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► **To cite this version:**

Dominika Alexa Teigiserova, Lorie Hamelin, Ligia Tiruta-Barna, Aras Ahmadi, Marianne Thomsen. Circular bioeconomy: Life cycle assessment of scaled-up cascading production from orange peel waste under current and future electricity mixes. *Science of the Total Environment*, 2022, 812, <10.1016/j.scitotenv.2021.152574>. <hal-03742897>

HAL Id: hal-03742897

<https://hal.science/hal-03742897v1>

Submitted on 2 Aug 2022

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Circular bioeconomy: Life cycle assessment of scaled-up cascading production from orange peel waste under current and future electricity mixes



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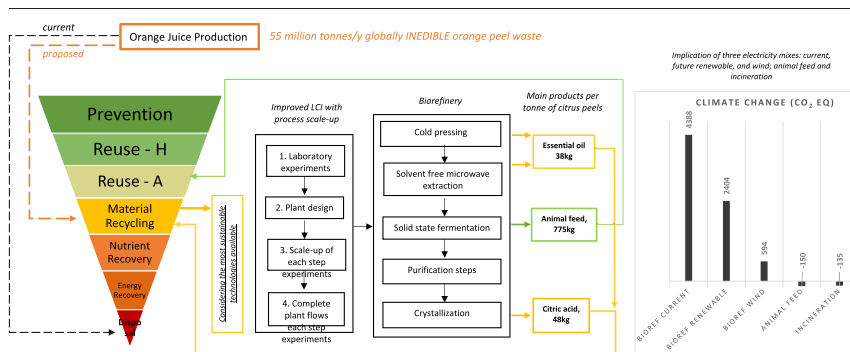
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HIGHLIGHTS

- Tens of million tonnes of orange peel waste is not managed sustainably.
- LCA inventories include scale-up of three plant sizes and three electricity sources.
- Climate change is 1.8 & 7.4 times lower for renewable & wind to current electricity.
- Biorefineries are not always the best option when compared to conventional options.
- Most environmental options stop at limonene and use the rest as dried animal feed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 14 July 2021

Received in revised form 14 December 2021

Accepted 16 December 2021

Available online 24 December 2021

Editor: Deyi Hou

Keywords:

Food waste

LCA

Circular economy

Biorefinery

Citric acid

Limonene

ABSTRACT

Orange peel waste (OPW) is present in large quantities both locally and globally, which makes them feasible input into the circular bioeconomy. However, due to their antimicrobial and anti-nutritional activity, they are problematic biomass, and proper waste management is yet to be determined. This life cycle assessment (LCA) quantifies the environmental performance of biorefinery producing limonene, citric acid, and animal feed from OPW generated from juice factories. Only previously assessed sustainable technologies were considered (cold press, microwave extraction, solid-state fermentation). The life cycle inventories were refined by a scale-up procedure to reflect industrial production at i) 0.5 t, ii) 100 t, and iii) 1000 t of OPW weekly. The data were translated per functional unit of 1 tonne OPW. Three electricity mixes and both attributional (average) and consequential (marginal) inventories were compared. Results showed that the environmental performance, in particular for climate change, was essentially dependent upon the electricity input, with 4388 CO₂ eq for current electricity mix, 2404 CO₂ eq using renewable, and 594 CO₂ eq using electricity from wind. Business-as-usual scenarios for OPW (incineration and animal feed) showed better performance in most scenarios, representing -150 CO₂ eq (animal feed) and -135 CO₂ eq (incineration) in the climate change. Lower impacts are reached due to avoided impacts of fossil fuel use and conventional feed cultivation. Renewable energy had better environmental performance than the current electricity mix, except for eutrophication, due to digestate spreading associated with biogas use, calling for mitigation action. Our results suggest that stopping the

Abbreviations: CP, cold press; EO, essential oil; LCA, life cycle assessment; LCI, life cycle inventory; OP, orange peel; OPW, orange peel waste; SFME, solvent-free microwave extraction; SSF, solid-state fermentation; TRL, technology readiness level; RoW, rest of the world.

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biorefinery processes immediately after the recovery of limonene via solvent-free microwave extraction process, with subsequent use of the dried OPW as animal feed, is the most environmentally performant option. This represents a feasible strategy for the circular bioeconomy and is in line with the updated food waste hierarchy.

1. Introduction

The recent EU ambitions towards sustainable production call for the use of waste as input material, shifting towards the circular economy (EC, 2014, 2020) and bioeconomy (EC, 2018). This also applies to organic waste. As highlighted in the updated food waste hierarchy (Sanchez et al., 2020; Teigiserova et al., 2020), inedible and unavoidable food waste should be prioritized for biorefinery production. Such waste is often generated by the food processing industry, representing homogeneous streams with regard to quantity and quality. One example of untapped potential comes from orange juice production, which generates around 55 million tonnes of inedible orange peel waste (OPW) globally per annum (Mahato et al., 2019). In general, OPW represents 50% of the whole fruit weight (Martín et al., 2018), with some factories having OPW output of hundreds of tonnes per day (juicing factory in Spain, Martín et al., 2018). Despite the EU's ban on landfilling, this waste is still landfilled or dumped into the ocean (Choi et al., 2015), which leads to a variety of environmental consequences, including the release of additional GHG emissions (Negro et al., 2017); while landfill prevention has been shown as achievable (Wang et al., 2020). OPW is also used as animal feed, albeit in limited amounts, due to its low pH (<3), anti-nutritional properties (limonene), and potentially toxic compounds (e.g., agrochemical residues), especially for monogastric animals (Ani et al., 2015). Currently, one of the main management strategies involves the incineration of OPW (Negro et al., 2017).

Other management strategies consider OPW as an input to bioenergy production (Oberoi et al., 2010; Wikandari et al., 2015). OPW can also be used to produce high-value compounds, among others pectin, which has been researched (Wang et al., 2008; Rezzadori et al., 2012; de la Torre et al., 2019) and implemented by the industry (CP Kelco, 2019).

In this study, we explore the extraction of chemicals like limonene and citric acid (CA) from OPW and also the production of animal feed with improved digestibility to raw OPW. Limonene is used as a flavoring and aromatic agent in food and cosmetics as well as a green solvent (Ciriminna et al., 2014). CA is a mild organic acid used in various applications, from cosmetics (safe to use on human skin in low amounts) and pharmaceuticals to food ingredients (around 65% of global CA use) (Singh Dhillon et al., 2011; Berovic and Legisa, 2007). Due to their versatile application, there is an increasing market demand for both CA (Torrado et al., 2011; Berovic and Legisa, 2007) and limonene (Ciriminna et al., 2014).

Using food waste fractions as input for new multi-product biorefineries in the circular economy has been stressed as one of the leading sustainable strategies in the EU (Caldeira et al., 2020; Sanchez et al., 2020) and beyond (Javourez et al., 2021). However, Life cycle assessment (LCA) is needed to assess the environmental performance of such strategies to anticipate the consequences of possible management changes, such as the one proposed herein for OPW. LCA is a widely applied approach to determine the environmental performance of a product, service, or system (Sala et al., 2021).

Previous LCAs concerning OPWs regarded the stream essentially as a waste to be managed. While the LCA of Beccali et al. (2010) focused on juice production (including cultivation packaging and cooling), the study of Negro et al. (2017) assessed ten OPW management scenarios. These include pyrolysis, incineration, anaerobic mono-digestion, anaerobic co-digestion with animal manure and seaweed (4 cases), landfilling, composting, and animal feeding. The study concluded that anaerobic digestion is the preferred option based on a preliminary cost flow analysis. However, OPW represents a potential for the circular bioeconomy, moving up the waste hierarchy to material recycling. Albeit these studies included valuable insights, the present study aims to go beyond the waste management perspective for OPW and consider their use for the sustainable production of chemicals. Yet, this has not been implemented and involves a

set of uncertainties regarding the life cycle inventories of the technologies to be used and their possible industrial performance. In order to supply meaningful insights for future sustainable investment decisions, there is a need to ensure that: i) green chemistry principles (Anastas and Eghbali, 2010) are applied throughout the cascading production, and that ii) realistic inventories integrating process modeling and scale-up are used.

The main goal of this study is to assess, for different production scales, the environmental performance and dependencies of using OPW from the juice industry in a biorefinery setting producing CA, limonene, as well as animal feed under different electricity scenarios. This will allow uncovering possible trade-offs between OPW management strategies that are in line with the updated food waste hierarchy and their environmental performance while highlighting possible performance dependencies upon the process scale and future electricity supply.

2. Methodology

2.1. LCA methodology, goal, and scope

2.1.1. LCA goal and scope

The goal of this LCA study is to quantify the environmental impacts of OPW-based bioeconomy value chains. As typical in waste management LCAs, the activities preceding the generation of the waste are excluded, as unaffected by the use of the waste (Negro et al., 2017). Capital goods (e.g., buildings, machinery, etc.) are included in background processes only. The functional unit (FU) is the management of 1 t of OPW after juicing (wet weight) for a plant producing 100 t of OPW waste per week, 52 weeks y^{-1} (excluding public holidays), modeled according to data representative of the Danish orange juice plant Rynkeby (Rynkeby Foods, 2019). The geographical scope is thus Denmark. The plant operation is envisioned for five days a week, with 16 h of daily running time. The initial OPW moisture content is 75%.

2.1.2. Life cycle inventory and impact assessment method

Inventory data for the foreground processes, i.e., those relating to the material and energy balances for the OPW transformation in a biorefinery, were obtained by combining literature information and process modeling, as further detailed in Section 2.5.1.

Background data (i.e., generic and unspecific to the system studied herein) were retrieved from the Ecoinvent v.3.6 LCI database. We distinctly considered both “average” inventory data (processes representing an average of current markets; these may be unable to react to a demand change and are often referred to as “attributitional processes”) and “marginal” data (referred to as consequential processes within Ecoinvent v3, these can and are foreseen to respond to a change in demand). While the use of average data implies reflecting the average operation (Plevin et al., 2014), the use of marginal data attempts to reflect the consequences of small-scale, long-term decisions (Weidema et al., 2013). Here, the scenarios using average life cycle inventory (LCI) data are termed “attributitional scenarios”, and those using marginal LCI data are termed “consequential scenarios”. This should not be confused with traditional modeling for attributitional LCA, which often uses allocation (e.g., Brandao et al., 2017); here, it only refers to the use of background processes.

Multi-functionality was handled with system expansion (Fig. 1), as recommended in the ISO standard for LCA (International Organization for Standardization, 2006a, 2006b).

In this study, we apply the EF life cycle impact assessment method (Environmental Footprint) as recommended by the European Commission (EC, 2013, 2017). The EF method was adjusted to account for biogenic CO₂. The assessment was facilitated with the Simapro LCA software, version 9.0. We

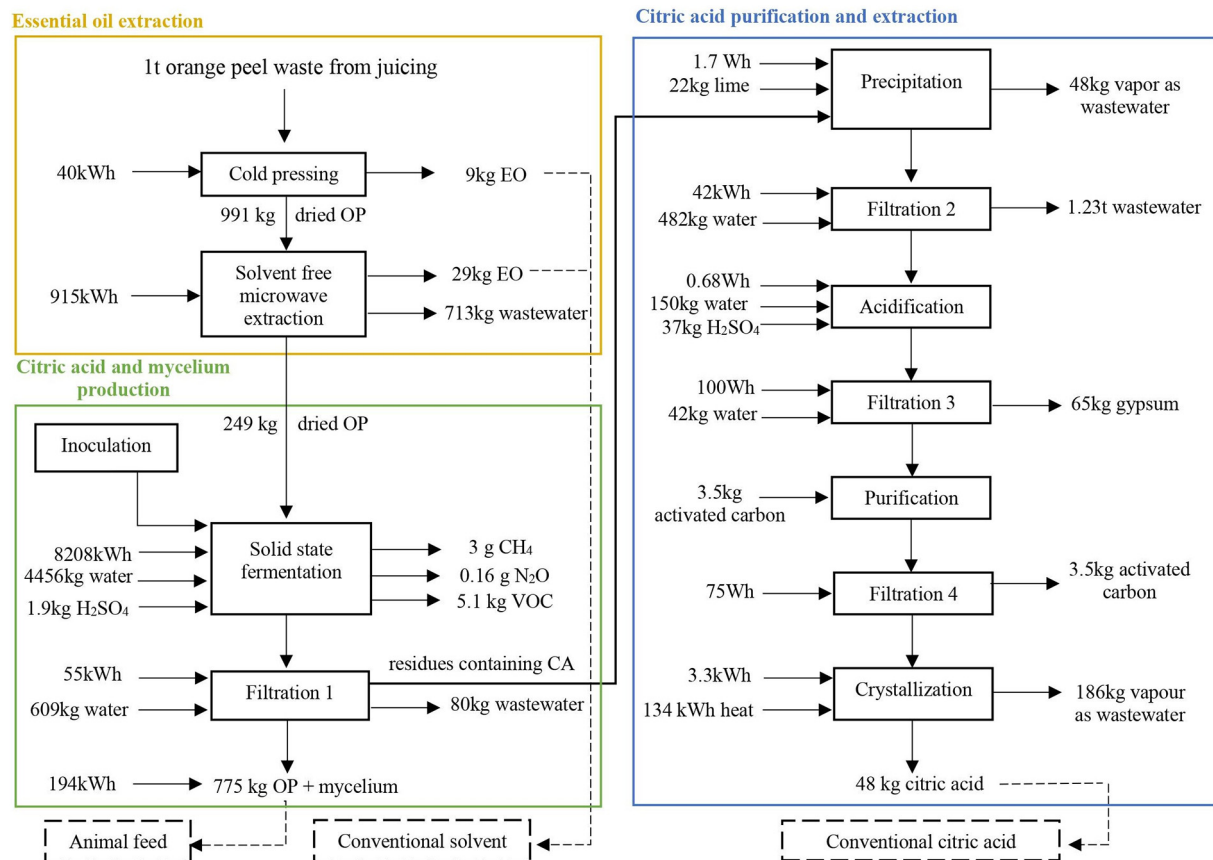


Fig. 1. Process flow diagram of the biorefinery scenario; design for a plant processing 100 t of wet OP per week. OP orange peel, EO essential oil.

report on six impact categories: climate change, land use, acidification, eutrophication marine, eutrophication freshwater, and human toxicity (Figs. 3 and 4), while the rest of the categories can be found in SI2. Similar categories were reported in previous OPW LCA (Negro et al., 2017).

2.2. Biorefinery description

The process chain was built up with the main objective of CA production, with the constraint of following green chemistry principles. The most relevant unit processes have been chosen based on their technical performance reported in the literature, even despite their low technology readiness level (TRL; Flak, 2020). One consequence of the biorefinery process architecture is the production of several co-products, here limonene, animal feed, and gypsum. The process is summarized into three main steps (marked by boxes in Fig. 1). In the essential oil extraction step, limonene extraction is performed via cold-press (CP) and solvent-free microwave extraction (SFME). Limonene is a high-value EO, and its extraction from the OPW is crucial to eliminate its inhibitory effect on microbial growth during the subsequent fermentation steps (de la Torre et al., 2019; Teigiserova et al., 2021). In the citric acid and mycelium production step, CA is produced via solid-state fermentation (SSF) with *Aspergillus niger*, which is grown on wetted OPW.

At the end of the SSF operation, filtration (Filtration 1, Fig. 1) is needed to separate the liquid fraction containing CA from the solid residue. The solid fraction is a mix of OP fiber-rich and mycelia (Show et al., 2015; Berovic and Legisa, 2007), which is used as animal feed (see Section 2.4).

Finally, citric acid purification and extraction include several purification steps to obtain a concentrated form of CA as the final marketable product. CA is precipitated from the aqueous phase as calcium citrate by adding calcium oxide (lime milk), which is a standard method used in the industry

to obtain high purity CA (Show et al., 2015; Berovic and Legisa, 2007). The mixture is filtered (Filtration 2 – Fig. 1), and the filtrate is washed with water. Afterward, the addition of sulfuric acid forms gypsum (calcium sulfate), which is filtered from the formed suspension. Gypsum is a by-product that can be further used in cement factories, and therefore it is not considered a waste in the system (Show et al., 2015). The CA filtrate is further treated by activated carbon removing impurities, followed by filtration step 4 (Filtration 4 – Fig. 1). Finally, a concentrated form of CA is subjected to vacuum crystallization with a forced circulating evaporator, and the final crystals of anhydrous CA are centrifuged (Show et al., 2015; Berovic and Legisa, 2007).

In this process chain, limonene extraction is performed through a combination of cold-pressing (CP) and solvent-free microwave extraction (SFME); yet there were several other options that could have been selected (Teigiserova et al., 2021). In selecting the processes, the following considerations were observed: i) green chemistry principles, ii) limonene extraction that maintains sugars in residue, iii) economic feasibility, and iv) scale-up potential. The rationale for the CP-SFME choice is detailed in (Teigiserova et al., 2021), while the well-researched SSF process (Mitchell et al., 2006) was chosen for citric acid production due to its simplicity and previously identified advantages over submerged fermentation (Lizardi-Jiménez and Hernández-Martínez, 2017; Torrado et al., 2011).

2.3. Scenarios and system boundaries

The management scenarios include a variation of the electricity and heat mixes for the biorefinery scenarios. The reason for this is the influence these mixes have on overall environmental performance as it affects most impact categories (Righi et al., 2018). The following mixes were considered: a) current electricity mix of Denmark (electricity as included in

Ecoinvent 3.6; for the consequential electricity process, it corresponds to mixes currently forecasted to react to a demand change in 2050 as detailed in Vandepaer et al., 2018); b) future renewable energy mix forecasted for Denmark (DEA, 2020; Ropenus and Klinge Jacobsen, 2015) and c) 100% wind for electricity and 100% biogas for heat production (more details in Section 2.5.2).

Decentralized production is envisioned as an on-site business expansion activity of juicing factories; therefore, transportation of OPW to the biorefinery is excluded. Two references, or business-as-usual (BAU), OPW management scenarios were considered, namely incineration and animal feed production. These were modeled according to Negro et al. (2017). Landfilling was excluded as BAU since the EU aims to avoid landfilling when possible (EC, 2014), and it has been previously documented as an environmentally harmful OPW treatment contributing to climate change (Negro et al., 2017).

In total, 10 main scenarios are assessed with LCA (Table 1). The influence of the plant size on environmental performance was also addressed. In addition to the capacity of 100 t OPW week⁻¹ considered in the baseline, two additional extremes were considered, namely size of 0.5 t OPW week⁻¹ (to represent small-sized plant) and 1000 t OPW week⁻¹ to reflect large plant sizes (one of the largest plant in the EU, Spanish juice factory producing up to 400 t of OPW ex-juicing per day (Martín et al., 2018)). The analysis for these extreme plant sizes considers both attributional and consequential processes for the “current” energy mix only (Table 1).

2.4. Marginal products

In line with the system expansion approach used for multi-functionality, the products generated are substituting products on the market, here those most likely to react to the change on the market (Weidema et al., 2013). Those products are hexane, substituted by limonene on a double mass basis as an organic solvent source (Negro et al., 2017); conventional CA production (substituted by the CA produced herein from OPW) and a mix of soy meal, maize, and palm oil, substituted by the OPW + mycelia mix as protein, carbohydrate, and lipid sources for animal feed. Maize is considered as marginal carbohydrate (145.4 kg avoided), soybean meal as marginal protein (16.3 kg avoided), and palm oil as marginal lipid (1 kg avoided). The substitution was modeled according to Tonini et al. (2016), with calculation details provided in Table SI3 Supplementary Information 1 (Table SI1.6). The mycelia from CA production via *Aspergillus niger* is a certified animal feed with a known composition (Citrique Belge, 2020; Jungbunzlauer, 2020).

These three products are independent of the energy variation, and the energy used in their background processes is the one considered in Ecoinvent 3.6 for both attributional and consequential processes.

Table 1

LCA scenarios. Background data refer to data in Ecoinvent 3.6.

Scenario specificities	Attributional	Consequential
Biorefinery (citric acid, animal feed, and limonene production)		
“Current” ^a electricity mix	Bioref Attr	Bioref Conseq
Renewable electricity mix	RE Attr	RE Conseq
Wind electricity	WIND Attr	WIND Conseq
Business-as-usual		
Animal feed	AF Attr	AF Conseq
Incineration with energy recovery	INC Attr	INC Conseq
Plant size variation		
Plant processing 0.5 t week ^{-1b}	0.5 t Attr	0.5 t Conseq
Plant processing 1000 t week ^{-1b}	1000 t Attr	1000 t Conseq

^a For the consequential process, this does not refer to today's electricity but includes 2050 marginal electricity (market response) currently forecasted under business-as-usual development pathways.

^b Compared using the current electricity (as the ratio of difference would be the same throughout different electricity scenarios).

2.5. Scale-up procedure and life cycle inventory

2.5.1. Biorefinery process modeling

Building life cycle inventories is especially challenging when emerging innovative technologies or processes are involved without proven efficiencies (Pizzol et al., 2021), yet, anticipating the consequences of technologies before their deployment is the very essence of LCA. To overcome this, it has been proposed to couple process design and modeling with LCA (Ahmadi and Tiruta-Barna, 2015; Mery et al., 2013) in order to identify the influence of the chosen unit processes, their operating conditions, and scale on environmental impacts. Process design modeling is necessary to fairly represent the flows, input, and activity data needed to represent potential biorefinery in a realistic way. Process design and modeling allow, as in conventional LCA, to build the process chain, from raw material to the final product, but it further refines inventories through a consistent dimensioning of the process equipment, calculating the material and energy balance at the desired scale and reflecting the actual operating conditions at the plant level.

The scale-up procedure selected the most relevant process design (as described in Section 2.2) to build the plant flowsheet. Predefined production capacity was considered as the main parameter for the equipment dimensioning and process scale-up. The general methodology combining process modeling and scale-up with LCA is presented in Fig. 2 and builds upon the methodology developed in (Ahmadi et al., 2019; Ahmadi and Tiruta-Barna, 2015; Bisinella De Faria et al., 2015; Mery et al., 2013), where logical and harmonized data selected from literature are used, along with plausible formulation of physico-chemical and thermodynamic phenomena, to simulate the process efficiencies at a larger scale.

Overall data were based on experiments at laboratory scale, reported in the literature. The plant design and scale-up procedure included the development of physico-chemical and energy models to determine process inventories (material and energy balances) based on the specificities of processes, including eventual fugitive emissions. Here, emissions of CH₄, N₂O, and VOC during the SSF process were modeled according to Maulini-Duran et al. (2015). No other process emissions were considered.

The Simulis package of the process simulation ProsimPlus® software (ProSim, 2020) was used for the calculation of fluid and thermodynamic properties of the processes involved. SI1 provides the general descriptions on the choice and suitability of equipment involved with a summary of process inventories as well as additional data and discussions on biomass composition and animal feed modeling. SI2 provides details on inventory calculations and equipment sizing.

The aim of the modeling phase was to use logical and harmonized data selected from literature for a reliable simulation of the process efficiency at a larger scale which can be obtained within reasonable computational costs. Inventory calculations and sizing of equipment are based on a reasonable formulation of physico-chemical and thermodynamic phenomena.

Energy consumption varies considerably on the process type, and operation parameters such as type of equipment, size, surface, materials, thermal insulation, and others, and therefore some simplifications and assumptions are expected. Here, this includes the composition of OPW and yields based on lab-scale experiments found in the literature; stoichiometry (such as the formation of CaO during precipitation); specification from industrial reactors (such as rotating drum chosen for SSF when using 1000 t OPW week⁻¹ (Maguin, 2020)), and other physicochemical parameters. It should be noted that any decision in scale-up needs to be in line with the economic consideration (such as the number of reactors).

At the start of the process modeling, the yields for all technologies considered were based on data reported for sweet orange with a focus on Valencia and Navel variety (*Citrus sinensis* L.): CP yield 0.009 g EO g⁻¹ dry OPW (Bousbia et al., 2009; Ferhat et al., 2016; NIIR Board, 2008), SFME yield of 0.029 g EO g⁻¹ dry OPW (Bousbia et al., 2009; Ferhat et al., 2016; Boukroufa et al., 2015; González-Rivera et al., 2016), CA production based on average sugar content 0.365 kg total sugar kg⁻¹ dry OPW reported in (Torrado et al., 2011), and CA yield of 0.403 g CA g⁻¹ dry OPW (Hamdy, 2013; Torrado et al., 2011). The details on the technology and composition of the OPW can be found in SI1, while details on yield

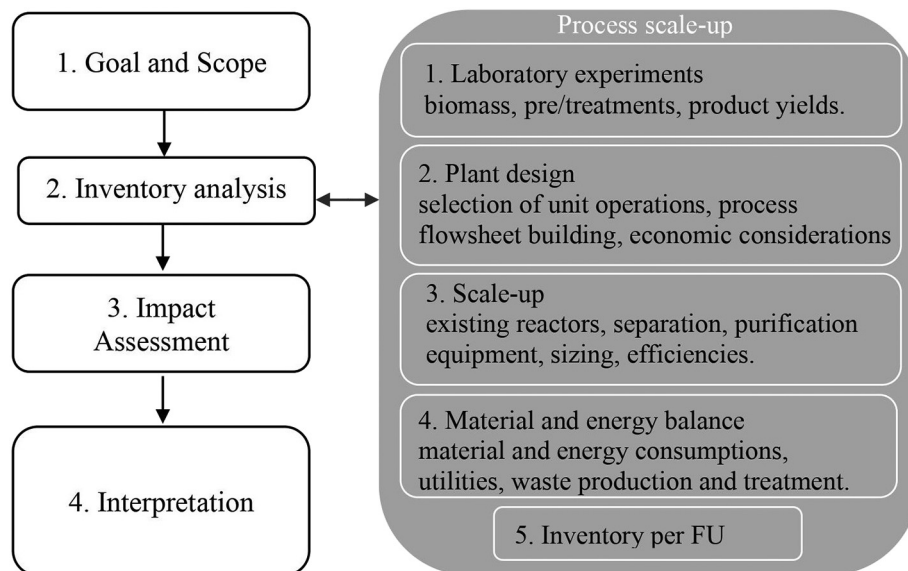


Fig. 2. Integrated process modelling and LCA. LCA is represented by four main steps, while scale-up procedure steps represent general points for ex-ante scale-up modelling, with step 5 included for LCA incorporation.

calculation can be found in sheet “Yields” in SI2. Details on the technology are included in SI1, with calculation details provided in sheet “Limonene extraction” for CP and SFME; sheet “SSF 100 t” and “Filtration 1” for CA and mycelium production; and sheet “Precipitation” — “Concentration & Drying” for purification and concentration steps. Even though laboratory production of CA can be achieved in three days (Torrado et al., 2011; Hamdy, 2013), we considered five-day production at the industrial scale (Berovic and Legisa, 2007). In this inventory, no additional nutrients are added to SSF.

2.5.2. Energy and heat projections for the renewable scenario

The future renewable electricity and heat mix was projected according to predictions from the Danish Energy Agency (DEA, 2020). When data were lacking or unclear, the projections were supplemented by the previous forecasts (Ropenus and Klinge Jacobsen, 2015), represented in Table 2. More details on the data are presented in SI1. As the Ecoinvent database 3.6 includes biogas as a rest-of-the-world process (RoW), it contains some unrealistic background processes for Danish biogas, such as fertilizer impact for global cotton cultivation. Therefore, the biogas was modeled according to manure mono-digestion inventories of Hamelin et al. (2014). This scenario includes avoided impacts of raw manure management and avoided marginal energy production.

Table 2

Electricity and heat mix projections for the renewable energy scenario.

Energy	%	Reference
Heat-future		
Heat pump	12	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)
CHP from municipal waste	32	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)
Electrification	16	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)
Biogas	30	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)
District heating from woodchips	10	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)
Electricity-future		
Wind offshore	40	(DEA, 2020)
Wind onshore	15.8	(DEA, 2020)
Solar PV	15	(DEA, 2020)
Biogas	14.6	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)
CHP with wood	14.6	(DEA, 2020; Ropenus and Klinge Jacobsen, 2015)

2.5.3. BAU scenarios

BAU Animal Feed from OPW ex-juicing was modeled according to Negro et al. (2017), which includes OPW pelletizing. We considered a transport distance of 100 km for 100 t and transported via lorry (Ecoinvent 3.6). This relatively short distance (considering the animal feed market) was selected as a maximum to reflect that animal feed from OPW is only profitable if kept local, as previously identified by Zema et al. (2018).

Incineration was modeled according to Negro et al. (2017) and data from the Ecoinvent 3.6 database. The distance was assumed as 20 km of fully loaded municipal collection by lorry. This distance was chosen based on the nearest incineration plant around the Rynkeby juicing plant in Denmark via Google maps.

2.6. Sensitivity analysis

The importance of uncertain assumptions on the results is addressed here through sensitivity analysis. These key sensitive assumptions were identified after a first contribution analysis of the LCA results. As the scenarios already investigate the sensitivity of the energy, the next most sensitive assumption identified relates to the choice of the solvent substituted by the produced limonene. To this end, a sensitivity analysis was performed considering acetone as an alternative organic solvent substituted by limonene (on an equal mass basis) as previously identified as a marginal organic solvent in Pourbafrani et al. (2013). Acetone is replaced on the equivalent mass basis (Pourbafrani et al., 2013), as opposed to hexane, which is replaced on a double mass basis due to limonene being a “stronger” solvent (Negro et al., 2017). It was here judged that the importance of these assumptions on the overall results interpretation was more significant than the variation of the numerical values used in the LCA, given the relatively small amount of data required for the system under investigation. Therefore, uncertainty was addressed in the light of sensitivity analysis only, which can be accounted as one of the limitations of this study.

3. Results and discussion

3.1. Life cycle impact assessment and interpretation (LCIA)

Figs. 3 and 4 present, for 6 of the 16 impact categories covered by the EF LCIA method, the LCA results with a breakdown per process, while the results for the other impacts are presented in SI3. The 6 impact categories

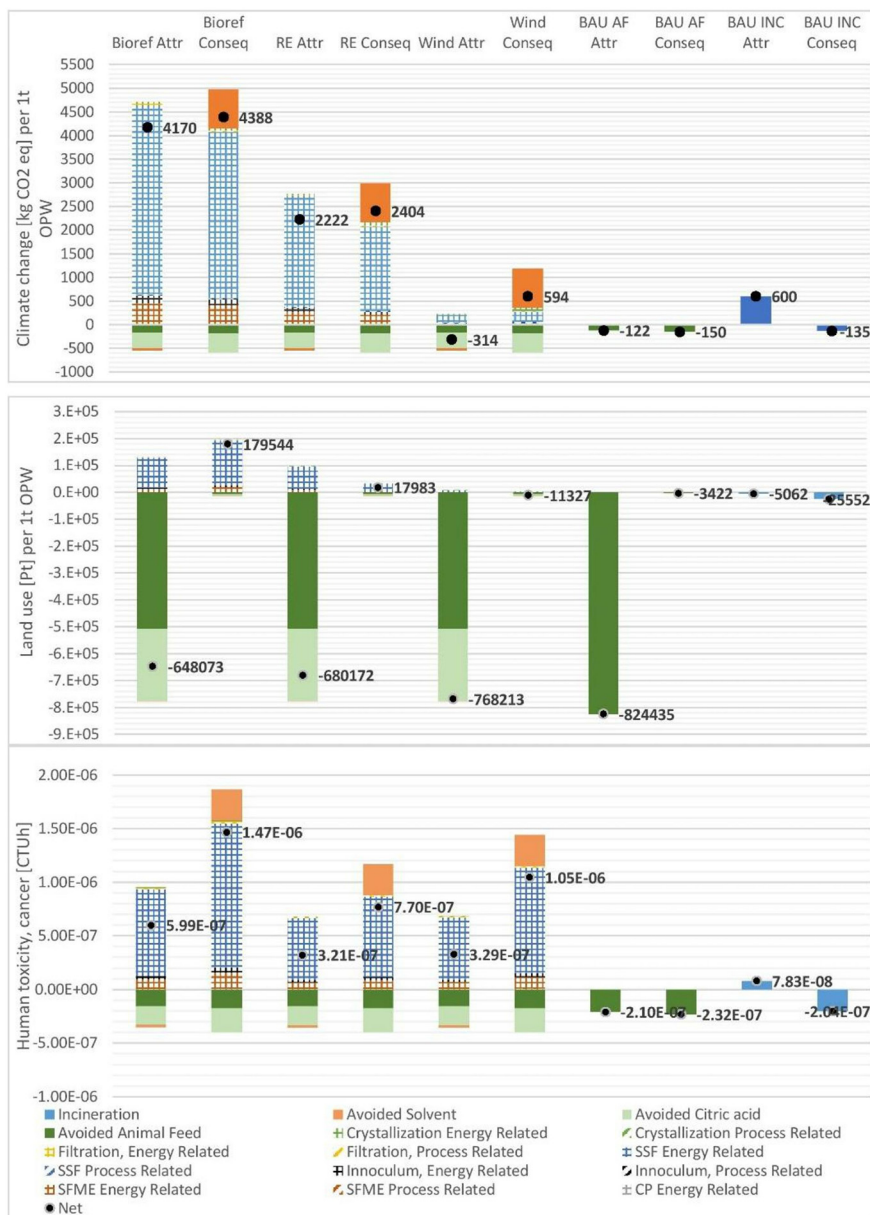


Fig. 3. Impact contribution for human toxicity, cancer (CTUh); land use (Pt); and climate change (CO₂ eq); the units are as defined in the Environmental Footprint method. Bioref Average: biorefinery with average market data and current average energy mix; Bioref Marginal: biorefinery with marginal market data and current marginal energy mix; RE Average biorefinery with average market data and future renewable average energy mix; RE Marginal: biorefinery with marginal market data and future renewable marginal energy mix; WIND Average: biorefinery with average market data and wind average energy, WIND Marginal: biorefinery with marginal market data and wind marginal energy mix; AF Average: Animal Feed Production with the average market; AF Marginal: Animal Feed Production with the marginal market; INC Average: incineration with energy recovery considering the average market; INC Marginal: incineration with energy recovery considering the marginal market.

were selected as commonly reported for waste management, including previous LCA (Negro et al., 2017). The positive values represent induced contributions, while the negative values signify avoided impacts. The net impact is a subtraction of avoided from induced impacts and shows the overall performance of the scenarios. Detailed contribution analyses in 3 levels (process, activity in the process, and contributing substance) are presented in SI3. It should also be noted that the variation of the energy mix does not affect the amount of biorefinery products manufactured (animal feed, citric acid, and hexane), hence their impacts being the same in all scenarios for a given impact category.

3.1.1. Climate change

The climate change impact category gradually improves (decreased impact) when the energy mix changes from “current” to future renewable, as displayed in Fig. 3. It is visible that energy-related emissions are the most

influential, with the SSF process having the most impact through the different scenarios. This was expected as SSF requires more energy than other biorefinery processes (Fig. 1). The most contributing background processes to the current electricity are hard coal (35% of the SSF contribution) and woodchips (25% of the SSF contribution) in the mix of the current attributional inventory dataset (Bioref Attr), with the main contributing substance being fossil CO₂ (62% contribution to the SSF impact). For the consequential dataset (Bioref Conseq), the main contributor is a global wood process in the electricity mix (91% of the SSF impact), biogenic carbon dioxide being the primary substance responsible for the impact (90% of the SSF impact). For the Bioref scenarios, the consequential energy mix was expected to perform better since it implicitly considers that the least competitive technologies are the first ones to be replaced, here coal and lignite. Yet, albeit a lower impact from energy consumption in the SSF process was observed, it was compensated, in the Bioref consequential scenario, by

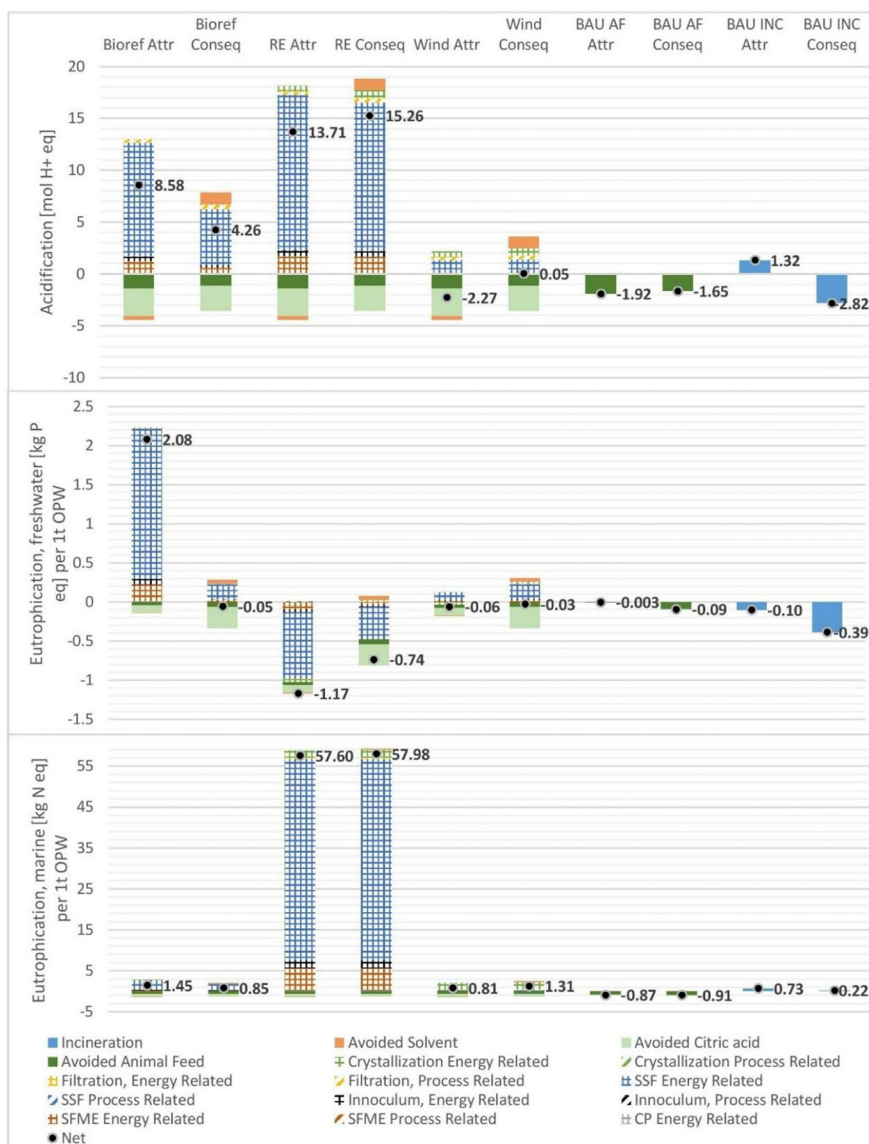


Fig. 4. Impact category graphs for Eutrophication, marine (kg N eq); Eutrophication, freshwater (kg P eq); and Acidification (mol H⁺ eq). Bioref Average: biorefinery with average market data and current average energy mix; Bioref Marginal: biorefinery with marginal market data and current marginal energy mix; RE Average biorefinery with average market data and future renewable average energy mix; RE Marginal: biorefinery with marginal market data and future renewable marginal energy mix; WIND Average: biorefinery with average market data and wind average energy, WIND Marginal: biorefinery with marginal market data and wind marginal energy mix; AF Average: Animal Feed Production with the average market; AF Marginal: Animal Feed Production with the marginal market; INC Average: incineration with energy recovery considering the average market; INC Marginal: incineration with energy recovery considering the marginal market.

effects related to avoiding hexane as a marginal solvent, which translated into net positive impacts. This reflects the fact that hexane is produced along with several co-products, including methylcyclohexane, and to a smaller extent, heptane, whose production has to be compensated for when avoiding hexane.

The RE scenarios' main impacts for both attributional and consequential dataset come from biogas generation. The biogas process includes the CO₂ and N₂O emissions from the digestate spreading on land, the fugitive losses of CO₂ and CH₄ during the digestion process, and the CO₂ and CH₄ emissions from the co-generation of heat and power, which are partially counterbalanced by avoiding the outdoor storage of raw manure (CH₄ and N₂O; Hamelin et al., 2014) and raw manure spreading (CO₂ and N₂O, as for digestate). In case 100% wind energy is used in the system, the contribution to climate change is mainly influenced by the materials needed for the construction of the wind turbines (such as iron and clinker), which correspond to 80% of fossil CO₂ for both attributional and consequential

scenarios in the SSF process. Wind energy with the attributional dataset is the best-performing among the biorefinery scenarios.

The climate benefits of avoided conventional CA, whether for the attributional or consequential dataset, are mainly due to CA's avoided production in China and its transportation (lorry, freight, and sea transport).

BAU scenarios are the best performing scenarios after WIND Attr. Animal feed scenarios were expected to have mostly negative impacts since it avoids the production of maize and soymeal, and it includes minimal energy for OPW pelletizing and local transport. Avoided maize is the main negative impact for both attributional and consequential datasets, albeit for different reasons (SI3). The difference between attributional and consequential incineration is due to the included avoided heat generation with consequential datasets, where the combustion of hard coal and lignite from Poland are avoided, among others.

Table 3 shows the impact of excluding biogenic CO₂ on the climate change category for biorefinery scenarios. Excluding biogenic CO₂ does

Table 3
Climate change impacts difference between including and excluding biogenic CO₂.

	Bioref Attr	Bioref Conseq	RE Attr	RE Conseq	Wind Attr	Wind Conseq
With biogenic CO ₂ (CO ₂ eq)	4170	4388	2222	2404	-314	594
Only fossil CO ₂ (CO ₂ eq)	2779	754	-2970	-1488	-400	546

affect the foreground model, and results have a significantly lower impact on climate change for the current and future RE energy mix. Bioref Attr has 33% lower total impact, Bioref Conseq 83% lower impacts, and RE Attr and RE Conseq results in 234% 162% lower, respectively. Other impact categories are not influenced, but for the climate change category, the conclusion in terms of ranking the scenarios from the most to the least performant would be affected. For instance, not accounting biogenic CO₂ renders the biorefinery with future renewable energy as the best performing scenario among the biorefinery options, instead of biorefinery with wind (Fig. 3). More details can be found in SI3.

3.1.2. Land use

Avoided animal feed, here translated as avoided maize, soy, and palm oil, is the process that avoids most of the land use, as seen in Fig. 3. For the attributional inventories, the animal feed process in the biorefinery as well as in BAU avoids mostly home composting used as fertilizer (which specifies 765 m² of green land per kg of maize in the attributional inventories) in maize production (97% of impact in BAU Attr). This generates an important avoided land use impact, mostly as a result of the flow “occupation, urban, green areas” of the avoided home composting process (this flow represents over 90% of the impact). Minor avoided processes coming from nitrogen fertilizer. On the other hand, the consequential dataset for (avoided) maize, soy and palm oil incorporate only minor inputs in terms of land occupation and transformation. Here, the biggest (avoided) contributing background processes are Maize grain production {RoW} and {US} (30% Rest-of-the-world production and 26% of the impact from US production). The largest avoided flows are “transformation, annual crop, non-irrigated” and “occupation, forest, intensive”.

The processes that contribute to SSF energy-related impacts for Bioref Attr and Conseq, as well as RE Attr and Conseq, are woodchips and their production from Sweden (mostly birch and pine) with varying contributions (usually around 25% for each kind of tree), which are used in the energy mix. Additionally, the current attributional electricity has a positive impact due to compost (43% of SSF), which is needed to cultivate some of the input biomass for energy generation. The wind has a much lower impact, mainly from the road occupation and forest transformation and occupation (consequential inventory), and green urban area occupation (attributional inventory) that are needed for the wind turbine production and the area it occupies.

The attributional incineration process avoids electricity production, primarily woodchips mixture in attributional electricity. This is counterbalanced by induced impacts by the electricity needed for pre-drying of OPW and internal use (i.e. for the incineration process itself). The consequential incineration has larger avoided impacts as the market's response leads to avoided heat generation that includes woodchips from Sweden and Germany. This avoids the occupation of the forest. More details can be found in SI3.

3.1.3. Human toxicity, cancer

Generally, human toxicity was not expected to have significant results, which is also in line with the previous LCA on OPW management (Negro et al., 2017). BAU for animal feed with the consequential inventories is the best performing scenario. This is due to avoided maize production (RoW) avoiding impacts of fertilizer with substances like chromium in the water compartment and coke (RoW) as background processes in the agricultural machinery. The attributional data also avoids these processes, but on a smaller scale, while ferrochromium input from the agricultural machinery is the second-largest impactful after avoided maize (RoW)

production. Otherwise, most of the impacts are ferrochromium and coke (RoW), which are background processes in wind turbine production, thus the Wind scenario is on an equal basis with other energy mixes. Consequential hexane induces heptane production, which has minor impacts in other categories, and the main consequences in the human cancer category are due to tetrachloroethylene and cumene background processes in heptane production.

The attributional incineration has positive impacts because it includes waste cement as background processes in the incineration. In contrast, the consequential dataset avoids background processes for steel and iron scraps. As visible from Fig. 3, the consequential scenarios have larger impacts for biorefinery and larger avoided impacts for BAU scenarios, induced by the market response where in feed avoids impacts from fertilizer, and energy mix includes in part avoided background processes. More details can be found in SI3.

3.1.4. Acidification

For acidification, the most impactful scenarios are due to the use of future renewable energy, as visible in Fig. 4. This is due to ammonia emissions to air in biogas management, which makes up 93% of SSF impact (“ammonia (air)” flow). While the background processes of avoided outdoor storage and spreading of raw manure lead to negative impacts, the induced emissions from digestate storage are more impactful. The impact of the current attributional electricity mix is due to hard coal (29% of the SSF impact) and wood chips use (11%). The consequential data include electricity from wood (70% of the SSF process) and copper needed for wind turbines (8%), with total nitrogen and ammonia emissions as the main contributing substances. Equally, several types of copper production processes are the most influential for the Wind energy scenarios for both attributional and consequential processes. The background process Copper {RAS}| production, primary contributing 39% in the consequential scenario (the most contributions in the SSF process) its attributional counterpart contributes 24% (the largest contribution for SSF Wind Attr). Other copper production stack up with smaller impacts (ex. “Copper {RU}| platinum group metal mine operation, ore with high palladium content”).

Avoided citric acid follows the same trend as in other categories. Its avoided production in China is the main negative impact, corresponding to avoided sulfur dioxide emissions (60% of the CA impact for both attributional and consequential datasets). Avoided feed, both for the biorefinery and BAU scenarios, prevents mainly maize production from around the world (RoW, US), including the associated ammonia and nitrogen oxide emissions. Interestingly, the highest avoided contribution for the attributional maize is due to palladium (Russia) being avoided in various agriculture equipment and machinery. BAU Incineration has a positive impact on the attributional data because of the internal energy use (the background process of “Electricity, for reuse in municipal waste incineration only {DK}| treatment of municipal solid waste, incineration”), while this is counterbalanced when using consequential datasets due to avoided heat processes, such as hard coal and lignite from Poland. More details can be found in SI3.

3.1.5. Eutrophication, freshwater (phosphorus)

The trends for this category are opposite to that of acidification for the energy impact, as seen in Fig. 4. RE scenario here avoids the most impacts and are the best performing due to avoided raw manure spreading for both datasets, while the attributional inventories also avoid spoil from hard coal as a background process in the avoided attributional electricity. The current attributional energy mix has positive impacts due to spoil processes from hard coal and lignite, which are present in the electricity mix, while the consequential processes also avoid sulfidic tailings from copper operation as background processes in wind turbine production. The latter is the main impact of the Wind scenarios with several processes from around the world, with China the largest one. The avoided hexane in the consequential dataset has spoil from lignite as the main contributor, which is due to the heat used for methylcyclohexane production, induced by avoiding hexane production (positive impact). Avoided animal feed

impacts are due to background processes of spoil from hard coal and lignite in the avoided maize drying activity and avoided due to maize transport by a freight train. The consequential processes avoid more activities involving these activities as a response to the market. A similar trend as previous categories follows for Incineration, where the attributional processes avoid electricity production, while in consequential incineration process avoids heat and therefore has larger negative impacts (78% due to spoil from lignite mining avoided in heat mixture from Poland). More details can be found in SI3.

3.1.6. Eutrophication, marine (nitrogen)

In this category, the high impact of the RE scenario is due to digestate spreading on the land with nitrate emissions to water responsible for almost all of the contributions. The positive contributions of the current energy scenario are due to coal and wood chips in the attributional mix and wood in the consequential dataset. As these are not influencing the Wind scenarios, here, the most contributing process is the heat needed for crystallization. The contributions are from the biogas, which is the same process used in the RE scenarios (positive impacts are from digestate spreading on land), but here it only accounts for the heat. The avoided impact of animal feed is again due to avoided maize production, mainly to nitrate and nitrogen oxides emissions coming from the nitrogen fertilizer use and its transformation, but also from tillage ploughing, packaging, and seeding activities. The incineration has impacts from the ammonia emissions from the incineration process (likely due wet scrubber process). The consequential incineration avoids heat generation as in previous categories (heat avoided from Poland) and therefore accounts for better results than the attributional. More details can be found in SI3.

3.2. Life cycle impact assessment of different plant sizes

Only the results for climate change impact categories are reported to illustrate differences in the energy efficiency of the individual processing steps. The main difference in the modeling of plant scales for 0.5 t and 1000 t per week are energy consumption of SFME, SSF, crystallization, and water needed for SSF. This is due to different specific requirements of the reactors of a different size; see details in SI2 (tabs “SSF 1000 t OP week” onwards, with main input and output on top of each sheet). As visible from Table 4, the smaller-scale reactors are more impactful, which was expected, these being more energy and resource-intensive. Larger reactors have usually better energy conservation and thus are more efficient per output processed when it comes to energy and water use. As energy is the main factor that influences all the impacts, less energy used translates directly into improved environmental performance. For example, the electricity required per functional unit during the SSF process is 7518 kWh, 8207 kWh, 9129 kWh, for 1000 t week⁻¹, 100 t week⁻¹ (baseline), and 0.5 t week⁻¹, respectively (line 15 in each tab for SSF). This latter scenario (0.5 t) has a 12% higher impact on climate change for Bioref Attr and a 9% higher impact for Bioref Conseq when compared to the baseline scenario. In contrast, the opposite is true if the larger scale is employed. The operation of a large-scale process is more efficient (due to the possibility of grey heat recovery), and therefore, the climate change impacts are 10% lower than the Bioref Attr scenario and 9% lower than the Bioref Conseq scenario. More, it can be observed that these impacts vary in a non-linear manner with the plant

Table 4
Net impacts per 1 t of OPW for different plant sizes.

Impact category	Unit	0.5 t Attr	0.5 t Conseq	1000 t Attr	1000 t Conseq
Climate change	kg CO ₂ eq	4673	4803	3734	4007
Land use	Pt	-6.3E05	2E05	-6.6E05	1.6E05
Human toxicity, cancer	CTUh	6.9E-07	1.6E-06	5.1E-07	1.3E-06
Acidification	mol H ⁺ eq	9.97	4.93	7.41	3.69
Eutrophication, freshwater	kg P eq	2.33	-0.03	1.87	-0.08
Eutrophication, marine	kg N eq	1.74	1.04	1.20	0.69

size, with a faster increase for the small size plant. This shows a promising prospect if such biorefinery is employed on a larger scale, especially when combined with future renewable energy. A similar trend can be observed in other impact categories, which leads to the conclusion that the economy of scale is also manifested by the reduction of environmental impacts.

3.3. Sensitivity analysis

As visible from Figs. 3 and 4, avoided hexane as the solvent has, for the consequential biorefinery scenarios, a non-negligible contribution to the different impacts analyzed. Table 5 presents the total impact results comparing hexane and acetone as avoided solvent from the production of limonene. For average background data, acetone and hexane yield comparable results for most of the categories, though acetone as the avoided solvent avoids double of the climate change but is four times less performant for land use. Marginal datasets show acetone having a better performance for all categories except land use. This is due to a simpler market reaction when acetone is used, which only leads to avoided products of acetone. On the other hand, the marginal dataset for hexane not only avoids but also induces production, as described in Section 3.1, and therefore leads to positive impacts for most of the categories.

The process for (avoided) CA included its production in China, involving coal energy. Alternative production in other countries can involve its production from industrial waste (such as steep corn liquor or molasses) and portions of future renewable energy in the mix. However, the avoided impacts from such a process would still not significantly change the overall conclusion, as visible from the CA portion in Figs. 3 and 4. Likewise, the overall impacts of the scenarios would remain similar when changing acetone for hexane, even when consequential processes of hexane have a significantly higher impact than acetone production.

4. Perspectives and limitation

4.1. Methodology and results

We acknowledge the limitation of the result representation regarding the methodology used. The authors chose to compare both attributional and consequential datasets using the Ecoinvent3.6 LCI database in order to highlight the variation when applying background processes. From the results, it is possible to see that the avoided products can steer the results towards different conclusions and are highly dependent on the choice of the background process data. For example, animal feed is the prominent avoided impact in the land use category for the attributional dataset, while avoided citric acid is significantly present only for the consequential dataset for Climate change, Human toxicity, and Acidification. The change is due to the anticipated market response when replacing hexane (avoided solvent). Its replacement by even more dangerous substances points out the need and perhaps future trend of phasing out those substances and replacing them with biobased (despite interactions with land use changes) or other less harmful chemicals.

Another difference in background data considers the incineration process for the climate change category, mostly prominent for categories of Climate, Human toxicity, Acidification, and Eutrophication freshwater. For the latter three, consequential data anticipate phasing out coal and lignite in the energy supply, which translate as more significant avoided impacts. While for the Climate change attributional process of “Municipal solid waste {DK}| treatment of, incineration” is the largest contributing process, while in the consequential dataset, the same process is counterbalanced by avoided heat production import from Poland.

Lastly, there is a significant difference between Bioref attributional and Bioref consequential datasets for the Eutrophication, freshwater category for the SSF process. Here, the impacts of the SSF process are mostly related to energy use, given the great amount of energy consumed per FU. The net positive impacts are due to spoils from coal and lignite caused by the import of electricity in the attributional dataset (49% coal, 45% lignite impacts to SSF), which is considerably smaller with the consequential data that

Table 5
Impact category comparison of acetone and hexane for the average and marginal background datasets.

Impact category	Unit	Acetone average	Acetone marginal	Hexane average	Hexane marginal
Climate change	kg CO ₂ eq	-94	-96	-44	827
Human toxicity, cancer	CTUh	-3.68E-08	-1.36E-08	-2.41E-08	2.90E-07
Acidification	mol H ⁺ eq	-0.456	-0.452	-0.404	1.148
Eutrophication, freshwater	kg P eq	-0.010	-0.008	-0.009	0.050
Eutrophication, marine	kg N eq	-0.068	-0.067	-0.058	0.317
Eutrophication, terrestrial	mol N eq	-0.735	-0.731	-0.632	3.369
Land use	Pt	-192	8	-773	-948

considers market change (16% coal impact to SSF). The process of “sulfidic tailings, from copper mine operation {CN}” is the largest contributor in the consequential data (21%), most likely due to copper use in wind power as wind energy included in the future market response. More details in SI3.

4.2. The restriction of raw OPW as animal feed

Despite animal feed being better performing than most scenarios in this study and the previous study by Negro et al. (2017), the use of OPW is restricted by several factors. Negro et al. (2017) discuss bitter taste, low nutritional content as decreasing OPW feed marketability, together with increased costs for required pelleting. Moreover, OPW can be potentially toxic in animal feed and have anti-nutritional properties, especially for monogastric animals (Ani et al., 2015). Therefore, despite the displacement of marginal feed calculated according to the lipid, protein, and carbohydrate content, OPW can be used in very limited amounts in the feed mix. In order for animal feed to be possible and economically feasible, it needs to be used locally (Zema et al., 2018). Therefore, using up to 100 t of OPW for animal feed weekly is an unlikely scenario and even more unlikely if more waste is present (such as 1000 t week⁻¹).

4.3. Considering only limonene biorefinery

Figs. 3 and 4 show that CA production has the highest impact in the biorefinery. Therefore, a more sensible option is to stop the biorefinery at the limonene extraction and use the residues for the animal feed. The biorefinery presented the option of using limonene as an organic solvent. However, limonene has further uses, which have limited evaluation for LCA. Ciriminna et al. (2014) provided an overview of limonene as a versatile chemical for the bioeconomy, focusing on production potential from oranges. It can be used as an insecticide, as an ingredient in environmentally friendly water-based industrial cleansers due to its low flammability, low odor, and VOC (Ciriminna et al., 2014). It also shows prospects for the production of polyamides, thermoplastic polycarbonates, and polyesters (Ciriminna et al., 2014). Limonene can also be used medical and pharmaceutical industries. It is a great solvent of cholesterol, relieves heartburn, and has shown chemopreventive activity against many types of cancers in clinical trials (Sun, 2007). Therefore, sustainable extraction of limonene from OPW is a promising valorization to be implemented in the near future.

The economic assessment is the next step to evaluate the feasibility of limonene extraction with subsequent use of residues for animal feed. Further, SFME provides dry residues that can be transported on longer distances and thus overcoming the limitation of being used only locally, as explained previously.

4.4. Energy transition dependency and insights for policymaking

Previous studies have highlighted the potential of using OPW for energy production, limonene, and animal feed (Negro et al., 2017; Pourbafrani et al., 2013), while this study adds the option of biorefinery for improved animal feed and citric acid production. Negro et al. (2017) has identified that landfilling and composting should be avoided and that biorefinery products (including energy) are the most likely route for OPW. And while incineration is better or equally well-performing considering

environmental impacts, its potential for the bioeconomy sector is minimal. Negro et al. (2017) has also identified that chemical and energy production is the most beneficial route for the environment and economy, although some incentives would be required. Therefore, the current study expands on that notion including energy transition towards a low-fossil renewable mix is crucial for future sustainable bioeconomy for the valorization of OPW. From the impact category results, it is evident that the lower impact can be achieved with the energy transition, with process emissions being negligible for most categories. However, the use of biogas can lead to increased impacts due to the emissions from the spreading of digestate. Biogas is planned to be used in the energy mix in Denmark for the future, and therefore special attention should be given to its management, i.e., its use as fertilizer. In our system, we did not consider the avoided fertilizer from the digestate to see the potential impact of this scenario when used in future sustainable agriculture. If such displacement is considered, the impacts would be lower.

5. Conclusion

Biorefinery of OPW for limonene, citric acid, and improved animal feed shows a clear dependency on energy transition for environmental production. The biorefinery was modeled with the engineering-based scale-up and therefore included realistic energy and material inputs, with results highly dependent on the energy mix. The background attributional and consequential processes show different results but with similar trends. Fully future renewable energy mixes show promise if the impact coming from the digestate use on the field is offset in the future by carefully implemented biogas management. The performance of different scales for biorefinery production has shown better results on a larger scale, with a non-linear dependency of the impacts on the plant's scale. The high dependency on energy for all the studied scenarios also highlights the need for renewable energy to keep the future circular bioeconomy of OPW sustainable.

Raw OPW used as animal feed and incineration showed an overall better performance, mostly due to the simplicity of the process compared to the biorefinery and reduced energy demand. However, both business-as-usual scenarios have limited use for the bioeconomy. OPW are anti-nutritional and, as a result, can be incorporated with limits in the feed, and the production needs to remain local to be economically feasible. Incineration represents a limited potential for the bioeconomy, especially because OPW has high moisture content and requires substantial energy for drying.

The sensible solution would be to only extract limonene as the most contributing impacts are due to SSF, i.e., citric acid production. The rest of the residue, now in a dry state after solvent-free microwave extraction, should be used as animal feed. The dry residues are more stable and can be transported on longer distances to displace a portion of environmentally-intensive feed, such as maize. This strategy is not only in line with the updated food waste hierarchy but also considers proven environmental technologies and thus represents one of the most optimal strategies for OPW waste management strategies and integration in the circular bioeconomy. The essential oil extraction can be realized as an add-on in current juice facilities, while dried residual OPW can be directly used as animal feed. The latter is of high importance to any region or country with large local animal production, such as Denmark.

CRediT authorship contribution statement

Dominika Alexa Teigiserova: Conceptualization, Methodology, Validation, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Lorie Hamelin:** Conceptualization, Methodology, Validation, Writing – review & editing, Supervision, Funding acquisition. **Ligia Tiruta-Barna:** Methodology, Validation, Investigation, Writing – review & editing. **Aras Ahmadi:** Methodology, Validation, Investigation, Writing – review & editing. **Marianne Thomsen:** Conceptualization, Validation, Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors do not report any declaration of interest.

Acknowledgment

The research of D.A. Teigiserova and M. Thomsen was funded by the Horizon 2020 project DECISIVE (Decentralised valorization of biowaste) under grant agreement N° 689229, Aarhus University's Centre for Circular Bioeconomy. The contribution of L. Hamelin was funded by the Cambioscop project, partly financed by the French National Research Agency, Programme Investissement d'Avenir (ANR-17-MGPA-0006) and Region Occitanie (18015981). L. Tiruta-Barna and A. Ahmadi participated as collaborators in the Cambioscop project.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.152574>.

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