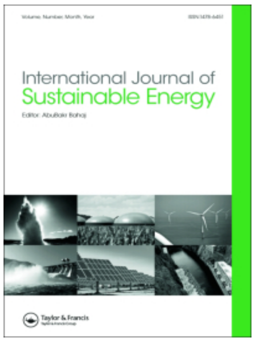

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Lignin valorisation: Life Cycle Assessment (LCA) considerations for enabling Circular Bioeconomy

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ABSTRACT

Lignin constitutes the sole renewable aromatic resource found in abundance on Earth and does not call into question the ethics of diverting land from food to energy production. In view of that, the potential of lignocellulosic biomass in promoting the Circular Economy and Bioeconomy concepts in future biorefinery operations is widely acknowledged. Biorefineries can convert lignocellulosic biomass into high-value products, lessen their environmental effect, and hasten the advent of a sustainable and renewable future by incorporating cutting-edge technologies and doing thorough Life Cycle Assessment analysis. However, converting lignin into added-value products is a challenging field from technological, environmental, and economic perspectives.

This work set out to determine the most critical environmental aspects expected to provide reliable evidence in support of lignin valorisation routes that enable the production of value-added products and bioenergy. Findings provide the necessary knowledge for decision-making in biorefineries aligned with the Circular Bioeconomy concept, like how decision-making relating to future biorefineries is facilitated.

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KEYWORDS
Lignocellulosic biomass; lignin; valorisation; biorefinery; LCA; environmental assessment; Circular Bioeconomy

HIGHLIGHTS

- Environmental burdens of lignin-derived products valorisation due to complex lignin structure
- A limited number of LCA studies on the valorisation of lignin to added-value products
- Future lignin-based biorefineries development should be assessed using holistic, sustainable approaches
- LCA methodologies on environmental performance of promising lignin valorisation routes
- Controversial aspects of lignin valorisation under the Circular Bioeconomy concept
- Raised concerns as a result of the lignin fraction expected high demand
- Identified controversial environmental aspects linking lignin valorisation to adverse impacts

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List of abbreviations

Abbreviations

AP	Acidification potential
CE	Circular economy
CED	Cumulative energy demand
EP	Eutrophication potential
Eq.	Equivalent
EU	European Union
EC	European Commission
FU	Functional unit
GHG	Greenhouse gas
GWP	Global warming potential
ISO	International Organisation for Standardisation
LIBRA	Lignin biorefinery approach
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LHV	Low heating value
MSW	Municipal Solid Waste
ODP	Ozone depletion potential
PAN	Polyacrylonitrile
POCP	Photochemical ozone creation potential
RES	Renewable energy source
SMC	Sheet moulding compound
TBC	Tert-butyl catechol

Symbols/units

$CFC-11$	Trichlorofluoromethane
CO_2	Carbon dioxide
C_2H_4	Ethylene
g	Grams
kg	Kilograms
MJ	Megajoules
PO_4^{-3}	Phosphate ion
SO_2	Sulfur dioxide

1. Introduction

A viable energy source that could aid in the endeavour to reduce greenhouse gas emissions is biomass, which is used efficiently in all three areas of high energy demand – electricity generation, home heating, and transportation (Christoforou and Fokaides 2015); (Zhao 2018). The biodegradable portion of goods, waste, and by-products from biological sources, including those from forestry, agriculture, and other associated industries, as well as the biodegradable portion of municipal and industrial wastes, is referred to as biomass (European Parliament 2009). Energy derived from biomass is seen as renewable since an equivalent amount of biomass grows to replenish it (Schuck 2006), and its significance to national economic frameworks is growing. Additionally, the availability of non-renewable energy sources (RES) is unevenly distributed throughout the world and is constrained in terms of both quantity and diversity (Zeren and Hizarci 2021).

Lignin constitutes one of the most abundant natural polymers, which contains aromatic structures and represents approximately 30% of the total organic carbon in the Earth's biosphere (Strassberger, Tanase, and Rothenberg 2014); (Xu et al. 2014). The black liquor typically contains 30 - 34% of lignin and is the main by-product of the Kraft process; the process that produces 80% of the world's chemical pulp (da Silva et al. 2009); (Saake and Lehnen 2003). For every tonne of pulp, the pulp mills also generate 1.7 - 1.8 tons of black liquor (Tustin 2007). Beyond that, lignin is also expected to form the main constituent of large residual streams in future cellulose ethanol plants and biorefineries (De Wild, van der Laan, and Wilberink 2010). Energy, chemicals, and materials can all be generated at existing biorefineries. Bio-based manufacturing chains can successfully implement the principles of the circular economy, which aim to reduce the amount of waste sent to landfills by encouraging its reuse and valorisation (Atabani et al. 2022) (Hoang 2021). The importance of the circular economy framework in promoting the use of renewable resources and minimising waste in biorefineries is crucial. The role of information technology in optimising the energy consumption and production processes in biorefineries, can contribute to reducing their environmental impact Hoang et al. (2022a).

Chemicals and fuels can be derived from the by-products of pulp mills, which include heat, power, and tall oil. In a traditional pulp mill, the residual product, known as black liquor, is burned in a recovery boiler to generate electricity and heat, and the chemical compounds are recirculated for the cooking process (Elomatic OY). Converting a pulp mill into a biorefinery that produces energy, chemicals, fibres for textiles, and biopolymers is a feasible scenario in the future (Figure 1). Due to its high organic content derived from dissolved lignin and carbohydrates, the black liquor is traditionally combusted in boilers to recover heat and chemicals at pulp mills. However, it has been indicated that such a valuable resource should not be wasted for energy purposes. Nevertheless, it could be exploited in a more sustainable way toward the production of value-added commodities. In general terms, the type of upstream processing used for lignin production defines the value of the products, which could be subsequently obtained (Smolarski 2012). Accordingly, higher-grade lignin, such as Kraft and organosolv lignins, have been shown to generate high-value products; on the other hand, lignosulfonates provide lower-value chemicals (Figure 2). Within the same context, Wild et al. [17] highlight that lignin valorisation is the key issue for developing economic lignocellulosic biorefineries. However, although lignin holds greater potential as a substitute for fossil fuels, current efforts in the biorefinery sector mainly focus on the valorisation of cellulosic and hemicellulosic biomass. This occurs principally due to the complex aromatic structure of lignin, constituting its conversion to value-added products as a challenging endeavour. At the same time, information and data on lignin's industrial availability, sustainable conversion, and applications, as well as pertinent to the environmental performance and economic viability of valorising this complex molecule, is still limited.

The primary objective of this study is to analyze the key environmental factors linked to the utilisation of lignin for the creation of valuable goods. This objective is accomplished by assessing the process of lignin valorisation using both the Life Cycle Assessment (LCA) approach and the

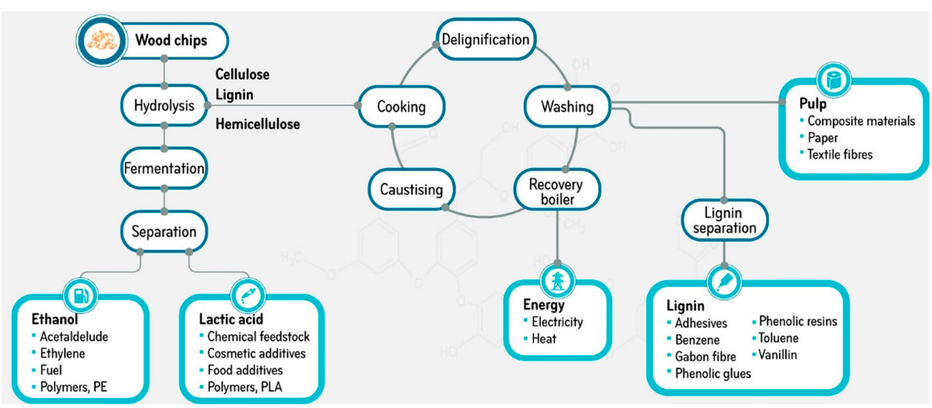

Figure 1. Pulp mill into a biorefinery conversion (reproduced diagram by Arkola H. (Elomatic OY)).

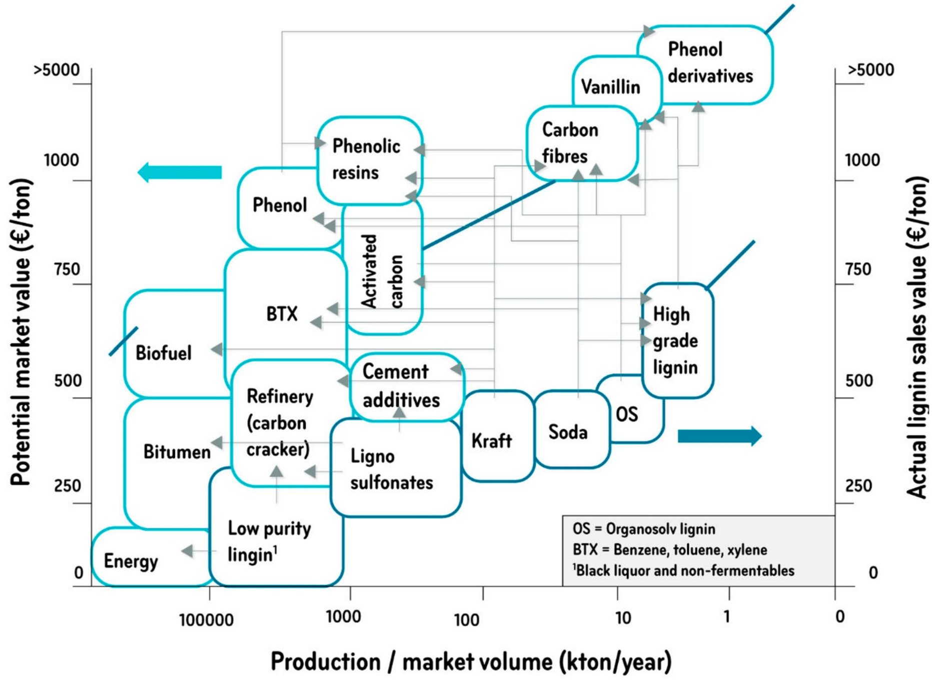

Figure 2. Lignin production and potential lignin-derived product market and value (reproduced diagram by (Gosselink 2011)).

Circular Bioeconomy framework. By applying the LCA methodology, the study aims to identify and understand the environmental impacts associated with lignin valorisation. Additionally, the research seeks to explore the potential of lignin as a sustainable resource within the context of the Circular Bioeconomy, considering its role in reducing waste and promoting resource efficiency. Section 2 provides background information on lignin, elaborating on its potential for conversion into value-added products, chemicals, and fuels, as well as describes the principles of conducting LCA studies in accordance with the ISO 14040 (2006a) series. A comprehensive review of the current state-of-the-art on the LCA of lignin valorisation routes is provided in Section 3, which also allows for defining the challenges met when conducting such a study. Section 4 introduces the relatively new Circular Bioeconomy concept and critically discusses whether lignin valorisation fits under this sustainable platform. Finally, the environmental considerations surrounding the valorisation of lignin are summarised in Section 5. In addition, the final deliberation on how lignin valorisation routes can contribute to fulfilling the European vision of Circular Bioeconomy is provided. This work provides valuable insights into the environmental performance of lignin valorisation, such as facilitating the decision-making relating to future biorefineries.

This study presents innovative aspects related to the valorisation of lignin, which include an emphasis on life cycle assessment (LCA) advancements and a focus on identifying the most critical environmental aspects that support the production of value-added products and bioenergy from lignocellulosic biomass. The approach taken in this study differs from previous studies on lignin valorisation as it specifically evaluates the environmental impacts of various lignin valorisation methods and provides a framework for decision-making in biorefineries aligned with the Circular Bioeconomy concept. Through this study, the authors provide valuable insights into the environmental and economic aspects of lignin valorisation, which can assist in the development of sustainable and efficient biorefinery operations. The findings from this analysis can also aid in the selection of appropriate lignin valorisation routes that maximise the value of lignocellulosic biomass while minimising its environmental impact.

2. Methodology

This study provides a comprehensive overview of the current state of knowledge regarding the LCA assessment of lignin, and identified areas for future research. The systematic review methodology allowed us to identify and analyze the most relevant and reliable sources of information, and provided a structured approach for synthesising the findings of the selected studies. The results of this study can be used to inform the development of sustainable lignin-based products and processes. Life cycle assessment (LCA) is a widely used methodology for the evaluation of the environmental impacts of a product, process, or service. The purpose of this study was to provide an overview of the current state of knowledge regarding the environmental impact of lignin production, extraction, and utilisation, and to identify areas for future research.

To achieve this goal, we conducted a systematic review of the literature on LCA assessments of lignin. The review was carried out according to the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines, which provide a framework for conducting and reporting systematic reviews. The sources were chosen based on specific criteria, including relevance to the topic, scientific rigour, and reliability of the data. The search was conducted using multiple electronic databases, including Web of Science, Scopus, and Google Scholar, with the following search terms: "lignin," "life cycle assessment," "environmental impact," "sustainability," and "renewable energy." The search was limited to articles published in the last 10 years, written in English, and focused on LCA assessments of lignin. After screening the titles and abstracts of the articles identified in the search, we excluded articles that were not relevant to our study, duplicates, and articles that did not meet the inclusion criteria. The full texts of the remaining articles were then reviewed in detail, and data relevant to the LCA assessment of lignin were extracted. The extracted data were then analyzed using a narrative synthesis approach, which involved summarising and synthesising the findings from the selected articles. The synthesis was structured around the main stages of the lignin life cycle, including production, extraction, and utilisation, and the environmental impact categories included in the LCA assessments.

The results of the review provided insights into the environmental impact of lignin production, extraction, and utilisation, and identified areas for future research. The study found that the environmental impact of lignin production and utilisation varies depending on the specific process and application, and that the use of lignin as a substitute for fossil-based materials can have a positive impact on reducing greenhouse gas emissions and improving resource efficiency.

3. Background information

3.1. Lignin valorisation

Biorefinery concepts should consider polysaccharides and lignin as equally important feedstocks for valorisation (Beckham et al. 2016). The upstream industrial processes required for raw biomass pretreatment are well-documented, producing high-purity lignin, but can additionally degrade the biopolymer into smaller molecules and result in chemical/structural transformations (Behling, Valange, and Chatel 2016). The European 2009/28/EC Directive (European Parliament 2009) on the development of RES, which aims for a share of 20% of the European energy demand to be met by RES, encourages the promotion of biomass. However, in order to identify and establish the best alternative, systematic methodologies must be used when making political decisions pertaining to the energy and environmental sectors (Kylili et al. 2016).

Various extraction techniques (enzymatic, chemical, and mechanical) can separate lignin, which results in intermediate by-products, including binders, carbon bres, dispersants, phenols, and plastics (Agrawal, Kaushik, and Biswas 2014). Chemical or steam treatment under high pressure is the standard method for removing lignin from plants (Wang et al. 2018). Vanillin (3-methoxy-4-hydroxybenzaldehyde) is one of the most popular flavouring compounds that has use in the culinary, fragrance, and medical fields since it is derived naturally from the vanilla plant's dried pods (da Silva et al. 2009). More than 90% of all carbon fibre in the world comes from polyacrylonitrile, made from petroleum. Carbon fibres may be created from lignin, and studies have demonstrated that they have the same qualities as carbon fibres made from petroleum-based precursors (Norberg et al. 2013) (Langholtz et al. 2014). Tanners, along with the chemical, pharmaceutical, food, and perfume sectors, stand to benefit from the phenolic compounds (Kleinert and Barth 2008). Phenolic resins are a very adaptable type of thermosetting polymers that find widespread application in many different fields, including construction, transportation, and electronics (Ghaffar and Fan 2014); lignin can partially replace phenol in this process (Zhao et al. 2016). Adhesives, aromatic chemicals, bioplastics, fertilizres, and biofuels are only some of the other products that could be created using lignin (Duval and Lawoko 2014) (Bi et al. 2018). Overall, lignin is an adaptable and important resource with many possible uses. Studies are now being conducted to uncover even more innovative methods for placing this complex polymer into practical use.

The types of lignin with the most applications include Kraft, Lignosulfonated, Organosolv, Pyrolytic, Steam Explosion, and Acidolysis lignin, where the isolated fractions demonstrate a range of important differences (e.g., intersubunit linkages, molecular weight, solubility) that negatively impact potential lignin valorisation methods (Ma, Xu, and Zhang 2015). The Sulphite process uses an aqueous solution of sulfur dioxide with different pH for obtaining lignins, which need further purification (e.g., by fermentation, chemical removal, ultrafiltration) for carbohydrate impurities removal. Soda lignin is produced through a dissolution of lignin from lignocellulosic biomass using sodium hydroxide and product recovery via acid precipitation, maturation, and filtration. The Kraft process employs sodium hydroxide and sodium sulfide solutions as the white liquor in which wood chips are boiled, allowing hydroxide and hydrosulphide anions to depolymerise lignin into smaller water/alkali-soluble fragments. The LignoBoost technology improves this process by lowering the pH with the use of CO_2 precipitating lignin from the black liquor. Although the Organosolv fractionation with ethanol–water has not yet been commercialised, this process yields high-purity lignin with <1 wt% residual carbohydrate content (Strassberger, Tanase, and Rothenberg 2014).

Acid-based pre-treatment of biomass has been shown to be effective in removing inorganic compounds and improving the subsequent pyrolysis conversion and product quality (Kumar et al. 2020) (Hoang et al. 2021) (Wang et al. 2015). Process design, quality, and safety can all be compromised by the corrosive nature of water and steam due to pressure fluctuations, acidic and oxidising environments, and high pH values (Pavlovic, Knez, and Skerget 2013). Maximum efficiency in a lignocellulose processing facility requires using all of the primary components of biomass (Chen et al. 2022). Hexoses, pentoses, and lignin should all be put to good use in value-added applications.

The process of lignin valorisation is accounted for in the downstream processing of lignin's life cycle, which includes depolymerisation and chemical modification of isolated lignin streams into commodity chemicals, fuels, and materials (Rinaldi et al. 2016). The most promising long-term sustainability technique is converting lignin into biopolymers and other high-value substances (Bilal et al. 2022). Epoxies, polyesters, polyurethanes, phenol resins, polyhydroxyalkanoates, poly(lactic acids), and many other important biopolymers have all benefited from advancements in this technology (Figure 3). However, although exploitation of lignin comprises a necessary approach for the development of a sustainable, cost-effective, and green bio-based industry, lignin biorefineries have not yet been established in commercial practice, and valorisation of the aromatic fraction still remains an active field of research (Chen and Wan 2017). The recovery and purification of lignin hamper industrial application, the complexity, and heterogeneity of lignin's aromatic structure and composition, as well as the reactivity of lignins which comprise major challenges for conversion of the macromolecule to value-added products (Xu et al. 2014). Thus, even though past research has concentrated on lignin conversion into valuable commodities, very little of this research has yielded commercial applications (Ragauskas et al. 2014). Current lignin applications comprise the production of (i) fuel, syngas, and power, (ii) macromolecules, and (iii) aromatic molecules (Yuan,

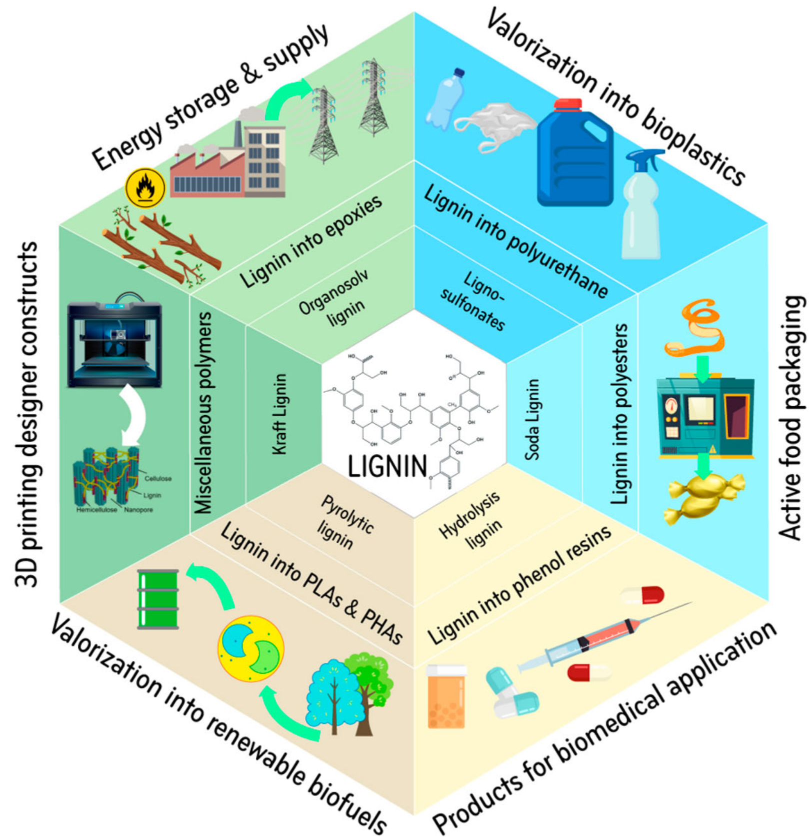

Figure 3. Lignin valorisation into different high-value products for diverse applications (reproduced diagram by (Bilal et al. 2022)).

Xu, and Sun 2013). However, intensive research has been conducted using a wide range of catalytic, thermal, and biological methods aimed at breaking down the biopolymer and utilising the monomers formed to generate added-value products (Beckham et al. 2016).

Thermochemical depolymerisation methods, such as hydrothermal carbonisation, pyrolysis, and gasification, have been widely applied for lignin conversion into valuable chemicals and fuels (Liu, Jiang, and Yu 2015). In comparison to other processes, such as gasification and combustion, followed by secondary gas-phase cracking and/or oxidation reactions to generate gaseous products, pyrolysis is in the leading phase (Hoang et al. 2021) (Hoang et al. 2022b). Brebu, Tamminen, and Spiridon (2013) found that bio-oil quality significantly improved. Converting a biomass-fast pyrolysis facility into a combined heat and power plant is a different potential approach that could boost pyrolysis's appeal as a commercially viable (Solantausta et al. 2013) and environmentally friendly means of producing energy. In particular, the latter part of the aforementioned assertion is supported by LCA modelling results showing a significant reduction in CO_2 emissions (72–99%) when compared with conventional fossil-based techniques for electricity production using the same integrated pyrolysis system (Iribarren, Peters, and Dufour 2012) (Fan et al. 2011). De Wild et al. (2009) studied pyrolysis of two different lignins, prepared from a mixture of hardwoods by the Alcell organosolv process as well as from grass and straw through soda pulping, in a bubbling fluidised bed at 400̊C. The results obtained indicate that the low molecular weight compounds formed (e.g., guaiacols, syringols, catechols) have economic potential as petrochemical substitution options for a range of commodities, including wood adhesives, bio-plastics, and pharmaceuticals.

Nevertheless, Beis et al. (2010) concluded that using different lignin feedstocks, kinetic parameters, and chemical composition vary substantially, and fast pyrolysis processes must be specified between different types of lignin. Moreover, a pyrolysis-based lignin biorefinery approach (LIBRA) was developed for lignin transformation into phenolic bio-oil and biochar using a bubbling fluidised bed reactor (De Wild, Huijgen, and Heeres 2012). The study suggested that bio-oil could replace petrochemical phenol in wood adhesives, resins, and polymer applications, while biochar could be used as fuel, soil-improver, solid bitumen additive, and precursor for activated carbon.

Fractionation of biomass constituents into lignocellulosic streams (e.g., glucose, lignin oil) enables valorisation through various approaches that often include catalytic processes (Galkin and Samec 2016). For lignin valorisation to aromatics, the refinery separates biomass into cellulose specialty fibres, subsequently fermented to second-generation bioethanol, and lignosulfonates used for vanillin production. Besides vanillin, lignin oxidation with molecular oxygen produces syringaldehyde and various aromatic aldehydes, while the majority of studies focus on understanding and improving the different catalytic processes for chemicals' production (Strassberger, Tanase, and Rothenberg 2014). In the work of Gosselink et al. (2012), organosolv hardwood and wheat straw lignins were converted in a supercritical fluid composed of carbon dioxide/acetone/water to phenolic oil (containing oligomeric and monomeric aromatics). The results suggested that competition occurred between lignin depolymerisation and recondensation of fragments. Furthermore, Díaz-Urrutia et al. (2015) compared the performance of three catalysts, namely homogeneous oxovanadium, copper, and cobalt complexes, in oxidation and depolymerisation of organosolv lignin, demonstrating that oxovanadium (V) complexes were the most selective and productive homogeneous oxidation catalysts for lignin valorisation. Although lignosulfonates are already used commercially in many applications (e.g., cement, polymers, resins), the valorisation of high-purity lignin can also produce lignin-derived carbon fibre, an alternative for polymers such as polyacrylonitrile (PAN). Moreover, da Silva et al. (2009) suggested an integrated process that includes reaction and separation steps for producing both vanillin – and lignin-based polyurethanes from Kraft lignin. Gosselink et al. (2011) performed catalytic pyrolysis and supercritical conversion of organosolv lignin, demonstrating that the use of end products from this process, in particular, phenolic oil for wood adhesives and bitumen, together with the gases for fuel, provides realistic scenarios for lignin valorisation in a future biorefinery.

Biological valorisation could be applied as a sustainable approach for lignin upgrading. White-rot fungi have been extensively studied for their capacity to efficiently degrade lignin, while the conversion of the biopolymer could also be achieved *in vitro* using laccases and peroxidases (Chen and Wan 2017). Bugg and Rahmanpour (2015) presented the advancements in using bacterial enzymes for converting lignin to renewable chemicals via fermentation and the potential for novel applications. However, although different bacteria have evolved catabolic pathways for lignin degradation and valorisation, common challenges such as low yield and productivity should be tackled for industrial application (Tian et al. 2014). Some natural strains can convert multiple aromatic species into one or two intermediates funnelled into central metabolism using a process known as 'biological funnelling' (Beckham et al. 2016). This topic has recently gained substantial interest as a viable biological approach for overcoming the major challenge of lignin heterogeneity, hampering the valorisation of this complex molecule to added-value commodities.

Lignin isolation methods aim to separate lignin, a complex biopolymer found in plant cell walls, from other components like cellulose and hemicellulose. A summary of commonly employed lignin isolation methods:

- Alkaline extraction involves treating lignocellulosic biomass with alkaline solutions, such as sodium hydroxide (NaOH) or ammonium hydroxide (NH4OH). The alkaline solution breaks down lignin-carbohydrate bonds, dissolving lignin while leaving cellulose and hemicellulose behind. Further acidification or precipitation is often performed to obtain lignin in solid form (Sun et al. 2021).

- Acidic hydrolysis involves subjecting lignocellulosic materials to acid treatment, commonly using sulfuric acid (H2SO4) or hydrochloric acid (HCl). The acid breaks down the polysaccharides, while lignin remains relatively unaffected. After hydrolysis, lignin can be recovered by filtration or solvent extraction (Huang et al. 2022).
- Organosolv process utilises organic solvents, such as ethanol, methanol, or a mixture of organic solvents, to selectively dissolve lignin from the biomass. The process often involves heating the biomass with the solvent in the presence of an acid catalyst. The resulting solution is then separated, and lignin can be recovered by evaporating the solvent (Parot, Rodrigue, and Stevanovic 2022).
- In steam explosion method, lignocellulosic biomass is exposed to high-pressure steam followed by rapid decompression. The sudden release of pressure causes the cellulosic fibres to rupture, facilitating the separation of lignin from cellulose and hemicellulose. Lignin can be recovered by washing, filtration, or subsequent solvent extraction (Akizuki et al. 2023).
- Enzymatic approaches use lignin-degrading enzymes, such as lignin peroxidase or laccase, to selectively degrade lignin. These enzymes break down the lignin structure, allowing for its separation from cellulose and hemicellulose. Enzymatic methods are often combined with other extraction techniques for more efficient lignin isolation (Paul et al. 2023).

3.2. Principles of Life Cycle Assessment

LCA comprises the most valuable and effective framework for assessing any product's or system's environmental impact throughout its life cycle. This systematic methodology, as specified by the international standards ISO 14040 (2006a) and ISO 14044 (2006b), can facilitate the decision-making regarding important life stages and processes with significant impact (positive or negative) on the overall environmental performance of the object under investigation. In line with this, LCA can assist in identifying opportunities for improvement of the environmental performance of the value chain.

An LCA study is distinguished into four phases: the goal and scope definition, the life cycle inventory (LCI) analysis, the life cycle impact assessment (LCIA), and the results interpretation phase. The first phase introduces the intended purpose of conducting the study and defines important parameters, such as the system boundaries, the functional unit (FU), the impact categories to be investigated, the relevant scenarios to be examined, any allocation methods used, and any assumptions made. LCI is an inventory of the input and output data that are relevant to the conduction of the study, including energy and materials required or released into the environment throughout the investigated object's whole life cycle. The key objective of the LCIA phase is to indicate the environmental significance of the inputs and outputs of the study quantitatively in terms of the potential of environmental impact categories. Categories include, but are not limited to, climate change, acidification, eutrophication, abiotic depletion of elements and fossil fuels, ozone depletion and photochemical ozone creation, and land, aquatic and human toxicity. In the final phase, the study's results are summarised and discussed as a basis for conclusions, recommendations, and decision-making in relevance to the intended scope of the study.

4. Life Cycle Assessment of lignin valorisation

4.1. Current state-of-the-art on the life cycle performance of lignin valorisation

The number of LCA studies on the valorisation of lignin to added-value products is currently very limited, resulting in a lack of validated information and quantified data on the environmental performance of lignin exploitation. In the up-to-date literature, the environmental and carbon emissions/trade-off assessment studies involve the valorisation of lignin into vanillin, epoxy resins, catechols, carbon fibre precursors, and soil amendments (Modahl and Vold 2011); (Llorach-Massana et al. 2017).

A work worth focusing on is the study of Modahl and Vold (2011) on the cradle-to-gate LCA of cellulose, ethanol, lignin, and vanillin of the Borregaard factories in Norway, products among which vanillin has the highest market value potential. The FU of this work was defined as one tonne of each useful product and 1 m^3 of ethanol in the specific case, while the analysis also included infrastructure, transport, and waste combustion. Given the scope of this study, it is admittedly the most comprehensive work found in the current literature. The LCIA results for this work's products of interest are provided in Table 1. The study demonstrated that vanillin includes the highest burdens, followed by cellulose, powder, and liquid lignin. Vanillin has the highest potential impact among the five investigated environmental categories, mostly due to high energy and chemical consumption. In contrast, the difference between liquid and powder lignin is that no energy is used to remove the water from liquid lignin. However, the uptake of these findings should also take into consideration that vanillin holds a 20 times higher market price than cellulose (Modahl and Vold 2011). The results also indicated that the energy used for the production and transportation of raw materials as well as internal processes at Borregaard is the most significant source of environmental burden, while infrastructure and transport to customers are insignificant – accounting for less than 3% of the total impact (except in the waste category). The study concludes that reducing the energy use at Borregaard will lower the impact across the majority of the categories; however, further work needs to be done to draw conclusions on whether environmental burdens will be reduced with the use of renewable energy. Furthermore, it is true that as long as biomass resources (forest wood and agricultural products) are readily accessible and near the establishment, biomass disposal and transportation costs stay stable, ensuring the long-term viability of such energy systems (Vallios, Tsoutsos, and Papadakis 2016).

Kosbar et al. (2000) employed cradle-to-grave LCA to compare the energy consumption and waste generated by manufacturing and using lignin/epoxy resins versus conventional epoxy resins. The findings indicated a 40% reduction in fuel usage for lignin-based resin production and manufacture due to lower energy requirements for raw materials production. Additionally, incineration of the board for disposal purposes produces reduced levels of greenhouse gases (GHG). At the same time, the boards disposed of at landfills could be easier to biodegrade due to the presence of fungi that break down lignin.

A comparative LCA study of tert-butyl catechol (TBC) from lignin against conventionally derived TBC from petrochemical phenol was performed by Montazeri and Eckelman (2016). The lignin-derived product indicated a reduction of 2%, 7%, and 59% in global warming potential, ecotoxicity effects, and fossil fuel depletion, respectively, while increases were noted in the rest of the categories investigated. The authors noted that the solvent used for lignin purification and the electricity used for lignin depolymerisation are greatly responsible for the negative environmental impact. Therefore, research should be directed towards alternative conversion processes for establishing more sustainable bio-based routes.

In the study of Das (2011), the life cycle energy and environmental impacts of alternative carbon fibre precursor materials (textile-type acrylic fibres and lignin) and production technologies

Table 1. Environmental burdens from cradle-to-gate for Borregaard's products (values obtained from Modahl and Vold (2011)).

Environmental impact category	Units	Lignin (liquid)	Lignin (powder)	Vanillin
Global Warming Potential (GWP)	$kg\ CO_2 - Eq.$	704	1227	1343
Acidification Potential (AP)	$kg\ SO_2 - Eq.$	7.1	10.4	11.7
Eutrophication Potential (EP)	$kg\ PO_4^{-3} - Eq.$	1.64	2.75	2.47
Photochemical Ozone Creation Potential (POCP)	$kg\ C_2H_4 - Eq.$	0.42	0.69	0.76
Ozone Depletion Potential (ODP)	$kg\ CFC - 11 - Eq$	4.3E-5	1,1E-4	9.7E-5
Cumulative Energy Demand (CED)	*MJ LHV*	18200	31500	36500
Waste	*kg waste*	37.8	59.6	82.8

(programmable powdered preforming process – P4 and sheet moulding compound (SMC)) were assessed for the production of a 30.8kg steel floor pan. The results showed that lignin carbon fibres have the potential to be 5% less energy intensive and to emit 22% less CO_2-equivalent GHG emissions as compared to conventional PAN-based textile grade acrylic fibres, without even accounting for the no energy consumption for lignin production, since it occurs as a by-product of the pulp and paper industry.

Pourhashem et al. (2013) and Llorach-Massana et al. (2017) investigated the potential of lignin as a soil amendment. In particular, the work of Pourhashem et al. (2013) examined life cycle GHG emission and techno-economic cost trade-offs for three alternative scenarios: the use of lignin as a soil amendment; lignin separation, drying, and commercialisation as a coal substitute; and lignin's conventional use for onsite electricity generation at biorefineries. The analysis revealed that among the three options examined, the scenario of using lignin as a soil amendment is the most economically and environmentally preferable, with the lowest GHG abatement costs. In particular, the carbon intensities of the three scenarios are: −25 – −2 g CO_2-Eq. MJ^{-1}; 4 – 32 g CO_2-Eq. MJ^{-1}; and 36 – 41 to −2 g CO_2-Eq. MJ^{-1}, respectively. Llorach-Massana et al. (2017) also showed that there is great potential to valorise tomato plant residues for biochar production due to the high lignin content included (19.7%) that produces higher yields of biochar with higher $\%_{Stable\text{-}C}$ when pyrolysed at low temperatures. However, the study indicated that percentages higher than 50 $\%_{Stable\text{-}C}$ in the biochar produced are required to ensure carbon sink with biochar production using urban agricultural feedstocks in pilot-scale plants.

4.2. Challenges in Life Cycle Assessment of lignin valorisation

Following the review of existing studies on the LCA of lignin valorisation, a number of challenges have been identified in relation to the practical implementation of such studies:

- The reason lignin valorisation is not yet considered an established process constitutes also the greatest challenge when conducting an LCA study on lignin valorisation. Different lignin resources are expected to include varying types of lignin polymers, which do not always result in the same targeted products and conversion efficiencies under the same process (Montazeri and Eckelman 2016). Therefore, the outcomes of an LCA study are also highly dependent on the production yield of target compounds and the conversion processes employed (González-García et al. 2016).
- At the industrial scale, liquid and powder lignin production occurs simultaneously and within the same process. The challenge to modelling this process in LCA is that it generates several different environmental profiles. To overcome this problem, Modahl and Vold (2011) modelled the production of the two types of lignin as two separate processes.
- Where LCA studies are conducted for quantification of the environmental and financial benefits that may arise from the alternative uses of lignin, the need for estimates to define the market value of a promising product provides a level of uncertainty in the results obtained (Pourhashem et al. 2013).
- Another challenge commonly met in various LCA studies on lignin valorisation is the data acquisition for the LCI (Kosbar et al. 2000). As in this case, the relevant stakeholders are not motivated to document all the details of the system's material and energy flows and/or are hesitant to share (confidential) information about their processes. The authors of this work also highlighted the necessity of financial incentives or external pressures, such as government regulations or the public interest, for the conduction of LCA studies that provide consistent and reliable results.

5. Sustainability considerations for enabling Circular Bioeconomy

5.1. Definition of Circular Bioeconomy

Circular Bioeconomy is defined as the intersection of bioeconomy and circular economy, as shown in Figure 4; building on the synergies of the two concepts (Hetemäki et al. 2017); (Newton et al. 2017). Although the two concepts have been developed in parallel until now, political and scientific communities encourage the merger or standardisation of the two concepts to address the challenges of sustainable development more efficiently.

The European Commission's Expert Group Report on the Review of the European Union (EU) Bioeconomy Strategy and its Action Plan thoroughly defines the interface between Bioeconomy, Cascading use, and Circular Economy (Newton et al. 2017). Bioeconomy covers all sectors and systems that rely on biological resources such as animals, plants, micro-organisms, derived biomass, and organic waste, their functions, and principles (EC 2018). It primarily addresses the production of renewable biological resources and their conversion into added-value products, ranging from food and feed to bio-based products and bio-energy (Newton et al. 2017). Then again, the Circular Economy (CE) attempts to close the loop in the life cycle of systems. It is conceptualised as an economy where the products, materials, and resources' value is maintained within the economy for as long as possible, such as minimising waste (EC 2015). The ideas of the CE are most effective in interconnected industrial ecosystems, where the by-products of one business can be used as resources by another. Wood can be used in novel ways, and these uses can be made possible through networking. A key principle of the CE is the prolonged use of existing raw materials. The value of these resources is preserved to the greatest extent feasible, emissions and waste are reduced, and the ability of nature to regenerate is guaranteed (Figure 5). To put it simply, the bioeconomy is a resource-efficient economy that makes use of renewable materials. The carbon dioxide in the atmosphere is removed from the forest's molecules through natural, regenerative processes. The climate would benefit greatly from a wood-based circular economy. Many different strategies are

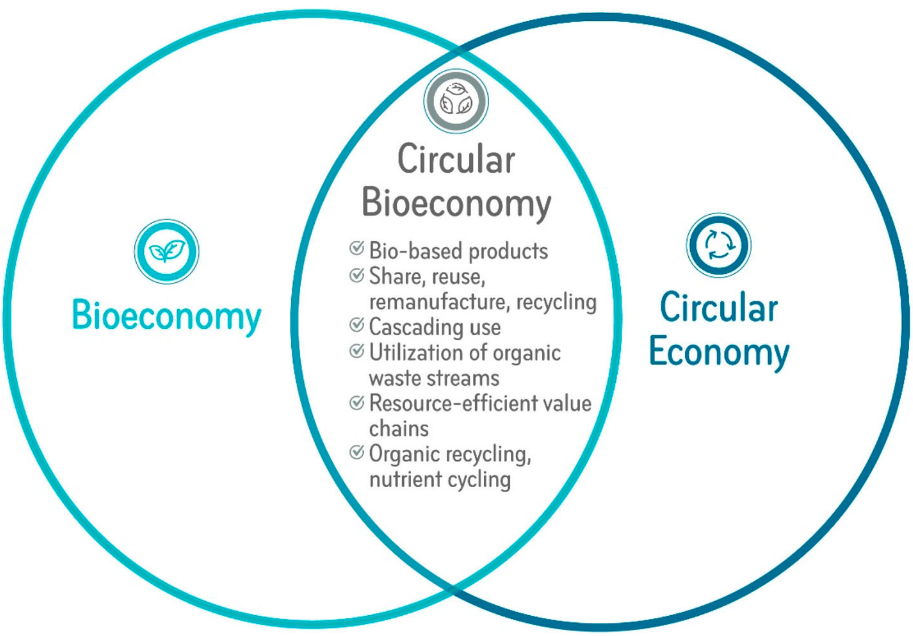

Figure 4. Circular Bioeconomy (reproduced diagram by (Newton et al. 2017)).

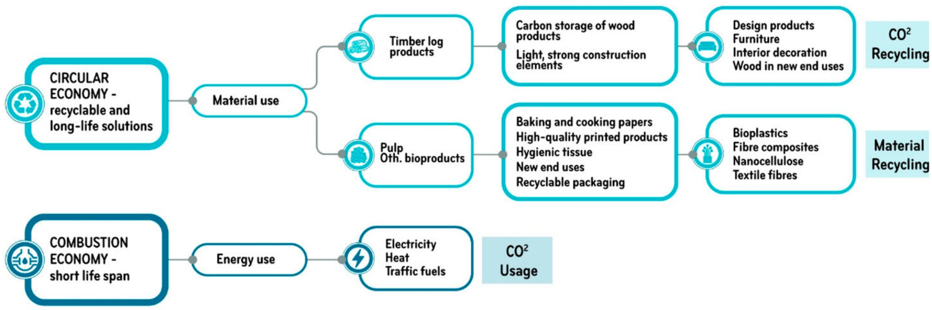

Figure 5. Material use of wood creates the highest value (reproduced diagram by Metsä Group's material).

being employed to combat climate change, including the use of carbon sinks and storage and the substitution of renewable materials for those derived from fossil fuels.

Both concepts incorporate a number of differences, although they share common objectives, complementary approaches, and they contribute to one another, with reference to improved use of resources, eco-efficiency, low GHG emissions, reduced demand for fossil-fuelled carbon, and valorisation of waste and side streams (Newton et al. 2017). Several material flows have not yet been included in the CE concept. At the same time, some others, including carbon utilisation, detergents, cosmetics, coating, and paints, cannot be part of the CE concept, given that collection or recycling of these materials is not feasible. Furthermore, the Bioeconomy is concerned with innovation using biological resources, such as the development of new chemical building blocks, new processing routes, and new functionalities and properties of products. At the same time, CE targets the maximisation of the products' life cycle under an industrial urban process perspective (Newton et al. 2017); (D'Amato et al. 2017). D'Amato et al. (2017) conducted a bibliometric review of almost two thousand scientific articles on Bioeconomy, Circular Economy, and Green Economy, concluding with significant suggestions regarding future research in the field as well as policy priorities, highlighting the need for further analysis of synergies and divergences among the various concepts with their harmonisation being the ultimate goal. This is in agreement with the work of Hetemäki et al. (2017), where establishing the connection between Bioeconomy and CE was deemed crucial for achieving significant societal objectives.

Given the differences between Bioeconomy and CE, several challenges and limitations arise with adopting the Circular Bioeconomy concept. Zabaniotou (2018) foresaw challenges in the bioenergy sector as a result of the new demand for bio-materials through the use of the same sources. The author highlighted the requirement for selecting bioenergy pathways based on ecoefficiency and to what level 'closing of the loop' has been achieved while taking into consideration cascading use. Cascading use is known to maximise resource effectiveness by applying residues and recycled materials to extend biomass availability within a given system; however, in some systems, the energy required for providing biomass multiple lifetimes cannot be justified in terms of GHG footprint.

The impacts of biobased products and cascading in biobased pathways should be assessed in combination with LCA methodologies (Newton et al. 2017); (Mohan et al. 2016). Moreover, another limitation of cascading that should be considered when assessing biobased pathways is the accumulation of toxic substances, which may prohibit further recycling or incineration of a material (Newton et al. 2017).

Extensive research on the benefits and challenges of the Circular Bioeconomy concept led to the update of Europe's Bioeconomy Strategy in 2012 (EC 2012). The Updated Bioeconomy Strategy specifies that a thriving European bioeconomy requires being both sustainable and circular, specifying 14 concrete actions expected to indicate the approach that should be adopted toward fulfilling this objective (EC 2018). The main action areas proposed by the Updated Bioeconomy Strategy include:

- strengthening and scale-up of biobased sectors, unlocking investments and markets;
- rapid deployment of local bioeconomies across Europe;
- understanding the ecological boundaries of bioeconomy.

5.2. Lignin valorisation under the context of Circular Bioeconomy

New technological innovations have enabled the production of value-added products from waste, and side steams of the food and feed, forest, and marine industries. As a co-product of the pulp and paper industry and the great potential of lignin in increasing its added value through conversion into aromatics, polymers, high-performance materials, and composites, this important molecule holds a significant position in the novel concept of Circular Bioeconomy. The forest industry is anticipated to allow for the utilisation and cascading of the highly efficient lignin-containing side-stream due to well-developed infrastructure and know-how in cascading paper (Newton et al. 2017). Proper forest management leads to increased forest growth and the ability to absorb more carbon dioxide. As forests expand, their ability to slow global warming increases because more trees are available to absorb more carbon dioxide as they are produced (Metsä Group 2019). With sustainable forestry practices in place, not only is biodiversity protected, but so is the recreational use of the very same forests that supply the raw materials for bioproducts. In Figure 6 is depicted the forest life cycle. An example of lignin for valorisation potential is demonstrated by the Borregaard factories in Norway, where some of the chemical structures of lignin are intentionally conserved in the factories' production processes (Antikainen et al. 2017).

The feedstock required in lignin valorisation processes is known as the only renewable source for industrial aromatics production that does not compete with food production and is available in large quantities (Gosselink et al. 2011); (Holladay et al. 2007). Lignin represents 30% of the non-fossil organic carbon found within the biosphere (Strassberger, Tanase, and Rothenberg 2014); (Xu et al. 2014). Indeed, LCA studies have validated the contribution of lignin-based products to sustainable development in terms of energy usage, solid wastes, and air – and water-borne emissions (Kosbar et al. 2000). Under the Circular Bioeconomy perspective, lignin exploitation offers significant environmental benefits but also stimulates controversial aspects, including processes that fit under the Bioeconomy concept but fail to satisfy the essence of the Circular Economy, which is the closing of the loop. Key considerations that should be taken into account in developing future biorefineries include wastewater treatment, leaching, transportation, and land use. Biomass

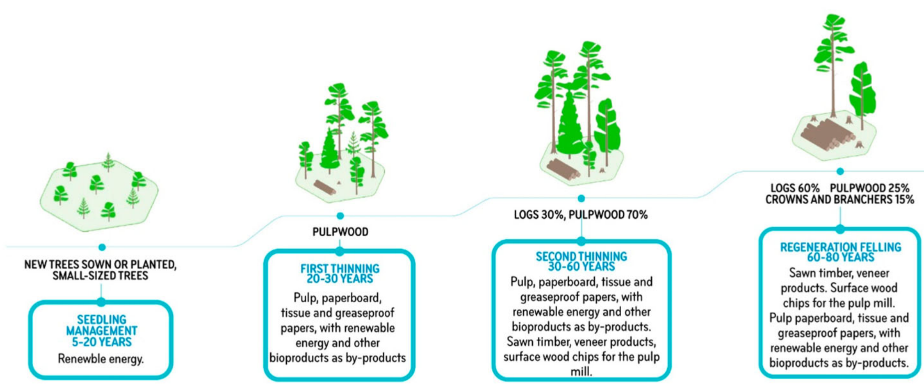

Figure 6. Forest life cycle (reproduced diagram by (Metsä Group 2019)).

utilisation is typically associated with wastewater treatment challenges, while concerns are also raised with relevance to the end-of-life management of lignin-based products, potentially polluting the aquatic environment (Bernier, Lavigne, and Robidoux 2013). Atmospheric pollution is also closely linked to biomass exploitation due to the need to transport large quantities of biomass to processing plants. Strassberger, Tanase, and Rothenberg (2014) suggested that the operation of future biorefineries processing lignocellulosic biomass should be based on a decentralised model, employing small and flexible units to overcome the specific challenge. The work of Montazeri and Eckelman (2016) also expressed concerns regarding the impact of the anticipated high demand for the lignin fraction to produce added-value products, which may lead to direct and indirect land use changes. According to Muritala and Adewole's most recent work (2022) on lignocellulose biomass, the potential value of lignocellulose biomass in microbial fuel cells for energy production has not been fully realised, particularly in developing countries. As a result of their study, they have also concluded that technological development in this area should be amplified to accelerate the commercialisation of lignocellulosic-based microbial fuel cells.

Considering all these aspects, it is concluded that lignin valorisation to added-value products should be assessed using holistic, sustainable approaches, accounting for direct and indirect impacts across all levels – economic, environmental, and societal – and across the whole life cycle. In addition, given the complexity of lignin's aromatic structure, the evaluation and decision-making regarding the development of a lignin valorisation process should be performed for each case separately, considering the specified detailed facts and data. Moreover, given that the Circular Bioeconomy concept is relatively new, several barriers exist not only from technological and environmental perspectives but also from a policy standpoint. It is a fact that the current bioenergy and biofuel policies favour the energetic use of biomass as compared to cascading (Carus, Dammer, and Essel 2015) (Newton et al. 2017). Thus, although countries such as Finland and Sweden are placed in the most favourable positions for benefiting from the adoption of a European Circular Bioeconomy, energy production from black liquor still plays a significant role in the overall energy mix and the fulfilment of renewable energy targets in these countries (Antikainen et al. 2017).

6. Conclusions

Undoubtedly, the potential of lignin to raise the sustainability level of future biorefineries is enormous. However, information and data on industrial availability, sustainable conversion routes and applications, environmental performance, and economic viability are still limited. The main objective of this work was to identify critical environmental aspects associated with lignin valorisation for the production of useful commodities. Therefore, a comprehensive review of studies employing LCA methodologies was conducted to examine the environmental performance of promising lignin valorisation routes. The review indicated that although bio-based processes of lignin-derived products can achieve small reductions in terms of GHG emissions, global warming, fossil fuel depletion, and ecotoxicity compared to conventional products' manufacturing, in the majority of impact categories, environmental burdens increase. The driver for these negative impacts is the complex lignin structure that requires intensive conditions and strong solvents for depolymerisation and further processing of value-added commodities. Therefore, it can be deduced that establishing optimal processes from a technological point of view allows the conversion of lignin into added-value bio-products, bio-chemicals, or bio-fuels and is also expected to deliver consistent and quantifiable data on the sustainability and environmental performance of lignin-based biorefineries. Yet, the development of optimal lignin processes is not the only challenge associated with the conduction of LCA studies on lignin valorisation, some of which were also identified in this work.

In support of the adoption of lignin bio-based routes for the production of value-added products, the compatibility of lignin valorisation with the Circular Bioeconomy concept is evaluated. Following a thorough description of the principles underlying this relatively new concept, specific environmental aspects pertinent to lignin valorisation that should be examined with caution when developing lignin-based biorefineries were identified. Despite the environmental benefits arising from lignin exploitation – an abundant renewable aromatic resource that does not compete with food – this work has identified controversial environmental aspects linking lignin valorisation to adverse impacts from wastewater treatment and leaching, biomass transportation, and land use change. In particular, concerns were raised regarding the pollution of the aquatic environment from the end-of-life management of lignin-based products, the atmospheric pollution occurring from the need to transport large quantities of biomass to processing plants over long distances, and the impacts of direct and indirect land use changes as a result of the expected high demand of the lignin fraction. Despite the fact that lignin has been demonstrated to have numerous potential uses, there are still difficulties to overcome in terms of optimising manufacturing methods, guaranteeing product consistency and quality, and identifying the most economically viable applications. Exploring the latest and most environmentally responsible means to use this resource necessitates continuous innovation and collaboration between academics, industry, and government in order to ensure a sustainable future through lignin utilisation. This work encourages the development of lignin-based biorefineries that are anticipated to alleviate global economic, ecological, and societal problems by incorporating green elements for longstanding economic benefits (Lorenz and Zinke 2005) – (Mohan et al. 2016). Nonetheless, these should be assessed in future research work using holistic, sustainable approaches, accounting for direct and indirect impacts across all levels and the whole life cycle in general, since there are substantial barriers yet to be overcome from a technological, socioeconomic, environmental, and political perspective.

Based on the findings of this study, there are several potential areas for future research. One of the main areas for future work is the development of more accurate and comprehensive LCA models for lignin, which could take into account a wider range of environmental impact categories, such as social and economic impacts. Another area for future research is the investigation of the potential for lignin to be used as a substitute for fossil-based materials in a wider range of applications, such as in the construction and automotive industries. This would require a detailed assessment of the environmental impacts and the technical feasibility of using lignin-based materials in these applications. Furthermore, there is a need for research on the development of new technologies and processes for the production and extraction of lignin, with a focus on reducing the environmental impact and improving the efficiency of the process. Finally, there is a need for research on the potential for lignin to contribute to a circular economy, by developing new business models and value chains for the use of lignin-based materials and by-products. This would require collaboration between different stakeholders, including industry, academia, and policymakers, to identify and overcome barriers to the development and adoption of lignin-based products and processes.

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