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The circularity of potential bio-textile production routes: Comparing life cycle impacts of bio-based materials used within the manufacturing of selected leather substitutes

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Abstract:

Nowadays the fashion industry faces increasing pressures to reduce the environmental impacts associated to the production of leather-based fashion products, particularly considering issues regarding public acceptance due to animal welfare standards and due to the toxicity of tanning chemicals. An alternative solution facilitated by the bio-textiles industry is the introduction of vegan and bio-based leather substitutes for the production of shoes, handbags, clothing's and upholstery i.e. on the basis of natural fibres, bio-based polymers, microbial cellulose and fungal mycelium composite products. Nonetheless, also these bio-based leather products cause negative environmental impacts i.e. related to land-use change and intensification, to water use and to energy use in polymer manufacturing. For further impact reduction and designing environmentally most sound solutions, design of leather substitute products should integrate best-practice interventions for increased circularity along the full product life cycles from fibre feedstock provisioning to polymer production and end-of-life recyclability and degradability. This study evaluated the current best practice scenarios for impact reduction when implementing circular design strategies in the production of bio-based fashion materials. Three case studies of alternative leather substitutes were considered, including respectively two sub-scenarios in a comparative Life Cycle Impact Assessment. Results for the aggregated single score using the Environmental footprint approach showed that principles of circularity (e.g. the feedstock type and by-product recovery for fiber and sugar feedstocks) have an influence of 65% between the best and worst performer in mitigating environmental impacts. Furthermore, enhancing the product durability of the leather substitutes against the temporal product replacement benchmark of animal leather would have an influence of 25- 70% in mitigating impacts concerning water scarcity and climate change. The most important conclusion of this work is that alternative leather substitutes can contribute to relative environmental advantages in impact reduction in 8 to 14 impact categories, but only as long as the material substitution is coupled with less frequent product replacement and preferably also low impact coating systems and impregnation agents.

Key words: Circular Bioeconomy, Bio-based textiles, Leather substitutes, Comparative Life Cycle Assessment, Circularity, Sustainable fashion products

1. Introduction

As of late, the fashion industry has demanded a shift in the textile sector as disruptive start-up entrepreneurs are rapidly prototyping an increasing variety of leather substitution materials. Many of these materials rely on circular design strategies and biofabrication technologies, and have been successively introduced in consumer markets (Earley and Goldsworthy, 2019; Myers and Antonelli, 2014; Qua, 2019; Wood, 2019).

A major reason for these substitution strategies and product innovation initiatives is the increasing policy pressure on textiles manufacturing for impact reduction. Overall, the textile sector has been forced to react to the market's dwindling acceptance of established products due to the use of toxic tanning agents, high environmental burden on water bodies from livestock farming, and poor international standards in animal welfare (Blackburn, 2009b; Bruckner et al; Charter, 2018; Chowdhury et al., 2017; Earley and Goldsworthy, 2019; Joseph and Nithya, 2009a). Some innovation processes used by experimental start-ups, such as biologically inspired designers and green fashion companies, offer a broad range of bio-based substitution options, circular strategies for sourcing secondary raw materials and agricultural residues, and novel manufacturing technologies, which can be introduced into the processing chain (Luis Quijano; Milly, 2018/2019; Qua, 2019; Wood, 2019). The use of agricultural residues is a possible approach to promote the shift toward circular resource mobilization strategies. One example of this includes the use of pineapple leaf fibers (PALF) from pineapple residues for fiber provisioning in the production of nonwoven products. Another example is the use of lignocellulose residues, e.g., straw and husks, for the cultivation of mycelium materials or for the production of bio-based coating options on the basis of lignocellulose-based polymers. Furthermore, leather tanning processes can be altered to a renewable resource base, e.g., producing tanning agents on the basis of oxalic acid (Alfarisi et al., 2017; Collet 2018; Goswami and O'Haire, 2016; Krishnaraj and Sani, 2019; Myers and Antonelli, 2014; Qua, 2019; Younes, 2017). In the testing and upscaling of biofabrication technologies, a common feature observed from emerging approaches is that the materials are manufactured with an aim to mimic the material properties of leather. This is done by cultivating eukaryotic cell tissues from yeast and bacteria cultures and of fungal mycelium composite materials (FMCM). The materials obtained include substrates, such as chitin structures, collagen proteins, and microbial cellulose sheets, that can be further processed into the desired fashion products (Andréa et al., 2017; Collet 2018; Ghalachyan, 2018; Kim et al., 2017; Qua, 2019). The benchmark of environmental footprints associated with circularity manufacturing processes for more environmental friendly leather production and alternative leather substitutes continues to face a vast quantity of questions regarding inventory collection and validation, indicator completeness in multi-criteria assessment frameworks, and fair baselines in equality-of-benefits comparisons (Blackburn, 2009a; Cayzer et al., 2017; Joseph and Nithya, 2009b; Laurenti et al., 2017; Qua, 2019).

Furthermore, for negating the option that impact decoupling is compromised by problem shifting, either by rebound effects or hidden externalization from developed to developing countries (Parrique et al., 2019), the circular design efforts of the fashion industry must be evaluated to determine if they represent showcases effective for public relation and marketing purposes, allowing the fast fashion business with high demands to exist in separated mass market divisions, or if they really contribute to circularity by full-line substitution on a mass market scale with durable, long lasting products (Andréa et al., 2017; Choi and Li, 2015; Franco, 2017; Stål and Corvellec, 2018; Younes, 2017). It is still not clear whether leather substitution and tanning agents with bio-based alternative and biomass residue feedstocks (Andréa et al., 2017; Collet 2018; Goswami and O'Haire, 2016; Myers and Antonelli, 2014; Qua, 2019; Younes, 2017) will contribute to impact the decoupling of the fashion industry in absolute figures in the mid-term perspective (Choi and Li, 2015; Franco, 2017) and therefore, must be evaluated. This assessment also relies heavily on valid benchmarking signals in circularity assessments, which inform consumer choices about the best practice materials and misleading green-labeled products, thereby promoting burden shifting instead of impact reduction. To make informed decisions, there must exist benchmarking both in the production phase as well as the use phase before waste recovery occurs. Further, it must simultaneously identify the potential impact reductions that can be achieved by introducing circular strategies on individual life cycle stages, and determine the benchmarking if the product use phase is comparable or if product replacements must be included for comparison with a common baseline due to differences in overall product durability.

Taking into account these considerations this study aims to apply an analytical framework of circularity along the value-added chains for three alternative leather substitute materials in order to compare their environmental sustainability profiles and identify strategic recommendations. Simply, this study aims to answer questions regarding the strategic levers and the general achievability of the impact decoupling in the metabolism of the bio-textiles industry and the influence of product durability and product replacement on the environmental impact profiles of leather substitute materials.

In particular, the assessment of a product’s circularity must combine traditional concepts of a circular economy with the novel understanding of a circular economy by going beyond waste treatment and enhanced recyclability to include the repurposing and holistic redesign of products.

The traditional concepts of circular economy have focused on shifting away from end-of-life treatment options, such as waste-to-energy valorization pathways, toward higher recovery rates in textile recycling as well as upcycling textile wastes with durable characteristic and high value-added potential for secondary product platforms. Today, increasingly accepted definitions of circular economy are applied from a life cycle perspective. When incorporating life cycle management strategies and circular economy design concepts into the life cycle stages of emerging bio-textile industry chains, a more cascading oriented end-of-life (EOL)-pathway is constructed, which furthers the concept of circularity by involving more efficient use of agricultural waste flows and enhancing the design-for-recyclability of finished textile products. Major expansions of circular economy definitions are seen in the extension of the waste-management oriented principle of 4R (reduce, reuse, recycle, and recover) toward 9R or even 10R frameworks (refuse, rethink, reduce, reuse, repair, refurbish, remanufacture, repurpose, recycle, and recover), which incorporate product design and clean production perspectives (Kirchherr et al., 2017; Reike et al., 2018). Concepts relying on the principles of regenerative design, biomimicry, and strategies of industrial ecology especially contribute to a more holistic optimization of value-chains in the fashion industry, leading them toward more nature-based solutions (Geisendorf and Pietrulla, 2018). Using these principles and their aim for decoupling environmental impacts from resource use and increasing demands for unsustainable raw materials for specific design problems on the micro level of fashion product design allows fashion companies to challenge their choices for basic materials, their functionality, and their upscaling of disruptive manufacturing innovations aligned with collaborators in their supply chains (Blackburn, 2009b; Franco, 2017; Koszewska, 2018).

2. Aim and Scope of the Circularity Assessment

2.1 Aim of the study

This study aims to evaluate the circularity of bio-based compounds by considering three case studies of alternative leather substitutes with two respective sub-scenarios, wherein each undergoes a comparative life cycle impacts assessment (LCIA). The results of the comparative LCIA are regarded as valuable support for benchmarking the reduction potentials of the environmental footprints of leather substitutes, thereby creating important data support for product designers and sector innovation platforms that can identify areas where further impact reduction could be feasible by implementing more ambitious circular design options. In addition to these three product-based assessments, the study assesses each individual product case with a sub-scenario that reflects how further integration of circular design options would contribute to an increased environmental impact reduction potential. These sub-scenarios were defined in order to quantify further potentials for reduction of environmental impacts when substituting biomass feedstocks, e.g., sugar crops, with lignocellulosic biomass or primary fiber resources with secondary fiber resources, e.g., hemp-flax-sisal materials with PALF-based natural fiber composites. All of these design strategies for leather substitute products are still in the market introduction stage. However, considering future market shares of bio-based leather substitutes and the associated increase in material diversification the obtained results are also expected to contribute to better assess the future circularity of more diversified markets for bio-based leather substitute products. The analytical framework and the sub-scenarios for the LCIA modeling are developed in a structured manner alongside the research questions presented in section 3.1. The analytical framework is discussed in section 3.2, the general specificities of the involved processes are described in section 3.3, and the LCIA modeling approach and datasets are evaluated in section 3.4.

2.2 Scope and Analytical framework for the comparative LCIA

To derive a realistic sustainability benchmark framework of the current developments of alternative leather products and the constraints and benchmarks they are facing during the optimization of their environmental impact profiles, the analytical framework has to include all the life cycle stages.

As shown in Figure 1, the scope of the LCIA is presented during the production stages of the product alternatives in order to compare the three different alternative leather substitute materials with the respective reference product of bovine leather.

Furthermore, Figure 1 shows the analytical framework, which is organized to cluster the research questions of the comparative LCIA alongside the circular design options in each life cycle (LC) stage from feedstock choice to product design for durability and the end-of-life scenarios.

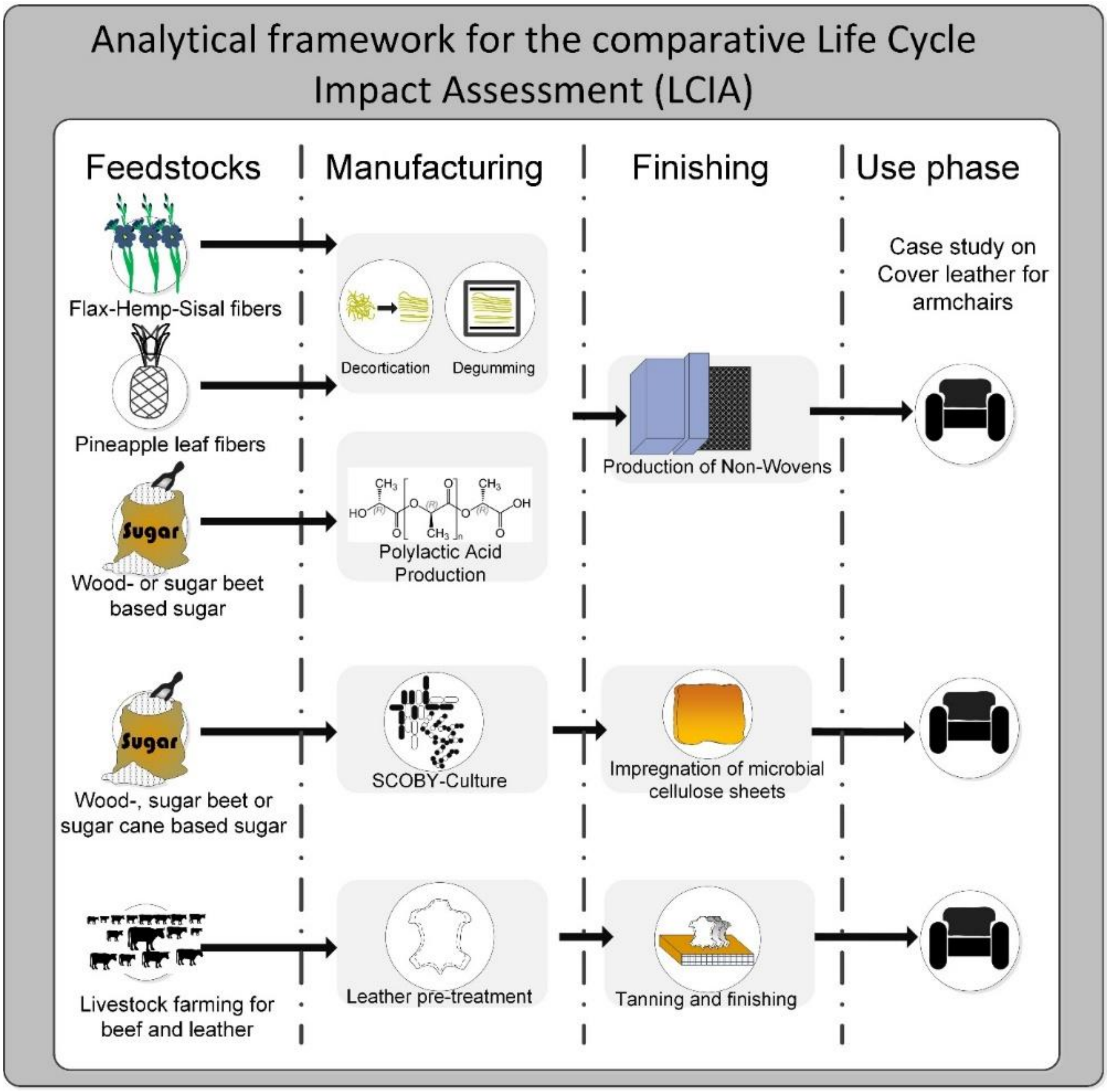

Figure 1: Scope and analytical framework in life cycle impact assessment (LCIA) of different leather substitutes

2.3 Research questions

In general, the circularity assessment must structure the alternative choices and the corresponding sub-scenarios for the comparative LCIA along the circular design choices, which can be considered throughout the production stages. Thus, the research questions involved whether: a. they relate to the resource base, b. they relate to the sensitivity of the results against the variances of the reference bovine leather production, or c. they relate to the sensitivity of the results against the variances during the use phase and end-of-life treatment scenarios.

2.3.1 Research questions regarding feedstock

Regarding feedstock, the following questions, which are summarized in Table 1, were identified to determine the quality of circularity, as well as the definition of allocation rules for the later impact assessment.

Table 1: Research questions concerning aspects on the feedstock side

Research question	Description	Motivation
1	How does the use of different fiber resources influence the comparative life cycle impacts assessment (LCIA)?	This depends on if a resource is cultivated as a main crop, e.g., sisal, hemp, flax, or kenaf, or if they are sourced as by-product from another main crop, such as fruit cultivation for the juice, and fruit production of pineapples for their leaf fibers. Regarding the case of pineapple leaf fibers (PALF), it is clear that they represent a type of agricultural by-product under a business-as-usual scenario. When comparing the order of magnitude this choice may have on the overall environmental impact footprint, it is useful to compare whether other fiber crops may have a substantially different environmental profile.
2	How does the feedstock choice, e.g., of wood-based sugar versus sugar-cane- or sugar beet-based sugar, influence the comparative LCIA?	Sugar was identified as a major input substrate in alternative leather production systems. Similar to the previous question, sugar-based input substrates can either be grown as main crop, e.g., in the case of sugar-beet based sugar, or it can be produced out of agricultural residues, such as lignocellulosic biomass.

2.3.2 Research questions regarding the influence of the finishing treatment for the enhanced durability on the LCIA results

When identifying the relevant specification factors for assessing the life cycle impact potentials of product finishes, its potential for durability enhancement and its associated influences on the use phase and end-of-life management options become critical considerations (Table 2). In particular, the material properties of biodegradability and durability both get an ambivalent character whether being a desirable or an undesirable property for further design preferences in product finishing.

Table 2 Research questions concerning the finishing treatment and the end-of-life treatment

Research question	Description	Motivation
5	How do different types of coating polymers and finishing agents influence the end-of-life options and consequently the comparative life cycle impacts assessment (LCIA)?	The alternative materials produced from microbial cellulose and fiber-based nonwoven materials not only face product design trade-offs between durability and biodegradability but also face issues regarding soaking water and lack of dimensional stability when prone to water. Therefore, impregnation and/or coating for increased hydrophobicity may be necessary to achieve good LCIA comparison results and reach internal design benchmarks.
6	How does the durability of the products vary concerning the application of different finishing agents and coating polymers, and consequently how does this influence the comparative LCIA?	When considering the finished products, whose leather-like properties were designed to exhibit durability similar to leather, it remains an important question whether a particular product in day-to-day application will indeed have the same product life expectancy as bovine leather products. In case of doubts or verified deviation from the durability, the product life time must be adjusted and a product replacement is introduced into the product-specific LCA modeling in order to compare all products against one common baseline.

Biodegradability is a preferable property as it helps direct alternative leather materials back into biological nutrient cycles.

Conversely, materials that are easily degradable often lack durability and therefore a coating or impregnation process may be a necessary design option. Moreover, the trade-offs between these two design choices and the natural durability of alternative materials are important factors that must be reflected when comparing impact factors and specific design options for each of the assessed products as their international benchmarks.

3 Materials and methods

3.1 System boundary, functional units, unit processes, and process inventories of applied life cycle assessment (LCA) approach

The LCA considers inventory data from cradle-to-gate system boundaries of particular process chains throughout the different material life cycle stages. On the upstream side of the processes, the unit processes included in the assessment encompass all agricultural provisioning processes for feedstock cultivation, harvesting, and transport, such as the supply of pineapple fibers, wheat straw, and husks from agricultural residues, and refined sugar for polylactic acid (PLA) production and microbial cellulose cultivation from sugar beet, sugar cane, and lignocellulosic biomass. The unit processes in the particular processing facility include all inventory data for required heating demand and electricity in the fermentation processes and polymer production. Herein, country-specific background emissions are not specified at the production sites. Rather, these data are used to evaluate the relative advantages against conventional leather products.

3.1.1 Functional units and the equality of benefits of the material alternatives and their references

The functional unit herein is 1 m^2 of leather patches applied as an upholstery leather substitute on armchairs. The statistical or estimated product life time, which is individual for each product type and its surface coating or impregnation type, is used as a benchmark for comparison to evaluate if product replacement is required for products with lower life time expectancies than that of conventional leather. The functional unit of the assessed alternative leather substitutes has to be brought on to a common basis for comparison. The equality of benefits is evaluated by identifying the density, thickness, tensile strength, and durability, wherein the four different materials need to be comparable regarding their material properties. These properties are summarized in Table 3 for each assessed alternative.

Table 3: Material properties of the assessed alternative upholstery materials

Material property	Alternative 1: pineapple leaf fibers (PALF) or flax-hemp-sisal nonwoven with polylactic acid (PLA) matrix and polyurethane (PU) cover [density > 30 kg/m^3]	Alternative 2: Impregnated microbial cellulose upholstery	Reference material: Bovine leather upholstery	References
1. Area of the upholstery [m^2]	1	1	1	Definition of functional unit
2. Thickness of upholstery sheets [mm]	1.6 ± 1	1.7	2	Basis for equality of benefits regarding tensile strength
3. Specific weight of the upholstery sheet dry tanned g/m^2	400	480	4 oz per ft^2 =141 g/ft^2 =1517 g/m^2	Calculated from 4.
4. Density kg/m^3 dry output finished	250 ± 15.6	285.00 ± 15	758.0 ± 45	(Damsin, 2019; J. Buljan, G. Reich, J. Ludvik, 2000)
5. Density kg/m^3 wet input	n.a.	999.0 ± 10 with 85 % water content	1433.0 ± 25 with 46 % water content	(Damsin, 2019; J. Buljan, G. Reich, J. Ludvik, 2000)
Replacement factor	Once every 5 years	Twice every five years	Once every five years a	due to abrasion and reduced durability

Only when the equality of benefits is ensured in definition of functional units for a set of product groups can more encompassing assessments be conducted, e.g., comparing full or partial market substitution strategies or additional benefits in meeting future market demand with alternative leather substitutes in contrast to further increasing bovine leather utilization.

3.1.2 Unit processes for biofabrication and nonwovens manufacturing assessed in the LCA

The production processes for the alternative leather substitute materials assessed herein involve mechanical needle fleece manufacturing and microbial production of microbial cellulose. In this section, the major features of the most relevant unit processes involved in leather substitute manufacturing are briefly described.

Biofabrication of Microbial cellulose

The biofabrication of bacterial cellulose relies on a variety of possible bacterial strains and symbiotic cultures of bacteria and yeast (SCOBY). Microbial organisms that have high yields of bacterial (nano-)cellulose exist in the acetic bacteria species, including *Gluconacetobacter xylinus*, *Komagataeibacter xylinus*, *Komagataeibacter hansenii*, *Komagataeibacter kombuchae, Komagataeibacter intermedius*, and the yeast *Zygosaccharomyces bailli* (Belgacem, 2008; Niyazbekova et al., 2018; Sederavičiūtė et al., 2019). Some major differences between plant cellulose and microbial cellulose are that microbial cellulose has a higher water content, is free of lignin and hemicellulose, and has a far higher crystallinity (Gandini and Belgacem, 2008). The choice of the feedstock for bacterial conversion is very important as it affects the environmental footprint of the entire production process, as well as the properties of the final products, e.g., flame retardancy and hydrophobic properties. However, trade-offs between the choice of substrates and process stability as well as product yield are also important factors that must be evaluated before substrate selection (Jozala et al., 2015; Żywicka et al., 2018, 2018). Microbial cellulose is a very versatile platform technology for production of a broad range of applications, including as stabilizers for emulsions, artificial textiles, sponges, water filtration devices, and medicinal artificial tissues (Ashjaran, 2013; Belgacem, 2008)**.** Therefore, the biofabrication of bacterial cellulose is regarded as a major bioeconomy technology, meaning its sustainability and associated footprint in the downstream processing and finishing phases should be closely monitored.

Manufacturing of nonwovens from natural fibers with biopolymer matrix

Nonwovens are a group of bio-based materials that have a broad range of technical and fashion related applications. Nonwovens are utilized in filter application, environmental textiles, agricultural uses, such as mulching techniques and erosion control, building processes for drainage and underlying systems, and in fashion products, such as artificial leather products (Geus, 2016). The production of nonwovens begins with the cultivation of fiber crops, followed by retting, decortication and/or fiber separation processes, and finishes with the manufacturing of nonwovens using the extracted bast fibers. The natural fibers used in nonwovens manufacturing can be sourced from different fiber plants, such as sisal, hemp, flax, cotton, and pineapple (Karthik et al., 2016; Peças et al., 2018; Sisti et al., 2018). These plant fibers can originate from bast, leaf, fruit, seed, wood, and grass fibers. The origin of the plant tissues is highly decisive as it determines which retting process is applicable and what fiber length compositions can be extracted (Sisti et al., 2018). Although the use of natural fibers in fiber reinforced composites, technical textiles, and leather substitutes is often regarded as more environmentally friendly than synthetic fibers, fiberglass, or bovine leather, sustainability issues, such as water resource depletion, pesticides use, and working conditions, must be closely monitored before conclusive results for individual products can be obtained (Rana et al., 2014). The retting process involves mechanical, chemical, and biological processes (Sisti et al., 2018). Once the natural fibers are extracted, separated, and cleaned, the manufacturing of nonwovens includes various processes, such as air laying, wet-laying, needle punching, stitch bonding, hydro entanglement, thermal bonding, and adhesive bonding, depending on both the intended application and the additional fabrication materials used, such as thermoplastic fibers and adhesives (Horrocks and Anand, 2000).

Using polylactic acid (PLA) polymers within nonwovens as biopolymer matrix

PLA monomers can be produced as L(+) and D(−) stereoisomers through fermentation processes involving starch and sugar crops, as well as treated cellulosic and lignocellulosic biomasses. Generally, through the condensation of lactic acid, low-molecular-weight PLA can be derived and further converted into L(+) and D(−) stereoisomers through depolymerization. It can then be further processed using chain growth reactions into a high-molecular-weight PLA. The main bacterial species that are deployed in the fermentation processes are from the *Lactobacillus* genus (Jamshidian et al., 2010).

The resulting PLA polymers can be converted into polymers granules for extrusion, injection molding, and blow molding processes, or be spun into fiber PLA. Fiber PLA can then be processed into nonwoven materials in combination with both fossil-based non-biodegradable polymer fibers and natural fibers and/or other bio-based biodegradable polymer fibers (Jamshidian et al., 2010). Herein, we assess the use of PLA polymers from both sugar and lignocellulosic sources for thermal bonding with needle fleece materials from PALFs (Qua, 2019), hemp, sisal, and kenaf fibers (Karthik, 2017).

3.1.3 Main factors of the life cycle inventory analysis

Inventory data were collected to establish material and energy flow balances and to specify the input substrates, input factors, and conversion factors.

The main feedstocks on the input side include sugar beet based and wood-based sugar for fermentation processes, nutritive supplements and nitrogen sources for fermentation processes, such as boiled tea, nutritive and amino acid supplements, and agricultural by-products, such as PALFs. Regarding energy and fuel input, transportation, including freight, truck, and train, and treatment processes, such as boiling, heating, and drying, were included in these calculations. The sources used were the most recent publications of manufacturers and researchers as well as benchmark studies on bio-based processes and technologies (Table 4).

Table 4: Specific input factors and conversion coefficients to produce 10 m^2 of cover leather sheets

	Leather alternative 1: pineapple leaf fibers (PALF) or Flax-Hemp-Sisal Fleece with polylactic acid (PLA) matrix	**Leather alternative 2: Impregnated Bacterial cellulose**	**Reference material: Bovine leather upholstery**	**References**
By-product ratios	90% pineapple fruits from input	90% kombucha tea from input	Meat	Supply chain parameter
Sugar mix	100% sugar beet vs. 100% wood-sugar	60% sugar beet vs. 40% sugar cane	Not applicable	Modeling parameter (relevant for sensitivity analysis and internal benchmarking)
Transport distances	18301 – 24250 km by sea freight for nonwovens (Manila to Rotterdam or Hamburg)	11500 – 16500 km by sea freight for tea from Kenya	31000 km as weighted average, global transport by sea freight, BRICHS to China and Italy to Northern Europe,	www.sea-distance.org, freight of bio-based commodities from most important international ports
Glucose conversion efficiency	65% – 85%, just applicable for PLA	45% – 68%	Not applicable	(Chawla et al., 2009; Iffland et al., 2015)

3.1.4 Compilation and justification of allocation rules applied in life cycle inventory modeling

For each of the resources involved in the production of the assessed bio-textiles, appropriate allocation rules need to be defined, selected, and applied. In particular, when assessing the material's influence on reducing environmental impacts, strategies for increasing the circularity within production chains might require definition, defense, and outlining of the chosen allocation rules (Table 5).

Table 5: Allocation rules applied in the life cycle modeling of the assessed leather substitute materials

Assessed leather substitutes	Involved material flows that demand for allocation	Allocation rules applied between by-products for the main scenario	Allocation rules for the sub-scenario
Alternative 1: pineapple leaf fibers (PALF) or Flax-Hemp-Sisal nonwoven with 20% polylactic acid (PLA) matrix and polyurethane (PU)-Coating	Pineapple fibers as by-product from Pineapple cultivation Industrial sugar feedstocks used for lactic Acid fermentation	Under the general assumption that there are no negative impacts allocated in the upstream chains of PALF provisioning, impacts of PALF- fibers can be balanced through allocation by price as future system expansions of pineapple cultivation could be positively influenced by fiber prices, rather than by fruit prices	Allocation by price is applied for sugar-based input substrates supplied by lignocellulosic feedstock biorefineries
Alternative 2: Impregnated Microbial cellulose sheets from symbiotic cultures of bacteria and yeast (SCOBY) fermentation processes	Industrial sugar feedstocks used as feedstock for yeast and bacterial growth and kombucha tea marketed as co-product	Allocation rules are applied for the marketing of kombucha tea as a main by-product of microbial cellulose production. The life cycle impacts assessment (LCIA) results are later compared considering allocation by price and mass.	Allocation by price is applied for sugar from lignocellulosic feedstock biorefineries

These allocations precisely differentiate between the main resources, business-as-usual production systems, and coupled use and co-production schemes that rely on agricultural residue and waste flows, which would be prone to mineralization or underuse in the absence of the proposed innovative production systems. Furthermore, when assessing novel biorefinery production platforms that produce tradeable feedstocks commodities, e.g., lignocellulosic feedstocks, a clear statement and selection of allocation rules may have a major influence on the outcome of the corresponding LCIA.

3.2 LCIA modeling with the associated process modules and the used LCIA datasets

LCIA modeling is conducted using the Software GaBi 6 based on the datasets of GaBi database Professional + ecoinvent integrated, the results of researchers from the UFZ Department of Bioenergy, and our own inventory processes modeled according to the systems analysis. The model includes all relevant unit processes in the two major production lines of nonwoven based and bacterial nano cellulose (BNC) based leather substitutes. These substitutes different sub-scenarios in feedstock supply and resource allocation, the unit processes of the individual production systems, and the by-products and their respective allocation rules are shown in Figure 2.

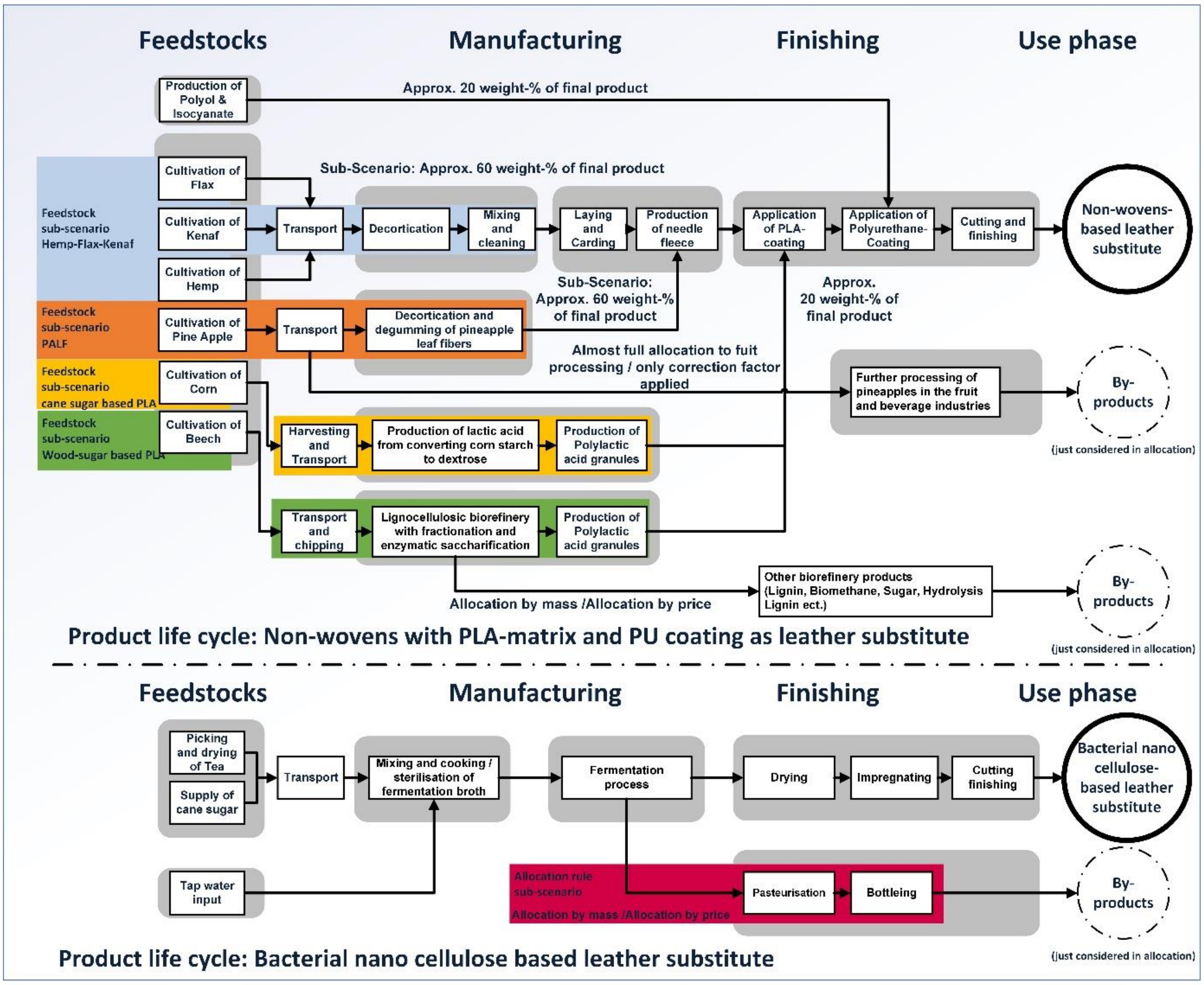

Figure 2: Overview of the unit processes, value-added chains, and sub-scenarios for production of alternative leather materials

The allocation rules shown in Figure 2 are the same as those listed in Table 4. The resources for production of polyol and iscocyanate remain fossil-based in this study, but feedstock substitution using biomass resources, such as castor oil, could be introduced. For the LCIA modeling, which is conducted alongside the unit processes as shown in Figure 2, the datasets used in the GaBi®-Software are summarized in Table 7.

The transport datasets include transportation by ship and truck, as listed in Table 6. The electricity grid mix applies to both Asian and European countries, which is where the fibers and nonwoven fleeces are produced.

Table 6: Unit processes of the life cycle impacts assessment (LCIA) model, and datasets used for LCIA modeling and their references

Product alternatives	Process type	Unit processes	References of datasets
Upholstery from pineapple leaf fibers (PALF) - nonwoven-with polylactic acid (PLA) matrix and polyurethane (PU) coating	Transport Nonwovens	Transoceanic ship, bulk, 100 – 200 k dwt	EMEP - CORINAIR Emissions Inventory Guidebook, 2011
	Production of PLA	a. Ingeo PLA b. PLA from life cycle biorefinery	EcoInvent 3.5 Budzinski und Nitzsche 2017
	Cover material backing	RER: Electricity grid mix - EU-28: Polyurethane (PU) flexible foam - TDI-based, no flame retardant, high density	Our modeling with GaBi Thinkstep database 2019
	PALF-PLA Nonwovens finishing	Electricity grid mix	GaBi Thinkstep database 2019
Sub-Scenario: Nonwovens from sisal-hemp-kenaf fleece mix	Production of a sisal-hemp-kenaf needle fleece	DE: Fleece from mixed fibre (flax, hemp, and sisal), agg.	GaBi Thinkstep database 2019
Upholstery from Impregnated microbial cellulose	Tea production	Tea production, Kenia	EcoInvent 3.5
	Tap water input	Tap water from groundwater	GaBi Thinkstep database 2019
	Sugar input for microbial cellulose production	ROW: Beet sugar production BR: Cane sugar production with ethanol	GaBi Thinkstep database 2019
	Tea brewing for fermentation broth	Heat input, tap water input and tea input	Own modeling with GaBi Thinkstep database 2019
Sub-Scenario Varying: Sugar supply mix	Kombucha Fermentation	Input of fermentation broth and input of living symbiotic cultures of bacteria and yeast (SCOBY) material for inoculation	Own modeling with yield factors and material and energy flow balances as depicted in table 3 and 4
	Impregnation of SCOBY	GLO: Crude coconut oil (including LUC)	ERASM Surfactant Life Cycle and Eco footprinting (SLE) Project: GaBi Thinkstep database 2019

Abbreviations: Agg: Aggregated, ROW : Rest of the World, GLO: Global, DE:Germany, BR:Brazil, RER:Europe, LUC: Land-use change

3.3 Robustness of model with regard to variances and uncertainty in the finishing processes

Further uncertainties are expected from variances in the finishing stage of the production life cycle of the alternative leather substitutes.

The main issue regarding product durability in different environments and under product safety constraint is ensuring hydrophobic properties, fire safety, e.g., through application of flame retardants, and the avoidance of brittleness, e.g., through the use of softening agents. These desirable properties can be guaranteed by certain biological, chemical, and physico-chemical material design options that either utilize additives in the finishing phase or additive shifts in the biological feedstock materials, or by physico-chemical treatment processes. Microbial cellulose sheet finishing requires additive materials for hydrophobic finishing, fire safety, and softening agents. The possible materials which can be used involve natural wax and/or fossil-based paraffin as a hydrophobic agent and bio-based oils as a softening agent.

4 Results

The LCIA compares the potential reduction of environmental impacts according to the Environmental Footprint (EF) 2.0 impact categories, and includes toxicity (tox) categories for individual processes to show the relative advantages compared with bovine leather upholstery as derived from the dataset EC, DG ENV 2018, which details finished leather for automotive and upholstery, preservation and tanning, and consumption mixes (section 4.1), at tanning plant (de)" (EC, 2018) in section 4.1.

Furthermore, the contributions analysis helps to identify environmental hot spots concerning individual product-specific unit process modules that cause negative environmental impacts, thereby highlighting areas that require impact reduction.

Finally, all three alternative leather substitutes are compared with an aggregated single score (Global equivalents) using EF 2.0 with tox categories in section 4.2.

4.1 Energy and material flow balances and their variances

The major material and energy flows associated with the biofabrication and manufacturing of leather, as compiled in table 7, include the water footprint, the cumulative energy demand for drying, boiling, pressing, and confectioning of the materials, and the upstream material and energy demands for supplementary materials.

Table 7: Material input and energy demand per functional unit of 10 m² and their respective variances

	Leather alternative 1: Nonwovens from leaf fibers or flax-hemp-sisal with polylactic acid (PLA) matrix as upholstery	Leather alternative 2: Impregnated microbial cellulose upholstery	Reference material: Bovine leather upholstery	References
Sugar input	0.256 kg/m²	100 g/l	N/A	(Damsin, 2019; Faida, 2017)
Water [m³/m²]	For pineapple leaf fibers (PALF)-cultivation the water use is allocated to fruit use, 0.144 – 1.44 for PLA, depending if rain-fed or irrigated	0.060 plus 0.2 for finishing	Between 0.030 and 0.350	(Chawla et al., 2009; Damsin, 2019; Jayabalan et al., 2014; Laurenti et al., 2017; Morão and Bie, 2019; Qua, 2019; Reich and Taeger, 2009)
Total energy demand [kWh/m²]	1,77 total and 0.58 non-renewable for PLA-Matrix, 27.5 as cumulative energy demand	26 kWh per kg of fermentation broth, including non-oxidized biogenic energy in fermentation products	2.8 – 16.0	(Laurenti et al., 2017; Morão and Bie, 2019)
Amino acid supplements [kg/m²]	N/A	32 kg Tea input per FU	N/A, protein fodder for livestock	(Jayabalan et al., 2014)

4.2 Results of the LCIA for the three leather substitute materials and their sub-scenarios

The results show that the allocation rules matter significantly for both microbial cellulose based leathers as well as nonwoven based alternative leather substitute materials.
For the nonwoven based materials, the land-use impact differs by almost 300% between the flax-hemp-sisal-based product and the bovine leather (greater than 200%) and PALF-based leather (greater than 450%), as shown in Fig. 3.

For microbial based cellulose, the different impact categories differ between 10% and 30% between the allocation by price and the allocation by mass, and between 50% and 260% between the allocation by mass and the replacement factor of 2 with allocation by price, as shown in Fig. 4.

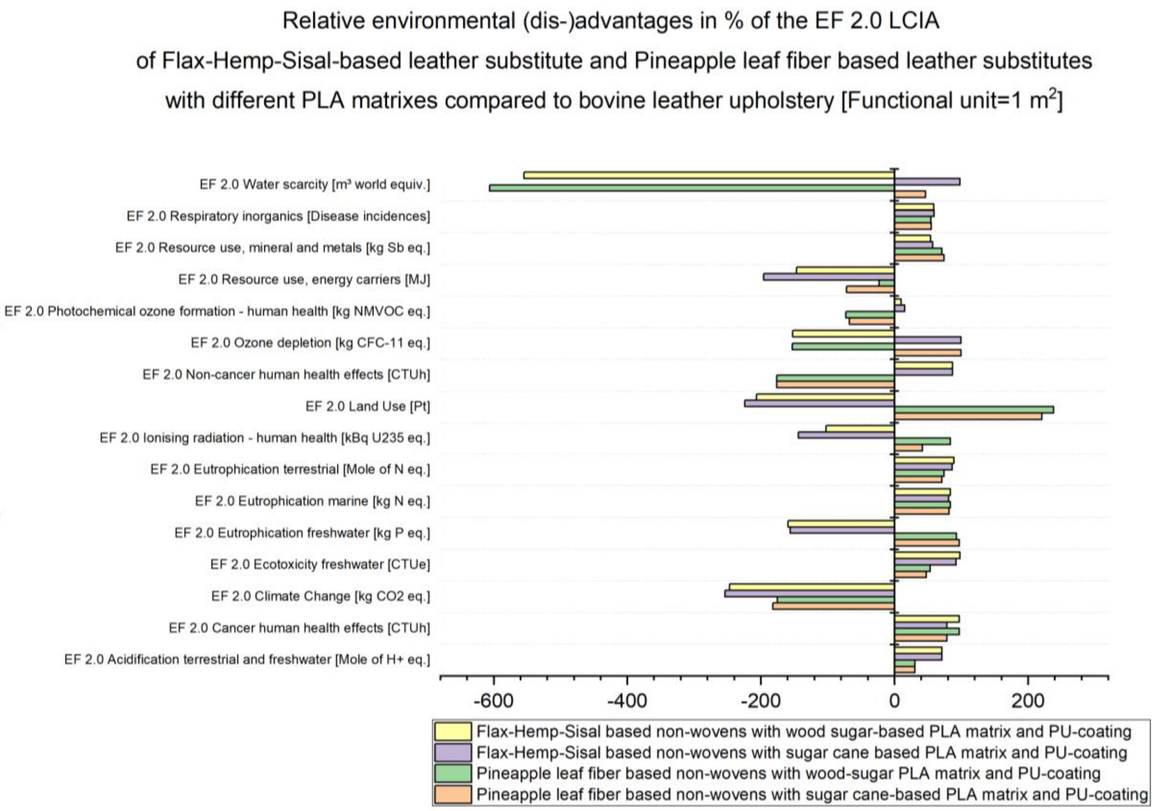

Figure 3: Relative comparison of Environmental Footprint (EF) 2.0 impact categories for four material compositions of fiber based nonwovens against bovine leather upholstery from dataset EC, DG ENV 2018 (EC, 2018), Remark: Allocation rules in the upstream system were not compared as they mainly include low-value organic fertilizers.

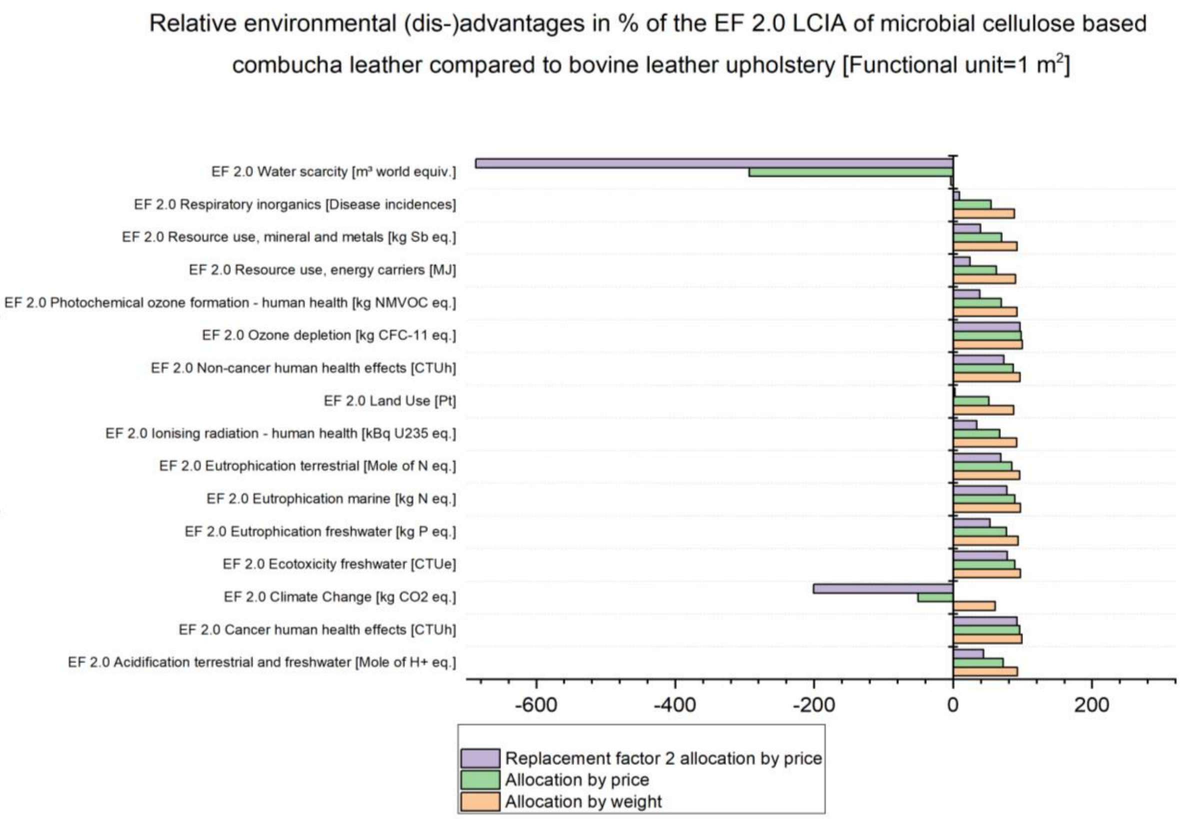

Figure 4 Relative comparison of Environmental Footprint (EF) 2.0 impact categories comparing case study three of microbial cellulose against bovine leather upholstery from dataset EC, DG ENV 2018 considering allocation rules for microbial cellulose by price and by weight against kombucha tea sales.

4.2 Results of the contribution analysis and the cumulated single score assessment comparing the EF 2.0 Global Equivalents

The results of the contribution analysis, which is presented in Fig. 5, revealed that in all 16 impact categories, the production of PLA matrix materials and flax-hemp-sisal fleece, and the upstream production systems of the PU based coating comprised the highest shares of potential negative environmental impacts. Individually, they contributed at varying degrees to the following categories:

- Water scarcity is highly attributed to the land use system and pre-treatment of lignocellulosic biomass for PLA production, whereas the land-use impacts category is dominated by the upstream cultivation systems of flax, hemp, and sisal fibers.
- The non-cancer health effects are attributed to the upstream production of raw materials for PU-coating, which is the largest contributor to this category.
- Concerning the resource use and energy carriers category, the shares between these three processes are nearly equal for the fleece production and PU coating upstream systems.

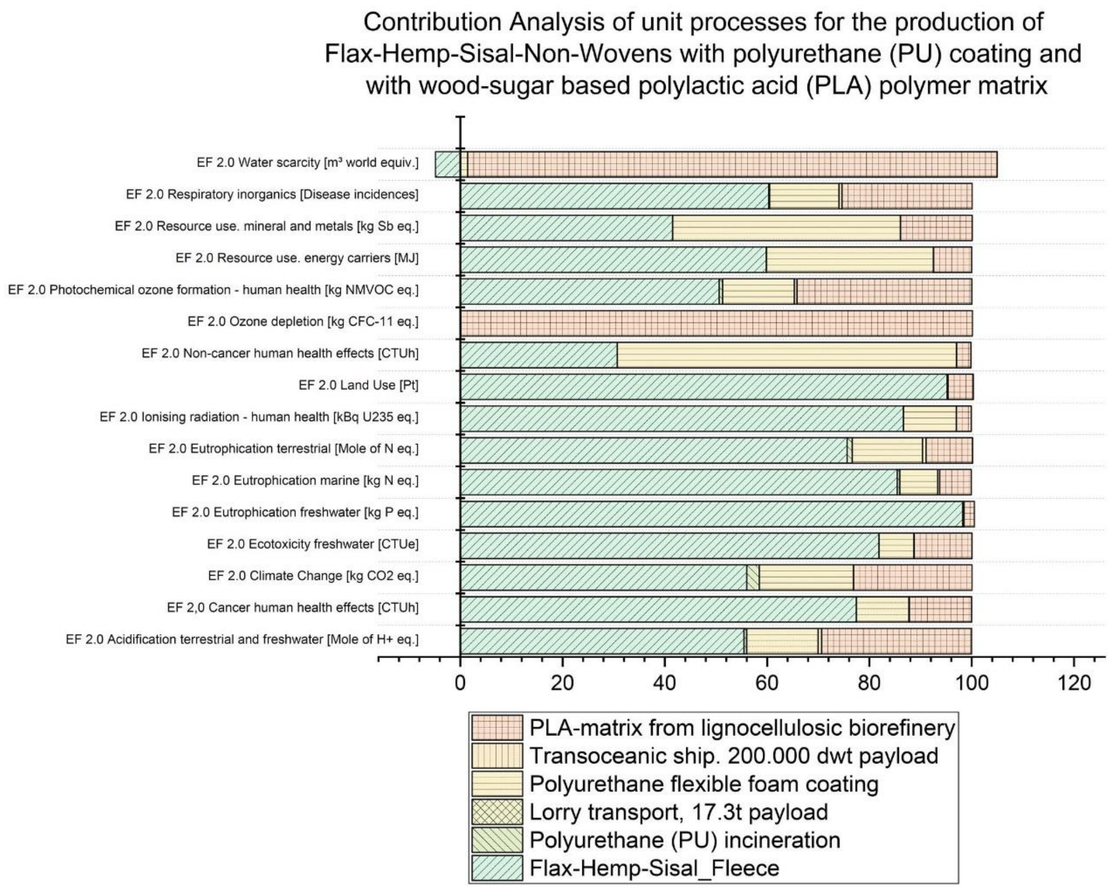

Figure 5: Contribution analysis of the unit processes for flax-hemp-sisal leather substitute production

When comparing the LCIA results of the flax-hemp-sisal based leather substitute with the production system, which relies on PALF as a fiber feedstocks, one can observe a significantly diversified picture as the LC impacts of the PALF-based non-wovens product are not dominated by the fiber production system. Regarding PALF nonwovens, wherein fiber production is not the dominant factor, the shipping emissions and impact of the PU coating are the major unit processes causing negative environmental impacts. Further, although the PLA polymer matrix comprises a significant fraction of the material composition, the associated environmental impacts are not represented in a quantity directly corresponding to its weight fraction. The results of the LCIA show that the use of PU coating for PALF and flax-hemp-sisal based nonwovens results in a comparable life time for each substitute. However, the contribution analyses dictate that a substantial fraction of the sensitivity (25% – 30%) of the LCIA contribution, e.g., resource use and energy carriers, is a result of PU-coating application. Relying on the contribution analysis for the microbial cellulose leather materials, as shown in Fig. 6, a major finding was that upstream impacts of tea production, the environmental impacts associated with the electricity and heat demand for the heating and pasteurisation of the fermentation broth, and the impacts of sugar cane production are key impact hot spots.

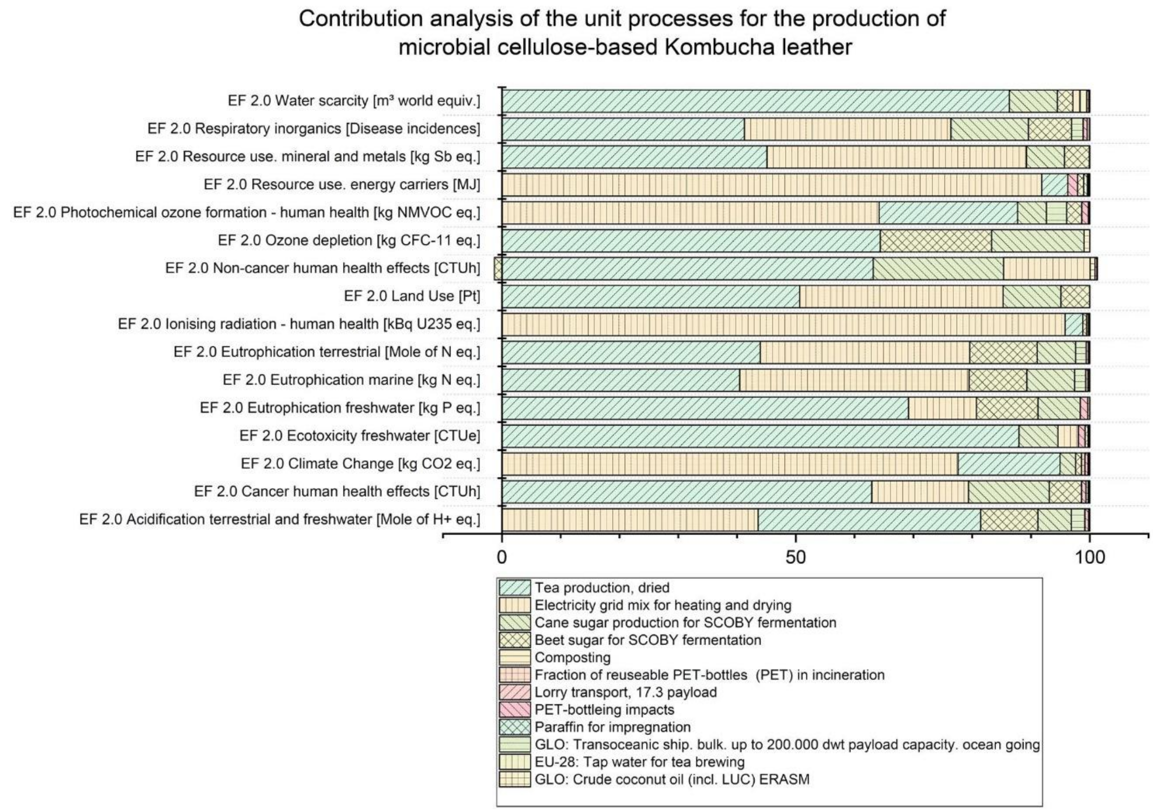

Figure 6: Contribution analysis for unit processes of microbial cellulose-leather production

4.3 Results of the cumulated single score assessment comparing the EF 2.0 Global equivalents

In this section, the results of the selected EF 2.0 impact categories are compared, as shown in Fig. 7. Weighting of the impact categories allows for comparison against a baseline weighted single score. Given this comparison, it is clear that the negative environmental impacts associated with microbial cellulose leather comprise only 80% of the total environmental impacts of PALF based leather, and that the leather-substitute based on flax-hemp-sisal fiber nonwovens has a negative environmental impact of approximately 10% higher than that of the PALF-based leather substitute during from the cultivation to factory gate stages.

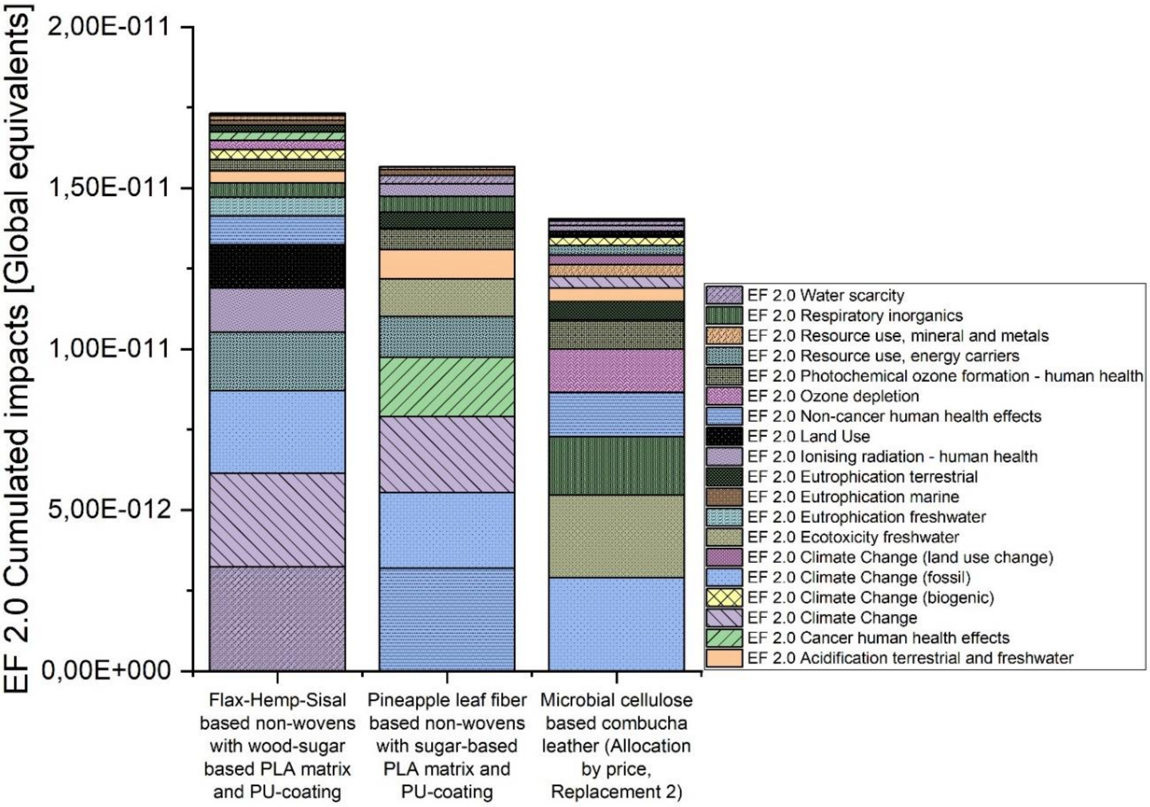

Figure 7: Comparison of the cumulated life cycle impacts for the three selected product compositions weighted according to Environmental Footprint (EF) 2.0 global equivalents

4.4 General results of the study with regard to the research questions

In this section, the results of this study are evaluated with regard to the potential answers to the four initial research questions based on the results of the LCIA, the contribution analysis, and the sensitivity analysis.

Regarding research question 1, which discusses decoupling from feedstock impacts, we determined that the influence of using different fiber resources on the comparative LCIA must be addressed in a very differentiated manner for the individual impact categories.

Some of our findings were straightforward, such as that of the PALF being a by-product of pineapple cultivation, wherein the land-use impact for PALF-based nonwovens are considerably low, whereas for the flax-hemp-sisal based nonwovens we found that they have a significantly higher land-use impact and higher water consumption even compared to the reference bovine leather upholstery allocated by mass after diverging from meat production.

Concerning research question 2, by comparing the environmental advantages of deploying sugar-beet based saccharose, the LCIA for microbial cellulose clearly shows that for a broad variety of impact categories the use of sugar-beet based sugars as a feedstock has a lower environmental impact. On average, for every impact category, the advantage would account for approximately 16%. For the category with the highest impact reduction potential (human health, no-cancer), the impact is approximately 100% lower (1/2) compared to the environmental impacts of sugar cane. The eutrophication potential, another major category, shows that sugar cane accounts for an approximately 75% lower environmental impact as a result of the environmental footprint methodology.

Concerning research questions 3 and 4, the results support the finding that even though the type of coating influences the end-of-life options, and consequently the comparative LCIA, the overall sensitivity of the results against the coating is rather low compared to the benefit of prolonging the durability of the product. The replacement factor of 2, which is used to ensure the quality of lifetime benefits, could cause an approximately 160% higher total environmental impact regarding the chosen functional unit compared with the approximately 10% to 15% higher impact associated with the enhanced durability from coating applications. Therefore, the biodegradability after its life time might be a company target for product claims and marketing, but long-term durability would have a better environmental impact. This trade-off will continue to be controversial as it might underpin fast fashion claims without sufficiently reducing environmental impacts. Note that only when further impact reductions of approximately 40% – 60% are achieved will the loss of life time through lower durability be outweighed by the environmental preferability of biodegradable coatings for more fast fashion applications.

5 Discussion

Considering the associated impacts of by-product flows, such as kombucha tea production and pineapple cultivation, clearly defined allocation rules are decisive factors for the accurate modeling of the associated environmental impacts. Although producers claim that the environmental impacts of pineapple cultivation or kombucha tea production and their downstream marketing can be neglected, the results herein show that under system expansion points of view, the actual market size and changes in agricultural management patterns induced by expanded leather substitutes production must be considered.

Market oversaturation of kombucha tea can lead to decreasing kombucha tea prices, thereby influencing the allocation by price as a higher share of revenues will be from alternative leather sales. Conversely, the production of PALFs requires additional collection forces and treatment spaces, thereby occupying a share of the land that would be otherwise useful for pineapple cultivation. We included these factors into our impact assessment by accounting for them according to the allocation rules described in the methods section.

The replacement factor and product life span are important factors for all leather products and substitutes in general. However, the life span of leather upholstery is assumed to be considerably lower than other leather products.

Consequently, herein we used for a rather conservative replacement factor. Even for the less durable leather substitutes it was assumed that a replacement factor between 1 and 2 was reasonable. When expanding this assessment toward more durable leather product applications, this reference value had to be raised to a replacement factor of the assessed leather substitutes, between 4 and 5. Meanwhile, due to the superior durability of the leather product in these cases no replacement would be accounted for in the LCIA of the leather upholstery production.

By evaluating our results in regard to the initial research questions, we identified further options for enhancing the circularity of these fashion materials, which could contribute to further reduction potentials of environmental impact during the life cycle stages. In sections 5.1 through 5.4, we discuss some of the most crucial findings and relevant intervention areas.

5.1 Further options for increasing the circularity regarding feedstock

To increase the circularity within microbial cellulose production, the use of waste substrates, e.g., from secondary brewing of waste substrates from tea brewing and instant tea production in the beverage industry (Pelvan and Özilgen, 2017) or pasteurized fruit waste (Abol-Fotouh et al., 2020) e.g. from fruit jelly production , could allow for a circular supply chain strategy (Jozala et al., 2015). Furthermore, integrating starch producing companies, such as potato starch production, with enzymatic saccharification combined with a later integration into existing wastewater infrastructures should be considered.

However, note that when the allocation by price or by weight starts to become obsolete in the waste-based microbial cellulose cultivation by losing the option for marketing of a valuable co-product, such as kombucha tea, the aqueous biowaste substrates will have to be treated as wastewater, resulting in a major drawback for burden allocation. An intelligent loop back into the existing waste and wastewater treatment infrastructures, as described for the case of potato starch, without further capacity increase could help mitigate these additional burdens, which would otherwise have to be allocated to a single product instead of two co-products.

5.2 Further options for increasing the circularity of production

Circularity regarding production can be increased when co-products are derived. For example, using the short fibers and leave residues from the non-used PALF leaf fractions and flax-hemp-sisal stems for further valorization streams, such as biogas co-substrates, including fungal cultivation co-substrates or further material use as biochar or composite supplements. Regarding kombucha brewing, further use of the spent tea substrate, such as insect cultivation for biogas processes, or for second brewing in production lines for non-beverage producing microbial cellulose should be considered.

5.3 Further options for increasing the circularity from an end-of-life perspective

The durability of bovine leather is a major product benefit concerning the reference system. Equal durability for each of the substitutes can only be achieved through an additional input of energy and supplemental materials, which is verified by the inventory and impact analysis. The shift in resource base from bio-based feedstocks for coating and impregnation agents does not lead to a substantial impact reduction if these coating agents are not derived from waste-based substrates.

5.4 General findings of this study

The life cycle based assessment conducted in this study helped strengthen the understanding of which life cycle stages in the process of manufacturing alternative leather materials are hot spots of environmental impacts. Furthermore, it revealed the challenges that should be solved as companies and the textile sector aim to upscale these bio-textile materials. The assessed materials herein each have specific areas where upscaling might create positive or negative spill-over effects, either in the beverage industry, fruit industry, or biotechnology supplement production.

Market uptake and the upscaling of by-products is a key factor that must be closely considered, as well as the types of coatings and/or impregnation agents applied to the substitute materials to increase durability.

Tea production and brewing for SCOBY biofabrication and PLA-biopolymer and PU coating production are the major impact factors identified along the value-added chains of alternative leather manufacturing.

6 Conclusions and further research demand

The study revealed that minimizing the environmental impacts of leather mimicking and substituting materials is a multi-facetted and multi-factorial endeavor, showing that slightly different changes in the processes can result in the offsetting of all the advantages associated with the bio-based alternatives.

6.1 General conclusions

In summary, we have determined that these alternative leather substitutes contribute to potential long-term strategies for reducing and replacing leather use, but only at a well-balanced mix when upscaling is predicted, and only with the leverage potential for deep impact decoupling. This demands that fast fashion applications do not compromise the attainable impact reductions but instead embrace further impact reduction strategies.

We also conclude that the full potential of impact reduction is still untapped, as the upscaling of beverage production lines and further high-value added by-product use, as well as internal resource recovery loops, are still unexplored with regard to their techno-economic performance and their impact on process stability or infrastructure trade-offs.

Further, we can conclude that relying on the natural synthesis of leather as a natural and free by-product of cost-intensive and high impact meat production is an increasingly misleading assumption when considering a world of increasingly scarce land, fodder resources, and growing population demand for meat consumption in absolute numbers. Therefore, allocation by price is an increasingly accurate allocation rule for differentiating the impacts between meat and leather, especially considering future cost drivers and land use pressures.

6.2 Further research demand.

There exists major demands for future research that considers even more holistic and integrated assessments by conducting complementary LCC assessment studies and social life cycle assessment studies to evaluate the economic and social sustainability of the presented value-added chains in nonwovens and microbial cellulose materials.

Furthermore, considering the technical and sectoral integration perspective we identified herein, there exists further research demand for in-depth integration into existing facilities of starch producing factories by means of industrial symbiosis, and the full transition of the assessed value-added chains toward entirely bio-based systems by also substituting fossil-based coating systems, such as the fossil-based PU-coating assumed in this study.

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