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Review

Review of life cycle assessments of lignin and derived products: Lessons learned

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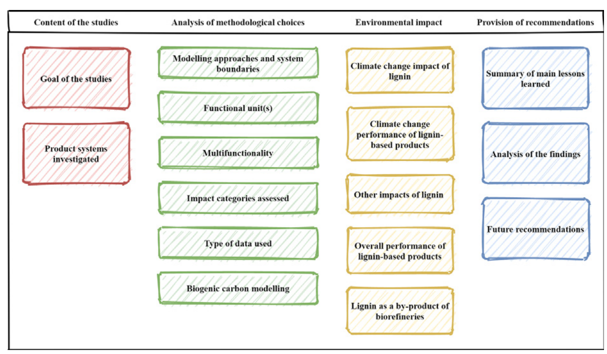
HIGHLIGHTS

- A first review of peer-reviewed LCAs of lignin and lignin-based products was conducted.
- Most of lignin-based applications showed promising climate change performances but trade-offs in other impact categories.
- The lack of harmonization in the application of LCA methodology hinders direct comparative analyses.
- Recommendations to increase consistency were provided.

GRAPHICAL ABSTRACT



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ABSTRACT

In the last decade, the use of lignin as a bio-based alternative for fossil-based products has attracted significant attention, and the first LCAs of lignin and derived products have been conducted. Assessing side-stream products like lignin and potential benefits compared to their fossil counterparts presents complex methodological issues. This article provides a critical review of forty-two peer-reviewed LCAs regarding lignin and derived products. Methodological issues and their influence on the LCA results include the choice of the modeling approach and system boundaries, functional unit definition, impact categories considered, type of data used, handling multifunctionality and biogenic carbon modeling. The review focused on climate change impacts, as this is also the main impact category considered in most studies. Other impact categories in the comparison between lignin-based products and counterparts were also discussed with examples from the studies. Based on ten lessons learned, recommendations were provided for LCA practitioners to increase future consistency of environmental claims made about lignin and lignin-based products. The finding suggest that the environmental performance of lignin-based products is significantly affected by both 1) LCA methodological problems such as allocation practices and biogenic carbon modeling and 2) technical aspects such as the percentage of lignin in the composition of products and the selection of the fuel to replace lignin in internal energy uses. Beyond this, the reviewed LCAs showed that often lignin-based products offer better environmental performances than fossil-based products, especially for climate change.

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Contents

1. Introduction

Next to cellulose and hemicellulose, lignin is the second most abundant natural biopolymer on Earth and accounts for about 30% of the organic carbon in the biosphere (Boerjan et al., 2003). In nature, lignin is an aromatic-composed binder that provides stiffness and strength to the stems of plants (Ragauskas et al., 2014). From a bioeconomy perspective, lignin is currently mainly used to produce bioenergy (electricity and heat) but has recently received attention as a renewable raw material for the production of chemicals and materials to replace petrochemical resources and sometimes provide also technical improvements. For example, plastic polymers can take advantage of the complexity of the lignin molecule to avoid the transformation steps to convert simple molecules into complex ones (Bernier et al., 2013). Other examples of interesting applications where lignin can be used to replace conventional materials are displacing urea-formaldehyde in adhesives (Yuan and Guo, 2017), bitumen in asphalts (Balaguera et al., 2018), polyacrylonitrile in carbon fibers (Hermansson et al., 2020), polyol in polyisocyanurate foams (Bernier et al., 2013) and liquid fuels (Obydenkova et al., 2017). Yet, lignin is currently largely unexploited for these purposes (Khan et al., 2019). Moreover, lignin can be used in other industrial applications that can benefit from the good surface activity of lignin (Czaikoski et al., 2020) such as adsorbents for CO_2 capture (Hao et al., 2017; Park et al., 2019) and catalysts (Cordeiro-Junior et al., 2020; Hernández-Ramos et al., 2020).

Lignin is mainly produced as a side stream of either the pulp and paper industry or from lignocellulosic biorefineries (Khan et al., 2019; Ragauskas et al., 2014). In the pulping industry, lignin can be extracted from black liquor which is a by-product of the wood pulping process of pulp mills (Bernier et al., 2013). In biorefineries, lignin is obtained as a non-fermentable side stream separated during biomass pre-treatment (Vera et al., 2020). With the expected development of lignocellulosic biorefineries more lignin is expected to become available. In both cases, lignin is currently mostly used internally to deliver energy needs (Ragauskas et al., 2014) but it can also be marketed (Bernier et al., 2013). Moreover, in both pulp mills and lignocellulosic biorefineries, the lignin extracted often exceeds the internal energy demand and can be sold externally (Ragauskas et al., 2014; Soam et al., 2016).

For pulp mills, extracting the lignin from the black liquor can be economically advantageous to have an extra source of revenue and diversify the products. Moreover, in most pulp mills, the recovery boiler works at maximum capacity since the upgrade of such a boiler is economically prohibitive (Axelsson et al., 2006; Culbertson et al., 2016). By extracting lignin, part of the solids from black liquor are taken away and the recovery boiler can be de-bottlenecked. This debottlenecking can increase the production of pulp and soap generating additional revenues (Axelsson et al., 2006; Hermansson et al., 2020).

The final application of technical lignin is largely influenced by the chemical and physical characteristics of the lignin (Arias et al., 2020). Beyond the feedstock used and the distinction between pulp mills and (lignocellulosic) biorefineries, the chemical structure of lignin is often influenced by the lignin production process and extraction techniques (Carvajal et al., 2016). For the different types of lignin and their extraction process, Fig. 1 provides examples of suitable applications and indicative market price ranges. Market prices depend among others on purity and potential application.

The Kraft process is the dominant process in the pulping industry (Cheremisinoff and Rosenfeld, 2010; Viikari et al., 2009). Other conventional pulping processes include the sulfite process and the soda process. With the Kraft process, lignin is obtained from hardwoods and softwoods using sodium hydroxide and sodium sulfide mixed in hot water (Bajwa et al., 2019). This mix is named white liquor and the residue of this process is the black liquor, from which lignin can be isolated. Among extraction techniques, acid precipitation through CO_2 and/or sulphuric acid (also commercially known as the Lignoboost process) are the most common (Bajwa et al., 2019). Alternatively, the organosolv process (solvent pulping) is a promising option that enables the extraction of relatively pure lignin but is only used at a small scale (Bajwa et al., 2019). Sulfite pulping allows isolation of lignosulfonates from spent sulfite liquor. In (lignocellulosic) biorefineries, the most common techniques for lignin separation from lignocellulosic biomass are steam explosion, acid pretreatment, enzymatic hydrolysis and alkaline hydrolysis pretreatment (Khan et al., 2019; Radotić and Mićić, 2016). Moreover, the abundant presence of aromatics in lignin makes it attractive for chemicals and fuels, and different depolymerization routes to aromatics (BTX and phenolic compounds) exist (Vural Gursel et al., 2019). Among novel routes for the production of bio-based aromatics from lignin, there are pyrolysis technologies, direct hydrodeoxygenation, and hydrothermal upgrading (Vural Gursel et al.,

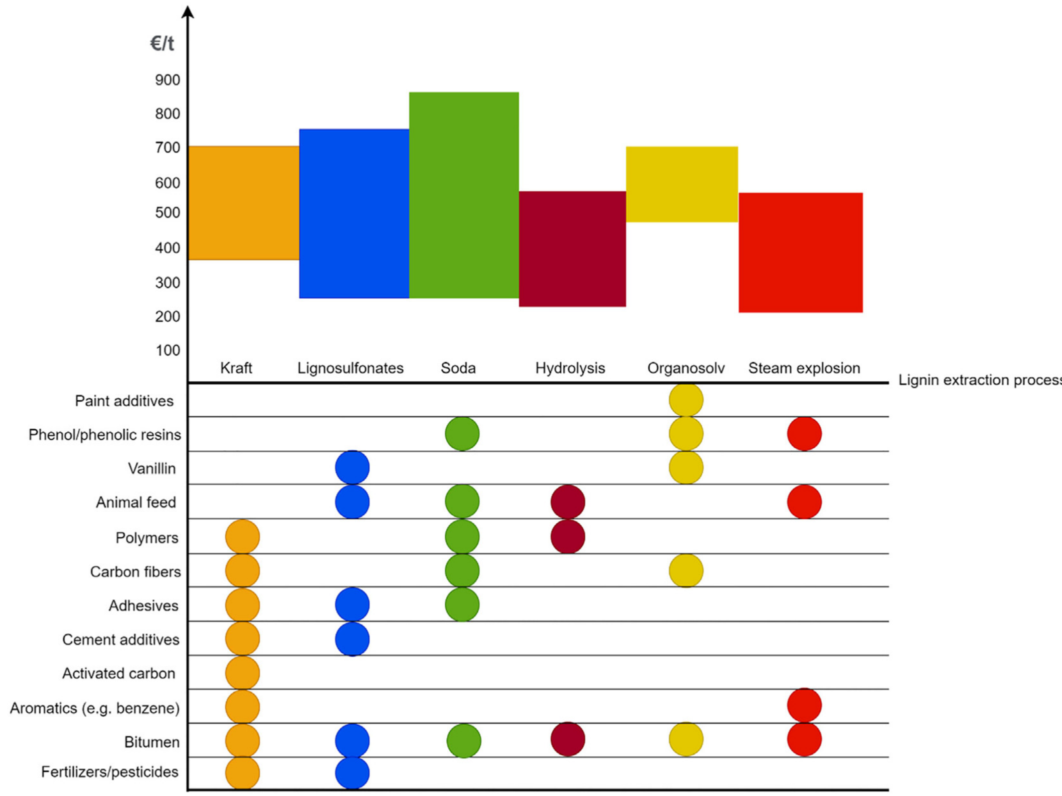

Fig. 1. Example of possible applications and indicative market price ranges of various types of lignin (Gosselink, 2011; Hodásová et al., 2015; Secchi et al., 2019).

2019). These routes aim to achieve good separation of the lignin from the cellulose and hemicellulose without changing it chemically or physically. This allows to utilize fully lignin's macromolecular structure in materials such as asphalt binders, adhesives, carbon fibers, resins and polymer composites. This has led to the lignin-first biorefinery concept that considers strategies to prevent structural degradation of lignin during biomass fractionation (Renders et al., 2017). Furthermore, vanillin can be produced from the oxidation of lignosulfonates that find use in foods and fragrances (Tarabanko and Tarabanko, 2017).

Lignin has the potential to substitute fossil fuels in both energy and non-energy use sectors to improve energy supply security and to contribute to climate change mitigation. For this reason, important development efforts are made by bioeconomy firms to make such a replacement possible. However, it is necessary to consider unambiguous sustainability criteria to assess if these alternative products allow actual environmental benefits compared to their fossil counterparts. In the bioeconomy, the tool that is often used to perform such a comparison between conventional products and bio-based alternatives is life cycle assessment (LCA) (Giuntoli et al., 2019). LCA is a standardized tool to model the entire life cycle of a product or system from resource extraction to final waste management (ISO, 2006a, 2006b). In the last decade, many peer-reviewed LCAs have been conducted to assess the environmental impact of lignin and the potential environmental benefits that lignin-based products can offer.

However, assessing the environmental impacts of bio-based products with LCA can be challenging since multiple life cycle modeling choices have to be defined by the practitioners (Broeren et al., 2017; Moretti et al., 2020c). In particular, the assessment of lignin and lignin-based products is among the most challenging case studies in bioeconomy. The origins of these challenges can be found in both *LCA methodological uncertainties* (e.g. handling co-products) that affect products from residual streams/bio-based by-products like lignin, and *data uncertainties* related to the low level of maturity of the production processes for lignin products for which often only lab-scale measurements are available. For these reasons, the environmental impact of lignin and lignin-based products is affected by high variability in the various LCAs reported in the literature (Montazeri et al., 2016). The carbon footprint of one kilogram of Kraft lignin can vary between a negative impact and 4 kg of CO_2eq depending on the selected allocation method (Hermansson et al., 2020) while the savings of GHG emissions allowed by lignin-derived adipic acid can range between −90% (savings) and + 100% (an increase of impact) depending on the data used and methodological choices applied (Montazeri et al., 2016).

This article is a critical review of peer-reviewed LCA studies of lignin and lignin-based products from the scientific literature. Given the methodological challenges in assessing lignin and lignin-derived products, the aim of this review is to obtain insights from the main findings of these studies and to evaluate qualitatively and quantitatively the methodological choices made in these LCAs and their consistency and robustness. Moreover, based on the results of these LCAs, potential environmental benefits of lignin-based products compared to the petrochemical products that they can replace are discussed. The insights from this review can be an important added value for LCA practitioners in the bioeconomy sector.

2. Material and methods

2.1. Selected studies

The LCA studies on lignin and lignin-based products were retrieved from the Scopus database (www.scopus.com) on July 8th 2020. In particular, the search[1] was based on two main keywords i.e. "life cycle assessment" or its acronym "LCA" and "lignin" looking at their presence

[1] The search string was: TITLE-ABS (("Life Cycle Assessment" OR lca) AND (lignin)) AND (LIMIT TO (LANGUAGE, "English")) AND (LIMIT-TO (SRCTYPE, "j")). It is possible that some biofuel studies that include biochemical processes making assumptions about lignin were not considered if they did not mention lignin in the abstract.

in titles and abstracts. Only studies published in English and documents published in scientific journals were considered. As a result of these parameters, 62 peer-reviewed articles were retrieved. After further screening, 48 articles concerning LCA of either lignin or lignin products were identified by excluding, for example, studies where the acronym LCA was not used as an acronym of life cycle assessment (but for "lignin-based activated carbon"). In particular, we focused on the studies published in the last decade which represents 41 studies out of 48. Of these 41 studies, about 85% were published in the last five years, which highlights the increasing interest in the topic. Moreover, a recent study published in August 2020 (Yadav et al., 2020) and not yet present in Scopus at the time of the search was also considered which resulted in the end in a total of 42 LCA studies to be assessed.

2.2. Aims and structure of the review

After the LCAs of lignin and lignin-based products were selected, the analysis was conducted in four main steps. The structure of the following sections resembles these four steps and the analyses conducted in each step (see Fig. 2).

The first step of the review was focused on understanding the content of the articles. In particular, we mainly answered these two questions: 1) what was the goal of the LCA studies? and 2) what lignin production system was investigated?. The first step aimed at providing recommendations to increase future consistency and was targeted at LCA practitioners only. To achieve this objective, we reviewed how the LCA methodology was applied in the lignin case studies taking ISO standards and major EU LCA guidelines as methodological reference documents. This allowed us to understand the state of art in assessing the environmental impact of lignin products. This part of the review can be found in Section 3.2 and sub-sections. The second step of the review aimed at comparing quantitatively the environmental impacts of lignin and lignin-based products reported in these LCAs. The result of this analysis (described in Section 3.3) was used to provide an overview of the impacts of lignin and the environmental performances of various lignin valorisation options compared to fossil counterparts. This second step was targeted as well to LCA practitioners, but insights could be also interesting for policymakers and lignin producers. A major focus was on climate change since it was the most considered impact in the selected studies (see Section 3.2.4), it is among priorities in policy agendas and the impact assessment methods are (almost) standardized allowing a (direct) comparison of the results. The other environmental impacts of lignin were also considered in Section 3.3.3 (and following). In the last step of the review, the main findings were summarised and recommendations for future research provided.

3. Results

3.1. Product systems

The 42 environmental LCA studies considered in this review (see supplementary materials for studies' categorization) can be divided into the following categories based on their *main* product system investigated:

- Assessing Kraft lignin (3 studies fall in this category (Bernier et al., 2013; Culbertson et al., 2016; Hermansson et al., 2020));
- Assessing organosolv lignin (1 study (Yadav et al., 2020));
- Assessing a biorefinery delignification process using natural malic acid (1 study (Yiin et al., 2018));
- Assessing lignin-based applications from various lignins (15 studies);
- Assessing major biorefinery products such as ethanol or lactic acid (21 studies);
- Performing a meta-analysis of life cycle energy and GHG emissions of bio-based chemicals (among them, some produced from lignin (1 study (Montazeri et al., 2016))

In the LCAs of Kraft lignin of Bernier et al. (2013) and Culbertson et al. (2016), the focus was on evaluating the environmental implications of introducing lignin extraction in Kraft mills. A similar study (Secchi et al., 2019) was also performed for biorefineries with and without marketed lignin.

Concerning the LCAs that look at the products using lignin, their aim was often twofold: identifying the environmental hotspots in the

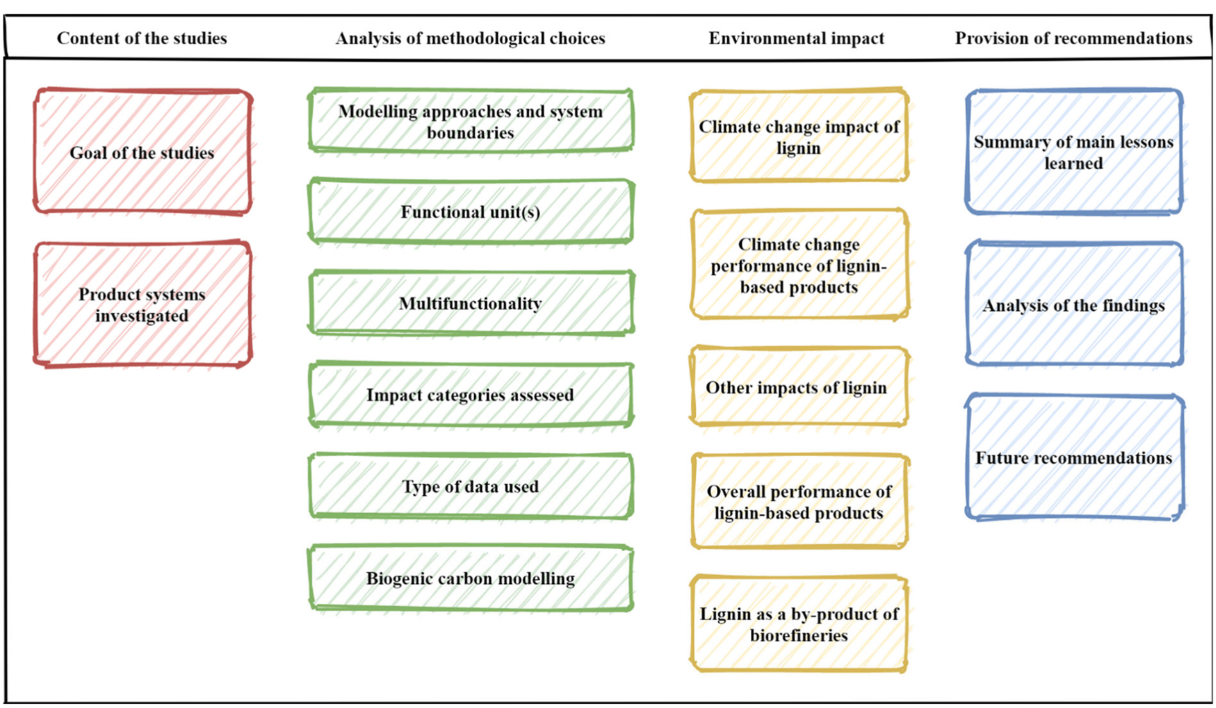

Fig. 2. Steps of the review.

production processes and evaluating possible environmental advantages in comparison with petrochemical products. Among the investigated lignin-based products, there were *adhesives* especially for wood fiberboards and laminates (Arias et al., 2020; Hildebrandt et al., 2019; McDevitt and Grigsby, 2014), *phenol* and *propylene* (Liao et al., 2020), *transportation fuels* (Obydenkova et al., 2017), *asphalt* (Tokede et al., 2020), *nanoparticles* (Koch et al., 2020), *polyurethane foams* (Manzardo et al., 2019), *fertilizers* (Krzyżaniak et al., 2019), *vanillin* (Isola et al., 2018), *adipic acid* (Corona et al., 2018; Van Duuren et al., 2011), *catechols* (Montazeri and Eckelman, 2016) and *carbon fibers reinforced polymers* (Das, 2011).

Concerning the studies investigating major biorefinery products (e.g. ethanol), some LCAs focused on assessing products of biorefineries where lignin is not a product since it is fully used for internal needs. For example, Vera et al. (2020) assessed a biorefinery producing ethanol and lactic acid which was equipped with a combined heat and power plant (CHP) where lignin was combusted for internal uses of the biorefinery without any surplus of heat and electricity. In other LCAs of biorefineries e.g. (Akmalina and Pawitra, 2020), the heat and electricity from the process were fully externally sourced from fossil fuels and all the lignin produced by the biorefinery was sold to generate electricity and chemicals outside. In other LCAs of biorefineries, lignin was only an intermediate product which was further processed to obtain biofuels and/or chemicals (Kumaniaev et al., 2020; Liao et al., 2020).

3.2. Analysis of methodological choices

The goal of the study has strong implications in all the choices that the practitioner has to make to conduct the LCA. In particular, it strongly affects the definition of the scope of the LCA. In fact, based on the goal, the following parts of the scope are strictly defined: the unit processes included in the system boundaries, the modeling approach to be used (and the type of data to be used), the functional unit (FU) and the methods to deal with co-products. All these aspects are crucial to interpret the results of an LCA and understand what can be concluded and what not from the LCA results. Accordingly, the goal of the selected studies and the modeling choices made by the LCA practitioners were noted in the following sections where relevant.

3.2.1. Modeling approaches and system boundaries

The appropriate modeling approach (attributional or consequential) is directly linked with the goal of the study. An attributional approach should be selected if the goal is to assess the environmental hotspots of a process or the determination of the environmental impact of a single bio-based product to compare with fossil products. A consequential approach should be used instead to assess a change in a specific system and the overall consequence of this change in the system and in the world outside. For example, in view of the worldwide environmental impact, is the current use of the black liquor in Kraft mills for internal energy better than isolating lignin from it to be marketed?

Despite the fact that ISO 14044:2006 does not distinguish between attributional and consequential LCAs, for many practitioners (Corrado et al., 2017; Nguyen and Hermansen, 2012) and handbooks (e.g. ILCD handbook (ILCD, 2010)), it is important to select the modeling approach based on the goal of the study. In a 2020 study of (Moretti et al., 2020a), using a text mining process, it was shown that 75% of the LCAs assessing multifunctional bioeconomy case studies did not clearly mention the modeling approach. When applying the same text mining method to the reviewed studies on lignin and lignin-based products, it was also found that 78% of the studies did not use the keywords "attributional" or "consequential" to specify the approach followed. Of the remaining studies, 8 LCAs were defined by the practitioners using the term attributional while only one article (Corona et al., 2018) defined the approach followed as consequential. The selection of the modeling approach also affects other decisions that the practitioners have to take to conduct an LCA. First of all, attributional studies require average data while in consequential studies marginal data are used. Second, depending on if the study is consequential or attributional, the system boundaries and the unit processes included within the system boundaries change (see a simplified example for dealing with electricity surplus in Fig. 3). Third, depending on the goal, the type of system expansion method that can be applied is different: enlargement (only expansion of the boundaries) or substitution (expansion followed by substitution). While enlargement can be used in both attributional LCAs (ALCAs) and consequential LCAs (CLCAs), the use of substitution as a system expansion method is inconsistent with attributional modeling (Majeau-Bettez et al., 2018). Further details about the use of system expansion to deal with multifunctionality can be found in Section 3.2.3.

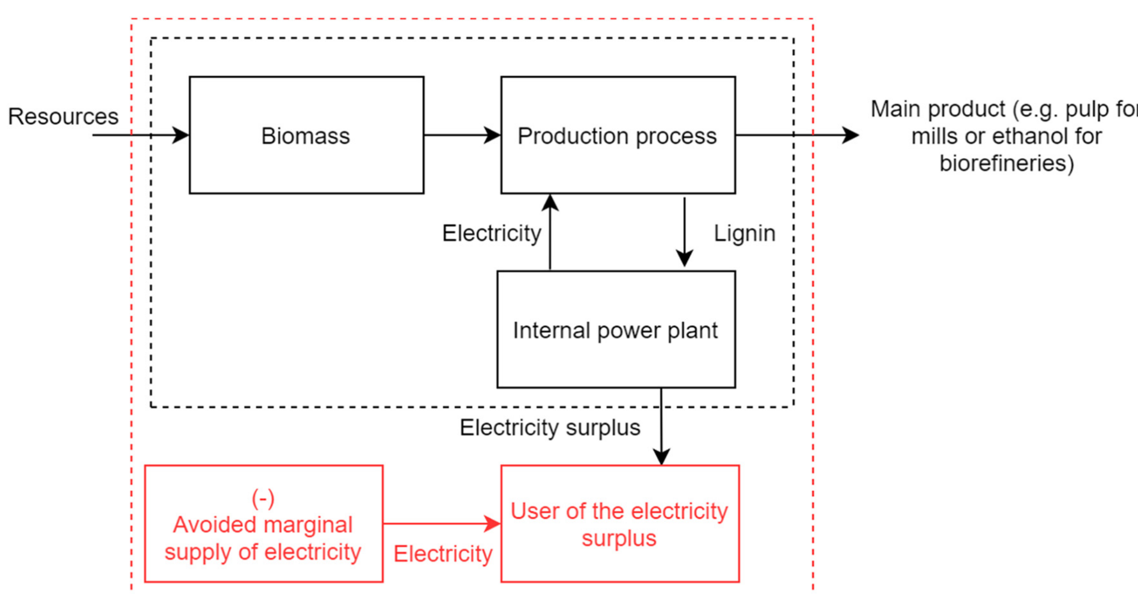

Fig. 3. A simplified example of differences between system boundaries in attributional (black) and consequential LCAs to deal with a surplus of electricity generated using lignin burned internally. In red, system boundaries and unit processes that would be included within the boundaries in a consequential analysis. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Concerning consequential LCAs, this type of study uses economic market modeling to forecast what will happen as a consequence of the assessed change. For example, market modeling is needed to understand what mix of technologies will be replaced by a by-product that is produced because of the assessed change in the system (e.g. lignin extracted from a pulp mill and no longer used for internal combustion). To avoid malpractices, it is necessary to understand what are the main market drivers of the system (what is the purpose of the system) and what products satisfy such market drivers. For example, the Kraft black liquor currently exceeds the demand for non-fuel uses (Bernier et al., 2013). So, the demand for paper (pulp) is the market driver of the Kraft mills and not the market applications of lignin (Bernier et al., 2013). Another aspect that is important to consider in a consequential LCA of Kraft mills is the energy source used to replace black liquor. Since black liquor is mainly combusted for internal energy needs, a likely scenario is that fossil fuels will be used to produce the part of steam that cannot be produced anymore from black liquor (Axelsson et al., 2006; Bernier et al., 2013). On the other hand, depending on regulations policy schemes in place, biomass is also an option especially if there is the availability of low-quality biomass in the vicinity (e.g. bark). Similarly, also in biorefineries, different alternatives are possible as replacement of lignin for internal energy purposes. Among them, the most probable options are the use of natural gas or biofuels such as wood chips or biogas (Obydenkova et al., 2017). While the use of natural gas is the most cost-effective (and better water footprint), the use of lignocellulosic biofuels is generally the best solution if the main goal is to minimize GHG emissions (Obydenkova et al., 2017).

Concerning the life cycle stages considered, most of the studies (87%) were cradle to gate studies i.e. the use and end of life of the products delivered by the system were not included in the assessment. As exceptions, the following cases were found:

- Well to wheel studies (Budsberg et al., 2016; Obydenkova et al., 2017; Raman and Gnansounou, 2015). In well to wheel studies, the combustion of the transportation fuels produced is considered (end of life = use phase).
- Investigation of a specific lignin extraction process (using natural malic acid), which was performed by Yiin et al. (2018). The boundaries of the systems were gate to gate: from the harvested oil palm empty fruit bunch to the extracted lignin.
- A full cradle-to-grave study for polyurethanes produced using lignin-derived polyols (Manzardo et al., 2019).
- A full cradle-to-grave study for adhesives used in fiberboard production (McDevitt and Grigsby, 2014). In this study, the same end of life (landfill) was assumed for bio-based and fossil-based products and the dataset for inert waste processed in a landfill was retrieved from ecoinvent (Ecoinvent, 2020). Hence, the different compositions were not taken into account.

3.2.2. Functional unit(s)

In LCA, the functional unit is the “quantified performance of a product system for use as a reference unit” (ISO, 2006a) and depends on the final function of the products delivered by the product system. Lignin can be used for products that have very different functionalities. How each product can fulfill a specific function has to be accounted for in the functional unit. For example, one of the main functions of polyurethane foams is to provide thermal resistance. Hence, in a comparative LCA of polyurethane foams, the differences in thermal resistances have to be accounted for by the functional unit. A good functional unit could e.g. be the amount of foam needed to achieve a specific thermal resistance (Manzardo et al., 2019). Only if the physical properties and mechanical characteristics that are important for the final applications are comparable, a simplified functional unit based on a mass or a surface is a possible option. For example, Hildebrandt et al., after checking that the tensile modulus and strength were comparable, defined the functional unit as $1 m^2$ of a laminate board (Hildebrandt et al., 2019). However, most studies, e.g. (Tokede et al., 2020), using simplified functional units did not make a similar check.

The functional units selected in these studies could be cataloged as follows:

- *Simplified mass* FU *based on the output*. For example, 1 t of pulp (e.g. used by (Culbertson et al., 2016)) or 1 g of vanillin (e.g. used by (Isola et al., 2018). This type of FU was used by about 50% of the LCAs;
- *Simplified energy* FU *based on the output*. For example, 1 MJ of ethanol (e.g. (Rahman et al., 2015)) or 1 MJ of jet fuel (e.g. (Budsberg et al., 2016)). This type of FU was used by about 15% of the LCAs;
- *Simplified volume FU based on the output*. This FU was used in two studies: 1 l of ethanol (Soam et al., 2018) and 1 m^3 of finished medium density fiberboard (Yuan and Guo, 2017);
- *Simplified area FU based on the output*. This FU (1 m^2) was used in two studies (Hildebrandt et al., 2019; McDevitt and Grigsby, 2014) assessing adhesives for wood fiberboards;
- *Input based FU*. This type of FU was used in the study of (González-García et al., 2016) where the functional unit was the input of the biorefinery (100 kg dried *Pinus pinaster* chips);
- *Entire biorefinery*. This FU was used by two studies (Ojeda et al., 2011; Shinde et al., 2020);
- *Multiple FUs (one per each main co-product)*. This type of FU was used by (Liao et al., 2020; Modahl et al., 2015; Vera et al., 2020);
- *Distance*. The FU of 1 km, which is typical for well-to-wheel assessments and was used by (Raman and Gnansounou, 2015);
- *Ultimate final application*. This functional unit was used by (Das, 2011) i.e. an automotive part under consideration.

In particular, an *input-based functional unit* or the assessment of the *entire biorefinery* allows to avoid the allocation between the co-products (among them, lignin). This approach is also one of the enlargement methods to solve the multifunctionality problem (for details, see Section 3.2.3). In this way, the modeling uncertainty generated by the multifunctionality problem is avoided. This approach is applicable if the goal of the study is the identification of the environmental hotspots of a process or an entire biorefinery.

However, this approach is not applicable if the goal requires the determination of the impact of a single co-product. When this is the case (for example to compare it with its fossil counterpart), multifunctionality uncertainty cannot be avoided (except the few cases where subdivision solves the multifunctionality problem, see Section 3.2.5). Under these circumstances, it is good practice to define multiple functional units to increase transparency and show what is the impact of all co-products after the allocation is applied. Only in this way, the reader of the LCA can directly understand the effect of the allocation method on the environmental performance of each co-product. For example, Modahl et al. (2015) defined the FUs of their LCA of a Norwegian biorefinery as 1 t of product for cellulose, lignin and vanillin and 1 m^3 for ethanol and showed the results per each FU.

3.2.3. Multifunctionality

Lignin is always a product of multifunctional systems, i.e. systems delivering multiple products. To perform an LCA of this type of system, the selection of a criterion to apportion the impact on each product is necessary. This selection is one of the main sources of uncertainty of LCAs (Klöpffer, 2012; Reap et al., 2008). In particular, ISO 14044:2006 provides a three-level hierarchy to deal with this problem. The first level of the hierarchy recommends avoiding allocation, either by dividing the process into sub-processes which are no more multifunctional or by system expansion e.g. by re-defining the boundaries of the system in a way that the system enveloped by the new boundaries is no more multifunctional. Applying subdivision in mills and biorefineries rarely solve the multifunctionality problem. System expansion can be applied in two ways: enlargement (system expansion alone) or (system expansion followed by substitution). A summary of possible enlargement methods can be found in (Moretti et al., 2020a). Enlargement is not a solution if the goal of the study requires the determination of the impact of a single output of the system and not of the whole system. For example, in the study of (Shinde et al., 2020), the impact of the entire biorefinery is considered avoiding the allocation to each of the three co-products ellagic acid (EA), lignin, and pectin. Substitution is the main option used in consequential studies. Considering Fig. 3, once the unit process representing the avoided marginal production is included in the system boundaries, substitution allows to subtract the impact of this unit to the one of the entire system inside the boundaries. In biorefineries and mills, lignin is often a by-product (and not the main product) contributing to less than 50% of the revenues of the system. Accordingly, in these cases, substitution can be an option (ILCD, 2010; Moretti et al., 2020a; Sandin et al., 2015). Nevertheless, substitution is sometimes used as a system expansion method in attributional studies leading to either erroneous results or misleading interpretations which emerge especially when multiple impact categories are assessed (Majeau-Bettez et al., 2018; Sandin et al., 2015). For example, Akmalina and Pawitra performed an LCA of ethylene from empty fruit bunch (residue of palm oil processing) and compared the obtained impact with the one of fossil ethylene (Akmalina and Pawitra, 2020). The interpretation of the results was that bio-based ethylene was much better than fossil ethylene from a climate change perspective (about half impact). However, Akmalina and Pawitra (2020) solved the multifunctionality issue by substituting the lignin produced with electricity and chemicals. This credit reduced the impact of bio-based ethylene by 83.9% (Akmalina and Pawitra, 2020), leading to a climate change impact of 1.15 kg of kg CO_2eq per kg of ethylene. However, the palm oil extraction unit alone had a contribution of 7.17 kg CO_2eq/kg ethylene. When applying allocation (partitioning) instead of substitution, a completely different conclusion would have been obtained.

The second level of the hierarchy recommends allocation methods reflecting the way "in which the inputs and outputs are changed by quantitative changes in the products" (ISO, 2006a), which in the literature has been often referred to as "physical causal relationships" allocation. To lignin, this allocation can be applied "by varying the quantity of lignin precipitated and then observing direct variations in the environmental loads" (Bernier et al., 2013). In general, the changes modeled using this type of allocation can be either marginal, incremental or average (listed in order of magnitude) (Azapagic and Clift, 1998). A physical causality allocation based on average changes was used by Bernier et al. (2013) who added/eliminated a functional output completely. Different from other allocation methods (e.g. energy or economic value), physical causality allocation does not apportion the impact of the system with a static share for all impact categories. This implies that if extracting lignin does not have consequences on the ratio wood chips/pulp, the land occupation impact caused by the wood chips used in the wood pulping process is not allocated to lignin.

The third (and last) level recommends allocation methods based on parameters such as mass, energy or economic value selected based on their ability to reflect other causal relationships. A comprehensive study on lignin allocation was conducted by Hermansson et al. (2020), who applied 12 types of methods to deal with the multifunctionality of a Kraft mill. Among the methods applied there were system expansion followed by substitution, allocations based on mass, energy, exergy, economic values, marginal allocation, substitution-based allocations and mixed allocations (e.g. mass plus energy). Based on the sample of allocation methods selected, Hermansson et al. (2020) concluded that the impact of Kraft-lignin and derived products could be significantly affected by the allocation choices. The results were highly influenced by the following allocation parameters: (1) the choice of the main product/function (driver of the system), (2) the price of lignin and (3) the choice of displaced outputs. With respect to the first parameter, Hermansson et al. obtained the highest variation of results because some of the allocation scenarios considered lignin instead of pulp as the main product. However, this is very unlikely, since lignin represents about 3–5% of the overall revenues of Kraft mills (Culbertson et al., 2016). So, apportioning all the impact of the Kraft mill to lignin (and no impact to the pulp) or substituting pulp looks unreasonable. When considering that pulp is the main driver of the system, the variation of the impact of lignin calculated by Hermansson et al. becomes much narrower (see Section 3.3.1) Concerning economic allocation, the price assumed for lignin should reflect the specific lignin under investigation. In fact, the price of lignin is highly variable depending on the source and quality of the lignin (see Fig. 1). However, specific quality-level lignin "has relatively stable prices through the years and seasons" (Hodásová et al., 2015). So, the scenario applied by Hermansson et al. (2020) assuming a tenfold increase in price for lignin in the future was also not considered in the ranges of climate change impacts identified in Section 3.3.

In some studies, the type of allocation used was not clear (e.g. in (Tokede et al., 2020)) while in most of the LCAs, a sensitivity analysis on the allocation method was performed. As an example, Culbertson et al. (2016) analyzed the impacts of producing pulp applying system expansion by substitution to the co-products in the baseline calculations. In particular, the two co-products (i.e. surplus electricity and lignin) were substituted with grid electricity and phenolic resin (Culbertson et al., 2016). In their sensitivity analysis, mass and economic allocations were used in combination with substitution (mixed approach) keeping the credit for the surplus of electricity.

A summary of the adopted multifunctionality practices in the selected LCAs is shown in Fig. 4. Although mass allocation was the most adopted method to deal with multifunctionality, Fig. 4 shows that a wide variety of methods were applied between the reviewed studies. The fact that various methods were used is not a problem per se. However, it becomes a problem if the different practices derive from a different interpretation of ISO 14044:2006 recommendations, which has not been uniform in the LCAs of bioeconomy systems in the literature (Moretti et al., 2020a). This problem emerges clearly from the case of lignin. For example, substitution was often used as both a system expansion method or as a basis for the application of an allocation. However, as Montazeri et al. (2016) observed in their meta-analysis, substitution "can produce distorted LCA results for biofuel systems in which coproducts constitute a significant fraction of total economic value, energy flow, or mass flow". For this reason, a check on physical/economical significance should be performed before applying substitution (ILCD, 2010). Since practices are not harmonized and to avoid the abovementioned problem, Montazeri et al. (2016) suggested that "to avoid such pitfalls, it is recommended that LCA practitioners, sustainability scientists, and the chemicals industry collaborate to form a consensus on a standardized LCA approach to account for coproduct flows for bio-based chemicals".

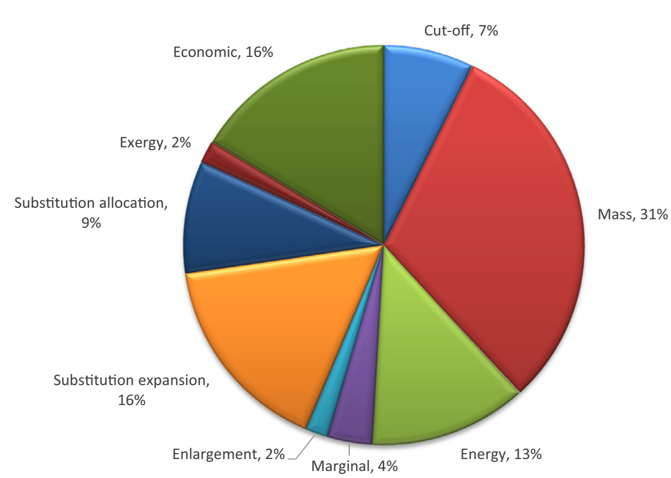

Fig. 4. Summary of the adopted multifunctionality practices in the selected 42 LCAs.

3.2.4. Impact categories assessed

The selection of the impact categories is part of the scope of the LCA. The impact categories considered are important, especially to understand the claims in the interpretation of the results such as "product A is more sustainable than product B". What does more sustainable mean? As it is possible to observe from Fig. 5, climate change impact was investigated in all the LCAs. The main reason is that climate change is the main driver for the development of bio-based products given the short time carbon cycle of the biomass used. Fossil depletion was also often investigated (55% of studies). This type of impact category has lower uncertainty than others and is linked with the results of climate change impacts. Hence, once data are collected to assess climate change, all data needed for assessing fossil depletion are available. Eutrophication and acidification were also assessed in more than 50% of the studies. These two impact categories are important for biomass products since agricultural production (and the emissions resulting from the application of fertilizers) can accelerate the decrease of the pH of the soil over time. Among the least assessed impact categories, there are land use and water depletion, which were assessed only in 13% and 10% of the LCAs respectively. These figures resemble the numbers presented by (Laurent et al., 2014) in their review of 222 LCAs of solid waste management systems. In their study, they showed that land use and water depletion were assessed in less than 15% of the LCAs. As Laurent et al. (2014) observed, the reason behind the lack of consideration of these two impacts can be found in the absence of consensus in their impact assessment methods. Doubtless, these two impacts are important for biomass systems and should be assessed. New methods are emerging for their assessment as for example the LANCA method for land use (Beck et al., 2010) and AWARE method for water depletion (Hélias, 2020). Despite this lack of consensus, at least an estimation of the hectares of land needed per functional unit and a water balance should be performed in an LCA of products derived from biomass.

3.2.5. Type of data used

Concerning inventory data used, in 55% of the studies, primary data were partially available. In most of these LCAs, these data were generated at the laboratory scale and then system modeling was conducted for their approximation on a large scale. In 28% of the studies, all data were generated through specific modeling software (without validation with lab experiments). In 18% of studies, all data were retrieved from the literature or LCA databases. Among the main literature sources for data of Kraft lignin, Culbertson et al. (2016), Benali et al. (2016) and Bernier et al. (2013) were the main sources used. For example, Culbertson et al. (2016) was used as a data source for Kraft lignin by (Hermansson et al., 2020) and (Manzardo et al., 2019).

Since lignin-based products are recently emerging, there is a problem with data availability and data quality. In particular, some lignin-based products have a technological readiness level below 5. Early-stage LCAs (e.g. Koch et al. (2020) for lignin nanoparticles) were conducted to support the development of the technology identifying environmental hotspots and possible modifications for environmental improvements. Early-stage assessments are characterized by problems related to lack of high-quality data and results are more affected by uncertainties than LCAs based on data collected from actual operating plants (Moretti et al., 2020b; Patel et al., 2012).

3.2.6. Biogenic carbon accounting procedure

The selected system boundaries and the timeframe influence how the biogenic carbon of the lignin is considered in the LCA.

In cradle-to-grave studies, one option is to include the biogenic carbon of lignin as stored in lignin (with credit) and in the future product (e.g. plastic application) derived from lignin. If during the lifetime of the product, the biogenic carbon from lignin embedded in the product does not degrade, the biogenic carbon is entirely sequestered in the product. However, if the product is, for example, incinerated within 100 years (global warming is often assessed over 100 years) after the production phase, a cradle to grave LCA would have to account for the CO_2 emissions from lignin. The way that biogenic carbon intake is accounted for in lignin studies can highly affect the results in climate change. Bernier et al. (2013) estimated in 0.6 kg of CO_2eq the cradle-to-gate impact of 1 kg of Kraft lignin. This value already includes a credit based on the biogenic carbon content of lignin (2.3 kg of CO_2eq per kg of Kraft lignin) (Bernier et al., 2013). The subtraction of such a credit in a cradle-to-gate study implies that the biogenic carbon remains stored for more than 100 years.

If the carbon content of the lignin-based product is released in less than 100 years, another option is to assign a characterization factor equal to zero for biogenic emissions over the entire life cycle. This is also an option in cradle-to-gate studies and is often referred to as the "carbon neutrality" assumption. This assumption was for example made by Shuai et al. (2016) and Hermansson et al. (2020). In most of LCA guidelines and policy recommendations, a zero discount rate is applied to biogenic emissions. This means that the time difference between the moment when the biomass absorbed the carbon and the moment when the carbon dioxide is released is not accounted for; it is as if they both happen at the same time. This approach is followed by European commission guidelines (European Commission, 2012, 2018; ILCD, 2010), European directives for renewable energies and alternative fuels (European Commission, 2016; European Parliament, 2015) and the US Environmental protection agency (EPA, 2011). Alternatively,

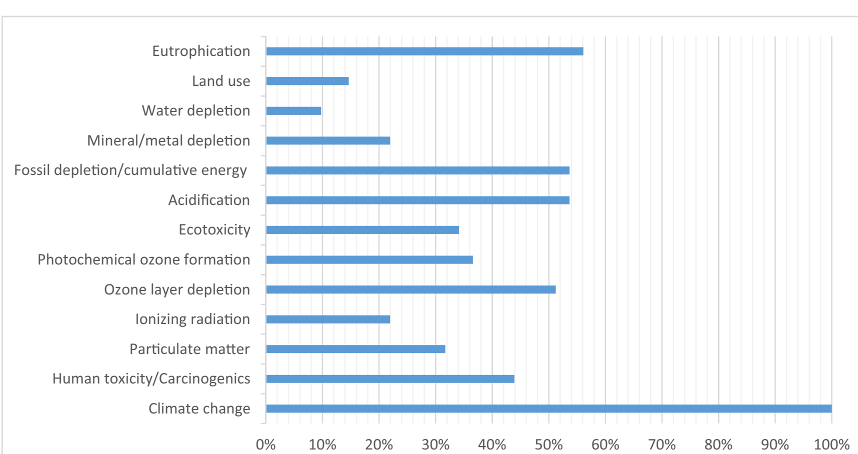

Fig. 5. Impact categories covered in the evaluated sample of LCAs.

the release of biogenic emissions and carbon storage can be discounted in time as proposed by UK PAS 2050 (BSI, 2011).

As an alternative to the carbon neutrality assumption, Culbertson et al. (2016) accounted for the biogenic intake with a characterization factor of −1. This elementary flow representing the biogenic intake was accounted for in the inventory of biomass (softwood). As a result, this flow was then allocated to all products with the allocation method applied to apportion the impact (and the biogenic credit) to the co-products. This method is consistent with the EU PEF guide and PEFCR guidance which recommends that the "allocation rules used for all other elementary flows shall also apply to model the biogenic carbon flows" (European Commission, 2012, 2018). However, this can lead to carbon accounting inconsistencies when the allocation rules applied do not reflect the actual biogenic content of the product. In such cases, should the biogenic emissions released when combusting the product correspond to its biogenic carbon content (as it would happen in reality), or to the allocated biogenic carbon content (as accounted for in the model)?. Using the method applied by Culbertson, a good practice is to separate the inventory and characterization results for climate change into two categories: fossil and biogenic, as done by (Hildebrandt et al., 2019) and suggested by recent EU LCA guidelines (European Commission, 2018)). Only in this way, it would be possible to use cradle-to-gate results (and inventory) as input to other cradle-to-grave studies. For this reason, the EU PEF guide and PEFCR guidelines recommend that "the biogenic carbon content at factory gate (physical content and allocated content) shall always be reported as additional technical information" (European Commission, 2012, 2018).

Moreover, since most LCAs were conducted from the cradle to the gate, the possible biodegradation of the carbon embedded in the lignin during the use phase and end-of-life of the products was not modeled in these studies.

3.3. Environmental impact

3.3.1. Climate change impacts of lignin

As mentioned in Section 2.2, this review pays increased attention to results for climate change than to other impact categories, since climate change was assessed in all the studies and offers higher comparability among studies. In particular, Fig. 6 shows the climate change impact of Kraft lignin as reported in the LCA studies assessing lignin from Kraft mills (Bernier et al., 2013; Culbertson et al., 2016; Hermansson et al., 2020) and two LCAs on lignin-based products for which it was possible to retrieve/back-calculate the values obtained from their inventory data.

From Fig. 6, it is possible to notice that the cradle-to-gate impact of 1 kg of dry Kraft lignin varies between 0.1 and 2.7 kg CO_2eq. But Fig. 6 is not self-explanatory and needs to be handled carefully. From Fig. 6, it appears as if Arias et al. (2020) estimated a much higher impact than the other 4 LCAs and that the impact calculated by Bernier et al. (2013) is perfectly in line with the upper values from Hermansson et al. (2020) and Culbertson et al. (2016) while the result of Tokede et al. (2020) is just a bit less than their lower values. On the other hand, these studies should be compared with consistent modelings for biogenic emissions. However, it is unclear how the biogenic carbon was accounted for in Tokede et al. and Arias et al. The other three studies used unharmonized accountings. Hermansson et al. used the carbon neutrality assumption, Bernier et al. subtracted the biogenic carbon content of lignin as a carbon dioxide credit and Culbertson et al. accounted for the biogenic intake from biomass with a characterization factor of −1, which was afterward allocated. Although different, the methods applied by Hermansson et al. and Culbertson et al. provide consistent cradle-to-gate results (as shown by Fig. 6 and considering that Culbertson et al. was also the main data source used by Hermansson et al.). Conversely, although the value reported by Bernier et al. looks numerically aligned with these two, the biogenic accounting is not accounted in a similar way. The value reported by Bernier et al. becomes consistent once the biogenic carbon intake (2.3 kg CO_2eq) is added, becoming about 2.9 kg CO_2eq and therefore much closer (and higher) to the value reported by Arias et al. (2020). This means that the kraft mill modeled by Bernier et al. is much more impacting on climate change than the one modeled by Culbertson et al. (2016). The key reason is the (allocated) consumption of natural gas per kg of lignin which is one order of magnitude higher in Bernier et al.

Regarding other types of lignin, in the LCA conducted by (Arias et al., 2020), the climate change impact of organosolv lignin from softwood was estimated in 1.85 kg CO_2eq (17% lower than Kraft lignin). This value falls also in the interval of values estimated by (Yadav et al., 2020), which was 1.4–2.1 kg CO_2eq per kg dry organosolv lignin from bark. In particular, the type of solvent used in the organosolv process affects the results significantly. For example, the use of either fossil-based ethanol/methanol or bio-based ethanol/methanol can lead to a completely different environmental footprint and insights (Koch et al., 2020). For instance, one of the insights of Koch et al. (2020) was to

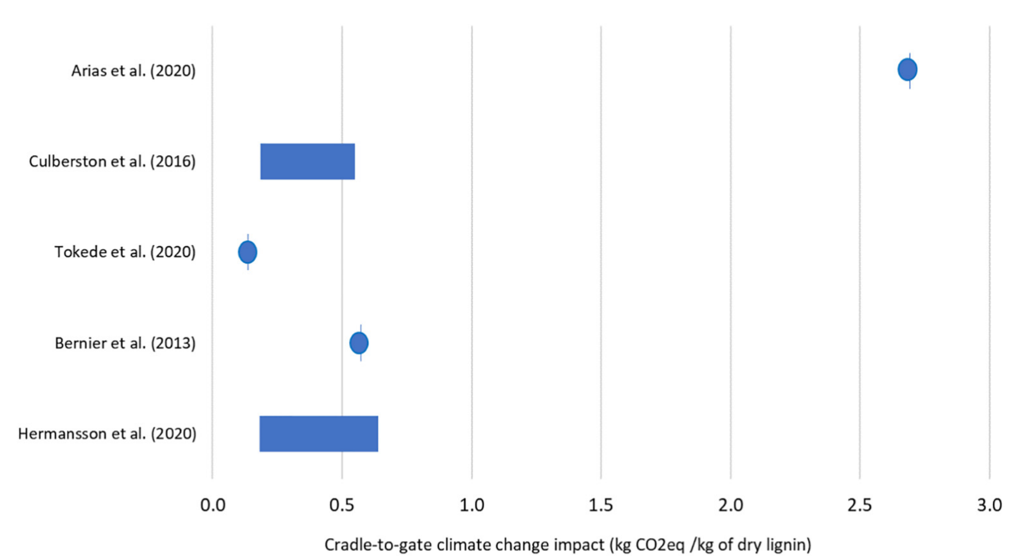

Fig. 6. Cradle-to-gate climate change impact of Kraft lignin as reported in the reviewed LCAs. Ranges represent mainly the testing of different allocation methods in the baseline calculations conducted in the LCA. With respect to the allocation methods tested by Hermansson et al. (2020), once the assumptions regarding the main product and current steadiness of lignin price were revised as mentioned in Section 3.2.3, the impact of 1 kg of Kraft lignin was in the range between 0.2 and 0.6 kg CO_2eq per kg dry lignin.

recover the fossil-based ethanol used as a solvent as much as possible. However, if bio-ethanol is used, the direction was "to not recover ethanol at all" (Koch et al., 2020).

Unfortunately, climate change values for lignin obtained from biorefineries are scarce, since in most of the LCAs of biorefineries lignin was neither used internally, nor the product in focus, and the functional unit was not defined in terms of lignin. The only LCA that reported the impact of lignin from the biorefinery was (Modahl et al., 2015), who applied multiple functional units. Modahl et al. estimated an impact of 1.12 kg CO_2eq per kg of lignin from a mix of timber and wood chips. However, it can be expected that for other biorefineries, the impact is very variable depending on the production process, allocation applied and feedstock used. The other two studies mentioned in Section 3.2.2 that used multiple functional units did not have lignin as a sold co-product.

3.3.2. Climate change performance of lignin-based products

Concerning lignin-based products and the potential reductions of climate change impact that they can allow in the replacement of fossil-based applications, two (conceptually) slightly different approaches are possible i.e. comparing final applications (e.g. asphalt with lignin versus conventional asphalt) or comparing ingredients (e.g. lignin for asphalts and bitumen).

The first approach assesses the two alternative products (with lignin and without lignin) considering the entire life-cycle. In each application, the percentage of lignin used compared to other input materials can be small or large. Based on how much percentage of materials input can be replaced with lignin, the importance of lignin on the final LCA outcome could be low or high. For example, 5% of the weight of asphalts is made of bitumen, which is one of the most environmentally impacting ingredients of asphalts' recipes, and lignin can replace reasonably up to 25% of this bitumen (Tokede et al., 2020).

In the second approach, one of the main aspects that is important to consider is the fact that, for most applications, lignin does not replace other ingredients with a 1:1 mass ratio. For example, 2 kg of lignin can replace 1 kg of carbon fibers or 3 kg of lignin can replace 1 kg of fossil raw materials for the production of tert-butyl catechols (Hermansson et al., 2020). A second aspect is that the use of lignin instead of fossil ingredients often leads to changes in the composition or manufacturing of materials, e.g. using lignin instead of bitumen changes the composition of the asphalt and the energy consumption of the production phase (van Vliet et al., 2017). Landa and Gosselink (2019) published the application of lignin in bio-asphalt showing a lower production temperature (130 °C) for this novel asphalt compared to conventional asphalt. If both asphalt composition and processing change significantly due to the use of lignin instead of bitumen, then it will not be possible to directly compare 1 kg of lignin with 1 kg of bitumen (or with a different mass ratio). For example, Arias et al. (2020) assessed bio-based adhesives made from Kraft lignin and organosolv lignin. An interesting finding of the study is that despite organosolv lignin can be used in higher percentage in the adhesive mix than Kraft lignin and its climate change impact was lower than for Kraft lignin, the climate change impact of organosolv lignin adhesives was higher than for Kraft lignin adhesives (15.5 kg CO_2eq versus 8.3 kg CO_2eq per kg of adhesive). The main reason was that the lignin glyoxylation process (required for the functionalization of lignin for this application) requires much (about 2.4 times) higher electricity consumption to process organosolv lignin than to process Kraft lignin. The study of Yuan and Guo (2017) calculated the impact of adhesives from lignosulfonates (hybrid ammonium lignosulfonates). They estimated that 1 kg of adhesives from lignosulfonates lignin generate 0.13 kg of CO_2eq,[2] which is much lower than the impact of the adhesive from Kraft and organosolv lignins calculated by (Arias et al., 2020).

Given the issues mentioned above about the second approach, most of the LCAs on lignin applications applied the first approach, which is more reliable. Fig. 7 shows the savings of GHG emissions that are achievable using lignin-based applications to replace conventional petrochemical products estimated by the reviewed LCAs.

As can be observed from Fig. 7, there are many applications where lignin can be used which are promising from a GHG emissions perspective. In particular, Obydenkova et al. (2017) reported that deriving a transportation fuel from lignin by pyrolysis that can replace diesel on the market could generate up to 90% of GHG emissions savings. However, the emissions savings vary in the range between 10% and 90% (Obydenkova et al., 2017) depending on two critical factors: 1) what source of energy is used in the biorefinery to replace the diverted lignin and 2) what type of allocation method is applied. For example, lignin could be replaced by either natural gas or biomass (e.g. corn stover) as energy sources, and biomass would be preferable from a GHG emissions perspective. However, (Obydenkova et al., 2017) estimated that the use of corn stover as fuel instead of natural gas would increase the cost by about 30%. Concerning the allocation method, the use of either energy allocation or cut-off allocation (all impact to ethanol) affected significantly the results of Obydenkova et al. (2017). However, a cut-off allocation does not seem fair since lignin cannot be considered a waste in LCA terms (ISO, 2006a) according to the waste management framework (European Union, 2008) adopted by the European Union in the Renewable Energy Directive (European Commission, 2016).

Adipic acid also seems a promising application from a GHG emissions perspective, allowing savings between 62% and 78% compared to petrochemical adipic acid (Corona et al., 2018). This range represents two different scenarios representing two different possible locations for the adipic acid plant. In particular, the study of Corona et al. was the only self-declared consequential LCA. Accordingly, inside the system boundaries, the unit processes representing the avoided production of heat and electricity internally to the biorefinery were included.

While most of the studies did not analyze the end of life of the products, Manzardo et al. (2019) conducted a full cradle-to-grave LCA of bio-based rigid polyurethane foams and compared their impact with the fossil counterpart. In particular, Manzardo et al. considered three different foams produced from bio-based polyols obtained from lignin. Bio-based polyurethane with lignin showed 6–32% savings of GHG emissions compared to the petrochemical polyurethane foam used as reference (Manzardo et al., 2019).

Among the applications that look less promising from a GHG perspective, lignin-based catechol, which is a chemical mainly used for fertilizer but also fine chemicals such as perfumes, shows savings of 2% (Montazeri and Eckelman, 2016), which is very minor compared to the uncertainty involved. Bio-based asphalts (Tokede et al., 2020) also showed low GHG emissions savings (about 5%) compared to conventional asphalts and this depends also on the percentage of lignin replacing bitumen assumed. On the other hand, the climate change impact per kg of kraft lignin assumed by Tokede et al. was also the lowest shown in Fig. 6. Changing methodological assumptions or Kraft mill might lead from a low GHG saving of emissions to higher impact than conventional asphalts.

3.3.3. Environmental performance of lignin-based products

In this section, the performance of lignin-based products is discussed considering other environmental impacts in addition to climate change with examples from studies.

Concerning bio-based adhesives derived from lignin, three LCAs were conducted and divergence was found in the insights on the overall performance in comparison with the petrochemical counterparts depending on the type of lignin considered and assumptions made. In particular, Arias et al. (2020) assessed two bio-adhesives used for manufacturing wood panels derived from two different lignins (from Kraft and organosolv) (Arias et al., 2020). These adhesives were compared with two alternative bio-based adhesives (from soy and

[2] This value was calculated from 20 kgCO_2eq per m^3 of finished fiberboard reported in Fig. 4 of (Yuan and Guo, 2017) and 154.2 kg/m^3 of ammonium lignosulfonate needed for the production of 1 m^3 of finished fiberboard (Yuan and Guo, 2017)

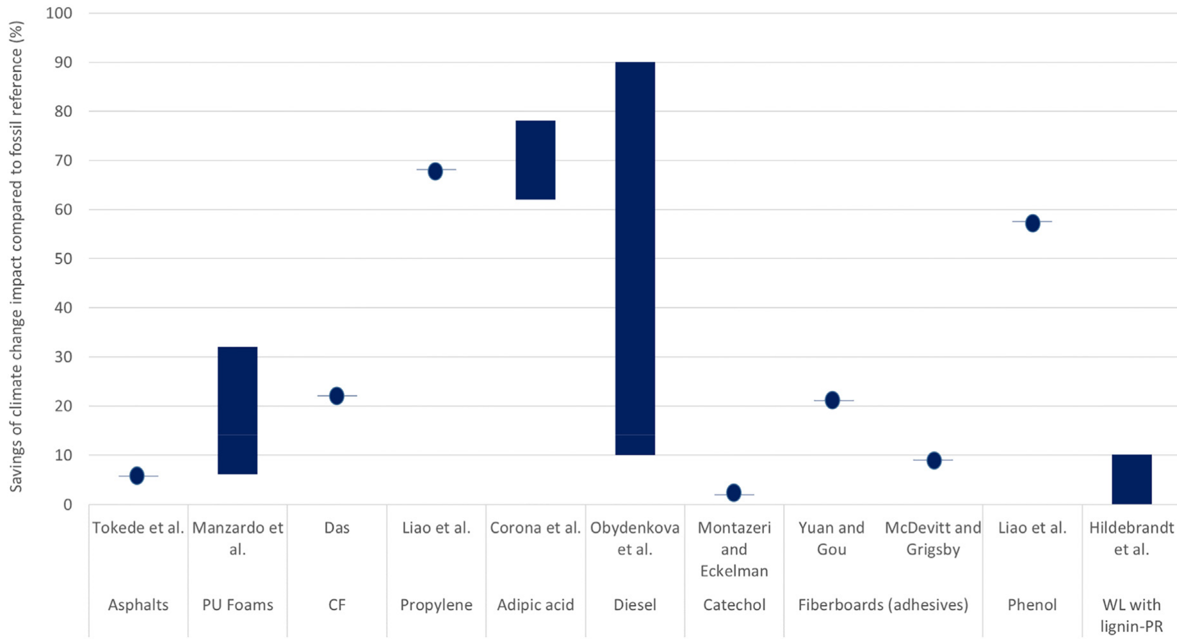

Fig. 7. Savings of climate change impact compared to fossil reference reported in the selected LCAs. PU=Polyurethanes, CF = carbon fibers, WL = wood laminate, PR = phenolic resin. The range of values from Manzardo et al. and Hildebrandt et al. refers to multiple formulations (e.g. varying shares of lignin content within the resin matrix). The range of values from Corona et al. represents the variation of the country where adipic acid is produced along with respective fossil reference and multiple feedstock scenarios. The wide range of values from Obydenkova et al. is due to testing both multiple allocation methods and alternative energy carriers.

tannin) and three conventional fossil resins (urea-formaldehyde, phenol-formaldehyde and melamine-urea formaldehyde). Nine impact categories were considered and the impacts were compared based on end-point results. On end-point bases, the comparison highlighted that lignin-based adhesives were performing much worse than other bio-based adhesives and conventional adhesives (between 2.5 and 4.5 times higher impact). On the other hand, the preliminary LCA conducted by (Yuan and Guo, 2017), based on endpoint results, concluded that wood panels made using lignosulfonates-based adhesives are environmentally better than wood panels using urea-formaldehyde. Similarly, also McDevitt and Grigsby concluded that Kraft lignin-based adhesives are environmentally better than urea-formaldehyde adhesives (about 22% lower impact on weighted bases (McDevitt and Grigsby, 2014)).

With respect to lignin-based polyurethane foams, Manzardo et al. (2019) found better performances compared to the petrochemical foam taken as reference in five out of the eight impact categories considered. In particular, they offer 9–33% savings in photochemical ozone formation, up to 29% in terrestrial eutrophication, 6–43% in freshwater eutrophication, and 14–36% in depletion of abiotic resources (elements).

Concerning lignin-based phenolic resins, the LCA of Hildebrandt et al. (2019) showed that wood-based fiber laminates using lignin-based phenolic resins perform better in nine out of eleven categories (with achievable reduction potentials up to 39% depending on the impact category considered) (Hildebrandt et al., 2019).

Concerning lignin-derived fertilizers, Montazeri and Eckelman assessed lignin-based catechols which are chemicals mainly used for the production of fertilizers. Their assessment showed that lignin-derived catechol, beyond negligible climate change benefits (see Fig. 6), potentially offer 7% and 59% environmental impact reductions respectively for ecotoxic effects and depletion of fossil fuels (Montazeri and Eckelman, 2016). However, in the other seven environmental impact categories, the fossil route was preferable (Montazeri and Eckelman, 2016). In particular, the solvent (Dichloromethane) used in the lignin purification process and electricity for lignin depolymerization were found as the dominant contributors to the environmental impacts of the bio-based route (Montazeri and Eckelman, 2016). Krzyżaniak et al. (2019) assessed the final application (cultivation using different fertilizers) and assessed the same impact categories of (Montazeri and Eckelman, 2016) concluding as well that lignin-based fertilizers are slightly better than mineral fertilizers. Specifically, Krzyżaniak et al. (2019) found that lignin was better than mineral fertilizers in four impact categories (climate change, particulate matter, terrestrial acidification and freshwater eutrophication) while worse in freshwater ecotoxicity, terrestrial ecotoxicity and human toxicity. Concerning the use phase, compared to mineral fertilizers, lignin-based fertilizers showed higher sequestration of organic carbon and lower field emissions in terms of particulate matter and acidification/eutrophication. For fossil depletion, the impact of lignin used as fertilizers was slightly worse. What appears interesting is that the categories where lignin used as fertilizer and lignin-based catechols (an ingredient for fertilizers) perform better or worse were opposite in the two LCAs (except for climate change) (Krzyżaniak et al., 2019; Montazeri and Eckelman, 2016). On the other hand, the two products assessed were not directly comparable except for the final use.

These examples show that, while lignin-based products are often preferable for climate change than their fossil counterparts, conversely, trade-offs occur in the other impact categories assessed. It is also not straightforward to summarize for which categories lignin-based products are generally better since it is very case dependent.

3.3.4. What fuel to use to replace lignin as an internal energy source?

One of the findings of the review is that the impact of lignin and lignin-based products depends significantly on the type of energy source that is used to replace the burning of lignin in biorefineries and paper mills.

Concerning Kraft lignin, most of the studies found that natural gas used to replace black liquor is the main environmental hotspot for most impact categories. However, Bernier et al. (2013) argue that using natural gas is one of the main drivers to equip old mills with lignin extraction since it is a cheap fuel whose combustion causes much lower local atmospheric emissions than black liquor. Alternatively, the additional steam required caused by lignin extraction can be provided by burning excess hog fuel (if available along with spare boiler capacity) (Bernier et al., 2013). In existing pulp mills, there is also a fraction of lignin that can be extracted without requiring an increase of natural gas consumption for energy use in the pulp mill (only a minor increase for the lignin extraction process) (Culbertson et al., 2016). This fraction of lignin has a lower impact than the part that requires additional energy for the pulp mill.

Secchi et al. (2019) performed an LCA on the effect of lignin extraction on the environmental impact of ethanol produced by a biorefinery and pulp produced by a Kraft mill. In particular, for the biorefinery, 40% of the lignin cake was assumed to be diverted from the internal energy use while for the Kraft mill, 50% of the black liquor was assumed to be removed (Secchi et al., 2019). Various fossil and biomass sources for energy production were considered to replace the fraction of lignin originally used as fuel and multiple allocation methods were applied (mass, energy and economic values). The results and conclusions were based on single score impacts calculated with ILCD normalization factors (Benini et al., 2014) combined with equal weighting. The two main outcomes of the study were that 1) the impact of ethanol and pulp does not increase if lignin is extracted and 2) using natural gas to replace lignin as an internal energy source is recommended in biorefineries while cogeneration using biomass is recommended in pulp mills (Secchi et al., 2019).

On the other hand, if the main goal of the biorefinery is the minimization of the GHG emissions and not of the total impact (climate change plus other environmental impacts more than climate change), the use of additional biomass instead of natural gas to compensate the diverted lignin might be preferable (Obydenkova et al., 2017). For example, in the case of lignin-derived transport fuels, the use of natural gas does not allow to fulfill the EU 60% GHG savings threshold of policy targets (Obydenkova et al., 2017) set by the EU renewable energy directive (EPA, 2017; European Commission, 2016) and U.S. renewable fuel standard. To fulfill this target, in the example of the biorefinery modeled by (Obydenkova et al., 2017), the use of corn stover also for internal energy purposes (and not only as feedstock for fuel production) was proposed.

3.3.5. Effects of lignin allocation on the LCAs of biorefinery products

In most of the LCAs of biorefinery, the focus was on the main products produced by the biorefinery and not on lignin, which was sometimes used for internal energy needs and some other times marketed for other purposes. This section report on how these LCAs dealt with lignin.

Turk et al. (2020) performed an LCA of nanofibrillated cellulose (Turk et al., 2020). In their study, the lignin produced from the biorefinery was considered a waste. Therefore, no impact was apportioned to lignin and the impact of one kg of nanofibrillated cellulose was as high as 800 kg CO_2eq (Turk et al., 2020). In the sensitivity analysis, mass allocation was applied to account for lignin as a by-product instead of waste. Since Soxhlet extraction and delignification represented a considerable part of the environmental burdens and were also allocated to lignin, the impact of one kg of nanofibrillated cellulose became about 400 kg CO_2eq (Turk et al., 2020). Hence, how the practitioners deal with lignin in assessing such a product has a major effect on the results.

Soam et al. (2016) assessed a second-generation biorefinery producing ethanol from rice straw in India. The study concluded that the ethanol produced offered major GHG emissions savings (77–89%) compared to gasoline. In particular, two assumptions were made: 1) the displaced electricity was coal-based electricity and 2) the carbon emissions from lignin combustion were carbon-neutral. Based on these assumptions the surplus of electricity generated combusting lignin led to major benefits (a credit of 40–45 g CO_2eq per MJ of ethanol over a total impact of about 55) (Soam et al., 2016). These same two assumptions were made also by (Shuai et al., 2016) in their assessment of ethanol from common reed produced in China. However, in their study, the credit generated by the replacement of the surplus of electricity was less important (2.5 g CO_2eq per MJ of ethanol over a total impact of 17.5 g CO_2eq). Hence, based on the amount of the surplus of electricity, the surplus of electricity might lead to a major credit or a small credit (this does not only depend on the quantity but also on the electricity mix displaced). One should wonder if, in the cases where a major credit was given, the surplus of electricity was a by-product of the system or the main product of the system. In the second case, the use of substitution would not be appropriate since the principle of physical/economic significance would not be respected.

Nascimento et al. (2016) assessed cellulose nanocrystal from coconut fiber. In the production process, lignin was produced as a by-product and was marketed. The two main environmental hotspots of the process were identified as the production of acetic acid and the electricity required. As an alternative, lignin could be burned for the internal electricity needs of the biorefinery. However, the results of the LCA conducted by Nascimento et al. (2016) showed that the use of lignin as an internal power source led to environmental impact increases in four (climate change, terrestrial acidification, water body eutrophication and marine eutrophication) out of six impact categories assessed. The main reason was that, if lignin were no more a by-product but were internally consumed, the impacts from milling and pulping processes would be attributed to cellulose nanocrystals only and would not be allocated anymore also to lignin. Thus, the benefits from the power generated from burning lignin were lower than the impact originally allocated to lignin in these four impact categories. However, looking only at the functional unit expressed in terms of nanocrystals and not to the overall system, this conclusion might have been affected by the mass allocation applied. In fact, if economic allocation were applied, a lower impact would have been allocated to lignin since the price of cellulose nanocrystals is higher than lignin (Nascimento et al., 2016).

Budsberg et al. (2016) noticed that the production of hydrogen is often the main environmental hotspots of the production of bio-jet fuels. In the biorefineries producing bio-jet fuels, often hydrogen is produced from natural gas and lignin is used as fuel for the internal demand for heat and electricity (Budsberg et al., 2016). Budsberg et al. wondered if, environmentally, this is the best solution or is better to gasify lignin to produce green hydrogen for internal needs. From a climate change perspective, their LCA showed that the current solution is better than using lignin to produce hydrogen (the impact would increase by 10%) due to the GHG emissions caused by the replacement of lignin with natural gas for the production of internal energy needs. However, their LCA showed that if hog fuel could be used instead of natural gas, then using lignin for hydrogen production could lead to important savings of GHG emissions (order of 50%) (Budsberg et al., 2016).

4. Conclusions

Lignin, which is a by-product of biorefineries and pulp mills, is currently (mainly) used for bioenergy but can be utilized to produce lignin-based products replacing fossil counterparts in various sectors. In the near future, the electricity mix is expected to be rapidly decarbonized. On the contrary, transport, heat and materials are much harder to decarbonize. Hence, we can expect that the use of lignin for producing bio-based products will start to play a more important role in the next decade. In parallel, the sustainability performance of such products should be monitored using accredited tools. Among them, LCA is the best candidate for sustainability assessment in bioeconomy sectors. Despite LCA is a standardized method, various methodological choices have to be taken by the practitioners leaving room for possible inconsistencies between the results of different studies. Forty-two studies concerning LCAs of lignin and lignin-based products were reviewed to detect the differences (and possible inconsistencies) in the application of the methodology and their influence on the life-

Table 1
Lesson learned and recommendations.

Lesson learned	Recommendations
Only a few studies considered the use phase and end of life of the product (see Section 3.2.1).	The end of life should be considered especially for the comparison between lignin-based products and their fossil counterparts. Realistic, average waste management should be investigated as well as the carbon degradation of lignin during the use phase and waste management.
78% of the LCAs did not explicitly specify the type of modeling approach followed i.e. attributional or consequential (see Section 3.2.1).	The approach followed should be specified since it helps to select properly the unit processes to be included in the system boundaries, the type of data to be used and what type of system expansion method is possible.
Most of the studies adopted a simple functional unit e.g. based on a mass basis (see Section 3.2.2). This type of functional unit does not state how well each product fulfills the function of the system.	In the definition of the functional unit, how well the function of the product system is fulfilled should be accounted for. Only if the function is fulfilled similarly by the investigated options, a simple functional unit could be used.
While climate change was investigated in all the selected LCAs, other impact categories were often neglected (see Section 3.2.4). Especially for land use and water use/depletion, one of the main reasons was probably the absence of consensus on the impact assessment method.	All relevant impact categories should be included. In particular, land use and water use are important for bio-based systems. The assessment method should be selected based on the recommendations from trusted sources (e.g. EU LCA guidelines). If an impact assessment method were not used, at least an estimation of the amounts of land and water needed should be provided.
In almost all LCAs (especially of biorefineries), data were mainly obtained from laboratory and process modeling (see Section 3.2.5). Few studies used primary (actual) data for kraft lignin production. These studies were also the main sources used in the LCAs that relied on secondary data.	It is important to collect new transparent primary data for lignin production from real operation at a large scale which are currently missing in the public domain.
Dealing with multifunctionality was identified as the major methodological problem in the assessment of lignin and lignin-based products since lignin is always the result of a multi-output process. Therefore, LCAs of lignin products are affected by higher uncertainties compared to other bio-based products. A standardized method for the selection of the allocation method exists and is provided by ISO 14044:2006. However, there is no shared interpretation in the LCA community and in LCA guidelines. As a result, multifunctionality practices in LCAs of lignin-based products are not harmonized (see Section 3.2.3).	A consensus on the interpretation of ISO 14044 hierarchy to deal with multifunctionality is urgently needed to have a standardized LCA approach to account for co-products. This is a problem not only of lignin production systems but of all bioeconomy. A ISO-compliant framework that keeps into account the major critical aspects identified during the review (e.g. the application of substitution without a check on physical/economic significance) is needed.
Biogenic carbon dioxide is treated differently in the studies (see Section 3.2.6). Often, it was treated as carbon-neutral while in other cases a carbon intake was accounted for based on the carbon content of lignin or based on the carbon intake during biomass growth. Moreover, often the carbon credit was integrated into the cradle to gate results for climate change and the accounting of the biogenic carbon intake was a key element for the better performance of lignin-based materials compared to their fossil counterparts. However, recent guidelines e.g. EU PEFCR recommends reporting the biogenic carbon separately in LCAs ending at the gate.	The first recommendation is that the choice regarding biogenic carbon accounting should be stated clearly in the LCA and also next to where the climate change results are shown. This would allow the user of the LCA to have a clear picture and, in case the LCA results were used for other studies, a double counting (or omission) in the assessment of the end-of-life phase would be avoided.
Comparing single lignin-based ingredients (e.g. lignin binder) with fossil-based ingredients (e.g. bitumen) can provide an erroneous picture. In fact, the utilities required during the production of the final application might change if lignin is used in the product. Moreover, sometimes, in order to have the same performances, also the other ingredients in the mixture have to be changed (e.g. proportions).	To have a full picture and a correct estimation of the potential savings that can derive from using lignin-based products to replace their petrochemical counterparts, the LCA should compare the final application (e.g. asphalts) rather than the chemical ingredient with its petrochemical counterpart.
The impact of biorefinery products (e.g. ethanol) is largely affected by how lignin is used in the system and how the practitioners deal with lignin in the LCA.	If lignin is exported as a product from the system, multiple functional units should be used. If the goal of the LCA requires the determination of the impact of a single function, a functional unit should be assigned to lignin and another one to the main product which is the focus of the investigation. Only in this way, the user of the LCA can (easily) understand how lignin was considered in the LCA and the effects of the allocation procedures applied.
Often, there is a trade-off between GHG emissions and economics in the selection of the best fuel to replace lignin in internal uses. Moreover, extracting lignin instead of using it for internal energy needs might affect importantly the environmental performance of biorefinery products.	The LCA should be conducted based on the most probable fuel and possible alternatives should be investigated by sensitivity analysis

cycle environmental impacts. Moreover, the climate change impact reported in LCAs of lignin and the GHG savings allowed by lignin-based products were quantitatively compared. The importance of other impacts in the comparison between lignin-based products and counterparts was also discussed with examples from the studies. The lesson learned from this exercise and possible recommendations are provided in Table 1.

Our list of recommendations could promote good practices and increase methodological harmonization in assessing the environmental sustainability of lignin and lignin-based products using LCA. On the other hand, even following these recommendations, conducting an LCA of lignin remains challenging from a methodological perspective. For this reason, the user of the LCA results needs to be very careful in checking the assumptions made by the practitioners. Moreover, using the results from different LCAs that compare lignin-based products and fossil-based products and concluding what option is the best is not straightforward. The reasons are both technical (e.g. using lignin as an ingredient changes also other parameters and lignin can substitute other ingredients with different shares) and methodological (allocation plays a major role, as does the way biogenic carbon storage is accounted). Beyond this, the reviewed LCAs showed that often lignin-based products offer better environmental performances than fossil-based products (especially for climate change), but if lignin is diverted from an energy application, the most probably alternative can have a substantial influence on the overall climate impact.

Declaration of competing interest

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

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Appendix A. Supplementary materials

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