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Life cycle assessment of suberin and betulin production from birch bark

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ABSTRACT

In this study, the environmental impact of suberin and betulin production from birch outer bark was evaluated using life-cycle assessment (LCA), and the ReCiPe 2016 Midpoint (H) method. The system boundary includes production of outer bark to betulin and suberin production, with bark residues utilized for energy production, including all inputs (materials and energy) and outputs. Three scenarios were used to examine using the allocation, extraction and hydrolysation of birch bark. In the baseline scenario (S1), the bark came from a biorefinery and processed in a separate unit, where the hydrolysed bark residue was incinerated to generate the heat; the integrated scenario (S2) and internal loop scenario (S3) considered production within a biorefinery but with different energy sources. The primary data were collected from laboratory experiments, and secondary data were taken from the literature and the ecoinvent 3.9.1 database. FU was 2.06 kg of polymer production that includes 1 kg of suberin and 1.06 kg of betulin production from the same process. The results showed that the global warming potential (GWP) for S1 was 15.5 kg CO_2 eq. per FU, 16.22 kg CO_2 eq./FU in S2 and 3.96 kg CO_2 eq./FU in S3. According to our results, increasing the recycling rate of ethanol (50%, 90% and 98%) in the process and using wood-based energy from the biorefinery instead of Finnish mixed electricity resulted in a significant reduction in the most important environmental impact categories except land use.

1. Introduction

The United Nations established the 17 Sustainable Developments Goals with the aim of creating a more sustainable society by replacing fossil-based products with bio-based renewable materials (United Nations, 2023). Forest biomass is a renewable resource that acts as a storage for carbon and supports biodiversity (Giuntoli et al., 2022). However, there are limits on how much biomass can be sustainably harvested from forests (Oettel and Lapin, 2021). These limits encourage more efficient use of harvested wood and side streams (Jyske et al., 2023). The circular bioeconomy combines these visions and can focused on achieving sustainable and resource-efficient biomass utilization through integrated production chains that use residues and waste, while increasing the value of biomass over time (Jyske et al., 2023). By developing innovative methods for manufacturing bio-based chemicals that can replace fossil-based alternatives is a crucial aspect of addressing current global challenges (Yadav et al., 2020; Harman-Ware et al., 2021).

In Finland, 75% of land area is covered by forest, making it one of the most forested countries in Europe. Finland's economy relies significantly on the forestry sector (Georgescu et al., 2024). The production of roundwood plays an important role in producing of various forest-related products (Georgescu et al., 2024). The roundwood production reached approximately 510 million m^3 worldwide in 2022 (Eurostat, 2022). Since then, these processing volumes gradually increased. In Finland commercial felling in 2023 totalled 65.9 million m^3 of roundwood (Eurostat, 2022; Luke, 2023). The main species were Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*). While, Broadleaf species included mainly silver birch (*Betula pendula*) and downy birch (*Betula pubescens*), which totalled 9.5 million m^3. The amount of bark in stem wood varies between 10% and 13.8% depending on the species and assortments, with birch pulpwood having the highest percentage (Luke, 2023). The Finnish Forest industries produced approximately 7 million m^3 of bark in 2021 (Jyske et al., 2024), of which 1.44 million m^3 was from birch (Luke, 2023). Green chipped bark can be used for mulching, but in industry it is almost solely incinerated as solid fuel for process energy or in combined heat and power units for the district heat or grid energy (Luke, 2022). As a non-wood lignocellulosic

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material, bark contains valuable chemical components, and has attracted attention in recent years for its potential in value-added applications (Korkalo et al., 2023). Recently, Ding et al. (2017), Rasi et al. (2019), Carlqvist et al. (2022) and Dou et al., 15 studied the extraction of softwood bark, containing hydrophilic compounds such as stilbenoids and tannins with valuable uses and used life-cycle assessment (LCA) to assess the environmental performance of the processes (Yadav et al., 2021; Kwan et al., 2022; Dou et al., 15).

Birch bark, a significant byproduct of biorefineries, and potential source for a variety of bioactive compounds has primarily been used for energy production. It is also a valuable resource for producing bio compounds, such as lignin, suberin, betulin, and other triterpenoids (Zhao et al., 2020) which are potential materials for hydrophobic coatings for packaging and textiles. Suberin is the most abundant of all these compounds, varying from 45 w/w% of the solid content in birch outer bark (Kumar et al., 2022). It cannot be isolated intact but can be depolymerized using, for example, alkaline hydrolysis, methanolysis or ionic liquid extraction (Li et al., 2016). Suberin is a hydrophobic biopolymer, and its exploitation in industrial processes is discussed in Pinto et al. (2009). It can potentially be used in hydrophobic coatings (Kumar et al., 2022), adhesives (Makars et al., 2022), foams (Cho et al., 2022; Ivdre et al., 2023) and biopolymers (Rizikovs et al., 2014, 2021). The properties of suberin-based polymers as a surface modifier of cellulosic substrates have been studied (Li et al., 2015a, 2015b, 2016). While the betulin content in silver birch outer bark is 24.5 w/w% (Demets et al., 2022), and it can be produced jointly with suberin processing. Betulin is the most popular of the known triterpenoids of the lupan series. It has valuable pharmacological properties including antibacterial, antitumor, antiviral, hypolipidemic and hydrophobic effect (Demets et al., 2022). Betulin-based polymers are environmentally friendly alternative for water-repellent textiles (Huang et al., 2019).

In the production of chemicals, however, bio-based precursors sourced from renewable biomass may not always offer a more environmentally sustainable substitute to synthetic chemicals. Transparent and credible evaluations of the environmental impacts associated with production are required before asserting any environmental benefits (Ögmundarson et al., 2020). LCA is a decision-making tool that assesses the environmental impacts and helps to identify the hotspots of any product or process during its life cycle (ISO, 2020a). Kumaniaev et al. (2020) conducted a LCA study of phenolic compounds from birch bark using organic solvents (MeOH, ethanol, and dioxane) and considered suberin as a co-product along with lignin and biofuel. They found that the main impacts on the process were due to using electricity as a source for heat. Kwan et al. (2022) reviewed mass balance studies of the production of bark-based chemicals. They discussed some properties of suberin and betulin, but the focus of the study was tannin production from bark.

To our knowledge, very few studies were found related to LCA of suberin and betulin production and none of the previously published studies have assessed the environmental impacts related to suberin and betulin production from birch bark using extraction and alkaline hydrolysis process. Although previously published LCA studies have included suberin as a side product from biorefineries, none has considered it as the main product. The present study assessed the environmental impacts of suberin and betulin production. A prototypical perspective of a biorefinery was modelled using different system boundaries (system expansion) and allocations. These variations were considered across three proposed scenarios (baseline S1, integrated scenario S2, and internal loop scenario S3) to identify the most environmentally friendly process among the scenarios. This analysis helps to identify environmental hotspots within the process.

2. Methodology

The present study follows international standardization organization ISO 14040 (ISO, 2020b) and 14044 (ISO, 2020a) guidelines for conducting LCA. The LCA involves the following steps: (a) goal and scope of the study, (b) data collection under life cycle inventory analysis, (c) environmental impact assessment, and (d) interpretation of the results.

2.1. Goal and scope definition

The goal of the present study is to identify the environmental impacts of the extraction and alkaline hydrolysis of birch bark for suberin and betulin production using LCA methodology. Different scenarios were assessed for the allocation of impacts between intermediate products and mass flows within the cascade process, final products, and the hydrolysis residue with contribution and sensitivity analysis.

2.1.1. Functional unit

Given that suberin and betulin have been found to have a number of potential functions, environmental impacts are calculated relative to a functional unit (FU) of kilogram of solid contents, even if the presented process does not include complete drying of the suberin and betulin. FU is used 2.06 kg of polymer production that includes 1 kg of suberin and 1.06 kg of betulin production from the same process.

2.1.2. System boundaries

The cradle-to-gate approach was adopted for all inputs (birch bark, energy, chemicals, and water) and outputs (suberin, betulin, and bark residues). Three scenarios were proposed (S1: baseline, S2: integrated, and S3: internal loop) that consider different allocations between the products and mass flows and operating environments for the production of suberin and betulin.

(a) Baseline scenario S1: The experiment process was considered as the baseline scenario. The procurement of the raw material, harvesting of the birch pulpwood, transportation to the biorefinery, industrial debarking, and the following extraction and hydrolysis processes were all included in the product system. In S1, mass allocation was used between the betulin I and extracted bark, and extracted bark residues and hydrolysis bark residues were considered as one of the process outputs that would be sold for commercial use in a district heat plant for incineration (Fig. 1).

(b) Integrated scenario S2: In scenario S2, as per Zoppi et al. (2023), it was assumed that the mill would be established in the proximity of a biorefinery and that the production of betulin and suberin from the birch bark flow was integrated with it. In S2, therefore, no environmental impacts load from bark and credits from bark residues were considered, and it was assumed that bark is an underused co-product of a biorefinery and does not release any emissions as a feedstock (Carlqvist et al., 2022). The energy needed for extraction and hydrolysis process was used from Finnish mix electricity grid. For the allocation of impacts between suberin and betulin fractions, bark extraction residue was considered as a waste flow reaching the end-of-waste state at hydrolysis. According to the cut-off method, all the impacts from drying the bark and extraction are allocated to betulin I (Fig. 1). Allocation between betulin II and suberin is based on their solid content yields from hydrolysis (mass allocation). It was assumed that hydrolysis residue bark can be used for energy production in the biorefinery due to calorific value and potential presence of ethanol in the bark residue. However, the study did not consider the changes in the calorific values of residue bark.

(c) Internal loop scenario S3: S3 is similar to S2 but does not account for any impacts from the mill's energy supply to the grid. Instead, the mill's internal energy cycle providing renewable heat and electricity needed for the process was considered, as per Metsä Group (2023). Therefore, the procurement of raw material and debarking are included in a separate process that provides energy from the biorefinery. However, the bark is assumed to be used in the process and then returned to the biorefinery, similar to S2.

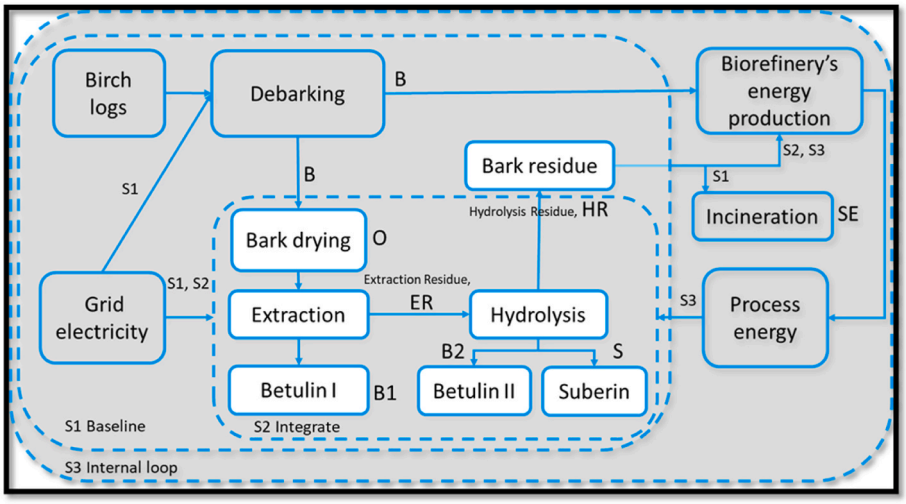

Fig. 1. System boundaries of baseline (S1), integrated (S2), and internal loop (S3) scenarios. White boxes indicate the extraction and alkaline hydrolysis process. The biorefinery's energy supply to the grid is not shown. Solid contents of bark (B), outer bark (O), betulin I (B1), betulin II (B2), suberin (S), extraction residue (ER), hydrolysis residue (HR), and system expansion (SE).

Fig. 1 shows a schematic diagram of material and energy flows within the process and system boundaries for the baseline (S1), integrated (S2), and internal loop (S3) scenarios. The recycling rate of ethanol was considered to be 98% for all scenarios.

2.2. Life-cycle inventory

Primary data were collected from the laboratory experiments in the natural resources institute Finland (Luke) as shows in Fig. 2 and secondary data were taken from the ecoinvent database 3.9.1 and from the scientific literatures. Table 1 presents inventories for 1 kg of suberin with betulin (1.06 kg) as a co-product. The background data to produce the chemicals sodium hydroxide, ethanol, sulfuric acid, and Finnish mix electricity were taken from the ecoinvent database 3.9.1 (Table S1). Energy consumption in each process phase was inventoried using specific heat capacities of materials and measured energy from previous comparable extraction experiments conducted in the natural resources institute Finland and literature sources (Statistics Finland, 2024). The Swedish background processes from ecoinvent database for wood extraction were assumed and used to represent Nordic practices in general, and energy profiles were chosen from those present in Finland. For other background processes, European geography was used for all scenarios. Detailed inventories are reported in Table S2, in supplementary material.

2.2.1. Alkaline hydrolysis of the extracted birch bark

Birch bark was collected from a plywood mill in Finland. The bark was dried at 60 °C for two days, and the particle size was decreased with a Weima WL2 (WEIMA Maschinenbau GmbH, Ilsfeld, Germany) shredder with a sieve with 10 mm diameter holes. The shredded bark was then fractionated with a Sweco round separator (SWECO, Florence, KY, USA) equipped with 1.8 mm and 0.4 mm screens. The bark fraction with particle size larger than 1.8 mm was collected and further crushed by the same shredder equipped with a smaller (6 mm) sieve. The obtained material (industrial bark) was further fractionated with the Sweco round separator, and the particle size under 1.8 mm was collected, stored, and used in the extraction and subsequent hydrolysis. The industrial bark sample was estimated to contain 50% outer bark.

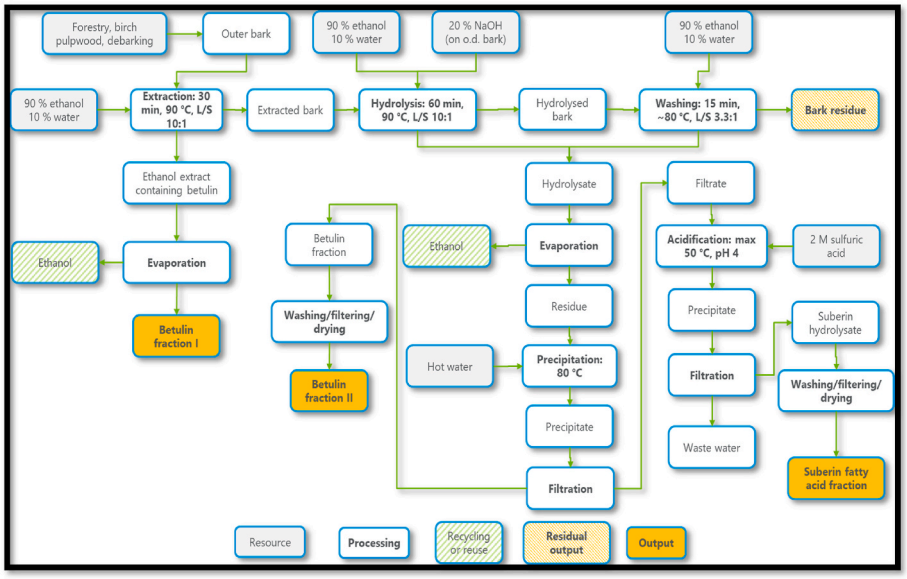

Fig. 2. Diagram of laboratory process used for suberin and betulin production from outer birch bark. L/S = liquid solids ratio.

Table 1
Inputs and outputs per 1 kg of suberin production.

Materials	Unit	Amount	Notes
Inputs, experimental laboratory data			
Pulpwood	m^3	8.94×10^{-2}	Estimated.
Industrial birch bark	kg (DM)	6.17	Sample batch size 1.25×10^{-1} (0.125) kg
Extracted bark	kg (DM)	5.15	After ethanol extraction.
Ethanol	L	85.17	
Water	L	105.13	
Energy			
Debarking	kWh	0.24	
Bark drying	kWh	5.82	
Extraction: 30 min, 90 °C	kWh	3.95	Based on the specific heat capacity of water.
Hydrolysis: 60 min, 90 °C	kWh	4.74	
Washing 15 min, precipitation, both at 80 °C	kWh	4.80	
Suberin fatty acid (acidification, washing, filtering, and drying)	kWh	2.24	
Energy for recycling of ethanol	kWh/kg	0.586	Pfeffer et al. (2007). Based on net energy demand of rectification preheating (2.1 MW), rectification reboiler (14.7 MW) and rectification condenser (12.4 MW), 60,000 tn production over 8000 h annual operation time.
Chemicals			
NaOH	kg	1.03	Experimental laboratory data.
H_2SO_4 (2 M)	litre	3.62	
Ethanol recycling (maximum)	%	98	Yadav et al. (2021)
Outputs			
Bark residues	kg	4.11	Experimental laboratory data.
Betulin I (B1)	kg	1.02	
Betulin fraction II (B2)	kg	0.04	
Suberin fatty acid or Suberin	kg	1.00	
Yield of suberin	%	16.2	Relative to the solid content of bark.
Yield of betulin	%	17.2	
Bark calorific value	MJ/kg	7.50	Statistics Finland (2024).

Additionally, outer bark was manually removed from freshly cut stems (diameter of the trees was 200–300 mm) grown in Punkaharju, Finland, and air dried at room temperature. The outer bark was then ground using a Pulverisette cutting mill (Fritsch, Idar-Oberstein, Germany) with a sieve with 4 mm × 4 mm square openings. The ground outer bark was then freeze-dried and stored in an airtight plastic bag in the dark at room temperature.

A sample of 150 g oven-dry bark was charged into a 2-L Büchi Glas Uster stirred autoclave (Büchi AG, Uster, Switzerland). A volume of 1200 ml (solvent to bark ratio 8 ml/g) of ethanol/water (9:1 v/v) mixture was added, and the reactor was closed. The temperature was raised from room temperature to 90 °C within 15 min, and the extraction was carried out for 30 min. The reactor was cooled, the extraction liquid was drained from the reactor via a cooling unit, and the volume was measured. The ethanol/water mixture was vacuum-evaporated by a Hei-VAP Precision vacuum rotary evaporator (Heidolph Instruments GmbH & C. KG, Schwabach, Germany). The residue was further dried in a Heraeus vacutherm vacuum oven (Thermo Scientific, Thermo Electron LED GmbH, Langenselbold, Germany) at 40 °C for 20 min and subsequently weighed. The residue containing mainly betulin was approximately 16.5% of the bark fraction (Table S3). For comparison, when using manually separated outer bark and a volume of 1500 ml (solvent to bark ratio 10 ml/g) of ethanol/water mixture, the yield (betulin I) of the residue was 20.9%.

The alkaline alcoholic hydrolysis of the extracted bark and the following betulin separation and suberin fatty acid precipitation was carried out using a slightly modified procedure (Korpinen et al., 2019). A volume of 1002 ml (solvent to bark ratio 8 ml/g) of ethanol/water (9:1 v/v) mixture including the solvent and water in the extracted bark was used in the hydrolysis (Fig. 2). Sodium hydroxide charge (20%) was calculated on completely dry extracted bark, and 25 g sodium hydroxide was dissolved in the water before the remaining ethanol of the solvent mixture was added. The hydrolysis was enhanced by using a stirred autoclave, and the temperature was increased to 90 °C. The hydrolysis time was 60 min, after which the hydrolysate was drained by using a cooler. The residual bark was washed with one third of the charged ethanol/water mixture (334 ml) at 80 °C and drained from the reactor using a cooler. The hydrolysate and the washing solvent were combined, and the solvent was evaporated by a Hei-VAP Precision vacuum rotary evaporator (Fig. 2). Boiling water was added to the evaporated residue, and betulin fraction was precipitated out. The precipitate was filtered out, dried, and weighed. The second betulin fraction accounted for 0.7% of the original charged 150 g industrial bark. In comparing the use of pure outer bark and the 10 mg/l solvent-to-bark ratio, the second betulin fraction II was 15.1% of the charged 150 g bark.

Finally, the filtrate containing suberin fatty acid soaps was acidified with 2 M sulfuric acid to a pH of approximately 4, and the suberin fatty acids were precipitated out. The precipitate was filtered and washed with water several times and collected in a filter paper. The obtained suberin fatty acid cake was weighed, and the dry solids content was determined. The amount of the suberin fatty acid fraction obtained from the industrial bark was 16.2%. As a reference, manually separated outer bark yielded 26.1% of the suberin fatty acid fraction.

2.2.2. *Allocation and assumptions*

According to ISO 14044, allocation should be avoided. However, if allocation cannot be avoided, it should reflect the physical relationships between the products and co-products throughout the product system. In scenario S1, system expansion was used for the hydrolysis residue by subtracting the avoided impacts of energy from the use of bark residue. It was assumed that a birch log's bark volume (13.80%) contains 25% outer bark. The basic densities of bark and wood material were assumed to be equal (500 kg/m^3); thus, allocations were based on these volumetric shares. Following the notation in Fig. 1, allocation for the betulin I and extraction bark residue was based on mass shares (betulin I = 16.5%, extraction bark residue = 83.5%). Credit was considered according to the caloric value in a district heat unit using woody materials. In S2 and S3, impacts from the extraction phase were not allocated to hydrolysis, and bark flow was not included. In all scenarios, allocation between betulin II and suberin after precipitation was based on solid contents (betulin II = 3.90%, suberin = 96.10%). In scenario S1, credits from using hydrolysed bark residue for district heating were subtracted from the impacts. In S2 and S3, bark flow had no direct maintained impacts; thus, betulin I had all (100%) the impacts from drying bark to extraction, betulin II and suberin allocations were done as per mass allocation. In scenarios S1 and S2, Finnish mix electricity was used; S3 used energy from the biorefinery. Primary inventories for the extraction and hydrolysis did not cover the energy consumption of the process. Following Piccinno et al. (2016), energy consumptions were based on the specific heat capacities of wood, water, and ethanol but without any corrections for possible heat losses. While the amount of wood material relative to ethanol was low and wood is not known as environmentally hazardous or energy-intensive, ethanol can be considered, *ex ante*, as a key source of environmental impacts. In each stage, it is necessary to evaporate ethanol from the extract and hydrolysate, and ethanol can be recycled into the process (Yadav et al., 2021). The efficiency of the recycling process was unknown, it was a subject for the sensitivity analysis. In all scenarios, the maximum recycling rate of the ethanol (98%) was assumed (Yadav et al., 2021), with energy consumption of 0.586 kWh/kg ethanol (Pfeffer et al., 2007).

2.3. Life-cycle impact assessment

Life-cycle impacts were assessed using the ReCiPe (2016) Midpoint (H) method (Huijbregts et al., 2017) as implemented in SimaPro v.9.5 (PRé Sustainability Database & Support team, 2023). The method is well suited for a global perspective. Most background data were adapted from the ecoinvent 3.9.1 database using the cut-off model. A full set of 18 impact categories are presented for all scenarios in the supplementary material (Table S4). The impact categories presented here are global warming potential (GWP), ionizing radiation potential (IRP), fine particulate matter formation potential (FPMP), and terrestrial acidification potential (TAP), which account for 80.30% of the imputed environmental costs according to the Environmental Prices method (CE Delft, 2018; PRé Sustainability Database & Support team, 2023) (Fig. S1). As land use potential (LUP) is usually critical for renewable raw materials and water consumption potential (WCP) for chemical processes, they were also analysed.

2.4. Sensitivity analysis

The LCA results are expected to be most sensitive to the ethanol used in the process. The default value of the ethanol recycling rate was used in all scenarios (98%). The sensitivity analysis was performed in all scenarios by changing the ethanol recycling rate to 0% (no recycling), 50% and 90%. In the laboratory experiment process in Luke, ethanol recycling was not possible, and so an ethanol recycling rate of 0% was used for the sensitivity analysis.

3. Results and discussion

Table 2 presents the environmental impact of various impact categories for the scenarios S1, S2 and S3. It is important to emphasize that the results reported in this study are dependent on the methodological choices (ReCiPe, 2016H), allocation method, assumptions, FU, and system boundaries in each scenario. The environmental impacts of the baseline scenario per FU were found for GWP (15.47 kg CO_2 eq.), IRP (18.61 kBq Co-60 eq.), FPMP (2.57×10^{-2} kg $PM^{2.5}$ eq.), TAP (7.35×10^{-2} kg SO_2 eq.), LUP (3.49 m^2) and WCP (0.63 m^3). Table 2 shows that GWP (5%), IRP (1.23%), FPMP (23%), TAP (8.22%) and WCP (1.90%) were higher in S2 than in S1. The differences in environmental impact between S1 and S2 resulted from the inclusion of the bark cycle and the credits associated with incinerating hydrolysed bark residue in S1, which was not considered in S2. Additionally, LUP was 59% lower in S2 than in S1, because direct land use impacts from bark were not considered in S2. Table 2 shows that in S1 the suberin impacts were higher than in S2 and S3. The variation in impacts due to allocation, where the majority of impacts (83.50%) were allocated to the extraction residue in S1 and the rest to betulin I, and the allocation to betulin II from hydrolysis output was 3.90%. The difference in impact between suberin and betulin varied significantly between S1 and S2. In S2, extraction and hydrolysis were considered separate processes. Scenarios S2 and S3 used the same allocation rules, but in S3 heat energy from the biorefinery was used instead of the Finnish mix electricity (S1 and S2). Consequently, the environmental impacts of S3 were significantly lower than those of S2, except for LUP, where the raw materials from the biorefinery had higher impact.

The GWP for betulin was 1.66 kg CO_2 eq. kg^{-1} in S1, 7.23 kg CO_2 eq. kg^{-1} in S2 and 1.40 kg CO_2 eq. kg^{-1} in S3. In S1, the production of betulin and suberin was treated as an interconnected process, where allocation after extraction was determined by mass flows involving bark, resulting in low impacts on betulin I and higher for hydrolysis outputs. In S2 and S3, the hydrolysis residue was redirected back to the biorefinery for energy production, and therefore the flow of bark was not considered in the allocations. Additionally, as extraction and hydrolysis were treated as separate processes, all the impacts related to bark drying and extraction were attributed to betulin, while the hydrolysis outputs were allocated based on the solid contents of betulin and suberin. In S3, heat was directly supplied from the biorefinery, leading to lower emissions from energy use compared to S2. The results for the 18 impact categories for all scenarios are presented in the supplementary material (Table S4).

If the product system were considered inseparable in outputs, the allocation can be based on the dry mass of betulin and suberin. This option was not included in the scenarios. However, the result can be easily derived from the total columns of Table 2 by dividing the effects in the ratio 1:1.06.

The functions of suberin and betulin vary such that any results found from the literature of comparable materials do not necessarily meet the criteria set for comparative analysis in ISO 14044 or 14067. As hydrophobic materials, however, suberin and betulin have a high potential as hydrophobic coating material solutions on the textile or packaging industry (Jumppanen, 2013). For example, polytetrafluoroethylene, a variant of which is widely used for the hydrophobic coating of textiles, has a high GWP:, in the ecoinvent database, a fluoroolefin used for polytetrafluoroethylene has GWP above 100 kg CO_2 eq. kg^{-1}. According to W.L. Gore and Associates (2013), a hydrophobic GORE-TEX® coating alone in a jacket had a GWP of 13.11 kg CO_2 eq., a figure that is already above the GWP per FU found in this study for suberin in S2 and S3 or for betulin in all scenarios. The European Environment Information and Observation Network (Eionet) has also identified knowledge gaps in the assessment of the whole life cycle of fluorinated polymers (Eionet, 2021). A previous result from another hydrophobic fossil-based polyurethane (7.8 kg CO_2 eq. kg^{-1}) (Bachmann et al., 2021) was higher than in scenario S3.

Table 2
Environmental impacts of per FU 2.06 kg biopolymer (total S1, S2 and S3) that includes 1 kg suberin and 1.06 kg betulin.

Impact Category	Unit	Baseline Scenario (S1)			Integrated Scenario (S2)			Internal loop Scenario (S3)		
		Suberin	Betulin I & II	Total S1	Suberin	Betulin I & II	Total S2	Suberin	Betulin I & II	Total S3
GWP	kg CO_2 eq.	13.72	1.75	**15.5**	8.57	7.66	**16.2**	2.48	1.48	**3.96**
IRP	kBq Co-60 eq.	16.5	2.12	**18.6**	9.52	9.32	**18.8**	0.452	0.112	**0.564**
FPMP	kg $PM^{2.5}$ eq.	2.38×10^{-2}	1.95×10^{-3}	**2.57×10^{-2}**	2.23×10^{-2}	9.34×10^{-3}	**3.16×10^{-2}**	1.53×10^{-2}	2.19×10^{-3}	**1.75×10^{-2}**
TAP	kg SO_2 eq.	6.95×10^{-2}	3.96×10^{-3}	**7.35×10^{-2}**	6.12×10^{-2}	1.78×10^{-2}	**7.89×10^{-2}**	4.91×10^{-2}	5.52×10^{-3}	**5.47×10^{-2}**
LUP	m^2a crop eq.	2.62	0.872	**3.49**	0.722	0.716	**1.44**	2.54	2.56	**5.09**
WCP	m^3 eq.	0.578	5.23×10^{-2}	**0.630**	0.422	0.220	**0.642**	0.231	2.61×10^{-2}	**0.257**

Note: Global warming potential (GWP), ionizing radiation potential (IRP), fine particulate matter formation potential (FPMP), terrestrial acidification potential (TAP), land use potential (LUP) and water consumption potential (WCP).

3.1. Contribution analysis

The contributions of different components and chemical used in the process (energy, virgin ethanol, bark, sodium hydroxide, sulfuric acid, and water) and system expansion are presented in Fig. 3. Finnish mix electricity was used in S1 and S2 and heat from a biorefinery in S3. Electricity contains all the energy needed for ethanol recycling, extraction, and hydrolysis as well as the processing of the outputs. The recycling rate of ethanol was the default 98%. The bark containing the bark only in S1 and the energy required for drying the bark in all scenarios. System expansion for the hydrolysed bark residue was only used in S1. Fig. 3a presents the contribution to GWP of different components in various scenarios. In S1 and S2, electricity was the primary contributor to GWP (12.17 kg CO_2 eq. total), while water had the least impact (0.03 kg CO_2 eq. total). However, in S3 the need of virgin ethanol is the main reason to GWP (1.66 kg CO_2 eq. total) (Fig. 3a). While, replacing Finnish mix electricity with the biorefinery's heat significantly reduced the GWP in S3. The GWP of other components used in the process, sulfuric acid,

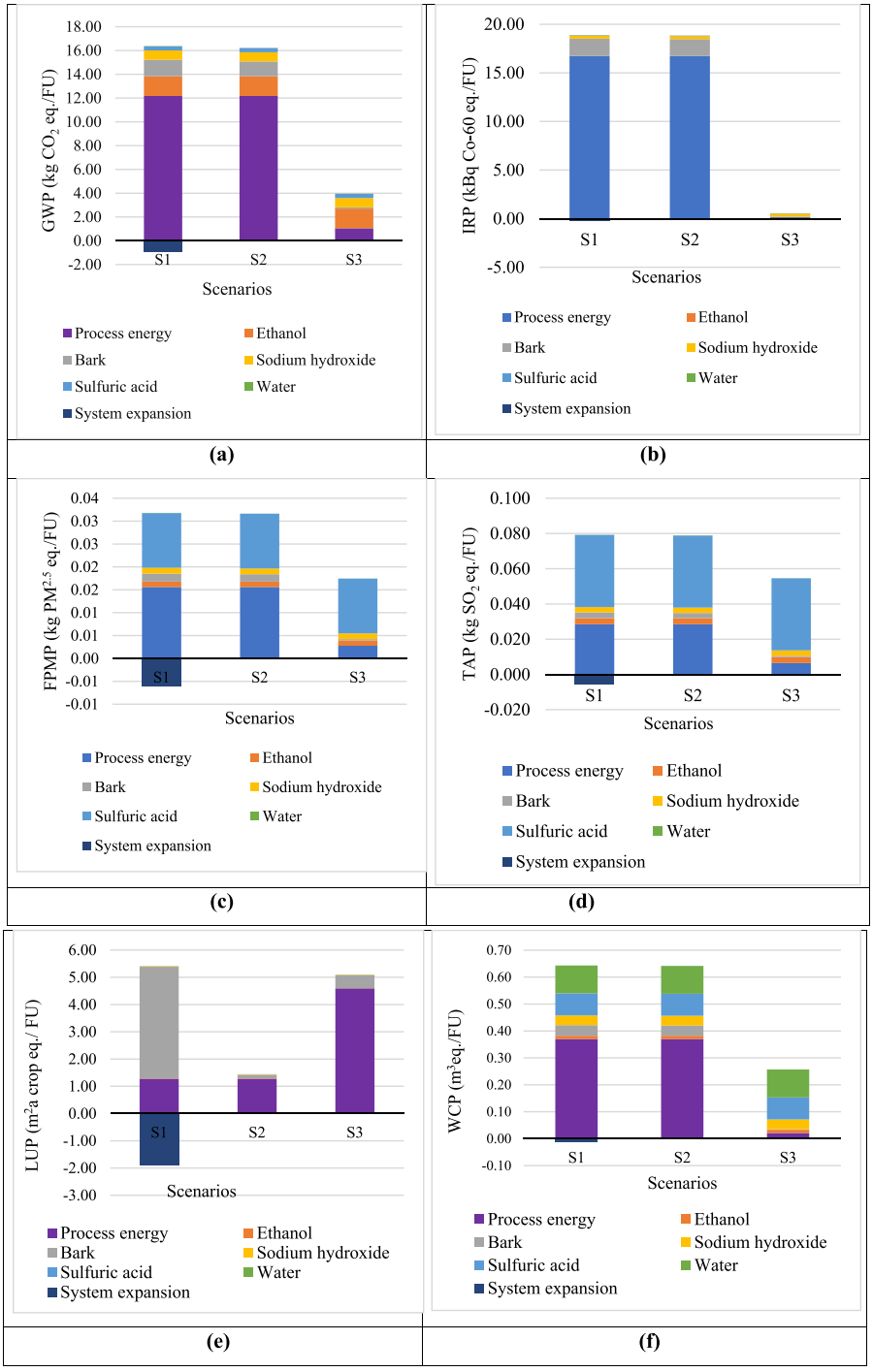

Fig. 3. Contribution analysis of components used per FU (1 kg suberin and 1.06 kg betulin) and their impacts on different environmental categories: (a) global warming potential (GWP), (b) ionizing radiation potential (IRP), (c) fine particulate matter formation potential (FPMP), (d) terrestrial acidification potential (TAP), (e) land use potential (LUP) and (f) water consumption potential (WCP). **Note:** Process energy refers to Finnish grid electricity in S1 and S2 and heat from the biorefinery in S3. Bark includes bark drying only in S2 and S3.

sodium hydroxide, and water, remained constant in all scenarios.

The process electricity was the main contributor to IRP (Fig. 3b) in S1 and S2. The contribution was the same (16.74 kBq Co-60 eq. total) in both scenarios and was due to the high share of nuclear energy (28.6%) in the Finnish grid. Moreover, bark's IRP was related to the energy used for drying the bark. For S3, IRP remained lower (0.17 kBq Co-60 eq. total) due to the use of wood -based heat. The use of energy and sulfuric acid in the process was the main contributor to FPMP (Fig. 3c). In the case of electricity, the high FPMP mirrored the relatively high shares of hard coal and peat in the Finnish grid electricity. The major impact on TAP (Fig. 3d) was due to the use of sulfuric acid in the process. In S1 and S2, the other contributors to TAP originated from the use of electricity and ethanol. In S3, process energy had a lower impact on TAP compared to S1 and S2 due to differences in energy sources. For S1, the LUP impact (Fig. 3e) was related directly to bark use and indirectly to the use of woody materials for electricity. In S3, the high LUP impact was related to black liquor, containing wood-based lignin and hemicelluloses, used for heat production at the biorefinery. WCP helps to assess the water consumption associated with a production process. The direct use of water in the process was low, but as shown in Fig. 3f indirect water consumption was channelled through Finnish electricity, contributing 0.40 m^3 of the total 0.63 m^3 in S1. Even if hydropower has a large share of local production, the main contribution to WPC originated from hard coal combined heat and power production (0.125 m^3) and energy imports from Sweden and Russia. In terms of raw materials, the sulfuric acid used in suberin production had impacts on the FPMP (46.50%), TAP (55.70%), and WCP (13%).

3.2. Sensitivity analysis

The contribution analysis showed that the process energy played a critical role in the environmental impacts. Given that the amount of process energy did not change between the scenarios, the 98% ethanol recycling rate accounted for 72.2% of the environmental impacts related to energy use in all scenarios. Therefore, aside from the source of the process energy, the way ethanol is used and processed appeared to be significant for the environmental impacts. The sensitivity analysis was performed using ethanol recycling rates of 0%, 50% and 90% relative to the default value 98%, and the results are presented in Figs. 4–6. It was assumed that recycling energy is not needed in 0% (=no recycling. In our model, ethanol recycling uses Finnish mix electricity (0.586 kWh kg^{-1} ethanol), whereas the production of virgin ethanol was inventoried according to the European ethanol markets from ecoinvent, in which environmental impacts are mainly related to ethylene production and hydration to ethanol.

As shown in Fig. 4a, there was an inverse relationship between the ethanol recycling rate and the GWP, FPMP, TAP and WCP impacts. At the 50% recycling rate of ethanol, the GWP of scenario S1 and S2 increased three times (51.9 kg CO_2 eq. FU^{-1}) and (51.6 kg CO_2 eq. FU^{-1}) from their default values, while GWP of S3 increased 11 times (43.4 kg CO_2 eq. FU^{-1}). The changes for FPMP (Fig. 5a) and TAP (Fig. 5b) were more moderate but followed the same patterns as the GWP. All these changes were dominated by a higher intensity of virgin ethanol use. The changes in IRP and LUP were opposite, due to the transition from Finnish energy to the European average in virgin ethanol. The composition of the Finnish electricity mix is nuclear (27.4%), biomass (25%), hydro (19.2%), wind (9.7%), solar (0.3%) and imported electricity from Sweden and Russia (18.4%) (Energy Statistics Finland, 2022; Yadav et al., 2024). In S1 and S2, IRP decreased together with the decreasing recycling rate due to the high share of nuclear energy in the Finnish electricity mix, whereas in S3 renewable energy was practically free of IRP, so that IRP increased with a higher share of virgin ethanol but remained at a significantly lower level than in S1 and S2. The increased LUP (Fig. 6a) in S3 was mainly related to the wood-based renewable energy. The recycling rate of ethanol 50%, WCP (Fig. 6b) increased by 24% from the baseline in S1 and S2, but by 109% in S3 due to the lower baseline WCP with equal increases in ethanol use in each scenario. The results of the sensitivity analysis for 18 impact categories for all scenarios are presented in the supplementary material (Table S5). According to the results, the effects of using ethanol without recycling had higher environmental impact. It was assumed that the ethanol-water solution used in the process was recycled or disposed of without considering the environmental impacts of treating ethanol wastewater. However, the calculations did not consider the possibility of treating used ethanol as a co-product of the process. Contaminated ethanol can be purified and used for other purposes. This scenario could have had a significant influence on the environmental impacts of using recycled and/or virgin ethanol in the process.

The sensitivity analysis regarding yield of suberin and betulin also performed in this study. The yields of betulin and suberin from manually separated pure outer bark are reported in section 2.2.1. In extraction stage, the yield of betulin I moderately increased from 16.6% to 20.9%. In hydrolysis, the yields increased from 0.7% to 15.5% for betulin II, and from 16.2% to 26.1% for suberin. Since the yields of suberin and betulin from the manually separated bark increase in different proportions in both extraction and hydrolysis, and the solvent/solids ratio is slightly higher in extraction, an exact comparison of the process yields is not possible. Using pure outer bark, the production of 1 kg suberin from manually separated bark yielded 0.8 kg betulin I and 0.578 kg betulin II. Table 3 shows the impact levels in relation to the baseline scenario presented in Table 2. This analysis assumes using the same quantities of suberin and betulin as the FU, for betulin I and II the process is scaled down to produce exactly 1.06 kg. The total impacts are roughly halved from industrially separated yields but distributed differently between the betulin and suberin fractions. In a cascade process, the results depend on the chosen allocation method and scenarios. In S1, increased

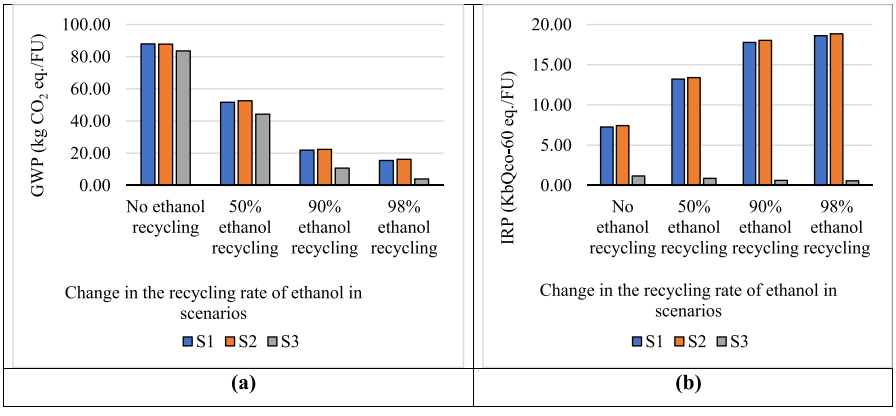

Fig. 4. Changes in (a) global warming potential (GWP) and (b) ionizing radiation potential (IRP) due to change in recycling rate of ethanol in all scenarios per FU.

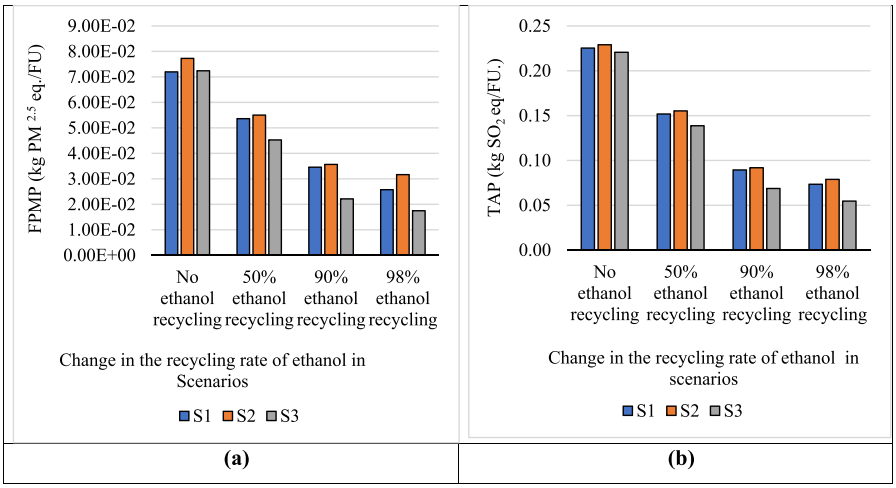

Fig. 5. Changes in (a) fine particulate matter formation potential (FPMP) and (b) terrestrial acidification potential (TAP) due to change in recycling rate of ethanol in all scenarios per FU.

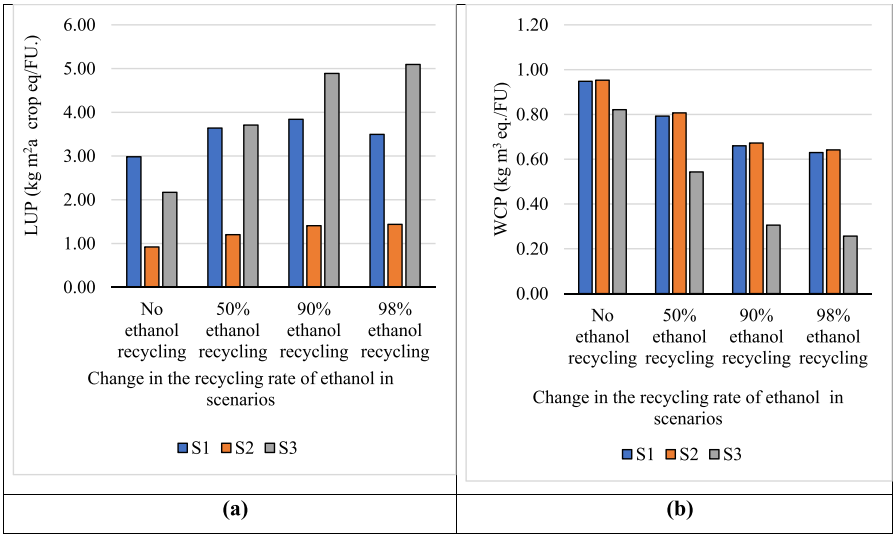

Fig. 6. Changes in (a) land use potential (LUP) and (b) water consumption potential (WCP) due to change in recycling rate of ethanol in scenarios per FU.

Table 3
Impact levels for processing manually separated outer bark relative to baseline scenario S1 in Table 2.

Impact Category	Unit	Baseline Scenario (S1)		
		Suberin (1 kg)	Betulin I & II, (1.06 kg)	Total S1
GWP	kg CO_2 eq.	44.5 %	64.9 %	46.8 %
IRP	kBq Co-60 eq.	43.4 %	64.7 %	45.8 %
FPMP	kg $PM^{2.5}$ eq.	57.3 %	66.4 %	58.0 %
TAP	kg SO_2 eq.	55.7 %	65.3 %	56.2 %
LUP	m^2a crop eq.	44.5 %	64.9 %	46.8 %
WCP	m^3 eq.	43.4 %	64.7 %	45.8 %

Note: Global warming potential (GWP), ionizing radiation potential (IRP), fine particulate matter formation potential (FPMP), terrestrial acidification potential (TAP), land use potential (LUP) and water consumption potential (WCP).

betulin I yield allocates more impacts to betulin I, so that the extraction residue for a hydrolysis input has lower maintained impacts. As betulin II yield higher than suberin yield, because some hydrolysis impacts were shifted from suberin to betulin II. As the total yields are higher for both products, the impact levels decrease for both.

4. Conclusion

Suberin and betulin are promising hydrophobic materials for various applications, and they can be produced in a cascade process using side streams from biorefineries or plywood mills. In this article, a cascade model of bark utilization was investigated, where betulin I was extracted from the outer bark of birch and suberin and betulin II were hydrolysed from the extracted bark. Bark residue from hydrolysis was used for energy recovery. Environmental impact was assessed for three scenarios [(baseline (S1), integrate (S2) and internal loop (S3)]. According to our results, the environmental impact of suberin and betulin production depends on the type of allocation used in the process, the source of energy and ethanol recycling rate in process. Suberin and betulin would not have significant environmental impacts if an efficient industrial solution can be found for the recycling or reuse of ethanol. The use of ethanol in the process was identified as a hotspot, with 72.70% of the process's energy utilized for recycling 98% of the ethanol. However, the sensitivity analysis was conducted based on ethanol recycling rate (0%, 50% and 90%). It was found increasing the ethanol recycling rate results in an increase the demand of energy inside the suberin and betulin

production process. A low recycling rate of ethanol decreased the Finnish mix energy demand but increased the overall impact on all the impact categories due to high demand of virgin ethanol in the processes. The yields of the process were also found critical, using the manually separated pure bark both betulin and suberin the yields increased such that the total impacts of the process were generally halved. Increased yields of the cascade process had considerable impact through consecutive impacts allocations between process flows and outputs. This complicates interpretation of results, but apart from (ISO, 2020a) there is no clear rule for the allocations choices other that being conditional to the goal and scope of the study. The key limitation of the research is the laboratory-level inventory, but from the results it can already be seen that the recycling of ethanol and the efficiency of the process are key factors of environmental impacts. This research into the use of suberin and betulin as hydrophobic materials is still in its infancy, but when solutions are found, suberin and betulin have potential as environmentally friendly functional materials.

CRediT authorship contribution statement

Pooja Yadav: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Risto Korpinen:** Writing – review & editing, Writing – original draft, Data curation. **Tarmo Räty:** Writing – review & editing, Writing – original draft, Validation, Software, Formal analysis, Data curation, Conceptualization. **Pasi Korkalo:** Writing – review & editing. **Kati Räsänen:** Writing – review & editing. **Jenni Tienaho:** Writing – review & editing. **Pekka Saranpää:** Writing – review & editing, Writing – original draft, Funding acquisition, Data curation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Pooja Yadav reports financial support was provided by Natural Resources Institute Finland. Pooja Yadav reports a relationship with Research Council of Finland that includes: funding grants. Project also funded by European Union – NextGenerationEU instrument and by the Academy of Finland under grant number 348870 (PN) and 349052. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2024.143570.

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