D.S., Pourhashem, G., Ullah, A.H., Bajwa, S.G., 2019. A concise review of current lignin production, applications, products and their environmental impact. Ind. Crop. Prod. 139, 111526.

Baral, N.R., Kavvada, O., Mendez-Perez, D., Mukhopadhyay, A., Lee, T.S., Simmons, B.A., Scown, C.D., 2019. Techno-economic analysis and life-cycle greenhouse gas mitigation cost of five routes to bio-jet fuel blendstocks. Energy Environ. Sci. 12 (3), 807–824.
Benali, M., Ajao, O., Jeaidi, J., Gilani, B., Mansoornejad, B., 2016. Integrated lignin-kraft pulp biorefinery for the production of lignin and its derivatives: economic assessment and LCA-based environmental footprint. In: Production of Biofuels and Chemicals from Lignin. Springer, Singapore, pp. 379–418.
Bernier, E., Lavigne, C., Robidoux, P.Y., 2013. Life cycle assessment of kraft lignin for polymer applications. Int. J. Life Cycle Assess. 18 (2), 520–528.
Chembid com, 2020. https://www.chembid.com/en/results/cluster/B/1/?q=ethanol&filterCurrency=usd&filterLocation=Sweden. (Accessed 4 May 2020).
Cherubini, E., Franco, D., Zanghelini, G.M., Soares, S.R., 2018. Uncertainty in LCA case study due to allocation approaches and life cycle impact assessment methods. Int. J. Life Cycle Assess. 23 (10), 2055–2070.
Cotana, F., Cavalaglio, G., Nicolini, A., Gelosia, M., Coccia, V., Petrozzi, A., Brinchi, L., 2014. Lignin as co-product of second generation bioethanol production from ligno-cellulosic biomass. Energy Procedia 45, 52–60.
Daylan, B., Ciliz, N., 2016. Life cycle assessment and environmental life cycle costing analysis of lignocellulosic bioethanol as an alternative transportation fuel. Renew. Energy 89, 578–587.
De Bruyn, S., Ahdour, S., Bijleveld, M., de Graaff, L., Schep, E., Schroten, A., Vergeer, R., 2018. Environmental Prices Handbook 2017: Methods and Numbers for Valuation of Environmental Impacts, vol. 18. CE Delft, Delft, 7N54.125.
De la Torre, M.J., Moral, A., Hernández, M.D., Cabeza, E., Tijero, A., 2013. Organosolv lignin for biofuel. Ind. Crop. Prod. 45, 58–63.
Ding, T., Bianchi, S., Ganne-Chédeville, C., Kilpeläinen, P., Haapala, A., Räty, T., 2017. Life cycle assessment of tannin extraction from spruce bark. iFor. Biogeosci. For. 10 (5), 807.
Dolezal, F., Spitzbart, C., Mötzl, H., 2014. Survey of Allocation Methods in Life Cycle Assessments of Wood Based Products. Performance and Maintenance of Bio-Based Building Materials Influencing the Life Cycle and LCA, p. 5.
Dong, X., Trembly, J., Bayless, D., 2017. Techno-economic analysis of hydraulic fracking flowback and produced water treatment in supercritical water reactor. Energy 133, 777–783.
Ecoinvent Database 3.5. https://www.ecoinvent.org/database/ecoinvent-36/ecoinvent-36.html. (Accessed 17 October 2019).
Edenhofer, O., Pichs-Madruga, R., Sokona, Y., Seyboth, K., Kadner, S., Zwickel, T., Eickemeier, P., Hansen, G., Schlömer, S., von Stechow, C., Matschoss, P. (Eds.), 2011. Renewable Energy Sources and Climate Change Mitigation: Special Report of the Intergovernmental Panel on Climate Change. Cambridge University Press.
Energias Market Research, 2018. https://www.globenewswire.com/news-release/2018/05/04/1496771/0/en/Tannin-Market-to-witness-a-CAGR-of-7-7-during-2018-2024-Energias-Market-research-Pvt-Ltd.htmlmay. (Accessed 22 August 2019).
Energias Market Research, 2020. https://www.globenewswire.com/news-release/2018/05/04/1496771/0/en/Tannin-Market-to-witness-a-CAGR-of-7-7-during-2018-2024-Energias-Market-research-Pvt-Ltd.html. (Accessed 30 April 2020).
FNR, E., 2009. Biogas-Messprogramm II. 61 Biogasanlagen im Vergleich. 1. Aufl. Gülzow: Fachagentur Nachwachsende Rohstoffe eV (FNR).
Himanshu, H., Murphy, J.D., Grant, J., O'Kiely, P., 2018. Synergies from co-digesting grass or clover silages with cattle slurry in in vitro batch anaerobic digestion. Renew. Energy 127, 474–480.
ISO, 2006a. Organización Internacional de Normalización, 2006. ISO 14044: Environmental Management, Life Cycle Assessment, Requirements and Guidelines. ISO.
ISO, 2006b. ISO-norm, I.S.O., 2006. Environmental Management-Life Cycle Assessment-Principles and Framework ISO 14040: 2006. ISO, Geneva, Switzerland.
Jönsson, A.S., Wallberg, O., 2009. Cost estimates of kraft lignin recovery by ultrafiltration. Desalination 237 (1–3), 254–267.
Kalogiannis, K., Matsakas, L., Aspden, J., Lappas, A., Rova, U., Christakopoulos, P., 2018. Acid assisted organosolv delignification of beechwood and pulp conversion towards high concentrated cellulosic ethanol via high gravity enzymatic hydrolysis and fermentation. Molecules 23 (7), 1647.
Kemppainen, K., Siika-aho, M., Pattathil, S., Giovando, S., Kruus, K., 2014. Spruce bark as an industrial source of condensed tannins and non-cellulosic sugars. Ind. Crop. Prod. 52, 158–168.
Koch, D., Paul, M., Beisl, S., Friedl, A., Mihalyi, B., 2019. Life cycle assessment of a lignin nanoparticle biorefinery: decision support for its process development. J. Clean. Prod. 118760.
Kumar, P., Barrett, D.M., Delwiche, M.J., Stroeve, P., 2009. Methods for pretreatment of lignocellulosic biomass for efficient hydrolysis and biofuel production. Ind. Eng. Chem. Res. 48 (8), 3713–3729.
Mackenzie, S.G., Leinonen, I., Kyriazakis, I., 2017. The need for co-product allocation in the life cycle assessment of agricultural systems-is "biophysical" allocation progress? Int. J. Life Cycle Assess. 22 (2), 128–137.
Matsakas, L., Nitsos, C., Raghavendran, V., Yakimenko, O., Persson, G., Olsson, E., Rova, U., Olsson, L., Christakopoulos, P., 2018. A novel hybrid organosolv: steam explosion method for the efficient fractionation and pretreatment of birch biomass. Biotechnol. Biofuels 11 (1), 160.
Matsakas, L., Raghavendran, V., Yakimenko, O., Persson, G., Olsson, E., Rova, U., Olsson, L., Christakopoulos, P., 2019. Lignin-first biomass fractionation using a hybrid organosolv-Steam explosion pretreatment technology improves the saccharification and fermentability of spruce biomass. Bioresour. Technol. 273, 521–528.
Matsakas, L., Rova, U., Christakopoulos, P., 2015. Sequential parametric optimization of methane production from different sources of forest raw material. Front. Microbiol. 6, 1163. https://doi.org/10.3389/fmicb.2015.01163.
Mesfun, S., Matsakas, L., Rova, U., Christakopoulos, P., 2019. Technoeconomic assessment of hybrid organosolv-steam explosion pretreatment of woody biomass. Energies 12 (21), 4206.
Mu, L., Wu, J., Matsakas, L., Chen, M., Rova, U., Christakopoulos, P., Zhu, J., Shi, Y., 2019. Two important factors of selecting lignin as efficient lubricating additives in poly (ethylene glycol): hydrogen bond and molecular weight. Int. J. Biol. Macromol. 129, 564–570.
Nitsos, C., Stoklosa, R., Karnaouri, A., Voros, D., Lange, H., Hodge, D., Crestini, C., Rova, U., Christakopoulos, P., 2016. Isolation and characterization of organosolv and alkaline lignins from hardwood and softwood biomass. ACS Sustain. Chem. Eng. 4 (10), 5181–5193.
Nitsos, C., Matsakas, L., Triantafyllidis, K., Rova, U., Christakopoulos, P., 2018. Investigation of different pretreatment methods of Mediterranean-type ecosystem agricultural residues: characterisation of pretreatment products, high-solids enzymatic hydrolysis and bioethanol production. Biofuels 9 (5), 545–558.
Özkan, A., Günkaya, Z., Tok, G., Karacasulu, L., Metesoy, M., Banar, M., Kara, A., 2016. Life cycle assessment and life cycle cost analysis of magnesia spinel brick production. Sustainability 8 (7), 662.
Paolini, V., Petracchini, F., Segreto, M., Tomassetti, L., Naja, N., Cecinato, A., 2018. Environmental impact of biogas: a short review of current knowledge. J. Environ. Sci. Health.Part A 53 (10), 899–906.
Pré-sustainability, 2020. Physical allocation. In: https://www.pre-sustainability.com/news/finding-your-way-in-allocation-methods-multifunctional-processes-recycling. (Accessed 15 April 2020).
Raghavendran, V., Nitsos, C., Matsakas, L., Rova, U., Christakopoulos, P., Olsson, L., 2018. A comparative study of the enzymatic hydrolysis of batch organosolv-pretreated birch and spruce biomass. Amb. Express 8 (1), 114.
Schlee, P., Hosseinaei, O., Baker, D., Landmér, A., Tomani, P., Mostazo-López, M.J., Cazorla-Amorós, D., Herou, S., Titirici, M.M., 2019. From waste to wealth: from kraft lignin to free-standing supercapacitors. Carbon 145, 470–480.
Shanmugam, K., Jansson, S., Gadhamshetty, V., Matsakas, L., Rova, U., Tysklind, M., Christakopoulos, P., Upadhyayula, V.K., 2019. Ecoefficiency of thermal insulation sandwich panels based on fly ash modified with colloidal mesoporous silica. ACS Sustain. Chem. Eng. 7 (24), 20000–20012.
Shanmugam, S., Krishnaswamy, S., Chandrababu, R., Veerabagu, U., Pugazhendhi, A., Mathimani, T., 2020. Optimal immobilization of Trichoderma asperellum laccase on polymer coated Fe3O4@ SiO2 nanoparticles for enhanced biohydrogen production from delignified lignocellulosic biomass. Fuel 273, 117777.
Statistics Sweden, 2019. https://www.scb.se/electrectrcity. (Accessed 27 November 2019).
Sun, Z., Fridrich, B., de Santi, A., Elangovan, S., Barta, K., 2018. Bright side of lignin depolymerization: toward new platform chemicals. Chem. Rev. 118 (2), 614–678.
Sveaskog, 2019. How much forest is there in Sweden. https://www.sveaskog.se/en/forestry-the-swedish-way/short-facts/brief-facts-1/. (Accessed 25 November 2019).
Swedish Energy agency, 2020. https://www.energimyndigheten.se/en/facts-and-figures. (Accessed 13 January 2020).
TAMU, 2016. Texas A&M University Libraries/chemical price. https://web.archive.org/web/20161125084558/http://www.icis.com/chemicals/channel-info-chemicals-a-z/. (Accessed 4 May 2020).
Thunman, H., Gustavsson, C., Larsson, A., Gunnarsson, I., Tengberg, F., 2019. Economic assessment of advanced biofuel production via gasification using cost data from the GoBiGas plant. Energy.Sci.Eng. 7 (1), 217–229.
Wells, K., Pors, D., Foan, J., Maki, K., Kouisni, L., Paleologou, M., 2015. CO_2 impacts of commercial scale lignin extraction at hinton pulp using the LignoForce process & lignin substitution into petroleum-based products. In: PACWEST Conference, pp. 10–13.
Zabaniotou, A., 2018. Redesigning a bioenergy sector in EU in the transition to circular waste-based Bioeconomy-A multidisciplinary review. J. Clean. Prod. 177, 197–206.