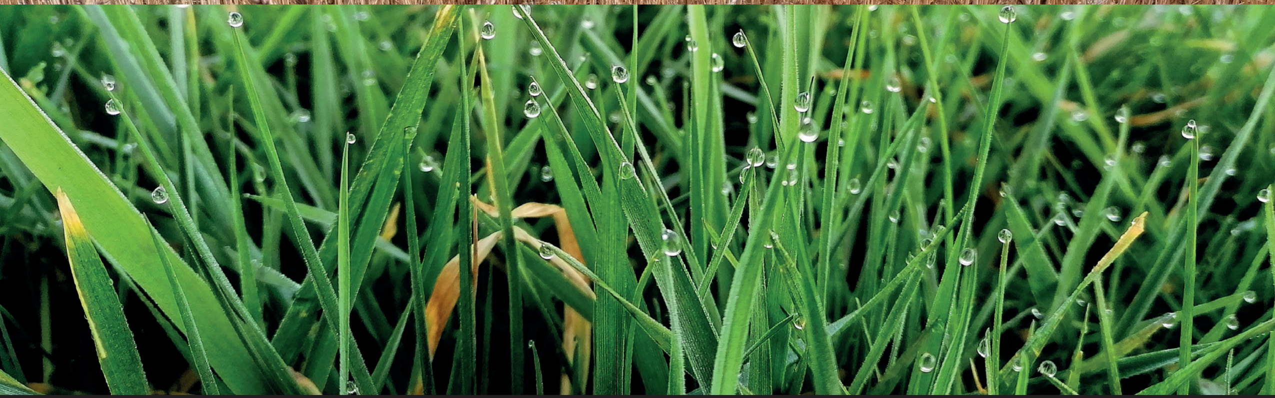

ANALYZING THE ENVIRONMENTAL IMPACT OF INNOVATIVE BIO-BASED PRODUCTS FROM MULTIFUNCTIONAL SYSTEMS

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CONTENTS

CHAPTER 1

Introduction

1.1 BIOECONOMY AND ITS ROLE IN THE EU GREEN TRANSITION

Bio-based products have emerged as a possible alternative to reduce the consumption of fossil resources in worldwide society. An economy relying on bio-based resources is often referred to as “bioeconomy” or “bio-based economy”. Using the definition of the European Commission, “bioeconomy comprises those parts of the economy that use renewable biological resources from land and sea – such as crops, forests, fish, animals and micro-organisms – to produce food, materials and energy” [1]. Hence, in simple words, bioeconomy is an economy relying on biomass conversion into a broad range of products, from more established ones such as food and feed to innovative products such as fuels for aviation and plastic materials.

The transition from the current fossil-based economy to a bio-based economy is seen as a new paradigm for a future sustainable economy, potentially generating environmental, societal and economic benefits [2–4]. Besides reducing dependence on fossil fuels, a bioeconomy is expected to reduce climate change impacts compared to the current fossil-based economy [5,6]. Such climate change benefits can be obtained by making use of the short carbon cycle of biomass and the possibility of storing biogenic carbon [4,7,8]. These environmental benefits could be accompanied by socio-economic benefits such as creating new job opportunities for sustainable economic growth [4,9].

The potential of bioeconomy to play a leading role in the sustainable transition towards climate neutrality has been widely recognized [10]. More than 50 countries have already created policy strategies related to bioeconomy [11,12]. In particular, the promotion of bio-based commodities is one of the main action plans of the European Green Deal launched by the European Commission [13]. The European Green deal looks at bioeconomy as a part of multiple policies related to renewable energy and chemicals and materials’ sustainability and circularity, and as a way to address the Paris Agreement’s climate objectives and meet the Sustainable Development Goals (SDGs) set by the United Nations [2,14,15].

Bioeconomy’s present economic volume is currently difficult to estimate due to a lack of statistics on emerging bio-based products and the fact that a broad range of industries is partially bio-based [16]. However, the European Commission-Joint Research Center has made a first attempt to estimate the value-added generated by bioeconomy based on 2017 data. In the EU-27 in 2017, the bio-based part of the economy generated €614 billion in value-added [17]. This value-added represents 4.7% of the 2017 EU-27 GDP [17]. The EU-27 bioeconomy employed 17.5 million full-time jobs in the same year, i.e. 8.9% of the EU-27 workforce [17]. Nowadays, the bioeconomy sectors employing most of the workforce are still the traditional sectors i.e. agriculture and food industries (see Table 1 for a detailed breakdown per sector).

Nonetheless, in the last decade, novel bio-based products have emerged in sectors traditionally dominated by fossil (or mineral) resources. Over the last decade, the bio-based chemicals and plastics industry experienced a 45% total added value growth accompanied by a 10% increase in the number of jobs created [18]. Electricity from biomass doubled its added value and almost tripled the number of jobs [18]. Conversely, the labor force in traditional bioeconomy sectors i.e. agriculture, and aquaculture, textiles and furniture/wood products declined significantly (see Table 1).

Table 1. EU-27 bioeconomy value-added and employment per sector (2017 data based on [17] and [18])

Sector	Value added 2017 (billion €)	Total growth of value added 2008-2017 (%)	Employment 2017 (million jobs)	Employment growth 2009-2017 (%)
Agriculture	189	37%	9.3	-16%
Forestry	25	50%	0.5	7%
Fishing and aquaculture	7	24%	0.2	-3%
Food and beverages	215	23%	4.4	6%
Natural textiles	21	16%	0.7	-21%
Furniture and other wood products	47	18%	1.4	-8.5%
Paper	42	25%	0.6	2%
Bio-based chemicals and plastics	60	45%	0.4	10%
Liquid biofuels	3	35%	0.02	-26%
Bioelectricity	4	105%	0.02	185%

The growing trend of innovative bio-based commodities is expected to continue under the push from the European Green Deal. In particular, the EU bioeconomy is expected to rise from the current 4.7% of GDP to 7.4% by 2050 to limit the global temperature rise to 2 °C above pre-industrial levels [19]. Besides the expected growth in the production of food products, most of the bioeconomy growth will be driven by innovative bio-based commodities i.e. bio-based chemicals, plastics and fuels [19]. Between 2020 and 2030, all these innovative commodities are expected to grow between 25% and 35% to reach the 2 °C target in 2050 [19]. In particular, the EU Bio-based Industry Consortium already targets at least 25% growth by 2030 [20]. Given this significant growth in the short term, there is an urgency to understand the climate and environmental impacts of innovative bio-based products.

1.2 THE INTEREST IN LOCALLY SOURCED AGRO-INDUSTRIAL AND FOOD-PROCESSING BY-PRODUCTS

Beyond the shared acknowledgement of the bioeconomy role in the EU green transition, there are various points of view regarding how a future bio-based economy should look, within which boundaries it should operate and its relationships to the rest of the economy [4,21]. In fact, the promotion of bio-based products in Europe has led to significant imports of bio-based products from outside the EU [22]. This fact may generate conflicts of interest in biogenic resources' international trade since sustainable biomass is a scarce resource and its availability in the EU is not sufficient to meet EU ambitions [2,21,23]. For example, EU ethanol imports doubled in 2019 compared to 2018, with imports from the US increasing by 233% [24]. Similarly, 43% of the bio-based solvents consumed in the EU are imported [25].

In particular, among the most discussed trade-offs generated by bio-based production, there are the competition for crops that might threaten food security [26] and the impacts on biodiversity of large-scale crop production [27]. Given the concerns about food security and land competition, locally sourced bio-based "residues" have attracted increasing attention to extend the amount of bio-based products produced sustainably in the EU [28,29].

This thesis investigates several innovative bio-based products from this type of feedstock that have been either recently commercialized or are close to reaching the market. The explored feedstocks are food processing residues i.e. potato by-products and used cooking oil, forestry residues i.e. wood chips and a by-product from paper/pulp industries i.e. lignin.

Besides avoiding land competition with food, these bio-feedstocks can be employed in biorefineries and bioenergy technologies that rely on local availability, reducing fossil fuel dependency and biomass imports [30]. Moreover, this type of residual by-products are usually cheaper than crops from dedicated cultivation [30] and their production usually has a lower climate change impact than dedicated crops [31–34].

1.3 LCA AS A KEY TOOL TO MEASURE THE IMPACTS OF INNOVATIVE BIO-BASED PRODUCTS

The last two decades have seen the development of numerous bio-based alternatives in sectors that have been (and currently still are) dominated by the fossil industry. Among these products, we can find bio-based plastics (in some cases also biodegradable ones), bioenergy, bio-based fuels, bio-based chemicals and other unconventional bio-based products (e.g. asphalts). These products are named “bio-based” since they are wholly or partly derived from materials of biological origin. Nevertheless, the name “bio-based” does not directly imply being environmentally friendly and their potential environmental impacts need to be minimized using a proper monitoring tool.

Life Cycle Assessment (LCA) is one of the main tools used by worldwide policymakers, scientists, and companies to monitor the environmental impact of bio-based products [9,21,35–37]. LCA is an internationally standardized method to assess the environmental impacts of products or services in all their life cycle, from raw materials extraction to waste management [38]. The results of LCAs have already been used to build environmental sustainability criteria in several policy instruments and mechanisms covering several bioeconomy sectors e.g. biofuels [3,35,39–41].

So far, policy instruments typically only cover a few impact categories (mostly climate and/or energy demand). However, trade-offs typically exist between bio-based and petrochemical products [7,42]. When the scientific literature considered other categories, the LCAs of bio-based products often showed higher eutrophication, acidification and water depletion impacts than their petrochemical counterparts due to biomass production [43,44]. So, it is necessary to apply LCA to provide a comprehensive overview of environmental impacts to understand if these tradeoffs could also be observed for the emerging products made from bio-based “residues” streams.

1.4 CHALLENGES OF MODELING BIO-BASED PRODUCTS IN LCA

The user of results from LCAs of bio-based products should be aware that several methodological uncertainties exist despite the existence of ISO LCA methodology standards. In particular, there are three major modeling uncertainties behind the LCAs of bio-based products: multifunctional processes, land-use changes (LUCs) and biogenic carbon [45–49]. Of these three methodological complexities, this thesis focuses on modeling multifunctional processes. This issue is considered one of the most "controversial" in the LCA scientific literature and with low convergence in LCA guides [50–55].

Since most innovative bio-based products are produced from multi-output systems [56,57], their LCAs regularly run into the "multifunctionality issue". The multifunctionality issue affects all LCA environmental impact categories, whereas LUCs and biogenic carbon predominantly affect the climate change impact of bio-based products. In addition, LUC impacts are generally more significant for products made from dedicated crops than bio-based by-products [58]. For these reasons, a major focus was given to the multifunctionality issue in this thesis.

Most methodological decisions made in LCA strictly depend on the goal and scope of the LCA. The goal and scope of the LCA have a direct link with the selection of the modeling approach and life cycle stages. For example, the modeling approach influences multifunctionality practices and the inclusion/exclusion of land-use changes in the system boundaries [59–61]. The literature distinguishes between two "major types" of modeling approaches i.e. attributional and consequential LCAs [59,62].

Attributional LCAs aim to understand which environmental impacts should be attributed to an economic activity [51]. Consequential LCAs aim to understand the direct and indirect changes in worldwide environmental impacts caused by changing or adding to some degree an economic activity [51]. Attributional LCAs usually apply an allocation share (e.g. based on the mass of co-products) to partition the environmental impacts of multifunctional processes and rarely include (indirect) LUCs [45,63,64]. In consequential LCAs, the so-called substitution method is often used to avoid allocation and both direct and indirect LUCs are generally included in the life cycle inventory [45,63,64].

The selection of the life cycle stages has a major effect on the modeling of biogenic carbon. The LCA modeling of biogenic carbon is under debate mainly for two aspects i.e. the credits from temporary or permanent biogenic carbon storage and the timing of carbon emissions [65]. The practices in the LCA literature regarding these two modeling aspects mainly vary depending on the inclusion or not of the end-of-life in the accounted life cycle stages and the type of product i.e. durable application or not (e.g. a fuel) [65].

1.4.1 THE LCA MULTIFUNCTIONALITY ISSUE AND THE ISO HIERARCHY INTERPRETATION ISSUE

The multifunctionality problem occurs every time a process provides more than one function. Since the LCA method calculates environmental impacts per functional unit, it is necessary to find a way to allocate the environmental impacts of a process between these functions or avoid such allocation [38]. Multifunctionality is usually present in bioeconomy systems since biorefineries, bioelectricity systems, paper industries and food industries produce more than one single economically valuable product.

ISO 14044:2006 recommends a three-level hierarchy that practitioners should follow in dealing with this problem:

1. The allocation should be avoided either by dividing the process into sub-processes or by system expansion.
2. Performing allocation based on the “physical relationships” between the co-products.
3. Using allocation methods reflecting “other relationships” that are present between the products (for example, their economic value).

Unfortunately, selecting the best allocation rule based on the abovementioned hierarchy is difficult. As pointed out by Wilfart et al. [66] among others, one of the reasons is that “following this decision hierarchy encounters several unresolved issues that ultimately result in a variety of strategies, some of which are supported only by dubious interpretations”. Consequently, the literature and sectorial/national ISO-compliant guidelines provide different (and sometimes contradictory) rules for applying such a hierarchy in practice [66–69].

There are two major debates on the interpretation of the ISO multifunctionality hierarchy. As explained in section 1.4, selecting the modeling approach (attributional or consequential) is crucial for selecting the multifunctionality method [61,69–71]. The first debate regards the use of the substitution approach as a system expansion method in attributional LCAs (ALCAs). This is not perceived as rationale by many LCA experts [51,52,68,71–73] since 1) ALCA modeling should not rely on perturbation logic or counterfactual notions, such as avoidance [51,59] and 2) the sum of the impacts accounted by ALCAs should sum up to the worldwide impacts, and this would not be valid anymore if substitution were applied [71,74]. However, some guides and authors consider system expansion as a synonym for substitution [75]. On this basis, substitution should also be used in attributional modeling instead of allocation since ISO prefers system expansion (e.g. [76–79]). This concept should be tracked back to the conceptual equivalency between substitution and system expansion illustrated by [80,81].

The second debate regards the meaning of “physical relationships”. Some guides and practitioners interpret the term “physical relationships” as “physical causal” relationships i.e. reflecting quantitatively how a change in the product mix changes the inputs, outputs and impacts of the system (causality concept) [41,82]. However, some guides and practitioners interpret allocation based on “physical relationships” as an allocation based on a physical parameter [83–85].

The rationales of the current interpretations of the ISO 14044 hierarchy have been summarized by Pelletier et al. [86]. However, it is not fully understood how the different implementation practices of the ISO hierarchy developed and how the discrepancies were generated, leading to current inconsistencies in various ISO-compliant LCA guides.

To picture the relevance of this interpretation problem, we can take a widely investigated case regarding a well-established product as an example, i.e. meat products. For the same meat production system, up to 7 different allocation methods can be applied following different LCA ISO-compliant guidelines [66], leading to a significantly different environmental impact for meat products [51,66,87]. As a result, since milk is a co-product of meat production, interpreting and comparing the results of LCAs of dairy products is challenging [87–92]. Economic allocation between milk and beef favors beef production, while substitution drastically reduces the environmental impact of milk [93]. Similar issues were pointed out by

Agostini et al. [94] in their review of the 100 most cited articles on LCA of bioenergy and by Brankatschk and Finkbeiner [69] for agricultural systems.

While the multifunctionality uncertainty on the climate change impact of established bio-based products has been broadly investigated, the effect of this issue on innovative bio-based products is still unknown. In fact, many bio-based products have been recently commercialized for the first time or are close to commercialization [10,95]. Moreover, multifunctionality practices in LCAs of bio-based products largely vary depending on the specific bioeconomy sector and the leading ISO-compliant guide followed by practitioners of that sector. While inconsistencies have been remarked already by several authors [52], a quantitative analysis of such inconsistent practices is still missing.

The relevance of the problem becomes even more important considering the following three facts:

1. the LCA literature has broadly acknowledged that using a different allocation method to assess the same bio-based product can change the results significantly, leading to conflicting recommendations to the study's commissioner and/or policymakers [50,96].
2. previous literature showed that comparing two products fulfilling the same function with multifunctionality practices consistent on both sides is important [69,97]. However, the same process might provide products with multiple applications for various sectors and actors involved. Consequently, part of the environmental burdens are probably "either unaccounted or doubly accounted for" [69], leading to limited comparability of the results.
3. the environmental performance of bio-based products is often benchmarked with petrochemical products, which are as well affected by the same multifunctionality issue at the refinery level. So, comparing the environmental impact of bio-based with petrochemical products has a double uncertainty related to the multifunctionality issue on both sides.

1.4.2 THE MULTIFUNCTIONALITY UNCERTAINTY IN LCAS OF BIO-BASED BY-PRODUCTS

As mentioned in section 1.2, among the main reasons for interest in bio-based products produced from bio-based "residues", there is the expected lower climate change impact than that of bio-based products from dedicated crops [31–34]. However, their lower climate change impact also originates from the allocation method applied to the bio-based by-products used as feedstock. A cut-off allocation does not assign upstream production burdens to the by-product, i.e. it is considered a “waste” in ISO LCA terms. Economic allocation assigns a much lower upstream impact to the by-product due to its lower market price than the main product.

A recent meta-analysis [65] of 30 LCA studies of plastic materials from bio-based residue or side streams highlighted that the most common multifunctionality practices are using a cut-off approach, i.e. no upstream production burdens are allocated to the feedstock (9 out of 30) and economic allocation (7 out of 30). Several studies did not clearly specify the allocation method adopted (5 out of 30). A few LCAs used mass allocation (3 out of 30). Other studies used system expansion to account for the consequences of redirecting the by-product feedstock from their current use (5 out of 30). One study compared economic allocation and system expansion [98].

Mass allocation assigns the same environmental impact per unit of mass to by-products and main products (e.g. 1 kg of rice and 1 kg of rice straw), leading to a much higher environmental impact for by-products compared to the impacts resulting from cut-off and economic allocations [65]. Accounting for the environmental consequences of diverting the by-product feedstock from their alternative fate is in line with consequential modeling and leads to a higher impact than cut-off allocation and sometimes also of economic allocation depending on the current use of the by-products [48,98,99]. However, the selection of approaches to deal with multifunctionality in the LCA literature is often inconsistent with the study’s goal and consequent modeling choice. As highlighted by a recent review of the 100 most cited LCAs of bioenergy products, the choice of the modeling approach based on the goal of the study, consequent selection of unit processes to be included in the system boundaries and multifunctionality approaches are made inconsistently in the majority of these 100 LCAs leading to misleading or inconsistent results in 67 of them [94].

Based on the experience gained by recent literature on established bio-based products from low-economic/physical significance streams, the environmental impact of this type of product is expected to be more affected than products from dedicated crops by the LCA multifunctionality uncertainty [55,57]. Given the lack of a shared interpretation of the ISO multifunctionality hierarchy and consequent inconsistency in multifunctionality practices, significantly different LCA results for products from low physical/economic magnitude flows like by-products are expected [55,57].

The reason for the high variability of LCA results for such products is straightforward for allocation with the following example. Suppose a pulp mill extracts lignin from the black liquor instead of using it as a fuel. Lignin becomes a co-product. The main product i.e. pulp, represents almost 90% of the mass of the two co-products (pulp and lignin) and a bit more than 90% of the economic revenues generated [100]. Suppose a slightly different assumption/data (e.g. on the price or moisture content), the allocation share of pulp could probably change to e.g. 85% and that of lignin (i.e. the by-product) to 15%. Therefore, the environmental burden allocated to the lignin by-product would increase by 50%, while the impact of the main product, i.e. pulp, would decrease only by 6%. Hence, the environmental impact of bio-based by-products is highly sensitive to the input data e.g. moisture content or prices used for determining allocation factors. As a result, the carbon footprint of one kilogram of lignin from the same pulp mill can vary between a negative impact ("substitution-based allocation") and 4 kg of CO_2eq/kg, by just varying the allocation method [55]. Consequently, the savings of climate change impact allowed by lignin-derived adipic acid can range between −90% (savings) and + 100% (an increase of impact) in the literature [67].

Therefore, it is important to increase consistency in selecting the multifunctionality practices to avoid generating arbitrary or extreme results with consequent erroneous interpretation and misleading recommendations to the study's commissioner and consequent decision-making [57,67]. Given the expected growth of innovative products from bio-based by-products/residues reaching the market, it is necessary to quantify the multifunctionality uncertainty behind the reported environmental impacts. However, the multifunctionality uncertainty for bio-based by-products has been investigated only incidentally so far when dealing with the climate change impact a specific product and rarely taking a holistic perspective on multiple products and at the same time on multiple impact categories [57].

1.5 RESEARCH OBJECTIVES AND STRUCTURE OF THE THESIS

This thesis aims to analyze the life cycle environmental performance of several innovative bio-based products from agro-industrial and food-processing by-products compared to their petrochemical counterparts. Since multifunctionality modeling causes high uncertainties for bio-based by-products, a major focus of this thesis is LCA multifunctionality. Accordingly, multifunctionality practices and uncertainties were deeply discussed for all investigated products for the entire spectrum of indicators considered useful for decision-making.

The ISO 14044:2006 hierarchy is the key selection tool for multifunctionality practices, but various ISO-compliant LCA guides have interpreted it differently. These different interpretations have led to inconsistent multifunctionality practices in LCAs of bio-based products. However, how these interpretations originated is partly unclear. Hence, the first research question of this thesis is:

1. What multifunctionality practices are adopted in LCAs of bio-based products and how can the consistency of the life cycle inventory model be improved?

A first review looks at multifunctionality practices and their effect on results' comparability for one of the feedstocks of interest i.e. lignin. A second review quantified the practices in implementing ISO 14044 multifunctionality recommendations in all LCAs of multifunctional processes in the literature (94% resulted in being somehow related to bio-based products). The same review investigates the debates on the interpretations of the ISO multifunctionality hierarchy and their historical origins.

Afterwards, this thesis aims to fill the lack of environmental LCAs of innovations from bio-based by-products that have been either only recently commercialized or are close to commercialization.

Hence, this thesis aims to answer the following research question:

2. What is the environmental impact of innovative bio-based products made from by-product/waste streams compared to their fossil counterparts?

Among products from bio-based "residues", the following products were analyzed: *polypropylene from used cooking oil* (Chapter 4), *bioenergy from wood chips* (Chapter 5), *bio-jet fuel from potato peels* (Chapters 6) and *lignin-based asphalts* (Chapters 7). An LCA was conducted based on primary data for their innovative conversion technologies for all these products. Moreover, LCAs of *other bio-based products from lignin* (Chapter 2) from the literature were also reviewed.

These emerging products belong to different sectors within bioeconomy. Therefore, they were considered suitable to provide an overview of the environmental impacts of innovative bio-based products in a holistic way. Compared to LCAs of other products from bio-based by-products in the literature, the spectrum of the environmental impacts was broadened to other categories than just climate change, such as acidification, eutrophication and human toxicity. In this way, it is possible to understand if the typical environmental trade-offs of bio-based products from dedicated crops also apply to products from bio-based by-products.

Based on the insights gained from their LCAs, e.g. on their environmental hotspots, this thesis aims to provide a list of recommendations to produce them sustainably. Potentially, these recommendations can be extended to similar innovative bio-based products to guide investment and research towards environmental sustainability.

As abovementioned, the environmental impacts of products from low-economic/physical significance streams are usually significantly affected by the multifunctionality issue. So, this thesis aims to answer a third research question:

3. How are the LCA results of such products influenced by the approach adopted to deal with the multifunctionality issue?

The answer to this research question is provided by broadly discussing the effect of the allocation approaches among the ones allowed by ISO 14044:2006. To show the magnitude of multifunctionality uncertainty on the environmental impacts of the investigated products, performing various sensitivity analyses is necessary. This exercise is essential to detect the impact categories for which the comparison with petrochemical products is hampered by such uncertainty. Based on the insights from the case studies and the reviews, this thesis aims to provide several recommendations for good multifunctionality practices for bioeconomy products.

Table 2 provides an overview of the topics of each chapter and their respective research objectives.

Table 2. Overview of chapters and corresponding research objective

Chapter	**Topic**	**Research question**		
		1	2	3
2	Review of Life Cycle Assessments of lignin and derived products: lessons learned	X		
3	Reviewing ISO compliant multifunctionality practices in environmental Life Cycle modeling	X		
4	Kraft lignin as a bio-based ingredient for Dutch asphalts: an attributional LCA		X	X
5	Combining biomass gasification and Solid Oxid Fuel Cell for heat and power generation: an early-stage Life Cycle Assessment		X	X
6	Attributional and consequential LCAs of a novel bio-jet fuel from Dutch potato by-products		X	X
7	Environmental Life Cycle Assessment of polypropylene made from used cooking oil		X	X

2

CHAPTER

Review of Life Cycle Assessments of lignin and derived products: lessons learned

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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 was 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.

2.1 INTRODUCTION

Next to cellulose and hemicellulose, lignin is the second most abundant natural biopolymer on Earth and accounts for approximately 30% of the organic carbon in the biosphere [101]. In nature, lignin is an aromatic-composed binder that provides stiffness and strength to the stems of plants [102]. 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 [103]. Other examples of interesting applications where lignin can be used to replace conventional materials are displacing urea-formaldehyde in adhesives [104], bitumen in asphalts [105], polyacrylonitrile in carbon fibers [55], polyol in polyisocyanurate foams [103] and liquid fuels [106]. Yet, lignin is currently largely unexploited for these purposes [107]. Moreover, lignin can be used in other industrial applications that can benefit from the good surface activity of lignin [108] such as adsorbents for CO_2 capture [109,110] and catalysts [111,112].

Lignin is mainly produced as a side stream of either the pulp and paper industry or from lignocellulosic biorefineries [102,107]. In the pulping industry, lignin can be extracted from black liquor which is a by-product of the wood pulping process of pulp mills [103]. In biorefineries, lignin is obtained as a non-fermentable side stream separated during biomass pre-treatment [56]. 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 [102] but it can also be marketed [103]. Moreover, in both pulp mills and lignocellulosic biorefineries, the lignin extracted often exceeds the internal energy demand and can be sold externally [102,113].

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 [100,114]. By extracting lignin, part of the solids from black liquor are taken away and the recovery boiler can be de-bottlenecked. This de-bottlenecking can increase the production of pulp and soap generating additional revenues [55,114].

The final application of technical lignin is largely influenced by the chemical and physical characteristics of the lignin [115]. 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 [116]. Figure 1 summarizes the different types of lignin according to their extraction process and main suitable applications and indicative market price ranges for each type of lignin. Market prices depend amongst others on purity and potential application.

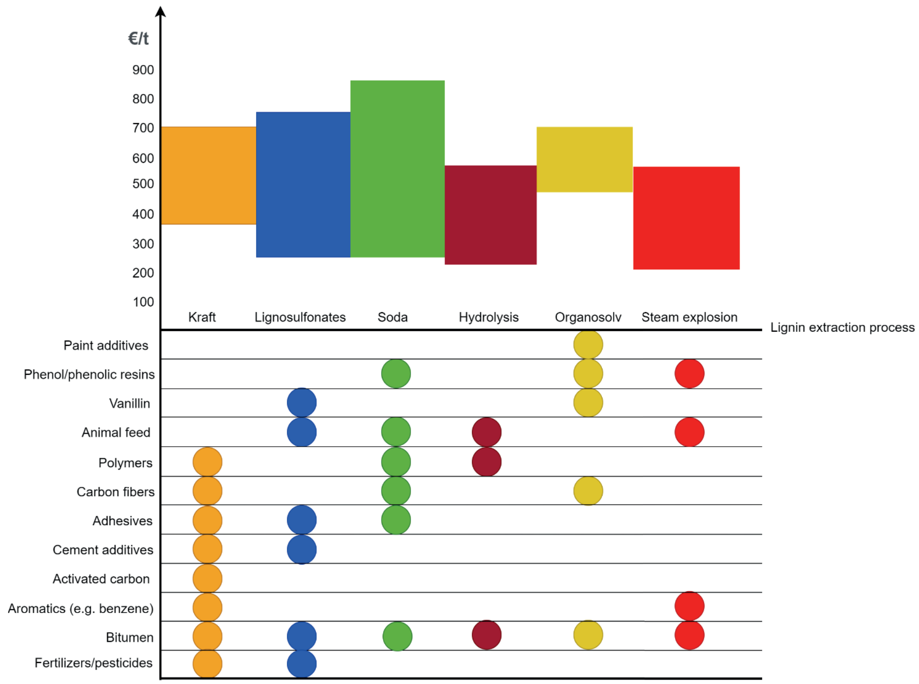

Figure 1. Example of possible applications and indicative market price ranges of various types of lignin [117–119].

The Kraft process is the dominant process in the pulping industry [120,121]. 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 [122]. 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 [122]. 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 [122]. 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 [107,123]. 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 [124]. Among novel routes for the production of bio-based aromatics from lignin, there are pyrolysis technologies, direct hydrodeoxygenation, and hydrothermal upgrading [124]. 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 [125]. Furthermore, vanillin can be produced from the oxidation of lignosulfonates that find use in foods and fragrances [126].

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) [35]. LCA is a standardized tool to model the entire life cycle of a product or system from resource extraction to final waste management [38,127]. 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 [42,128]. 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 [67]. The carbon footprint of one kilogram of Kraft lignin can vary between -23 kg CO_2eq to 4 kg of CO_2eq depending on the selected allocation method [55] 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 [67].

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.

2.2 MATERIAL AND METHODS

2.2.1 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 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

[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.

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 [129] 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.

2.2.2 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 Figure 2).

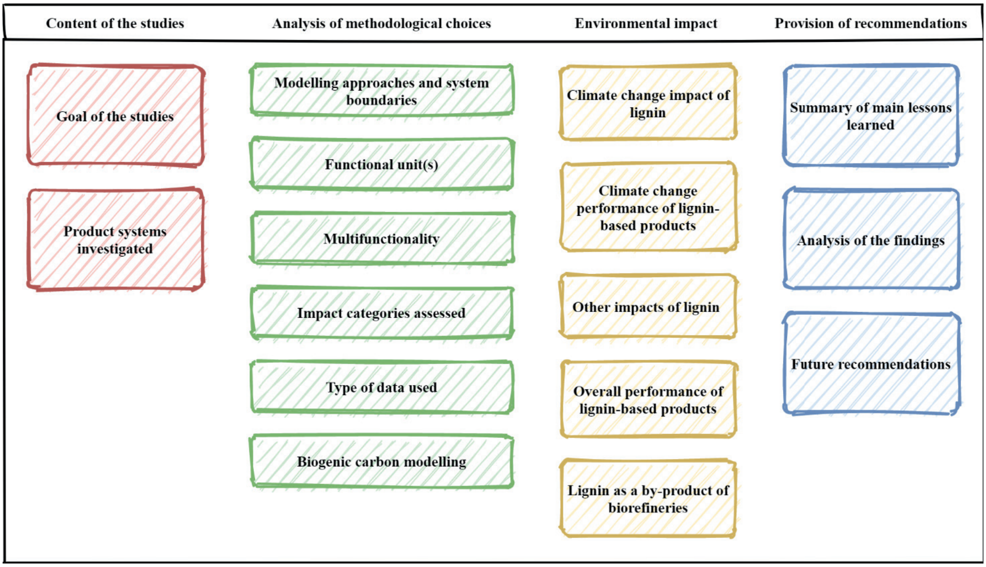

Figure 2. Steps of the review

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 identify the state of art in assessing the environmental impact of lignin products. This part of the review can be found in section 2.3.2 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 2.3.3) 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 out on climate change since it was the most considered impact in the selected studies (see section 2.3.2.4), 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 2.3.3.3 (and following). In the last step of the review, the main findings were summarised and recommendations for future research provided.

2.3 RESULTS

2.3.1 Product systems

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

- Assessing Kraft lignin (3 studies fall in this category [55,100,103]);
- Assessing organosolv lignin (1 study [129]);
- Assessing a biorefinery delignification process using natural malic acid (1 study [130]);
- 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 [67])

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 [119] 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 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 [115,131,132], *phenol* and *propylene* [133], *transportation fuels* [106], *asphalt* [134], *nanoparticles* [135], *polyurethane foams* [136], *fertilizers* [8], *vanillin* [137], *adipic acid* [138,139], *catechols* [140] and *carbon fibers reinforced polymers* [141].

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. [142], 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 [133,143].

2.3.2 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.

2.3.2.1 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 [61,144] and handbooks (e.g. ILCD handbook [145]), it is important to select the modeling approach based on the goal of the study. In a 2020 study [146], using a text mining process, it was shown that 75% of the LCAs assessing multifunctional 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 as attributional while only one article [138] 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 Figure 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 [51]. Further details about the use of system expansion to deal with multifunctionality can be found in section 2.3.2.3.

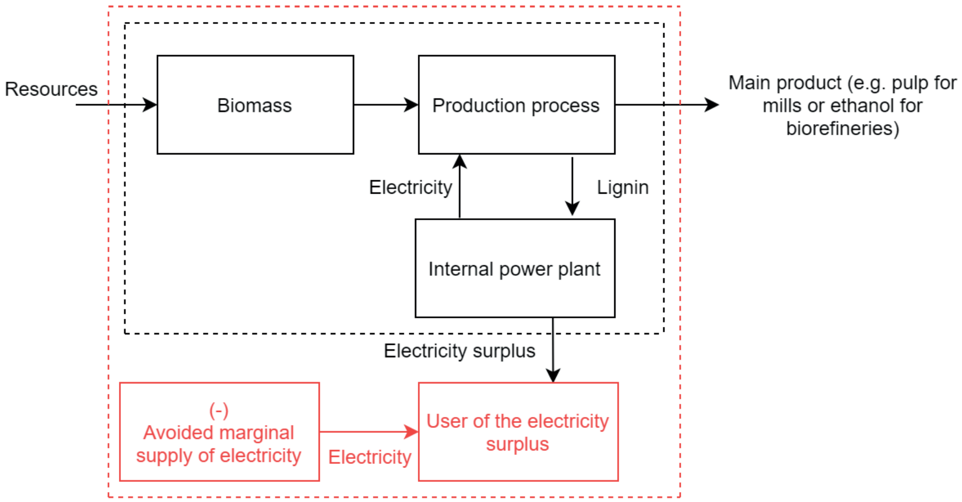

Figure 3. A simplified example of differences between system boundaries in attributional (black) and consequential LCAs to deal with a surplus of electricity generated using lignin burned internally. In red, system boundaries and unit processes that would be included within the boundaries in a consequential analysis.

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 [103]. So, the demand for paper (pulp) is the market driver of the Kraft mills and not the market applications of lignin [103]. 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 [103,114]. 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 [106]. 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 [106].

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 [106,147,148]. 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 [136].
- A full cradle-to-grave study for adhesives used in fiberboard production [132]. 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 [149]. Hence, the different compositions were not taken into account.

2.3.2.2 Functional unit

In LCA, the functional unit is the “quantified performance of a product system for use as a reference unit” [38] 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 [136]. 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 1m^2 of a laminate board [131]. However, most studies, e.g. [134], 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 tonne of pulp [100] or 1 g of vanillin [137]. 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 [150] or 1 MJ of jet fuel [148]. 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 liter of ethanol [151] and 1 m^3 of finished medium density fiberboard [104];
- *Simplified area FU based on the output*. This FU (1 m^2) was used in two studies [131,132] assessing adhesives for wood fiberboards;
- *Input based FU*. This type of FU was used in the study authored by González-García et al. [152] 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 [153,154];
- *Multiple FUs (one per each main co-product)*. This type of FU was used by several studies [56,133,155];
- *Distance*. The FU of 1 km, which is typical for well-to-wheel assessments and was used by one study [147];
- *Ultimate final application*. This functional unit was used by one study [141] 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 2.3.2.3). 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 2.3.2.5). 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 tonne of product for cellulose, lignin and vanillin and 1 m^3 for ethanol and showed the results per each FU.

2.3.2.3 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 [50,156]. 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. [146]. 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 by Shinde et al. [154], 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 Figure 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 [57,145,146]. 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 [51,57]. 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 [142]. The interpretation of the results was that bio-based ethylene was much better than fossil ethylene from a climate change perspective (about half impact). 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% [142], leading to a climate change impact of 1.15 kg of kg CO_2 eq per kg of ethylene. However, the palm oil extraction unit alone had a contribution of 7.17 kg CO_2 eq/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” [38], 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” [103]. In general, the changes modeled using this type of allocation can be either marginal, incremental or average (listed in order of magnitude) [157]. A physical causality allocation based on average changes was used by Bernier et al. (2013) who 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 mill [100]. 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 2.3.3.1). Concerning economic allocation, the price assumed for lignin should reflect the specific lignin under investigation. In fact, the price of lignin is highly variable dependent on the source and quality of the lignin (see Figure 1). However, specific quality-level lignin "has relatively stable prices through the years and seasons" [117]. 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 2.3.3.

In some studies, the type of allocation used was not clear (e.g. in [134]) 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 [100]. In their sensitivity analysis, mass and economic allocations were used in combination with substitution (keeping the credit for the surplus of electricity).

A summary of the adopted multifunctionality practices in the selected LCAs is shown in Figure 4. Although mass allocation was the most adopted method to deal with multifunctionality, Figure 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 [146]. 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 co-products 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 [145]. Applying substitution without this check, Hermansson et al., (2020) obtained negative climate change impacts for Kraft lignin. 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".

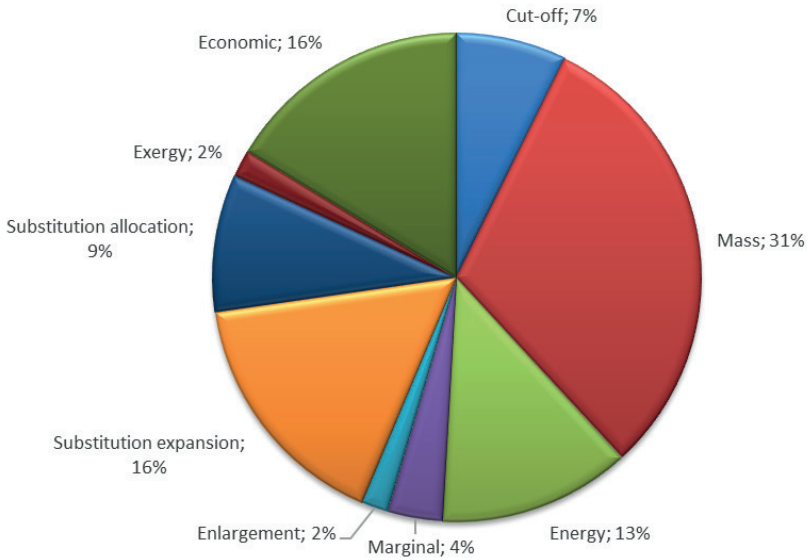

Figure 4. Summary of the adopted multifunctionality practices in the selected 42 LCAs.

2.3.2.4 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 Figure 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 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 [158] 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 [159] and AWARE method for water depletion [160]. 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.

2.3.2.5 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 [55] and [136].

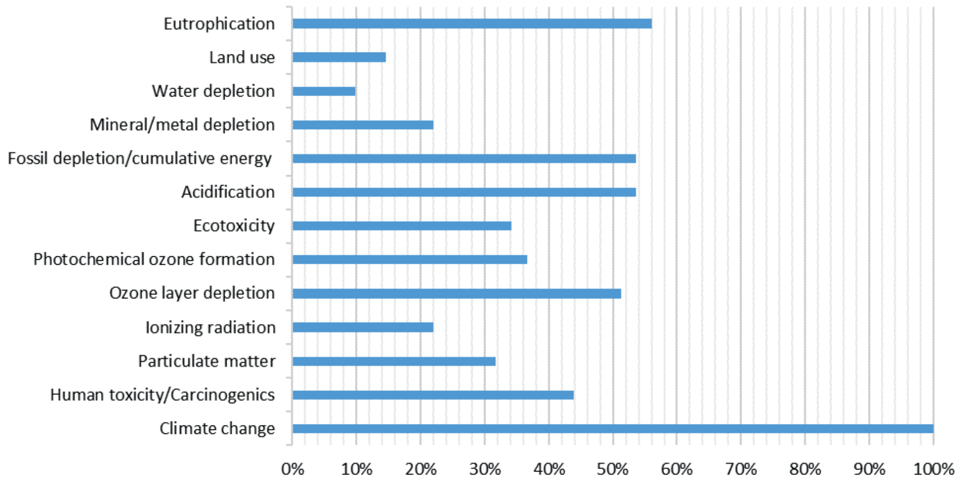

Figure 5. Impact categories covered in the evaluated sample of LCAs.

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 [162,163].

2.3.2.6 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 deriving from biomass 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) [103]. 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 [85,145,165], European directives for renewable energies and alternative fuels [9,37] and the US Environmental protection agency [166]. Alternatively, the release of biogenic emissions and carbon storage can be discounted in time as proposed by UK PAS 2050 [167].

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" [85,165]. 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 [131] and suggested by recent EU LCA guidelines [165]). 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" [85,165].

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.

2.3.3 ENVIRONMENTAL IMPACT

2.3.3.1 Climate change impacts of lignin

As mentioned in section 2.2.2, 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, Figure 6 shows the climate change impact of Kraft lignin as reported in the LCA studies assessing lignin from Kraft mills [55,100,103] and two LCAs on lignin-based products for which it was possible to retrieve/back-calculate the values obtained from their inventory data.

From Figure 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 Figure 6 is not self-explanatory and needs to be handled carefully. From Figure 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 Figure 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 modelled by Bernier et al. is much more impacting on climate change than the one modelled 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.

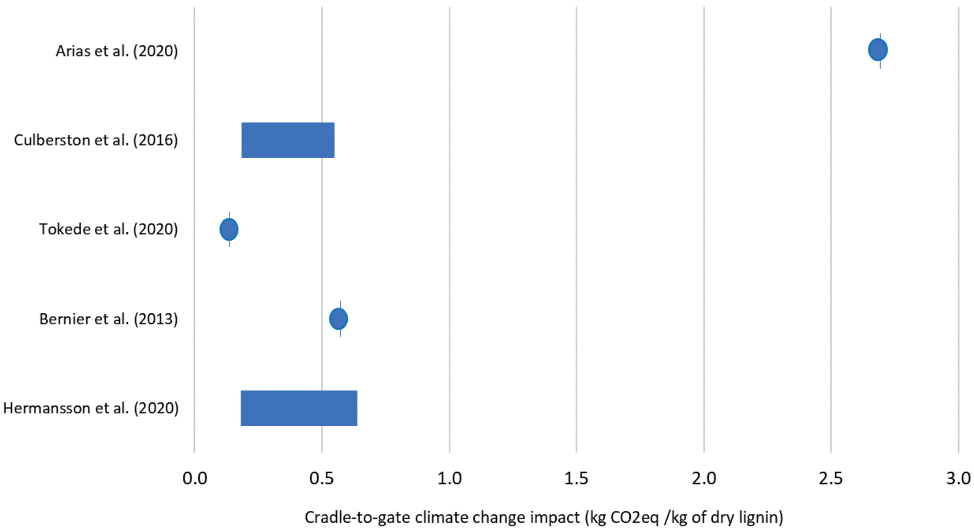

Figure 6. Cradle-to-gate climate change impact of Kraft lignin as reported in the reviewed LCAs. Ranges represent mainly the testing of different allocation methods in the baseline calculations conducted in the LCA. With respect to the allocation methods tested by Hermansson et al. (2020), once the assumptions regarding the main product and current steadiness of lignin price were revised, as mentioned in section 2.3.2.3, 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.

Regarding other types of lignin, in the LCA conducted by [115], 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 [129], 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 [135]. For instance, one of the insights of Koch et al. (2020) was to 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” [135].

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 [155], 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 2.3.2.2 that used multiple functional units did not have lignin as a sold co-product.

2.3.3.2 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 [134].

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 [55]. 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 [168]. Landa and Gosselink (2019) published the application of lignin in bio-asphalt showing a lower production temperature of 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 [115].

Given the issues mentioned above about the second approach, most of the LCAs on lignin applications applied the first approach which is more reliable. Figure 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 Figure 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%

[2] This value was calculated from 20 kgCO2eq per m3 of finished fiberboard reported in figure 4 of [104] and 154.2 kg/m3 of ammonium lignosulfonate needed for the production of 1 m3 of finished fiberboard [104]

and 90% [106] 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, [106] 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 [38] according to the waste management framework [170] adopted by the European Union in the Renewable Energy Directive [9].

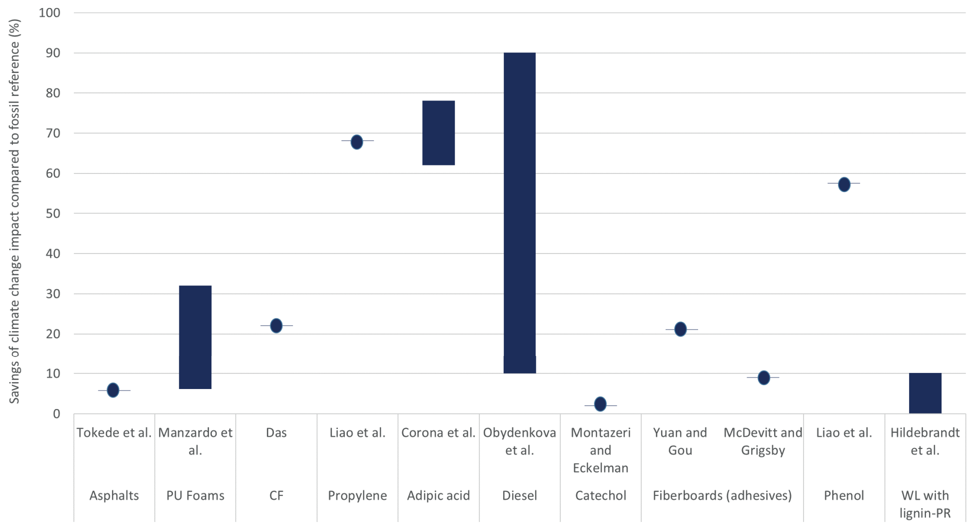

Figure 7. Savings of climate change impact compared to fossil reference reported in the selected LCAs. PU=Polyurethanes, CF= carbon fibers, WL=wood laminate, PR=phenolic resin. The range of values from Manzardo et al. and Hildebrandt et al. refers to multiple formulations (e.g. varying shares of lignin 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.

Adipic acid also seems a promising application from a GHG emissions perspective, allowing savings between 62% and 78% compared to petrochemical adipic acid [138]. 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 [136].

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% [140], which is very minor compared to the uncertainty involved. Bio-based asphalts [134] 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 Figure 6. Changing methodological assumptions or Kraft mill might lead from a low GHG saving of emissions to higher impact than conventional asphalts.

2.3.3.3 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) [115]. These adhesives were compared with two alternative bio-based adhesives (from soy and 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 [104], 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 [132]).

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 at 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) [131].

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 Figure 7), potentially offer 7% and 59% environmental impact reductions respectively for ecotoxic effects and depletion of fossil fuels [140]. However, in the other seven environmental impact categories, the fossil route was preferable [140]. 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 [140]. Krzyżaniak et al. (2019) assessed the final application (cultivation using different fertilizers) and assessed the same impact categories of [140] 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) [8,140]. 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.

2.3.3.4 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) [103]. 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) [100]. 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 [119]. 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 [171] 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 [119].

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 [106]. 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 [106] set by the EU renewable energy directive [9,39] and U.S. renewable fuel standard. To fulfill this target, in the example of the biorefinery modeled by [106], the use of corn stover also for internal energy purposes (and not only as feedstock for fuel production) was proposed.

2.3.3.5 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 [172]. 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 [172]. 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 [172]. 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) [113]. These same two assumptions were made also by [164] 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 [173].

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 [148]. 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%) [148].

2.4 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-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 3.

Table 3. Lesson learned and recommendations. Continues in the following two pages.

Lesson learned	**Recommendations**
Only a few studies considered the use phase and end of life of the product (see section 2.3.2.1).	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 specify the type of modeling approach followed i.e. attributional or consequential (see section 2.3.2.1).	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 2.3.2.2). 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 2.3.2.4). 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 2.3.2.5). 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.

Lesson learned	Recommendations
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 harmonised (see section 2.3.2.3).	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 2.3.2.6). 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.

Lesson learned	Recommendations
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

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.

CHAPTER 3

Reviewing ISO compliant multifunctionality practices in environmental Life Cycle modeling

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ABSTRACT

The standard ISO 14044:2006 defines the hierarchical steps to follow when solving multifunctionality issues in Life Cycle Assessment (LCA). However, the practical implementation of such a hierarchy has been debated for twenty-five years, leading to different implementation practices from LCA practitioners.

The first part of this study discusses the main steps where the ISO hierarchy has been implemented differently and explores current multifunctionality practices in peer-reviewed studies. A text-mining process was applied to quantitatively assess such practices in the 532 multifunctional case-studies found in the literature. In the second part of the study, citation network analysis (CNA) was used to identify the major publications that influenced the development of the multifunctionality-debate in LCA, i.e. the key-route main path. The identified publications were then reviewed to detect the origins of the different practices and their underlying theories.

Based on these insights, this study provides some “food for thought” on current practices to move towards consistent methodology. We believe that such an advancement is urgently needed for better positioning LCA as a tool for sustainability decision making. In particular, consistent allocation practices could be especially beneficial in bioeconomy sectors, where production processes are usually multifunctional, and where current allocation practices are not harmonized yet.

3.1 INTRODUCTION

Life Cycle Assessment (LCA) is supposed to be a standardized methodology to measure the life cycle impacts of products or services. LCA is currently ruled by ISO 14040:2006 and ISO 14044:2006 [38,127]; these standards have been the basis of the LCA methodology for the last two decades. Nevertheless, in the scientific community, some experts wonder if the detail presented in these standards is enough to guide LCA practitioners in practice [57,71,174].

One of the most debated problems in LCA is the so-called "multifunctionality" issue (or commonly, "allocation") [50,83,175]. Multifunctionality issues need to be dealt with when different product systems share a process, e.g. manufacturing processes delivering more than the studied product, or end-of-life activities providing both waste management service and a recovered or recycled product. In these cases, apportioning environmental burdens among the co-products, or rather co-functions, becomes necessary. According to ISO 14044:2006, multifunctionality should be solved by using the following three-levels hierarchy [38]:

1. Avoiding allocation, by subdivision (dividing the unit process into two or more sub-processes) or system expansion ("expanding the product system to include the additional functions related to the co-products");
2. Allocation following underlying physical relationships (i.e. an allocation that quantitatively reflects how the inputs and outputs are changed by changes in the amount of each product of the system);
3. Allocation (partitioning) based on other relationships (e.g. economic value);

The same hierarchy applies also to "open-loop" recycling i.e. when a material is recycled as a different product because it is no longer suitable to replace the original product directly. Only in such open-loop recycling, ISO 14044:2006 provides further guidance on the third level of the hierarchy, where physical properties (e.g. mass) are preferred to economic value, which in turn is preferred to the number of subsequent uses of the recycled material [38].

The existence of the ISO's multifunctionality hierarchy should avoid the use of inadequate approaches, e.g. determined by the interests of the stakeholders or the ones of the study's commissioner [57]. Nevertheless, the apparent lack of sufficient guidance has fed different implementation practices [68]. Consequently, although most LCAs claim compliance with the two ISO standards, practitioners have applied different allocation procedures in LCAs assessing the same or similar products [94]. Since the choice of the allocation method typically affects the outcome of the LCA significantly [50,94,96,128,176], this problem has led to different conclusions and therefore low reliability and robustness of the LCA results [177]. Moreover, due to the lack of a shared view in the LCA community, some authors decide not to follow the ISO hierarchy (see [178]), while other authors select the allocation method based on their subjective decision (see e.g. [179] and [180]). Other researchers choose allocation methods that are "commonly" applied in similar case studies in the literature (see e.g. [181]), others calculate also an average allocation parameter considering common parameters (e.g. [182]) or others use "conservative" allocation methods that provide the highest impacts (e.g. see [183]).

This article presents a literature review on the main practices and debates on using ISO 14044:2006 recommendations to solve multifunctionality problems. A critical literature review on multifunctionality methodology development was combined with quantitative analysis of current multifunctionality practices, and a bibliometric review based on citation network analysis (CNA). The quantitative analysis was performed by a text-mining process in 532 multifunctional case-studies found in the literature.

The CNA was used to identify the main knowledge flow on multifunctionality in LCA, also known as "the main path". Tools and software based on the "main path" method are used for many applications: tracking the evolutionary trajectory of a science field or the development of a specific technology, or the evolving changes of legal opinions of courts [184,185]. The "main path" was investigated to detect the historical origins of the different practices currently present in the literature and their underlying theories. The use of such a tool overcomes some limitations of the traditional systematic reviews conducted so far on this topic, which were based on "human" selection of the articles (e.g. through criteria such as the number of citations).

In the literature, the definitions used to characterize the multifunctionality issue are not harmonized. For this reason, we provide an appendix reporting the definitions used in this review to distinguish the different types of products, multifunctional processes, modeling approaches and system expansion approaches.

3.2 METHODOLOGY

Figure 8 summarizes the three main steps followed in this literature review. First, the literature search was performed. Second, a critical review was conducted to identify the main issues and bottlenecks in the LCA literature when implementing the ISO allocation procedures. The critical review was combined with a text mining process to quantitatively assess the current practices in the LCA literature (focusing on all the LCA case studies selected by the query). Third, a bibliometric analysis was performed based on citation network analysis (CNA).

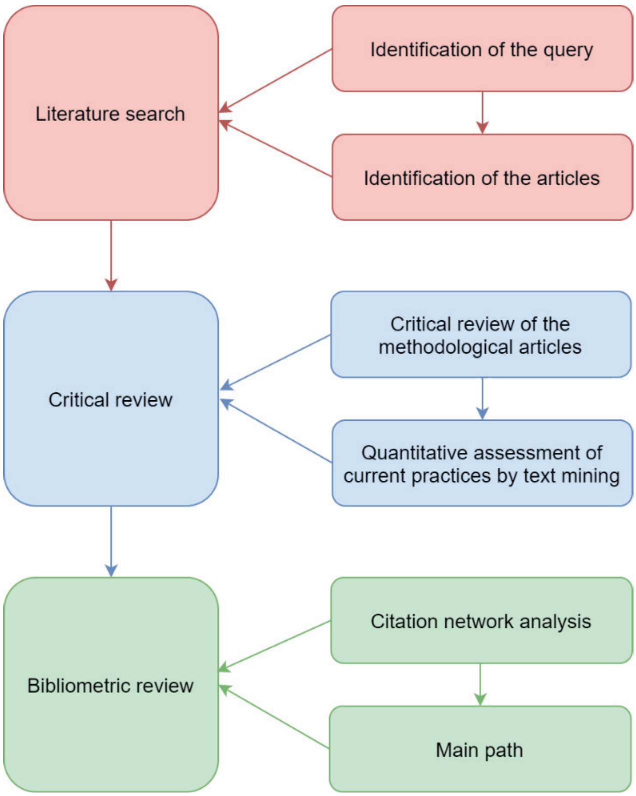

Figure 8. The three steps followed in the literature review

The literature search was based on data collected from the Scopus database in February 2019. The searched publication fields were: title, abstract, and keywords. The search string was characterized by the terms: "Life Cycle Assessment", "LCA", "multifunctionality", "allocation" and "multi-output". Since allocation approaches are also used in other fields (e.g. in business management), the query was first limited to environmental assessment or engineering-related fields. Because of this, the documents were reduced from 1310 documents to 1152. This allowed us to exclude 145 documents belonging to business management, 6 related to veterinary science and 7 others. Our analysis was further refined by considering articles only from the category of Scopus "journals". By applying this last adjustment, the articles resulting from the search became 930. Since only the research articles were analyzed, some relevant books or conference proceedings may have been excluded from the analysis. Nevertheless, often books resume the contributions previously published as articles and some excluded documents might have been considered by some of the reviews reviewed. Figure 9 shows the number of publications per year, highlighting the growing interest in the topic.

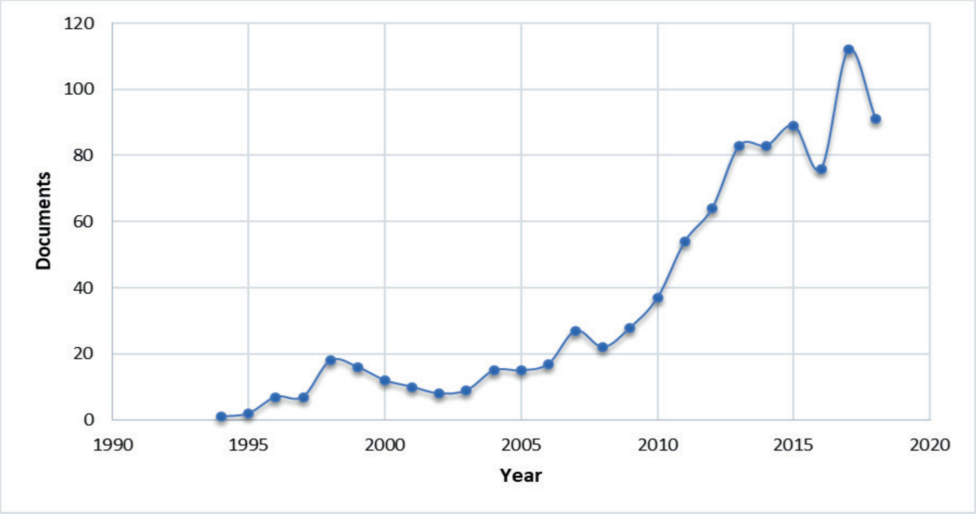

Figure 9. The time distribution of the articles on multifunctionality in LCA published in scientific journals per year retrieved from Scopus.

The corpus of documents on which the analyses were performed included the 930 articles retrieved from Scopus and the main LCA guides and standards, i.e. ISO technical reports and standards (also withdrawn ones like ISO 14041:1998) [186–189], the International reference Life Cycle Data system (ILCD) handbook [145], the Product Environmental Footprint (PEF) guide [85] and the Product Environmental Footprint Category Rules (PEFCR) guidance [165]. Out of the 930 documents, 307 studies were identified through their title and abstract as "methodological articles" (of which 117 were review articles focusing on a specific sector where LCA is applied). These methodological articles focused either on the general methodological debate about multifunctionality procedures, or discussed a specific method, or introduced a new model to solve multifunctionality. The most relevant articles in this group were critically reviewed to understand the main issues when solving multifunctionality in LCA while claiming compliance with ISO. This critical review focused mainly on the articles cited more than 20 times ("most cited ones") and the articles published after 2015 ("recent ones").

The critical review was combined with a text mining process whose aim was to quantify the current practices when solving multifunctionality issues. The text mining process was manually performed on the remaining 532 case studies. These 532 case studies resulted from a further refinement which excluded 91 articles that either did not apply full LCA or were not environmental LCA studies. Concerning the multifunctional case studies retrieved from the literature, we observed that specific parts of the bioeconomy namely agriculture (63 case studies), bioenergy (185), bio-based materials (52) and anaerobic digestion (21), are the ones most affected by the issue of multifunctionality together with related sectors namely aquaculture (14), dairy and meat products (79), fossil counterparts (34) and waste management (50). These sectors together represent 94% of the 532 case studies identified by the query.

A text mining software can detect relevant terms or keywords in the corpus of literature with less time and cost than a person [190]. However, when the keywords represent technical concepts, dedicated software typically achieves low to medium efficiencies (e.g. 25-65%) [190]. For instance, software could not understand when the concept "system expansion" was used as an alternative expression for substitution or for system enlargement. To increase the efficiency of the text mining method, the quantitative estimation was performed directly by the analyst. When the terms representing the concepts of interest (e.g. "allocation") were encountered, the context of their use was assessed by reading the surrounding text.

In the third step, i.e. the bibliometric review, the 930 articles were investigated by CNA. The CNA was performed using Pajek software [191]. Documents are considered "nodes" and the citations are the "links" between these documents. The type of nodes is defined therefore based on the type of document. The "sources" are the documents that are cited but cite no other documents and therefore represent the origins of the knowledge. The "sinks" are the documents that cite other documents but are not cited and therefore could represent the "current stage" of the knowledge stream. Intermediate documents cite other previous documents and are also cited by more recent documents [184]. Our CNA aimed at identifying the main path of research. This path represents the main knowledge flow in a specific topic, i.e. the major contributions that have influenced the development of the research, which does not mean directly the most cited ones overall [192,193]. The main path was obtained by using an algorithm that computes what citations between articles have been more significant. In particular, such a significance was calculated through the key-route method [194]. This method identifies the main chain of articles by considering the highest transversal count [193,194]. The transversal counts measure the significance of a citation link i.e. by counting the times a citation link is traversed [194]. The transversal count adopted was the search path count (SPC). The SPC assigns as value to each link the number of paths traversing the link among all possible paths connecting all the sources to all the sinks [184,185].

3.3 THE CRITICAL REVIEW COMBINED WITH TEXT MINING

When critically reviewing the methodological articles on multifunctionality, it emerged that these articles present two main "debates" regarding ISO-compliance practices. These two debates concern the application of system expansion (explained in section 3.3.1 and related sub-sections) and the identification of relevant partitioning criteria (see section 3.3.2). In particular, Pelletier et al. (2015) identified three "schools" distinguished by the way they interpret the ISO hierarchy with respect to these two aspects: (1) the consequential thinking school interprets system expansion as substitution, (2) the natural-science attributional school applies system expansion as enlargement and prioritizes allocation based on a physical parameter, and (3) the socio-economic attributional school applies system expansion as enlargement but prefers economic allocation. According to Pelletier et al. [68], these three schools are "internally consistent" but "mutually exclusive".

3.3.1 Debate on the interpretation of ISO's system expansion

The system expansion debate focuses on how and when should the substitution method be applied. ISO 14044:2006 recommends system expansion as a way to avoid allocation, but no further specification is provided regarding the differences between enlargement and substitution (see appendix for detailed definitions), and about its implementation in attributional or consequential LCAs. Substitution is often used as a system expansion approach in attributional LCAs (ALCAs), which is not perceived as correct by many LCA experts [51,68,71–73,195]. According to these practitioners, ALCA modeling should not rely on perturbation logic or counterfactual notions, such as substitution or avoidance of other products/ processes (as also highlighted e.g. by Majeau-Bettez et al. (2018). It is argued that the sum of the impacts accounted by attributional LCAs should add up to the worldwide impacts, and this would not be valid anymore if substitution were applied [71,74]. For this reason, Chen et al. (2010) concluded that the "allocation methods, even if perfectible, are still preferable to the system expansion method" (used as synonymous of substitution), because "system expansion does not ensure a global coherency between various LCA studies" [196]. On this bases, the use of substitution as a system expansion method in ALCA is not supported by any of the schools of interpretation identified by Pelletier et al. [68]. Similarly, Bailis and Kavlak, after applying substitution for the by-products of a biofuel, concluded that "the large disparity between system expansion and other methods raises questions about the validity of system expansion" [197]. Concerning system expansion by enlargement, this cannot be applied when the goal of the study requires to obtain the impacts of just one of the co-products or by-products. In these cases, allocation cannot be avoided. For example, "In a milk production system that also produces beef, system expansion without substituting would lead to a system with a function of delivering both milk and beef" [198].

Other authors argue that ISO 14044:2006 does not acknowledge substitution as a system expansion approach. The reason is that ISO refers only to the addition of functions (i.e. enlargement) and not to the substitution of functions [57,71,199–201]. On these bases, several authors argue that a distinction ALCA/CLCA should be present in future ISO 14044 [68,202] since, for them, this distinction is crucial to select the appropriate system expansion method (enlargement or substitution) (as also pointed out by [61,144]). By contrast, other authors argue the opposite i.e. that substitution is generally recognized as a valid method for avoiding allocation within attributional LCA [203,204]. For many practitioners, substitution is considered as synonymous with system expansion [205,206]. Under this argument and considering the ISO hierarchy, substitution should be preferred to any allocation method [76–

79,207]. Pelletier et al. [68] suggested that the equivalence substitution-system expansion might have been originated in a study of 1994 authored by Tillman et al. [81]. The reason was that Tillman et al. [81] is a frequent citation when justifying the equivalence of substitution with system expansion. However, their study was published previous to the publication of the ISO standards.

3.3.1.1 Current practices in specifying the modeling approach

Although the choice of modeling approach (ALCA or CLCA), which depends on the goal, clearly determines the outcome of an LCA study, our text mining process found that the keywords “attributional” and “consequential” were missing in 75% of the LCAs involving multifunctional systems (see Figure 10 for a detailed breakdown, per product sector). This percentage refers only to the portion of articles published after 2004 when the term consequential LCA was clearly established (see section 3.4.3). There are several possible reasons for this low specification rate of the modeling approach: (1) practitioners could still not be aware of the relevance to differentiate between consequential and attributional approaches (since consequential approaches were still not common), (2) practitioners may not specify the modeling approach because it is a direct consequence of the goal description, (3) they may not agree with a strict distinction between ALCA and CLCA, (4) they may be strictly following current ISO standards (that do not distinguish between the two approaches) or (5) they may have followed the recommendations of a policy directive or national/international guide that does not make such a distinction. Actually, some ALCA studies are emerging that combine attributional modeling with consequential thinking [94]. These approaches aim mainly at accounting for some specific counterfactual effects or credits, and at the same time, limit complexity and uncertainties [35].

The text mining process found that 31% of the self-declared ALCA studies (using the keyword “attributional”) used substitution as a system expansion approach to avoid allocation. However, this percentage varies depending on the sector under consideration, ranging from 19% to 45% (see Figure 11). The highest rate of substitution approaches in ALCAs was found in studies related to bio-based materials (45%). On the other hand, the LCAs investigating fossil products that self-declared to be attributional studies are few since substitution is rarely an option for fossil products since they are usually the “substituted ones”.

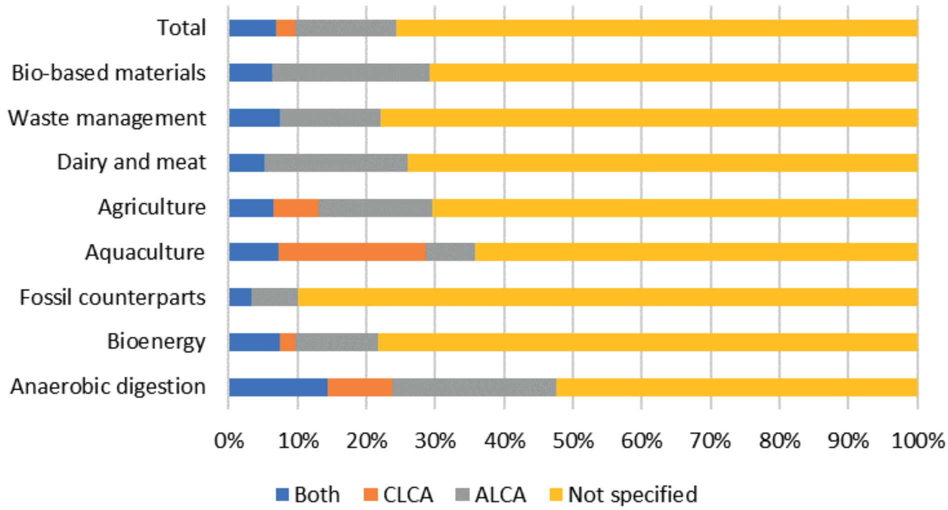

Figure 10. Percentage of articles which applied on the same case study both CLCA and ALCA approaches (Both), self-declared attributional studies (ALCA), self-declared consequential studies (CLCA) and studies which did not declare the approach followed (Not specified). Only case studies published after 2004 were considered (504).

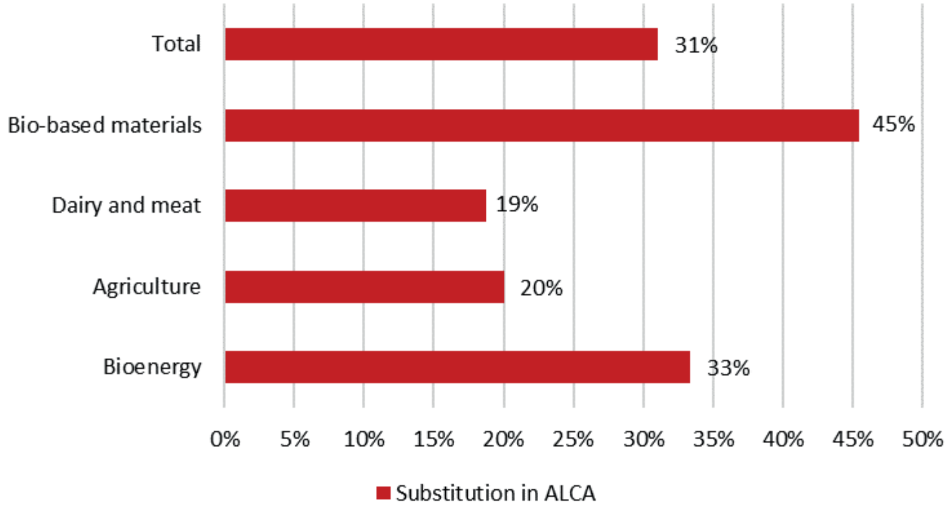

Figure 11. Percentages of self-declared attributional studies (ALCAs) which applied substitution as a system expansion approach. Only the sectors with a significant amount (more than 10) of self-declared attributional studies are included in this graph.

3.3.1.2 The application of substitution as system expansion method

Beyond the use of substitution in ALCA, two other critical aspects of substitution have been discussed in the LCA methodological articles. First, the high uncertainties introduced by the use of the substitution approach since it can lead to different results depending on the choice of substituted and/or substituting by-product [57,208,209]. A sensitivity analysis should be therefore recommended. Second, when substitution is suitable, the substitution of co-products should be avoided by checking the physical/economical significance of the products delivered by the multifunctional process [57,145]. However, some authors (for example [57]) argue that the importance of the co-products' physical significance is not emphasized enough in ISO 14044:2006 and the ILCD handbook. When physical significance is not checked and a by-product is credited for the replacement of co-products, the practitioner could obtain significantly distorted results [57,210]. A common practice to account for physical significance is to select the primary functions based on the main source of revenues [51,211]. In cases where the primary co-function(s) cannot be directly identified, the ILCD handbook proposes that they should be assumed to be those that jointly contribute to more than 50 % of the combined market value of all co-functions of the analyzed multifunctional process [145].

Clear rules for differentiating by-products from the co-products are important, because, in substitution, all the credits from substituting by-products are attributed to the main co-product. If another LCA on the same process is made in which a by-product is considered to be the main product, the impacts of the process "get counted twice", so that the impacts for different products no longer add up to the total for the process (this would be a problem in an attributional model- for further details see the next section).

3.3.1.3 Using substitution as the allocation method

Another point of debate is the use of substitution for allocation (and not as system expansion method). This type of allocation has been mentioned in the literature with different names such as substitution-based allocation [68] and "proxy-based disaggregation" by substitution [51] and various versions of this method have been proposed (e.g. see Hermansson et al. who applied two different versions of this method to assess Kraft lignin [55]). By many practitioners, this option is perceived as the attributional way of using substitution. PEFCR guidance and PEF guide [85,165] propose that, when a by-product of a multifunctional system directly substitutes another product, such substitution might be considered as an allocation reflecting physical relationships. When this is the case, such substitution has to be based on a direct and empirically demonstrable relationship [85,212]. Pelletier et al. [212] stated that this is different from substitution based on marginal market models applied in consequential LCAs [212]. An example of such a substitution is when "manure nitrogen is applied to agricultural land, directly substituting an equivalent amount of the specific fertilizer nitrogen that the farmer would otherwise have applied" [85,212]. Hence, it is assumed that the impact caused in the system by the production of the substituted by-product corresponds to the impact of the production of the replaced product (as shown in [213]). With substitution, the impact of a by-product should equal that of the product it substitutes, and so is independent on the actual process that produces it. Moreover, the application of this substitution-based allocation can lead to a negative impact that in ALCA would mean that the model has been built inconsistently [51]. As an example, this happens when the wrong product is chosen as the main product of the multifunctional system [213] or the substituted product is not a minor product even if representing less than 50% of market value [163]. Even if the substituted co-products are chosen carefully (i.e. they represent small percentages in physical and economical terms), this method sometimes fails in ALCAs assessing multiple impact categories resulting in negative impacts for some of them [128,163]. Moreover, PEFCR guidance and PEF guide [85,165] also allow the possibility of using indirect substitution as a form of allocation based on "other relationship". "Indirect substitution may be modeled as a form of allocation based on some other relationship when a co-product is assumed to displace a marginal or average market-equivalent product via market-mediated processes" [85].

3.3.2 Selection of the ISO allocation criterion

The main discussion on the allocation criterion concerns the nature of the so-called ISO "physical relationships" and "other relationships" [175,198].

The authors in line with the socio-economic school argue that allocation can be based on physical relationships only when the ratio of the output products can be varied since this allows the establishment of physical causality between functional units by mathematical modeling [97,157,214–217]. For example, Bernier et al. [103] assessed the impact of Kraft lignin and applied the physical causality principle to allocate the impact between pulp and lignin “by varying the quantity of lignin precipitated and then observing direct variations in the environmental loads”. They also specified that this type of allocation was selected based on ISO standards, which recommends this type of allocation over allocation based on mass, energy or economic values. They, therefore, interpret “physical relationships” as “physical causality relationships” and interpret “other relationships (e.g. economic value)” as “other causal relationships”. Accordingly, they consider the allocation by other relationships as the only possible approach when it is not possible to change the ratio of production of the functional outputs of the system [215,217]. The practitioners following the view of this school often argue that, at this level, economic allocation is the recommended option, and only when it is not possible to use economic allocation, the allocation can be based on a physical parameter that should be selected based on the best proxy for economic revenues (e.g. see [175]). For example, this happens when there is a lack of market prices for one specific product [218]. However, the approximations of these causal relationships have always been a debated scientific issue [219]. For example, these relationships can be based on the common function of all co-products (as done by [220]).

The text mining process revealed that only 28% of the LCA case studies selected an allocation method based on ISO relationships that could be interpreted as “causal relationships”. The percentage of studies following this interpretation varies significantly depending on the sectors considered (see Figure 12). In particular, it is very low in the studies focusing on anaerobic digestion, bioenergy and bio-based materials (5-16%). On the other hand, this interpretation is largely present in the fossil fuels sector where the allocation of the emissions to the single products is often based on linear programming models calculating the marginal emissions by varying the amount of the functional units [221,222]. Another example where this interpretation is largely present is in the dairy sector. The main reason is that many practitioners assessing dairy products selected their allocation choice based on the recommendations of the International Dairy Federation [223], which adopts this interpretation.

Conversely, the practitioners belonging to the natural-science school often refer to an allocation by physical parameter as ISO-second level allocation by interpreting “physical relationships” as allocation based on a physical parameter e.g. mass or energy value [83,179,224–226]. On this basis, they prefer allocation based on a physical parameter (e.g. mass) over economic allocation because “ISO 14044

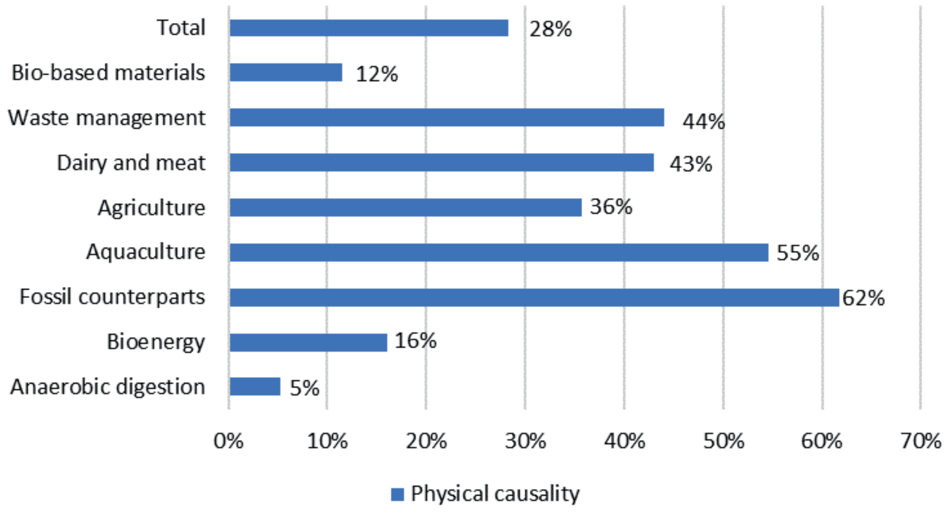

Figure 12. The percentage of studies in each research area that used causality as the principle for the allocation choices per (sub-)cluster and overall. The number of studies per sector: anaerobic digestion (21), bioenergy (185), fossil counterparts (34), agriculture (63), aquaculture (14), dairy and meat (79), waste management (50), bio-based materials (52).

standard mentions economic allocation when no other possibility is available" [227]. Economic allocation may be selected by the practitioners following this view if allocation based on physical parameter "result in the attribution of a large proportion of burdens to low-value co-products" [68]. The same school often argue that an allocation based on a physical parameter is preferred over economic allocation, since it is not affected by price fluctuations [225,228]. As a response, authors in line with the socio-economic school argue that the price fluctuation is not the important parameter for the allocation method, but the ratio of prices among all products, which is much less variable because it mainly depends on the fluctuating price of the inputs common to the process [229].

The preference expressed by the natural-science school is adopted by PEFCR guidance and PEF guide, which prefer allocation based on physical keys (e.g. mass or energy) to economic ones [85,165]. In the PEF guide, ISO "physical relationships" might have been interpreted as allocation based on physical parameters (this emerges from our understanding of annex X of PEF guide), leading to the preference for physical allocation keys. On the contrary, the ILCD handbook adopts the interpretation of "ISO physical relationships" from the socio-economic school

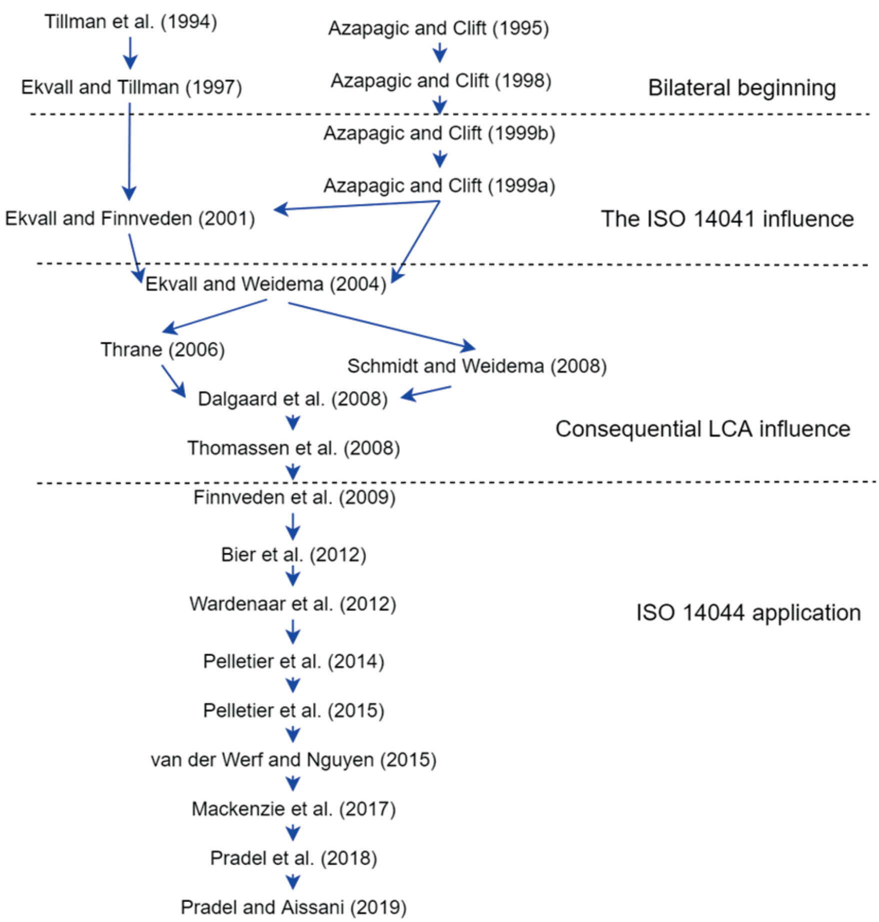

Figure 13. The key-route main path of research on LCA multifunctionality (output from Pajek calculations), obtained from the citation network analysis.

and states that only when it is not possible to find clear physical *causal* relationships between the co-functions, allocation based on economic relationships can be used [145]. However, differently from what is usually preferred by the socio-economic school, the ILCD handbook does not give preference to economic allocation over non-causal physical properties such as energy content [145]. The ILCD handbook also adds a footnote to remark that energy allocation is not an allocation based on ISO causal physical relationship but a simplified allocation based on a physical property that is not causal [27].

To make an example of the implication of adopting one interpretation or the other, we can consider a biorefinery example that produces fuels (e.g. ethanol) and chemicals for materials (e.g. lactic acid) [56]. The natural-science school would prefer energy or mass allocation (considered by them as ISO second level) over economic allocation (considered ISO third level). Conversely, the socio-economic school would prefer economic allocation arguing that mass and energy allocations (all considered ISO third level) are meaningless for such a biorefinery because of not representing any causality mechanism. Also, they would argue that it is not appropriate to use energy allocation when not all the co-products are used for their energy content or to use mass allocation when there are energy products among the co-products.

3.4 THE BIBLIOMETRIC REVIEW BASED ON MAIN PATH ANALYSIS

The main path of research identified using CNA is shown in Figure 13 and includes 21 articles. The evolution of the multifunctionality discussion in the scientific community can be divided into four periods, which have been defined as 1) Bilateral beginning, 2) The ISO 14041 influence, 3) Consequential LCA influence, and 4) ISO 14044 application.

3.4.1 Bilateral beginning (1994-1998)

The discussions on the LCA multifunctionality issue were initially developed following two parallel routes (see Figure 13). On the first route, Tillman, Ekvall and their co-authors developed different types of allocation methods for multi-output systems and open-loop recycling [80,81]. It is crucial to notice that at that time, the ISO 14041 was not yet released [188]. Tillman et al. [81] focused their article on the choice of system boundaries based on the purpose of an LCA. They defined three LCA purposes: 1) process tree (PT), today known as ALCA and applied to processes where there are one main product and some by-products, 2) technological whole system (TWS), similar to what today is known as ALCA and applied to processes delivering several co-products, and 3) socio-economic whole system (SWS), similar to current CLCA [81]. In this article, the word *expansion* was used once with respect to SWS, indicating that such a system accounts for economic and social factors and therefore “may lead to further expansion of the system” [81]. In 1994 and 1996, two conferences were held with sessions on allocation and life cycle inventory. Clift, who was also co-author of the publications in the second parallel route, published the reports of such sessions [230,231]. These reports concluded that allocation must, when possible, be based on causal relationships. Ekvall and Tillman discussed this conclusion, arguing that causal relationships could be either cause-oriented or effect-oriented [80]. An example of the first one is the manufacture of a product that occurs because the company expects customers to be willing to pay for it (cause). An example of an effect-oriented relationship is a system delivering a recycled product, which reduces the amount of virgin product in another system (effect). This second type of relationship resembles the current CLCA thinking. To represent effect-oriented relationships, they argue that the effects of the investigated product on other life cycles can be included in the LCA through the expansion of system boundaries. As the expansion of the system boundaries, they cited the approach developed by Tillman et al. (1994), which today is known as "substitution". Moreover, they argued that when LCA is used as a tool for decision support, the allocation procedure should generally be effect-oriented rather than cause-oriented. Therefore, it is possible to identify the probable origin of the consequential school in the study of 1997 of Ekvall and Tillman [80] and the study of 1994 of Tillmann et al. [81].

The four articles of the second route were authored by Azapagic and Clift. In the first article (1995), they proposed linear programming (LP) modeling to solve the multifunctionality issue and to calculate the optimized environmental impact of plastic resins production, such as polypropylene and polystyrene [232]. The inputs and outputs of the system are then allocated to each of the co- and by-products through marginal changes in its production [232]. The marginal allocation coefficients correspond to the variation of the environmental burdens associated with a marginal variation of one of the co-functions [232]. The second article (1998) focused as well on LP as a tool for solving the problem of allocation and was applied to systems producing borate products. They highlighted that 1) "the main characteristic of this kind of modeling is that it is based on physical and technical relationships between the inputs and outputs [...] describing the underlying physical causation in the system" [157] and 2) that the allocation by causal relationships provided by the model is obtained ''by exploring how the burdens change when the quantity of one function is changed with the quantities of all the other functions kept constant'' [157]. These changes can be marginal, incremental, or average ones; however, LP can only be applied when system behavior can be linearized, which does not usually happen in average changes (i.e. substantial changes as for example the elimination of a functional output completely) [157].

In 1996, the first draft [233] of the ISO hierarchy for solving multifunctionality was released, as reported by Ekvall and Tillman [80]. This hierarchy was very similar to the one still present in the current ISO 14044:2006. System expansion was indicated in the first level [233]: “by expanding the system boundaries so that inputs, outputs and recycles remain within the system” (retrieved from [80]). From the literal statement, it appears clear that it was intended as an enlargement of the system boundaries to include all the co-functions within the boundaries (see Figure 3A.1). Such an approach is different from the system expansion method (substitution) indicated by Tillman’s SWS, where functions are avoided instead of added.

On the second level, it was stated [233]: “where allocation cannot be avoided, the allocation should be based on the way in which the inputs and outputs are changed by quantitative changes in the products or functions delivered by the system” (retrieved from [80]). There was no use of the term “physical relationships” as in ISO 14044:2006. Hence, ISO was proposing allocation methods such as the marginal allocation developed by Azapagic and Clift [232], which are based on quantitative changes in the products or functions delivered by the system.

Finally, the last level allowed the allocation of different functions based on economic relationships, excluding allocation by physical properties. This preference for economic values could be due to their cause-oriented essence (the function is provided because one is willing to pay for it). Based on the analysis of this bilateral beginning and this ISO draft [233], it seems that the socio-economic ALCA school represents the first version of the ISO hierarchy. In fact, they distinguish themselves by applying system expansion by only adding (and not subtracting) functions, and preferring economic allocation to an allocation based on a physical parameter (excluded option by this first draft version).

3.4.2 The ISO 14041 influence (1999-2003)

In the third article of the second route, Azapagic and Clift used the boron product system to examine the different allocation methods recommended by ISO 14041, which was just released in 1998 [188,234]. ISO 14041:1998 introduced the same three levels of the hierarchy of the present ISO 14044:2006. However, the second level included the following clarification: “the resulting allocation will not necessarily be in proportion to any simple measurement such as mass or molar flows of coproducts''. Azapagic and Clift argued that, following ISO 14041, allocation by physical (causal) relationships had to be the result of mathematical system modeling [97,234]. Nevertheless, the ISO 14041 allocation underlying physical relationships allowed also allocations based on the "cause of the limits" of the amount of product output. This aspect emerges from the annex of ISO 14041:1998, where mass or volume allocations are suggested as representing physical relationships for road transportation because the quantity of materials transported is limited by the maximum load that the vehicle can carry [188]. Although these two approaches may at first appear contradictory, they are in fact in line with Azapagic and Clift's work, who also concluded that in some cases (which include the transportation example), allocating based on a physical quantity leads to the same results obtained by marginal allocation [97,234]. In these cases, it may be correct to allocate based on a physical parameter representing the physical causation involved, and therefore, not arbitrarily [97,234].

Azapagic and Clift (1999a) highlighted that system expansion (enlargement) is not always applicable. This approach is not possible when the goal of the study requires to determine the impacts of only one of the products [234]. The reason is that, by expanding the functional unit to include the co-functions, the results at the level of one single product would not be available. They also investigated allocation in heat and power cogeneration plants. Due to lack of data, they could not model the system to represent physical causalities and therefore applied the "avoided burdens approach" (in later research "substitution"). Azapagic and Clift argued that substitution is a conceptually equivalent alternative to system expansion, and is suitable when one co-product displaces its production elsewhere, such as for energy recovery from waste or cogeneration [234].

Actually, annex B of ISO 14041 quoted the same example of system-expansion/substitution applied to energy from waste incineration [188]. Nevertheless, annex B specified that the expansion of the boundaries like this requires 1) that the goal of the study is aimed at assessing a change, "i.e. a comparison between two alternative scenarios for the same product" and 2) that the modeled change which will actually occur because of the decision supported by the LCA can be predicted with a fair degree of certainty [188]. To apply this type of expansion, the LCA should aim, therefore, to answer the question of what would have been the long-term marginal effect if the service had not been performed [188]. Hence, substitution became a possible system expansion approach in what nowadays is understood as consequential thinking. This annex with allocation examples is no more included in the current ISO 14044:2006.

In the lowest level of the allocation hierarchy of ISO 14041 [188], economic allocation became an example, and no more the only acknowledged allocation method as it was reported in the previous draft version [233]. Hence, in some cases, allocation based on a physical parameter could be preferred to an economic allocation, and this might have given birth to the natural-science ALCA school. Moreover, ISO 14041:1998 specified that the environmental impact should be allocated only to the products causing the release of the emissions (causality principle). ISO 14041:1998 proposed the example of a multi-input incineration process releasing cadmium emissions which should be allocated only to the input wastes that contain cadmium.

The fourth article [97] refers once again to LP-based marginal allocation, stating that this modeling applies “when the functional outputs can be varied independently” i.e. in partial joint production or combined production (see appendix for more details about these definitions). A naphtha cracking was proposed as an example of a system where the outputs can be independently varied (within physical and thermodynamic limits) by changing the operating conditions [97]. When that is not the case (i.e. full joint production; with a fixed ratio of products), “allocation by physical causality cannot be implemented” [97]. Linked to this impossibility, they provided the -many times cited- example of the ratio of sodium hydroxide (NaOH) and chlorine (Cl_2) produced by electrolyzing brine, which is fixed by stoichiometry. Other examples that they mentioned about this impossibility are rapeseed oil/residue (ratio fixed by the chemical structure of the plant) and beef/leather (fixed by the physical structure of the animal) [97]. In these cases, the authors stated that ISO recommended economic allocation because it reflects “the socio-economic demands which cause the multiple-function systems to exist” [97]. They concluded that “allocation on an arbitrary basis, such as mass or energy flow, must be avoided’’ and “where physical causality between functional units and environmental burdens exists, the allocation should always be based on these causal relationships’’[97]. The authors based their methodological choices on the 1997 voting draft of ISO 14041.

The ISO 14041:1998 was complemented by ISO/TR 14049:2000 [186]. This technical report defined system expansion as the addition of functions but lost the concept of system expansion with substitution when the goal is to assess a change. This is still missing in the current ISO 14044:2006 and ISO/TR 14049:2012 [38,187].

This ISO/TR provided two examples related to the disposal phase of the life cycle. The first example showed how to expand boundaries to compare two processes with different outputs, A and B, using the same inputs. As illustrated in Figure 3A.1d in the appendix, the system boundary for each process needs to be expanded with an alternative process for making the other product. Then the two systems under comparison produce the same functional unit A+B. Moreover, it specified that the added processes shall be those that "would actually be involved when switching between the two analyzed systems" [186]. In the second example, open-loop recycling is solved with a closed-loop procedure that includes the entire recycling processes into the same system boundaries (like case c of Figure 3A.1 in appendix).

Concerning allocation by physical property (e.g. mass or viscosity), ISO/TR 14049:2000 [186] emphasized that this type of allocation should be preferred to economic allocation only when it reflects the way in which the inputs and outputs are changed by quantitative changes in the products, (as, for example, in the transportation example in ISO 14041:1998, quoted above). This had to be proven by varying the ratio of co-products [186].

In 2001, Ekvall and Finnveden published a critical review on allocation in ISO 14041:1998 [217]. Ekvall and Finnveden stated that system expansion (in the form of substitution) could be used in a broader range of LCA goals than the one for which it is recommended by the annex of ISO 14041. For example, it can be used to account for indirect effects [217], similar to how the substitution method is used today in CLCAs.

In the same review, Ekvall and Finnveden (2001) identified the marginal allocation of [97] as a method corresponding to the second level in the ISO hierarchy (the first connection between the two parallel routes in Figure 13). In particular, Ekvall and Finnveden [217] explained that there were two possible interpretations for ISO allocation based on physical-causal relationships. Under the first interpretation, the "environmental burdens allocated to a function should be the burdens avoided if that function is no more delivered while the other functions are unaffected" [217]. This type of allocation is applicable when the environmental burdens are linear with the quantity of each of the functions delivered and, therefore, it is possible to eliminate the functions independently [217]. The second interpretation is that "the environmental burdens allocated to each of the functions should be proportional to the partial derivatives at the point of operation" [217]. This is a generalized description of the LP modeling of Azapagic and Clift.

Concerning the third level of ISO hierarchy, they emphasize that a rigorous interpretation of the standard leads to an allocation based on other causal relationships, e.g. economic value, and not in non-causal relationships (e.g. allocation based on "arbitrary physical property of the products such as mass, volume or energy content") [217]. As aforementioned, this strict approach was also the only one foreseen in the first draft of ISO hierarchy [233] and favored by the ALCA socio-economic school. At this point, the main path stops to be bilateral and starts a period of interconnection that led to the development and definition of what today is categorized as "consequential thinking".

3.4.3 Consequential LCA influence (2004-2008)

In 2004, the keyword *consequential LCA* appeared for the first time [235]. In the same article, Ekvall and Weidema delineated the consequential LCA as commonly defined today. They stated that CLCA avoids allocation by applying substitution-type system expansion, using marginal data [235].

Following the main path of research, we found several articles on CLCA case-studies. In the first article, Thrane conducted a CLCA of fish products [236]. The second article authored by Schmidt and Weidema [237] is focused on how to identify the marginal vegetable oil to be substituted in a CLCA of agricultural systems providing food and oil. Thrane [236] pointed out that, generally, the ISO allocation hierarchy can also be considered valid for CLCA. In fact, when system expansion (either by enlargement or by substitution) or subdivision is not applicable, it is necessary to allocate by physical or other relationships also in CLCA [236]. Dalgaard et al. [238] then performed a CLCA of soybean meal and avoided allocation by applying the substitution of marginal vegetable oil [238]. The fifth article of this period authored by Thomassen et al. compares attributional and consequential LCAs of milk production [89]. They showed that depending on the modeling approach (ALCA or CLCA), the results significantly vary for the same system because of the different ways of dealing with multifunctionality (allocation versus system expansion with substitution). In the middle of this period, ISO 14044:2006 was released.

3.4.4. ISO 14044:2006 application (from 2009)

At this stage, the most cited article identified by the search was published by Finnveden et al. [62]. This article repeated that the underlying physical relationships of ISO14044:2006's second hierarchy level should represent physical, chemical, or biological causation (as also specified before in [157]). Consequently, economic, mass, or energy allocations were intended to be used only as the third level option [62].

Following the main path, we found an LCA on a bio-based plastic product derived from a blood meal [239]. Bier et al. highlighted how different approaches for solving allocation issues in LCAs of bio-based materials could widely vary the results. The next two articles of the main research path discussed the choice of allocation approaches to use in LCAs aimed at informing policy-making. Wardenaar et al. [240] pointed out that methodological uncertainty within ISO leads to significantly different results due to the influence of the allocation approach, and argued that the policy context could benefit from new guidelines [240]. Concerning the ISO hierarchy, they stated that "several authors have argued that substitution is equivalent to system expansion" referring as an example to [80]. However, "conceptually equivalent does not mean that system expansion and substitution provide the same results" because there are "large differences between these two methods" [240]. As a consequence of this assumed conceptual equivalency, some authors "use this implicit argument to choose for substitution, while still claiming compliance to ISO" [240]. Concerning allocation based on a physical parameter, Wardenaar et al. argued that the physical parameter should be the one reflecting the physical characteristics related to "the purpose or use of the product" i.e. the relevant characteristic for which they are sold [240].

Following the main path, we found a study on the Environmental Footprint guidelines published by the European Commission [212]. The study of 2014 of Pelletier et al. [212] highlighted that in ISO's first level system expansion, the functional unit is expanded to include the other co-functions (enlargement), and the impacts are therefore reported at the system level, i.e. at the level of all co-products [212]. This was claimed to be the "literal interpretation of ISO 14044" [68]. Accordingly, the PEF guide [85] does not consider substitution as a system expansion approach, but only enlargement (similarly the more recent PEFCR guidance [165]). However, the ILCD handbook allows system reduction as an option only in CLCA, and for those ALCAs whose aim is to include also the interactions with other systems [145].

The key-route main path analysis allowed us to identify the origins of the ”equivalency” substitution-system expansion i.e. the articles on the side of the bilateral beginning period originated by Tillman et al. [81]. The suggestion of Pelletier et al. [68] that this equivalency was originated from the study of 1994 of Tillman et al. [81] was therefore confirmed by our analysis. Nevertheless, the article by Tillman et al. [81] was published before any ISO standard and, therefore, did not refer to the system expansion method as intended by ISO.

In the next publication of the main path, Pelletier et al. (2015) observed that, despite the ISO hierarchy, consistent implementation of this hierarchy in the literature was limited, and presented the three schools of thought (consequential, socio-economic ALCA and natural-science ALCA) mentioned at the beginning of section 3.3.

The next two articles in the research path are focused on finding allocation parameters for agricultural systems. The first article proposed an allocation based on plant physiological construction cost for plant compounds, which should represent the underlying physical relationships between co-products i.e. the physiological mechanism involved in plant growth [241]. Hence, they concluded that, according to ISO, such a method should be preferred to allocation based on common properties of co-products, such as energy or economic content [241]. Subsequently, Mackenzie et al. [215] studied similar biophysical allocation methods and concluded instead that these methods might not represent the causal physical mechanisms of these systems because they overlook the interconnectivity between co-products [215] as instead, LP would do. Therefore, they concluded that allocation by economics is preferable [215]. Mackenzie et al. also pointed out that many practitioners often choose an allocation based on an arbitrary parameter (e.g. their mass or energy content) also when it does not reflect such a cause-effect mechanism [215].

The last two articles of the main path are focused on how to allocate burdens to by-products which were previously considered wastes [242,243]. These by-products are scarce wastes that can be converted into valuable products. In particular, Pradel et al. constructed a novel allocation method based on relevant causal relationships obtained by mathematical modeling [242]. This model was applied to wastewater treatment plants delivering sludge (by-product) and clean water (main product) and calculated the allocation factor for sludge and water.

3.5 DISCUSSION AND CONCLUSIONS

Despite the existence of a hierarchy for solving multifunctionality in ISO 14044:2006, the complexity of the multifunctionality problem, the lack of sufficient guidance, its difficult interpretation, and the discrepancies in other "ISO-compliant" guides or handbooks have led to a wide variety of allocation procedures in the literature. Such variety is especially present in the system expansion approaches and in the choice of the allocation key.

ISO 14044:2006 does not distinguish between attributional and consequential modeling. For many practitioners, distinguishing between attributional or consequential LCAs is a crucial key to select the method to deal with multifunctionality. For other practitioners, some mixed approaches can be considered advancements in the methodology. We found that only 25% of the LCAs clearly state the approach followed using the terms "attributional" and "consequential". Are practitioners not specifying it because they assume it to be "intrinsically clear" from the goal description, or because they do not agree with such a distinction? Some mixed approaches are also proposed in the literature.

The first major reason for debate on ISO's multifunctionality hierarchy is the application of substitution as a system expansion method in ALCA (found in 31% of the self-declared attributional studies explored through text mining). Such practice is perceived as inappropriate by many practitioners. However, some practitioners who do not acknowledge substitution as system expansion in ALCA recognize the use of substitution as an allocation method for ALCA. Concerning the use of substitution, another aspect that many practitioners pointed out is that a future ISO standard should emphasize more the criterion of physical/economical significance as a prerequisite to apply substitution to avoid incorrect interpretations of the results.

The second reason for the debate is the meaning and application of the "ISO relationships" criterion for the selection of the allocation method. A first interpretation (found in 28% of the case studies) is that the ISO refers to "causal physical relationships" as relationships mathematically modeled, while "other causal relationships" relate to other relationships (e.g. based on physical or economic parameters) selected based on the best proxy for physical relationships. The second interpretation is that allocation by "physical relationships" refers to an allocation by physical parameters (e.g. mass or energy) while "other relationships" refer to economic relationships.

Most (94%) of the LCAs of multifunctional case studies found in the literature search are linked to bioeconomy (agriculture, biofuels, bioenergy, and biomaterials) and its linked sectors (fossil fuels and petrochemical plastic materials and dairy products). This has generated inconsistencies within each area, but also at the boundaries between these sectors, because of their multiple links. As an example, biogas can be produced from the manure of a farm, which produces dairy products with animals that eat dried distillers grains with solubles coming from ethanol fermentation. Such ethanol production may have a pre-treatment process shared with a lactic acid fermentation. This lactic acid may be used to produce poly-lactide, which on the market replaces polypropylene. The biogas above can then be used to generate electricity, that can be partly consumed in the farm, and partially injected in the grid, substituting power from fossil fuels. How many double counting or inconsistencies arise when ISO 14044:2006 is interpreted differently in each of these sectors?

The bibliometric review based on the analysis of the main path obtained tracing the citation network allowed us to 1) reconstruct how the implementation practices of the ISO hierarchy developed in the last 25 years, 2) identify the origin of the different interpretations and their rationales, and 3) understand how the discrepancies found in the critical review were generated. It emerged that, originally, the ISO hierarchy [233] recommended the approach followed by the "socio-economic ALCA school". The socio-economic ALCA school interprets system expansion as enlargement but prefers economic allocation to allocation based on physical parameters representing a proxy for causality. The origin of the "natural-science ALCA school" was traced to ISO 14041:1998 [188], when, allocation by physical parameter, as well as economic allocation, was permitted as an example of ISO "allocation by other relationships". The natural-science ALCA school interprets system expansion as enlargement and applies allocation based on a physical parameter (for a part of the practitioners subscribing to this view, this choice is justified only when a physical parameter representing causality principles is identified). Its role was promoted by the release of the PEF guide and PEFCR guidance [85,165] which expressed a preference for allocation based on physical parameters over economic ones. Another important view is the one of the "CLCA school" interpreting system expansion as substitution and selecting the allocation method based on causality principles. The birth and development of the CLCA school were found in the annex of ISO14041:1998 and in the publications of Ekvall and co-authors [80,217,235]. They were the first ones (in the main path) to acknowledge the suitability of the substitution method to avoid allocation and account for counterfactual effects (originally proposed by Tillman et al. [81]) and the assumption of "conceptual equivalency" of substitution with the system expansion method.

Summarizing, following one or the other school of thought, a different method is often preferred for the same system, goal and decision context. Applying these different methods often leads to different conclusions, and sometimes, opposite conclusions.

To increase the consistency and reliability of LCA, we believe that a future revised ISO should:

1. State clearly if distinguishing between attributional and consequential LCA is a key principle to implement the hierarchy. If yes, then: it should differentiate the hierarchy for the two approaches and clarify if the hierarchy allows substitution as a system expansion method in attributional LCAs.
2. Clarify the meaning of allocation by "physical relationships" and "other relationships" providing more examples and details than the ones reported in ISO 14044:2006 and ISO 14049:2012.

3.A APPENDIX

Type of products

In this article, three definitions are used for different types of products and services: *co-products*, *by-products and wastes*.

Co-products are the ones satisfying the main (primary) function[3] that a production system or process is intended to deliver. Conversely, by-products represent only secondary functions of the system. A by-product is a substance resulting from a production process whose primary function is not the production of that item but either it is inevitably produced or could, in principle, be avoided by the system without altering the main functionality of the process (e.g. a farm offering also tourist accommodation services).

The primary function of a product system is identified by evaluating the purpose of such a system. For example, for the internal combustion engine of a car, the primary product is the mechanical power needed by the car to carry people (primary function). A secondary function of the same engine can be the production of heating (by-product) to keep a proper temperature in the car. Nevertheless, the distinction between primary and secondary functions can be particularly difficult for some unit processes (e.g. sunflower oil vs meal). When such difficulty is encountered, the primary function should be selected by assessing what function of the multifunctional process generates more revenues for the investigated process [51,145,211], within the temporal scope of the LCA. Nevertheless, there are processes whose aim is the generation of several functions of comparable value. In such a case, there can be multiple primary functions. For example, a biorefinery can produce various chemicals and fuels as primary functions (co-products) and provide district heating as a secondary function (by-product).

The shared environmental impact of a process shall be apportioned between co-products and by-products, but not to wastes [38]. According to ISO 14044:2006, wastes are “substances or objects which the holder intends or is required to dispose of” [38]. There is, however, a fine line between wastes and by-products. For example, manure is nowadays used as feedstock for biogas plants, used cooking oil is used for biodiesel production, and residues of the potato industry are used

[3] As highlighted by Majeau-Bettez et al. (2018), co-products have also been defined with the term ‘’primary’’, ‘’determining”, and others.

for animal feed. When these alternative uses make these wastes find a market demand represented by market values, they should be considered, therefore, as by-products. We adopt the distinction waste/by-product provided by the Waste Directive Framework [170]. A “waste” becomes a by-product when the “following conditions are met: 1) further use of the substance or object is certain; 2) the substance or object can be used directly without any further processing other than normal industrial practice; 3) the substance or object is produced as an integral part of a production process; 4) further use is lawful, i.e. the substance or object fulfills all relevant product, environmental and health protection requirements for the specific use and will not lead to overall adverse environmental or human health impacts” [170].

Type of multifunctional processes

As highlighted by several authors, the terminology reported in the literature for distinguishing the different types of multifunctional processes is not harmonized [51,71]. This article follows the terminology defined by Majeau-Bettez et al. [51], who differentiated between *full-joint production*, *partial joint production*, and *combined production*. *Full joint production* takes place when the co-products are produced simultaneously, with a fixed ratio of production (e.g. fixed by the stoichiometry of a chemical reaction, or by natural processes such as the proportions between wheat grains and wheat straw). *Partial joint production* occurs when there is an intermediate level of technological linkage between the different co- and by-products (e.g. an oil refinery as a whole or the production of milk and meat or the transportation of two different products) and *combined production* when there is not technological linkage (e.g. a gasoline station also offering shop services). According to this definition, the ratio of production of the co- and by-products could be varied in every case except for the full joint production.

Type of modeling approaches

The selection of the modeling approach is based on the goal of the study and the decision context. Generally, when the goal of a study is to describe the status of a system, an attributional LCA (ALCA) approach is followed to calculate the environmental impact of providing a specific amount of the functional unit [71]. When the goal is to describe the effect of a change due to a decision, a consequential LCA (CLCA) approach is followed to estimate how this environmental impact would change in response to a *change* in the output of the functional unit (i.e. it is change-oriented) [89]. The current conceptualization of the CLCA approach was first publicly discussed in the 2001 international workshop on electricity data for life cycle inventories [244].

One of the main principles of ALCA is the so-called 100% additivity [51]. This principle means that "results of a separate analysis of all economic activities should add up to the result of an analysis of the total economic activity"[74], so ALCA is suitable for attributing the total impacts to a defined function (product or service), but, for example, it does not indicate to policy makers the impact of policy changes, when these cause an incremental change from the *status quo*.

By contrast, CLCA determines the *change* in impacts due to a *change* in the production of the product or service, or to a change in policy. So it attempts to consider *all* the impacts of the change, also on other sectors that are influenced, for example as a consequence of the use of by-products [51]. CLCA is therefore preferred to ALCA for estimating the impact of policy changes [64]. CLCA usually uses market-driven modeling to forecast what will happen once the product or service of interest is introduced [35]. This means that in CLCA, marginal processes are considered, rather than average ones, including the activities displaced by by-products. This is typically modeled through the so-called substitution approach, whereby CLCA considers only the activities reacting to the change in demand for the functional unit, keeping the total of other services constant. Therefore, the quantification of displaced activities depends on the market characteristics of competing products [177].

Type of system expansion approach and substitution

System expansion means the enlargement of the boundaries of the system under investigation to include additional processes and functions. As mentioned above, expansion of the boundaries can be used to avoid allocation. There are two possible approaches to avoid allocation by expanding the boundaries: enlargement (see Figure 3A.1 for different types of enlargement) and substitution (see Figure 3A.2). By considering the subtraction as "a negative addition" [205], substitution is considered by some LCA practitioners as a form of system expansion used to isolate the impact of just one function from a multifunctional process.

One can apply system enlargement by modifying the functional unit to include all co-functions (case a of Figure 3A.1). This approach is not possible when the goal of the study requires to determine the impacts of only one of the products because the results at the level of one single product would not be available. System enlargement is also often used for comparative assessments. In Figure 3A.1b, the aim is to compare process P1 (providing functions A and B) with process P2 (providing only function A). One needs to add to P2 another process for producing B in order to allow the comparison for the same outputs. Similarly, in Figure 3A.1d, the aim is to compare a process producing A with a process producing B (for example, comparing the impacts of two products which could be made from the same raw material). In this case, one needs to add alternative processes for making both A and B in order to make a meaningful comparison. Even though these processes are not initially multifunctional, system enlargement is applied to allow for a fair comparison. One can also apply system enlargement in open-loop recycling systems. In the example of system enlargement from ISO/TR 14049, open-loop recycling is solved with a closed-loop procedure that includes the entire recycling processes into the same system boundaries (like case c of Figure 3A.1).

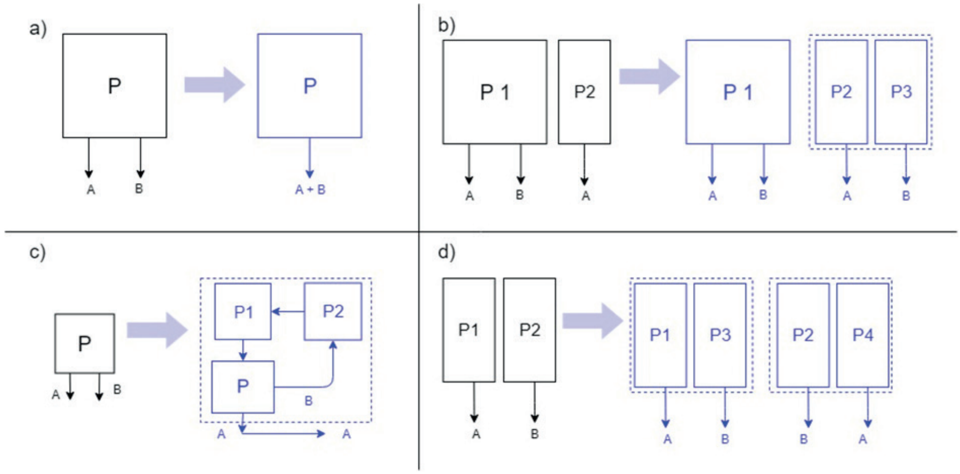

Figure 3A.1 Different ways to apply system expansion as enlargement-addition of functions. In black: multifunctional process before applying system expansion. In blue: process after the expansion of the boundaries/addition of functions. a) Changing the FU to avoid allocation. b) Adding extra processes (P3, delivering B) to a system (P2, delivering A) that is compared with another system (P1) delivering several functions (A and B). c) Applying closed-loop recycling to a system (P) where one of the outputs (B) is used as a material input in the same product system (P2 represents the intermediate processing of B that allows its re-use). d) Adding extra processes (P3 and P4) to compare systems that provide different functions and that at the beginning were not multifunctional

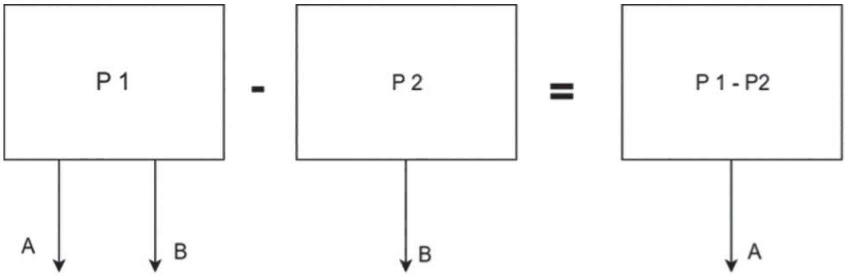

Figure 3A.2 System expansion by substitution (reduction of functions). The investigated system (P1) delivers two products (A and B). Alternatively, product B can be produced by another system (P2). The substitution method proposes that the impact of producing A (only) by process P1, corresponds to the difference of impact between P1 and P2.

CHAPTER 4

Kraft lignin as a bio-based ingredient for Dutch asphalts: an attributional LCA

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ABSTRACT

In the last decade, lignin has received much attention as a feedstock to produce bio-based products. This study investigates the potential benefits of using lignin to mitigate the environmental impact of the road construction sector. An environmental life cycle assessment (LCA) of various top-layer bio-based asphalts using kraft lignin was conducted. From a cradle-to-grave perspective, lignin-based asphalts were compared with conventional asphalts.

The results of the LCA revealed that the climate change impact of lignin-based asphalts could be 30-75% lower than conventional asphalts. For the other ten impact categories, trade-offs were observed. Overall, two key factors to make the environmental impact of lignin-based asphalts lower than conventional asphalts are 1) increasing the amount of bitumen-substituted and 2) using low-grade biomass fuels for process steam in the pulp mill. The substitution of weak filler with lignin was beneficial only for climate change and could lead to a worse overall environmental performance than conventional asphalts. Similarly, higher environmental impacts for lignin-based asphalts could be obtained if the pulp mill consumed natural gas to complete the energy balance to replace the part of the black liquor from which lignin is extracted.

This study also includes an in-depth discussion on methodological choices such as the allocation methods for lignin, functional units, and asphalt layers considered. We believe that such a methodological discussion could be helpful to support future Product Category Rules for asphalt mixtures.

4.1 INTRODUCTION

10% of the GHG emissions of the transportation sector are caused by the construction of roads [245]. This amount corresponds to more than 5% of the total greenhouse gas (GHG) emissions generated in the European Union (EU) [246]. Picturing, this amount is twice as much as the impact contributed by the aviation sector [246]. To contribute to the EU climate change reduction targets, the Netherlands aims to make this sector carbon-neutral by 2030 [247]. Some circular economy practices, such as the recovery and recycling of deteriorated asphalt, are already well-established all around Europe [248,249]. However, such practices alone are not sufficient to achieve carbon neutrality. To achieve such a goal, it is necessary to couple asphalt recycling with alternative construction and production methods as well as alternative (e.g. bio-based) materials and renewable energy sources [134,249].

Projects in the construction sector are often assigned via public tenders. The environmental aspects are becoming more often part of the public tenders in the EU member states [250,251]. With this purpose, the results of environmental Life Cycle Assessments (LCAs) have recently been increasingly used in public tenders of infrastructure projects such as roads, airports and railways [250,251]. LCA, which is standardized by ISO [38,127], is acknowledged as a rigorous methodology and should provide unbiased and comparable environmental calculations. Hence, combining properly the environmental impacts calculated using LCA with project costs should avoid that low-cost materials with high environmental burdens are selected in investments using public funding [251]. With this purpose, tenders in the Dutch construction sector include an environmental cost indicator that simplifies and unites various environmental impacts into a single monetary value score representing the avoided damage cost or shadow cost [250,252].

Bitumen production is one of the most important environmental burdens of asphalts [134,252]. For this reason, some renewable alternatives to bitumen such as plant-based binders, municipal wastes, and live-stock manures are currently under investigation [134,253]. Lignin, a bio-based by-product of pulp mills and lignocellulosic biorefineries, has received attention as a renewable binder to replace bitumen in asphalt mixtures [134,254]. Several roads using lignin-based asphalts have been paved in the Netherlands in the last five years [255]. However, there is a lack of clear evidence supporting the environmental benefits of producing lignin-based asphalts. To our knowledge, there is only one peer-reviewed LCA [134] of lignin-based asphalts. The results of this LCA suggest that lignin-based asphalts with 25% replacement of the bitumen binder with lignin allow almost a 6% reduction in asphalt production GHG emissions [134]. However, this LCA study [134] presents some research gaps and leaves space for further research. For instance, the use- and end of life- phases were not considered in the study, and it only focused on climate change impacts. Moreover, the life cycle inventory of lignin production lacks transparency for what concerns data, allocation methods and biogenic carbon accounting, which are all crucial aspects to interpret the results of an LCA of a lignin-based application [254].

Our study presents the cradle-to-grave LCA of various top-layer lignin-based asphalts using kraft lignin, assessing 11 impact categories together with an environmental cost indicator. The cradle-to-gate (pulp mill gate) environmental impact of kraft lignin is also discussed. Since the Netherlands is the geographic scope of this LCA, beyond ISO 14040 and ISO 14044 [38,127] and EN 15804 [256], the Dutch Product Category Rules (NL-PCRs) [257], SBK bepalingsmethode 3.0 [258] and Dutch LCA asphalt sector report [252] were used as reference documents to conduct the LCA. Nevertheless, even following the best LCA practices recommended by these methodological documents, multiple sources of methodological uncertainty exist and the results of LCAs should be carefully interpreted [50,146,208]. For this reason, the effect of the allocation methods, functionali units and product systems' definition was broadly discussed through sensitivity analyses.

4.2 MATERIALS AND METHODS

4.2.1. Goal and scope definition

This study aims to assess and compare the environmental impact of lignin-based asphalts with conventional asphalts. The intended audience of this study is made of technology developers and researchers working on lignin-based applications. Given the valuable research conducted concerning LCA methodological implications presented in the discussion section, we believe that this study can be relevant for the LCA community involved in the construction sector. Moreover, the results of this study might be of interest for stakeholders that look for options to reduce the carbon footprint of road construction, e.g. policymakers, the road construction sector. The geographic scope is the Netherlands, and the temporal scope is 5-10 years from now.

In comparative LCAs, a proper functional unit, i.e. the reference unit of comparison, should be carefully selected. Hence, how well a product system fulfills a specific function should be accounted for in the functional unit. In particular, asphalts are required to maintain multiple rheological performances, especially in terms of stiffness/deformation resistance but also workability, durability, permeability and resistance to possible damages caused by moisture or fuels and oils [134,259,260]. Moreover, one of the key mechanisms of failure of asphalts affecting their lifetimes is the occurrence of fatigue cracks (predominately) within the binder of asphalt [134,261,262]. For this reason, various compositions for lignin-based asphalts were tested via standardized methodologies in the laboratory by Asfalt Kennis Centrum (project partner) to guarantee similar functional properties and performances to conventional asphalts. Empirical and functional tests were conducted according to respective documents of EN 13108 series (1 and 20) and EN 12697 series (8-12-23-24-25-26-31-33-35). The empirical tests included run on gyrator samples, hollow space percentage and water sensitivity and were conducted for all asphalt types investigated. The functional tests included performing on gyrator test pieces and rolled test plates, tri-axial test for rutting resistance, stiffness on test beam sawn from the rolled test plates, fatigue on the same beams. The functional tests were applicable only to asphalt concretes. Using these compositions, based on the goal of the study, a functional unit of 1 tonne (t) of top layer asphalt was selected in line with the reference unit used by the Dutch LCA asphalt sector report [252]. Adopting a different functional unit representing the whole asphalt product made of several layers is also considered in section 4.4.2. The baseline calculations refer to stone mastic asphalts (SMAs). Asphalt concretes (ACs) and porous asphalts (ZOABs with the Dutch acronym) were also assessed as alternative product systems. The LCA has a cradle to grave scope. Figure 14 illustrates the sub-division into life cycle stages with coding according to the Dutch reference documents [252,257]. As indicated for environmental product declarations based on the European standard EN 15804 used in the construction sector [263], this LCA is conducted using attributional modeling. By attributional modeling, all the environmental burdens of all processes involved in the life cycle were accounted for with representative average data [68,146]. As mentioned in the introduction, the cradle-to-gate environmental impact of kraft lignin is also discussed in this study. For such a product, a functional unit of 1 kg of dry lignin was adopted.

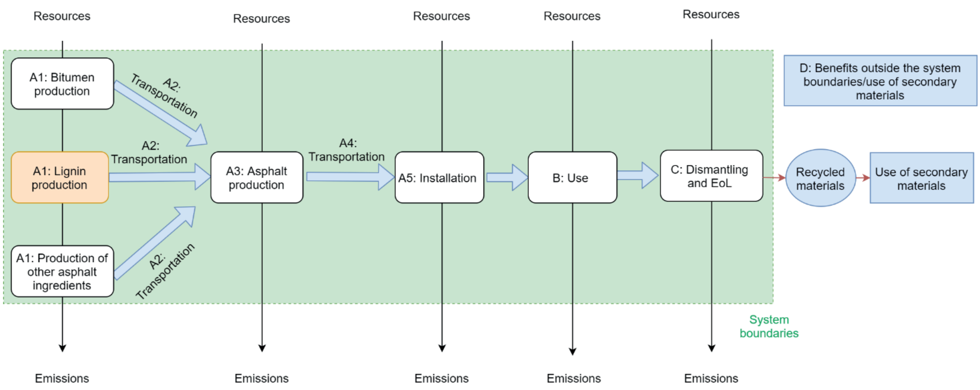

Figure 14. Flow chart of lignin-based asphalt with cradle-to-grave life cycle stages based on [252]. *The unit process named "A1:Lignin production" starts from the cradle to the gate of the pulp mill where lignin is delivered.*

The impact categories and respective weighting factors (see Table 4) were selected based on the calculation rules of SBK bepalingsmethode 3.0, which is based on the recommendations from EN15804 + A2 (2019) [256]. In particular, the weighting factors used in the Netherlands are the so-called MKI weighs [258], which are based on the shadow price method (internationally often referred to as environmental cost indicator). The shadow price corresponds to the cost of the preventive measures for the government to avoid that environmental impact [252].

Table 4. Environmental impact categories and weighting factors [258].

Impact category	**Unit**	**Weighting factor (€/kg or €/MJ)**
Abiotic depletion	kg Sb eq	0.16
Abiotic depletion (fossil fuels)	MJ	7.7E-05
Global warming (GWP100a)	kg CO_2 eq	0.05
Ozone layer depletion (ODP)	kg CFC-11 eq	30.0
Human toxicity	kg 1,4-DB eq	0.09
Freshwater aquatic ecotoxicity	kg 1,4-DB eq	0.03
Marine aquatic ecotoxicity	kg 1,4-DB eq	0.0001
Terrestrial ecotoxicity	kg 1,4-DB eq	0.06
Photochemical oxidation	kg C_2H_4 eq	2.0
Acidification	kg SO_2 eq	4.0
Eutrophication	kg PO_4--- eq	9.0

4.2.2. Life cycle inventory

4.2.2.1 Extraction and processing of raw materials (A1)

The compositions of the asphalts compared were determined with the objective to have similar functionality performances. For this reason, the same type of asphalt (SMAs, ACs or ZOABs) with lignin or without lignin were assumed to have the same lifetime. The compositions of lignin-based and conventional asphalts are reported in Table 5. From this table, it is possible to notice that lignin replaces bitumen and a fraction of weak filler. For ZOABs, lignin replaces only weak filler and not bitumen. Table 5 also reports the background inventory data. As for the Dutch LCA asphalt sector report, the background data were retrieved from the ecoinvent library named "Allocation cut-off by classification" and the Dutch National Environmental Database [264], which contains various datasets representing the environmental performance of buildings in the Dutch context. In particular, the ecoinvent system model "Allocation, cut-off by classification" is an attributional database where the primary (first) production of materials is always allocated to the primary user of a material [265]. Accordingly, if "a material is recycled, the primary producer does not receive any credit for the provision of any recyclable materials", and the recyclable materials are available burden-free to recycling processes and bear only the impacts of the recycling processes [265]. However, in the Dutch asphalt sector, the credits generated outside the system boundaries are allocated to the final environmental impact of the asphalt (see section 4.2.2.6).

4.2.2.1.1 Lignins

There are various technologies and feedstocks from which lignin can be derived from pulp mills or lignocellulosic biorefineries [254]. The kraft process is the dominant process of the pulping industry [122]. With the kraft process, wood is converted into wood pulp, which is used in the production of paper. Black liquor is a by-product of the wood pulping process in a pulp mill. The major component of black liquor is lignin. The black liquor is usually burnt in a recovery boiler to produce internal process energy [103]. Alternatively, kraft lignin can be obtained from black liquor through precipitation and separation processes; for example by the LignoBoost process [266].

Table 5. Composition of 1 t of top layer asphalt based on industrial partner's primary data and background inventory datasets used. Lifetimes=15 years for SMAs and ACs and 12 years for ZOABs. B=bio-based, C=conventional.

Raw material (amounts expressed in kg)	**B-SMA**	**C-SMA**	**B-AC**	**C-AC**	**B-ZOAB**	**C- ZOAB**	**Inventory dataset**
Recycled content	0	0	288	300	0	0	Burden-free
Cellulosefiber	3	3	0	0	0	0	SBK_ Cellulosevezels [264]
Bitumen	44.5	65	21.1	40	43	43	ESU NL-PCR bitumen [252]. The selection of the bitumen dataset is discussed in section 4.4.1.1.2
Crusher sand	88	75	154	171	106	106	Sand {CH}| gravel and quarry operation | Cut-off from Ecoinvent 3.6
Natural sand	88	75	76	57	0	0	SBK 296 Industriezand [264]
Crushed stone (Morene)	0	0	411	410	811	811	Gravel, crushed {CH}| production | Cut-off from Ecoinvent 3.6
Crushed stone (Porfier)	700	710	0	0	0	0	Gravel, crushed {CH}| production | Cut-off from Ecoinvent 3.6
Weak filler	35	72	23	22	0	40	Limestone, crushed, washed {CH}| production | Cut-off from Ecoinvent 3.6
Lignin	40	0	24	0	40	0	Modelled based on literature sources (see section 4.2.2.1.1)
Linseed oil	1.5	0	2.9	0	0	0	Linseed seed, at farm {CH}| linseed seed production, at farm | Cut-off from Ecoinvent 3.6

In our LCA model, kraft lignin is separated from the black liquor and used in asphalts. For the production of kraft lignin, two studies were considered as main data sources: Culbertson et al. (2016) & Bernier et al. (2013). The background data can be found in the Appendix. In these two studies, the precipitation step is carried out by injecting liquid carbon dioxide in the black liquor together with sulfuric acid [100,103]. Three lignin production "scenarios" were modeled, one based on Culbertson et al. (2016) and two based on Bernier et al. (2013):

- "kraft1-BIOM" was modeled based on the inventory data from Culbertson et al. (2016). The pulp mill modeled by Culbertson et al. is equipped with a hog fuel boiler, has an internal power plant that sells an electricity surplus to the electric grid and uses natural gas only for the lignin extraction process.
- "kraft2-NG" was modeled based on the pulp mill modeled by [103]. The electricity is instead supplied to the pulp mill by the grid and natural gas is used to produce the part of the energy that is no longer produced from burning black liquor (due to the extracting of lignin). Since the consumption of natural gas (NG) to produce the kraft lignin is much higher in Bernier et al. (2013) than Culbertson et al.(2016), the lignin modeled based on Bernier et al. (2013) will be referred to as "kraft2-NG".
- "kraft2-BIOM" was modeled as well based on the pulp mill modeled by Bernier et al. (2013). According to Bernier et al., the same pulp mill could also potentially use hog fuel (chips of wood bark) instead of natural gas to compensate for the loss of steam production from the recovery boiler. Since the heat source is a key factor affecting the environmental impact of lignin [254], this third scenario represents the use of hog fuel instead of natural gas to compensate the loss of steam from the recovery boiler.
- Details on inventory data can be found in Appendix.

4.2.2.1.2 Allocating the environmental burdens to lignin

Since the production of kraft lignin is the result of a multifunctional process, it is necessary to apply an allocation method to apportion the environmental burdens of the pulp mill between lignin and pulp. In the LCA literature, it is acknowledged that the environmental impact of kraft lignin is importantly affected by the allocation method applied between pulp and lignin [55,254]. For this reason, how the allocation method was selected based on the reference documents is here extensively illustrated. The Dutch Product Category Rules [257] recommends following the directions from EN 15804 [256].

EN 15804:2012+A2:2019 specifies that the allocation between the co-products should be avoided by subdivision every time possible. The other typical option to avoid allocation i.e. system expansion is not allowed by EN 15804:2012+A2:2019. This is a common statement in rules for environmental product declarations to avoid that a substitution approach (one of the methods to perform system expansion) is used as a system expansion method in attributional LCA modeling [267,268]. In fact, the use of the substitution as a system expansion method in attributional LCAs is considered misconduct by many LCA practitioners and often leads to misleading interpretations or erroneous results [51,57,68]. If subdivision is not possible or data for sub-processes are not available, the allocation can be based on the underlying physical causality relationship i.e. reflecting "the way in which the inputs and outputs are changed by quantitative changes in the products or functions delivered by the system" [38].

This type of allocation is often referred to as "physical causality allocation [146,214,215]. A physical causality allocation relies on the mathematical modeling of the changes in operating conditions of the process under investigation to establish a physical causality relationship between functional units[4]. Accordingly, the PCR 2019:14 v.1.0 for construction products [268] and ISO 21930:2017 [269] i.e. the ISO core rules for environmental product declarations of construction products remark that physical causality can be established only if "each of the co-products can be produced without the other(s) or the ratio of the co-products typically varies in normal production", which is the only case when allocation by physical causality can be modeled [97,146,215,216]. According to EN 15804, allocation based on simple physical properties as mass can be used as a proxy for physical causality only if the difference in revenue from the co-products is low (defined as max. 25% difference) [256]. In all other cases, the allocation shall be based on economic values [256].

Accordingly, the pulp mill modeled by Culbertson et al.(2016) generating kraft1-BIOM was subdivided as much as possible. In this way, the lignin extraction process itself was not allocated to the pulp and bleaching was not allocated to lignin. For the other processes, keeping the pulp output constant and extracting kraft lignin leads to a change in the ratio between pulp and the surplus of electricity output produced (and a minor change in the soap output). So, allocation by physical

[4] Sometimes, such a type of relationship can be reflected by a simple physical parameter for volume allocation in a truck transporting empty packaging or mass allocation in a truck transporting full packaging [187].

causality cannot be established. Moreover, since the difference in revenue from the co-products is not “low”, economic allocation was used. To calculate the economic allocation share for lignin, the following prices were used: 535 €/t for kraft lignin [254], 788 €/t for pulp [270], 111 €/t for soap and 0.08 €/kWh for the surplus of electricity [271]. By using these prices, an allocation factor (A_L) of 7.3% to lignin was calculated. Two sensitivity analyses varying the price of kraft lignin and using mass allocation can be found in section 4.4.1.1.

In the case of the kraft2-NG lignin and kraft2-BIOM, electricity is supplied from the grid. Hence, a physical causality allocation was applied for this pulp mill. This type of allocation is based on the “physical causal relationships between burdens and functional outputs, which in turn requires a model of the system behavior” [157]. Taking the words of Azapagic and Clift, who are the authors that more than others have emphasized the importance of this allocation method [146], “when the causal relationships are represented by a model which describes the real behavior of the product system, the model can be used to allocate burdens between different functions by exploring how the burdens change when the quantity of one function is changed with the quantities of all the other functions kept constant” [157]. In order of magnitude, there are three types of changes that can be modeled, i.e. marginal-, incremental- and average [146,157]. Based on the approach adopted by the data source [103], average changes were used as the base for the allocation. Average changes are substantial changes such as eliminating or adding a functional output [146,157]. In Bernier et al. (2013), this type of allocation is “performed by comparing emissions with and without lignin recovery for the same mill and assigning the differences to lignin”.

Both allocation methods preserve the additivity principle of environmental impacts, one of the key aspects of attributional modeling [51,146]. Such a principle for the two allocation methods applied is expressed by equation 1.

$$I_t=I_p+I_L=I_t*A_{pp}+I_t*A_{Lp}=I_t*A_{pe}+I_t*A_{Le} \quad \text{Eq.1}$$

Where:

- I_t is the total impact of the pulp mill producing both lignin and pulp
- I_p is the impact of the pulp mill if producing only pulp
- I_L is the additional impact generated by the pulp mill when lignin is extracted compared to when pulp only is produced

- A_{pp} is the physical causality allocation factor for pulp ($=I_p/I_t$)
- A_{Lp} is the physical causality allocation factor for lignin ($=I_L/I_t$)
- A_{pe} is the economic allocation factor for pulp
- A_{le} is the economic allocation factor for lignin.

4.2.2.2 Transport to the producer (A2) and production of asphalt (A3)

The transportation distances were mostly based on primary information from the asphalt industry. For the transportation of other materials for which primary information was not available, standard values from the Dutch LCA asphalt sector report [252] were used. A summary can be found in Table 6.

Based on primary data collected from one industrial producer (Roelofs Groep), natural gas consumption during the production phase differs between lignin-based and conventional asphalts. Each t of lignin-based asphalts require on average 5.3 Nm^3 of natural gas (NG), while conventional asphalts require on average 7.5 Nm^3 of natural gas. According to Schwarz et al. (2020), asphalts with recycled material (PR) use more natural gas due to the overheating stage. The overheating stage is responsible for 6% of the total natural gas use of asphalt production, and the overheating of one kg of PR requires 0.0015 Nm^3 of natural gas (Schwarz et al., 2020). Therefore, the natural gas usage of each asphalt type can be calculated by using equation 2:

$$\text{NG usage } [Nm^3] = \text{average NG usage } [Nm^3] * (1-0.06) + 0.0015 * \text{PR [kg]} \qquad \text{Eq.2}$$

Filling equation 2, the natural gas requirement is calculated as 5.0 Nm^3 for SMA and ZOAB and 5.4 Nm^3 for AC. Conventional SMAs and ZOABs require 7.1 $Nm^{3,}$ while ACs require 7.6 Nm^3. Moreover, based on primary data from the same industrial producer, per each tonne of asphalt, on average, 5.6 kWh of electricity and 0.17 liters of diesel are needed during the production phase. For electricity, the dataset *Electricity, medium voltage {RER}| market group for | Cut-off* from ecoinvent 3.6 was used updating the shares of electricity per source based on the 2030 EU reference scenario for the Netherlands [272].

In the Dutch LCA asphalt sector report [252], for conventional asphalts, natural gas and electricity consumptions are 5-15% higher than assumed in our study based on primary data, while diesel consumption is 40% lower. A sensitivity analysis considering such a relatively reasonable variation of data (probably linked to differences in the plant configurations or younger/older plant) was not conducted. On the other hand, in the Results section, the LCA results of lignin-based asphalts were compared with the calculated impact for conventional asphalts and the impacts for that type of asphalt reported in the Dutch LCA asphalt sector report. In this way, not only the possible differences in utility consumptions but also the ones in the assumed compositions (and transportation distances) were considered.

Table 6. Transportation distances assumed for the asphalt mix's ingredients

	km by lorry truck	km inland shipping	km transoceanic shipping	Data source
Cellulosefiber	177	0	0	Standard value from the Dutch LCA asphalt sector report [252]
Bitumen	64	0	0	primary information from the asphalt industry
Crusher sand	7	0	0	primary information from the asphalt industry
Natural sand	86	0	0	primary information from the asphalt industry
Crushed stone (Morene)	25	660	0	Standard value from the Dutch LCA asphalt sector report [252]
Crushed stone (Porfier)	25	53	933	Standard value from the Dutch LCA asphalt sector report [252]
Weak filler	7	0	0	primary information from the asphalt industry
Lignin	146	0	1822	primary information from the asphalt industry
Linseed oil	150	0	0	primary information from the asphalt industry

4.2.2.3 Transport to the construction site (A4) and installation (A5)

The distance for the transportation to the construction site was taken to be 50 km by lorry (*Transport, freight, lorry 16-32 metric ton, EURO4 {RER}| transport, freight, lorry 16-32 metric ton, EURO4 | Cut-off*) based on [252]. During the installation of the product, 0.26 liters of diesel [252] are consumed (*Diesel, burned in building machine {GLO}| market for | Cut-off* from ecoinvent 3.6).

4.2.2.4 Use phase (B)

The service lifetime has currently no influence on the environmental impact calculations per tonne of asphalt conducted following the Dutch NL-PCR [252]. Based on the Dutch LCA asphalt sector report [252], the only process from the use phase included in this LCA is the leaching of substances from top layers to the soil under the influence of precipitation, which mostly happens in the first years after construction. For this reason, such an effect is considered independently of the lifetime of the asphalt [252]. The leaching emissions of various types of conventional asphalts determined with laboratory tests of factory samples were retrieved from [252]. Based on industrial partners (Asfalt Kennis Centrum and Holding de Vier Ambachten-H4A), there is a piece of the first evidence that the leaching emissions of lignin-based asphalts are in line with the ones from conventional asphalts, but further research is needed. So, for lignin-based asphalts, the same inventory data for leaching were used.

Other environmental burdens during the use phase, such as the ones due to the maintenance of the road surface and repairs, are not included [252].

The NL-PCR Asphalt [257] also considers a 10% mass loss of bitumen in top layers due to the effects of erosion [252]. This material loss occurs in nature as an inert material [252]. Similarly to the bitumen binder, it is assumed that a 10% loss due to erosion is also present in lignin and linseed oil.

4.2.2.5 End of life (C)

The end of life phase of asphalts is made up of several stages. The first stage is the demolition i.e. the removal of the asphalt, which requires an average of 23.0 MJ of diesel [252]. As background data for such a diesel consumption, the process *Diesel, burned in building machine {GLO}| market for | Cut-off* from ecoinvent 3.6 was used. Then, the recovered asphalt is transported for 50 km to the processing site to make road construction mixtures with a percentage of recycled asphalt. For the modeling of this transport step, the process *Transport, freight, lorry 16-32 metric ton, EURO4 {RER}| transport, freight, lorry 16-32 metric ton, EURO4 | Cut-off* from ecoinvent 3.6 was used. At the asphalt plant, the recovered asphalt is processed via breaking followed by blending/mixing, which requires 13.4 MJ of diesel per tonne of asphalt [252]. As background data, also for such a diesel consumption, the process *Diesel, burned in building machine {GLO}| market for | Cut-off* from ecoinvent 3.6

was used. Based on industrial partner information (Holding de Vier Ambachten-H4A), only 1% of the recovered asphalt (at the net of material losses described in section 4.2.2.6) ends in a landfill (*Waste concrete {Europe without Switzerland}| treatment of waste concrete, inert material landfill | Cut-off* from ecoinvent 3.6). Consistently with the selection of the ecoinvent library named “Allocation cut-off by classification”, the entire environmental impact of the end of life (C) is apportioned to the primary asphalt. So when an asphalt uses recycled content (secondary material), this material is free of environmental burden in module A1.

4.2.2.6 Benefits and loads beyond the boundaries (D)

Based on the NL-PCR and Dutch LCA asphalt sector report [252,257], module D is “outside the system boundaries” but included in the calculation of the environmental performance of the asphalt. In module D, the benefits resulting from recycling primary raw materials and the loads resulting from the loss of secondary raw materials are accounted.

Therefore, module D includes the benefits generated by the avoidance of the extraction (A1) and the burdens of transport of recycled asphalt granulates to the asphalt plant (A2) but also the loads of material that was obtained via recycling from the previous cycle but doe not get recycled in the current cycle. According to the NL-PCR [257], the amount of materials recovered during recycling is calculated considering two types of material losses. The first loss is due to the fraction of asphalt that leaves the asphalt system with no further use (e.g. a small fraction goes to landfill) or reused in a low-value function other than asphalt e.g. towards foundation layers. In the NL-PCR [257], this fraction accounts for 29% (L_1). Such a figure was based on the results of the investigation conducted by [273] and applies to all asphalt layers. The second material loss (L_2) is represented by the 10% loss of bitumen (and lignin and linseed oil by assumption) due to erosion in the use phase of top layers described in section 4.2.2.4. Moreover, based on the NL-PCR [257], two quality losses were also accounted respectively for bitumen quality (4% quality loss Q_1), which was also applied to lignin, and the quality loss of crushed stones (Q_2=10% in the top layer and 5% for sub-layers). With respect to anti-drip inhibitors, they remain in asphalt but no longer fulfill any function [257]. So, they do not receive any credit in module D.

Equation 3 from NL-PCR [257] shows the mass of each raw material *i* that obtains credits or loads for their A1 and A2 impacts. In particular, the loads are represented only by the second part of the equation where m_R is the mass of recycled content and M_{Ri} is the fraction of raw material *i* in the recycled content. As a derogation from NL-PCR [257], the mass of the recycled content was assumed to be the same as the one of the top layer asphalt. In particular, based on the compositions assumed (see Table 5), ACs only have recycled content and therefore a small percentage of loads (minor compared to the benefits) using Equation 3.

$$m_i*(1-L_{1i})*(1-L_{2i})*(1-Q_{1i}-Q_{2i}))-(m_R*M_{Ri}*L_{1i}*(1-L_{2i})*(1-Q_{1i}-Q_{2i}))_{i\neq R} \qquad \text{Eq.3}$$

4.2.2.5 Biogenic carbon

Lignin-based asphalts contain two bio-based ingredients, which are lignin and linseed oil. The biogenic carbon content of kraft lignin was measured in the lab as 2.4 kg CO_2eq/kg. The biogenic carbon content of linseed oil was assumed as equivalent to 2.9 kg CO_2eq/kg based on [274]. One of the main updates to EN 15804:2012 (A2:2019) is that, in the new version, it is specified that “the biogenic carbon content shall not be allocated but always reflect the physical flow” [263]. Additionally, the NL-PCR [257] specify that “if the submitter can demonstrate based on tests and other technical evidence that the CO_2 is permanently stored in the asphalt product, the CO_2 stored could be included in the calculation of the CO_2 impacts”.

The biogenic carbon from the bio-based inputs that is lost as an inert material (or ends to landfill) is assumed not to biodegrade since lignin does not biodegrade under landfill anaerobic conditions [275,276] and is mixed with bitumen in the asphalt matrix. So, it can be considered permanently stored during the life cycle due to losses/landfill since it is expected to remain stored for 100 years. Moreover, the lignin and linseed oil contained in the asphalt that is recycled (the part that is not lost) will go to sub-layers and over a time horizon of 100 years, it will be permanently stored in other asphalt products or e.g. lost as inert material in foundations in the following life cycles. The top layer asphalt contributed to store such an amount of biogenic carbon for a percentage of time, and after 100 years, it will be permanently stored. Hence, the top layer should be entitled to a percentage of the credit for the permanent storage of biogenic carbon over 100 years, which is therefore accounted (for SMAs and ACs, 15% over 100 years, for ZOABs 12%). This credit was included in module D.

4.3. RESULTS

4.3.1 Kraft lignins

The cradle-to-gate climate change impact of the three kraft lignins assessed is shown in Figure 15.

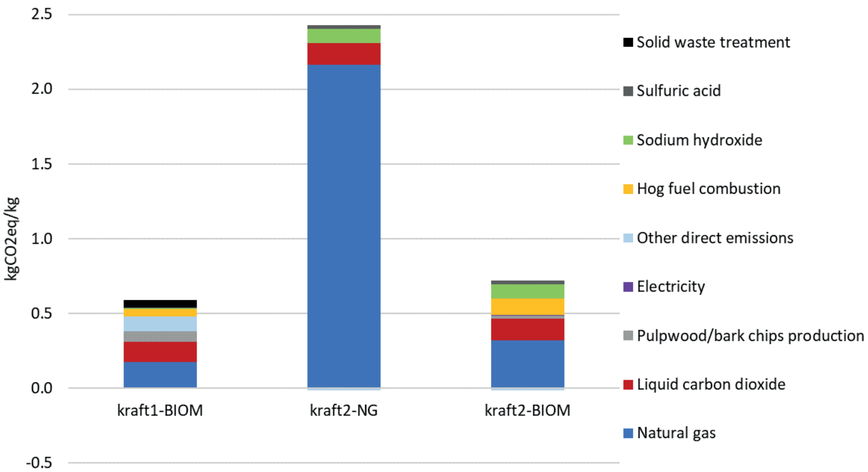

Figure 15. Cradle-to-gate climate change impact (excl. biogenic carbon removal) of 1 kg of kraft lignin. Biogenic carbon content expressed in terms of carbon dioxide equivalent: 2.4 kg CO_2eq/kg.

As highlighted by the authors of the LCAs from which the data were retrieved [100,103], the climate change impacts of kraft lignin production are mainly caused by the consumption of natural gas (mainly emissions from combustion i.e. 83% of climate change impact) and the production of liquid carbon dioxide. For kraft1-BIOM, other relevant impacts are direct emissions from the calcination reaction (carbon dioxide) and from the combustion of hog fuel (direct emissions of dinitrogen monoxide and biogenic methane and electricity to operate the furnace) and the impact (mainly emissions of biogenic methane) from the treatment of the solid waste generated by the pulp mill. For kraft2 (both -NG and -BIOM), the additional sodium hydroxide, which is necessary to make up for sodium losses caused by lignin extraction, is also important. In particular, beyond natural gas, the differences in impacts are also linked to the different allocation methods. For example, the direct emissions from the calcination reaction are allocated via economic value to lignin in kraft1-BIOM while they were not allocated to kraft 2 using physical causality allocation (since their amount does not change extracting or not lignin). Similarly, the impact of sodium hydroxide is much higher for kraft2 (both -NG and -BIOM) than for kraft1-BIOM since the additional sodium hydroxide that is necessary due to lignin extraction was entirely apportioned to lignin via physical causality allocation. Conversely, given the impossibility to subdivide the process in which it is used, this additional sodium hydroxide was also allocated to the pulp and only a negligible fraction to lignin (via economic allocation see section 4.2.2.1.1).

Figure 16 shows the cradle-to-gate environmental impact of kraft lignin expressed in terms of MKI score. The environmental impact per each impact category can be found in Appendix.

From Figure 16, it is possible to observe that the difference of impact between the kraft lignins assessed is much lower than in the case of climate change (see Figure 15). For all lignins, the reason is that the impacts of carbon dioxide and sulfuric acid production are much higher in terms of MKI scores than for climate change. The reasons (see Figure 16 and details for these categories in Figure 17) can be found in their marine acquatic ecotoxicity and human toxicity impacts. In particular, for liquid carbon dioxide production, the main causes are the materials (mainly copper and steel) used to construct the chemical plant and the production of monoethanolamine (MEA). Similarly, for sulfuric acid production, the main impact is caused by the materials consumed for the construction of the chemical plant.

An important contribution to the impact is these categories is also caused by hog fuel combustion for kraft1-BIOM and kraft2-BIOM. For human toxicity, the main source of impact of hog fuel combustion is made from direct emissions (mainy benzene to air). For marine acquatic ecotoxicity, the electricity for operation of the furnace and materials for the construction of the furnace are the main impacts. For kraft2-NG and kraft2-BIOM, a small negative impact is generated by the reduction of direct emissions from the combustion of black liquor (see Appendix for further information about the meaning of negative emissions using physical causality allocation). Regarding kraft2-NG lignin, the important impact of natural gas is mainly caused by its combustion (76%).

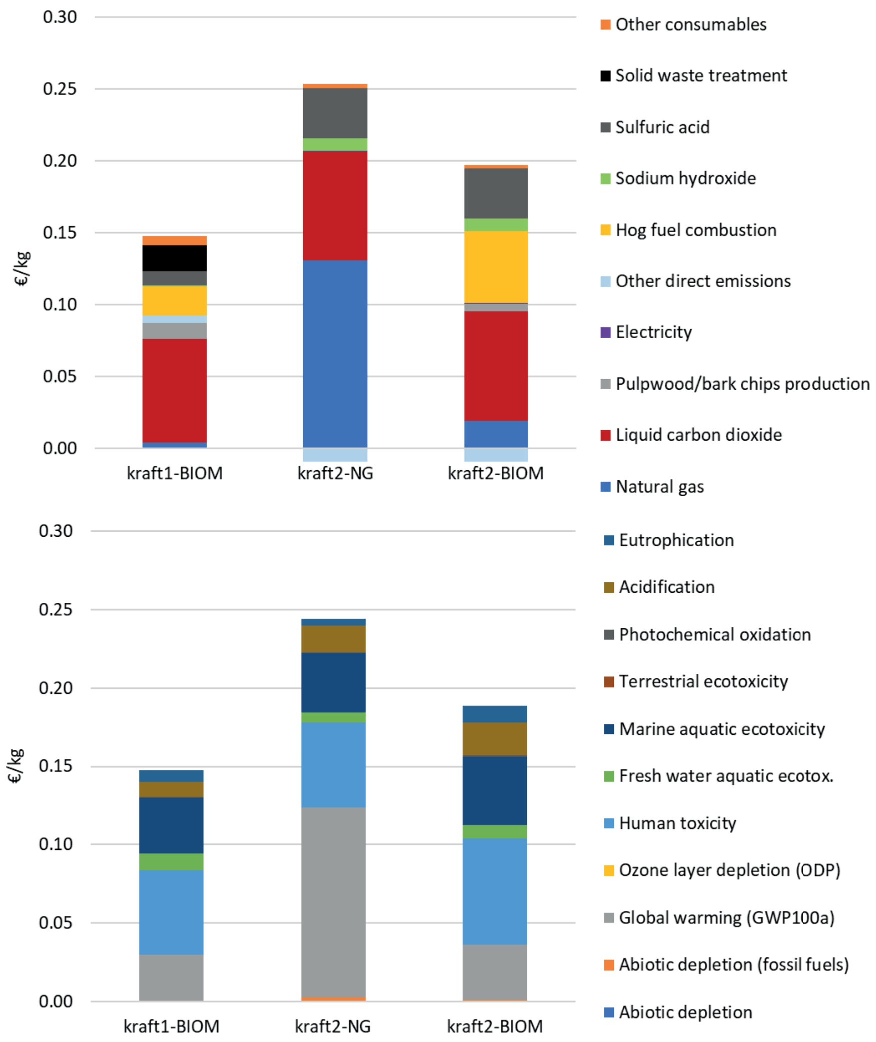

Figure 16. Cradle-to-gate MKI score of 1 kg of kraft lignin. Breakdown per process contribution (top bars) and impact category (bottom bars). In this figure, the MKI score is without biogenic carbon removal, which accounts for 0.12 €/kg.

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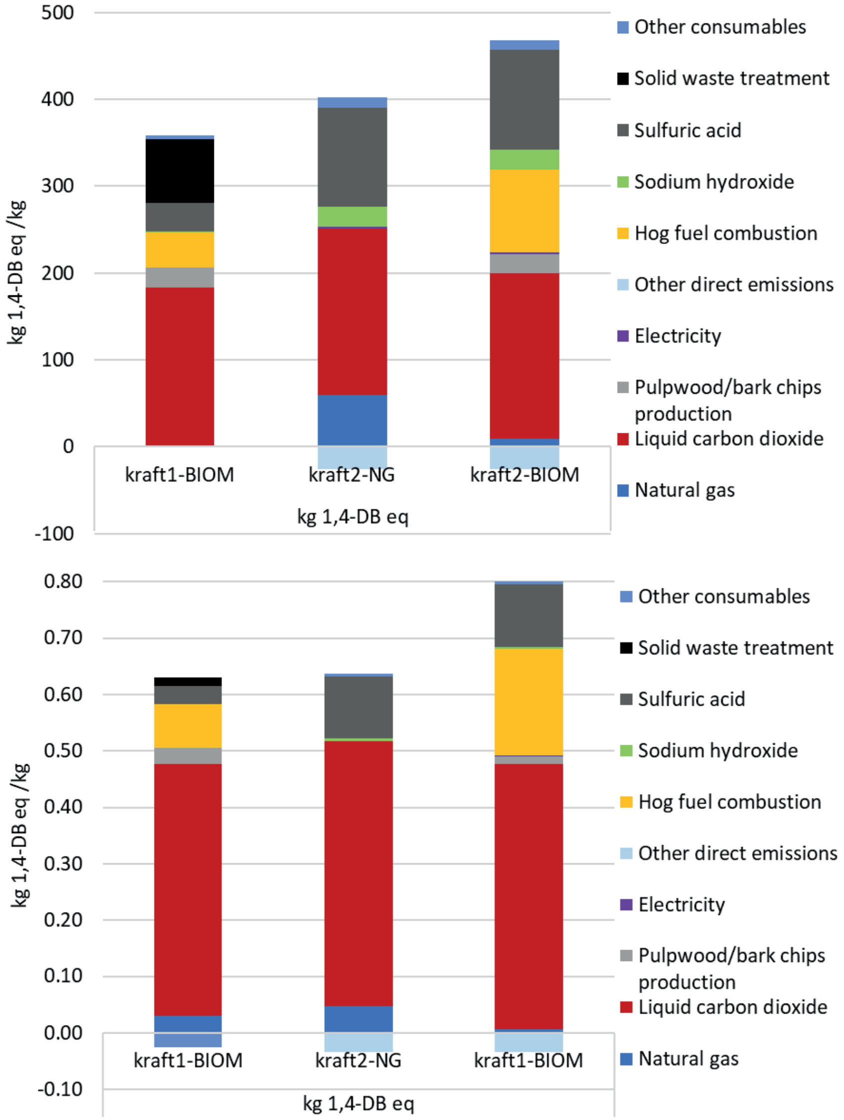

Figure 17. Cradle-to-gate impacts of 1 kg of kraft lignin in marine aquatic ecotoxicity (top bars) and human toxicity (bottom bars).

4.3.2 SMAs

Figure 18 shows the climate change impact and environmental profile (in MKI score) for SMAs divided into life cycle stages.

In all SMAs investigated, the extraction and processing of raw materials (A1) make the largest contribution to the total climate change impact and MKI score. What is immediately noticeable is that the environmental impact of the asphalt components (other than lignin) is smaller when lignin is used. The main reason is the replacement of a fraction of bitumen (and for a minor fraction by the biogenic carbon content of linseed oil, which is higher than the climate change impact generated by the production of linseed oil). For climate change, the impact of lignin is negative when hog fuel is used for steam production (kraft1-BIOM and kraft2-BIOM) since the biogenic carbon stored in lignin outperforms the climate change impacts caused by their production. Conversely, in the case of kraft2-NG lignin, the biogenic carbon content of lignin does not outperform the climate change impact caused by the production of lignin but it is almost entirely compensated (almost 95%). Regarding the climate change benefits of accounting module D, these benefits are important for kraft2-NG lignin and conventional SMAs since bitumen (and other materials for a minor fraction) is recycled. However, in the case of kraft1-BIOM lignin and kraft2-BIOM, module D represents a small positive impact since the climate change benefits of recycling bitumen and lignin (and other materials) are lower than the biogenic carbon removal that from the lignin in the assessed asphalt will be incorporated in the recycled asphalt in the following cycle. Overall, all lignin-based SMAs offer a reduction of climate change environmental impacts i.e. 78% using kraft1-BIOM lignin, 34% using kraft2-NG lignin and 75% for kraft2-BIOM.

In terms of MKI score, only SMAs using kratf2-NG shows higher MKI scores than conventional SMAs. The reason can be found in the high environmental impact of the production and combustion of natural gas for kraft1-NG (see section 4.3.1).

The MKI score of conventional asphalt was calculated as 8.5 €/t, which is 0.3 €/t lower than calculated in the Dutch LCA asphalt sector report [252] assuming a different composition for conventional SMA and supply chains. Taking this value 8.8 €/t as a reference would further improve the environmental competitiveness of lignin-based SMAs.

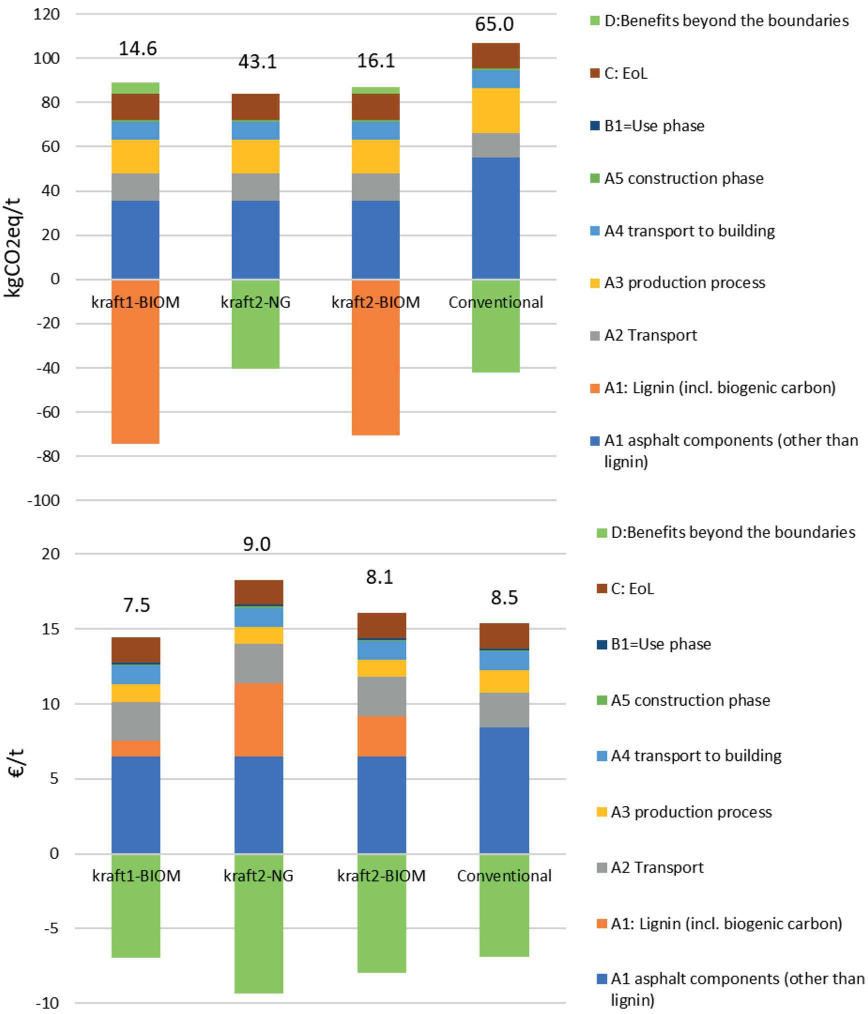

Figure 18. Cradle-to-grave climate change impact (top bars) and MKI score (bottom bars) of 1 t of SMA asphalt. Breakdown per life cycle stage.

4.3.3 ACs

Figure 19 shows the climate change and environmental profile (in MKI score) for ACs divided into life cycle stages.

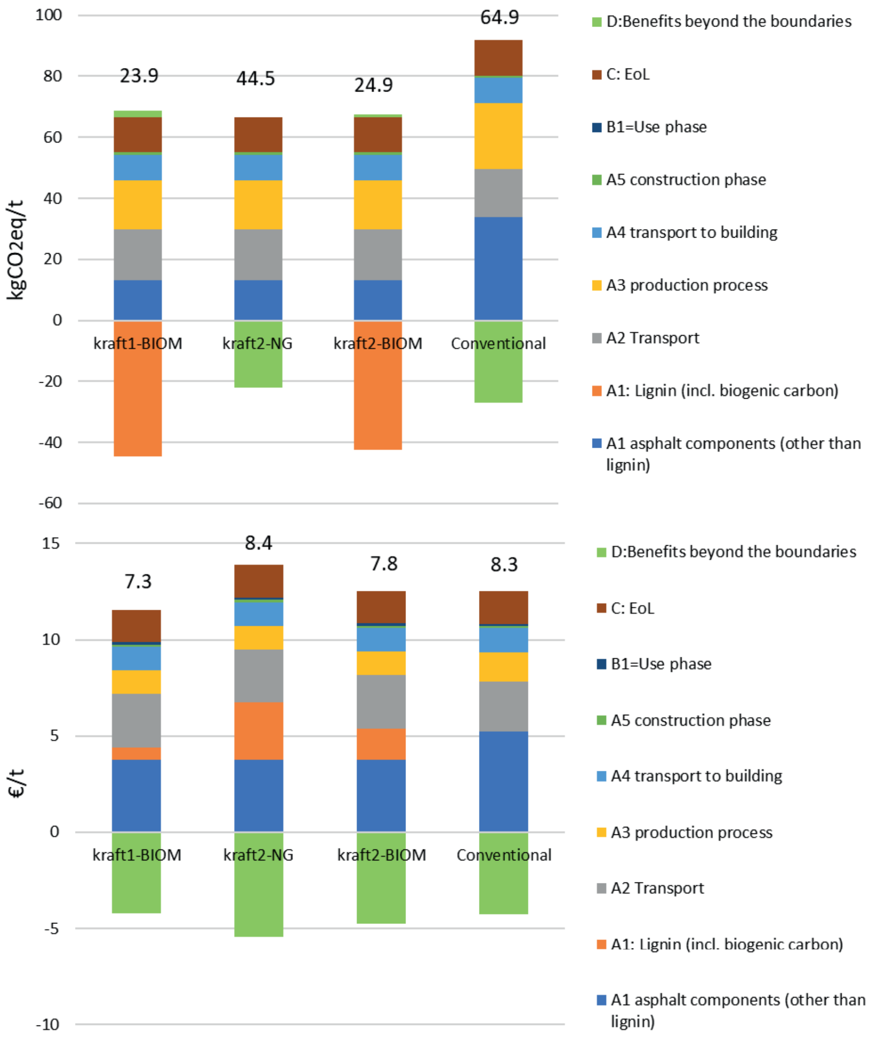

Figure 19. Cradle-to-grave climate change impact (top) and MKI score (bottom) of 1 t of AC asphalt. Breakdown per life cycle stage.

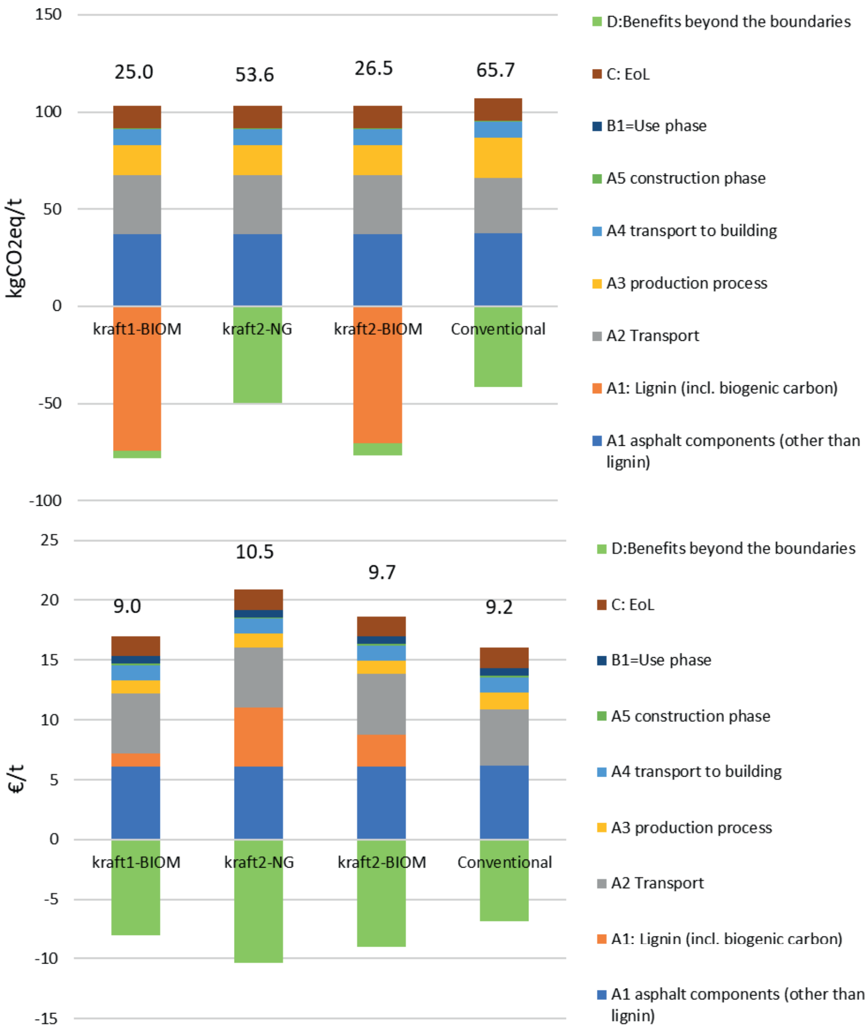

Figure 20. Cradle-to-grave climate change impact (top bars) and MKI score (bottom bars) of 1 t of ZOAB asphalt. Breakdown per life cycle stage.

Compared to SMAs, the extraction and processing of raw materials (A1) make a lower contribution to the total climate change impact and MKI score since 1) the amount of bitumen in ACs is lower than in SMAs, 2) ACs have a fraction of recycled component (i.e. burdens free) and 3) the amount of lignin is as much lower compared to SMAs (see section 4.2.2.1). Comparing lignin-based and conventional ACs, similar trends to SMAs are observed for climate change and MKI scores. The

MKI score of conventional AC was calculated as 8.3 €/t, which is in line with the range 7.4-8.8 €/t reported for several compositions of ACs in the Dutch LCA asphalt sector report [252]. However, considering this broader range instead of 8.3 €/t, it is not possible to claim what asphalt between lignin-based and conventional ACs is better except for the AC using kraft1-BIOM.

4.3.4 ZOABs

Figure 20 shows the climate change (top) and environmental profile (bottom, in MKI score) for ZOABs divided into life cycle stages.

In the case of ZOABs, lignin substituted weak filler instead of bitumen. This means that this type of asphalt has benefits related to the biogenic carbon content of lignin, which is beneficial for climate change (and consecutively for the MKI score). However, for the same amount in mass, the impact of weak filler is negligible compared to bitumen. For this reason, ZOABs show a good performance in terms of reductions of climate change impact (up to 60%) but MKI scores are in line or higher than conventional ZOAB. The MKI score of conventional ZOAB was calculated as 9.2 €/t, which is higher than 7.3-8.5 €/t reported in the Dutch LCA asphalt sector report [252]. So, taking this range, it can be concluded that lignin-based ZOABs substituting weak filler with lignin leads to a (slightly) higher MKI score than conventional ZOABs.

4.4 DISCUSSION

4.4.1 Multifunctionality

4.1.1.1 Lignin allocation

Previous literature showed that by-products like lignin are more affected than other products by the LCA issue of multifunctionality due to their lower physical/economic significance than the main product [57,128,254]. The price of lignin with the same quality specifications (mainly measured in terms of impurities) has been quite stable over time. However, lignin prices can vary significantly depending on its quality specifications (mainly impurities) [117,254].

In our calculations, an average market price of 535 €/t was assumed for kraft lignin. This price is in line with $600/t assumed by Dessbesell et al. (2018) but higher than

250$/t assumed by Abbati De Assis et al. (2018) and Culbertson et al. (2016). For a kraft lignin that meets the quality requirements to be used in asphalt, a reasonable price range is between 370 and 700 €/t [254]. This price range was used for a sensitivity analysis on the economic allocation applied to the pulp mill producing kraft1-BIOM. To assess the impact of allocation methods, mass allocation was also applied. The results of this sensitivity analysis are shown in Table 7. Based on the results shown in Table 7, climate change impact and MKI scores of asphalts using kraft1-BIOM lignin are affected by the allocation method respectively within the order of ±10% and ±4%. Since the price of pulp in €/t is higher than the lignin price (see section 4.2.2.1.1), mass allocation results in a higher environmental impact of lignin compared to economic allocation.

Table 7. Sensitivity analysis on the allocation method applied to kraft1-BIOM.

Sensitivity	**1 kg kraft1-BIOM**		**1 t SMA kraft1-BIOM**		**1 t AC kraft1-BIOM**		**1 t ZOAB kraft1-BIOM**	
	GWP 100 (kg CO_2eq)	**MKI (€)**	**GWP 100 (kg CO_2eq)**	**MKI (€)**	**GWP 100 (kg CO_2eq)**	**MKI (€)**	**GWP 100 (kg CO_2eq)**	**MKI (€)**
Min price	0.47	0.13	13.0	7.2	22.7	7.1	23.4	8.7
Baseline (avg. price)	**0.58**	**0.15**	**14.6**	**7.5**	**23.9**	**7.3**	**25.0**	**9.0**
Max price	0.67	0.17	16.1	7.7	25.0	7.5	26.6	9.3
Mass allocation	0.71	0.17	16.8	7.8	25.5	7.6	27.2	9.4

4.4.1.2 Bitumen allocation and dataset

In the baseline calculations, the prescribed process map of the NL-PCR asphalt i.e. ESU NL-PCR bitumen [257] was used. This dataset was based on data from Energie-Stoffe-Umwelt (ESU) consultants, which uses energy allocation [252]. However, bitumen, like lignin for biorefineries, is a low-economic/physical significance product of oil refineries and therefore, its environmental impact is affected strongly by allocation [222,279]. Moreover, applying energy allocation to bitumen seems in contrast with what is recommended by ISO 14049:2012 [187], which is the ISO technical report illustrating how to apply ISO 14044:2006. In such a report, ISO makes an example of bitumen production for which economic allocation is used since no physical parameter (“mass, feedstock energy, thermal conductivity, viscosity, specific mass, etc.”) reflects the underlying physical relationship between bitumen and the other co-products.

In literature, there are some alternatives available that LCA practitioners in the construction sector often use. For example, the previous version of the Dutch LCA asphalt sector report [280] was using a dataset for bitumen (SBK bitumen) that was based on data from Eurobitume and economic allocation was applied [252]. Ecoinvent is also often used to retrieve datasets for petroleum products. For bitumen, two possible datasets are *Bitumen, at refinery/RER* and *Bitumen adhesive compound, hot {RER}| production | Cut-off*. Both these datasets use energy allocation [252].

Alternatively, a practitioner could follow a simplified approach and imagine the oil refinery as a black box. In literature, this approach is also referred to as allocation at the aggregate refinery level [222,279,281]. By using this method, the whole upstream and oil refinery emissions are allocated to oil products using allocation shares based on the production volumes (mass allocation), or economic value (economic allocation) or their energy value (energy allocation).

Typical efficiencies (kg of products/kg of petroleum oil input) of oil refineries are between 90 and 94% [281]. This means that for 1 kg of petroleum input (from ecoinvent, *Petroleum {GLO}| market for | Cut-off*), 0.92 kg of petroleum products are produced on average (among them gasoline, diesel and bitumen). The remaining 0.08 kg of petroleum input is burned for refining such products (from ecoinvent, *Refinery gas, burned in furnace/kg/RER*). Based on this simplified modeling, the environmental impact generated by burning fuels in the refinery and producing the upstream crude oil can be allocated to the oil refinery products using the abovementioned allocation shares. In Europe, bitumen represents about 3% of the economic share of oil refineries [282]. Mass and energy allocation shares for bitumen were assumed as 4.8% and 5.0% using petroleum coke as a proxy [222].

Figure 21 shows the climate change and MKI scores of producing 1 kg of bitumen. From Figure 21, it is possible to notice that 1) the dataset for bitumen from the NL-ESU PCR [257] is the one with the highest impact compared to all other alternatives compared, 2) economic allocation is the method that provides the lowest environmental impact for bitumen (in line with the effect of allocation on lignin shown by Table 7) while energy/mass allocation the highest. Moreover, the use of a different dataset and/or a different allocation method can influence the climate change impact up to a factor of 2-3 and the MKI score up to a factor of 4. It also emerges that the difference of data and not only of allocation play an important role in the difference of environmental impacts between the current and previous bitumen datased used by the Dutch LCA asphalt sector report. Figure 22 shows the effect on the comparative analysis between the environmental performances of lignin-based and conventional asphalts taking the lowest environmental impact for bitumen. Consequently, the environmental impacts of both lignin-based and conventional asphalts would be lower since both contain bitumen. On the other hand, assuming a lower impact for bitumen, while the trends observed for climate change in the baseline analysis are conserved, the MKI scores of SMAs and ACs using kraft2-BIOM lignin becomes worse than the ones of the conventional asphalts while they were better in the baseline calculations. In particular, the impact of ACs is the least affected by the allocation applied to bitumen since its bitumen content was the lowest compared to SMAs and ZOABs.

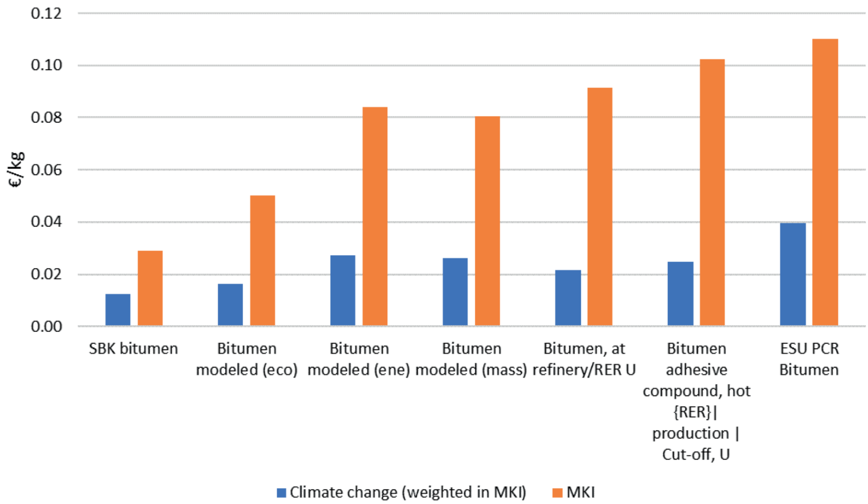

Figure 21. Climate change impact (weighted in terms of MKI score) and MKI score of 1 kg of bitumen using various data and allocation methods. Eco=economic; ene= energy.

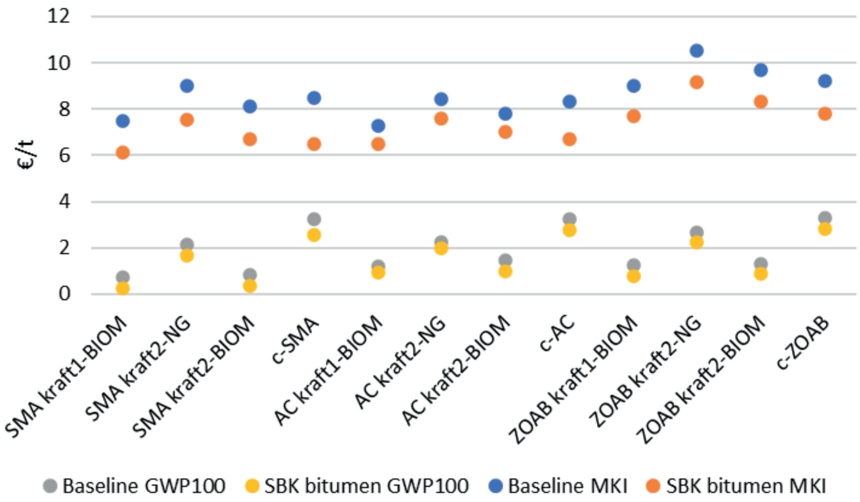

Figure 22. Climate change impact (weighted in terms of MKI score) and MKI score of 1 t of top layer asphalt using a different dataset for bitumen (SBK bitumen). C=conventional

4.4.2 Change of functional unit (to 1 m²) and product system

In this LCA, the functional unit was defined as 1 tonne (t) of top layer asphalt. In the LCA literature, it is acknowledged that different functional units could lead to different results for the same product system [50,254,283]. Besides a mass-based functional unit, a common functional unit for asphalts is a surface-based functional unit [249,257,284]. Taking a functional unit of 1 m^2, it becomes a reasonable option to include in the product system also the middle and base layer asphalts that are under the top layer asphalt. A sensitivity analysis was conducted taking a functional unit of 1 m^2 of asphalt made of three layers. The compositions, lifetimes, densities and thicknesses of each layer can be found in Appendix. Depending on the type of asphalt, the mass of asphalt made of three layers under a surface of 1 m^2 has a mass between 0.39 and 0.40 t.

It is necessary to remark what follows to interpret the results of this sensitivity analysis correctly. Sub-layers have important recycled percentages. For lignin-based asphalts, the recycled part was tested with lignin-based recovered asphalt. The recycled material is free of the environmental burden in module A1 (that for lignin was accounted as environmental impact at the net of biogenic carbon

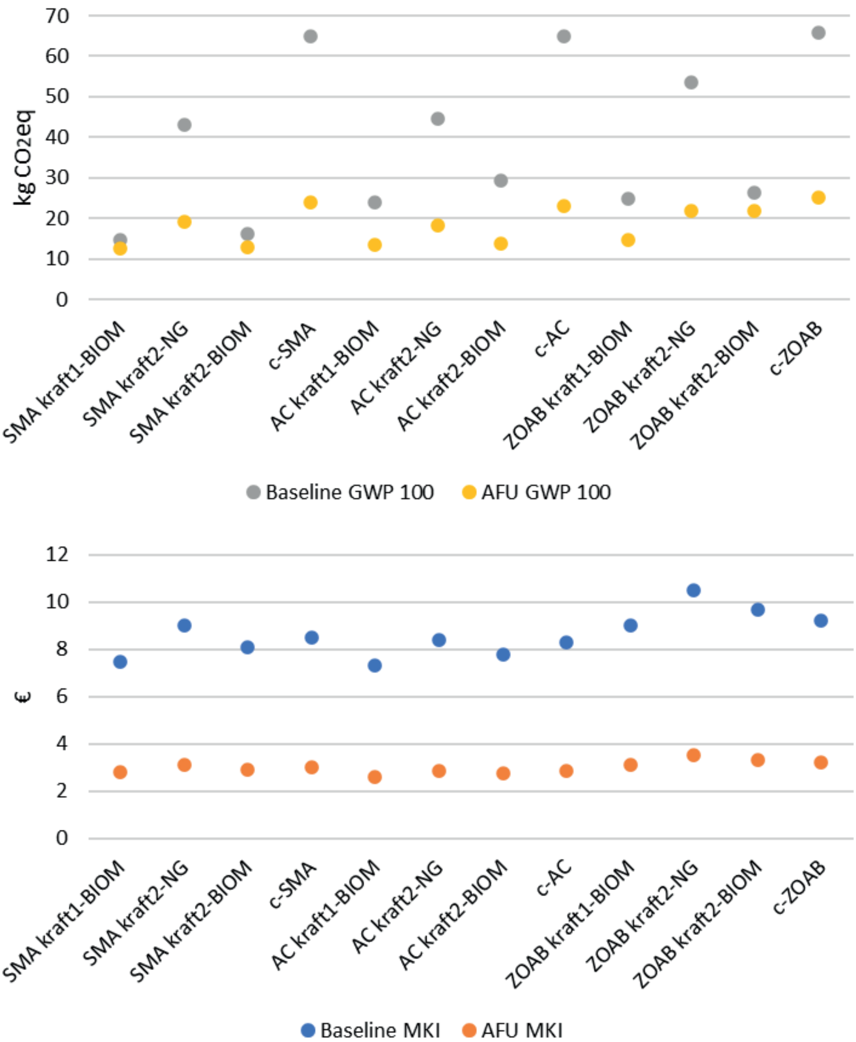

Figure 23. Cradle-to-grave climate change impact (top graph) and MKI score (bottom graph) of baseline (1 t of top layer asphalt) versus alternative functional unit (AFU) i.e. 1 m² of asphalt made of three layers.

removal). Accordingly, asphalt mixtures that already contain secondary material (recycled asphalt) do not include this percentage as environmental benefits in module D. Conversely, they are charged with a fraction of environmental burdens in module D as a result of the 29% loss of these secondary raw materials leaving the product system that cannot be used in the following cycle [257]. These charges are net of the quality losses that these materials would have had in the following cycle in the sub-layers where they would have been utilized. The distance for the asphalt to be recycled to the transportation to the construction site was assumed as 50 km [252]. Leaching does not take place in sub-layers since they are installed above groundwater level and do not come into contact with precipitation [252]. Virgin lignin used in the middle and bottom layer takes a 30% credit on the biogenic carbon that will be stored in the following cycles and was stored by this product system for 30 years.

Figure 23 shows the results of changing functional unit and product system. Changing functional unit does not lead to other trends. On the other hand, 1) the total impact is reduced in terms of surface since 1 m^2 contains less than 1 t of asphalt and 2) the differences between lignin-based and conventional asphalts are reduced since middle and base layers are 50% made of recycled asphalt (free of burden) and middle layer does not have any virgin lignin and base layer only 1%.

4.5. CONCLUSIONS

This study contributes to the existing research in the field of environmental sustainability of bio-based asphalt products. Various asphalts (stone mastic asphalt, asphalt concrete and porous asphalt) using kraft lignin were assessed using LCA methodology and compared with their conventional counterparts. The major LCA methodological choices were mostly based on the Dutch Product Category Rules (NL-PCRs) for asphalts. The data for kraft lignin were retrieved from two studies from the literature. The effects of allocation methods and defining a different functional unit were broadly discussed as well as the effect of using a different fuel (natural gas or hog fuel) for steam production.

The results of the LCA revealed that using lignin in asphalts could reduce the climate change impact of top-layer asphalt products over their life cycle substantially (order between 30% and 75% depending on the type of asphalt considered). The highest reductions are achieved if the current use of lignin (contained in the black liquor) for process energy is replaced with low-grade biomass fuels, for example hog fuel. Using natural gas to replace the heating value of lignin for energy production leads to a significant/strong reduction in the climate change benefits.

Considering also other impact categories, on a weighted basis expressed in terms of economic cost (MKI scores), the advantages of using hog fuel instead of natural gas in the pulp mill on the impact of the asphalts are in part mitigated since other categories are less affected by such shift of burdens. Nonetheless, given the important advantages in the climate change category, using hog fuel instead of natural gas also showed advantages in total MKI scores.

For this reason, stone mastic asphalt containing lignin produced from a pulp mill where hog fuel is used to replace the part of the energy that is no longer produced from burning black liquor showed environmental advantages also in terms of MKI scores compared to their conventional counterpart. Conversely, if natural gas is used to deliver the required process heat, the MKI score of lignin-based stone mastic asphalts was worse than the conventional counterpart. In other words, the climate change impact (and, to a lesser extent, overall environmental impact) is largely determined by how additional process heat is produced in the pulp mill. This conclusion is in line with previous research on other lignin-based applications [106,119,254].

For asphalt concretes, it was not possible to identify an "environmental winner" between lignin-based and conventional asphalts since the differences in impact were in line with the uncertainties. For porous asphalts with lignin used only as weak filler, lignin-based asphalts perform worse than conventional asphalts. The reason is that the impact of limestone filler is minor compared to kraft lignin. To maximize environmental benefits, the maximum substitution of bitumen with lignin should be targeted, whereas the substitute of weak filler should be limited.

Regarding the allocation method, using mass allocation instead of economic allocation would lead to much higher environmental impacts for both lignin and bitumen. Using a physical causality allocation avoids that impacts that are typical of pulp production and are unaffected by extracting or not lignin are allocated also to lignin. On the other hand, once the credits for their recycled are accounted for, the effect of the change in allocation method becomes smaller.

Regarding the sensitivity analysis on the functional unit and product system considered, since middle and base layers have a higher percentage of recycled content and a lower amount of bitumen/lignin, once the mix of the three layers is considered as the product system instead of the top layer only, the MKI score difference between the two asphalt options becomes minor. Conversely, looking at climate change only, the difference is still significant given the importance of the biogenic carbon storage for this category.

4.A Appendix

Inventory data

Table 4A1. Life cycle inventory data for the production of 1 t of kraft1-BIOM lignin. Based on Culbertson et al. (2016)

Flow	Data	Unit	Process (ecoinvent)	Multifunctionality
Sulfuric acid for lignin extraction	0.07	t	Sulfuric acid {RER}| market for sulfuric acid | Cut-off	Subdivison
Natural gas for lignin extraction	1034	MJ	Natural gas, burned in industrial furnace >100kW/RER U	Subdivison
Liquid carbon dioxide for lignin extraction	0.28	t	Based on (Young et al., 2019), 3.56 MJ of heat are necessary to separate the carbon dioxide using MEA from waste gas (burdens free) from ammonia production. For heat from natural gas, the process Natural gas, burned in industrial furnace >100kW/RER was used. The amount of MEA, electricity consumption, losses of MEA to air and water flows of carbon dioxide separation using MEA were retrieved from ecoinvent 3.6 dataset Carbon dioxide, {RER}| production | Cut-off.	Subdivison
Sodium hydroxide	0.10	t	Sodium hydroxide, chlor-alkali production mix, at plant/RER	Economic allocation
Lime	0.41	t	Lime {RER}| market for lime | Cut-off	Economic allocation
Natural gas	21029	MJ	Natural gas, burned in industrial furnace >100kW/RER U	Economic allocation
Softwood	49.9	m^3	Pulpwood, softwood, measured as solid wood under bark {Europe without Switzerland}| market for | Cut-off	Economic allocation
Combustion of hog fuel	152040	MJ	Retrieved from Heat, central or small-scale, other than natural gas {CH}| heat production, softwood chips from forest, at furnace 50kW, state-of-the-art 2014 | Cut-off. Background data for the electricity input for the operation of the furnace updated using 2030 EU reference scenario for the Netherlands (Carpos et al., 2016)	Economic allocation
Waste to landfill	1.07	t	Municipal solid waste {CH}| treatment of, sanitary landfill | Cut-off	Economic allocation
Sulfur dioxide direct emissions	27	kg	Sulfur dioxide, NL (direct emissions)	Economic allocation
Fossil carbon dioxide direct emissions	1402	kg	Carbon dioxide, fossil (direct emissions)	Economic allocation

Table 4A.2. Life cycle inventory data calculated using physical causality allocation based on average changes for the production of 1 t of "kraft2-NG" and "kraft2-BIOM" [103]. When the name of the flow does not specify the name of the two scenarios, that flow is the same for the two scenarios.

Flow	**Data**	**Unit**	**Process (ecoinvent)**
Sodium hydroxide	0.107	t	Sodium hydroxide, chlor-alkali production mix, at plant/RER
Sulfuric acid	0.23	t	Sulfuric acid {RER}| market for sulfuric acid | APOS, U
Lime	0.23	t	Lime {RER}| market for lime | APOS, U
Natural gas (kraft2-NG)	31500	MJ	Natural gas, burned in industrial furnace >100kW/RER U
Natural gas (kraft2-BIOM)	4700	MJ	Natural gas, burned in industrial furnace >100kW/RER U
Hog fuel production (kraft2-BIOM)	1.87	t	Bark chips, wet, measured as dry mass {CH}| bark chips production, softwood, at sawmill | Cut-off, U
Combustion of hog fuel	26800	MJ	Retrieved from Heat, central or small-scale, other than natural gas {CH}| heat production, softwood chips from forest, at furnace 50kW, state-of-the-art 2014 | Cut-off. Background data for the electricity input for the operation of the furnace updated using 2030 EU reference scenario for the Netherlands (Carpos et al., 2016).
Liquid carbon dioxide	0.3	t	Carbon dioxide, liquid {RER}| market for | APOS, U
Water	4.85	t	Tap water {RER}| market group for | APOS, U
Electricity	10	kWh	2030 EU reference scenario for the Netherlands (Carpos et al., 2016)
Reduction of combustion emissions from black liquor	-1	t	Emissions from avoided combustion of black liquor (Corona et al., 2018)

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Environmental impacts

Table 4A.3. Cradle-to-grave environmental impacts of 1 t of top layer asphalt.

Impact category	**Unit**	**SMA kraft1-BIOM**	**SMA kraft2-NG**	**kraft2-BIOM**	**c-SMA**	**AC kraft1-BIOM**
Abiotic depletion	kg Sb eq	3.92E-01	3.92E-01	3.92E-01	5.72E-01	2.25E-01
Abiotic depletion (fossil fuels)	MJ	1.58E+03	2.07E+03	1.64E+03	1.96E+03	1.24E+03
Global warming (GWP100a)	kg CO2 eq	1.46E+01	4.31E+01	1.61E+01	6.50E+01	2.39E+01
Ozone layer depletion (ODP)	kg CFC-11 eq	8.52E-06	1.32E-05	9.27E-06	8.71E-06	8.56E-06
Human toxicity	kg 1,4-DB eq	2.53E+01	2.53E+01	2.78E+01	1.69E+01	2.24E+01
Fresh water aquatic ecotox.	kg 1,4-DB eq	1.28E+01	1.07E+01	1.19E+01	6.36E+00	1.34E+01
Marine aquatic ecotoxicity	kg 1,4-DB eq	1.92E+04	1.94E+04	2.05E+04	1.46E+04	1.81E+04
Terrestrial ecotoxicity	kg 1,4-DB eq	6.54E-01	6.55E-01	6.75E-01	1.53E-01	1.29E+00
Photochemical oxidation	kg C2H4 eq	5.27E-02	5.08E-02	5.63E-02	6.69E-02	3.30E-02
Acidification	kg SO2 eq	3.21E-01	3.51E-01	3.64E-01	3.20E-01	2.64E-01
Eutrophication	kg PO4--- eq	6.03E-02	5.44E-02	6.54E-02	4.48E-02	6.49E-02

It continues

AC kraft2-NG	**AC kraft2-BIOM**	**c-AC**	**ZOAB kraft1-BIOM**	**ZOAB kraft2-NG**	**ZOAB kraft2-BIOM**	**c-ZOAB**
2.25E-01	2.25E-01	4.28E-01	3.79E-01	3.79E-01	3.79E-01	3.79E-01
1.59E+03	1.28E+03	1.70E+03	1.64E+03	2.13E+03	1.70E+03	1.62E+03
4.45E+01	2.49E+01	6.49E+01	2.50E+01	5.36E+01	2.65E+01	6.57E+01
1.19E-05	9.10E-06	9.02E-06	9.51E-06	1.42E-05	1.03E-05	9.28E-06
2.24E+01	2.42E+01	1.64E+01	2.89E+01	2.89E+01	3.14E+01	1.94E+01
1.19E+01	1.27E+01	6.83E+00	1.58E+01	1.36E+01	1.49E+01	1.03E+01
1.83E+04	1.90E+04	1.49E+04	2.48E+04	2.50E+04	2.61E+04	1.92E+04
1.29E+00	1.31E+00	1.29E-01	1.53E-01	1.54E-01	1.74E-01	1.27E-01
3.16E-02	3.56E-02	5.10E-02	5.11E-02	4.92E-02	5.47E-02	4.62E-02
2.86E-01	2.96E-01	2.72E-01	3.26E-01	3.55E-01	3.69E-01	2.86E-01
6.07E-02	6.86E-02	4.80E-02	6.84E-02	6.25E-02	7.35E-02	5.43E-02

Compositions of asphalt mixtures

Table 4A.4 Compositions, densities, thicknesses assumed for SMA (bind and base layer assumed as ACs).

Composition	**Lignin based asphalt**			**Bitumen based reference**			**Unit**
	Top layer	**Bind layer**	**Base layer**	**Top Layer**	**Bind layer**	**Base layer**	
Cellulosefiber	3			3			kg/tonne
Recycled content		500	500		500	500	kg/tonne
Bitumen 40/60	44.5		5.5		18	18	kg/tonne
Bitumen 70/100		18		65			kg/tonne
Crusher sand	88			75			kg/tonne
Natural sand	88	200	187.5	75	200	200	kg/tonne
Crushed stone (Morene)		267	280		267	267	kg/tonne
Crushed stone (Porfier)	700			710			kg/tonne
Weak filler	35	15	15	72	15	15	kg/tonne
Lignin	40		10				kg/tonne
Linseed oil	1.5		2				kg/tonne
Asphalt density	2250	2375	2375	2255	2375	2375	kg/m^3
Asphalt lifetime	15	30	30	15	30	30	Years
Asphalt thickness	3.5	5	8	3.5	5	8	cm

Table 4A.5. Compositions, densities, thicknesses assumed for ACs

Composition	**Lignin based asphalt**			**Bitumen based reference**			**Unit**
	Top layer	**Bind layer**	**Base layer**	**Top Layer**	**Bind layer**	**Base layer**	
Recycled content	288	500	500	300	500	500	kg/tonne
Bitumen 40/60	21.1		5.5	40	18	18	kg/tonne
Bitumen 70/100		18					kg/tonne
Crusher sand	154			171			kg/tonne
Natural sand	76	200	187.5	57	200	200	kg/tonne

Composition	Lignin based asphalt			Bitumen based reference			Unit
	Top layer	Bind layer	Base layer	Top Layer	Bind layer	Base layer	
Crushed stone (Morene)	411	267	280	410	267	267	kg/tonne
Weak filler	23	15	15	22	15	15	kg/tonne
Lignin	24		10				kg/tonne
Linseed oil	2.9		2				kg/tonne
Asphalt density	2335	2375	2375	2340	2375	2375	kg/m^3
Asphalt lifetime	15	30	30	15	30	30	Years
Asphalt thickness	3.5	5	8	3.5	5	8	cm

Table 4A.6. Compositions, densities, thicknesses assumed for ZOABs

Composition	Lignin based asphalt			Bitumen based reference			Unit
	Top layer	Bind layer	Base layer	Top Layer	Bind layer	Base layer	
Recycled content		500	500		500	500	kg/tonne
Bitumen 40/60			5.5		18	18	kg/tonne
Bitumen 70/100	43	18		43			kg/tonne
Crusher sand	106			106			kg/tonne
Natural sand		200	187.5		200	200	kg/tonne
Crushed stone (Morene)	811	267	280	811	267	267	kg/tonne
Weak filler		15	15	40	15	15	kg/tonne
Lignin	40		10				kg/tonne
Linseed oil			2				kg/tonne
Asphalt density	1950	2375	2375	1950	2375	2375	kg/m^3
Asphalt lifetime	12	30	30	12	30	30	Years
Asphalt thickness	5	5	8	5	5	8	cm

Comment: about negative emissions using physical causality allocation versus substitution

The second level of ISO 14044:2006 multifunctionality hierarchy recommends an allocation underlying the physical relationships that exist between the products. Physical relationships are described as those relationships that are at the basis of how inputs and outputs change depending on changes in functions delivered. This type of allocation is referred to in the literature as physical causality allocation [82,216,285]. As described by ISO 14044:2006, ISO 14049:2012 and their previous versions, this type of allocation implies modeling some changes in the system under investigation. In this study, an average change was modeled to allocate the impact in one of the two pulp mills (i.e. the one that allowed such type of modeling). Conceptually, emissions based on mathematical modeling of physical causality represent the additional amount of emissions that the system would emit to produce one unit (marginal, incremental or average) more of one co-function. In the case of linear relationships (applicable only to marginal or incremental changes), this amount would also correspond to the emissions that the investigated system would avoid if it could prevent the production of one unit of that co-function. The concepts “marginal”, “avoided” and “change” should not make readers link this type of allocation with system expansion by substitution in consequential modeling. The principle of additivity of the emissions typical of attributional modeling is respected by physical causality allocation as for any other allocation, i.e. the sum of the emissions allocated to each product with a physical causality allocation corresponds to the sum of the emissions of the system. This is not the case for substitution used as a system expansion method [286].

Some authors use substitution in attributional modeling to calculate an allocation factor (as a form of “proxy-based disaggregation”) instead of as a system expansion method [51]. For example, EU footprint guides [85,165] suggest that substitution can also potentially be used as an allocation method by “other relationships” (ISO third level). Accordingly, Hermansson et al. (2020) applied two substitution-based allocation methods to kraft lignin. This type of allocation often leads to negative impacts, which means this method has been erroneously used and is wrongly representing the physical causalities of that system [286]. This often happens when the wrong product is chosen as the main product of the multifunctional system (Cherubini et al., 2011) and the credit of the subtracted functions (potentially avoided) is higher than the total impact (see Figure 4A.1). For example, in the case of lignin, this happens if the impact of lignin is calculated as the impact of the total system subtracting the impact of an alternative technique producing a product fulfilling the same functions of pulp [55].

The generation of negative impacts by physical causality allocation has a different meaning [222,287] instead. For example, in the allocation model used in the European legislation for biofuels, negative refining emissions are obtained for heavy fuel oil [222]. The excess of heavy fuel oil is produced due to the need for refining more remunerative products (e.g. gasoline) and not because demanded by the market. So, the extra hydrogen needed by the refinery to desulphurize the extra heavy fuel is caused by gasoline and not by heavy fuel oil. Therefore, the most valuable products take an additional impact because they cause the production of this surplus of low-value products and such additional hydrogen consumption (causality). This additional impact gets a sign minus for the low-value product based on the attributional additivity of emissions calculated with mathematical modeling.

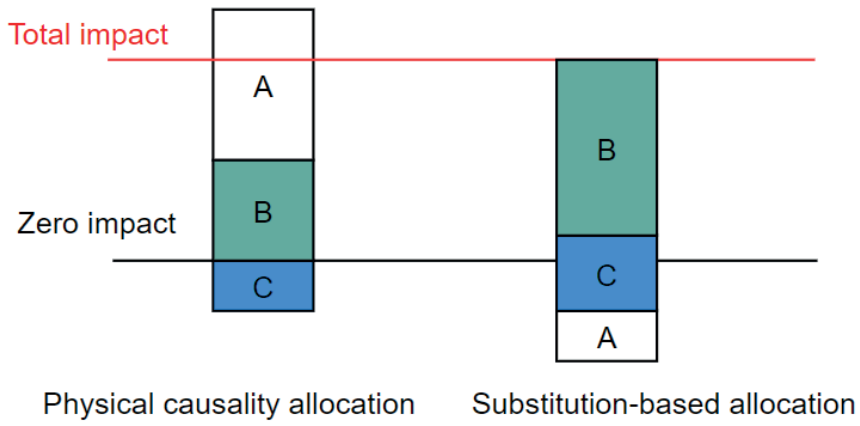

Figure 4A1. Negative emissions by physical causality allocation vs negative emissions by (direct) substitution-allocation. The red line (Total impact) represents the total impact of the system producing A, B and C that need to be allocated. A is the main product, B is a co-product, and C is the by-product with less physical/economical significance. By physical causality allocation, product C may get negative emissions. By substitution-based allocation, product A may get negative emissions after substitution-based allocation. The substitution method does never agree with physical causality impacts in case of negative impact i.e. in such cases, it fails in representing the causality of the system.

CHAPTER 5

Combining biomass gasification and Solid Oxid Fuel Cell for heat and power generation: an early-stage Life Cycle Assessment

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ABSTRACT

Biomass-fueled combined heat and power systems (CHPs) can potentially offer environmental benefits compared to conventional separate production technologies. This study presents the first environmental life cycle assessment (LCA) of a novel high-efficiency bio-based power (HBP) technology, which combines biomass gasification with a 199 kW solid oxide fuel cell (SOFC) to produce heat and electricity. The aim is to identify the main sources of environmental impacts and to assess the potential environmental performance compared to benchmark technologies. Also, the use of various biomass fuels and alternative allocation methods were scrutinized.

The LCA results reveal that most of the environmental impacts of the energy supplied with the HBP technology are caused by the production of the biomass fuel. This contribution is higher for pelletized than for chipped biomass. Overall, HBP technology shows better environmental performance than heat from natural gas and electricity from the European grid. When comparing the HBP technology with the biomass-fueled ORC technology, the former offers significant benefits in terms of particulate matter (about 22 times lower), photochemical ozone formation (11 times lower), acidification (8 times lower) and terrestrial eutrophication (26 times lower). The environmental performance was not affected by the allocation parameter (exergy or economic) used. However, the tested substitution approaches showed to be inadequate to model the environmental impacts of CHP plants under the investigated context and goal.

5.1 INTRODUCTION

Compared to separate production of heat and electricity from fossil fuels, combined heat and power systems (CHPs) can potentially allow for significant reductions of climate change impact [288,289]. In Europe, coupling heat and electricity generation from renewable sources is also one of the most cost-effective decarbonization strategies [290–292]. In particular, solid biomass has attracted increasing interest by policymakers and investors especially due to the high availability of local biomass from forests and wood processing industries in some regions [293]. The environmental performance of biomass-fueled CHPs depends not only on the type of technology but also on the type of biomass, its supply chain and the environmental impact categories in focus [294,295].

Mature CHP technologies using solid biomass as fuels have often shown restricted fuel flexibility, limited electric efficiencies and high particulate matter emissions [296]. To overcome these three limitations, a novel technology has been developed during the H2020 HiEff-BioPower[5] project [297]. This novel technology (see Figure 24) is based on a fixed-bed updraft gasifier coupled with a novel primary gas treatment zone, a novel gas cleaning unit (GCU) and a solid oxide fuel cell (SOFC). Its current technological readiness level is between 4 and 5 (based on the definition adopted by the European Commission [298]). The biomass fuel is converted into product gas in the gasifier. Syngas derived from biomass (e.g. wood chips) contains HCl, H_2S and tars [299] making it not suitable for direct utilisation in fuel cells [300], which require purified gaseous fuels. Therefore, the syngas from gasifier is first pre-treated in a primary gas treatment unit (first tar reforming step) and then purified in the GCU. The GCU is one of the key innovations of this technology. It combines the use of ceramic filter candles and sorbents [297]. Syngas cleaning is processed in five steps: primary tar reforming, high-temperature particle filtration, HCl sorption (after cooling the product gas), H_2S removal by sorbents and tar reforming (after re-heating). After re-heating the product gas is then fed into the SOFC unit to generate electricity. The off-gases from the SOFC unit are then burnt in a catalytic afterburner to recover heat. Most of biomass CHPs are suited for medium and large-scale plants (1-100 MWel). The HBP is available also in small size (about 200 kW of electricity output)[296]. For this size, among biomass technologies, one of the main competitors is the organic Rankine cycle (ORC) technology [301].

[5] http://www.hieff-biopower.eu/home/

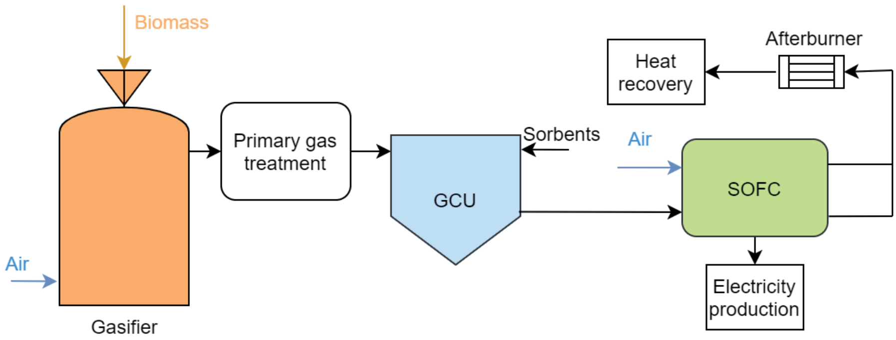

Figure 24. Concept of HBP technology. GCU=Gas cleaning unit. SOFC=Solid oxide fuel cell.

At this stage of the Hieff-BioPower project, the assessment of life-cycle potential environmental impacts of the current design configuration can help the technical development of the HBP, by minimizing the impacts already at an early stage of development. In particular, the literature reports a few Life cycle assessments (LCAs) of heat and power from SOFC-based CHPs, and several ones of CHPs involving biomass gasification processes but no one on their combination[6]. These studies provided the following main findings: (1) the investigated CHPs present lower impact in terms of climate change compared to conventional technologies [289,302] and (2) the biomass fuel production has the highest contribution to total life cycle impacts [303,304]. These studies also highlighted several methodological uncertainties of LCAs that can lead to significantly different results. Such uncertainties are mainly linked to the multifunctional nature of the CHPs. A CHP is a system producing two products, heat and electricity. Depending on the goal of the LCA, it may be necessary to apportion the overall impact of the system to each of the co-products. Finding the right criterion for the allocation of impacts to each co-product is generally understood as a multifunctionality problem [38]. When a multifunctionality issue is encountered, the practitioner has to properly select the functional units and allocation methods [305,306]. The selected criterion could affect the outcome of the LCA significantly and, for this reason, this selection is broadly discussed in the literature [51,56].

The environmental LCA presented in this study has a twofold aim: 1) to identify the main sources of the environmental impact of this new technology and 2) to

[6] From Scopus database (October 2019): TITLE-ABS-KEY (lca AND chp AND (gasification OR gasifier) AND sofc) 0 documents, TITLE-ABS-KEY (lca AND chp AND sofc) 9 documents, TITLE-ABS-KEY (lca AND chp AND gasification OR gasifier) 21 documents

assess its ecological competitiveness compared to separate production of heat and electricity, and to one of its main competitor, i.e. Organic Rankine Cycles (ORC). Moreover, this case study is used to analytically discuss the influence of the allocation method in the LCA results for CHP plants and provide methodological recommendations for better allocation practices.

5.2 MATERIAL AND METHODS

5.2.1 Goal and scope definition

The LCA has been conducted according to ISO 14040:2006 and ISO 14044:2006 [38,127]. The intended audience of this LCA consists of technology developers, researchers involved in the field of bioenergy and LCA practitioners. An attributional LCA (ALCA) approach is followed since the goal of this study is to identify the activities within HBP causing the highest contribution to the environmental impacts, and not the consequences of changes in these activities [51,68].

Two technologies are considered for environmental comparison: 1) a combination of the electricity mix (EMIX) from the German national grid plus heat provided from a natural gas boiler (NG) and 2) biomass-based organic Rankine cycle (ORC) CHP.

The HBP plant delivers two different functions simultaneously, namely the supply of heat and power. Since the ORC Rankine cycle has a different heat to electricity ratio compared to the HBP, the definition of two functional units was preferred to the definition of a single functional unit with a fixed heat/electricity ratio. Hence, two functional units were defined as follows: 1 kWh of electricity or 1 MJ of heat.

The HBP technology finds one of the main strengths in its fuel flexibility [297] since it can operate with various biomass feedstocks in the forms of chips or pellets. To explore the effect of different feedstocks on the environmental impacts of the HBP CHP technology, this study explored the use of three different types of biomass fuels: wood chips, wood pellets, and Miscanthus pellets. The operation with wood chips was considered as the baseline scenario (WC), while the operation with wood pellets (WP) and the operation with Miscanthus pellets (MP) as alternative scenarios. The baseline scenario with wood chips was also used for comparison with the competing technologies, i.e. the ORC technology (fueled with wood chips as well) and the combination of grid electricity plus natural gas boiler. Additionally, this last competing option was also compared to the WP and MP alternative scenarios.

Figure 25 shows the process diagram of the HBP product system. The system boundaries follow a cradle-to-gate approach. As shown in Figure 25, all the life cycle stages from the extraction of the raw materials to the final dismantling and waste treatment are included. The final distribution and consumption of the products, i.e. heat and electricity, are not included in the LCA. Also, after the power plant is dismantled and parts are recycled, the use of the recycled materials is outside of the system boundaries. Biomass transport stages from the forest to the processing plant and from the processing plant to the HBP plant were included in the study. The transportation of plant components (e.g. the gasifier) from the production site to the power plant location and the construction activities of the plant were not included in the analysis. The exclusion of these activities was based on their expected minor contribution to the total environmental impacts, as also found in similar studies, e.g. [303].

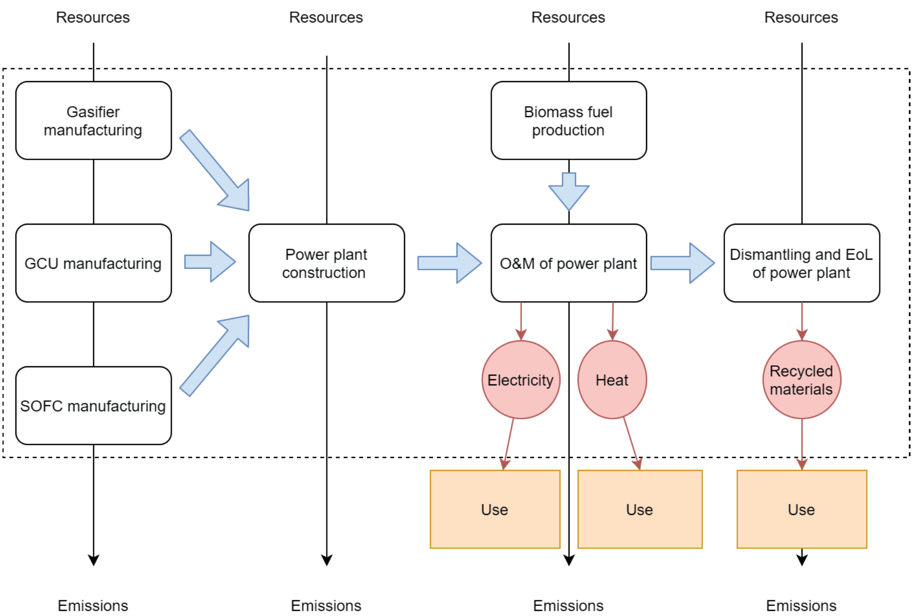

Figure 25. Flowchart of the HBP product system, including system boundaries (dashed lines).

The temporal scope of the study is placed in the near future (the next 5-10 years) when the HBP technology should be commercialized. The HPB is assumed to be installed in Germany, being the country with the maximum potential sales for the

HBP technology in Europe [307]. Nevertheless, some components for the HBP (e.g. the gasifier) might also be manufactured outside Germany (in other EU countries).

Seven mid-point impact categories were selected and the adopted impact assessment models for each impact category were selected following the ILCD recommendations [145] (see Table 8). Climate change (CC) and depletion of mineral, fossil, and renewable resources (MFRD) were chosen because they are considered top priorities in the current societal and political challenges [308]. Particulate matter (PM) and photochemical ozone formation (POF) are selected because of their relevance to the energy sector [309]. Acidification (AC), Terrestrial eutrophication (TE) and Water resource depletion (WRD) were selected because of their relevance for agricultural systems, and therefore for biomass production [42].

Table 8. Selected impact categories and models

Impact Category	**Unit**	**Impact assessment models**
Climate change (CC)	kg CO_2eq	IPCC 2013 Global Warming Potential 100 years [310]
Particulate matter (PM)	kg PM2.5 eq	Premature death or disability from particulates/respiratory inorganics from [311]
Photochemical ozone formation (POF)	kg NMVOC eq	Potential contribution to photochemical ozone formation for Europe from [312]
Acidification (AC)	molc H+ eq	Accumulated Exceedance (AE) characterizing the change in critical load exceedance of the sensitive area from [313]
Terrestrial eutrophication (TE)	molc N eq	Accumulated Exceedance (AE) characterizing the change in critical load exceedance of the sensitive area from [313]
Water resource depletion (WRD)	m^3 water eq	Freshwater scarcity: Scarcity-adjusted amount of water used from Swiss Ecoscarcity 2006 [314]
Mineral, fossil and renewable resource depletion (MFRD)	kg Sb eq	Depletion of resources based on the scarcity model from [315]

To assess the robustness of the results, two sensitivity analyses were conducted. As anticipated in the introduction, a comprehensive sensitivity analysis was performed on the allocation choices to explore their influence in the outcome of the LCA (and as recommended by ISO [38]). The second sensitivity analysis was performed to explore parameters that are potentially sensitive for the results and that might environmentally improve or make less attractive the technology in the future.

5.2.2 Life cycle inventory

5.2.2.1 Unit processes, data, and assumptions

This study assesses the small scale configuration of the HBP technology, which has a nominal electricity output of the SOFC of 199 kW. Its main characteristics during the average lifetime (assumed 18 years) are reported in Table 9.

For the foreground system, data on the gasifier and the GCU were collected from the industrial partners involved in the H2020 HiEff-BioPower project. For the SOFC, secondary data based on the scientific literature [304] were used due to the unavailability of specific primary data. The background data were largely based on the ecoinvent database (version 3.4). For unavailable data, assumptions were made based on literature (see the following sub-sections for details regarding each phase of the life cycle).

Since the system provides two different products (and functional units), it was necessary to determine an allocation key to partition the overall impact to the two functional units. Allocation by physical causality was not applied under the absence of a representative mathematical model (to model the causality relationships) [128,215,234]. Among the possible remaining allocation methods, the exergy key was chosen because it can represent both quantity and quality of both functional outputs, and is common practice for CHPs (e.g. ecoinvent uses such key [316] and is also recommended by RED II [9]). Table 9 shows the biomass input, intermediate performance indicators, and energy outputs in terms of their exergy and economic value based on the two biomass feedstocks. The exergetic outputs expressed in percentage reported in Table 9 represent also the allocation factors used for the baseline calculations. The economic values are based on three years (2015-2017) average prices for medium size industries without VAT, in Germany, retrieved from Eurostat [317]. The prices were 0.079 € per kWh of industrial electricity and 0.0086 € per MJ of industrial heat.

Table 9. Characteristics of the small scale HBP technology (8000 hours of operations per year). Modeled values.

Flow (unit)	**Wood chips (30 wt.%)**	**Pellets (from wood and Miscanthus) (5 wt.%)**
Biomass fuel (kW)	548.5	570.0
Biomass fuel (kg/h)	164.1	115.7
Gross electric power (kW)	170.5	190.0
Thermal power output (kW)	288.5	292.0
Electrical efficiency gross (%)	31	33
Electrical efficiency net (%)	30	32
Thermal efficiency (%)	53	51
Exergy output as heat (%)	24.6	22.7
Exergy output as electricity (%)	75.4	77.3
Economic output as heat (%)	41.8	39.3
Economic output as electricity (%)	58.2	60.7

5.2.2.2 Inventory data for chips and pellets

To model the life cycle of wood chips, the ecoinvent 3.4 dataset “Wood chips, wet, measured as dry mass {CH}| market for | APOS” was used. This dataset includes both wood chips from industrial activities and forest management and represents the average Swiss market (assumed to be a good proxy for Germany). In particular, wood chips from forest management represents an 85% share of the modelled Swiss wood chip market.

For wood pellets, the ecoinvent 3.4 dataset “wood pellet, measured as dry mass {RER}| market for wood pellet | APOS” was used.

For Miscanthus pellets, a similar dataset was not available in ecoinvent. Hence, the inventory data from [318] were used together with the best practices reported in [319]. An average dry yield value of 23.5 t Miscanthus (85% dry matter) per hectare was used to estimate the land requirements to provide enough fuel for the HBP plant for one year. The planting rate of 16’000 Miscanthus per ha was taken from [319]. As Miscanthus is a perennial crop, field preparation activities such as herbicide application, harrowing and plantation, occur only during the first year. The lifetime of the crop was assumed to be 18 years [319] and therefore 1/18 of the impact from field preparation activities was apportioned to one year of operation of the HBP plant. Once the Miscanthus is collected from the field, it is necessary to transport it to the pelleting plant. The transport distance to the pelleting plant was assumed to be 10 km by tractor [318]. For the chipping of Miscanthus, the energy consumption of the chipper and the amount of lubricating oil were retrieved from the ecoinvent 3.4 datasets “Wood chips, wet, measured as dry mass {CH}| wood chips production, hardwood, at sawmill | APOS”. For the pelleting of Miscanthus, the amounts of electricity, heat, lubricating oil and water were retrieved from the ecoinvent 3.4 dataset “Wood pellet, measured as dry mass {RER}| wood pellet production | APOS”. The transportation of miscanthus pellets to the HBP plant was assumed to occur by truck and with an average distance of 100 km [318].

5.2.2.3 Inventory data for the manufacturing of the power plant

The HBP manufacturing consists of three sub-processes: the manufacturing of the gasifier, the manufacturing of the SOFC stack and its balance of plant (BoP), and the manufacturing of the GCU. The data for the manufacturing of the gasifier is based on HBP project data and shown in Table 10.

Table 10. Materials of the gasifier including the primary gas treatment zone

Material	Amount	Process dataset
Steel (low alloyed) (kg)	6770	Steel, low-alloyed {RER}| steel production, converter, low-alloyed | APOS
Stainless steel (kg)	585	Steel, chromium steel 18/8, hot rolled {RER}| production | APOS
Iron-nickel-chromium alloy (kg)	220	Iron-nickel-chromium alloy {RER}| production | APOS
Concrete fireproof (kg)	4480	Concrete block {DE}| production | APOS
insulating material (kg)	1220	Glass wool mat {CH}| production | APOS

5.2.2.3.1 Inventory for the SOFC stack

The production of the SOFC stack was modeled considering secondary data from scientific literature and, to a lower extent, from ecoinvent database[7]. The literature data was retrieved from studies where the SOFC stacks had a similar power capacity as the HBP technology. The amount of electricity, nickel oxide, solvents, materials for the binder, carbon black, and chromium steel, as well as direct emissions (released during the production of the stack) to the air of carbon dioxide, methyl ethyl ketone and benzyl alcohol were taken from [304] and adjusted proportionally

[7] https://www.ecoinvent.org/

to the power capacity (factor of 0.793 based on 199 kWe of HBP SOFC versus 250 kWe of SOFC in [304]).

The data for the manufacturing of the anode, cathode, electrolyte, and the required ceramic materials (Lanthanum Strontium Manganite (LSM) and Yittria Stabilised Zirconia (YSZ)) were retrieved from [320].

The other secondary data for the SOFC stack, which were not available in [304,320], were retrieved from the already existing inventory in ecoinvent 3.4 called “Fuel cell, stack solid oxide, 125kW electrical, future {CH}| production APOS” and multiplied times 1.59 to account for the different size (assumption of linear proportionality of materials to the size as before).

For the production of the SOFC’s BoP, data for the inputs of steel and energy were retrieved from [304]. The other data were instead retrieved from ecoinvent 3.4 dataset “Fuel cell, solid oxide, 125 kW electrical, future {CH}| production | APOS”, which was modified as well by multiplying times 1.59.

5.2.2.3.2 Inventory for the GCU

The materials for manufacturing the cage of the GCU were assumed to be similar to the ones of the cage of the external reformer of the SOFC provided in ecoinvent 3.4. The 96 filter candles which are present in the GCU system at the beginning of the operation were included within the manufacturing stage. These candles are made from calcium-magnesium-silicate high-temperature fiberglass. The processes “Calcium borates {GLO}| market for | APOS”, “Magnesium {GLO}| market for | APOS” and “Silica sand {DE}| production| APOS” from ecoinvent 3.4 were used as a proxy for $CaMgO_4Si$. It was further assumed that 1.1 kg of material input would generate 1kg of filter candles. The mass of each candle was derived from the technical sheet of the candles [321].

5.2.2.4 Inventory data for operation and maintenance

The system operation includes all the material and energy inputs needed to operate the plant during one year of service (e.g. gas cleaning sorbents, water), waste outputs (e.g. ash which needs to be disposed of) and direct emissions to the environment (e.g. pollutant gas released to air).

The resulting direct emissions to air from the HBP are summarised in Table 11. Data for such emissions were only available for wood chips and wood pellets. The emissions from the operation with Miscanthus pellets were assumed to be the same as for wood pellets. Data on the ash formation (grate ash and fly ash) was retrieved from [301].

Table 11. Direct emissions (mg) to air per MJ of overall energy output (heat and electricity). OGC=organic gaseous compounds, TSP=total suspended particle, NO_x=nitrogen oxides. Maximum values shown in the table are used in the LCI.

Fuel	**Direct emissions, in mg/MJ**			
	CO	**OGC**	**TSP**	**NO_x**
Wood chips	<20	<0.01	<0.01	<0.01
Pellets	<20	<0.01	<0.01	<0.01

The operation of the gasifier needs 2.36 kg of natural gas for start-up operations and about 80.0 t of tap water per year for gasification air humidification (based on simulations from project data). According to measurements performed downstream the primary gas treatment zone, i.e. at GCU inlet, the syngas composition during utilisation of wood chips is as follows (in volume percentage): 15.4% CO, 10.6% CO_2, 1.8% CH_4, 8.3% H_2, 21.8% H_2O, 41.1% N_2. During the multiple tests performed, such a composition showed to be stable. After the primary treatment unit, the syngas typically shows contaminant concentrations in the range of 30 ppm for sulphur and 20 ppm for chlorine (on wet basis) when wood chips are used as fuel. The tar concentration at the inlet of the GCU was lower than 2.0 g/Nm^3 on dry basis and particulate matter contents (TSP) of about 200 g/Nm^3 on dry basis have been determined.

For the operation of the GCU, about 1.2 t of zinc oxides per year are needed for H_2S removal. The GCU also requires 1200 Nm^3 of Nitrogen per year for the cleaning of the filter elements. One year of operation of the GCU requires also 4800 kg of dolomite mixed with 900 kg of sodium bicarbonate[8] as coating materials respectively for Cl-sorption. The GCU has been designed to feed the SOFC with a product gas containing less than 5 ppm of chlorine, less than 1 ppm of sulphur, and less than 100 ppm of particulate matter (TPS < 0.1 mg/Nm^3, on wet basis). Since the composition of the syngas is expected to be stable (confirmed also by the first test runs), the uncertainty about the simulated electric power output of the SOFC

[8] From ecoinvent 3.4. Soda ash, dense {GLO}| market for | APOS

is expected to be very low.

The maintenance stage includes all the components which are replaced during the lifetime of the HBP plant. The SOFC stack and the GCU have a shorter lifetime than the average lifetime of the HBP CHP plant. Since the SOFC stack currently investigated for the HBP technology has an estimated lifetime of 5 years, the production of 1/5 extra SOFC stack per year was added to the maintenance stage. The GCU used for the HBP technology has a lifetime of 10 years, therefore, the production of 1/10 extra GCU per year was added to the maintenance stage. All other maintenance inputs (steel components and deionized water consumption for start-ups) for the SOFC were retrieved from [304] and scaled for the capacity of the SOFC under investigation. For the filter candles, an average of 30% of candles is estimated to be replaced each year of operation of the HBP technology. Therefore, the production of 29 extra candles per year was included in the maintenance stage.

5.2.2.5 Inventory data for end-of-life disposal

The main material employed in the components of the HBP is steel and can be recycled at the end of the life of each component. Based on the amount of steel present in the components (and their replacements), it was assumed that about 1900 kg of steel are recycled per average year of operation. The model included the energy for pressing and crushing the steel crap (based on [322]), a recycling efficiency (referred to as RRE in Equation 4) of 88% [323], and a transportation distance of 100 km from [322]. Such transportation was assumed to occur mainly by 16–32 t lorries Euro 3 [322].

A recycling process is a typical example of a multifunctional process fulfilling two functions i.e. the treatment of waste and the production of a recycled product. Based on our goal, the modeling approach (i.e. attributional), and the recommendations by ISO 14044:2006, mass allocation was applied[9]. The impacts arising from transportation (E_T), recycling (E_{RC}) and the extraction and processing of the primary material (E_v) were therefore allocated by mass between this life cycle and the following one (see equation 4 expressing the allocated impact to our functions). The resulting mass allocation factor (1/(1+RRE)) was 53% (1/1.88). The second part of equation 4 related to the virgin material takes into account the fact

[9] ISO 14044:2006 prioritizes allocating by a physical property for open-loop recycling over the economic value or number of uses (among ISO third level allocations i.e. by other relationships). Additionally, system expansion cannot be applied since we want to isolate the first function (first use of the material which led to its treatment) from the second function (next use or cycle of the material).

that the primary production was already accounted entirely in the manufacturing phase, and therefore the corresponding burdens (e.g. extraction of raw material) that belong to the following life cycle should be subtracted.

$$E_{steel\ disposal} = RRE(E_T + E_{RC})\frac{1}{1+RRE} - \frac{RRE}{1+RRE}\,E_v \qquad \text{Eq.4}$$

There are some precious metals (e.g. used as catalytic materials) used in the power plant that, depending on the recovery efficiency and initial concentration, might be economically convenient to recover, though e.g. hydrometallurgical treatment [324]. Nevertheless, such specific recovery processes were not modeled because of the unavailability of LCI data. Materials other than the steel used in the power plant components consist of hazardous waste (24 kg) and inert waste (10t per year in the chips scenario, and 20t per year for the pellets scenarios). The treatment of the hazardous waste was modeled through the ecoinvent dataset *Hazardous waste, for underground deposit {DE}| treatment of hazardous waste, underground deposit | APOS*. The inert waste consists mainly of materials for sorbents and was modeled through the ecoinvent 3.4 dataset *Inert waste, for final disposal {CH}| market for inert waste, for final disposal | APOS.*

5.2.2.6 Inventory data for the competing technologies

For the comparative analysis, the ecoinvent 3.4 datasets "1 MJ Heat, district or industrial, other than natural gas {CH}| heat and power co-generation, wood chips, 2000 kW, state-of-the-art 2014 | APOS" and "1 kWh Electricity, high voltage {CH}| heat and power co-generation, wood chips, 2000 kW, state-of-the-art 2014 | APOS" were used for the ORC. This dataset represents a state of the art ORC co-generation plant equipped with an electrostatic precipitator for particulate emission reduction and includes the infrastructure. For the separate production of heat and electricity, the ecoinvent 3.4 datasets "1 MJ Heat, central or small-scale, natural gas {CH}| heat production, natural gas, at boiler condensing modulating <100kW | APOS" and "1 kWh Electricity, medium voltage {DE}| market for | APOS" were used.

Following the description provided in ecoinvent 3.4 for the ORC ecoinvent dataset, the capacity of the ORC plant is 1000 kW thermal, and 200 kW electric (similar to the electric output of the HBP technology). This information was used to estimate the exergy allocation factor of 46% for heat (assumed district heating provided at 90°C as for HBP) and 54% for electricity. Based on 2015-2017 average prices for

Germany, the economic allocation shares for ORC would be 66% for heat and 34% for electricity. Since the total power input (as wood chips) is 2000 kW, this ORC plant has an overall energy efficiency of 60%, i.e. 10% electrical efficiency plus 50% thermal efficiency.

5.3 RESULTS

5.3.1 Environmental impact of the HBP technology

5.3.1.1 Baseline (wood chips)

Figure 26 shows the breakdown of the impact of the HBP technology for the seven investigated impact categories (see appendix for absolute values).

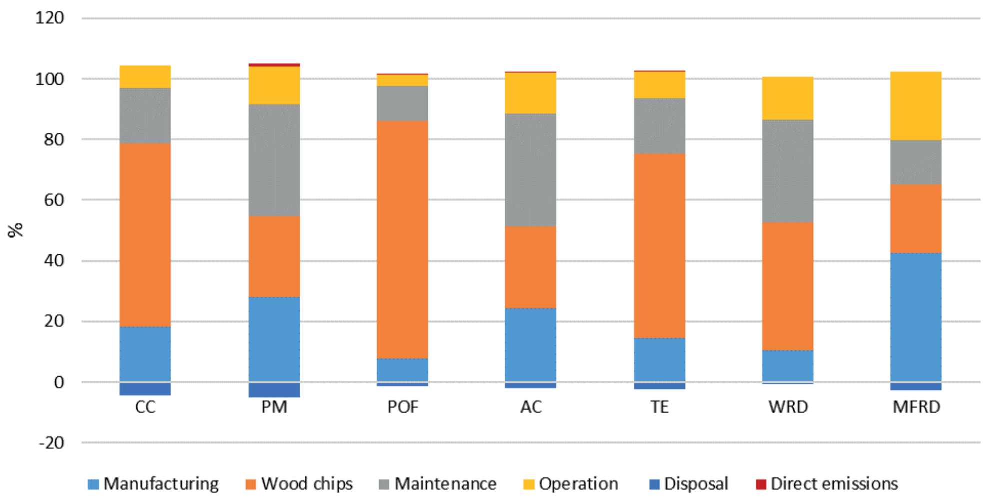

Figure 26. Main contributions to the cradle-to-grave environmental impact of producing heat and electricity with the HBP technology using wood chips as biomass fuel. The presented breakdown is valid for both functional units. CC=Climate change, PM=Particulate matter, POF=Photochemical ozone formation, AC=Acidification, TE=Terrestrial eutrophication, WRD=Water resource depletion, MFRD=Mineral, fossil and renewable resource depletion.

The main contributions to the cradle to grave environmental impact are the wood chips used, followed by the maintenance phase and manufacturing phase.

The impact of wood chips is made of two components i.e. their transportation and their production. The impact of transporting wood chips (based on the Swiss supply chain assumed by the dataset retrieved from ecoinvent) represents 18-27%

of the impact of wood chips for all impact categories, except for photochemical ozone formation (9%) and water depletion (2%). In all impact categories, except water depletion, the impact of the production of wood chips is mainly caused by the production and combustion of diesel and petrol (60-80%) used in power sawing machines, skidders and chippers. The production of the lubricants used in the three processes mentioned above causes about 2-10% of the impact of wood chips production in all impact categories except for water resource depletion for which it represents 80% of the impact. The water depletion impact of wood chips is mainly due to the fraction of vegetable oils used for lubricating the chains during power sawing activities (in absolute terms, this impact is quite low, see Figure 27 and Figure 31).

The main impact of the manufacturing stage is due to the production of the SOFC system, which contributes to 63%-100% of the impacts in this stage (depending on the category). Within the SOFC system, the production of the SOFC stack and the inverter. This is mainly due to the large electricity consumption during the manufacturing of the stack (as also highlighted by Rillo et al. [304]) and the manufacturing of chromium steel (mainly caused by the production of ferrochrome [304]).

Concerning the maintenance impacts, the maintenance of the SOFC system contributes to 63-100% of the impact depending on the impact category. The major contributor (95-99%) to the impact of the maintenance of the SOFC system is the replacement of the SOFC stack, which requires the production of a new SOFC stack every five years of operations

The operation phase is dominated by the operation of the GCU (mainly zinc oxide used and sodium bicarbonate) expect for water depletion whose impact is mainly caused by the water used for the operation of the gasifier. The contribution of direct emissions is negligible in all impact categories. The particulate matter caused by the operation of the HBP technology was only 1% of the total particulate matter impact.

5.3.1.2 Alternative scenarios (wood and Miscanthus pellets)

Figure 27 shows the environmental impact of the baseline scenario in comparison to the alternative scenarios.

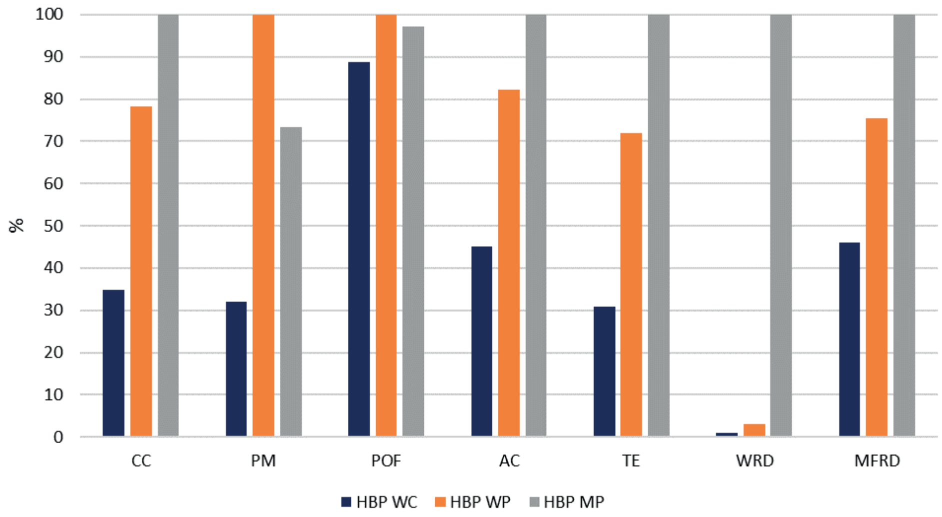

Figure 27. HBP technology fuelled with various biomass fuels (the same graph applies to both 1 MJ heat or 1kWh electricity). Values are normalized taking as 100% the impacts of the most impacting scenario. CC=Climate change, PM=Particulate matter, POF=Photochemical ozone formation, AC=Acidification, TE=Terrestrial eutrophication, WRD=Water resource depletion, MFRD=Mineral, fossil and renewable resource depletion. WC=wood chips, WP=wood pellets, MP=Miscanthus pellets.

The results show that, in all impact categories, the total life cycle impact is the lowest for the operation with wood chips compared to the other two biomass scenarios (wood pellets and Miscanthus pellets). Since the inventories for manufacturing and maintenance are the same, the main difference between the three scenarios is the production of the biomass fuel (wood chips have lower environmental impacts than the two pellets).

The impact of the WC scenario is between 10% and 70% lower than for the WP scenario (with the highest impact difference for water depletion and particulate matter). For water depletion, the impact of wood pellets is almost entirely caused by the electricity consumption of the pellet factory. For particulate matter, the shaving process accounts for about 54% of the impact of producing wood pellets. Shaving is, therefore, the main cause of the significantly higher particulate matter impact in the production of wood pellets compared to wood chips. The impact of shaving is mainly caused by its drying process (87%), which leads to high particulate emissions due to the combustion of industrial wood.

5

Except for particulate matter and photochemical oxidant formation, the Miscanthus scenario presents higher environmental impacts than the wood pellets scenario. The characterized results indicate between 18% and 28% lower impacts for the wood pellets scenario than for the Miscanthus scenario in the categories of acidification, climate change, resource depletion (mineral, fossil and renewables) and terrestrial eutrophication. The particulate matter impact is lower (-27%) in the case of Miscanthus pellets because the shaving process -that was the main source of impact for wood pellets- is not used to produce Miscanthus pellets.

The difference in impact is even higher for the water depletion category, which scores 97% lower in the wood pellets scenario than in the Miscanthus pellets scenario. The irrigation needed during its cultivation is the main cause of the significantly higher water depletion in the scenario with Miscanthus pellets (see Figure 28). Other activities that are an important source of impacts for Miscanthus pellets are the electricity for pelleting, the diesel burnt during the harvesting stage and the emissions caused by fertilizing (see Figure 28 for the single contributions in each impact category).

Similar to the baseline case of wood chips, direct emissions have a negligible impact for the operation with wood pellets and Miscanthus pellets. This aspect is particularly important in the case of biomass technologies installed in heavily populated areas.

Since Miscanthus is an energy crop, it is important to assess the impacts due to land use. As for the other impact categories, the selection of the method was based on ILCD recommendations [145]. Accordingly, the carbon deficit caused by land use was assessed using the Soil Organic Matter model of [325]. This model accounts for the changes in soil quality caused by the occupation and transformation of the land. Land occupation generates changes in soil quality which depend on the amount of area occupied and the duration of such an occupation. Land transformation generates changes in soil quality which depend on the extent of changes in land properties and the area affected. In this model, the deficits in soil organic matter content are assessed and expressed by an indicator whose unit is kilograms of carbon deficit[10].

[10] These deficits are caused by the effects of agricultural practices on degradation rates. The changes can also be additions of soil organic matter. For example, these additions can be caused by the application of manure or crop residues. It should be observed that this modeling of land use impacts does not account for the counterfactual effects caused by land use changes modelled in consequential LCAs of bioenergy.

The production of 1 MJ of heat using Miscanthus pellets generates a 0.86 kg C deficit. Such an impact is much higher than for 1 MJ of heat generated using wood chips (0.12 kg C deficit) and using wood pellets (0.13 kg C deficit). The reason is that Miscanthus is an energy crop. Hence, differently from the feedstock for wood chips and pellets, it requires dedicated cultivation.

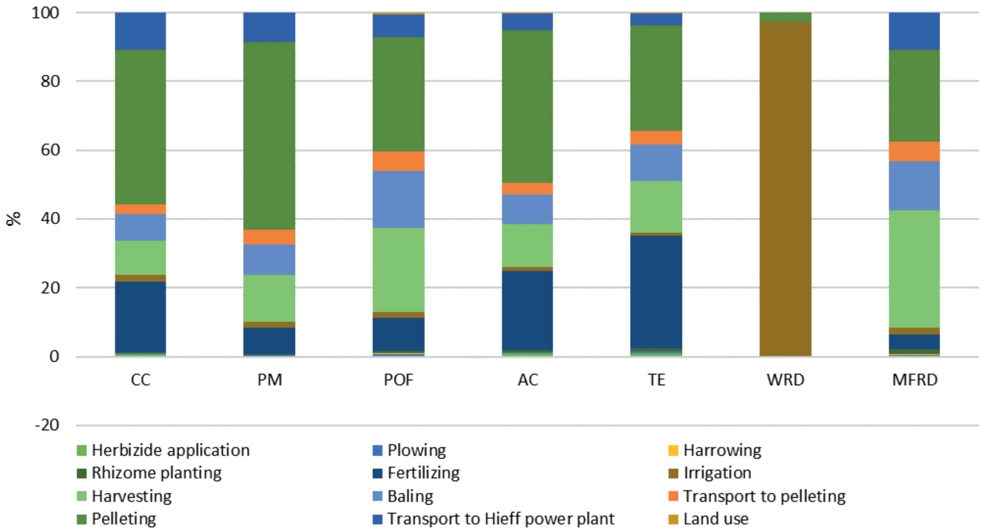

Figure 28. Main contributions to the environmental impact of Miscanthus pellets supplied to the HBP CHP plant. CC=Climate change, PM=Particulate matter, POF=Photochemical ozone formation, AC=Acidification, TE=Terrestrial eutrophication, WRD=Water resource depletion, MFRD=Mineral, fossil and renewable resource depletion

5.3.2 Benchmarking with competing technologies

5.3.2.1 Comparison with ORC technology (both fueled with wood chips)

Figure 29 shows the comparison between HBP technology and ORC technology both fueled with wood chips.

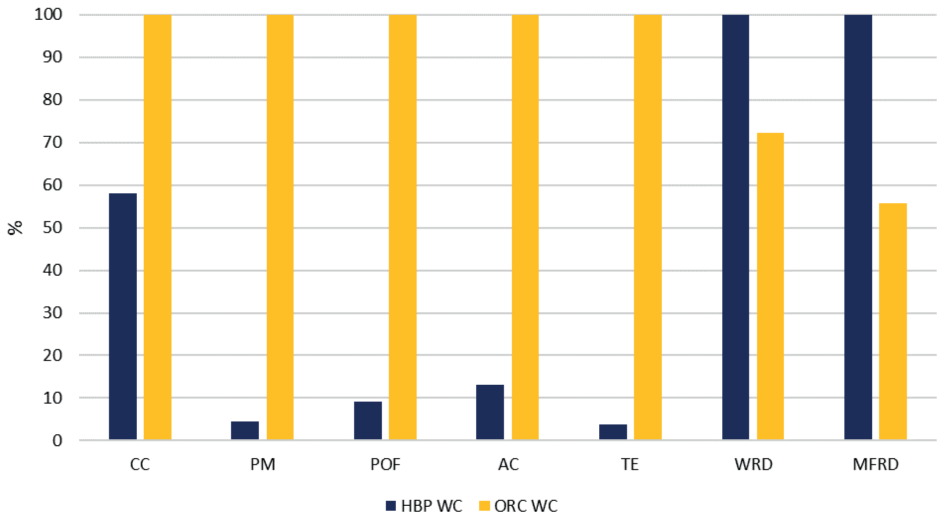

Figure 29. Comparison of HBP technology with ORC technology for 1 MJ of heat. The graph for 1kWh electricity shows some minor differences due to a slightly different Carnot factor assumed for ORC techology. Values are normalized with respect to the impacts of the most impacting scenario. CC=Climate change, PM=Particulate matter, POF=Photochemical ozone formation, AC=Acidification, TE=Terrestrial eutrophication, WRD=Water resource depletion, MFRD=Mineral, fossil and renewable resource depletion. WC=wood chips.

The heat co-generated by the HBP technology shows lower environmental impact compared to the same amount of heat produced by the ORC technology allowing -42% impact on climate and -87/-96% impact in terms of particulate matter, photochemical ozone formation, acidification and terrestrial eutrophication. These differences can be explained by two main advantages of the HBP technology: 1) the HBP has higher energy and exergy efficiencies, and therefore less biomass is needed for producing the same amount of energy and exergy as outputs and 2) HBP avoids the external combustion of biomass occurring in the ORCs, and therefore releases less particulate emissions (the particulate matter impact of the ORC technology is for 97% caused by direct emissions of particulates). Although the HBP technology has the same thermal efficiency as the ORC technology, its electric efficiency is three times higher. On the other hand, the HBP technology shows higher water depletion (+38%) and resource depletion (+79%). For water depletion, the high impact is caused by the replacements of the SOFC stack. For depletion of resources (minerals, fossil and renewables), the main cause can be found in the production of the SOFC stacks. All these components are not present in the case of an ORC.

Similar results were obtained when comparing electricity production from HBP and ORC. The HBP shows a lower impact of -45% for climate change, -96% for particulate matter, -91% for photochemical ozone formation, -88% acidification and -96% for terrestrial eutrophication. On the other hand, the HBP technology shows an increased impact in terms of water (+31%) and depletion of resources (MFRD) +70%.

5.3.2.2 Comparison with conventional production of heat and electricity

Figure 30 and Figure 31 shows the comparison between the HBP technology operating with the three investigated biomass fuels and conventional separate productions of heat and electricity.

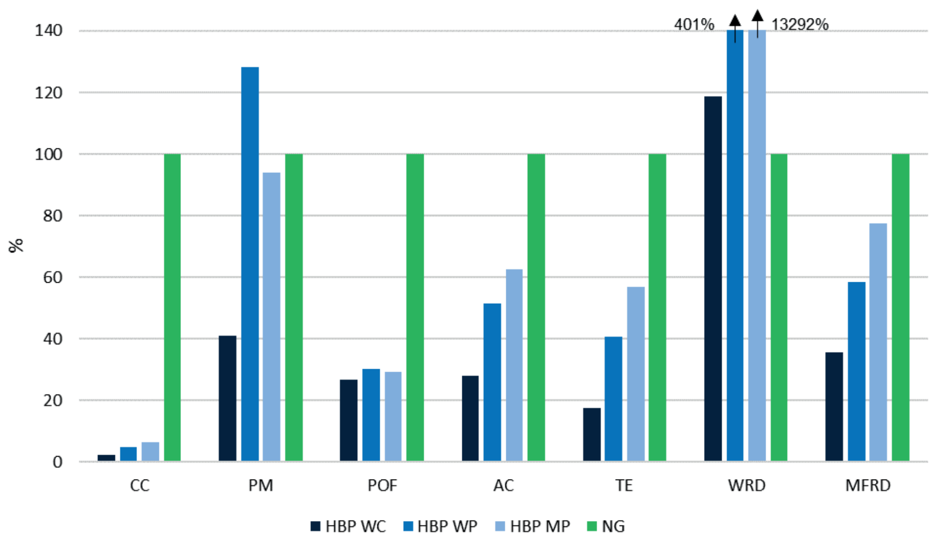

Figure 30. Comparison of HBP technology with competing technologies (for 1 MJ heat). Values are normalized taking as 100% the impacts of the natural gas boiler. CC=Climate change, PM=Particulate matter, POF=Photochemical ozone formation, AC=Acidification, TE=Terrestrial eutrophication, WRD=Water resource depletion, MFRD=Mineral, fossil and renewable resource depletion. WC=wood chips, WP=wood pellets, MP=Miscanthus pellets.

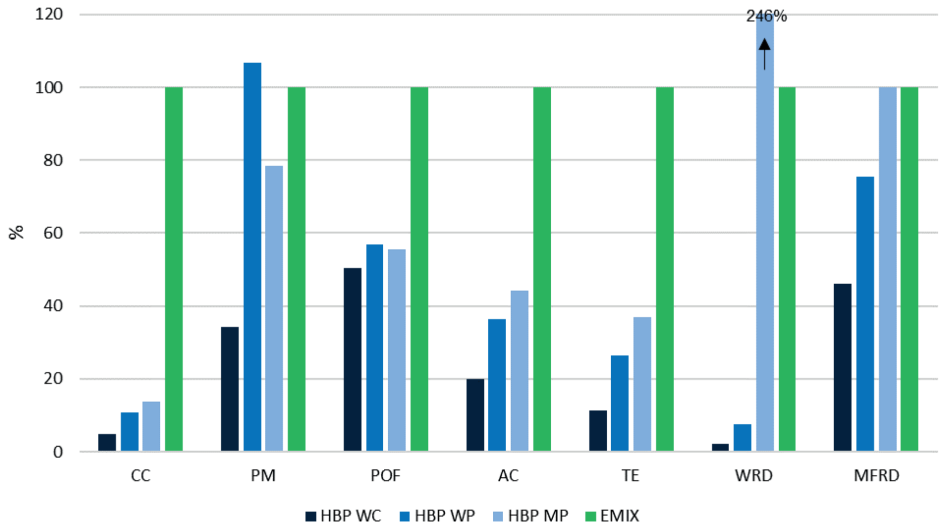

Figure 31. Comparison of HPB technology with competing technologies (for 1 kWh electricity). Values are normalized taking as 100% the impacts of the German electricity mix. CC=Climate change, PM=Particulate matter, POF=Photochemical ozone formation, AC=Acidification, TE=Terrestrial eutrophication, WRD=Water resource depletion, MFRD=Mineral, fossil and renewable resource depletion. WC=wood chips, WP=wood pellets, MP=Miscanthus pellets.

The heat co-generated by the HBP technology shows a lower environmental impact compared to the heat produced by a condensing boiler burning natural gas. Even considering the least preferred fuel scenario in each impact category, the impact differences are at least -94% in terms of climate change, -70% in photochemical ozone formation, -37% in acidification, -43% in terrestrial eutrophication and -22% in depletion of resources. In particular, the significant difference in climate change is mainly generated by the biogenic carbon dioxide emissions (which are assumed to be carbon neutral) instead of fossil ones.

On the other hand, the HBP technology causes +28% impacts in particulate matter in the WP scenario (caused by the high particulate matter released when producing wood pellets) and significantly higher water depletion for the MP scenario (+13000%), due to the water used for irrigation in the cultivation of Miscanthus (the only scenario with irrigation). When wood chips are fed instead of pellets, the HBP shows a much lower impact in terms of particulate matter (-59%) but still a relatively higher impact in water depletion (+19%), due to indirect water consumption in different life cycle activities.

The electricity co-generated by HBP technology shows a lower environmental impact compared to electricity produced by the German electricity mix (EMIX). In particular, even considering the worst fuel scenario, the differences of impact are at least -86% for climate change, -43% for photochemical ozone formation, -56% for acidification and -63% for terrestrial eutrophication. Nevertheless, the HBP using Miscanthus pellets as fuel can lead to an increase in particulate matter (+7%) and water resource depletion (+146%; caused by irrigation of Miscanthus) and the same impact in terms of depletion of mineral and fossil resources. When operating with wood chips, the HBP shows a much lower impact than the EMIX, leading for example to -66% impacts in particulate matter, -98% in water depletion and -54% in depletion of resources (MFRD).

5.3.2.3 Comparing with other LCAs of SOFC CHPs

In the literature[6], 8 LCAs of SOFC CHPs have been conducted along with a review of LCAs on SOFC systems. In most of these LCAs, the fuels used in the SOFC CHPs assessed were natural gas and biogas and the capacity of the SOFC was only a few kilowatts (1-20 kW) of electricity. Among these 8 LCAs, LCAs on SOFC CHPs of larger capacity (comparable to the one of the HBP) were conducted by [326–328]. Our results for the climate change impact of the HBP technology (0.03-0.09 kgCO2eq/ kWh_{el} depending on the fuel considered) indicate considerably lower impacts than for the SOFC CHPs assessed by these LCAs.

These lower impacts are especially found for the SOFC CHPs using natural gas as fuel because of the avoidance of direct emissions of fossil CO_2 allowed by the HBP which is fueled with a biofuel instead of fossil fuel. In particular, among the LCAs of SOFC CHPs whose size is comparable to the HBP and operating with natural gas, Strazza et al. [326] assessed a 230 kW_{el} SOFC CHP with electric efficiency of 53.4%. The resulting impact was 0.47 $kgCO_2eq$ per kWh of electricity, which is at least 5 times higher than for the HBP. An older study [328] assessing a 125 kW_{el} SOFC CHP operating with natural gas, calculated an impact of 0.9-1.0 $kgCO_2eq$ per kWh of electricity, which is at least 10 times higher than for the HBP. Staffell et. [320] assessed a 1 kW_{el} micro-SOFC CHP fueled with natural gas and calculated an impact of 0.32-0.37 $kgCO_2eq/kWh_{el}$, which is a least 3-4 times higher than for the HBP.

The impact of the HBP is also at least 44% lower than for SOFC CHPs operating with biogas. In this case, the main reason can be found in the different fuel production processes and composition of the fuel used and consequent different composition of the direct emissions (e.g. methane emissions released with the exhaust gases). In particular, Strazza et al. [326] calculated an impact of 0.16 $kgCO_2eq$ for a 230 kW_{el} SOFC CHP with 52.2% electric efficiency operating with biogas from sewage sludge. For the same type of system but with a capacity of 125 kW_{el}, Sadhukhan [327] calculated an impact of about 0.19 $kgCO_2eq$ per kWh_{el}. Concerning this last figure, Sandhukhan used a different functional unit (1 ton of sewage sludge processed through anaerobic digestion) and we derived it by applying exergy allocation on the energy outputs.

Similarly to the HBP, for multiple impact categories, Strazza et al. found that the impact of this system, independently on the fuel considered (natural gas or biogas) was dominated by the production of the fuel. The only exception was the climate change impact of the operation with natural gas, whose impact was mainly caused by the operation phase (mainly direct emissions of fossil CO_2 of the system).

5.3.3 Alternative methods for solving multifunctionality

Exergy allocation was used to partition the total environmental impact between heat and electricity, as explained in section 5.2.2. In the literature, the two most applied alternative approaches to address the multifunctionality of SOFC CHPs are system expansion (enlargement) and economic allocation [306]. The first approach was only applied in studies where it was not necessary to differentiate between the impacts of heat and those of electricity.

Although the substitution method has a clear link with consequential analyses, it has been often applied in the literature for attributional LCAs with goals similar to the one of this study [57,71,329]. By the substitution method, the impact of the main product is obtained by subtracting the impact of the marginally avoided secondary products from the impacts of the overall system [51,213]. In particular, the main product is defined as the one providing the highest share of revenues within the analysed product system (physical/economical significance) [51].

A sensitivity analysis was performed to understand the influence of the method on the results of the study. This analysis explored the variation of the results, when applying economic allocation and the substitution method for the WC scenario, ORC scenario, and separate productions of heat and electricity.

When applying substitution, the first step is identifying the main product. Based on the economic heat/electricity ratio (see Table 9), electricity is the main product for the HBP. The production of heat by the HBP technology can marginally avoid the production of heat from natural gas on the market (Heat, central or small-scale, natural gas {CH}| heat production, natural gas, at boiler condensing modulating <100kW | Conseq from ecoinvent 3.4).

On the other hand, the HBP heat could also avoid the production of heat by an average biomass boiler (Heat, district or industrial, other than natural gas {CH}| heat production, softwood chips from forest, at furnace 300kW | APOS from ecoinvent 3.4). The choice of a biomass boiler as substituted technology can be considered as an "alternative activity allocation" i.e. a form of "proxy-based disaggregation" [51]. This type of allocation is performed through the subtraction of impacts but differs from the substitution performed in consequential LCAs because it is not based on modeling of marginality [51]. Instead, this allocation takes as substituted processes the ones providing "primary productions of identical products and not of products that fall under different categories" [51]. This approach might, therefore, be an option also in attributional LCAs when reflecting the underlying physical relationship between the main and subsidiary products [51]. This sensitivity analysis considered both approaches, the substitution of a marginal activity (heat from a natural gas condensing boiler) and the substitution of an alternative activity (heat from a biomass boiler, marked as (a) in Figure 32 and Figure 33).

Based on the economic heat/electricity ratio of the ORC technology (see Table 9), heat is the main product for the ORC technology. In the case of the ORC, the electricity produced from the ORC avoids the production of marginal electricity from the electricity mix (this process is represented in the model by the ecoinvent dataset Electricity, high voltage {DE}| market for | Conseq).

The sensitivity analysis (see Figure 32 and Figure 33) indicated that, compared to exergy allocation, the economic allocation method apportions more impacts on heat (+70% in every category) while it decreases by 23% the impacts of electricity. The same applies to the ORC technology (+40% and -36% respectively for heat and electricity). For CHPs, it is therefore important to show the impacts for both heat and electricity when an allocation method is applied, so that a full picture of its environmental impacts is provided.

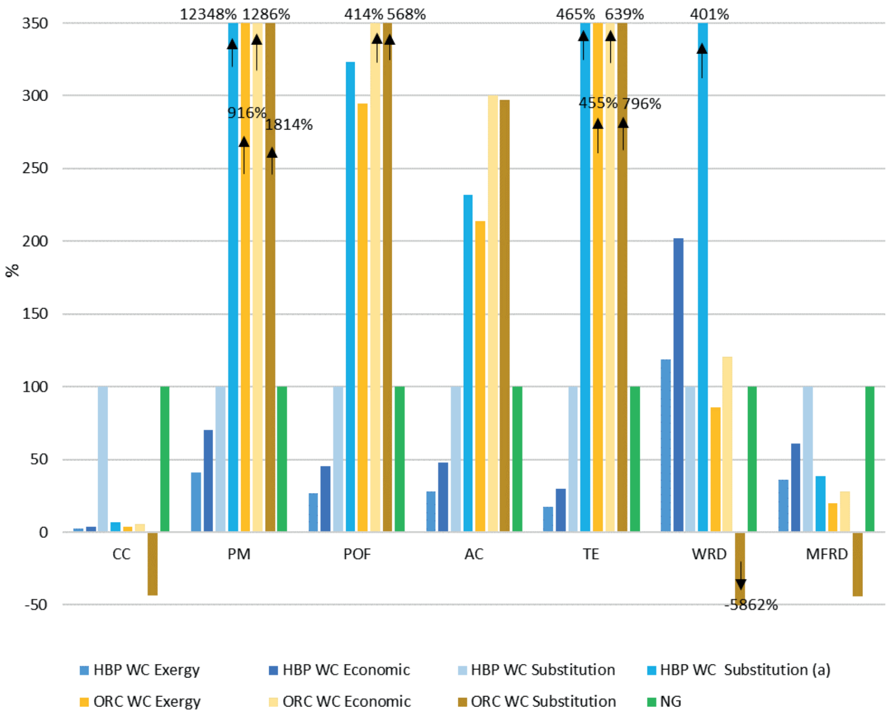

Figure 32. Sensitivity on allocation method for the generation of 1 MJ with HBP technology and competing technologies. Boiler running with natural gas taken as 100%. ORC=Organic Rankine Cycle, NG=Natural gas boiler, (a)= substitution of heat from a biomass boiler.

On the other hand, the conclusions of the comparative assessment did not change when applying exergy or economic allocation methods. This was true for all three comparisons: 1) between HBP with wood chips and ORC with wood chips, 2) between the three different biomass fuels scenarios and 3) between the HBP and the separate productions. For instance, the impact of the HBP per MJ of heat with both allocation methods was lower than for ORC in climate change, particulate matter, photochemical ozone formation, acidification and terrestrial eutrophication, but it was higher in the two categories concerning the depletion of resources (see Figure 32). On the other hand, the percentages of potential environmental impact savings or intensifications compared to separate production can change significantly. For example, for climate change, the savings of impact of the HBP compared to ORC was 42% when using exergy allocation while it was decreased to 30% with economic allocation. However, for particulate matter, there was no difference.

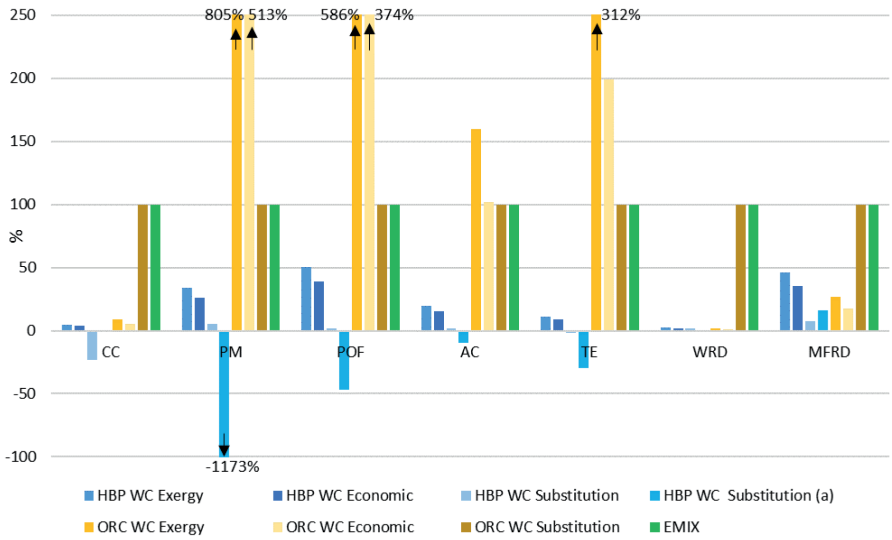

Figure 33. Sensitivity on allocation method for the generation of 1 kWh with HPB technology and competing technologies (1 kWh electricity). Electricity mix taken as 100%. ORC=Organic Rankine Cycle, EMIX=Electricity mix, (a)= substitution of heat from a biomass boiler.

Concerning substitution (see Figure 32 and Figure 33), the variations compared to other allocation methods were small or large depending on the impact category considered and the type of substitution applied. Moreover, both types of substitution approaches and the alternative activity method led to negative results in some impact categories. This last aspect highlights that the modeling was not consistent with the attributional goal of the study, which is not aimed at assessing a change in demand, and therefore, it should provide negative emissions for a single product of a multifunctional process whose overall impact is positive [51]. When a physically/economically significant product (the substituted function was 42% of total revenues for HBP and 34% for ORC) is substituted in attributional LCAs (by assuming that its impact corresponds to the one that would be replaced in the market), the results are often not aligned with other allocation methods and contrasts with the attributional aim of the LCA. This aspect emerges clearly when multiple impact categories are investigated in the same LCA study resulting in conclusions in contrast with other allocation methods and of difficult interpretation.

5.3.4 Sensitivity analysis on potentially sensitive parameters

5.3.4.1 Internal parameters

The results of the analysis indicated that the production of the biomass fuel (23-78% for the baseline scenario WC, depending on the category) and the SOFC stacks (10-43%) have a high contribution to the total impacts.

The ecoinvent dataset used for wood chips included both wood chips obtained as by-products of sawmill activities (15%) and from forest management (85%). To reduce the environmental impact, a scenario with only sawmill wood chips as fuel could be used. This type of wood chips presents a lower impact compared to wood chips from forest management because an important percentage of the impact of the upstream activities occurring in the forests is allocated to the main products of the sawmills. This scenario was assessed by sensitivity analysis to estimate the potential variation in the impact of the HBP (see the second column of Table 12). By using only sawmill wood chips, the environmental impact of HBP technology can be significantly reduced (indicatively by 10-40%).

Table 12. Sensitivity analysis on the reduction of the environmental impact of the HBP technology by either increasing the SOFC stack lifetime or using only wood chips produced as industrial by-products.

Impact category	Industrial wood chips only (measured in % variation)	Longer SOFC stack lifetime (measured in % variation)
Climate change (CC)	-37%	-4%
Particulate matter (PM)	-15%	-6%
Photochemical ozone formation (POF)	-47%	-2%
Acidification (AC)	-16%	-7%
Terrestrial eutrophication (TE)	-37%	-4%
Water resource depletion (WRD)	+8%	-10%
Mineral, fossil and renewable resource depletion (MFRD)	-10%	-3%

Alternatively, the impact of HBP technology could be improved by acting on the SOFC stack. Since the stack needs to be replaced every five years, the environmental impact could be improved by increasing the SOFC stack lifetime and therefore reducing the number of replacements over the plant lifetime. The second column of Table 12 shows the reduction of the environmental impact that could potentially be achieved by increasing the lifetime of the SOFC from five to seven years. This would lead to a decrease between 2% and 10% of the impacts of the wood chips scenario (baseline).

5.3.4.2 External parameters

Since the technology will be deployed after 2025, it is important to explore how the comparative evaluation will change taking into account the current trends of decarbonization, which should lead to a decrease in the share of coal-produced electricity by shifting to renewables. In particular, the expected decarbonization of the European electricity grid will diminish the environmental benefits of HBP technology.

Table 13. Sensitivity analysis on the savings of environmental impacts of 1 kWh of electricity produced by the HBP compared to the grid electricity mix (EMIX).

Impact category	**EMIX Germany, ecoinvent 3.4**		**Future EMIX Germany, EU reference scenario 2030**		**Future EMIX EU, IEA current policy scenario 2030**	
	1 kWh electricity	**Savings (%) HBP vs EMIX**	**1 kWh electricity**	**Savings (%) HBP vs EMIX**	**1 kWh electricity**	**Savings (%) HBP vs EMIX**
Climate change (kg CO_2eq)	6.41E-01	-95%	5.61E-01	-95%	3.96E-01	-92%
Particulate matter (kg PM2.5 eq)	7.87E-05	-66%	7.84E-05	-66%	6.74E-05	-60%
Photochemical ozone formation (kg NMVOC eq)	6.02E-04	-49%	6.06E-04	-50%	4.98E-04	-39%
Acidification (molc H+ eq)	1.58E-03	-80%	1.35E-03	-77%	1.12E-03	-72%
Terrestrial eutrophication (molc N eq)	4.33E-03	-89%	3.86E-03	-87%	3.49E-03	-86%
Water resource depletion (m^3 water eq)	2.75E-03	-98%	1.32E-04	-54%	4.47E-04	-87%

Impact category	EMIX Germany, ecoinvent 3.4		Future EMIX Germany, EU reference scenario 2030		Future EMIX EU, IEA current policy scenario 2030	
	1 kWh electricity	Savings (%) HBP vs EMIX	1 kWh electricity	Savings (%) HBP vs EMIX	1 kWh electricity	Savings (%) HBP vs EMIX
Mineral, fossil and renewable resource depletion (kg Sb eq)	7.47E-06	-54%	7.89E-06	-56%	7.41E-06	-54%

To assess this variation, the electricity mix based on two future scenarios for 2030 were considered: the EU reference scenario for Germany [272] and the IEA current policy scenario [330]. Due to the unavailability of the IEA current policy scenario for Germany, the IEA average mix of 2030 for the EU was taken as a proxy. This second scenario represents a more decarbonized electricity sector and includes other countries where the HBP could be commercialized. In particular, the IEA current policy scenario has only 13.7% coal and 44.7% renewables. The future savings of environmental impact allowed by the HBP is shown in the two columns on the right in Table 13. The environmental savings from the HBP technology will be only slightly affected (order of 5% overall) by the change expected in the electricity mix for 2030.

5.4 CONCLUSIONS

This article presented the first life cycle assessment (LCA) of a novel technology integrating biomass gasification and SOFC technologies. This technology is currently under development in the H2020 HiEff-BioPower (HBP) project and allows for the use of various biomass types as feedstock. This LCA assessed the environmental impacts when operating the technology with three different fuels: wood chips, wood pellets and Miscanthus pellets. The impact of producing heat and electricity with the HBP technology was compared to the state of the art competing technologies. The results showed that most of the impacts of producing heat and electricity with the HBP technology are generated during the production (including transportation) of the biomass fuels (between 23% and 99%

of the total impacts depending on the category and the fuel). The use of wood chips as fuel generates much lower impacts per functional unit than the operation with wood pellets (11-70% lower) and Miscanthus pellets (9-99% lower), in all impact categories. The next highest contributor to the life cycle environmental impacts is the SOFC stack, due to both the high energy intensity (especially in electricity consumption) and material intensities of its manufacturing processes, and its short lifetime (the stack should be replaced every 5 years). Beyond increasing the fuel efficiency of the technology and therefore reducing the consumption of biomass fuels, the main recommendation to technology developers would be to increase the lifetime of the SOFC stack. Increasing the SOFC stack lifetime could decrease the environmental impacts of 2-10%, depending on the category.

The comparison of the HBP technology with separate productions of heat and electricity (from natural gas condensing boilers and the German electricity grid) indicated significantly lower impacts for the HBP technology, especially in climate change (86/94% lower), photochemical ozone formation (-43/-70%), acidification (-37/-56%) and terrestrial eutrophication (-43/-63%). Overall, HBP showed also better performance than ORCs, since they have higher exergy efficiencies and almost zero particulate emissions resulting in 86-96% lower impact in the category particulate matter.

The sensitivity analysis on the allocation method for heat and electricity provided useful insights for the choice of allocation methods in CHP plants, and led to the following recommendations: 1) the attributional LCAs of CHPs should always provide the results for both heat and electricity to allow for better interpretation of results, independently of the allocation method, 2) LCA results from different CHP plants should not be compared if they assumed different allocation approaches and 3) substitution is not recommended in attributional LCA (especially if the substituted product is not a minor by-product) because it provides results which are not in line with the attributional aim (e.g. negative emissions) and lead to conclusions in contracts with the ones from applying allocation methods which are proven to be a good proxy of physical causality for CHPs and therefore preferable.

5.A APPENDIX

Table 5A1 presents the characterized environmental impacts per functional unit of heat and electricity for each fuel scenario.

Table 5A1. Cradle-to-grave environmental impacts per functional unit and biomass fuel. WC=wood chips, WP=wood pellets, MP=Miscanthus pellets.

	1 MJ of Heat			**1 kWh of electricity**		
Impact Category	**Wood chips**	**Wood pellets**	**Miscanthus pellets**	**Wood chips**	**Wood pellets**	**Miscanthus pellets**
Climate change (kg CO_2eq)	1.52E-03	3.42E-03	4.36E-03	3.05E-02	6.87E-02	8.77E-02
Particulate matter (kg PM2.5 eq)	1.34E-06	4.19E-06	3.07E-06	2.70E-05	8.41E-05	6.17E-05
Photochemical ozone formation (kg NMVOC eq)	1.51E-05	1.71E-05	1.66E-05	3.04E-04	3.43E-04	3.33E-04
Acidification (molc H+ eq)	1.57E-05	2.87E-05	3.49E-05	3.15E-04	5.78E-04	7.02E-04
Terrestrial eutrophication (molc N eq)	2.46E-05	5.72E-05	7.96E-05	4.93E-04	1.15E-03	1.60E-03
Water resource depletion (m^3 water eq)	3.01E-06	1.02E-05	3.36E-04	6.03E-05	2.04E-04	6.76E-03
Mineral, fossil and renewable resource depletion (kg Sb eq)	1.71E-07	2.80E-07	3.71E-07	3.43E-06	5.63E-06	7.46E-06

CHAPTER 6

Attributional and consequential LCAs of a novel bio-jet fuel from Dutch potato by-products

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ABSTRACT

To mitigate the climate change impact of aviation, jet fuels from bio-based by-products are considered a promising alternative to conventional jet fuels. Life cycle assessment (LCA) is a commonly applied tool to determine the environmental impacts of bio-jet fuels. This article presents both attributional and consequential LCA models to assess an innovative bio-jet fuel produced from potato by-products in the Netherlands. The two models led to opposite conclusions regarding the overall environmental performance of this bio-jet fuel. The attributional LCA showed that this bio-jet fuel could offer about a 60% GHG emissions reduction compared to conventional jet fuel. In comparison, the consequential LCA estimated either a much lower climate change benefit (5-40%) if the potato by-products taken from the animal feed market are replaced with European animal feed or a 70% increase in GHG emissions if also imported soybean meals are used to replace the feed. Contrasting conclusions were also obtained for photochemical ozone formation. Conversely, the attributional and consequential LCAs agree on acidification, terrestrial eutrophication and depletion of fossil fuels. Although the consequential LCA was affected by higher uncertainties related to the determination of the actual product displaced, it allowed understanding the consequence of additional animal feed production. This process was not included in the system boundaries of the attributional LCA.

6.1 INTRODUCTION

The substitution of petrochemical fuels with low-carbon fuels is crucial for reducing anthropogenic greenhouse gas (GHG) emissions. Although the direct emissions from the aviation sector were responsible for only 2% of pre-covid-19 worldwide GHG emissions [331], the sector is expected to continue growing [332]. The role of biomass for the aviation sector is essential as there are limited options for decarbonization, especially in the next twenty years [333,334]. Unlike road transport, it is challenging to equip aircrafts with electric-powered engines for long distances [334]. Other alternatives such as hydrogen or other fuels based on renewable electricity and CO_2 are at the early stages of development [335]. Therefore, the decarbonization of aviation is still far away from practical implementation.

6

In the short term, the most promising option for GHG emissions reduction is using drop-in jet fuels from sustainable biomass feedstocks [333,334]. Bio-jet fuels can be blended with petroleum fuels and used in existing engines without modifications [47]. Despite that, to date, hydroprocessed esters and fatty acids (HEFA) is the only commercial technology available for bio-jet fuels [333]. However, HEFA bio-jet fuel production volumes are still considerably limited, accounting for less than 1% of total jet fuels worldwide [336]. The high production costs (2 to 6 times higher than conventional jet fuels) and strict sustainability requirements to be incentivized (60-65% GHG emissions' savings depending on the country) result in unfavorable industry development conditions (de Jong et al., 2015; IRENA, 2017; O'Connell et al., 2019). Currently, used cooking oil is the only alternative applied on industrial levels that is near cost-competitive and delivers low life cycle GHG emissions (O'Connell et al., 2019; Pavlenko et al., 2019). Nevertheless, the availability of used cooking oil is limited. Used cooking oil is also demanded for road and marine transport fuels and chemicals [128,341,342].

Many emerging bio-jet fuels have been recently certified by the American Society for Testing and Materials [343]. Among them, Fischer-Tropsch (FT) and alcohol-to-jet (ATJ) pathways are the production routes closest to commercialization [334].

For policy decision-making, it is crucial to assess the potential environmental benefits/impacts of bio-jet fuels compared to petrochemical jet fuels. Generally, these assessments are carried out using Life Cycle Assessment (LCA), a method for environmental assessments standardized by ISO 14041 and ISO 14040 [38,127]. This article investigates the environmental performance of an innovative ATJ route developed in the Netherlands to use local potato by-products from the food processing industry. This abundant low-price carbohydrate by-product is currently used (mainly) as animal feed but can be potentially transformed into valuable bio-based products such as biofuels, materials and chemicals [344–346].

Despite being a standardized approach, the LCA results can be affected by significant variability. The option of different methodological assumptions can steer the life-cycle environmental impact of a process/product, even when assessing the same fuel and feedstock [47,48]. A recent review of LCAs for HEFA biofuels [47] identified the method used to deal with multifunctional processes and the inclusion/exclusion of land-use change (LUC) emissions as the most important sources of variability. For example, the GHG emission intensity of a bio-jet fuel could increase by 2-3 times using either a different allocation method or making a different assumption for LUC [47]. Similar findings for bio-jet fuel pathways were also reported by Capaz et al. (2020). However, note that these issues also apply to other biofuels in general [45,46].

Multifunctionality practices and the inclusion of land-use changes depend on the specific LCA's goal and scope and consequently, the choice of modeling approach [45,347]. In the literature, two main modeling approaches are distinguished: attributional and consequential LCAs [244]. The attributional approach attempts to quantify the portion of global burdens associated with the specific products under assessment [59]. The environmental impact of such products is determined by analyzing the production system using representative average data [51,68]. Via attributional LCAs (ALCAs), the environmental impacts of each co-product are obtained by distributing the burden based on allocation parameters such as energy or market values [57,68,128]. In this way, the so-called additivity principle of ALCAs is respected (i.e. the sum of the attributional LCAs of all worldwide products corresponds with the total environmental burdens worldwide [59]. In attributional LCAs, looking at the status-quo and not to what has happened in the past or the future, LUC is generally not included. However, when LUC is included, only direct LUC is addressed and indirect LUC is not considered [45,63,64]. Consequential LCAs (CLCAs) focus on modeling the relative changes in the entire techno-sphere when the decision supported by the LCA is adopted. Hence, the CLCA provides information on both direct and indirect environmental burdens that occur due to the changes in demand for a product caused by such a decision [59]. Therefore, a CLCA allows assessing all the causal-effect relations within the market by changing product demand using marginal data [48,51,177]. In CLCAs, multifunctionality is addressed by a substitution approach (or often referred to as "displacement method"), and both direct and indirect LUC are included in the life cycle inventory [45,63,64].

While there is plenty of methodological literature regarding the effects of the modeling approach on the LCA result, the practice of applying both modeling approaches to the same product system before drawing recommendations about a product system is rare. No more than 8% of the peer-reviewed LCAs on bioenergy products in the Scopus database applied both modeling approaches to their case study [146]. Only 3% of peer-reviewed LCAs on bioenergy products declared to adopt a consequential modeling and the remaining studies used an attributional or unspecified modeling approach [146]. A recent review of the 100 most cited LCAs of bioenergy products highlighted that ambiguous results in these LCAs are mainly due to choices in the inventory modeling approach that are inconsistent with the goal of the study [94].

To our knowledge, there is only one peer-reviewed LCA applying both modeling approaches to a bio-jet fuel [348]. In their study, the CLCA resulted in an environmental benefit mainly due to the substitution of the power surplus. Their ALCA using allocation instead of substitution could not confirm such a benefit. While there is only one study applying both modeling approaches to a bio-jet fuel, there are several comparisons between the two modeling approaches for the case of animal feed [89,237]. Among them, a recent study by van Zanten et al. (2018) applied attributional and consequential modeling approaches to understand the environmental advantages of replacing soybean meal with an alternative protein source (peas or rapeseed meal) in pig diets. While their ALCA concluded that this practice could lead to environmental advantages, the CLCA concluded the opposite [349].

Given the lessons learned by the abovementioned literature regarding the effects of the modeling approach on LCA results, this study presents both attributional and consequential LCAs of this bio-jet fuel. In particular, the attributional LCA aimed to investigate the environmental impacts caused by the production of this jet fuel starting from the total environmental impact of the production system (i.e. the full pie). The attributional LCA of the bio-jet fuel attributes a piece of the pie to the bio-jet fuel. Technically, other practitioners could have a different aim leading to investigate one of the co-product of the same production system.

These attributional LCAs attribute the remaining shares of the pie to the other system's co-products. The sum of each functional output's environmental impact corresponds to the total impact of the product system (additivity of these ALCAs). Hence, the ALCA modeling serves to answer the research question: how much each of the functions, as entailed in the different product outputs, is responsible for the production system's total environmental impact? The company producing such bio-based products will be interested in consulting these LCAs. The results of these ALCAs allow understanding the environmental performance of the entire spectrum of bio-based products produced, usually benchmarking with their petrochemical counterparts, as well as possible process improvements.

While attributional modeling has a producer perspective, consequential modeling has a policy perspective instead. CLCA aimed to answer the following research question: how could introducing the new bio-jet fuel to the market potentially change the overall environmental impact of the supply chain of bio-jet fuel production and the economic sectors affected by the change? For example, the consequential LCA in our study aimed to understand the environmental impact of this novel bio-jet fuel at the net of the effects of displacing the potato by-products from the animal feed market.

6.2. MATERIALS AND METHODS

6.2.1. Goal and scope definition

Both attributional and consequential LCAs aimed to assess the environmental impact of a novel bio-jet fuel. Based on this goal, a functional unit of 1 MJ of bio-jet fuel was defined. The results of the two LCAs were compared with the environmental impact of conventional jet fuel (assumed kerosene). Such a comparison was used to evaluate if the two modeling approaches lead to similar findings regarding the environmental performance of this novel bio-jet fuel. This exercise aimed not only to provide a comprehensive picture of environmental impacts of the bio-jet fuel using a food processing by-product, but also to join the current LCA debate about the influence of the type of modeling approach used on the environmental performance of bio-based fuels and materials using by-products with low economic significance or waste as resources [45,146]. Hence, the intended audience of this LCA includes both technology developers and the LCA community.

Since this novel bio-jet fuel has been developed in the Netherlands from a local feedstock, the geographic scope of the LCA is the Netherlands. The temporal scope is the year 2030, when this fuel could be commercialized on a large scale.

A cradle-to-grave scope was adopted in the two LCAs of the bio-jet fuel. The respective process flow diagrams are shown in Figure 14. For both ALCA and CLCA, the common unit processes included are: acetone-ethanol-butanol (ABE) fermentation, swing adsorption, alcoholic condensation, hydrotreatment, distribution and combustion. The production of the aircrafts and the biorefinery plant and their decommissioning were neglected in both models. The exclusion of capital goods (including infrastructures) and their end of life is common practice for LCAs of biofuels in the EU. In fact, the well-to-wheel approach, which is the main tool adopted for the EU biofuels, excludes the production of the vehicles and plants and their decommissioning from the LCA scope [9,37]. The reason is that the contribution of capital goods on the total life cycle of fuels is expected to be small and very similar for biofuels and conventional fuels [36,350]. The exclusion of capital goods from this LCA is also in line with the update of the EU Product Environmental Footprint (PEF) recommending the exclusion of capital goods from the LCA "unless there is evidence from previous studies that they are relevant" [351].

In the attributional LCA, the flow diagram starts with potato cultivation and the potato processing industry generating the potato by-products. In the CLCA modeling, the unit processes representing the production of the feedstock (cultivation and potato industry) are not included because they are not affected by the change that the CLCA aims to assess. In fact, if the potato by-products were not used for this application, they would be produced anyway. The use of potato by-products for this application changes neither the raw potatoes inputs nor the utilities utilized by the factory per tonne of the main food product. Hence, introducing this bio-jet fuel as a substitute for petrochemical kerosene (i.e. the change assessed) does not affect these processes.

In the Netherlands, the potato by-products that can be used for this fuel are made of potato peels (80%), grey starch (15%) and press-pieces (5%) [347]. Most potato by-products (potato peels and potato pieces) are currently used in animal feed. Grey starch might be used for other purposes [352]. Since grey starch represents only 15% of potato by-products, we can assume that taking away the potato by-products used as feed affects mainly the animal feed supply. If the animal feed demand is to be maintained, additional feed needs to be supplied from other sources. For this reason, the flow diagram of the CLCA includes the unit process named “marginal animal feed production”, representing the production of such additional animal feed from other sources.

Furthermore, attributional modeling was also used to determine the environmental impact of the co-products in the value chain, namely animal feed (from fermentation residue), bio-based hydrogen, bio-based carbon dioxide and biolubricants. In this way, it is possible to transparently present how the environmental impact of each process contributes (i.e. is allocated) to the environmental impact of each function delivered by the system. The impacts between the main product (jet fuel) and these by-products are partitioned by physical or economic relations. The allocation choices between the co-products can be found in the next section. For CLCA, system expansion is applied to these by-products. The by-products are assumed to replace the marginal productions of the same or similar products (see Section 6.2.2); therefore, the environmental impacts of the individual co-product cannot be quantified.

In the ALCAs, the environmental impacts of each co-products were compared with their current market benchmarks. This comparison is in the interest of the producer of such bio-based products. The company wants to avoid burden-shifting between their co-products i.e. producing one bio-based product with a good environmental performance at the expense of a bad environmental performance for the other bio-based products produced. In case of opposite conclusions between the two LCAs of the bio-jet fuel for certain impacts, it is possible to use the results of the other ALCAs to immediately understand if the main reason is linked to the allocation method. For example, we could notice if most of the environmental impact in that category is caused by a certain unit process and was allocated mainly to a co-product instead of the bio-jet fuel. If this is not the case, we can exclude this effect of allocation and look for other reasons not linked with the allocation applied in the ALCA. For example, a reason can be the inclusion of a different process in the system boundaries (e.g. animal feed displacement effect) or marginal versus average data.

The selection of the market benchmarks for the ALCAs and the substituted products in the CLCA can be found in sections 6.2.2.5 and 6.2.2.7).

For the attributional LCAs of the bio-based co-products of the jet-fuel, the functional units were defined as 1 kg of each product, i.e. 1 kg of animal feed or 1 kg of hydrogen or 1 kg of carbon dioxide or 1 kg of lubricants (see Table 16 and Table 17 for the actual production ratios). Since these co-products are delivered at the bio-jet fuel production process gate and are usually not combusted, their LCA follows a cradle-to-gate scope. Given the additivity principle of ALCA, summing the environmental impact of the functional units of all ALCAs conducted leads to the total environmental impact of the system shown in Figure 34 on the left.

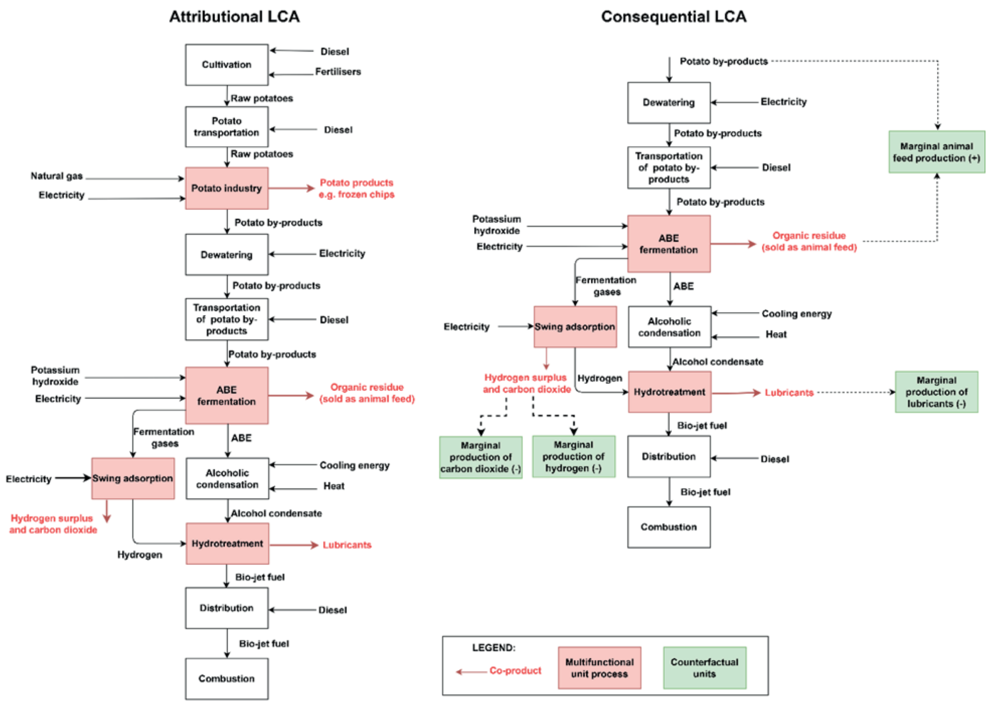

Figure 34. Cradle-to-grave process flow diagram detailing inputs and co-products for attributional (left) and consequential (right) LCAs of the investigated bio-jet fuel.

Five midpoint impact categories were selected (see Table 14). Besides climate change and depletion of fossil fuels, which are priority indicators in the current environmental decision-making [308], other four impact categories were considered. Among them, photochemical ozone formation, acidification and terrestrial eutrophication are recognized as key environmental problems linked to nitrogen oxides in the Netherlands [353]. These three categories are acknowledged as important impacts for bio-based products' policy and investment decisions. Previous work showed that a fair comparison with petrochemical products with reasonable uncertainties is possible for these five impact categories [43,354]. As for the sustainability assessment of bio-based fuels used in the European Union [9,37,350], the direct biogenic carbon dioxide emitted to the atmosphere from the combustion biofuels was considered carbon neutral. Based on the European legislation for biofuels and supporting documents (mentioned above), the only exception regards the consequences of land use changes on the soil organic carbon (biogenic). Accordingly, the carbon removal from the original biomass matter was neglected except for net effects related to LUC (for details regarding the data used for LUC modeling, please refer to section 6.2.2.6.

Table 14. Environmental impact categories and impact assessment models.

Impact Category	Unit	Impact assessment models
Climate change	kg CO_2eq	[310]
Photochemical ozone formation	kg NMVOC eq	[312]
Acidification	molc H+ eq	[313]
Terrestrial eutrophication	mol N eq	[355]
Depletion of fossil fuels	MJ	[315]

6.2.2. Life cycle inventory

6.2.2.1 Feedstock production (ALCA) and transportation

Potato by-products are a low economic significance by-product of the potato processing industry. In the flow diagram of the ALCA (see Figure 34), the product system starts with the cultivation of potatoes. After the raw potatoes are harvested, they are sent to the potato industry to be processed into food products. The main inputs to the potato industry are electricity and heat. The quantities are reported in

Table 15. Economic allocation was applied to the potato industry unit process (for details, see section 6.2.2.4).

The dewatered potato by-products are then transported by lorry (16-32 t) to the refinery, which is assumed in Rotterdam. Based on the location of potato processing factories in the Netherlands, the potato by-products need to be transported on average for 105 km [347].

Table 15. Mass and energy inputs of the potato industry per tonne of potato by-products and background data sources for these inputs. Processed potatoes output based on confidential data from the industry and not disclosed.

Activity level data	**Quantity per tonne of potato byproduct**	**Foreground data sources**	**Background data**
Inputs			
Raw potatoes, wet (t)	4.6	[356]	Agri-footprint 5.0 (Potatoes, market mix, at regional storage/NL) This dataset already includes the average transportation of potatoes in the Netherlands.
Heat (GJ)	12.4	[356]	Heat, district or industrial, natural gas {Europe without Switzerland}| heat production, natural gas, at boiler condensing modulating >100kW | APOS, from ecoinvent 3.6.
Electricity (kWh)	558.9	[356]	For electricity, the dataset Electricity, medium voltage {RER}| market group for | APOS from ecoinvent 3.6 was used updating the shares of electricity by fuel mix based on the 2030 EU reference scenario for the Netherlands [272].

6.2.2.2 From potato by-product to bio-jet fuel

At the refinery location, the potato by-products are processed via ABE fermentation, which produces a mix of valuable alcohol/ketone made of butanol and acetone with a minor fraction of ethanol. Table 16 reports on the ABE fermentation process's inputs and output, which uses pervaporation for in-situ butanol recovery, increasing the ABE yield [357,358].

The mix of ABE alcohols is then processed through alcohol condensation [360], avoiding an important part of energy consumption to separate the alcohols and acetone to purity [357,361]. The water produced by alcohol condensation is recycled back to a second pervaporation membrane for water separation, which needs to be treated before being discharged to the environment. In parallel, the fermentation gases (a mix of carbon dioxide and hydrogen) are separated via swing adsorption, with 344 kWh of electricity consumed per t of carbon dioxide separated [362]. The alcoholic condensate is then further deoxygenized via hydrotreatment using part of the separated bio-hydrogen [347,360]. Table 17 reports on the inputs and outputs of the thermochemical upgrading to bio-jet fuel.

Table 16. Mass and energy inputs and outputs of ABE fermentation (including in-situ recovery pervaporation) per t pure ABE from [347] *and background data sources for the LCA.*

Flow	Amount	Comment/ background data
Inputs		
Potato by-products (t) wet	46.6	As the output from the potato processing industry. Corrected based on dry base content for the data source [356].
Enzymes (kg)	2.2	Electricity (1.9 kWh per kg of enzyme) and steam (4 MJ per kg of enzyme) to produce α-amylase enzymes based on [359]. For electricity in the attributional modeling, the dataset Electricity, medium voltage {RER}| market group for | APOS from ecoinvent 3.6 was used to update the shares of electricity per source based on the 2030 EU reference scenario for the Netherlands [272]. For electricity in the consequential modeling, Electricity, medium voltage {NL}| market for | Conseq was used. For steam, the dataset Heat, from steam, in chemical industry {RER}| market for heat, from steam, in chemical industry | APOS or Conseq from ecoinvent 3.6 was used.
Potassium hydroxide (t)	0.1	Potassium hydroxide {RER}| production | APOS or Conseq from ecoinvent 3.6.
Electricity (kWh)	1000	For electricity in the attributional modeling, the dataset Electricity, medium voltage {RER}| market group for | APOS from ecoinvent 3.6 was used to update the shares of electricity per source based on the 2030 EU reference scenario for the Netherlands [272]. For electricity in the consequential modeling, Electricity, medium voltage {NL}| market for | Conseq was used. The marginal electricity dataset, which was based on 2014 data, was not modified since marginal technologies are more stable in time than data based on average technologies [211].
Nitrogen (kg)	2.8	Nitrogen, liquid {RER}| market for | APOS or Conseq from ecoinvent 3.6
*Co-outputs (*Details about allocation are reported in section 6.2.2.4)		
Organic residue (t)	3.5	Sold as animal feed.
Part of fermentation gases made of hydrogen (t)	0.05	Partly used for hydrotreatment while the surplus is sold.
Part of fermentation gases made of biogenic carbon dioxide (t)	1.6	Sold.

Table 17. Mass and energy inputs and outputs of the thermochemical upgrading (alcohol condensation plus hydrotreatment) per t of bio-jet fuel from (Moretti et al., 2021) and background data sources for the LCA.

Flow	Data	Background data
Inputs		
Pure ABE	1.5 t	
Heat	4.6 GJ	Heat, district or industrial, natural gas {RER}| market group for | APOS or Conseq from ecoinvent 3.6.
Cooling energy	1.7 GJ	Cooling energy {CH}| from natural gas, at cogen unit with absorption chiller 100kW | APOS or Conseq from ecoinvent 3.6.
Bio-hydrogen	36.5 kg	From swing adsorption.
Co-outputs		
Lubricants	41.7 kg	Mass allocation. Details in section 6.2.2.4.
Wastes		
Wastewater	0.6 t	Wastewater, average {Europe without Switzerland}| market for wastewater, average | APOS from ecoinvent 3.6

6.2.2.3 Distribution and combustion

For the distribution of the bio-jet fuel and petrochemical kerosene to the "tanks", a distance of 65 km was assumed, representing the transportation from Rotterdam to Amsterdam. As background data, the process Transport, freight, lorry >32 metric ton, EURO4 {RER}| transport, freight, lorry >32 metric ton, EURO4| APOS or Conseq from ecoinvent 3.6 was used. The technology under assessment is similar to the ATJ-synthetic paraffinic kerosene pathway where isobutanol or ethanol are upgraded to jet fuel via alcohol dehydration and condensation reaction followed by hydrotreatment [363]. For this reason, the combustion emissions for the bio-jet fuel were assumed to be the same as ASTM-certified synthetic paraffinic kerosene. The carbon dioxide from the bio-jet fuel is biogenic and was assumed carbon neutral as currently assumed in biofuels' sustainability calculations according to the EU legislation [9,37]. The inventory for distribution and combustion can be found in Table 18. The lower heating value (LHV) of the bio-jet fuel was assumed to be 44.1 MJ/kg, which is typical of synthetic paraffinic kerosene [364]. An LHV of 43.2 MJ/kg was assumed for petrochemical kerosene [364]. Combustion emissions were based on a single-aisle passenger aircraft operating on an average distance and with an average payload for both jet fuels. With these assumptions, the so-called payload fuel energy intensity is 8.62 $kJ/kg_{payload}$-$km_{great\text{-}circle\ distance}$ for the bio-jet fuel [364] and 8.65 $kJ/kg_{payload}$-$km_{great\text{-}circle\ distance}$ for conventional jet fuel.

Table 18. Inventory of combustion emissions per kg of jet fuel retrieved from [364]

Emissions to air	Data (grams/kg) for bio-jet fuel	Data (grams/kg) for petrochemical kerosene
CO_2	3100	3151
CO	4.0	4.0
N_2O	0.01	0.01
NO_x	14.5	15.5
SO_x	0	1.4
CH_4	0.005	0.005

6.2.2.4 Multifunctionality in the attributional model

In attributional modeling, the allocation method was based on each unit process's "causality mechanism" as recommended by ISO 14044:2006. The purpose of the potato industry is to produce revenues selling potato food products. Hence, an economic allocation was used. Based on [356], approximately 1% of the environmental impact of the potato industry was allocated to the potato by-products (based on the five-year average price).

For the ABE fermentation process, energy allocation was applied because the process is driven to produce an energy product, even though the fermentation residue used for animal feed has a higher mass content compared to the alcohols. The following allocation factors were retrieved from [347]: 34.4% to ABE, 6% to the fermentation gases and 59.6% to the animal feed.

The swing adsorption process aims to remove the fermentation gases continuously and separate carbon dioxide and hydrogen. Between these two co-products, carbon dioxide is more significant from both economic and mass perspectives. Given that this unit process's main product is not an energy product, economic allocation was also applied for this multi-output process. As a result, allocation shares of 59.4% and 40.6% were calculated respectively for carbon dioxide and hydrogen [347].

Together with the bio-jet fuel, the hydrotreatment process also delivers a minor co-product (i.e. lubricants). The goal of the hydrotreatment process is to generate bio-jet fuel (energy). Energy allocation should be applied to the hydrotreatment process. Since the lower heating value of the lubricants is unknown, a mass allocation was used as a proxy for energy allocation resulting in 4% of the environmental impact allocated to lubricants. Since lubricants are a minor product, the allocated environmental impact to each t of fuel is only slightly affected by the applied allocation method. Conversely, the impact of the lubricants is more affected by the allocation method based on how different is the actual lower heating value of the lubricants compared to the one of the bio-jet fuel.

6.2.2.5 Multifunctionaility in the consequential model

6.2.2.5.1 By- and co-products

The consequential modeling included avoiding the production of all bio-jet fuel's co-products in the system boundaries through the so-called system expansion by substituting marginal production. For most commodity products, the marginal production data were taken from the Ecoinvent Consequential datasets. Lubricants were substituted with Lubricating oil {RER}| production | Conseq. The surplus of hydrogen was substituted with Hydrogen (reformer) E from PlasticsEurope since nowadays hydrogen production mainly comes from steam reforming of natural gas [365]. The current market demand for carbon dioxide is mainly for the promotion of plant growth, the creation of inert environments or as a heat transfer medium, as a refrigerant, or as a chemical for the production of a variety of other chemicals [366,367]. For these markets, commercial carbon dioxide is obtained as a "waste gas" from ammonia and hydrogen production processes [366,367]. In the ammonia production process, an intermediate mix of gases made of hydrogen, nitrogen and carbon dioxide is produced. However, only the first two gases are required to make ammonia. So, carbon dioxide needs to be removed from the gas stream. A gas with a similar composition that needs to be separated is also generated in hydrogen production from reforming natural gas.

The impact associated with CO_2 production is led by extraction and purification of the abovementioned "waste" gas (that comes burdens-free) [367]. In our LCA, it was assumed that the CO_2 is extracted and purified from the waste gas of ammonia production using monoethanolamine (MEA), which is a common practice [367,368]. Based on [368], 3.56 MJ of heat are necessary to separate 1 kg of CO_2 from waste gas from ammonia production using MEA as extraction solvent. For heat from natural gas, the process *Heat, district or industrial, natural gas {Europe without Switzerland}| heat production, natural gas, at industrial furnace >100kW | Conseq* from ecoinvent 3.6 was used. The amount of MEA, losses of MEA to air and water flows of carbon dioxide separation using MEA were retrieved from ecoinvent

3.6 dataset *Carbon dioxide, {RER}| production | Conseq*. This modeling could be considered also a good proxy for the purification of carbon dioxide using MEA from gases from other industrial processes and not only from ammonia production. The electricity input for carbon dioxide liquefaction was not considered since the carbon dioxide delivered by the swing adsorption process is in a gas state.

6.2.2.5.2 Additional animal feed to replace potato by-products

Concerning the additional animal feed production, it was necessary to identify the mix of marginal technologies that will fulfill such marginal demand. A decision tree is typically used to determine the marginal technologies, i.e. the technologies affected by a small change in market demand [211]. Since the additional production of animal feed affects a market more than a specific process, it is necessary to understand the trend in the volume of the affected market [211].

Potato peels are a balanced feed with good fibre, starch and protein contents [369]. The dry content and starch content (on a dry basis) of this mix of potato by-products were measured as 12.3% and 55.7%, respectively [347]. It could be argued that potato peels are also used to provide calorific values of the feed for its high content of starch. Due to commercial confidentiality, it is not possible to disclose the exact feed markets of potato peels supplied by the specific potato processing industry in the project. We used generic feed market information about potato peel-based feed to determine the types of displacement feed. In the baseline analysis, the protein content is used to quantify the equivalent function of different types of animal feeds because the nutrient value of feed is determined by the protein content.

The protein content (on a dry basis) was measured only for the sample of potato peels as 16.0%. In this study, the protein content of potato peels (being the main fraction) was assumed for the entire mix of potato by-products. Potato by-products are a minor amount in the Dutch animal feed market currently used mainly for pigs [370,371] and cattle [369,372]. In the Netherlands (and nearby countries), the compound feed production for pigs is declining and for cattle is stagnating [373,374]. Therefore, there is a potential to provide a marginal increase in production since the production system is not saturated, and thus the current market is the marginal technology. The animal feed produced in Europe is made mostly of grains produced in Europe (71%) and imported oil meals (24%) [373]. The grains are mostly made of wheat (32%), corn (30%) and barley (25%), while soybean meal makes 58% of all the oil meals used in the EU and is mainly imported from Argentina (58%) and Brazil (33%). These major crops were considered to build the marginal market that can be found in Table 19. The sensitivity of the choices made the marginal feed production is discussed in Section 6.4.2.

Table 19. Animal feed actual/average (used for the comparison conducted in the ALCA) and marginal market proportions (used for the CLCA) based on [373] and background sources; n.a. = not applicable.

Flow	**Actual/ Average market (%)**	**Marginal (%) market based on the rescaling of the major crops**	**Amount assumed (kg) for the marginal animal feed modeling in the CLCA***	**Background data sources for ALCA (benchmark used for comparison) and CLCA (marginal animal feed used in system expansion)**
Barley grain (EU)	17.8%	23.9%	1.5	Barley grain {DE}| barley production | APOS or Conseq
Corn gran (EU)	21.3%	28.6%	1.8	Maize grain, Swiss integrated production {CH}| production | APOS or Conseq
Wheat grain (EU)	22.7%	30.5%	1.9	Wheat grain {DE}| wheat production | APOS or Conseq
Other grains	9.2%	0%	0	Oat grain {FI}| oat production | APOS
Total grains	71%	83%	5.2	n.a.
Soybean meal (AR)	8.1%	10.9%	0.7	Soybean {AR}| soybean production | APOS or Conseq. Soybean production used as proxy for soybean meal.
Soybean meal (BR)	4.6%	6.2%	0.4	Soybean meal {BR}| market for soybean meal | APOS or Conseq
Other oil meals	11.3%	0%	0	Rapeseed, at farm/NL Economic from Agri-footprint 5.0. Rapeseed production as a proxy for rapeseed meal.
Total oil meals	24%	17%	1.1	n.a.
Neither gains nor oil meals	5.0%	0%	0	Neglected

** to obtain 1 kg of total protein considering the dry matter and protein content of the dataset used as background data.*

6.2.2.6 Land use changes

There are still many concerns around the sustainability of biofuels' production; many of these are related to direct or indirect carbon stock changes from land-use transitions [375]. The induced direct carbon stock changes from bio-jet fuel production are proven to play a critical role in their environmental performance [376]. Simultaneously, the role of carbon stock changes becomes increasingly relevant when the impact on other lands from the displacement effect of biomass production is accounted for (indirect land-use change) [377]. Therefore, it is paramount to include such direct and indirect effects of land-use change (iLUC) from biofuels systems.

In our study, LUC impacts were retrieved from the Ecoinvent database (version 3.6) and Agri-footprint database (version 5.0). Under these databases, LUC is assessed following the methodological principles recommended by PAS2050 [167]. A 20-year time horizon for carbon pools to reach equilibrium is considered [378]. Carbon stock changes are addressed over the four main carbon pools [379]: aboveground biomass, belowground biomass, dead organic matter and soil organic carbon. Note that the LUC Ecoinvent model follows the WFLDB Quantis adapted version (country-level perspective) of the Blonk tool to assess direct LUCs [380]. The LUC model of Agri-footprint 5.0 used for the direct land-use change of potato product relies on the Blonk tool [380].

To illustrate, carbon stock change effects from potato production within the same country are accounted for directly as they occur on the same land as the potato production land use. In the ALCA model, no indirect effects are generated from the current potato production as there is no effect of using land already dedicated for this purpose.

In contrast, in the CLCA model, animal feed production's displacement effects outside the production country's boundaries are accounted as indirect. The upstream displacement effects of the potato by-products attributed to the marginal animal feed production are assumed with a direct casual-effect relationship. Consequently, the displacement effect from the marginal feed (corn, wheat, barley, and soybean) production and derived carbon stock changes in the EU, Brazil and Argentina are attributed 100% to this displacement effect and accounted for entirely in this supply chain. The data of carbon stock exchanges of the marginal feed produced in the EU, Brazil and Argentina are obtained from ecoinvent version 3.6.

6.2.2.7 Petrochemical kerosene and other reference products

For petrochemical kerosene, the dataset *Kerosene {Europe without Switzerland}| kerosene production, petroleum refinery operation* from the libraries of ecoinvent 3.6 named APOS and Conseq respectively for the ALCA and CLCA. This dataset represents the European production of kerosene at the factory gate of the oil refinery. Distribution and combustion were modeled as detailed in section 6.2.2.3. The process flow diagram detailing inputs and co-products for petrochemical kerosene can be found in the appendix.

For the other co-products, the comparison was conducted with the following reference products:

- for hydrogen, the dataset *Hydrogen (reformer) E from PlasticsEurope* [381];
- for carbon dioxide, the inventory was modeled as illustrated for the marginal carbon dioxide using average data i.e. 1) assuming that 3.56 MJ of heat are necessary (*Heat, district or industrial, natural gas {Europe without Switzerland}| heat production, natural gas, at industrial furnace >100kW | APOS* from ecoinvent 3.6) and 2) retrieving the amount of MEA, losses of MEA to air, water flows of carbon dioxide separation using MEA from the ecoinvent 3.6 dataset *Carbon dioxide, {RER}| production | APOS*.
- for lubricants, the dataset *Lubricating oil {RER}| production | APOS* from ecoinvent 3.6;
- for animal feed, the average European mix has been considered and can be found in the second column of Table 19.

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6.3. RESULTS

The results section is structured as follows. First, the environmental impact hotspots of the ALCA of the bio-jet fuel are presented. Second, the environmental hotspots of the CLCA of the bio-jet fuel are presented. Third, the environmental impacts of the bio-jet fuel assessed with both modeling approaches are compared to petrochemical kerosene. Afterward, the results of the ALCAs of the bio-based co-products are presented, along with a comparison with market benchmarks. The last section of the Results (section 6.3.4.4) reports on the “attributional pie”, showing how much each co-product contributes to the environmental impact of the entire product system.

6.3.1 ALCA: identification of the environmental hotspots

The overall breakdown of the cradle-to-grave environmental impact of 1 MJ of bio-jet fuel is illustrated in Figure 35 (numerical results can be found in the appendix).

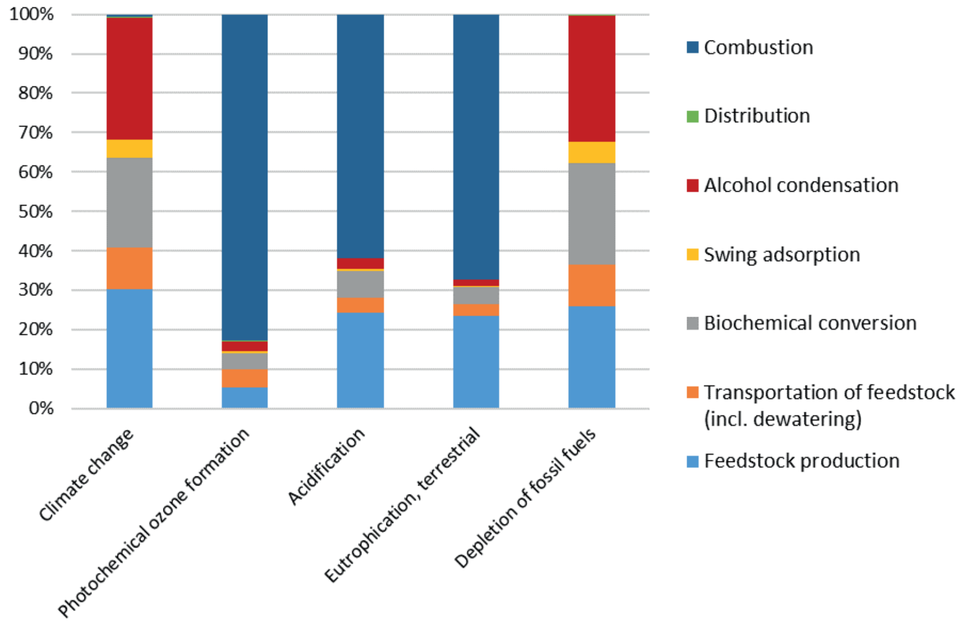

Figure 35. Breakdown of the cradle-to-grave attributional environmental impact of 1 MJ of bio-jet fuel per key-unit process.

From Figure 35, it can be observed that feedstock production is a relevant environmental hotspot (23-30% of the cradle-to-grave environmental burden) in all categories except for photochemical ozone formation. Both biochemical conversion and alcohol condensation generate about 30% of climate change and depletion of fossil resources, adding up to 60% of the environmental impact for these two categories. The bio-jet fuel combustion dominates the photochemical ozone formation, acidification and terrestrial eutrophication impacts (60-80% of the cradle-to-grave impact in these categories).

Heat from natural gas used for feedstock production contributes to 45% of the impact share for climate change and 56% for the depletion of fossil fuels. The impact of electricity for feedstock production is relatively small for all categories (less than 18%) and minor (3%) for acidification. Conversely, for all categories, raw potato production represents a significant impact share of feedstock production. This share ranges from 25% in depletion of fossil fuels to up to 94% in acidification. The main environmental impact sources from raw potato are the cultivation stage and their transport to the processing industry. The transport process is responsible for 20% of climate change impact and 35% of fossil fuel depletion. For climate change, 45% of the impacts in the cultivation stage are caused by dinitrogen monoxide emissions to air. These emissions result from applying fertilizers, manure and crop residues. In addition, 20% of the impact is caused by diesel use in agricultural machinery and 14% by calcium ammonium nitrate production. The acidification and terrestrial eutrophication from feedstock production are related to ammonia emissions from fertilizer application. The diesel burned in agricultural machinery also represents 38% of the depletion of fossil fuels caused by feedstock production. Regarding the small impact (5%) of the feedstock production on the cradle-to-grave photochemical ozone formation, it is mainly caused by the transportation of the potatoes to the processing food industry and by the diesel burned in the agricultural machinery during the cultivation phase. The direct carbon stock change caused by the cultivation of potatoes processed in the Netherlands has a negligible impact since there is no displacement effect. Long-term established agricultural land is used in the EU for the production of potatoes. Thus, the carbon stock changes within this land are relatively small as the land is already dedicated for agricultural purposes.

In all categories, biochemical conversion is important and has high impact especially for climate change and fossil fuel depletion. The environmental impact of this process is caused mainly by the production of electricity (44-72%) and potassium hydroxide (26-51%). The impact of alcohol condensation represents 30% of climate change and depletion of fossil fuels, respectively, and 12% of photochemical ozone formation. For these impact categories, the main sources of impact are the production of steam (55-56%) and cooling energy (43-45%). The production of steam impact is related to the combustion of natural gas. Swing adsorption impact is minor and generated by the allocated electricity (1-5%). Since hydrogen is the only consumable of hydrotreatment and comes from a closed-loop flow, hydrotreatment has no impact. The environmental impact of hydrogen production is already accounted for in the biochemical conversion and the swing adsorption process.

Feedstock transport is an important environmental hotspot for climate change (9%), photochemical ozone formation (18%) and use of fossil resources (9%), mainly due to the production and combustion of diesel. The distribution of the bio-jet fuel to the tank has a negligible contribution to the cradle-to-grave environmental burden in all categories. The combustion of the bio-jet fuel releases nitrogen oxide emissions resulting in high photochemical ozone formation, acidification and terrestrial eutrophication.

6.3.2 CLCA: potential environmental impacts due to changes in demand

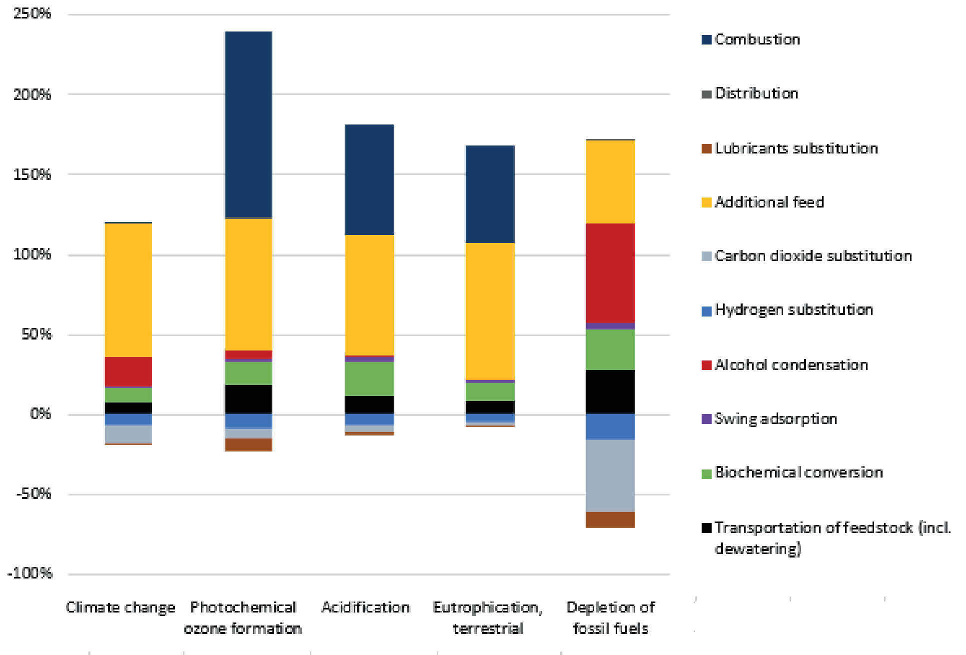

Figure 36. Breakdown of the cradle-to-grave consequential environmental impact of 1 MJ of bio-jet fuel per key-unit process. Numerical results can be found in the appendix (see Table 20 for LUC contribution).

Figure 36 shows the breakdown of the cradle-to-grave CLCA. Similar to the results from the ALCA, the environmental impacts caused by the utilities in the biochemical conversion are high in most of the impact categories. Similarly, the impact of alcoholic condensation is high in climate change and depletion of fossil resources (marginal data did not change that). The environmental impact caused by the combustion of the bio-jet fuel remains important also for the CLCA for photochemical ozone formation, acidification and terrestrial eutrophication.

Different from ALCA, the consequential modeling leads to the result that the additional amount of animal feed is one of the most important contributors of impact for all impact categories. The impact of the additional animal feeds can be divided into biomass production and LUC (see Table 20).

Table 20. Breakdown of the environmental impacts of the additional feed needed to replace the potato by-product, contributed by biomass (crop) production and land use changes.

Impact category	**Biomass (crop) production**					**LUC**		**Total**
	Barley grain (EU)	**Maize grain EU**	**Soybean from Argentina**	**Soybean from Brazil**	**Wheat grain EU**	**Soybean from Argentina**	**Soybean from Brazil**	
Climate change (%)	11	9	3	4	16	46	11	100
Photochemical ozone formation (%)	18	16	17	9	23	13	5	100
Acidification (%)	10	52	12	4	14	6	2	100
Eutrophication, terrestrial (%)	18	40	5	0	27	7	2	100
Depletion of fossil fuels (%)	26	21	12	2	36	0	0	100

For biomass production, producing fertilizers and resulting field emissions from their application are the major environmental impacts. For barley, the impact of producing nitrogen fertilizers is high in all categories, ranging from 32% for photochemical ozone formation to 79% for acidification. Similarly, for wheat grain, the production of nitrogen fertilizers contributes between 39% (photochemical ozone formation) and 93% (acidification) of the environmental impact. Tillage is also an important source of environmental impact for barley in three categories i.e. photochemical ozone formation (26%), acidification (19%) and depletion of fossil fuels (13%). The impact of tillage is also high in these categories for wheat grain (but with slightly lower percentages for wheat grains). For maize production, direct field emissions cause 33% of climate change due to dinitrogen monoxide and carbon dioxide released into the air. They also cause 86% of acidification and 80% of terrestrial eutrophication (80%), mainly due to ammonia to air.

The effects of LUC are relevant for climate change, photochemical ozone formation, acidification and terrestrial eutrophication. The LUC impacts are attributed primarily

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to soybean production. LUC is responsible for 94% and 74% of the climate change impacts of Argentinian and Brazilian soybeans, respectively. The LUC climate change impact of the Argentinean soybeans is higher than that of the Brazilian soybeans, given the larger share of high carbon stock lands converted to arable land. For Argentina, the land-use transition to arable land involves mainly removing forest land, from which 85% is secondary and 8% is primary. A lower share of forest removal characterizes the land-use transition in Brazil, with 75% clear-cutting of primary forest. In addition, 5% of the land-use transition is attributed to the removal of grasslands. Grasslands contain considerably lower carbon stocks than forests.

For photochemical ozone formation, 18% of the impact of marginal animal feed is caused by LUC. Of this 18%, 13% is caused by clear-cutting of secondary forest to arable land in Argentina and 4% by clear-cutting of primary forest to arable land in Brazil while the remaining 1% by clear-cutting of primary forest to arable land in Argentina. For clear-cutting of secondary forests in Argentina (and similarly for Brazil with slightly different shares), the photochemical ozone formation impact is caused by air emissions of ethene (29%), nitrogen oxides (21%), formaldehyde (11%) and propene (9%) due to the burning of vegetation, for 7% by diesel combustion in tractors and for 5% by petrol combustion in power sawing.

The small acidification and terrestrial eutrophication impacts caused by LUC is mainly due to Argentinian soybeans (see Table 20). More than 90% of acidification and terrestrial eutrophication related to Argentinian soybeans' LUC is due to the clear-cutting of secondary forests converted into arable land. The acidification due to such clear-cutting is caused by ammonia emissions to air (61%) and nitrogen oxides (26%) due to the burning of vegetation. For terrestrial eutrophication, these percentages become 64% for ammonia emissions and 34% for nitrogen oxides.

The marginal production of carbon dioxide (given the high amount of carbon dioxide produced) is the main environmental credit for climate change, depletion of fossil fuels and photochemical ozone formation. In these three categories, the credit is mainly caused by avoiding the production and combustion of natural gas with smaller credits for the avoidance of the production of the MEA solvent used for carbon dioxide separation.

The credit for substituting the surplus of hydrogen is lower than the one for carbon dioxide in climate change and depletion of fossil fuels, because the quantity of the surplus H_2 is much smaller than that of CO_2.. Nevertheless, the surplus H_2 offers environmental credits for photochemical ozone formation (98% caused by avoided nitrogen oxides emissions to air from fuel combustion) and acidification (70% nitrogen oxides and 30% sulfur dioxide to air from fuel combustion).

However, in the future, the production of hydrogen and carbon dioxide and other chemical products might become less carbon-intensive (see section 6.4.1 and section 6.4.2 for discussion regarding the uncertainties on future benchmark technologies).

The credit for the substitution of lubricants is high only for photochemical ozone formation and depletion of fossil fuels. 90% of such credit is caused by the avoidance of direct air emissions of non-methane volatile organic compounds (NMVOC) during the production of the lubricants. For depletion of fossil fuels is caused by avoided production of crude oil (75%).

6.3.3 Comparison with petrochemical kerosene

As shown by Figure 37, the environmental performance of the bio-jet fuel compared to conventional jet fuel is very different depending on the LCA modeling selected.

Based on both ALCA and CLCA results, the bio-jet fuel assessed offers a lower fossil fuel depletion impact compared to petrochemical kerosene (55% reduction based on ALCA and 49% based on CLCA).

For climate change, the ALCA and CLCA models lead to different conclusions. The ALCA model shows benefits for the bio-jet fuel (58% lower impact), whereas the CLCA calculated a 68% higher climate change impact than conventional jet fuels. In the CLCA model, LUCs due to the additional animal feed production is influential for climate change (47% of the cradle-to-grave climate change) but not significant for the depletion of fossil fuels (see Table 20).

For photochemical ozone formation, the differences between the bio-jet fuel and the petrochemical jet fuel are insignificant (1% difference) based on the ALCA model, whereas based on the CLCA model, a 50% higher impact is observed for the bio-jet fuel compared to petrochemical kerosene. In the CLCA model, the credits for photochemical ozone formation from co-products substitution are only 30% of the impact from the additional production of animal feed. Moreover, the impact of the transportation of potato by-products becomes 3 times higher in all categories (with slightly different percentages 2.9-3.1 times given the shift from average to marginal data). The reason is twofold: 1) this impact is no more allocated to the co-products but entirely apportioned to the bio-jet fuel and 2) this unit process does not have co-products to substitute.

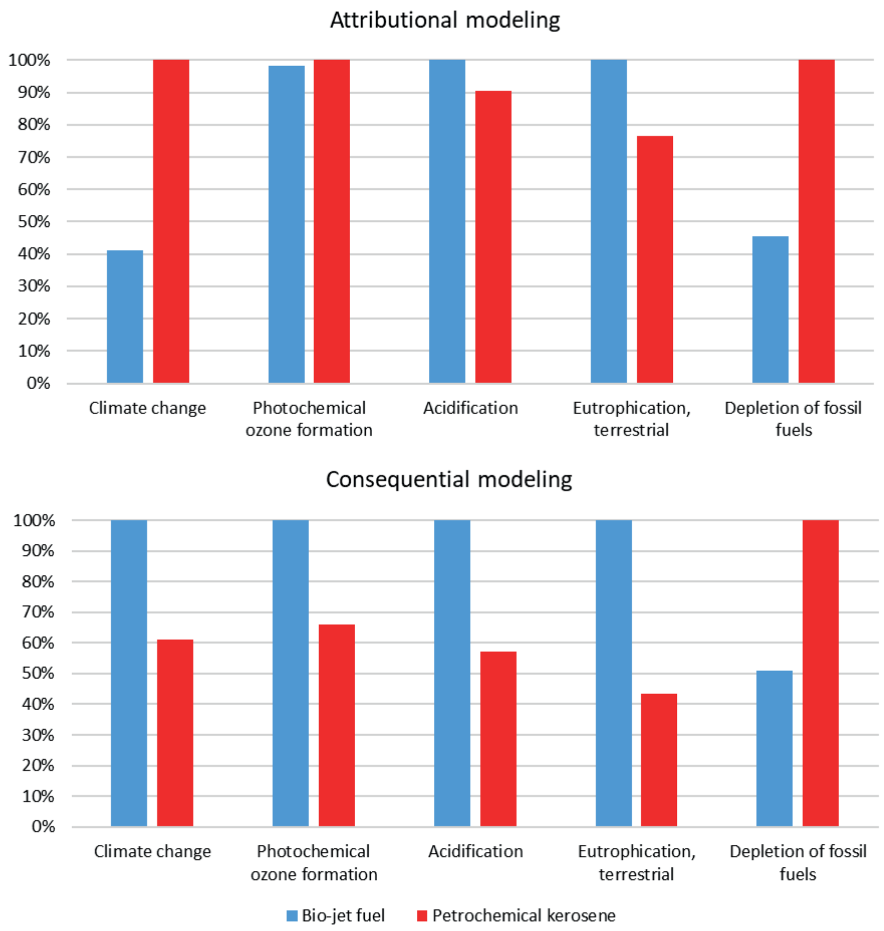

Figure 37. Comparing the cradle-to-grave environmental impacts of 1 MJ bio-jet fuel from potato by-products: the attributional model (top) and the consequential model (bottom). Characterized impacts can be found in the appendix.

The bio-jet fuel has a higher impact on acidification than the petrochemical jet fuel (10% in ALCA and 75% higher in CLCA). In the CLCA model, the benefits from co-product substitution do not compensate for the impact of the additional animal feed production i.e. the benefits are only 17% of the additional impact from animal feed production). Similar patterns are observed for terrestrial eutrophication between the results of ALCA and CLCA.

Based on the comparison of the results obtained with CLCA and ALCA approaches (see Figure 37), it can be concluded that the recommendations for producing this bio-jet fuel would be very different depending on the approach used with respect to climate change and photochemical ozone formation impacts. In fact, for these categories, the bio-jet fuel has a significantly higher impact once the additional production of marginal animal feed is considered. The attributional modeling overlooks this aspect.

6.3.4 ALCA: Environmental performance of the other bio-based products

This section presents the ALCA comparisons between the environmental impacts of the co-products of the bio-jet fuel production process and their reference products.

6.3.4.1 Animal feed (organic residue)

Figure 38 shows the cradle-to-gate comparison between the organic residue from ABE fermentation sold as animal feed and the average European animal feed consumed (see Table 19). In four out of five impact categories, environmental benefits ranging between 65% and 87% were observed. A 38% higher impact was observed for the depletion of fossil fuels (see Figure 38 for the sources of impact). Figure 38 also shows the shares of the contribution of the various unit processes to the cradle-to-gate environmental impact of the animal feed co-product. We can observe that, since the allocation is applied only after subdividing the process as much as possible, the animal feed is not responsible for any environmental impact caused by either the swing adsorption process or alcohol condensation process. Similar considerations also apply to the other co-products. For this reason, the environmental impact of each co-product has its own “recipe” of environmental burdens’ contributors, which differ among them.

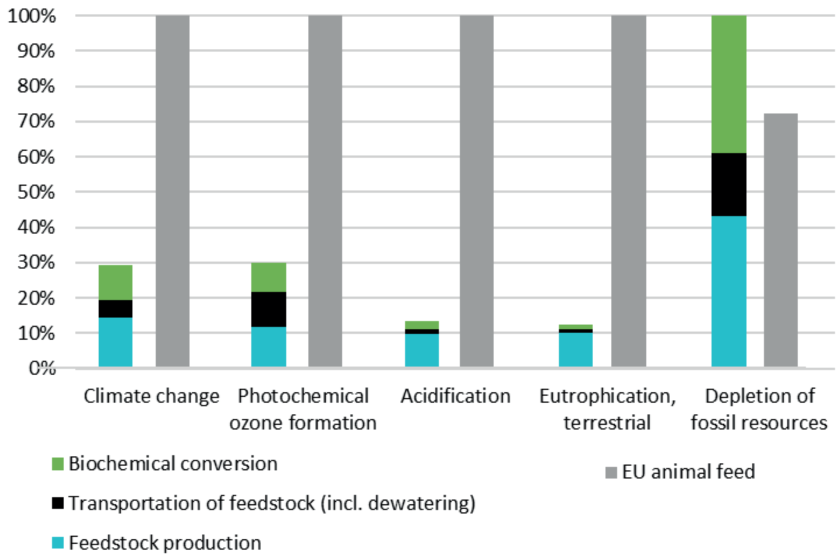

Figure 38. Cradle-to-gate comparison between the animal feed by-product (left bar) obtained from the bio-jet fuel production process (with process contributions highlighted) and the average European animal feed. Values are normalized taking the most impacting value per each category as the reference.

6.3.4.2 Bio-based carbon dioxide and bio-based hydrogen

Figure 39 shows the cradle-to-gate comparison between the carbon dioxide separated via swing adsorption and conventional carbon dioxide (see section 6.2.2.7). In three out of five impact categories, environmental benefits ranging between 16% and 65% were observed. However, the acidification and terrestrial eutrophication caused by the bio-jet fuel are higher than for conventional jet fuel (see Figure 39 for the impact sources).

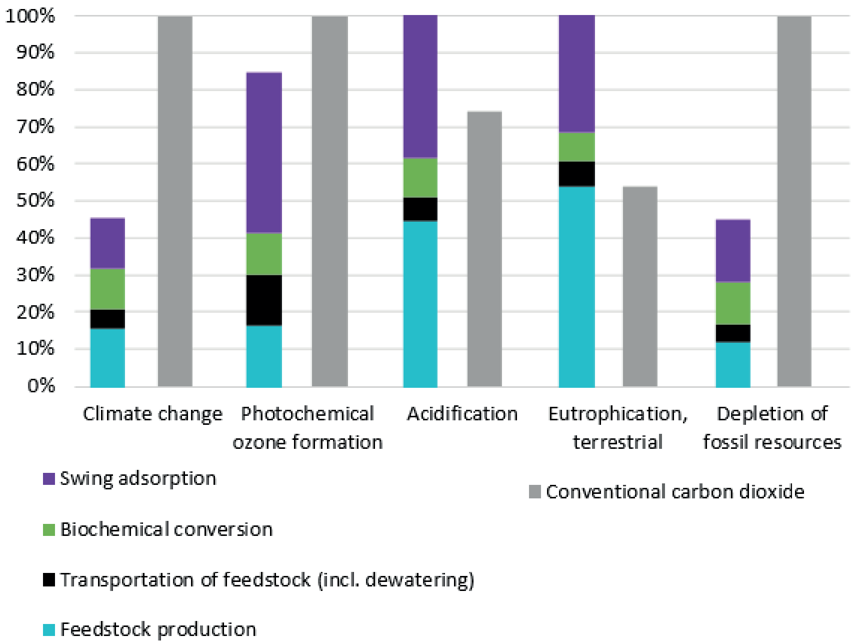

Figure 39. Cradle-to-gate comparison between the bio-based carbon dioxide (left bar) delivered by this innovative process (with process contributions highlighted) and conventional carbon dioxide. Values are normalized taking the most impacting value per each category valued as 100%.

Figure 40 shows the cradle-to-gate comparison between the bio-based hydrogen separated via swing adsorption and conventional hydrogen from reforming (see section 6.2.2.7). In four out of five impact categories, environmental benefits ranging between 45% and 78% were observed compared to reforming hydrogen (see Figure 40 for process contributions). For bio-hydrogen, the breakdown of the environmental impact corresponds to the one of bio-based carbon dioxide since they are both co-products leaving the system from the swing adsorption process.

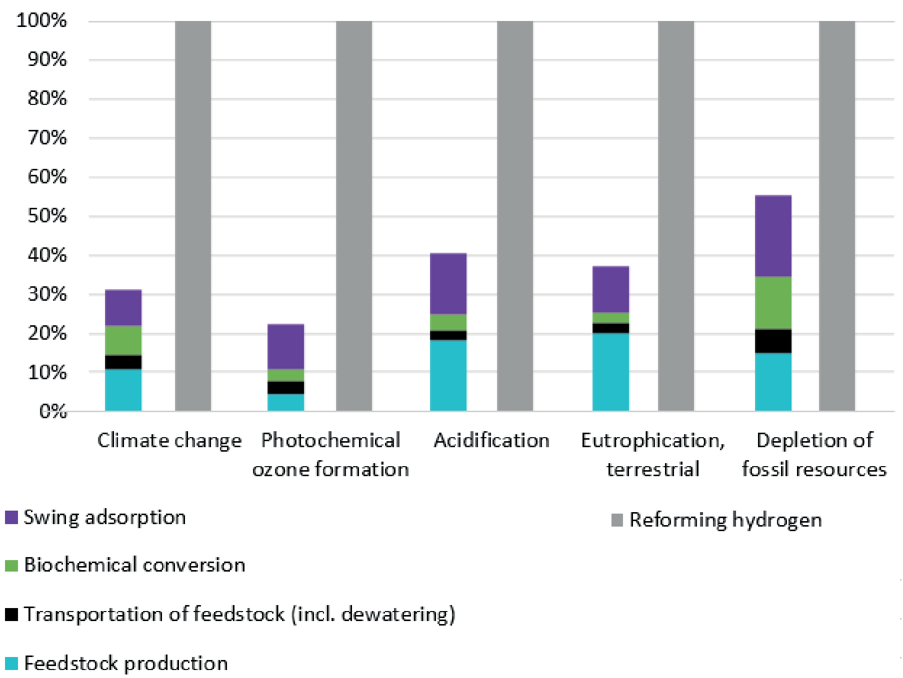

Figure 40. Cradle-to-gate comparison between the bio-based hydrogen (left bar) delivered by this innovative process (with process contributions highlighted) and reforming hydrogen. Values are normalized by taking the most impacting value per category valued as 100%.

6.3.4.3 Biolubricants

Figure 41 shows the cradle-to-gate comparison between the biolubricants produced from hydrotreatment and petrochemical lubricants (see section 6.2.2.7 for petrochemical lubricants). In four out of five impact categories, environmental benefits ranging between 12% and 88% were observed compared to petrochemical lubricants. For climate change, the impact of biolubricants was 18% higher than petrochemical lubricants (see Figure 41 for process contributions). For bio-hydrogen, the breakdown of the environmental impact corresponds to the one of bio-based carbon dioxide since they are both co-products leaving the system from the swing adsorption process.

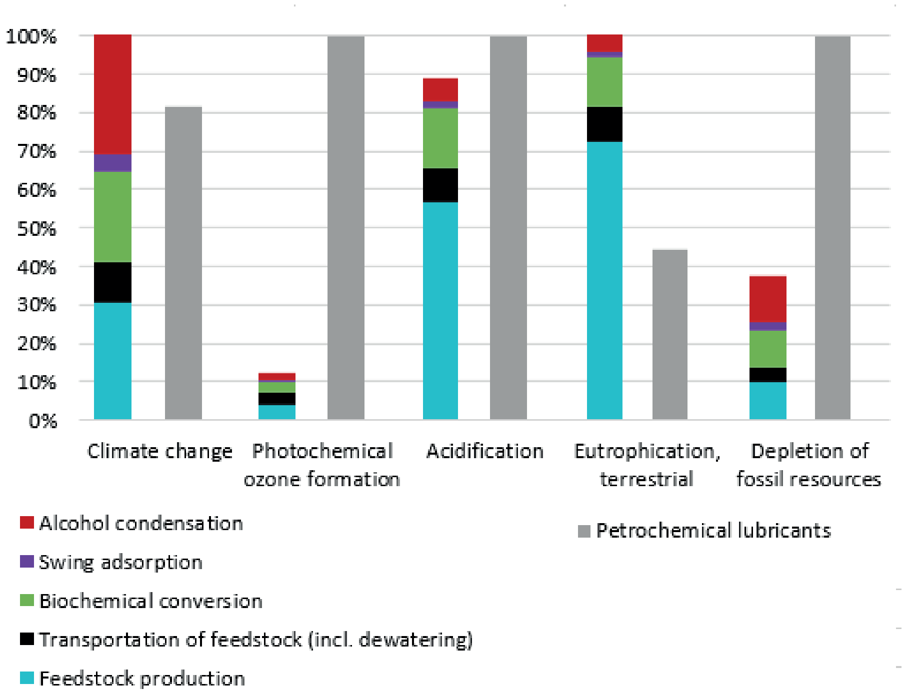

Figure 41. Cradle-to-gate comparison between the biolubricants (left bar) delivered by this innovative process (with process contributions highlighted) and petrochemical lubricants. Values are normalized taking the most impacting value per each category valued as 100%.

6.3.4.4 The "attributional pie"

By summing up the environmental impacts of each bio-based product (i.e. each piece of the pie), it is possible to visualize their contributions to the total pie i.e. the environmental impact of the entire "attributional" product system shown in Figure 34. Figure 42 shows the whole pie with its pieces.

The bio-jet fuel is the main cause of the environmental impact of the investigated product system. In fact, being responsible for the existence of the entire system product, the bio-jet fuel is the product that got allocated the environmental impact of all unit processes in the system (at least partially). The animal feed obtained as the organic residue also received a significant fraction of the environmental impact of the product system. The reason is that it got allocated most of the environmental impact at the level of the ABE fermentation due to its significant mass ratio compared to the other co-products (e.g. >5 kg of animal feed/kg of bio-jet fuel). The other co-products have minor "responsibilities" instead.

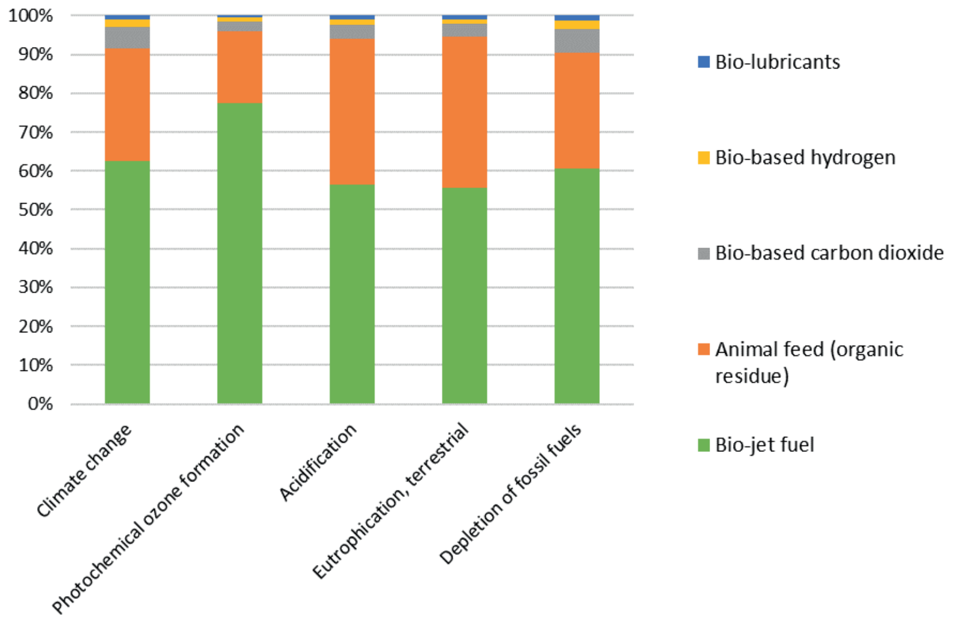

Figure 42. The attributional environmental impact of the overall production system and its allocation to each of the bio-based co-product.

6.4. DISCUSSION

6.4.1 Uncertainties in the attributional model

The ALCA has modeling uncertainties that are usually related to the application of a different allocation method or the price fluctuation for economic allocation. Applying energy or mass allocation at the level of the potato processing industry would mean a significant increase of the environmental impact of the potato by-products since they have physical characteristics similar to the main food products but much lower market price. As we argued in our earlier work [347], such a type of allocation leads to distorted results and would not respect the ISO causality principle that the allocation criterion should reflect. Regarding the price fluctuation of potato by-products, the market trend of the last five years showed a small variation (order of 3%) that would have a negligible effect on the results of the ALCA. The economic allocation factor applied at the level of the swing adsorption unit is also affected by uncertainties related to the prices of carbon dioxide and hydrogen. For both these products, novel technologies with a lower carbon footprint and/or using greener energy are expected to take place in the next decade. Carbon capture and storage (CCS) might also play a role in many chemical and energy processes in the longer term (e.g. the next two decades). Despite the deployment of CCS has been very slow so far (accounting for less than 0.5% of global investment in clean energy), robust climate targets could increase CCS investments [382]. Greener energy and CCS could affect the price of carbon dioxide and/or hydrogen, affecting the allocation share applied. Uncertainties also apply to the allocation factors regarding the fermentation residue sold as feed and lubricants, but they are not affected by future price fluctuations since based on energy and mass values respectively. The environmental impact of petrochemical kerosene is also influenced by the allocation applied at the level of the oil refinery, but with a minor effect on the overall environmental impact, for two reasons: 1) combustion emissions dominate the environmental impact of conventional kerosene (70-90% of cradle-to-grave impact) in all categories except depletion of fossil fuels and 2) there is small difference in energy, mass or economic shares for European kerosene over the total EU refinery system output [222].

6.4.2 Uncertainties in the consequential model

It is still challenging to address (without significant uncertainties) the historical question [89] "which feed ingredient will meet the increased protein demand?". A different answer to this question can significantly impact the final numerical results of the LCA [349].

For our study, taking different animal feeds to replace potato by-products does change the conclusion for climate change (see Figure 43). In fact, the climate change impact of the bio-jet fuel could become between 4% and 23% lower than petrochemical kerosene if the company currently utilizing the potato by-products replaced them with European animal feed only. Conversely, the climate change impact of the bio-jet fuel could double than calculated for the baseline scenario (and therefore significantly higher than petrochemical kerosene) importing soybean from South America to replace the potato by-products diverted from the European animal feed market. For the other four categories, the conclusions of the CLCA regarding the comparison between the bio-jet fuel and petrochemical kerosene are unaffected by the market displacement from the animal feed market. However, the numerical results could change significantly depending on the type of animal feed assumed and the impact category considered.

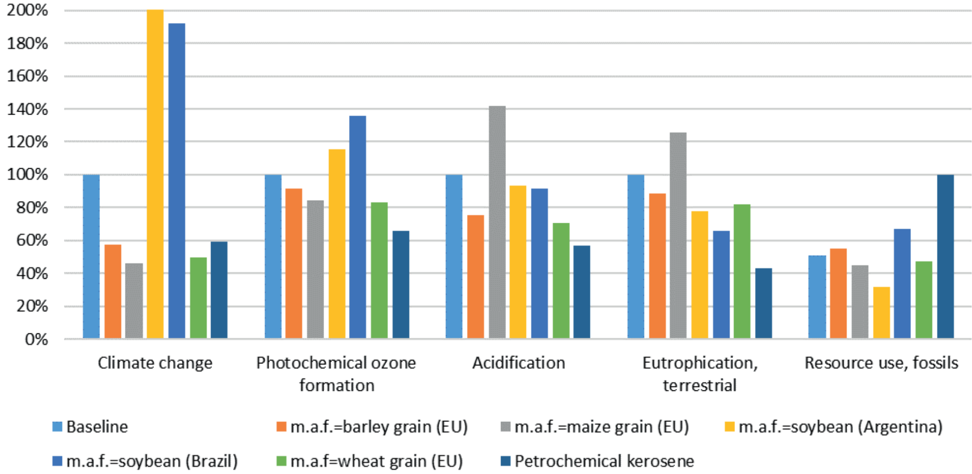

Figure 43. Consequential cradle-to-grave comparison between the innovative bio-jet fuel from potato by-products and petrochemical kerosene for 1 MJ of fuel varying the type of marginal animal feed (m.a.f.). Values are normalized taking the most impacting value per each category between the bio-jet fuel with baseline calculations and petrochemical kerosene (i.e. 100% taken as Figure 37).

Moreover, in our study, the displacement of animal feed with potato by-products was made on a protein basis. Alternatively, an energy basis could be assumed since animals do not need only proteins from the feed but also energy (the same apply to humans, who are the final user of the food products). The gross energy of potato by-products is 17.2 MJ/kg_{dry} [370]. Keeping the same composition of the marginal mix, the mass of animal feed to be substituted would increase by 10% mainly because soybean meals have much higher protein content than potato by-products but only slightly higher energy content. Alternatively, if one type of animal feed only were consumed as an alternative for potato by-products on the market, it should be considered what follows. Assuming that the primary function of potato by-products used as animal feed is to provide calorific energy and not protein to animals, it would be incorrect to compare 1 MJ of soybean meal only with 1 MJ of potato by-products. In fact, what matters is the primary function and the causality mechanism behind the provision of such a function [41,223]. For the same amount of energy, soybean meal would also provide a much higher amount of nutrients (proteins) to animals than the original potato by-products (hence, the primary function of soybean meal is to provide proteins). This means that the product's primary function that is included in the system boundaries would no more correspond to the primary function of the displaced product. Figure 44 shows the sensitivity analysis results where the market displacement was assumed based on energy as the primary function.

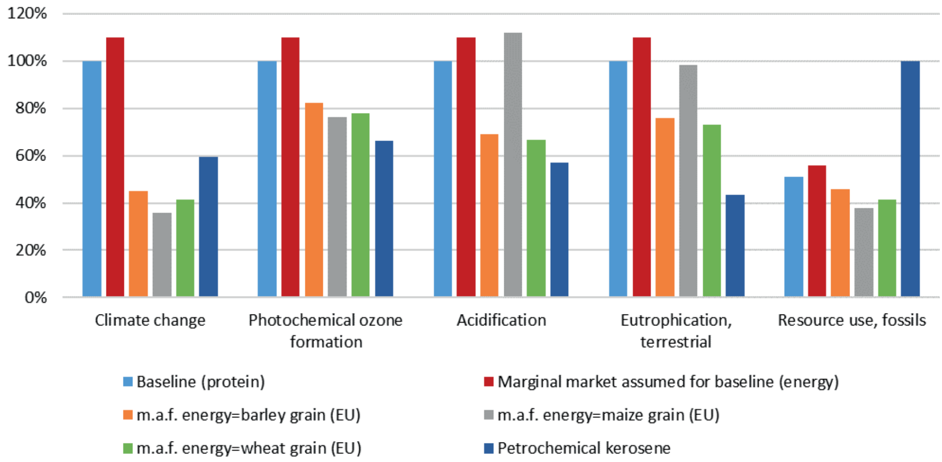

Figure 44. Sensitivity analysis results based on energy as the primary function. Consequential cradle-to-grave comparison for 1 MJ of fuel varying the type of marginal animal feed (m.a.f.). Values are normalized taking the most impacting value per each category between the bio-jet fuel with baseline calculations and petrochemical kerosene (i.e. 100% taken as Figure 37).

As for the previous sensitivity analysis, what emerges is that the outcome of the CLCA on the better or worse performance between the bio-jet fuel and conventional jet fuel was affected only for climate change. However, the environmental performance of the bio-jet fuel for photochemical ozone formation, acidification and terrestrial eutrophication can change significantly. The case of potato by-products displaced by maize is the best scenario for climate change and depletion of fossil resources. In particular, almost 40% reduction of climate change impact could be achieved compared to petrochemical kerosene. On the other hand, the acidification and eutrophication impacts would increase in the case of maize compared to barley and wheat grains.

Given the temporal scope (the year 2030), the production method of the products substituted in the CLCA based on market trends such as the surplus of hydrogen and carbon dioxide are affected by uncertainty. In fact, in the future, environmental impact reductions could be achieved if carbon dioxide and hydrogen production will be produced on a large scale using renewable energy (see discussion in section 6.4.1 for future effects on benchmark technologies). CLCAs with a future scope need to be revised if a significant market shift occurs and such shift was not accounted. Otherwise, such LCAs would consider an outdated production method instead of the one actually affected [237]. In the results of our CLCA, the credits for substituting carbon dioxide and hydrogen were (much) lower than the environmental impact of the marginal production of animal feed in four out of five categories. However, there is high uncertainty in the impacts of the additional animal feed (see Figure 43 and Figure 44). In the future, more sustainable animal feeds might be marketed e.g. food waste enriched with proteins using insects [383], or the market demand for meat may decline due to a switch in diet choices [384]. Furthermore, we assumed that the entire fraction of potato by-products are taking away feed from the animal feed supply. However, as detailed in section 6.2.1, 15% of potato by-products are not used as animal feed and would need waste treatment or can be used for lower-value applications.

6.4.3 Indirect land use change

The iLUC impacts from the displacement effects from potato by-products accounted for almost 45% of the climate change impact category in the baseline calculations. The results suggest that iLUC impacts steer to a large extent the biofuel environmental performance. However, iLUC impacts are subject to high uncertainty [385] and were significant only for imported soybeans. Carbon stock changes from land-use transitions were directly retrieved from publicly available LCA databases and not modeled directly, given the scope of the study. Therefore, impacts from carbon stock changes are conditioned by the assumptions and methods carried out in such databases.

Ecoinvent (WFLDB Quantis-adapted version of the Blonk tool) assumes that mainly forests are converted to arable land. For Brazil, 75% of the total converted land corresponds to forests. However, recently it’s been suggested that the effect of soybean as an (in)direct driver from forest loss in Brazil is more significant than previously understood [386]. Therefore, if a larger share of forest loss were accounted for, the share increase would lead to higher carbon losses and overall CO_2 emissions.

Forest contains considerably higher carbon stocks than other land categories such as grasslands, shrublands, or cropland [378]. Contrastingly, the change from any of the mentioned categories towards soybean would result in a lower iLUC impact.

Inherently, iLUC occurs somewhere else where biomass is produced and often with a significant time-related lag effect [387]. Therefore, attributing causality from displacement effects is extremely challenging.

In this paper, we attributed 100% of the carbon stock change impacts to the displacement effect. However, several economic, social, and environmental variables affect such direct casual effect attribution. Therefore, in reality, the displacement effect between shifting the land use in one location to the marginal animal feed production in another one is submitted to numerous conditions such as market conditions [63]. Thus, the causal-effect relationship might not be as direct as assumed in this study, and the potential impacts from carbon stock changes could decrease. In addition, land-use carbon stock changes are highly location and context-specific. These conditions can vary considerably driven by biophysical characteristics (e.g. temperature), management practices (e.g. land intensification), and socio-economic conditions, which can vary significantly even within the same region [388,389]. Thus, the real iLUC impact from bio-jet fuel production due to displacement effects is difficult to determine. Still, when accounted for in biofuels, iLUC generally results in an unfavorable environmental performance [390]. Note that iLUC processes and impacts are valid for any land-based service. Future research should focus on including adequate measures to estimate the percentage of attribution from iLUC process that help to reduce the uncertainty nature from these processes and understand better the overall performance of biofuel supply chains.

6.4.4 Advantages and disadvantages of attributional or consequential LCAs

There has been an open discussion, for more than 20 years, on which modeling approach (attributional or consequential) is better for environmental product labeling and policy making [146,177]. Currently, two (or more) well-established “internally consistent but mutually exclusive schools” exist [68]. Each of these schools claims that there is “general agreement in the literature” [391] that supports their modeling choices over the rest [391,392]. However, other researchers believe that both modeling approaches are necessary and both approaches should be kept well distinguished [68,146,393,394]. As shown by our LCA investigation and highlighted by previous literature [47,48,64], attributional and consequential LCAs of the same product or system could lead to different conclusions.

Table 21 shows a summary of major differences between attributional and consequential LCAs in general and in our case study. In our case, the two modeling approaches led to contrasting results for climate change and photochemical ozone formation (see section 6.3.3). The ALCA showed that the bio-jet fuel and its co-products offer environmental impact reductions in most categories compared to their conventional counterparts. A similar conclusion was drawn by Djomo et al. (2008), who concluded that "using potato steam peels to produce hydrogen along with feeding animals with its by-products offer more environmental benefits than using the potato steam peels directly for animal fodder". However, like our ALCA, the LCA of [395] and other peer-reviewed LCAs investigating bio-based products from potato by-products [347,396,397] overlooked the effects of diverting potato by-products from the animal feed market. However, the fact that we do not know precisely which animal feed market the potato peels are mainly sold for and with what feed would be probably replaced is a major uncertainty in our study. On the other hand, checks on the consequences linked with the indirect effects of biofuels production (and resulting environmental impacts) are needed for a policy perspective aiming to avoid unintended counterfactual effects.

From our case study, we have learned what follows. Contradictory trends in the outcome of attributional and consequential LCAs of a fuel from a bio-based by-product can be expected in certain impact categories if the three circumstances are in place. The first one is that the by-product feedstock is already marketed to be utilized by another process. The second is that the process from which the by-product feedstock is diverted is not part of the processes delivering the final bio-based product (the bio-jet fuel in our case). In our case study, the two LCAs would have led to less contradictory outcomes, e.g. if the potato industry itself would be the current user of the potato by-products they generate e.g. to produce biogas used internally. In that case, the potato industry would need to replace that biogas with an alternative energy input that both types of LCA would include. Third, the displacement of a by-product from its current use leads to more contradictory results between the two LCAs if their conversion process does not have a high yield. Consequently, i.e. requiring a large amount of by-product feedstock per t of the final product (the bio-jet fuel in our case).

Table 21. Summary of aims, product systems, multifunctionality, uncertainty and application of attributional and consequential modeling approaches in general and in our study. (continues to the next page)

Most critical difference	**Attributional LCA (ALCA)**	**Consequential LCA (CLCA)**
Aim		
General	"Provide information on what portion of global burdens can be associated with a product (and its life cycle). In theory, if one were to conduct attributional LCAs of all final products, one would end up with the total observed environmental burdens worldwide" [398].	"Provide information on the environmental burdens that occur, directly or indirectly, as a consequence of a decision (usually represented by changes in demand for a product)"[398].
In our case study	Investigate the environmental impacts caused by the production of an innovative bio-jet fuel starting from the total environmental impact of the production system (i.e. the full pie). LCAs of the other co-products of the bio-jet fuel aimed to understand how much each of the co-product is responsible for the production system's total environmental impact.	Investigate how introducing an innovative bio-jet fuel to the market could potentially change the overall environmental impact of the supply chain of bio-jet fuel production and the economic sectors affected by the change. Hence, the aim was to understand the environmental impact of this novel bio-jet fuel at the net of all the displacement effects i.e. the potato by-products from the animal feed market and the products potentially replaced by the co-products of the bio-jet fuel.
Product system definition		
General	"The systems analysed ideally contain processes that are actually directly linked by (physical, energy, and service) flows to the unit process that supplies the functional unit or reference flow". [398].	"The systems analysed in these LCAs are made up only of processes that are actually affected by the decision, that is, that change their output due to a signal they receive from a cause-and-effect chain whose origin is a particular decision". [398].
In our case study	The most critical aspect was the choice to incorporate additional animal feed production within the system boundaries in the consequential LCA. Conversely, the ALCA accounted for the impact of producing the potato by-products but did not include the effect on the animal feed market.	
Multifunctionality		
General	Allocation of the inputs and outputs of processes among co-products based on certain allocation keys.	The approach aims to reflect cause-and-effect chains including the expansion of the system to include affected unit processes outside the supply chain and substitution i.e. avoided burden effect due to the effects of introducing co-products in the market leading to the displacement of conventional market products.

Most critical difference	Attributional LCA (ALCA)	Consequential LCA (CLCA)
In our case study	Various allocation methods were applied at unit process-level (for details, see section 6.2.2.4).	Our setup is driven by the change in potato by-products' final use. Currently, potato by-products are used as animal fed. However, shifting the use for bio-jet fuel production results in a supply-side deficit, which requires to be covered by producing additional feed. Substitution was applied to the co-products.
Uncertainty related to product system definition and multifunctionality		
General	The modeling uncertainties in ALCAs are often related to the application of a different allocation method or the price fluctuation for economic allocation.	The fact that CLCAs are more sensitive to uncertainties than ALCAs due to the inclusion of market prospects is already broadly acknowledged by the literature [35,89,348].
In our case study	For this specific case study, the uncertainty of the allocation practices on the outcome of the study can be considered minor (for details, see section 6.4.1). The inclusion of the additional animal feed production in the system boundaries of the CLCA was affected by high uncertainties depending on both the type and functionality of the animal feed assumed (for details, see section 6.4.2) as well as uncertainties regarding indirect land use changes (for further details, see section 6.4.3).	
Applications		
General	Attributional LCAs have been broadly applied for ecolabeling and policy support since ALCA results are usually less sensitive to assumptions and have lower uncertainties [94,391,399]. Regarding policy application, results from attributional LCAs have been used in EU legislation to place thresholds on GHG emissions savings.	Consequential LCA have been applied mainly for policy support on understanding consequences of possible policy decisions especially for biofuels. The production of biofuels and current drastic changes in the energy and materials sectors are occurring due to policy interventions. Although the consequences of a decision might be uncertain, policy interventions could be supported to prevent unwanted effects from happening.
In our case study	Consequential modeling becomes a key tool to avoid unintended effects that might lead to significant environmental damages. In fact, an ALCA could not prevent the chance that diminishing the environmental impact of the aviation sector could push up the impact somewhere else e.g. increasing the impacts of meat production due to shifts in the animal feed market. For this reason, while ALCA could be a proper tool for market regulation, both ALCA and CLCA should be used to support policy making. For example, if the current user of potato by-products shifts towards maize as animal feed to replace potato by-products, 40% GHG savings can still be reached by this bio-jet fuel compared to conventional jet fuel. The directives of such a shift are in the hands of policy making.	

6.5. CONCLUSIONS

The main goal of this article was to compare the results of attributional and consequential LCAs to evaluate a future bio-jet fuel produced in the Netherlands.

For this specific case study, in all impact categories assessed, the environmental burdens were higher when using consequential modeling than attributional modeling, leading to contrasting conclusions in this fuel's environmental performance than conventional jet fuels. The reason was that, besides the major environmental hotspots related to the bio-jet fuel conversion processes, the impact of the production of additional animal feed could be much higher than the credits from co-product displacement in the consequential LCA.

So, even if the results of consequential LCAs by including market prospects and indirect land-use changes are more sensitive to uncertainties than attributional LCAs, we believe that both LCAs are necessary for decision making to mitigate possible indirect effects on the affected markets. In this specific case, our consequential LCA highlights the environmental issues arising if the potato by-products diverted from the European animal feed market to produce the bio-jet fuel are replaced (in part) with imported soybean meals from South America. Such an aspect was instead overlooked using attributional modeling. To mitigate indirect environmental impacts on the animal feed market, it is necessary that the market is steered with a holistic perspective and both ALCA and CLCA become necessary.

The technology investigated in our study is at a pilot scale and was successfully tested using potato by-products. However, other bio-based residual streams could be converted via ABE fermentation. Further research may provide data for assessing the operation of this innovative technology with other feedstocks.

6.A APPENDIX

Figure 6A.1. Process flow diagram conventional jet fuel (kerosene)

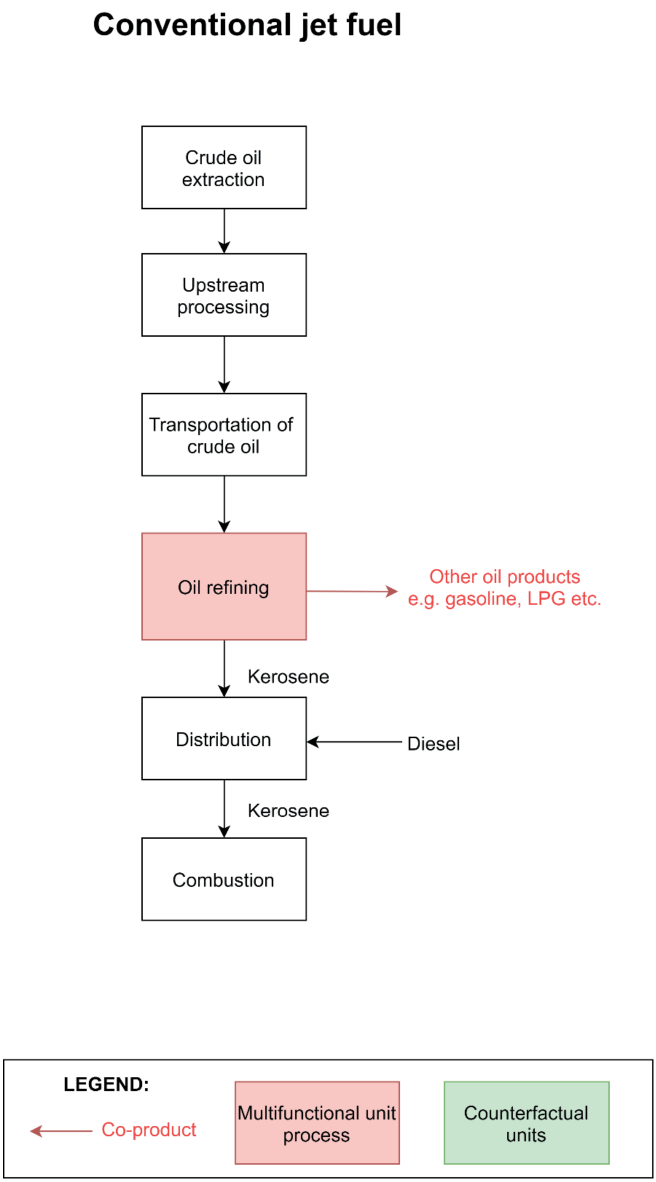

Table 6A.1. Cradle-to-grave attributional environmental impact of 1 MJ of bio-jet fuel with numerical breakdown

Impact category	Feedstock production	Feedstock transportation	Biochemical conversion	Swing adsorption	Alcohol condensation	Distribution	Combustion	Sum
Climate change	1.05E-02	3.68E-03	7.99E-03	1.60E-03	1.07E-02	1.33E-04	2.05E-04	3.49E-02
Photochemical ozone formation	2.28E-05	1.91E-05	1.77E-05	2.82E-06	1.07E-05	7.57E-07	3.54E-04	4.28E-04
Acidification	1.19E-04	1.80E-05	3.30E-05	3.55E-06	1.24E-05	6.79E-07	3.02E-04	4.88E-04
Eutrophication, terrestrial	5.21E-04	6.72E-05	9.42E-05	1.06E-05	3.14E-05	2.54E-06	1.50E-03	2.22E-03
Resource use, fossils	1.37E-01	5.61E-02	1.35E-01	2.81E-02	1.69E-01	2.08E-03	0.00E+00	5.26E-01

Table 6A.2. Cradle-to-grave consequential environmental impact of 1 MJ of bio-jet fuel with numerical breakdown

Impact category	Transportation of feedstock (incl. dewatering)	Biochemical conversion	Swing adsorption	Alcohol condensation	Hydrogen substitution
Climate change	1.08E-02	1.26E-02	1.63E-03	2.48E-02	-9.61E-03
Photochemical ozone formation	5.81E-05	4.08E-05	8.06E-06	1.40E-05	-2.78E-05
Acidification	5.21E-05	9.40E-05	9.80E-06	8.20E-06	-2.93E-05
Eutrophication, terrestrial	2.03E-04	2.83E-04	3.98E-05	9.65E-06	-1.16E-04
Resource use, fossils	1.61E-01	1.48E-01	2.42E-02	3.65E-01	-9.11E-02

It continues

Carbon dioxide substitution	Additional feed	Lubricants substitution	Distribution	Combustion	Sum
-1.57E-02	1.16E-01	-1.38E-03	1.35E-04	2.05E-04	1.39E-01
-1.71E-05	2.54E-04	-2.50E-05	7.62E-07	3.54E-04	6.60E-04
-1.89E-05	3.31E-04	-6.71E-06	6.46E-07	3.02E-04	7.42E-04
-5.29E-05	2.10E-03	-1.21E-05	2.53E-06	1.50E-03	3.95E-03
-2.65E-01	3.04E-01	-6.05E-02	2.08E-03	0.00E+00	5.88E-01

Table A6.3. Cradle-to-grave environmental impacts of 1 MJ bio-jet fuel from potato by-products and 1 MJ petrochemical kerosene. Results for ALCA and CLCA (baseline calculations). For a conventional product like petrochemical kerosene, the change between average and marginal data led only to a minor difference for acidification impacts.

Impact category	ALCA		CLCA	
	Bio-jet fuel	Petrochemical kerosene	Bio-jet fuel	Petrochemical kerosene
Climate change	3.49E-02	8.28E-02	1.39E-01	8.28E-02
Photochemical ozone formation	4.28E-04	4.36E-04	6.60E-04	4.36E-04
Acidification	4.88E-04	4.42E-04	7.42E-04	4.24E-04
Eutrophication, terrestrial	2.22E-03	1.71E-03	3.95E-03	1.71E-03
Depletion of fossil fuels	5.26E-01	1.16E+00	5.88E-01	1.16E+00

CHAPTER 7

Environmental Life Cycle Assessment of polypropylene made from used cooking oil

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ABSTRACT

Used cooking oil (UCO) has received much attention as feedstock for the production of renewable fuels and bio-based materials. This study aims to assess the environmental impact of UCO-based polypropylene (PP) by a cradle-to-factory gate Life-Cycle Assessment (LCA). 16 impact categories were assessed. The results were interpreted with normalization and weighting steps. For several multi-output processes, different allocation procedures were scrutinized.

On a normalized and weighted basis, the environmental impacts of UCO-based PP are dominated by climate change (28%), fossil resource use (23%) and water use (11%). The following environmental hotspots are identified: the polymerization process (38%), the production of hydrogen (21%), the production of LPG (18%) and the combustion of LPG (8%). Compared to petrochemical PP, cradle to factory gate impact reductions of 40-62% for climate change and 80-86% for fossil fuel resource use can be achieved by UCO-based PP, depending on the allocation approach chosen. Moreover, if renewable propane and methane produced from the biorefinery could be used to replace LPG, the overall weighted environmental impact of UCO-based PP could be potentially further reduced by 34%.

7.1 INTRODUCTION

It was 1954 when Natta and his research group first polymerized propylene to a crystalline isotactic polypropylene (PP) [400]. Already at that time, Natta realized that the properties of polypropylene could have introduced new trends in the world of plastics [401]. Nowadays, PP represents one of the most widely used plastics in Europe (nearly 20% of the total consumed plastics) [402]. In 2018, the global production of polypropylene resin was 56 Mt. The demand is projected to increase to 88 Mt by 2026 [403]. The high demand for polypropylene is due to its versatility, which allows its use in many applications such as food packaging, construction pipes, and automotive parts. The properties that especially make polypropylene multipurpose are associated with its high melting point, low density, excellent stiffness and strength [404].

In the last decade, due to the growing demand for plastics and related concerns about their climate change impact, bio-based plastics have attracted attention as a possible option to replace petrochemical plastics. Many studies have highlighted that bio-based plastics could potentially offer a lower carbon footprint [7,56,405–407]. However, their environmental performance depends on the type of polymer, the impact category in focus, the selected system boundaries, the type of biomass feedstock and its final application and the supply chain [407–410]. In 2017, the global production capacity of bio-based plastic materials reached 2.05 Mt and was expected to increase to 2.44 Mt by 2022 [411]. In particular, bio-based PP entered the market in 2019 with a production capacity of about 19 kt, which is predicted to increase by about six folds by 2024 [412]. The three main synthesis routes for bio-based PP are: 1) using bio-ethanol from sugar fermentation [413–415], 2) using bio-syngas [415,416] and 3) using hydrotreatment of used cooking oil (UCO) [417].

To our knowledge, only two peer-reviewed environmental life cycle assessments (LCAs) of bio-based PP are publicly available [415,418], and no peer-reviewed LCA has been conducted for the third route, i.e. UCO-based PP. Mayumi et al. performed a cradle-to-factory gate LCA of biomass-derived PP and polyethylene (PE) at the design stage [418]. In the study, they quantified greenhouse gas (GHG) emissions of polyolefins made from the waste wood-syngas route. They found out that bio-based PP and PE could lead to higher GHG emissions compared to the petrochemical counterparts. The impacts of the bio-based polymers are dominated by biomass production and conversion processes. Kikuchi et al. (2017) investigated PP and

PE made from sugarcane-ethanol and woody-biomass-derived syngas based on data from demonstration plants and simulations. Defining one liter of ethanol as functional unit, they found that the highest GHG reduction can be obtained by using ethanol for PE production (a saving of about 50% compared to petrochemical PE), followed by use of ethanol as transport fuel (a saving of about 40% compared to gasoline), and PP production (a saving of about 15-20%). They concluded that PP production from bio-syngas offers relatively limited GHG emission reduction.

This study aims to investigate the environmental impacts of bio-based PP obtained through the third route, i.e. via hydrotreatment of UCO. UCO is a waste and thus does not compete with food and feed. It also avoids potential land use changes. In the EU-28, the total potential UCO available from the gastronomy sector, food processors and households is estimated at around 4 Mt per year [419]. These features make it an attractive feedstock for future transportation fuels and material production [341,420]. Primary data were collected from Neste Oyj based on a new commercial production facility in Europe [421].

The primary objective of this study is to provide a full picture of the environmental footprint of UCO-PP. Unlike the published LCAs of bio-based PP, which focused on climate change only, this study assesses 16 mid-point environmental impact categories following the recommendation of impact category selection by the European Commission's Product Environmental Footprint Category Rules (PEFCR) guidance (draft version 6.3) [165]. Such a wide selection of indicators has been rarely reported for the environmental assessment of bio-based chemicals and products [7,42]. For bio-based materials, environmental indicators such as land use, eutrophication, and acidification should not be ignored before investment or a policy decision is made [42,407]. Since there are several multifunctional processes involved, the second objective of the study is to gain insights into how the allocation choices could influence the LCA results. Despite that LCA is a standardized methodology, multifunctionality is one of the main remaining issues in LCA [50]. The findings of this LCA should be used both to understand the full environmental impacts, originated from both resources and emissions, and also to provide recommendations for future EU policy directives on the allocation choices for innovative bio-based plastics. The policy-level allocation recommendation is so far only available for renewable transport fuels and bioenergy [9]. The insights gained from this case study could help to reveal some complexity and demonstrate the influence of the allocation decisions in the LCAs of innovative bio-based products.

7.2 MATERIAL AND METHODS

7.2.1 Goal and scope definition

Life Cycle Assessment (LCA) is a widely applied standardized method used to assess the environmental impacts of a product or a service. In particular, this LCA study focuses on UCO-based PP and has been developed within the EU Bio-SPRI[11] project. The LCA was conducted according to ISO 14040 and ISO 14044. The draft PEFCR guidance was used as practical guidance when the guidance from the ISO standards were insufficient [38,127,165]. The main goal of this LCA is to assess the impact of UCO-based PP to identify the environmental hotspots of the life cycle. The second goal of this LCA is to add further valuable research to the open debate of solving multifunctionality of biorefineries (as detailed in section *Multifunctionality*). The results of this study are intended to be used by the industry for further process improvement, policy makers and the LCA community.

Based on the goal of the study, the functional unit (FU) is defined as 1 kg of polypropylene. A cradle-to-factory gate scope and an attributional approach are adopted in the LCA. Figure 45 shows the process diagram of the production of UCO-based PP. Used cooking oil is converted into high value hydrotreated vegetable oil (HVO) by Neste NEXBTL technology. Together with the HVO renewable diesel grade product, a renewable HVO naphtha grade product is obtained from the hydrotreatment process. This study focuses on the cracking of this "bio-naphtha" to obtain propylene via a process equivalent to petrochemical steam cracking.

Accordingly to the location of Neste Oyj's biorefinery (Rotterdam), the geographic scope is defined as the Netherlands. This reflects the specific situation of UCO collected from the Netherlands and nearby regions and all the major processes for the conversion into polypropylene occurring in the Netherlands as well with the exception of steam cracking occurring in a neighbor country. Nevertheless, when a specific inventory for the Netherlands was not available, or a specific process occurs in another EU country, average European data have been used.

The temporal scope is current (2018) to the near future (5-10 years), and the technological scope is defined as the status-quo technology which is ready for commercialization (technology readiness level 8, based on definition reported in [298]).

[11] BIOSPRI (Bioeconomy: Support to Policy for Research and Innovation) project funded by DG RTD of the European Commission.

Sixteen mid-point impact categories are selected to analyze the full environmental footprint. The adopted impact assessment models for each impact category are listed in

Table *22*. Their selection is based on the recommendation of PEFCR draft guidance (version 6.3), which was the version available at the time when the study was conducted. Differently, from the PEFCR guidance, particulate matter (PM) and land use are assessed using the methods recommended by the PEF guide [85].

Due to the many impact categories considered, normalization and weighting are applied to identify the *overall* environmental hotspots. To ensure that the same impact assessment models are used for characterization and normalization, the following selection has been done. For water use and resource depletion categories, the normalization factors are retrieved from PEFCR guidance v.6.3. For all the other impact categories, per capita, EU 27 normalization factors (2010) are retrieved from ILCD 2015 [171].

Table *22* reports the normalization and weighting factors applied in this study.

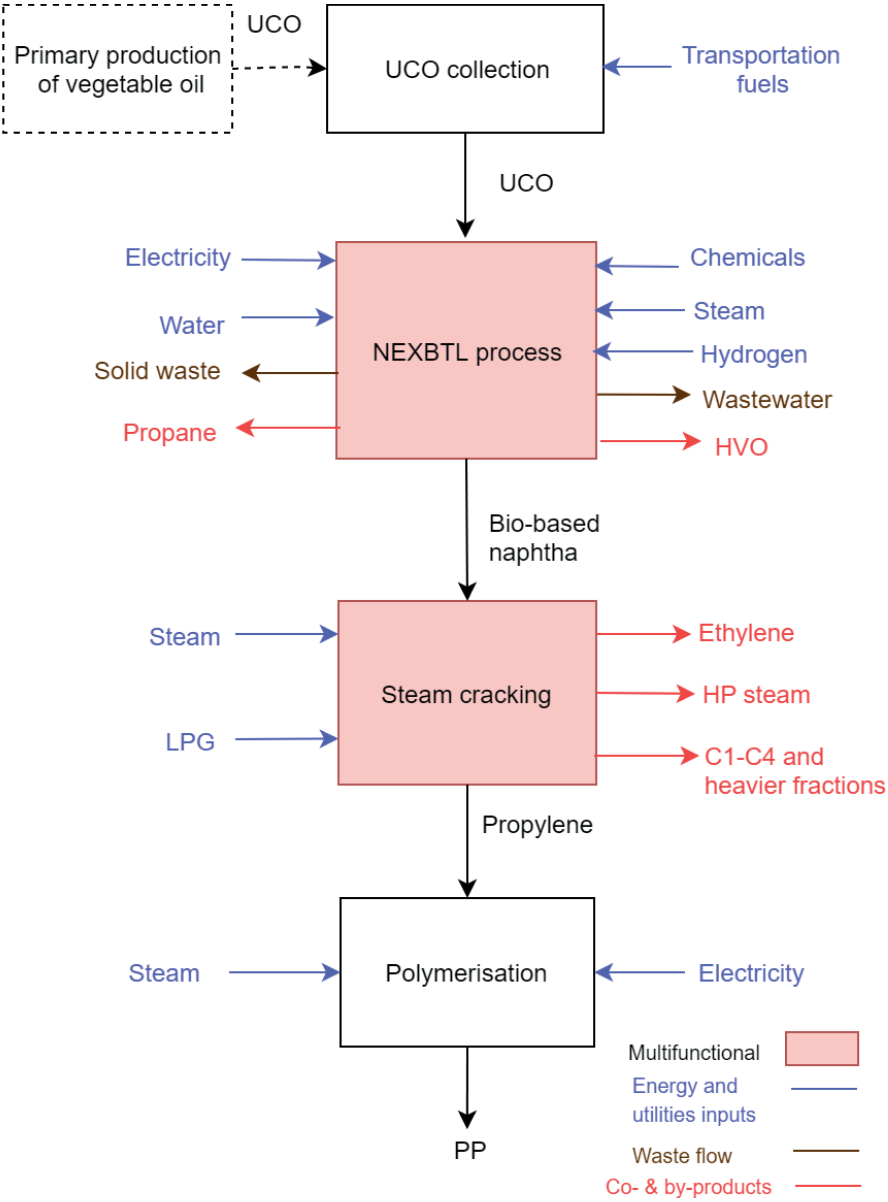

Figure 45. Flow diagram of the production of polypropylene from used cooking oil (UCO). The dashed box represents the primary production of vegetable oil, which is a unit process that is out from the system boundaries in the baseline analysis.

Table 22. Selected impact categories and models, normalization and weighting factors

Impact Category	Unit	Impact assessment models	Normalization factors EU 27 per person [165,171]	Weighting factors with toxicity [165,171]
Climate change	kg CO_2eq	IPCC 2013, GWP 100a with carbon climate feedback [310]	9.22E+03	21.06
Ozone depletion	kg CFC-11 eq	[422]	2.16E-02	6.31
Human toxicity, non-cancer effects	CTUh	USEtox (recommended + interim) [423]	5.33E-04	1.84
Human toxicity, cancer effects	CTUh	USEtox (recommended + interim) [423]	3.69E-05	2.13
Particulate matter	kg PM2.5 eq	[311]	3.80E+00	8.96
Ionizing radiation Human Health (HH)	kBq U235 eq	[424]	1.13E+03	5.01
Photochemical ozone formation	kg NMVOC eq	[312]	3.17E+01	4.78
Acidification	molc H+ eq	[313]	4.73E+01	6.20
Terrestrial eutrophication	molc N eq	[313]	1.76E+02	3.71
Freshwater eutrophication	kg P eq	[355]	1.48E+00	2.80
Marine eutrophication	kg N eq	[355]	1.69E+01	2.96
Freshwater ecotoxicity	CTUe	USEtox (recommended + interim) [423]	8.74E+03	1.92
Land transformation	kg C deficit	Soil Organic Matter model [325]	7.48E+04	7.94
Water use	m^3	AWARE factors [425]	1.15E+04	8.51
Resource use, minerals and metals	kg Sb eq	[315]	5.79E-02	7.55
Resource use, fossil fuels	MJ	[315]	6.53E+04	8.32

7.2.2 Life cycle inventory modeling

7.2.2.1 Unit processes, data, and assumptions

For the foreground system, primary data were collected from Neste. Those are site-specific data. The background data were largely based on the Ecoinvent database (version 3.4) and PlasticsEurope's Eco-profiles [404]. For unavailable data, assumptions were made based on literature and/or validated by Neste. Important assumptions have been scrutinized by sensitivity analyses in the discussion section (*Data uncertainty*).

The production of UCO-based PP starts from the collection of UCO. In the baseline calculations, UCO has been considered as waste and, therefore, entering the system "free of burdens'' (cut-off approach). A sensitivity analysis of this approach can be found in the discussion section (*Model uncertainty: multifunctionality*). The impacts of the collection are taken into account in the LCA. UCO is sourced from restaurants and commercial buildings mainly in the nearby regions of the bio-refinery country of location. The collection of UCO from restaurants and other users to the biorefinery is carried out by trucks and by water and distances assumed based on Neste's specific supply chain.

During the NEXBTL process, the oil is pre-treated and deoxygenized under high pressure to transportation fuel quality using hydrogen. The triglycerides of UCO are converted to saturated straight and branched-chain hydrocarbons and oxygen (of the triglycerides eliminated). In the chemical reaction shown by equation 5, a triglyceride with formula $C_{57}H_{104}O_6$ is taken as an example. Hydrogen is produced by steam reforming of natural gas and fuel gas.

$$C_{57}H_{104}O_6 + 15\,H_2 \rightarrow 3\,C_{18}H_{38} + C_3H_8 + 6\,H_2O \qquad \text{Eq. 5}$$

The main inventory data related to the NEXBTL unit process are collected in Table 23. For each ton of pre-treated oil, 1.02 t of UCO are needed [426]. Steam is produced in the refinery while the Dutch national grid supplies the electricity. The amounts of steam reported in the inventory tables are equivalent kilograms, recalculated to keep into account the different energy content between the actual steam flows and the chosen dataset (equivalent kilograms on an energy basis). The chosen dataset represents the production of 1 kg of steam used for heating in the chemical industry. The heat is produced with the average fuel mix used in the European chemical industry [427]. The process releases wastewater that is treated on-site.

Table 23. Input and output data per 1 kg of bio-based naphtha from UCO (NEXBTL unit process-hydrotreatment plus pre-treatment)

Flow	**Data**	**Process**	**References on which foreground data are based on**
Inputs			
Collected UCO	49.0 kg		
Phosphoric acid	28.0 g	Phosphoric acid, industrial grade, without water, in 85% solution state {RER}| purification of wet-process phosphoric acid to industrial grade, product in 85% solution state | APOS	[36]
Process chemicals	0.1 kg	Chemical, inorganic {GLO}| production | APOS	
Water	5.0 kg	Water, decarbonised, at user {RER}| water production and supply, decarbonised | APOS	[36,426]
Sodium hydroxide	48.0 g	Sodium hydroxide, chlor-alkali production mix, at plant/RER (PlasticsEurope. Industry data 2.0 project)	[36,428]
Electricity	6.0 MJ	Electricity, medium voltage {NL}| market group for | APOS	[36,426]
Steam	10.0 kg	Steam, in chemical industry {RER}| production | APOS	[36,426]
Hydrogen	1.7 kg	Hydrogen (reformer) E (PlasticsEurope. Industry data 2.0 project)	[36,381,426]
Nitrogen	1.6 g	Nitrogen, liquid {RER}| market for | APOS	[36,426]
Outputs			
Bio-based naphtha,	1.0 kg		Along with HVO diesel and propane whose percentages are not disclosed.
Wastes			
Wastewater (output to technosphere: waste to treatment)	8.8 L	Wastewater, average {Europe without Switzerland}| market for wastewater, average | APOS	[36,426]
Solid waste going to incineration	0.5 kg	Final waste flow, waste to incineration	

After hydrotreatment, bio-based naphtha is transported by train[12] to the steam cracking unit (distance based on Neste's specific case). During steam cracking, bio-based naphtha is diluted with steam and cracked into smaller hydrocarbons such as propylene and ethylene. The reaction is highly endothermic; therefore the feed is heated in a furnace burning fuel gas. The mass and energy inputs of the steam cracking unit in real operation are kept confidential and therefore the process has been modeled based on literature and is not Neste- specific. Process data from Karimzadeh et al. 2009 were modified considering the same feed capacity as the real operating pilot. The direct emissions released by the combustion of propane and butane are retrieved from the US Environmental Protection Agency dataset for industrial boilers [429]. In the calculations, The mix of 50% propane and 50% butane is assumed according to the dataset used for the production of LPG [430]. A selection of the main inventory data of this unit process is shown in Table 24.

The last process is polymerization of the propylene to obtain PP. The polymerization of bio-based propylene is identical to that of petrochemical propylene. The polymerization process has been extrapolated from the most recent PlasticsEurope's Eco-profiles [404] as follows: assuming that 1.02 kg (based on [432]) of propylene are required to produce 1 kg of polypropylene, the impact of the polymerization process has been obtained by subtraction. The PlasticsEurope's Ecoprofile [433] has been widely used as benchmarks for comparison. However, they are often regarded as "black box" data because of a lack of transparency constrained by confidentiality.

As a consequence, it is challenging to interpret the impacts of the polymers fully. This can be considered a limitation of the study because of the restricted interpretation of the polymerization process due to the use of this dataset (see section *Data uncertainty*). Nevertheless, polymerization has also been modeled according to the Matter (MATerials Technologies for CO_2 Emission Reduction) study of 1998 for sensitivity analysis, and when necessary the comparison has been used as a validation test [432]. Despite the Matter study is twenty years old, it has been selected since it is the only transparent dataset for PP polymerization available in the public domain.

[12] From Ecoinvent 3.4: Transport , freight train {Europe without Switzerland}| market for | APOS

Table 24. Selected inventory data of the steam cracking process per 1 kg of bio-based propylene made from bio-based Naphtha.

Flow	Foreground data	Adopted background processes from Ecoinvent or modeled direct emissions	References on which these data are based on
Inputs			
Bio-based Naphtha	2.67 kg		Naphtha from hydrotreatment. [431]
LPG	0.63 kg	Liquefied petroleum gas {CH}| market for | APOS	[429,431]. Notice that only 0.43 kg of this LPG is burnt while the rest pumped with the feed.
Steam	2.9 kg		[431]. This steam is the sum of diluted steam (66%) and boiling feed water (34%).
Outputs			
Propylene	1.0 kg		
Steam	5.1 kg	Steam, in chemical industry {RER}| production | Conseq (dataset used for net production)	[431] Steam conditions: 520 °C and 112 bar
Other cracked gases and heavier products	Not disclosed		Ethylene is the major co-product and is accompanied by several other by-products (e.g. hydrogen, methane, benzene) whose percentages are not disclosed.
Direct Emissions			
Nitrogen oxides	1.3E-03 kg	Emissions to air due to LPG burning	[429,431].
Carbon dioxide of fossil origins	1.3E+00 kg	Emissions to air due to LPG burning	[429,431].
Carbon monoxide	7.5E-04 kg	Emissions to air due to LPG burning	[429,431].
Methane	1.9E-05 kg	Emissions to air due to LPG burning	[429,431].
Dinitrogen monoxide	8.5E-05 kg	Emissions to air due to LPG burning	[429,431].
Particulate <2.5 um	5.7E-05 kg	Emissions to air due to LPG burning	[429,431].

Flow	Foreground data	Adopted background processes from Ecoinvent or modeled direct emissions	References on which these data are based on
Particulates, > 2.5 um, and < 10um	1.9E-05 kg	Emissions to air due to LPG burning	[429,431].
VOC, volatile organic compounds	2.5E-05 kg	Emissions to air due to LPG burning	[429,431].
Sulphur oxides	1.6E-06 kg	Emissions to air due to LPG burning	[429,431].

7.2.2.2 Multifunctionality

According to ISO 14044, multifunctionality (or commonly also referred to as ‘’allocation”) should be solved using the following hierarchy [38]:

1) Avoiding allocation by subdivision or system expansion (i.e. expanding the product system to include the additional functions related to the co-products);

2) Allocation underlying physical relationships (i.e. an allocation that reflects how the inputs and outputs are changed by quantitative changes in the products of the system); and

3) Allocation (partitioning) based on other relationships (e.g. economic allocation).

In this context, the framework followed in this LCA to identify the impact of the choice of allocation approach is detailed. In the product system studied, there are two processes where more than one useful product is delivered: hydrotreatment and steam cracking.

The hydrotreatment process mainly delivers three products: hydrotreated vegetable oil (renewable diesel), propane and bio-based naphtha whose percentages are not disclosed. In this study, bio-based naphtha is the precursor of propylene while the other two co-products are sold. In this LCA, the problem of how to assign the environmental impact to the multiple products of hydrotreatment has been solved through energy allocation. Concerning the hierarchy above, the allocation is not avoided due to the inapplicability of subdivision (the process cannot be further subdivided). System expansion, both enlargement and reduction approaches, is also not possible. In fact, according to the goal and scope of the study, the functional unit cannot be enlarged to include all the co-functions. Concerning the inapplicability of system reduction, bio-based naphtha is a non-dominant by-product and therefore, the criterion of physical significance is not respected (i.e. the mass ratio of bio-naphtha is in the magnitude of a few percents to that of HVO, see Table 23). Allocation by physical causality is not applied because this would require a mathematical model (commonly based on linear programming) that is not available [215,222,234]. Among the possible remaining allocation methods, energy has been chosen according to RED recommendations when dealing with transportation fuels [9,434]. Although naphtha is not used as transportation fuels, it is a by-product of fuel production (HVO) and therefore, RED recommendations are followed to respect the energy balances.

Nevertheless, it should be noticed that the RED focuses only on greenhouse gas emissions while 16 impact categories are analyzed in this study. For this reason, the authors have performed a sensitivity analysis on all possible allocation approaches used in the study (see the discussion section 7.4.2). Energy allocation has been adopted for the hydrotreatment process.

The steam cracking unit delivers two main products, i.e. propylene and ethylene, other cracked gases and steam (see Figure 45 and Table 24). Propylene is in the focus of this study. Nevertheless, ethylene represents the biggest mass fraction among the cracked gases. The steam cracking process also delivers 2.2 kg (6 MJ) of net industrial steam per kg propylene. Similar to the hydrotreatment unit process, subdivision, system expansion, and physical causality are not applicable. Avoiding allocation by substituting all propylene co- and by-products is not possible because a non-dominant product is in focus. For this unit process, a hybrid method has been chosen. Steam is directly used for other processes of the same biorefinery, and, otherwise, should be produced as marginal production of refinery steam from Ecoinvent (Steam, in chemical industry {RER}| production | Conseq). Direct substitution has been therefore applied to the net production of high pressure (HP) steam as, in this case, it may represent physical causality better than other arbitrary allocations (as highlighted by the PEFCR draft guidance). Energy allocation has been applied to all the other co-(by-) products resulting in 20% of the remaining impacts allocated to propylene.

7.2.2.3 Biogenic carbon accounting

The carbon content of UCO-based PP originates from the CO_2 sequestered by biomass. According to the PEFCR draft guidance, only biogenic carbon emitted later than 100 years after its absorption shall be considered permanent storage [165]. Permanent storage results in a carbon credit to be assigned to the bio-based product. When biogenic carbon is emitted earlier than 100 years, no carbon credits must be assigned for temporally carbon storage or delayed emissions [165]. In particular, the PEFCR guidance recommends to not assign carbon credits for cradle-to-user assessments [165]. Nevertheless, the biogenic carbon content at the factory gate ''shall be reported as additional information'' [165]. Accordingly, the authors have therefore proceeded as follows: the climate change impact is reported both with and without biogenic carbon removals while only the second one has been considered for the weighted results. Considering the chemical formula of propylene C_3H_6, per kg PP, the biogenic carbon removal corresponds to 3.14 kg CO_2eq.

7.3 RESULTS

7.3.1 Impact assessment and interpretation

The cradle-to-factory gate environmental impact results are shown in Table 25, while the breakdown results of each unit process are illustrated in Figure 46 (see Appendix A for numerical values behind the figure).

From Table 25, it can be seen that climate change (28%), fossil resource use (23%) and water use (11%) as the most important environmental impacts of UCO-based PP. shows that the NEXBTL process (hydrotreatment plus pre-treatment), steam cracking and polymerization are the three most significant key-unit processes in terms of environmental impact. On a weighted basis, polymerization contributes 38%, steam cracking 26% and hydrotreatment 29% of the total cradle-to-factory gate impacts[13]. The impact is almost entirely caused by the production (18%) and combustion of LPG (8%) and the production of hydrogen (21%) respectively for steam cracking and hydrotreatment (environmental hotspots). In particular, 7% out of 8% share of LPG combustion is caused by releasing GHG emissions. UCO collection and transportation of naphtha account only for 5% and 2% respectively. Pre-treatment represents the remaining 5%.

[13] These shares of impact highlighted by the weighted results are not affected by the choice of applying weighting with toxicity instead of without toxicity. Indeed, changing weighting factors, only the shares of steam cracking and polymerization vary to 28% and 36% respectively.

Table 25. Cradle-to-factory gate environmental impacts of 1 kg UCO-based PP

Impact Category	**Unit**	**Value**	**Normalized and weighted scores (Total 100%)**
Climate change (without biogenic carbon removal (BCR))	kg CO_2eq	0.63	28%
Climate change, with biogenic carbon removal (BCR)	kg CO_2eq	-2.51	Not applicable
Ozone depletion	kg CFC-11 eq	9.0E-08	1%
Human toxicity, non-cancer effects	CTUh	1.1E-08	1%
Human toxicity, cancer effects	CTUh	5.5E-09	6%
Particulate matter	kg PM2.5 eq	1.2E-04	5%
Ionizing radiation HH	kBq U235 eq	6.4E-02	6%
Photochemical ozone formation	kg NMVOC eq	1.9E-03	6%
Acidification	molc H+ eq	2.1E-03	5%
Terrestrial eutrophication	molc N eq	6.0E-03	2%
Freshwater eutrophication	kg P eq	8.7E-06	0%
Marine eutrophication	kg N eq	5.6E-04	2%
Freshwater ecotoxicity	CTUe	2.2E-01	1%
Land transformation	kg C deficit	1.1	2%
Water use	m^3	7.4E-01	11%
Resource use, minerals and metals	kg Sb eq	3.2E-07	1%
Resource use, fossil fuels	MJ	9.3	23%

The hydrotreatment process has a significant contribution (20-40%) in the following impact categories (see): climate change, human toxicity without cancer effects, human toxicity with cancer effects, particulate matter, photochemical ozone formation, acidification, terrestrial eutrophication, marine eutrophication, and resource use categories. For almost all the above impact categories, hydrogen production from steam reforming is the most (88-97%) relevant source of environmental impacts of the hydrotreatment process. The only exception is human toxicity without cancer effects, where most of the impact is due to producing electricity (37%) and phosphoric acid (31%).

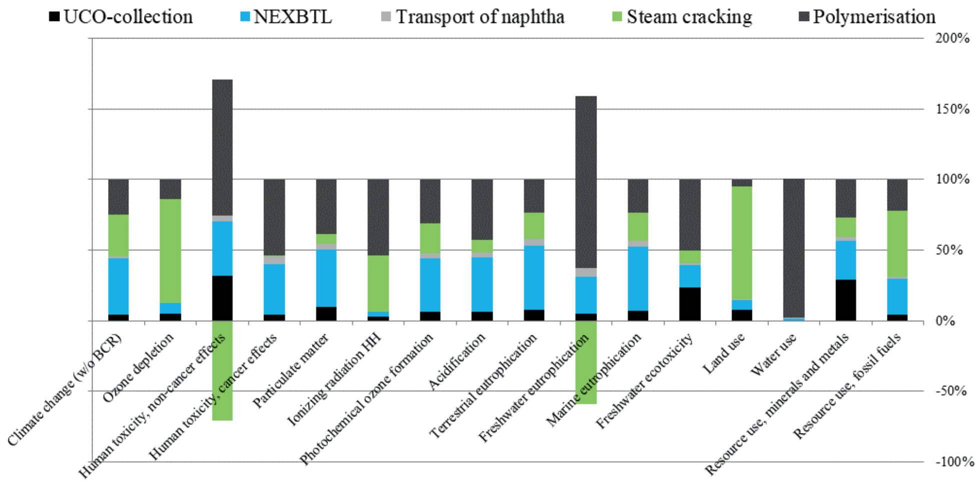

Figure 46. Breakdown of cradle-to-factory gate environmental impact of UCO-based PP per key-unit process (climate change shown without biogenic carbon removal (BCR))

Since a PlasticsEurope's "black box" Ecoprofile has been used for polymerization (see section *7.2.2.1*), an interpretation at the activity level is not possible. Nevertheless, it is possible to identify the five impact categories where polymerization contributes with the highest share (on a weighted basis) on the total impact (see Appendix A): water use (10%), climate change (7%), resource use of fossil fuels (5%), human toxicity (cancer) (3%) and ionizing radiation (3%).

From, it is possible to see that steam cracking shows negative impacts in two categories: *human toxicity without cancer effects* and *freshwater eutrophication*. The ''credit'' received from the substitution of the net steam produced during the steam cracking overcompensates the two impacts caused by LPG production and combustion. The steam cracking process is responsible for a significant share of environmental impact in several impact categories: namely, climate change (ca. 30%), ozone depletion (ca. 75%), ionizing radiation HH (40%), land use (80%) and fossil fuels resource use (ca.50%). In particular, LPG production and its combustion account respectively for 18% and 82% of the cradle-to-factory gate impact on climate change. The production of LPG alone is instead entirely responsible for the impact caused in the other four impact categories.

7.4 DISCUSSION

7.4.1 Data uncertainty

This section reports several sensitivity analyses related to data uncertainty. The first assumption that is discussed is related to UCO collection. UCO was assumed to be sourced locally in the nearby regions of the bio-refinery, based on Neste's specific case. This assumption led to a small overall impact (5%) from UCO collection. Nevertheless, UCO has attracted much attention as one of the bio-based feedstocks to achieve European renewable energy and greenhouse gas (GHG) emissions targets. This has led to an increase in demand and the international trade of UCO [435]. Accordingly, it is reasonable that in the case of larger scale production of bio-based PP, UCO may be collected globally. Globally sourced UCO has been modeled considering the macro-areas, which together account for more than 90% of the 2017 Dutch consumption: Asia (40%), US (25%), West EU (20%) and Netherlands (15%) [436]. According to a market study by [437], China has the highest UCO collectable potential. We assume that UCO is shipped[14] from China (port Shanghai) to represent the imported Asian UCO. For the US and Western Europe, New York and London have been considered for calculating the transportation distances, while the collection in the Netherlands has been modeled assuming the same distances used for the baseline calculations. The first column of Table 26 shows the changes in the results of environmental impact results when UCO is globally sourced. Compared to the baseline analysis, the change of UCO sources leads to a significant increase in all 16 impacts. The most affected categories are particulate matter (+60%), photochemical ozone formation (+48%), acidification (+78%), terrestrial and marine eutrophication (+55-57%) and human toxicity without cancer effect (+22%). Overall, on a weighted basis, the environmental impact increases by 19%.

Another important assumption is related to the dataset used for hydrogen production, which is among the main environmental hotspots of this route. In the investigated system, hydrogen is produced from steam reforming of natural gas. According to the selected dataset [381], the production of 1 kg of hydrogen generates 9.4 kg CO_2eq. Nevertheless, literature reports GHG emissions from steam reforming of natural gas ranging from 8.9 to 12.9 kg CO_2 eq./kgH_2 [438]. Accordingly, the climate change impact of bio-based PP production could vary in the range

[14] From Ecoinvent 3.4: Transport, freight, sea, transoceanic ship {GLO}| market for | APOS)

from -2% to 12% considering other datasets. In the future, environmental impact reductions could be achieved if hydrogen could be produced using renewable energy, i.e. via electrolysis powered by photovoltaics [439]. In such a case, the climate change impact of UCO-PP could be reduced by a third. However, as green hydrogen is not expected to be commercially viable within the next decade, it is out of scope for our LCA, and therefore not investigated in more detail.

Table 26. Variation of cradle-to-factory gate impact assessment results by different assumptions on UCO collection and process optimization of using renewable propane and methane

Impact Category	**Increase of impact (%) shifting from UCO locally sourced to sourced globally**	**Increase of impact (%) (negative values stand for a decrease) changing from conventional LPG in the baseline to the renewable propane and methane scenario**
Climate change	10%	-42%
Ozone depletion	11%	-86%
Human toxicity, non-cancer effects	22%	-44%
Human toxicity, cancer effects	7%	2%
Particulate matter	60%	-31%
Ionizing radiation HH	6%	-41%
Photochemical ozone formation	48%	-5%
Acidification	78%	-17%
Terrestrial eutrophication	57%	4%
Freshwater eutrophication	12%	-15%
Marine eutrophication	55%	4%
Freshwater ecotoxicity	6%	-12%
Land transformation	13%	-95%
Water use	1%	0%
Resource use, minerals and metals	4%	2%
Resource use, fossil fuels	10%	-64%
Total weighted results	19%	-34%

LPG production and combustion has been identified as the second environmental hotspot of this route. The composition of LPG is variable depending on the location where it is sourced. For example, it can be 25% propane/75% butane in Italy and 95%/5% in Sweden [440]. In the baseline, we assumed that petrochemical LPG

7

is used to produce the UCO-based PP. However, it is possible to optimize the process energy requirement by using the propane from UCO obtained from the hydrotreatment to meet the need of LPG for dilution of the steam cracking and by using the methane produced in the steam cracking as energy source (see Figure 45). The environmental impact of UCO-based PP would vary as shown in the third column of Table 26. The overall environmental footprint of UCO-based PP would be reduced by 34%. In particular, these reductions of impact are high for climate change (-42%), ozone depletion (-86%), human toxicity without cancer effects (-44%), particulate matter (-31%), ionizing radiation HH (-41%), land use (-95%) and fossil resources (-64%). It is possible to notice that these reductions are even higher than the percentage of the impact caused by steam cracking on the overall environmental impact. This is consistent with the substitution approach used for steam produced. In fact, for these impact categories, the impact caused by steam cracking becomes negative when renewable propane and methane are used instead of LPG. The reason is that the impact caused by the production and combustion of renewable propane and methane (not the allocation of impact to methane produced by steam cracking) becomes lower than the credit for steam production (substitution).

Another important assumption is related to the datasets selected to model polymerization. As highlighted in the previous section, the impact of polymerization is particularly significant in five impact categories (on a weighted basis). For these impact categories, the share on the total impact has been validated using the data from the Matter study [432]. According to the Matter study, PP polymerization requires 2.1 MJ of electricity[15] and 1.3 MJ of steam [16] (averaged[17]) per kg of polypropylene. Using the inventory data from Matter study instead of PlasticsEurope (see section *Life cycle inventory modeling*), the share of polymerization would shift from 96% to 88% for water use, from 25% to 46% for climate change, from 22% to 40% for resource use of fossil fuels, from 54% to 48% from human toxicity (cancer) and from 54% to 59% for ionizing radiation. Overall, considering all the impact categories, the total share of polymerization would shift from 38% to 48% on a weighted basis. The authors consider the 9% difference in line with the different temporal scope of the two datasets (due to improvements in process efficiencies that have occurred

[15] Dataset used from Ecoinvent 3.4: Electricity, medium voltage {RER}| market group for | APOS

[16] Dataset used from Ecoinvent 3.4: Steam, in chemical industry {RER}| production | APOS

[17] Ranging between 0.8 and 1.8 MJ depending if the polymerization occurs in liquid phase, gas phase or suspension

over 20 years). Figure 47 summarises the results of the sensitivity analyses on data uncertainty on weighted bases. The variations are shown using the baseline values as 100%.

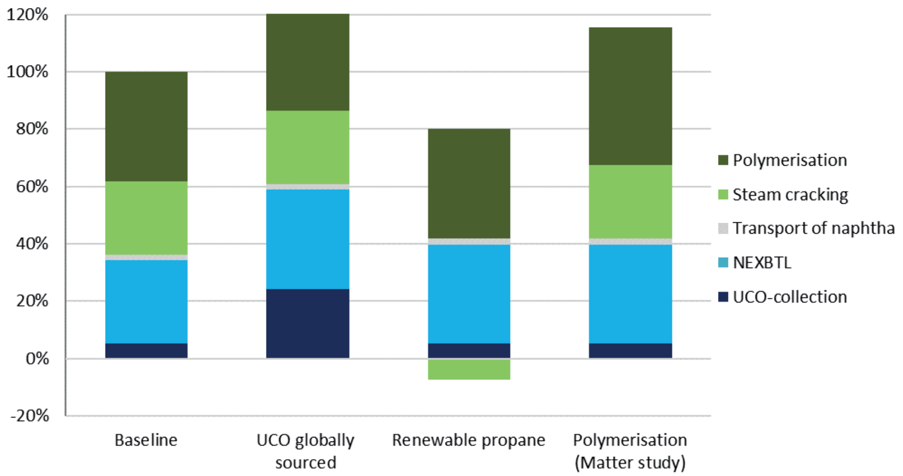

Figure 47. Sensitivity analysis on data uncertainty (normalized and weighted results with baseline values taken as 100%). Steam cracking using renewable propane and methane is shown as a negative impact, as their production and combustion have a lower impact than the credit for steam production (substitution). For more details, see the main text.

7.4.2 Model uncertainty: multifunctionality

This section provides a sensitivity analysis of the allocation approaches that were selected for the baseline calculations. The first sensitivity is related to UCO, which has been considered as a waste, being its use promoted in the European Union [9]. Accordingly, UCO has been treated with a cut-off approach in the baseline calculations. Nevertheless, the increasing demand for UCO for renewable diesel production has driven the high price of UCO in the past decade [435]. Accordingly, it might be argued that UCO should be considered as a by-product rather than a waste and a part of the impact caused in the first life (e.g. vegetable oil production) should be assigned to the recycled function (e.g. PP). For this sensitivity analysis, the 50/50 method instead of the cut-off approach is used. The 50/50 method assigns the credits and the burdens due to recycling to both previous (50%) and subsequent life cycle (50%). Accordingly, we allocate 50% of the impact of the primary production of vegetable oil to UCO. This method is the most conservative and it is usually applied for open-loop recycling when it is not known whether the use of the recycled material should be promoted [71].

Table 26 shows the variation of the environmental impact when the 50/50 method is applied to UCO. UCO is an oil waste derived from the use of oils and fats in cooking activities. For this sensitivity analysis, palm oil and soybean oil, whose data were available in the Ecoinvent database, were considered[18].

When 50% of the environmental impacts of the production of the vegetable oils are allocated to the second life, the impacts of UCO-PP are significantly increased. On a weighted basis, the impacts of UCO-based PP would increase from 25% to 160% depending on the types of primary vegetable oil. In particular, larger variations are obtained when UCO origins from soybean oil. For climate change and the use of fossil resources, the environmental impact would increase by 17%-58% and 10-28%, respectively.

Table 27. Variation of cradle-to-factory gate impact assessment results by using 50/50 method on UCO open-loop recycling

Impact Category	Increase (%) (negative values stand for a decrease) 50/50 Method palm oil	Increase (%) (negative values stand for a decrease) 50/50 method soybean oil
Climate change	17%	58%
Ozone depletion	6%	28%
Human toxicity, non-cancer effects	-12%	-3701%
Human toxicity, cancer effects	16%	166%
Particulate matter	113%	485%
Ionizing radiation HH	4%	12%
Photochemical ozone formation	42%	164%
Acidification	37%	127%
Terrestrial eutrophication	40%	155%
Freshwater eutrophication	37%	1282%
Marine eutrophication	55%	789%
Freshwater ecotoxicity	20%	3150%
Land transformation	3%	2496%
Water use	26%	20%
Resource use, minerals and metals	38%	347%
Resource use, fossil fuels	10%	28%
Total weighted results	25%	160%

[18] From Ecoinvent 3.4, Palm oil, crude {GLO}| market for | APOS, Soybean oil, crude {GLO}| market for | APOS.

The second sensitivity is related to the multifunctionality of hydrotreatment and steam cracking. In the baseline calculations, energy allocation was used for hydrotreatment, while direct substitution for net steam and energy allocation is applied for the steam cracking process. Alternatively, different allocation methods could have been followed:

- *Energy allocation (only, not combined with direct substitution)*. In this approach, energy allocation is applied to all the co-products of steam cracking including steam. All the products have been valued with their lower heating values (LHVs) while the energy value of steam has been considered to be its enthalpy. Unlike the hybrid method applied for the steam cracking process in the baseline, no credits for steam substitution have been assigned (strictly attributional LCA and consistent with RED).
- *Exergy allocation (only, not combined with direct substitution)*. Compared to the baseline, the only difference is that exergy allocation has been applied to the steam cracking unit (for hydrotreatment using exergy or energy allocation key is indifferent). The reason behind this choice is that exergy can account for different quality in energy carriers. The superheated conditions of the steam released by steam cracking are 520 °C and 110 bars. This flow has been valorized with the exergetic value of this steam at such conditions. This means that it is assumed that this steam is entirely recovered and directly used, e.g. as process steam input in other refinery processes. The other co-products are energy and chemical products whose exergy value has been approximated with their LHVs. For the steam in input to the steam cracking unit, Ecoinvent database has been used[16].
- *Cut-off*. Differently, from the baseline calculations, a cut-off approach is applied to the hydrotreatment unit process for bio-based naphtha. Due to the minor production share of bio-based naphtha compared to renewable diesel, all the environmental burdens of hydrotreatment are assigned to the renewable diesel. In this case, bio-based naphtha comes into the system as an 'emissions-free' input and, therefore, no impact has been apportioned for UCO collection and NEXBTL process. This is consistent with the model for this conversion route developed by the Joint Research Centre (JRC) of the European Commission [350]. In their model, HVO was the investigated product and the JRC applied a cut-off approach, neglecting the very small fraction of naphtha. The hybrid approach used for the steam cracking process remains unchanged.

7

- *Cut-off & energy allocation.* In this case, the cut-off approach is applied to the hydrotreatment unit process. Differently from the previous case, solely energy allocation is applied to the steam cracking unit.
- *By-products substitution.* In this case, system expansion followed by substitution is applied to all the by-products of hydrotreatment and steam cracking (but not to the co-products, i.e. diesel and ethylene). All the credits are assigned to the co-products diesel/naphtha and propylene/ethylene that is then partitioned by energy allocation. In the hydrotreatment unit, renewable propane has been assumed to replace petrochemical propane[19]. For the steam cracking unit, the following by-products are substituted with the conventional processes which would be avoided: steam, hydrogen, bio-methane, bio-based benzene, and bio-butadiene[20]. The other by-products (C5, C7, and C8) that have not been substituted have been considered as neutral, i.e. neither burdens or related credits are caused by them. For the baseline calculations, they were instead accounted in the energy allocation shares.

Figure 48 shows that energy and exergy allocation worsen the environmental footprint of bio-based PP. The only exception is resource use (minerals and metals) with energy allocation, although the difference is minimal (1%). This increase in impacts is caused by disregarding the credit from the substitution of steam in the baseline. Moreover, energy and exergy allocation lead to very similar results but impacts are higher in the case of exergy allocation. The reason behind this is that the exergy value of steam is lower than the enthalpy value. The weighted impact increases by 35%. Hence, it is concluded that exergy allocation is the most conservative among the assessed approaches.

The cut-off and by-product substitution approaches provide a significantly lower environmental footprint for bio-based PP. On the other hand, the by-products substitution approach is also the one assigning the highest impact for ionizing radiation HH and resource use of minerals and metals. The reason for this is that the credits for direct substitution of the by-products above do not compensate for the higher amount of impact apportioned to propylene. Moreover, negative impact results are obtained when system expansion followed by substitution is applied in

[19] From Ecoinvent 3.4, Propane {GLO}| market for | Conseq

[20] From Ecoinvent 3.4, Steam, in chemical industry {RER}| production | Conseq, Natural gas, from medium pressure network (0.1-1 bar), at service station {GLO}| market for | Conseq. Butadiene {RER}| production | Conseq. From PlasticsEurope's Ecoprofiles, Hydrogen (reformer) E from PlasticsEurope [381]), Benzene, at plant/RER based on PlasticsEurope Industry 2.0 database [449]

the following impact categories: human toxicity without cancer effects, particulate matter, freshwater eutrophication, freshwater ecotoxicity and resource use of fossil fuels. This is caused by the by-products of hydrotreatment (propane) and steam cracking (especially bio-methane and benzene), which displace products that have high impacts for these five categories.

Conversely to other methods, the weighted impact decreases by (-) 113% by using the by-products substitution method (overall negative impact) compared do the baseline values. These negative flows violate the desirable characteristics of an attributional LCA [51]. This negative impact means that the perturbation logic of substitution has created ''links between emissions and activities that are not mediated by product or service flows'' [51].

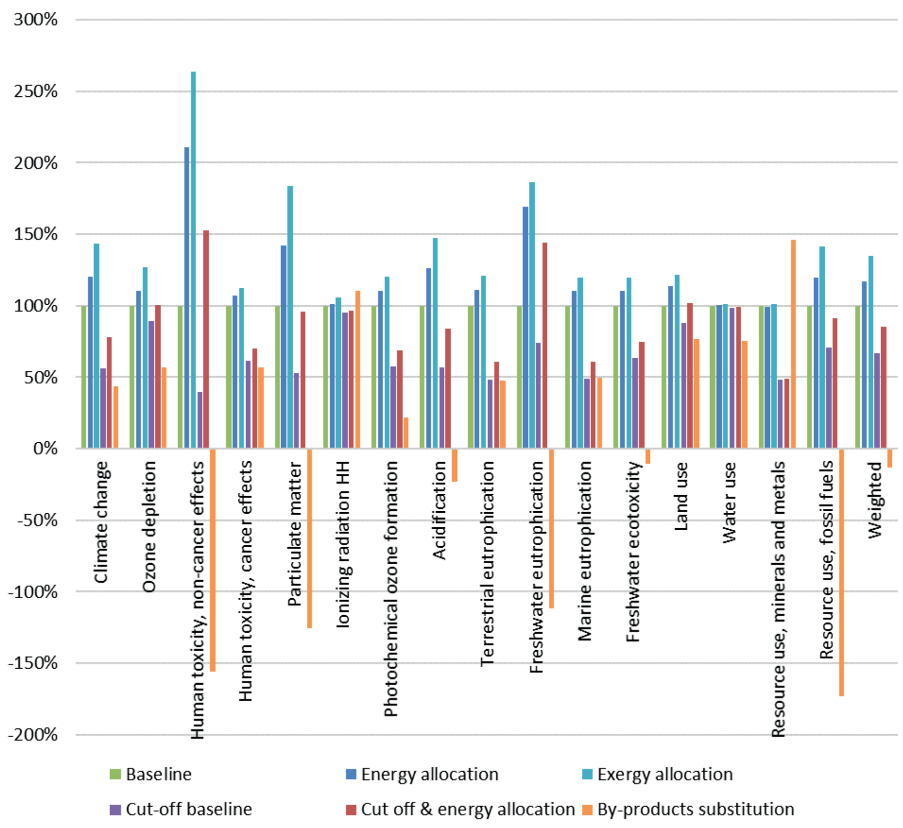

Figure 48. Sensitivity analysis of the allocation approaches for hydrotreatment and steam cracking units (numerical values in Appendix B). Baseline results expressed as 100%.

Moreover, the results presented in Figure 48 confirm the findings of Sandin et al. [57] that when a non-dominant product is in focus: 1) the results are sensitive to the choice of the allocation method and 2) the substitution method provides results in contrast with other allocation methods.

These two findings apply only when non-dominant products are in focus. Thamsiriroj and Murphy, who studied the same route but with renewable diesel (HVO) in focus (the physically dominant product), concluded that UCO HVO is relatively unaffected by allocation methodology (energy allocation, substitution, and cut-off approaches) [441]. Moreover, these results show that the RED statement that energy allocation provides results generally in line with the substitution method fails when a non-dominant product is investigated and cannot be extended to more impact categories than climate change.

The authors, therefore, recommend avoiding the use of the substitution method in attributional studies because its application can lead to results in contrast with other methods. The only exception can be the use of direct substitution for by-products when this can represent physical causality. Nevertheless, this should be based on a direct and empirically demonstrable relationship [212]. This would also be in line with ISO 14044 recommending allocation by-physical causality shall be preferred to other allocation methods. When a mathematical model is not available, the use of direct substitution shall be validated by comparing it with other allocation (partitioning) methods. For this case study, the baseline results, where direct substitution has been applied, have been validated by comparison to energy, exergy, and cut-off allocation methods. Moreover, the baseline results can be considered the most representative being an average among the possible allocation methods for all 16 impact categories and on a weighted basis.

7.4.3 Environmental benchmarking of UCO-based PP

The lack of harmonization in LCA method limits the direct comparison between bio-based and petrochemical materials studies, especially when considering multiple impact categories [7]. Specifically, such a comparison is often reliable only for climate change impacts [7]. In particular, this issue emerges when different black box datasets for petrochemical PP are compared (see Appendix C). From the analysis in Appendix C, it is possible to consider also resource use (fossil fuels) along with climate change as a reliable impact category (less than 10% variation).

From cradle to factory gate, 1 kg of petrochemical PP causes 1.65-1.78 kg CO_2eq GHGs and the resource use (fossil fuels) ranges between 67 -74 MJ (see Appendix C). To be conservative, the two lower values have been used for comparison. Hence, UCO-based PP (baseline) has a 62% lower impact on climate change and 86% for resource use (fossil fuels) compared to petrochemical PP.

Moreover, these reductions are almost unaffected when UCO is globally imported (58% and 85% respectively). By using the 50/50 method, these reductions are reduced to the minimum of 40% and 82% respectively for climate change and fossil resources use when UCO is derived from soybean oil. When exergy allocation is used for hydrotreatment and steam cracking, these environmental impact reductions are instead reduced to a minimum of 45% in terms of climate change and to 80% as resource use of fossil fuels. From these results, it can be concluded that UCO-based PP is a favorable alternative option to petrochemical polypropylene in terms of climate change and fossil fuel resources.

It should be kept in mind that these impact reductions are for the cradle-to-factory gate scope and the differences above in climate change impact do not account for BCR. Thus, UCO-based PP has the potential advantages to act as a biogenic carbon sink if the material is recycled. Moreover, biogenic carbon emissions are released when burned in a waste-to-energy system. These advantages are out of the scope of this LCA and therefore not estimated.

Comparing our results with the LCA published by Kikuchi et al. [415], it is found that UCO-based PP shows about 80/90% lower impact on climate change compared to bio-based PP made from sugarcane and woody biomass at factory-gate.

It should be recognised that UCO is a very limited feedstock. The European Commission already promotes its use for renewable diesel as a second-generation biofuel [434]. UCO-based PP is developed from the bio-based naphtha, which is a by-product of this renewable diesel. From an environmental perspective, it would be therefore interesting to assess what is more attractive between replacing petrochemical diesel or polypropylene. “Sidestream naphtha” case was chosen as baseline due to the current strong market demand for renewable diesel (main product). Nevertheless, the cracking of all HVO diesel and naphtha would be technically feasible. Increasing the mixture of bio-based HVO naphtha and diesel used for bio-based PP would not lead to a different environmental impact compared to the baseline calculation due to the slightly different LHVs assumed for bio-based naphtha and diesel (energy allocation). It is known that UCO renewable diesel could lead to GHG emissions saving up to 88%, which are much higher compared to 40-62% savings by other biodiesels [435]. These savings could appear higher than the ones allowed by UCO-based PP. Nevertheless, we are not able to answer this question properly without proper modeling for the end of life of polypropylene.

7.5 CONCLUSIONS

The first objective of the study is to identify the major environmental burdens in the cradle-to-factory gate life cycle of UCO-based PP. The environmental footprint of UCO-based PP is dominated by the polymerization process (38%), the production of hydrogen (21%), the production of LPG (18%) and the combustion (8%) of LPG. Climate change (28%), fossil resource use (23%) and water use (11%) have resulted in being the most important environmental impacts of UCO-based PP.

It is found that if the renewable propane and methane from UCO could be used instead of LPG, the environmental footprint of UCO-based PP could be potentially further reduced by 34%. Bio-based PP from UCO might also offer significantly better environmental performances in terms of climate change compared to bio-based PP from sugarcane and woody biomass (-80/90% at factory gate).

Compared to petrochemical PP, UCO-PP offers substantial impact savings for climate change (62%) and for fossil fuel resource use (86%). These savings remain substantial even in the cases of 1) globally imported UCO (58% and 85% respectively), 2) when UCO is considered a by-product instead of waste (40% and 82% respectively based on the 50/50 method) or 3) when a different allocation approach is used for hydrotreatment and steam cracking (savings of 45% and 80%, respectively). From these results, it can be concluded that bio-based PP from UCO is a promising alternative option to replace petrochemical polypropylene in terms of climate change and fossil fuel resources.

It should be reminded that the comparisons made above are for the scope of cradle to factory gate. UCO-based PP has the further advantage of having a 100% biogenic carbon content embedded in the product, potentially for the long term (e.g. in a durable application). The full biogenic carbon balances should be accounted for in a future cradle to grave LCA when a final product made from UCO-PP is analysed.

The second objective of the study is to scrutinize how the allocation procedures used to solve multifunctionality affect the results of this LCA. It is found that exergy allocation leads to an increase of 35% of the environmental footprint of UCO-based PP compared to the baseline in which a hybrid direct substitution and energy allocation is applied. Conversely, the environmental footprint would become negative by using system expansion followed by substitution. The negative footprint obtained by using substitution is because bio-based naphtha and propylene, which are the precursors of UCO-based PP, are two physically non-dominant output-products of multifunctional processes. Such a negative impact is a clear violation of the desirable characteristics of an attributional LCA. It is recommended to avoid the use of system expansion followed by substitution in attributional studies. The only exceptional cases are the ones where direct substitution of by-products can represent physical causality like applied to the steam produced during steam cracking. This steam produced is directly used by other processes of the same biorefinery, and, otherwise, should be produced as marginal production of refinery steam. In fact, this would result in line with ISO 14044 recommending allocation by physical causality shall be preferred to other allocation methods. As a mathematical model is not available to model physical causality relationships, the use of direct substitution for steam has been validated by comparing with other allocation methods, showing alignment in all the impact categories assessed.

It is concluded that economic significance should be considered as an important requirement to fulfill before applying substitution. When this requirement is not respected, direct substitution shall be avoided.

Last but not least, UCO has been used as feedstock for a variety of applications of chemicals and fuels. The increasing demand for UCO has driven the price up in the past years. In this study, the impact of considering UCO as a by-product instead of a waste was assessed. It is found that the LCA results could significantly vary depending on the type of original vegetable oils as well as how the allocation is performed (e.g. based on the 50/50 approach). The results provided in this study should be used to elicit the discussion in the context of assessing the impacts of products in a future circular and bio-based economy.

7.A APPENDIX: DETAILED BREAKDOWN PER LIFE CYCLE STAGE AND INTERPRETATION

Table 7A.1 provides the numerical breakdown of the environmental impact assessment per unit process. Table 7A.2 shows the same results on weighted basis.

Table 7A.1. Breakdown of cradle-to-factory gate impact assessment results (1 kg of bio-based PP)

Impact Category	Unit	UCO-collection	NEXBTL	Transport of naphtha	Steam cracking	Polymerization
Climate change (w/o BCR)	kg CO_2eq	4%	40%	2%	29%	25%
Ozone depletion	kg CFC-11 eq	5%	7%	0%	74%	14%
Human toxicity, non-cancer effects	CTUh	32%	38%	5%	-71%	96%
Human toxicity, cancer effects	CTUh	4%	36%	6%	1%	54%
Particulate matter	kg PM2.5 eq	10%	40%	4%	6%	39%
Ionizing radiation HH	kBq U235 eq	3%	4%	1%	40%	54%
Photochemical ozone formation	kg NMVOC eq	7%	37%	4%	21%	31%
Acidification	molc H+ eq	6%	39%	4%	9%	43%
Terrestrial eutrophication	molc N eq	7%	46%	5%	19%	23%
Freshwater eutrophication	kg P eq	5%	26%	6%	-59%	121%
Marine eutrophication	kg N eq	7%	45%	4%	19%	24%
Freshwater ecotoxicity	CTUe	24%	16%	1%	9%	50%
Land transformation	kg C deficit	8%	6%	1%	80%	5%
Water use	m^3	0%	1%	0%	0%	99%
Resource use, minerals and metals	kg Sb eq	29%	28%	2%	14%	27%
Resource use, fossil fuels	MJ	4%	25%	1%	47%	22%

Table 7A.2. Breakdown of cradle-to-factory gate impact assessment results on weighted basis

Impact Category	Unit	UCO-collection	NEXBTL	Transport of naphtha	Steam cracking	Polymerisation
Climate change (w/o BCR)	kg CO_2eq	1%	11%	0%	8%	7%
Ozone depletion	kg CFC-11 eq	0%	0%	0%	0%	0%
Human toxicity, non-cancer effects	CTUh	0%	0%	0%	-1%	1%
Human toxicity, cancer effects	CTUh	0%	2%	0%	0%	3%
Particulate matter	kg PM2.5 eq	1%	3%	0%	0%	2%
Ionizing radiation HH	kBq U235 eq	0%	0%	0%	2%	3%
Photochemical ozone formation	kg NMVOC eq	0%	2%	0%	1%	2%
Acidification	molc H+ eq	0%	2%	0%	0%	2%
Terrestrial eutrophication	molc N eq	0%	1%	0%	0%	1%
Freshwater eutrophication	kg P eq	0%	0%	0%	0%	0%
Marine eutrophication	kg N eq	0%	1%	0%	0%	0%
Freshwater ecotoxicity	CTUe	0%	0%	0%	0%	0%
Land transformation	kg C deficit	0%	0%	0%	2%	0%
Water use	m^3	0%	0%	0%	0%	10%
Resource use, minerals and metals	kg Sb eq	0%	0%	0%	0%	0%
Resource use, fossil fuels	MJ	1%	5%	0%	11%	5%

7.B APPENDIX: MULTIFUNCTIONALITY SENSITIVITY ANALYSIS

Table 7B.1 provides the numerical results of the sensitivity analysis on multifunctionality.

Table 7B.1. Numerical values of sensitivity analysis on multifunctionality

Impact Category	Unit	Energy allocation	Cut off & energy allocation	Exergy allocation	Baseline	Cut-off baseline	By-products substitution
Climate change (w/o BCR)	kg CO_2eq	7.6E-01	4.9E-01	9.1E-01	6.3E-01	3.6E-01	2.8E-01
Ozone depletion	kg CFC-11 eq	9.9E-08	9.0E-08	1.1E-07	9.0E-08	8.0E-08	5.1E-08
Human toxicity, non-cancer effects	CTUh	2.3E-08	1.7E-08	2.9E-08	1.1E-08	4.4E-09	-1.7E-08
Human toxicity, cancer effects	CTUh	5.9E-09	3.9E-09	6.2E-09	5.5E-09	3.4E-09	3.1E-09
Particulate matter	kg PM2.5 eq	1.7E-04	1.1E-04	2.2E-04	1.2E-04	6.2E-05	-1.5E-04
Ionizing radiation HH	kBq U235 eq	6.4E-02	6.1E-02	6.7E-02	6.4E-02	6.0E-02	7.0E-02
Photochemical ozone formation	kg NMVOC eq	2.1E-03	1.3E-03	2.3E-03	1.9E-03	1.1E-03	4.2E-04
Acidification	molc H+ eq	2.6E-03	1.7E-03	3.0E-03	2.1E-03	1.2E-03	-4.8E-04
Terrestrial eutrophication	molc N eq	6.7E-03	3.7E-03	7.3E-03	6.0E-03	2.9E-03	2.9E-03
Freshwater eutrophication	kg P eq	1.5E-05	1.3E-05	1.6E-05	8.7E-06	6.4E-06	-9.7E-06
Marine eutrophication	kg N eq	6.1E-04	3.4E-04	6.7E-04	5.6E-04	2.7E-04	2.8E-04
Freshwater ecotoxicity	CTUe	2.4E-01	1.6E-01	2.6E-01	2.2E-01	1.4E-01	-2.4E-02
Land transformation	kg C deficit	1.2E+00	1.1E+00	1.3E+00	1.1E+00	9.6E-01	8.3E-01
Water use	m^3	7.4E-01	7.3E-01	7.4E-01	7.4E-01	7.3E-01	5.6E-01
Resource use, minerals and metals	kg Sb eq	3.1E-07	1.6E-07	3.2E-07	3.2E-07	1.5E-07	4.6E-07
Resource use, fossil fuels	MJ	1.1E+01	8.5E+00	1.3E+01	9.3E+00	6.6E+00	-1.6E+01
Weighted	p	6.0E-03	4.3E-03	6.9E-03	5.1E-03	3.4E-03	-6.8E-04

7.C APPENDIX: PETROCHEMICAL POLYPROPYLENE?

Large differences in the most of the impact categories can be noticed when comparing PlasticsEurope[21] and Thinkstep Gabi[22] datasets for petrochemical PP. The result of this comparison is presented in Figure 7C.1. Due to the large variations, these dissimilarities cannot be justified only by the different geographic scope (Europe and Germany).

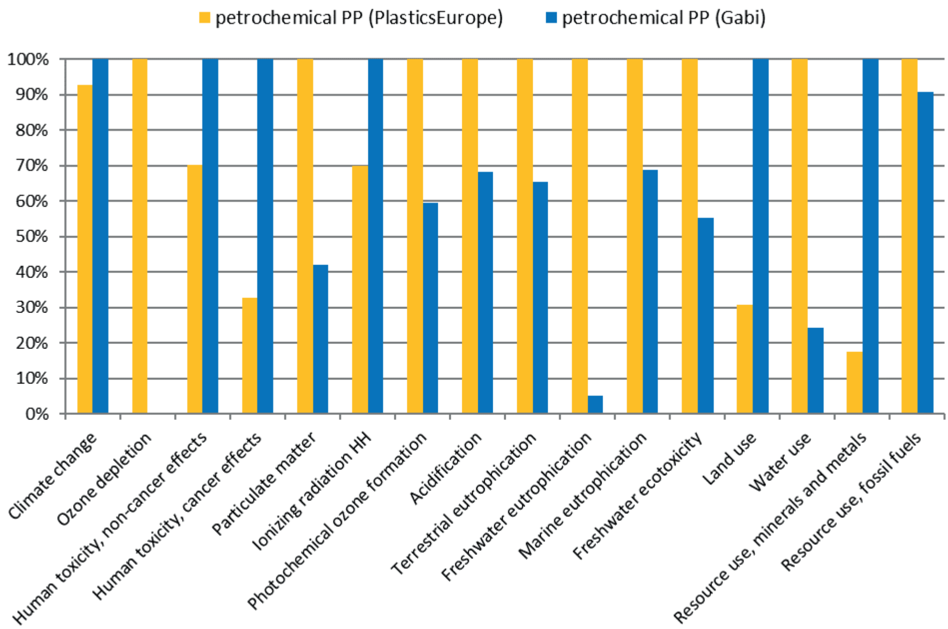

Figure 7C.1. Variation of impact for petrochemical PP comparing different datasets.

[21] Polypropylene, PP, granulate, at plant/RER

[22] DE: Polypropylene granulate (PP) mix ts

CHAPTER 8

Summary, conclusions and recommendations

8. SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

There is a growing interest in converting local bio-based “residues” streams into high-value products i.e. chemicals, energy and materials. Compared to bio-based products from dedicated crops, this type of bio-based product can limit potential trade-offs such as land competition, potential biodiversity damages and food security issues. Moreover, local bio-based residues do not require significant imports of bio-based products and biomass from outside the EU. Furthermore, avoiding dedicated cultivation, products from residual flows are expected to have a much lower environmental impact than bio-based products from dedicated crops.

However, the literature has previously emphasized that calculating the life cycle environmental impact of products from bio-based residues is challenged by several methodological uncertainties. In particular, one of the most relevant uncertainties regards the so-called LCA multifunctionality issue, i.e. the allocation of environmental burdens among the process co-products. In fact, a variation in the allocation type or in the input data used for determining allocation factors has a much higher effect on the environmental burdens of low-economic/physical significance streams than for the main products.

On top of it, selecting the multifunctionality approach is challenged by various inconsistencies in the reported interpretation of the so-called ISO multifunctionality hierarchy in ISO-compliant LCA guides. Since different guides are prominent references in different bioeconomy sectors or countries, inconsistent multifunctionality practices are claimed as “ISO-compliant” in LCAs of bio-based products. Consequently, significantly different results for products with similar life cycle inventory data are reported in the literature. The literature has acknowledged such inconsistency several times. However, the origins of such inconsistent interpretations are not fully understood and the magnitude of such inconsistency among practices applied in different bioeconomy sectors has never been quantified so far.

For this reason, the first research question (RQ) that this thesis aimed to answer was: *what multifunctionality practices are adopted in LCAs of bio-based products and how can the consistency of the life cycle inventory model be improved?*

In this thesis, current practices in implementing ISO 14044 multifunctionality recommendations in LCAs of bio-based products have been quantitively reviewed for the first time. The same review also contains an investigation of the origins and rationales of ISO multifunctionality hierarchy's interpretations. This investigation was fundamental to select the multifunctionality practices adopted in the LCA case studies presented in this thesis by considering only those fully "ISO compliant". While the ISO-guidelines review (Chapter 3) quantified multifunctionality practices for products in various bioeconomy value chains, the lignin–use review (Chapter 2) looked at those applied in LCAs related to one of the feedstocks of interest, i.e. lignin.

Besides reviewing the LCAs of various lignin applications, four LCAs were conducted for bio-based asphalts utilizing lignin, bioenergy from wood chips, a bio-jet fuel from potato by-products and polypropylene from used cooking oil. The selection of these feedstocks prioritized 1) bio-based by-products that are locally available in the EU (with a major focus on the Netherlands) and 2) those for which the collection of primary data for modeling the respective innovative conversion technologies was possible.

These LCAs were conducted to answer this thesis's second research question (RQ2) is: *What is the environmental impact of novel bio-based products made from by-product/waste streams compared to their fossil counterparts*?

All the investigated high-value bio-based products are produced using innovative emerging technologies to convert agro-industrial and food-processing by-products. This thesis presents the life cycle environmental impact of several innovative technologies for the first time as well as their environmental performance compared to their petrochemical counterparts. More than just climate change, this thesis aims to look at the broad picture of the environmental impacts of innovative bio-based products from residual streams and their environmental hotspots in multiple impact categories. In this way, it was possible to understand if the lower impact of residue-based products compared to bio-based products from dedicated crops also applies to other environmental issues than climate change. Accordingly, environmental impacts that are generally higher for bio-based products than petrochemical products, such as eutrophication and acidification, were considered. The analysis of the environmental hotspots was fundamental to recommend possible ways to reduce the overall environmental footprint of each product robustly. For each proposed action, potential trade-offs in burdens-shifting were analyzed. Furthermore, several comparisons between products from bio-based by-products and comparable bio-based products from dedicated crops were also performed.

Since the scientific literature highlighted the magnitude of multifunctionality uncertainty on the reported climate change impacts of bio-based products from residual streams, this thesis aims to answer the third RQ: *How are the LCA results of such products influenced by the approach adopted to deal with the multifunctionality issue*?

Various sensitivity analyses on the multifunctionality approach were conducted for the LCAs performed. Previous literature assessing bio-based by-products mainly performed this type of analysis on climate change and rarely on a broad spectrum of environmental indicators. Conversely, this thesis aims to understand the environmental impact categories for which the multifunctionality uncertainties hamper the comparison with the petrochemical counterparts, making it inconclusive.

8.1 RESEARCH QUESTION 1

1. *What multifunctionality practices are adopted in LCAs of bio-based products and how can the consistency of the life cycle inventory model be improved?*

The first part of this research question i.e. *what multifunctionality practices are adopted in LCAs of bio-based products*? was provided in two reviews. The review presented in Chapter 2 focused on the effect of multifunctionality practices in comparing the results from 42 peer-reviewed LCAs regarding lignin and derived products.

The review presented in Chapter 3 looked at practices in implementing ISO 14044 multifunctionality recommendations in LCAs of products belonging to diverse bioeconomy sub-sectors and LCAs of the petrochemical products used as the benchmark to calculate their environmental performance.

The second part of RQ1 i.e. *how can the consistency of the life cycle inventory model be improved*? was answered based on the review presented in Chapter 3 using key-route main path analysis to identify the major publications that influenced the debate under the interpretation of the ISO-multifunctionality hierarchy. This exercise allowed us to detect the origins of the different interpretations, their underlying theories and not-(fully) ISO compliant practices. These insights were fundamental to 1) select the allocation method in all LCA case studies reported in the following chapters and 2) prioritize fully ISO-compliant methods as alternative methods applied in the sensitivity analyses.

8.1.1 Lignin use-review

Lignin is the second most abundant natural biopolymer on Earth and accounts for about 30% of the organic carbon in the biosphere [56,266]. The complexity of the lignin molecule and its physical characteristics make the conversion into higher-value products interesting and challenging at the same time. Chapter 2 presented a review of 42 peer-reviewed LCAs regarding lignin and derived products. Most emerging high-value lignin-based applications presented considered by these 42 studies seem to offer promising climate change performances. From a climate change perspective, transportation fuels, adipic acid and propylene seem the most promising applications currently at the development stage to utilize lignin.

Lignin, which is currently mainly used as low-value on-site fuel, is always a by-product from a multifunctional production process of pulp mills or lignocellulosic biorefineries. As shown by the summary of the adopted multifunctionality practices in the 42 reviewed LCAs illustrated in Figure 4, a wide variety of multifunctionality practices were applied. This fact is not a problem per se but is a problem if the rationale for their selection is inconsistent or they derive from inconsistent application of ISO 14044:2006 recommendations. Mass allocation was the most adopted method to deal with lignin multifunctionality (13 out of 42 studies). However, this method has the limit of assigning the same impact per kg of product to lignin and the main products of the multifunctional system (pulp mill or lignocellulosic biorefinery). This does not reflect the value of lignin (e.g. energetic or economic) and the fact that lignin is a by-product and not the main product of pulp mills and lignocellulosic biorefineries. Three studies also applied the cut-off method. However, a cut-off allocation does not seem appropriate if lignin cannot be considered a waste in both ISO LCA and EU legislation terms (e.g. see the EU waste framework directive and the updated EU Renewable Energy Directive). This circumstance applies to lignin with a market price that cannot be treated as a waste. Moreover, if lignin is no more internally combusted and a different fuel is used, the environmental impact of pulp or the bio-based products from the lignocellulosic biorefineries is affected.

Substitution was adopted as a system expansion approach by four studies. However, this method could also lead to numerically negative impacts for lignin if the main product is erroneously substituted. Regarding such a result, some authors of the articles reviewed in Chapter 2 referred to inconsistent results caused by doubtful interpretation of ISO 14044:2006. Accordingly, they state 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 co-product flows for bio-based chemicals" [65]. <u>However, applying substitution to obtain the impact of lignin by subtracting an alternative production of pulp is conceptually not correct and not an option allowed by ISO 14044:2006.</u> In fact, even in consequential LCAs, only by-products can be substituted and not the main product. <u>A check on co-products' economic significance is necessary before applying substitution to avoid erroneous application (as remarked before also by several authors [51,211] and the ILCD handbook [70]). However, some authors [57] remarked that ISO does not emphasize this aspect enough.</u> Conversely, the substitution method would be applicable for those LCAs focusing on the main product of a system co-producing lignin. In those cases, lignin could be substituted if it is not an economically significant product i.e. a by-product.

Even for climate change only, allocation practices could significantly affect the environmental impact of lignin and derived applications. Taking the example of lignin-derived transportation fuels, the allocation method applied was one of the two reasons for the significant change of the emissions savings compared to diesel ranging between 10% and 90% (the energy source i.e. biomass or fossil fuels, was the second main factor). The cut-off allocation (lignin considered a by-product free of burden) led to the highest climate change savings for lignin-based transportation fuels.

<u>Applying allocation to the biogenic carbon was one of the main comparability issues for the reported climate change impacts of lignins in the literature. However, the actual biogenic carbon content of the final products should be preferred to an allocated biogenic carbon, since there is no allocation parameter (physical or economic) that can reflect how the carbon from biomass ends in the co-products.</u> For example, bio-based products with no carbon content can be derived from biomass feedstock with an important carbon content (e.g. bio-based hydrogen could be obtained from wood chips).

The reviewed case studies showed promising climate change performances and trade-offs in other impact categories with some conflicting results for similar lignin-based applications. For example, divergence was found in the outcome of comparisons with petrochemical counterparts of three LCAs of lignin-based adhesives [104,115,132] for the same end-point categories i.e. damage to human health, ecosystem quality and resources. Arias et al. [115] concluded that lignin-based adhesives have between 2.5 and 4.5 times higher impact than conventional adhesives. At the same time, the other two LCAs showed a better performance for lignin-based adhesives in the same categories. However, the study from Arias et al. lacks transparency on the allocation practices and handling of biogenic carbon for lignin, limiting a full understanding of the reasons leading to the different outcomes. The same lack of transparency on these critical methodological choices was reported in the only LCA of lignin-based asphalts found and reviewed in Chapter 2 [134]. Since dealing with allocation and biogenic carbon significantly affects the outcome of the LCAs of bio-based products, it is strongly recommended to illustrate their modeling transparently.

8.1.2 ISO guidelines -review

Regarding substitution, the review of Chapter 3 clarified that the concept of equivalency between substitution and system expansion originally illustrated by Tillman and Ekvall [80,81] does not mean that they are equivalent in practice. In fact, system expansion and substitution do not provide the same results (as remarked already remarked before by several authors [240]). Moreover, when the first ISO hierarchy was drafted, system expansion as enlargement and not as substitution was the only option. The ISO compliance of system expansion in the form of substitution originated from the publication of the annex of ISO 14041. However, this annex stated that substitution is possible only if the following conditions are met: 1) the LCA study's goal is assessing the long-term marginal effect of a change and 2) the change modeled can be predicted with low uncertainty. This annex was then removed from the current ISO 14044:2006. However, nowadays, the LCA goal and modeling described in that annex are well-known under "consequential LCA". However, this term became recognized much later than when this annex was published.

Accordingly, in the attributional LCAs presented in this thesis, system expansion was applied only via enlargement, i.e. as literally explained in ISO 14044: "expanding the product system to include the additional functions related to the co-products," e.g. modifying the functional unit to include all co-products. Such expansion by enlargement is applicable in both attributional and consequential LCAs. Still, it cannot be applied if the study's goal requires obtaining the impacts of just one of the co-products or by-products because such results would not be available (e.g. also remarked by [234,442]). In these cases, ALCA cannot avoid allocation while applying substitution as a system expansion method is instead possible for CLCA.

On this basis, distinguishing ALCA/CLCA and using substitution as a system expansion method only in CLCA could be a way to increase consistency in future LCAs of bio-based products (and others). Comparing ALCA results only with ALCA results and not with CLCA results would increase the comparability of the results in more categories than climate change. The text mining process applied in Chapter 3 highlighted that the practice of strictly distinguishing between attributional and consequential modeling is already implemented in 25% of the LCAs in the scientific literature with a highly variable percentage among bioeconomy subsectors (see Figure 10). Some guides [85,267,443] specify their modeling approach as attributional and accordingly do not allow substitution as a system expansion method. However, some authors [68,202] recommend that the distinction ALCA/CLCA should also be present in future ISO 14044 to avoid substitution in attributional LCAs.

The review presented in Chapter 3 also clarified the nature of the so-called ISO "physical relationships" since they have been interpreted uniformly by the studies belonging to the main path. Physical relationships used to define the allocation criterion in ISO's second level should represent "causal relationships" instead of relationships based on any physical parameter. Such relationships can be modeled only if the ratio of the output products can be varied i.e. the functional outputs can be varied independently. This allows establishing physical causality between functional units via mathematical modeling by changing the operating conditions (a concept already remarked by several authors [97,157,214–217]). Hence, a future ISO 14044 should remark on this concept. However, this interpretation is currently present only in 28% of LCAs on multifunctional bioeconomy systems (see Figure 12 in Chapter 3).

Conversely, such interpretation was adopted far more widely (see Figure 12 in Chapter 3) in LCAs of petrochemical products (>60%). Oil refining processes have been historically the most cited example of applying physical causality allocation based on linear programming (LP). For this reason, it is still common practice to use this method for oil products. For example, LP-based marginal refinery emissions are incorporated in the so-called fossil fuel comparator i.e. the reference value of carbon emissions of fossil fuels used to calculate the GHG emissions savings of biofuels in the EU legislation [9,37]. The selection of the allocation method based on physical causality is also common practice in LCAs of dairy and meat products (≈40-45%). In this case, the reason is that physical relationships are interpreted as physical causal relationships by a leading guide developed by the International Dairy Federation (IDF) [223]. Physical parameters can seldom represent physical causality. The most cited exception is the transportation of heavier products that linearly increase the truck's fuel consumption. As remarked by ISO 14049:2012 (i.e. the ISO report on applying ISO 14044 to the inventory analysis), the reason is the "linear interdependence of the fuel consumption and the load mass" independently if the additional mass is of product A or B. Such linear independence implies that mass allocation quantitatively reflect how the input (fuel) is changed by changes in the amount of each product of the system i.e. the transported load (mass of e.g. product A).

Except for these rare cases, if detailed modeling (e.g. LP) is not possible/too time-consuming or the ratio between products is fixed, it is necessary to allocate by "other relationships". The third-level ISO allocation is predominant in bioeconomy LCAs, but the selection procedure adopted is often not transparent with missing sensitivity analyses [83,94,285].

The review of Chapter 3 clarified that "other relationships" should be interpreted as other causal relationships whose allocation key (e.g. economic value, energy content, or mass basis) should be based on a proxy of physical causality (e.g. earlier remarked by [201,215,222,232]). Hence, the frequent allocation practice of using an arbitrary parameter for which no causality can be justified should be avoided. If the co-product ratio cannot be varied, no physical property can be used as a proxy for physical causality (on this last point, there seems to be a shared consensus by experts on LCA multifunctionality, as remarked recently by [444]). If the ratio can be varied, the guidance for justifying the "other relationship" selection is given (insufficiently) only by ISO 14049:2012 and, unfortunately, not in the more known main text of ISO 14044:2006. How to approximate causal relationships in this last case has been a debated scientific issue for two decades.

According to ISO 14049:2012, economic allocation is often the preferred option (see the example of bitumen). Economic allocation well represents why the products are produced (i.e. to generate revenues). However, the preference for a physical parameter is often defended if the goal of the LCA is to "understand and manage the environmental implications of the material and energy flows associated with efforts to meet human needs through economic activity" [444]. In this way, it is possible to directly link inputs, outputs, and impacts via allocation by physical parameters. However, material flow analysis more than LCA is the correct tool to provide the information required by this goal definition (as remarked recently by [444]).

On the other hand, there are also a small number of cases when economic allocation does not well represent (other) causal relationships. In this case, the choice of the allocation parameter remains value-laden. A typical example is a waste management process requiring the allocation of its environmental burdens and benefits to the different waste inputs. Waste management is not a human activity meant only to generate revenues selling a product e.g. energy or a recycled bottle, but also a necessary service to treat various wastes and/or give them a new life. Suppose the treated products are uniform, e.g. an average mix of plastic wastes with food contamination. It is necessary to allocate the environmental burdens (and benefits) from their treatment to the original plastic products and food products. The price of the original food product contaminating the plastic waste might be much higher than that of the plastic item e.g. a packaging film. In that case, the environmental impact and energy generated by incinerating such wastes can be assumed to be linearly proportional to the energy contents of the treated plastic wastes (input 1) and food contamination (input 2). At the same time, there is a much lower proportionality to the economic values of food and plastics. Suppose the same waste flow is treated via recycling. In that case, the environmental impact of the recycling process and the amount of recycled plastic varies linearly with the mass of plastic waste (input 1) and not with the mass of food contamination (input 2). For this reason, ISO 14044 states that mass/energy allocation, the number of useful cycles and economic allocation are all possible options for open-loop recycling.

8.1.3 List of recommendations

Based on the lessons learned from Chapters 2 and 3, the following summary of recommendations to properly deal with the multifunctionality issues in LCAs of bio-based products can be formulated:

1. The rationale of the multifunctionality approach should always be transparently illustrated;
2. Allocation parameters not reflecting the ISO causality principle should be avoided;
3. By-products with a market value should not be considered wastes, and as such cut-off allocation should be avoided.
4. LCA results of a bio-based and conventional product should be compared based on allocation methods following the same principle (i.e. ISO causality);
5. Such an allocation should reflect the aim of the process of producing the main product(s) and not the by-product.
6. Using the distinction between attributional and consequential modeling becomes an important way to increase the consistency and comparability of results further, and ISO should provide their definitions and uses since those proposed by SETAC are not universally accepted (as recently remarked by Schaubroeck et al. [59]); On this basis, consequential LCA modeling should be used complementarily to attributional LCAs and not using hybrid modeling for a correct picture of the environmental impacts.
7. The use of substitution should be linked with a specific goal of the LCA, i.e. assessing a change via consequential modeling;
8. Before applying substitution in consequential LCAs, a check on the economic significance of co-products is necessary. Such a check would avoid an erroneous generation of negative impacts by erroneously substituting main products instead of by-products
9. Allocating the biogenic carbon content of bio-based materials should be avoided, preferring the tracking of the physical biogenic carbon content in the final products. In fact, there is no single allocation parameter (physical or economic) that can reflect physical causality for the carbon content of co-products.

8.2 RESEARCH QUESTION 2

What is the environmental impact of novel bio-based products made from by-product/waste streams compared to their fossil counterparts?

8.2.1 Lignin-based asphalts

Given lignin's high biogenic carbon content, there is interest in developing a durable and circular application that could store such biogenic carbon permanently. For this purpose, roads with bio-based asphalts using lignin to replace bitumen are currently tested worldwide, e.g. from the Netherlands [255] to Australia [134]. Chapter 4 investigated the environmental impact of various bio-based asphalts with kraft-lignin in the Dutch context. A cradle-to-grave attributional LCA was conducted, i.e. from the extraction of all asphalt's ingredients to its recycling at the end of life, for a functional unit of 1 t of top-layer asphalt. The environmental impact was calculated for 11 impact categories and was weighted to obtain an environmental cost indicator, i.e. a single monetary value score representing the avoided environmental damage cost or shadow cost [257]. Various top-layer asphalts were considered. i.e. stone mastic asphalts, asphalt concretes and porous asphalts. Both natural gas and biomass hog fuel were considered as possible steam sources to replace the part of black liquor no more burnt in a recovery boiler.

For both climate change and the weighted environmental damage, the production of the raw materials, i.e. bitumen and lignins (excl. biogenic carbon intake), were the main environmental impacts of the asphalt. After recycling, the benefits of their second life largely compensated for the impacts of lignin and bitumen productions. Such benefits were calculated using the formula proposed by the respective Dutch Product Category Rules. Regarding kraft lignin production, steam production with natural gas dominated the climate change impact. Together with steam production (with natural gas or hog fuel), liquid carbon dioxide and sulfuric acid were the other main source of environmental impact for lignin production in the other ten categories. On the life cycle of the asphalt, another main source of environmental impact was the energy for the production process (i.e. natural gas and electricity).

The results of the LCA revealed a climate change impact reduction between 30% and 75% for top-layer asphalts using lignin compared to conventional asphalts. Hence, the cradle-to-grave LCA of bio-based asphalts showed that storing the biogenic carbon for 100 years in a durable application that is recycled over time could be a way to mitigate the climate change impact of the road construction sector. The percentage of lignin replacing bitumen and the steam source used by the pulp mill to replace the fraction of black liquor were the two main factors influencing bio-based asphalts' overall environmental cost performance.

A sensitivity analysis was conducted on the assumed functional unit and product system. A functional unit of 1 m^2 of asphalt of three layers (top, middle and base) instead of 1 t of top-layer asphalt was selected in such a sensitivity analysis. Changing the functional unit, i.e. from 1 t to 1 m^2 and product system, i.e. from top-layer only to the entire asphalt block made of three layers, did not alter the outcome of the comparison between bio-based and conventional asphalts. On the other hand, the difference in weighted environmental damage between lignin-based and conventional asphalts became minor since lower layers have a higher percentage of recycled content (burdens-free) and a lower amount of bitumen/lignin (less binder needed in lower layers).

8.2.2 Bioenergy from wood chips

Chapter 5 of this thesis presented the LCA of a novel CHP technology (close to commercialization) integrating biomass gasification with a 199 kW solid oxide fuel cell (SOFC). This technology can produce heat and electricity from wood chips from sustainable forest management and sawmills.

Wood pellets and Miscanthus pellets were also assessed as feedstocks. For the seven impact categories considered, the bioenergy produced using this technology utilizing these three fuels was compared with conventional energy technologies i.e. heat from natural gas and electricity from the German grid.

The production of biomass fuels was the main source of environmental impacts of the energy produced using this technology. Depending on the category and the fuel, biomass fuels' production (including transportation) represented between 23% and 99% of the total impacts. For all categories, energy from wood chips as fuel generates much lower impacts than energy from wood pellets (11–70% lower depending on the category with the highest impact difference for water depletion and particulate matter) and Miscanthus pellets (9–99% lower depending on the category with the highest impact difference for water depletion). This confirms that utilizing bio-based by-products as a feedstock usually leads to a lower environmental impact than dedicated crops (i.e. Miscanthus); also for other categories than climate change.

Besides the production of the feedstock, another important contributor to the environmental impact was identified in the SOFC stack due to both the high energy intensity and material consumption of its manufacturing process and its short technical lifetime (5 years).

Compared to heat from natural gas and electricity from the German grid, this technology showed significantly lower impacts for climate change (86/94% lower), photochemical ozone formation (-43/-70%), acidification (-37/-56%) and terrestrial eutrophication (-43/-63%). Since this technology achieves high exergy efficiencies and almost zero particulate emissions, it is also performing environmentally better than organic Rankine cycles using wood chips.

8.2.3 Bio-jet fuel from potato by-products

Food processing by-products also have high availability all around Europe and are largely unexploited for innovative bio-based products. Potato by-products are used as low-price animal feed but could be potentially transformed into higher value bio-based products [383]. The Netherlands is the fourth country in EU28 for potato production [445], with 20% of the processed potatoes becoming a by-product [446]. For this reason, potato by-products have been recently tested as a feedstock to produce bio-jet fuels to partly substitute the 4000kt/y of jet-fuels consumed in the Netherlands [347].

Chapter 6 investigated the environmental performance of the innovative bio-jet fuel derived from Dutch potato by-products mentioned above. Both attributional and consequential LCA models were applied. Besides climate change, photochemical ozone formation, terrestrial eutrophication, acidification and depletion of fossil fuels were investigated.

The bio-jet fuel's climate change impact was 60% lower than conventional jet fuel when applying attributional modeling. A lower climate change benefit (5-40% reduction) was calculated by consequential modeling, assuming that European animal feed would replace the potato by-products on the animal feed market. In an extreme case, assuming the use of imported soybean meals to replace the potato by-products, the climate change impact of the bio-jet fuel could become three times the one of petrochemical kerosene due to the high land-use change impact of soybean.

Opposite outcomes between the two LCAs were obtained for photochemical ozone formation. Conversely, both modeling approaches confirmed that the investigated bio-jet fuel causes higher acidification and terrestrial eutrophication impacts than petrochemical kerosene but lower fossil fuel depletion.

8.2.4 Polypropylene from used cooking oil

Used cooking oil from the gastronomy sector, food industry and households could provide 4 Mt of used cooking oil per year in EU 27 [419]. Used cooking oil is already largely exploited worldwide for renewable diesel. However, together with renewable diesel grade products, other grades can be obtained from the hydrotreatment process, mainly naphtha and propane fractions [421,447]. A new commercial production facility converting such bio-based naphtha fraction into bio-based polypropylene has recently started operation [421].

Chapter 7 presented a cradle-to-gate LCA of polypropylene (PP) from used cooking oil considering 16 impact categories. Once normalization and weighting are applied to these 16 impact categories, the environmental impact of UCO-based PP was dominated by its climate change impact (28%), fossil resource use (23%) and water depletion (11%). The following overall environmental hotspots (with respective contributions) were identified: the polymerization process (38%), the production of hydrogen (21%), the production of LPG (18%) and the combustion of LPG (8%). Compared to petrochemical PP, significant environmental impact reductions can be achieved for climate change (40-62%, depending on the allocation method used at the process level) and fossil fuel resource use (80-86%). Moreover, these climate change impact reductions are for the cradle-to-factory gate scope without biogenic carbon removal. So, further benefits could be achieved by storing such carbon. The PlasticsEurope's Ecoprofile for petrochemical PP was used as the benchmark for comparison. PlasticsEurope provides mainly "black box" data due to confidentiality. As a consequence of the lack of transparency, it was challenging to fully interpret the impacts of petrochemical PP. So, it was not possible to provide robust conclusions for categories such as toxicity, ozone depletion and freshwater eutrophication.

Compared to bio-based PP made from sugarcane and woody biomass, PP from

UCO shows about 80/90% lower impact on climate change, from cradle to gate, without taking into account biogenic carbon removals. So, for climate change, bio-based PP from UCO can be considered a better alternative to both petrochemical and bio-based PP from dedicated crops. The reductions on impact in the climate change category were almost unaffected if UCO is globally imported instead of locally sourced. Moreover, compared to petrochemical PP, PP from UCO has the further advantage of having an embedded biogenic carbon content. Storing such carbon in a durable application combined with recycling could lead to further environmental benefits from a cradle-to-grave perspective. Using this advantage, bio-based PP from UCO could reach the reduction of climate change impact allowed by renewable diesel from UCO compared to oil diesel, which is up to 88% and is as well higher than the reductions allowed by other biodiesels (40–62% savings).

For both UCO-based diesel and PP, additional environmental impact reductions could be achieved using renewable hydrogen locally produced via electrolysis powered by photovoltaics or wind power. For PP from UCO, renewable propane and methane produced as co-products could be used to (partially) replace LPG consumption for steam cracking instead of being marketed. Greener electricity and steam used for polymerization could lead to further benefits for PP.

8.2.5 Summary of lessons learned and recommendations of RQ2

This section summarizes lessons learned regarding the life cycle environmental impacts of emerging products from bio-based by-products (“residues”).

1. Bio-based products from by-products (“residues” streams) have usually lower climate change impact than their fossil counterparts.

Figure 49 shows a summary of the reduction of climate change impacts from using bio-based by-products compared to the fossil counterpart was observed in this thesis. Climate change impact reduction between 30 and 70% can be achieved by lignin-based asphalts (details in Chapter 4) and between 86% and 96% by producing energy (heat or electricity) from wood chips via an innovative technology combining gasification and SOFC (details in Chapter 5). An innovative bio-jet fuel from potato by-products showed a 5-60% lower climate change impact than conventional jet fuel (details in Chapter 6). A climate change reduction in the range of 40–62% was calculated for PP from used cooking oil compared to petrochemical PP (details in Chapter 7). Moreover, the lignin-use review also showed a reduction of climate change impacts of 62-78% for lignin-based adipic acid and 6–32% for lignin-based polyurethane (details in Chapter 2). It is worth remarking that these climate change performances are specific to the product assessed with respective technologies and feedstocks and cannot be straightforwardly generalized to the product category, e.g. bio-jet fuels or bio-based asphalts. Even for the same product e.g. lignin-based asphalt, various factors and environmental optimization play a role in achieving a positive environmental performance e.g. type of lignin, supply chain, the composition of the asphalt etc.

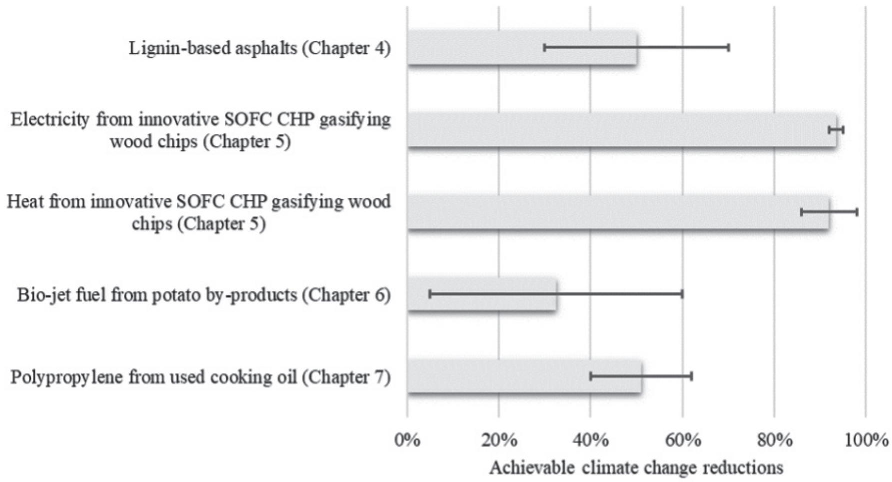

Figure 49. Climate change benefits (expressed as a percentage compared to the fossil counterparts taken as 100%) calculated in the LCAs conducted in this thesis. Variations represent both data uncertainties e.g. future energy mixes and methodological uncertainties e.g. caused by allocation choices. The LCA of polypropylene from used cooking oil had a cradle-to-gate scope and excluded potential benefits of biogenic carbon removal if stored in a durable application. All other LCAs had a cradle-to-grave scope.

2. <u>Moreover, climate change impacts are not the only relevant environmental impacts. In most cases, the savings in terms of climate change impacts were also reflected by savings of similar magnitude for the depletion of fossil resources. Trade-offs with conventional (fossil) products occur instead in other categories.</u> Previous LCA literature has widely observed this for bio-based products from dedicated crops. This thesis confirms that this also applies

to products from bio-based by-products. For example, the investigated bio-based products performed significantly worse than their fossil counterparts for acidification and terrestrial eutrophication. These two impacts are mainly due to atmospheric pollution caused by fuel combustion (e.g. in tractors) and fertilizers volatilization releasing Nitrogen (N) and Sulphur (S) emissions. Hence, they are strictly linked with agricultural activities. The allocation of even a small percentage of e.g. fertilizer-related impacts to the bio-based by-product feedstock leads to higher acidification and eutrophication impacts than petrochemical products (not requiring fertilizers). Since forestry residues/ wood chips assessed in Chapter 5 do not require any cultivation, higher eutrophication and acidification impacts were not observed for this by-product feedstock. Furthermore, the pretreatment of bio-based feedstocks often requires acids or other chemicals with high environmental impacts in toxicity categories. For example, this was the case for sulfuric acid and liquid carbon dioxide solvent for lignin.

3. The decision-maker has to be careful if by-products ("residues") are already utilized to a large degree for other purposes. In such a case, particular attention is needed, especially when converting a low-grade bio-based feedstock into a high value-added product has a low yield, leading to a high feedstock consumption. This was the case of potato by-products converted via ABE fermentation that are otherwise used mainly as animal feed (see Chapter 6). Their current user would be affected and should not replace them with a much less environmentally sustainable product. This was also the case for the overall performance of lignin-based asphalts. The part of the black liquor from which lignin is extracted would normally be combusted in the boiler of the pulp mill. If the pulp mill consumes natural gas to complete the energy balance, lignin-based asphalts could potentially lead to a higher overall environmental impact than conventional asphalts.

4. Process energy plays an important role in the climate change reduction achievable using bio-based products. If the goal is to maximize climate change benefits, low-value biomass (e.g. hog fuel) or renewable gaseous fuels (e.g. renewable propane or biogas) are key choices. However, this choice often has an economic trade-off with natural gas or LPG (see Chapters 2 and 4 and 7 for details regarding the entities of the environmental burden shifting using one fuel or the other). Similar considerations apply to low carbon intensity

chemicals and the use of renewable electricity. Electricity generally has a much higher relevance for the environmental impact of bio-based chemicals and fuels than their petrochemical counterparts.

5. For durable applications, the modeling of biogenic carbon involves high uncertainties. Since durable applications like asphalt or plastics (e.g. PP) can be recycled multiple times, accounting properly for the biogenic carbon storage is crucial for better climate change performance compared to their petrochemical counterparts. While most guidelines, e.g. the Dutch-product category rules for the construction sector, consider the biogenic carbon storage as permanent after 100 years, the new recommendation of the EU PEF [351] of not accounting for credits for permanent (and temporary) carbon storage could penalize this type of bio-based products.

8.3 RESEARCH QUESTION 3

How are the LCA results of such products influenced by the approach adopted to deal with the multifunctionality issue?

8.3.1 Lignin-based asphalts

In Chapter 4, lignin is used as an alternative ingredient to bitumen in asphalts. For asphalts using kraft lignin, an allocation is necessary at the level of the pulp mill to apportion the environmental impact between the main product (pulp) and lignin. Applying mass allocation instead of economic allocation to the unit processes of the pulp mill that lignin shares with pulp showed a much higher environmental impact for lignin.

Bitumen is as well a product from a multi-output process, i.e. the distillation of crude oil. Hence, the environmental impact of bitumen is also the result of an allocation procedure. As for kraft lignin, the sensitivity analysis showed that shifting from economic allocation to mass allocation at the level of the oil refinery leads to a higher environmental impact allocated to bitumen.

The cradle-to-gate environmental impacts of both asphalts were significantly affected by the allocation method chosen for the lignin and bitumen production processes. Hence, the cradle-to-gate comparison between lignin-based asphalts and conventional asphalts was highly affected by the allocation method applied to lignin or bitumen.

However, once the credits for recycling lignin and bitumen are accounted, a high fraction of the environmental impact of lignin and bitumen is credited. Such a credit makes the type of allocation method (i.e. economic, mass or energy) less influential over the cradle-to-grave life cycle. Hence, the cradle-to-grave comparison between the environmental impacts of the two asphalts was less affected by the allocation applied to lignin and bitumen (if the same method is applied to both). This applied to both the comparison per impact category or on a weighted basis.

8.3.2 Bioenergy from wood chips

Chapter 5 presented an LCA of an innovative CHP combining gasification and SOFC technologies. As for most CHPs producing heat and electricity in similar magnitudes, the allocation between these two products can significantly change the allocated environmental impact in most impact categories. Scrutinizing the effect of the allocation method applied between heat and electricity, the importance of providing the LCA results for both heat and electricity produced by CHPs emerged. In this way, it is possible to understand the effects of the allocation applied and increase transparency to allow for the right interpretation.

The comparison between the innovative SOFC CHP and an ORC CHP using the same fuel (i.e. wood chips) showed the importance of comparing LCA results between CHP technologies based on the same allocation method. In fact, different CHPs can produce heat and electricity with very different ratios. Only if the same allocation parameter (independently if exergy or economic) is applied to both CHPs, heat from the investigated technology will have a lower impact than heat from ORC in five categories (climate change, particulate matter, photochemical ozone formation, acidification, and terrestrial eutrophication) and higher in the two related to resource depletion. Conversely, if exergy allocation is applied to one CHP and economic allocation to the other CHP, the outcome could be misleading in some categories.

Testing the use of substitution in attributional modeling provided negative environmental impacts in several impact categories that 1) conflicts with the attributional aim and 2) cannot be replicated using exergy and economy allocation methods, i.e. allocation practices that the LCA literature has broadly acknowledged as a good proxy of causality for CHPs. This fact confirms the importance of the recommendation provided in section 8.1.3 that the use of substitution should be linked with a specific goal of the LCA, i.e. assessing a change via consequential modeling;

8.3.3 Bio-jet fuel from potato by-products

Chapter 6 investigated the effect of applying a different modeling approach (i.e. attributional or consequential) to investigate an innovative bio-jet fuel. Regarding the allocation method applied in the attributional LCA, the effect on the environmental impact of the price fluctuation on the allocation share between potato by-products and potato food products played a minor role. However, applying energy or mass allocation at the level of the potato processing industry would significantly change the environmental impact of the potato by-products. These two last methods would allocate impacts of similar magnitude to the potato by-products and the potato food products since they have similar physical characteristics (while the economic allocation reflected the much lower market price of the potato by-products). Nevertheless, these two types of allocation would not respect the ISO causality principle that should be reflected by the allocation criterion (i.e. the potato processing industry works to generate revenues and not energy products). So, energy and mass allocations should not be used in this case.

An attributional LCA was also conducted to determine the environmental impact of the co-products of the bio-jet fuel, namely animal feed (from fermentation residue), bio-based hydrogen, bio-based carbon dioxide, and biolubricants. In this way, it was possible to understand how physical or economic relations partition the environmental impact of each process and how the sum of the environmental impact of each product delivered by the system composes the total environmental impact of the system.

As often emphasized in LCA literature, consequential modeling has a much higher uncertainty than attributional modeling. A deep investigation was conducted on the marginal production of animal feed assumed since it could significantly change the consequential LCA outcome. The attributional LCA cannot capture any displacement effects on the animal feed market. In fact, the system analyzed in attributional LCAs aims only to assess the processes directly linked by (physical, energy, and service) flows to the unit process supplying the functional unit and not at all processes affected by the decision based on a cause-and-effect chain. However, the additional animal feed production in the system boundaries of the consequential LCA led to major uncertainties linked to the specific type of animal feed assumed to be used as a substitute for potato by-products. Furthermore, the displacement of the animal feed itself presents two other major sources of uncertainties: 1) the functionality assumed for animal feed, i.e. provision of proteins or energy to the animals and 2) indirect land-use changes if soybean is part of the displaced animal feed. Other major uncertainties of the consequential modeling arise from the future production method of the co-products substituted (surplus of hydrogen, lubricants and carbon dioxide). Since the investigated bio-jet fuel will take some years before being marketed and major changes towards decarbonization are expected in the EU chemical sector, forecasts of future markets can significantly differ from the future markets as assumed. So, the substitutions applied in the CLCA of the bio-jet fuel could lead to significant changes in the results.

8.3.4 Polypropylene from used cooking oil

The production of polypropylene from used cooking oil analyzed in Chapter 7 is obtained via two main multi-output processes, i.e. hydrotreatment and steam cracking. Polypropylene from used cooking oil at the production gate already showed better climate change and fossil fuel resource use than petrochemical PP.

The allocation method used at the process level can vary the climate change mitigation potential range between 40% and 62% and fossil fuel resource use between 80% and 86%. Such uncertainties consider both UCO as a by-product instead of waste or a different allocation approach used for hydrotreatment and steam cracking. Despite multifunctionality uncertainty being relatively small for these two impact categories, it was much higher in other categories. For example, by using the so-called 50/50 method on UCO open-loop recycling, the total weighted impact could increase between 25% and 160% and e.g. become up to 4 times higher for particulate matter impact depending on the types of primary vegetable oil.

Regarding the allocation method for hydrotreatment and steam cracking units, exergy allocation was apportioning the highest impact to bio-based PP in most categories compared to energy allocation or cut-off allocation. Conversely, as already experienced with the bioenergy case study presented in Chapter 5, the substitution method would lead to a negative footprint for bio-based PP. This negative footprint is due to the low physical (and economical) significance of bio-based naphtha and propylene, i.e. UCO-based PP's precursors. However, such a negative impact should be considered the result of a wrong methodological choice and not part of the uncertainty since physical significance should be checked before applying substitution to avoid substituting by-products.

8.3.5 Summary of lessons learned and recommendations of RQ3

This section summarizes lessons learned regarding the uncertainty caused by the approach adopted to deal with the multifunctionality issue in the LCAs of emerging products from bio-based by-products ("residual flows") assessed in this thesis.

1. <u>Applying mass or energy allocation to the unit process delivering the by-product feedstock often leads to a much higher environmental impact for by-product feedstocks.</u> This was the case for lignin in Chapter 4 (and also its fossil counterpart i.e. bitumen in the same chapter) and potato by-products in Chapter 6. Nevertheless, these two types of allocation do not reflect ISO causality relationships. In fact, both the potato processing industries and pulp mills do not operate to produce lignin and potato by-products but to generate revenues selling pulp and food products, and the most desired product reflects "why these processes exist". There might be cases when the most desired product for the same process might change in the future and a change of economic allocation share would reflect this.
2. In comparative LCAs between bio-based and conventional products, <u>the practitioner can expect that multifunctionality uncertainty can make the comparison with conventional products inconclusive for some categories (e.g. see the comparison between the LCA impacts of bio-based PP from UCO and petrochemical PP in Chapter 7).</u> In attributional LCAs, this means that the environmental performance of a bio-based product in several categories might depend on the adopted allocation method. Hence, an allocation key might apportion a higher share of a unit process' impact to a co-product than another co-product and another key would have done the opposite. Accordingly, the allocation applied in the attributional LCA of a certain co-product can affect the outcome of the comparison with its fossil counterpart. For this reason, increasing transparency in illustrating allocation choices and respective data (e.g. prices assumed) is the only way to allow for the

right interpretation (as strongly remarked under RQ1). Moreover, it is worth remarking that attributional LCA results of all co-products of a system should always sum up to the total impact of the system (e.g. see the deep illustration of this mechanism for the case of heat and electricity produced by SOFC-CHP in Chapter 5 and bio-jet fuel and its co-products in Chapter 7).

3. For the same product, the cradle-to-gate LCA results might have relatively high multifunctionality uncertainty, but the cradle-to-grave LCA might have low multifunctionality uncertainty (or vice-versa). For example, Chapter 4 showed that the environmental impacts of lignin and bitumen have high multifunctional uncertainties. However, when used in asphalt production, the uncertainties associated with the productions of lignin and bitumen are less significant because unit processes such as the asphalt production process are responsible for a significant percentage of the overall impacts. Therefore, summing a high-impact monofunctional process like asphalt production lowers the relevance of the multifunctionality uncertainty of some ingredients in the overall impact of the product system. Furthermore, asphalt recycling allows the recovery of a significant fraction of lignin and bitumen. So, the total net mass (consumed minus recovered) of lignin and bitumen allocated to the first life cycle is lower once recycling is included in the system boundary, decreasing their relevance on the overall environmental impact of the asphalt. Hence, large credits received due to recycling can lower the effect of the choice of the allocation method at the process level. However, the credits regarding products from multifunctional processes should rely on the same allocation method for correctness. For example, suppose the impact of oil refining was allocated to bitumen using a certain allocation approach. In that case, the credited impact for recovered bitumen should be based on the impact of bitumen calculated via the same allocation. The importance of comparing LCA results based on consistent allocation methods was strongly remarked under RQ1. This should also apply to credits from recycling activities when crediting the virgin production from a multifunctional process.
4. The case studies presented in this thesis highlighted that the environmental impact of by-products/residues used as feedstock for bio-based products are generally not trivial in the life cycle impact of the bio-based products and are highly affected by multifunctionality uncertainty. In fact, a small change in the allocation share (e.g. due to a sudden and substantial increase in demand) of the main product can significantly change the allocation share of the by-product.

5. Consequential LCA modeling is a key tool for policy support if the bio-based residues used as feedstock already have other (low-value) applications. Their utilization for a different use may, in turn, cause high indirect environmental effects. Using a consequential LCA, these potential burden shifts with overall potentially negative side effects can be foreseen/mitigated in advance and monitored, e.g. through policy measures. This was the case of potato by-products used for bio-jet fuel and currently used as animal feed (see Chapter 6). Since the current use of potato by-products is not part of the supply chain of the potato by-products, the attributional LCA neglecting this aspect showed a 60% climate change mitigation potential for the investigated bio-jet fuel. The consequential LCA estimated no or much lower climate change benefit (5-40%).
6. Testing substitution-based allocation in attributional modeling provided negative environmental impacts in several impact categories for bio-energy from SOFC CHP in Chapter 5 and polypropylene from used cooking oil in Chapter 7. The allocation of negative impacts to these products was not in line with the results generated by other allocation methods that are broadly acknowledged as a good proxy of causality relationships. Hence, such negative impacts conflict with the attributional aim, confirming once more the importance of using substitution only under specific LCA goals requiring the assessment of a change in demand and consequent adoption of consequential modeling.

8

8.4 OVERARCHING LIMITATIONS OF THIS THESIS AND FUTURE RESEARCH

This thesis aimed to analyze the life cycle environmental performance of several innovative bio-based products from agro-industrial and food-processing by-products and methodological complexities in conducting their environmental LCA. A major focus of the chapters of this thesis was on LCA multifunctionality, outlying unresolved issues in the implementation of ISO recommendations, and their effect on the environmental impacts of bio-based products from residual streams. As a result, this thesis provided recommendations to improve the consistency of life cycle inventory models of multifunctional product systems delivering bio-based products.

This thesis does not presume to have solved and covered all issues related to multifunctionality practices and LCAs were conducted only for a number of LCA cases for bio-based products from residual streams. Besides the research conducted in this thesis regarding the multifunctionality issue in LCAs of products from bio-based residual steams, future research is still necessary to investigate LCA multifunctionality for other relevant products from bio-based residues. When more than one allocation method is suitable for these products, evaluating the related uncertainty varying allocation methods is necessary.

Moreover, the case studies analyzed in this thesis and collected data could have been suitable to cover (broadly) other major aspects behind the life cycle impacts of bio-based products of high relevance nowadays. In particular, the following are two alternative research questions on two major aspects currently under debate by the scientific literature on the life cycle impacts of bio-based products.

The first aspect to investigate is: *what are the implications of the temporal scope on the data used for prospective LCAs?* In this thesis, all LCAs regarded innovative bio-based products that have just or not reached commercialization yet. So, even if all LCAs were based on primary data as much as possible, primary data does not mean directly high data quality for emerging technologies. The chapters of this thesis looked at various technologies at an early stage of development to assess their environmental performance and provide the necessary guide for investment and research. However, pilot plants might significantly differ from future commercialized technology. Major reasons behind higher data uncertainties for innovative products than commercialized products are due to: 1) potential process design changes, 2) size scaling effects, 3) process synergies that could be optimized, 4) future technological learning and 4) external factors (e.g. a future infrastructural change of the electricity mix) [448]. All these aspects have been considered in the performed LCAs, but a higher focus could have been given.

A second research question to investigate could be: *what are the most environmentally friendly and cost-effective ways of using bio-based by-products as feedstocks?* In fact, bio-based by-products (“residues”) have limited availability and their best use should be preferred. Furthermore, products from bio-based by-products generally have a (much) higher production cost than petrochemical products and their production needs support from national and international policy initiatives. The chapters of this thesis provided only a limited overview of bio-based products that could be obtained from the same feedstock and respective reductions of environmental impacts compared to their fossil counterparts. Moreover, techno-economic analysis was not applied in the chapters of this thesis. So, cost implications were completely neglected in the comparison between bio-based based and petrochemical alternatives.

Despite the challenges faced by LCA practice, LCA is a powerful tool to understand the environmental impacts of products and services. An increasing number of governments and companies consider LCA the key environmental management decision-support tool. The strength of LCA is its capacity to detect burden-shifting from one impact category (e.g. climate change) to another (e.g. human toxicity) and from one life cycle stage (e.g. production process) to another (e.g. combustion during fuel use). Such issues cannot be detected by other available metrics such as green chemistry metrics, circularity metrics, and process-related parameters (e.g. consumption of energy or water) and are crucial in the environmental comparisons to support decision making.

CHAPTER 9

English summary

9. ENGLISH SUMMARY

In the last decade, the need to reduce dependence on fossil resources has led to the emergence of numerous bio-based alternatives in European sectors traditionally dominated by petrochemical products. The *European Green Deal* and *EU bioeconomy strategy* could further accelerate the growing trend of innovative bio-based commodities worldwide. This significant growth in the short term should be accompanied by scientific evidence on the environmental impacts of innovative bio-based products. In fact, to achieve the targets of bioeconomy policies, investments should be guided towards environmentally sustainable bio-based products. Science-based evidence can confirm if a certain bio-based product achieves the expected lower environmental footprint than its petrochemical counterpart.

Locally sourced *bio-based "residues"* are a key feedstock to extend the amount of bio-based products produced sustainably in the EU. This feedstock does not generate concerns about food security and land competition, is usually cheaper than dedicated crops, and does not require transoceanic imports. Products from this type of feedstock were the major focus of this thesis. In particular, one of three main research questions was: *what is the environmental impact of innovative bio-based products made from by-product/waste streams compared to their fossil counterparts*?

This thesis explored the conversion of various local bio-based residues into a heterogeneous range of innovative products from an environmental perspective. Among the explored feedstocks were *food processing residues,* i.e. potato by-products and used cooking oil, *forestry residues,* i.e. wood chips, and a by-product from *paper/pulp industries,* i.e. lignin.

The life cycle environmental impacts of innovative bio-based products from these agro-industrial and food-processing by-products were compared to their petrochemical counterparts using *Life Cycle Assessment* (LCA) methodology. LCA is an internationally standardized method to assess products and services' life cycle environmental impacts, from raw materials extraction to waste management. Various policy decision instruments and regulation mechanisms already rely on LCA results to assess the environmental performance of bio-based products.

In particular, this thesis presents for the first time the LCAs of *four* emerging products:

1. *polypropylene* from *used cooking oil* via hydrotreatment,
2. *bioenergy* from a solid oxide fuel cell (SOFC) combined heat and power plant (CHP) relying on gasification of *wood chips*,
3. *a bio-jet fuel* via acetone-butanol-ethanol fermentation of *potato by-products* and,
4. *bio-based asphalts* with *lignin* from the kraft pulping process used as the binder.

These four LCAs were based on primary data for the respective innovative conversion technologies. All four LCAs scrutinized a broad range of environmental impact indicators.

LCA modeling needs to be applied consistently on both sides to perform a meaningful comparison between the life cycle impacts of bio-based products and their petrochemical counterparts. Although LCA practice is standardized, one of the historically most debated points with lower convergence of views is the so-called *multifunctionality issue*. The multifunctionality issue occurs if a process in the production system under assessment provides more than one function or product. The hierarchical steps to follow when solving multifunctionality in LCA are based on the recommendations of ISO 14044:2006.

However, the *interpretation* of such recommendations has been debated for twenty-five years leading to different implementation practices in the LCA scientific literature and ISO-compliant LCA guides. Since the production processes of bio-based products are frequently multifunctional, a criterion for allocating environmental impacts between the co-products is often necessary when conducting LCAs of bio-based products. Hence, the lack of a shared interpretation of ISO recommendations for selecting multifunctionality approaches and consequent lack of consistent allocation practices closely affect LCAs of bio-based products. So, a major research question of this thesis was: *what multifunctionality practices are adopted in LCAs of bio-based products and how can the consistency of the life cycle inventory model be improved*?

In particular, all feedstocks explored in this thesis are by-products obtained through the production process of a more economically valuable product. Hence, the feedstock originates from a multifunctional process for all four case studies. Based on the experience gained by previous LCAs of more established products from bio-based residues, multifunctionality is a major source of LCA *uncertainty* for products from this type of feedstock. In fact, for such products, not only the modeling of the production process but also the feedstock itself is affected by the LCA multifunctionality issue. So, for an accurate interpretation of the LCA results, it is necessary to understand *how the LCA results of such products are influenced by the approach adopted to deal with the multifunctionality issue.*

Accordingly, different multifunctionality practices both at the level of the feedstock and at the processing level were deeply discussed in the four LCAs presented. The multifunctionality issue in LCAs of bio-based products was further investigated via two *reviews*. A first review scrutinized multifunctionality practices and their effect on LCA results' *comparability* for one of the feedstocks of interest, i.e. lignin. A second review focused on the existing interpretations of the multifunctionality solutions recommended by ISO 14044:2006, the related debates and their historical origins. In the same review, a *text mining* process was adopted to quantify ISO-compliant multifunctionality practices in all LCAs of bio-based products in the literature. The knowledge gained from these two reviews allowed detecting the different interpretations' origins and underlying rationales. Such knowledge was fundamental to select only (fully) ISO-compliant multifunctionality practices in the LCAs of the four investigated bio-based products. Based on the lessons learned from the two reviews, nine key *recommendations* to deal with the multifunctionality issues of bio-based products were formulated. These recommendations relate to the context of the use of substitution approaches, ISO-causality as the key principle for selecting allocation methods, transparency in the rationale for their selection and a strict distinction between attributional and consequential modeling approaches.

The LCAs of all four products from bio-based residues showed a generally better *climate change* performance than their fossil counterparts. The climate change impact reductions were quantified as 30-70% for top-layer lignin-based asphalts, 86-96% for energy (heat or electricity) from wood chips via gasification-SOFC CHP, 5-60% for an innovative bio-jet fuel from potato by-products and 40–62% for polypropylene from used cooking oil. Moreover, the review of LCAs of lignin-based products showed promising climate change impact reductions of 62-78% for lignin-based adipic acid and 6–32% for lignin-based polyurethane. *Low carbon-intensity process energy* was confirmed to play an important role in achieving high climate change reductions for all these products. Hence, low-quality biomass or renewable gaseous fuels (e.g. renewable propane or biogas) and renewable electricity are key choices to maximize climate change benefits.

For all four products, the reduction of climate change impacts compared to petrochemical counterparts was also reflected by similar reductions for the *depletion of fossil resources*. Trade-offs with conventional (fossil) products were instead observed for other impact categories. The allocation of even a small percentage of e.g. fertilizer-related impacts to *agricultural by-products* led to higher *acidification and eutrophication* impacts than their petrochemical counterpart for products from this type of feedstock. Moreover, if the pretreatment of the feedstock to be converted into high-value bio-based products requires chemicals like sulfuric acid and liquid carbon dioxide solvent for lignin, higher impacts than their petrochemical counterparts can be observed in *toxicity categories*.

Besides multifunctionality, the modeling of *biogenic carbon storage* for durable bio-based products and the modeling approach (*attributional or consequential*) for by-products utilized to a large degree for other purposes may lead to high uncertainties. Accordingly, in such cases, selecting a different multifunctionality or modeling approach can make the comparative LCAs between bio-based and conventional products inconclusive for some impact categories. Despite these methodological challenges, the case studies investigated in this thesis highlighted once more the strength of LCA as a key tool to provide decision support on the environmental impacts of bio-based products to avoid undesirable shifts of environmental burdens from one ecological issue to another one.

CHAPTER 10

Nederlandse samenvatting

10. NEDERLANDSE SAMENVATTING

In het afgelopen decennium heeft de noodzaak om de afhankelijkheid van fossiele grondstoffen te verminderen geleid tot de opkomst van tal van bio-based alternatieven in Europese sectoren die traditioneel worden gedomineerd door petrochemische producqten. The European Green Deal en EU bioeconomy strategy kunnen deze groeiende trend voor innovatieve bio-based goederen wereldwijd verder accelereren. Deze significante groei zou op korte termijn gepaard moeten gaan met wetenschappelijk onderbouwing van de milieu-impacts van innovatieve bio-based producten. Om de doelen van het bio-economie beleid te behalen, moeten investeringen gericht gestuurd moeten worden met betrekking tot de duurzamheid van bio-based producten . Wetenschappelijk bewijs kan bevestigen óf verschillende bio-based producten de ecologische voetafdruk reduceren ten opzichte van de petrochemische tegenhanger.

De inzet van Lokale bio-based 'residuen' als grondstoffen zijn belangrijk om meer bio-based producten duurzaam te kunnen produceren. Deze grondstoffen zijn geen grond tot zorg met betrekking tot voedselveiligheid en land competitie. Daarnaast zijn ze gewoonlijk goedkoper dan - gewassen, en import is vaak niet noodzakelijk . Producten uit residuen van dit type grondstof stonden centraal in dit proefschrift. Een van de drie hoofdonderzoeksvragen was: Wat is de milieu-impact van innovatieve bio-based producten gemaakt van bijproduct/afvalstromen in vergelijking met hun fossiele tegenhangers?

Dit proefschrift onderzoekt de conversie van verschillende lokale bio-based reststoffen naar een heterogeen aanbod van innovatieve producten vanuit een milieuoogpunt. Tot de onderzochte grondstoffen behoorden voedselverwerkingsresiduen, dat wilt zeggen aardappelbijproducten en gebruikt frituurvet, bosbouwresiduen, houtsnippers, en een bijproduct van de papier-/pulpindustrie, namelijk lignine.

De milieueffecten over de levenscyclus van innovatieve bio-based producten van deze agro-industriële en voedselverwerkende bijproducten werden vergeleken met hun petrochemische tegenhangers met behulp van Life Cycle Assessment (LCA)-methodologie. LCA is een internationaal gestandaardiseerde methode om de milieueffecten van de levenscyclus van producten en diensten te beoordelen, van de winning van grondstoffen tot afvalbeheer. Verschillende beleidsinstrumenten en reguleringsmechanismenzijn deels gebaseerd op LCA-resultaten om de milieuprestaties van bio-based producten te beoordelen.

In het bijzonder presenteert dit proefschrift voor het eerst de LCA's van vier opkomende producten:

1. olypropyleen uit gebruikt frituurolie via waterstofbehandeling,
2. bio-energie uit een vasteoxidebrandstofcel (SOFC) gecombineerd met warmtekrachtkoppeling (WKK) op basis van vergassing van houtsnippers,
3. bio-kerosine via aceton-butanol-ethanolfermentatie van aardappel-bijproducten, en
4. biobased asfalt met lignine uit het kraftpulpproces als bindmiddel.

Deze vier LCA's waren gebaseerd op primaire gegevens voor de specifieke innovatieve conversietechnologieën. In het kader van de vier LCA's is een breed scala aan milieu-impact indicatoren onder de loep genomen.

LCA-modellering moet van beide kanten consistent worden toegepast om een zinvolle vergelijking te maken tussen de levenscycluseffecten van bio-based producten en hun petrochemische tegenhangers. Hoewel de LCA-praktijk gestandaardiseerd is, is een van de historisch meest besproken punten (met tevens een lage mate van convergentie van standpunten) het zogenaamde multifunctionaliteitsprobleem. Het multifunctionaliteitsprobleem doet zich voor als een proces in het te beoordelen productiesysteem meer dan één functie of product levert. De te volgen hiërarchische stappen bij het oplossen van multifunctionaliteit in LCA zijn gebaseerd op de aanbevelingen van ISO 14044:2006.

Er wordt echter al vijfentwintig jaar gedebatteerd over de interpretatie van dergelijke aanbevelingen, wat heeft geleid tot verschillende implementatiepraktijken in de LCA-wetenschappelijke literatuur en ISO-conforme LCA-gidsen. Omdat de productieprocessen van biobased producten vaak multifunctioneel zijn, is bij het uitvoeren van LCA's van biobased producten vaak een criterium nodig voor de toerekening van milieueffecten tussen de co-producten. Vandaar dat het ontbreken van een breed gedeelde interpretatie van ISO-aanbevelingen voor het selecteren van multifunctionaliteitsbenaderingen en het daaruit voortvloeiende gebrek aan consistente toewijzingspraktijken nauw van invloed zijn op LCA's van biogebaseerde producten. Een belangrijke onderzoeksvraag van dit proefschrift was daarom: welke multifunctionaliteitspraktijken worden toegepast in LCA's van bio-based producten en hoe kan de consistentie van het levenscyclusinventarisatiemodel worden verbeterd?

In het bijzonder zijn alle grondstoffen die in dit proefschrift worden onderzocht, bijproducten die worden verkregen door het productieproces van een economisch waardevoller product. De grondstof is dus afkomstig uit een multifunctioneel proces voor alle vier casestudies. Op basis van de ervaring die is opgedaan met eerdere LCA's met meer gevestigde producten uit bio-based residuen, is multifunctionaliteit een belangrijke bron van -onzekerheid voor producten uit dit type grondstof. In feite wordt voor dergelijke producten niet alleen de modellering van het productieproces, maar ook de grondstof zelf beïnvloed door het LCA-multifunctionaliteitsprobleem. Voor een nauwkeurige interpretatie van de LCA-resultaten is het dus noodzakelijk om te begrijpen hoe de LCA-resultaten van dergelijke producten worden beïnvloed door de aanpak die is gevolgd om het multifunctionaliteitsprobleem aan te pakken.

Dienovereenkomstig werden verschillende multifunctionaliteitspraktijken zowel op het niveau van de grondstof als op het verwerkingsniveau uitvoerig besproken in de vier gepresenteerde LCA's. Het multifunctionaliteitsprobleem in LCA's van bio-based producten is verder onderzocht via twee reviews. Een eerste review onderzocht multifunctionaliteitspraktijken en hun effect op de vergelijkbaarheid van LCA-resultaten voor een van de van belang zijnde grondstoffen, namelijk lignine. Een tweede review richtte zich op de bestaande interpretaties van de multifunctionaliteitsoplossingen aanbevolen door ISO 14044:2006, de gerelateerde debatten en hun historische oorsprong. In dezelfde review werd een tekstdelvingsproces gebruikt om ISO-conforme multifunctionaliteitspraktijken in alle LCA's van bio-based producten in de literatuur te kwantificeren. De kennis die uit deze twee reviews werd verkregen, maakte het mogelijk om de oorsprong en onderliggende beweegredenen van de verschillende interpretaties te detecteren. Dergelijke kennis was van fundamenteel belang om alleen (volledig) ISO-conforme multifunctionaliteitspraktijken te selecteren in de LCA's van de vier onderzochte bio-based producten. Op basis van de lessen die uit de twee beoordelingen zijn getrokken, zijn negen belangrijke aanbevelingen geformuleerd om de multifunctionaliteit van bio-based producten aan te pakken. Deze aanbevelingen hebben betrekking op de context van het gebruik van substitutiebenaderingen, ISO-causaliteit als het belangrijkste principe voor het selecteren van allocatiemethoden, transparantie in de reden voor hun selectie en een strikt onderscheid tussen attributie- en consequentiële modelleringsbenaderingen.

De LCA’s van alle vier producten uit biobased residuen lieten over het algemeen betere prestaties zien met betrekking tot klimaatveranderings dan hun fossiele tegenhangers. De reductie van de impact op klimaatverandering werd gekwantificeerd als 30-70% voor de toplaag op lignine gebaseerd asfalt, 86-96% voor energie (warmte of elektriciteit) uit houtsnippers via vergassing-SOFC WKK, 5-60% voor een innovatief bio-kerosine uit aardappelbijproducten en 40–62% voor polypropyleen uit gebruikt frituurvet. Bovendien toonde de review van LCA’s van op lignine gebaseerde producten veelbelovende verminderingen van de impact op klimaatverandering van 62-78% voor op lignine gebaseerd adipinezuur en 6-32% voor op lignine gebaseerd polyurethaan. Er werd bevestigd dat procesenergie met een lage koolstofintensiteit een belangrijke rol speelt bij het bereiken van een sterke vermindering van de klimaatverandering voor al deze producten. Daarom zijn het gebruik van biomassa van lage kwaliteit, hernieuwbare gasvormige brandstoffen (bijv. hernieuwbaar propaan of biogas) en hernieuwbare elektriciteit belangrijke keuzes om de reductie van klimaatverandering te maximaliseren.

Voor alle vier de producten werd de vermindering van de gevolgen van klimaatverandering in vergelijking met petrochemische tegenhangers ook weerspiegeld in vergelijkbare verminderingen voor de uitputting van fossiele hulpbronnen. Voor andere impactcategorieën werden daarentegen trade-offsen met conventionele (fossiele) producten waargenomen De toekenning van zelfs een klein percentage van bijvoorbeeld kunstmest-gerelateerde impacts aan agrarische bijproducten leidden tot hogere effecten op verzuring en eutrofiëring dan hun petrochemische tegenhanger voor producten van dit type grondstof. Bovendien, als de voorbehandeling van de grondstof die moet worden omgezet in hoogwaardige bio-based producten chemicaliën vereist zoals zwavelzuur en vloeibaar kooldioxide-oplosmiddel voor lignine, kunnen grotere impacts worden waargenomen in toxiciteitscategorieëndan hun petrochemische tegenhangers..

Naast multifunctionaliteit kan het modelleren van biogene koolstofopslag voor duurzame bio-based producten en de modelleringsaanpak (attributioneel of consequentieel) voor bijproducten die in hoge mate voor andere doeleinden worden gebruikt, tot grote onzekerheden leiden. Dienovereenkomstig kan in dergelijke gevallen het selecteren van een andere multifunctionaliteit of modelleringsaanpak de vergelijkende LCA’s tussen bio-based en conventionele producten onbeslist maken voor sommige effectcategorieën. Ondanks deze methodologische uitdagingen, benadrukten de case studies die in dit proefschrift werden onderzocht eens te meer de kracht van LCA als een belangrijk hulpmiddel om beslissingsondersteuning te bieden over de milieueffecten van bio-based producten om ongewenste verschuivingen van milieubelasting van het ene ecologische probleem naar het andere te voorkomen.

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