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Quantifying the environmental implication of cotton-Fiber-based Nanocrystalline cellulose: A life-cycle assessment

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ABSTRACT

Considering the increasing demand for nanocrystalline-cellulose in the industry, due to its exceptional physical and biological properties, cheaper and more efficient production processes are sought. Addressing environmental concerns, especially within the framework of EU policies, this study employs Life Cycle Assessment (LCA) to evaluate the environmental performance of a novel nanocrystalline-cellulose production procedure, encompassing biomass depolymerization, rinsing, and bleaching. The LCA aims to identify environmental hotspots, explore mitigation measures, and enables comparisons with other LCA studies on nanocrystalline-cellulose. The results are calculated and reported for 19 environmental impact categories, using the ReCiPe 2016 impact assessment method. The production of 1 kg of dry nanocrystalline-cellulose using the novel process emits 63.7 kg CO_2 equivalent, which is lower than the literature average (68 kg CO_2 equivalent). The solvent (e.g. diethylene glycol) is the major contributor to the global warming potential and fossil-fuel depletion potential in the product stage of the nanocellulose, while the electricity requirements and glycerin represent environmental hotspots regarding 15 of the 19 impact categories assessed. In terms of the water-consumption potential, the environmental hotspot is production of raw materials (e.g. cotton fibers). Electricity contributes more than 50 % of the burden to the impact categories associated with ionizing radiation, the pollution of aquatic ecosystems and human toxicity related to cancer. It also holds a significant share of the burdens for terrestrial acidification (48 % of the impact), the formation of fine particulate matter (46 % of the impact), and human toxicity related to non-cancer diseases (37 % of the impact). This underscores the importance of optimizing the production process, possibly through upscaling. Additionally, incorporating on-site renewable energy sources and utilizing biomass-derived diethylene glycol can enhance the environmental performance of nanocrystalline-cellulose.

1. Introduction

Biomass is unique in its ability to supplement fossil-fuel-based raw materials, particularly liquid fuels and chemicals like methanol, ethanol, biodiesel, etc. (Antar et al., 2021). Regrettably, the practice of burning biomass undermines its potential benefits in diverse applications (European Commission, 2019). However, the cascade use of biomass promotes the biomass components with the highest added value, combined with the extraction of components of lower added value, and only at the end of the chain is the residue burnt as a source of energy (Haberl and Geissler, 2000).

Nanocellulose is a valuable component of biomass. Its efficient production is at the core of many different research efforts, with have an ever-increasing number of applications (Dalli et al., 2018; George and Sabapathi, 2015). Nanocellulose can be used in various construction materials, as well as in composites, additives, coatings, etc. (Foroughi et al., 2021). But the demand for cheaper and more efficient production also comes from other industries, such as packaging, bio-composites and bio-plastics, paper and textiles (Phanthong et al., 2018; Trache et al., 2020).

Nanocellulose can improve the mechanical characteristics, the barrier properties (Jayshree Samuel Jacob, 2022), and the product printability (Lengowski et al., 2019), while different technologies can be used to produce it from biomass input materials such as wood, cellulose, straw, and sugar beet (Das et al., 2022; Sharma et al., 2018). There are three types of nanocellulose: nanofibrillated cellulose, nanocrystalline cellulose and bacterial cellulose (Arvidsson et al., 2015).

In this study we investigated the nanocrystalline cellulose extracted

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from cotton fibers. This is a renewable nanomaterial suitable for biomedical applications due to its exceptional physical and biological properties, such as surface chemistry, low toxicity, and biodegradability, as well as biocompatibility (Jayshree Samuel Jacob, 2022) and applications in medical implants, drug-delivery systems, wound healing, tissue engineering, cardiovascular disease, and antibacterial/antimicrobial activities (Bhat et al., 2017).

In recent years there has been a surge in the amount of research focused on the use of cotton fibers for the production of nanocrystalline cellulose. Natural cotton, waste cotton, and cotton products can be used as raw materials for the production of nanocrystalline cellulose (Pandi et al., 2021; Wang et al., 2017).

The synthesis of cellulose nanocrystals from cotton using ultrasound-assisted acid hydrolysis was reported by Pandi et al. (2021), while Theivasanthi et al. (2018) reported the extraction of cellulose nanocrystals from natural cotton through acid hydrolysis using sulfuric acid. In terms of their physical and chemical characteristics, cellulose nanocrystals extracted from natural cotton have a high crystallinity, a small particle size, and stable thermal properties (Kunaver et al., 2016; Pandi et al., 2021). The potential applications of such nanocrystalline cellulose include electronics, cosmetics, tissue engineering, polymer fillers, and food processing (Pandi et al., 2021). Cellulose nanocrystals can also be extracted from waste clothing, particularly cotton-based textile waste (Vanzetto et al., 2021). The extraction involved acid hydrolysis with sulfuric acid. The reported yield was between 51 % and 62 % by weight. The properties of the nanocrystalline cellulose extracted in this way were comparable to those extracted from virgin sources (Ruiz-Caldas et al., 2022).

The environmental advantages of the cascade use of biomass materials can be evaluated with a Life Cycle Assessment (LCA). This is a standardized technique with which the environmental footprint is calculated for the production, use and end-of-life phase of a product, material or service. Different scenarios (e.g., recycling versus incineration) can be compared, but there can also be other aims, like finding production hotspots and strategic planning to reduce environmental impacts (Guinée et al., 2002).

The overall objective of this study was to investigate the environmental performance of a novel, nanocrystalline-cellulose production process that has three steps: depolymerization, rinsing and final bleaching. The LCA had several goals: (i) the identification of environmental hotspots during the product stage and finding opportunities for mitigation, (ii) the provision of a holistic environmental evaluation that can be used as a reference for comparisons of the LCA results with those of future studies and (iii) the comparison of the LCA results of this study with those of nanocrystalline-cellulose manufacturing reported in the literature.

2. Literature review

The number of LCA studies that consider nanocrystalline cellulose is relatively small; however, most of them are recent, so the number of studies is expected to increase. The available studies are mostly cradle-to-gate LCAs, meaning that they focus on the product stage. For example, they evaluate the different manufacturing systems (cf. de Nascimento et al., 2016; Leao et al., 2017; Zhang et al., 2022) or the different scale-up approaches (Carneiro and Rudrigues, 2022). These LCAs assess the environmental performance of cellulose nanocrystals extracted from various organic materials and via different production routes. While some studies focused only on the global warming potential (GWP) (Carneiro and Rudrigues, 2022; Husgafvel et al., 2016), others considered a few additional impact categories (de Nascimento et al., 2016; Gu et al., 2015; Zargar et al., 2022, etc.). However, none of these studies encompassed the entire set of impact categories listed in the corresponding LCA standards and handbooks.

Several studies discussed environmental hotspots in the nanocrystalline-cellulose production process, offering recommendations for process optimization to reduce the environmental footprint. Some, such as de Figueirêdo et al. (2012), proposed ways to enhance process yields, decrease energy consumption, and minimize water use, thereby reducing the environmental footprint of nanocrystalline cellulose. Another approach to mitigating environmental impacts involves adjusting the input volume and/or changing the reuse frequency of solvent solutions (Zargar et al., 2022).

In many LCA studies, various production processes for nanocrystalline cellulose were benchmarked. For instance, comparisons were made between the production of nanocrystalline cellulose using different source materials, such as the extraction from unripe coconut fibers versus the extraction from white cotton fibers (de Figueirêdo et al., 2012). This comparison highlighted the differences in energy and water consumptions, as well as the emissions impacting the environmental outcomes.

Another approach involved comparing the environmental performance of different extraction methods for producing cellulose nanocrystals from the same raw materials. These extraction methods can vary in the hydrolysis process itself, such as using diluted sulfuric acid, concentrated sulfuric acid, ammonium persulfate, and high-power ultrasound (de Nascimento et al., 2016). Alternatively, distinctions might arise in the pre-treatment (Leao et al., 2017) or the separation of sulfuric acid from a hydrolysate mixture (Zhang et al., 2022). The latter process can be executed through gravity settling, low-speed centrifugation, and ceramic-membrane microfiltration. This route was found to be the most environmentally sustainable due to its high recovery ratio (65.0 %), relatively low acid concentration, and moderate water consumption (Zhang et al., 2022).

Some studies tried to identify the most feasible process for nanocrystalline-cellulose production using a specific source material. An example is the evaluation of twelve extraction approaches from sugarcane bagasse fibers via acid hydrolysis, considering both the technical, i.e., yield and crystallinity, and environmental aspects, i.e., GWP and water footprint (Leao et al., 2017). Additionally, the feasibility of an LCA to enhance the environmental performance of nanomaterials during the innovation process was presented by de Nascimento et al. (2016).

Some studies aim for novelty by evaluating environmental impacts in the production of nanocrystalline cellulose from biomass source materials not previously addressed by others. Teh et al. (2019) conducted a study comparing three process routes using oil-palm empty fruit bunches as the source material. Two production routes were based on acid hydrolysis, and one was based on TEMPO oxidation.

Studies focusing on the use of nanocrystalline cellulose and the end-of-life stages of nanocellulose-based products remain scarce. Petrucci et al. (2018) conducted a cradle-to-gate LCA of a nanocellulose-based product, namely limonene plasticized poly(lactic acid) (PLA) film containing nanocrystalline-cellulose produced from *Phormium tenax* leaf fibers via acid hydrolysis. The study compared the environmental performance of this product with acetyl tributyl citrate plasticized PLA films containing organo-modified montmorillonite. Both products exhibited similar mechanical properties, and the LCA results demonstrated comparable environmental performance. The production of fillers based on nanocrystalline cellulose is energy intensive, which is a reason why the alternative product does not yield a much better environmental performance than the traditional product.

There are also studies of the environmental performance of nanocellulose production; however, these studies refer to nano-fibrillated cellulose (cf. Gallo Stampino et al., 2021; Hervy et al., 2015; Turk et al., 2020).

Compared to the aforementioned studies, our research introduces a novel, nanocrystalline-cellulose production process based on the liquefaction of biomass, specifically cotton fibers. While the existing scientific literature predominantly focuses on acid hydrolysis and considers only a limited number of impacts (focusing mostly on the GWP of the nanocrystalline cellulose), our study conducts a comprehensive assessment of a broader set of environmental impacts. This holistic evaluation

provides valuable insights into the environmental implications of the novel nanocrystalline-cellulose production process. Our research not only serves as a reference for future LCA studies on nanocrystalline-cellulose production routes but also highlights specific environmental hotspots during the product stage and proposes targeted measures for their mitigation.

By addressing these aspects, we aim to contribute to a broader understanding of the sustainability challenges associated with nanocrystalline-cellulose production, thereby providing valuable insights for future research and industrial applications.

3. Methods

3.1. Life-cycle assessment

The LCA was conducted in accordance with the principles and guidelines of international standards (ISO 14040, 2006; ISO 14044, 2006) using an attributional LCA approach, which focuses on assessing the environmental impact of a product or process at a specific point in time and typically considers the current state of technology and the existing system boundaries (Ekvall et al., 2016). The Ecoinvent database, which is integrated into the GaBi software, was used.

3.1.1. Description of the production process

Nanocrystalline cellulose is produced via the liquefaction of biomass with glycols. Methane sulphonic acid is used as a catalyst. The production process consists of three steps: (i) the depolymerization of lignin, hemicelluloses and the amorphous part of the cellulose fibrils, (ii) the centrifugation and rinsing of the reaction product with a mixture of glycols and water, and (iii) the final bleaching with sodium hypochlorite and rinsing with water.

The reaction conditions for the depolymerization process were chosen according to previous experience of the liquefaction of wood and similar biomasses (Kunaver et al., 2016). The mixture of diethylene glycol and glycerol was charged into a stainless-steel reactor equipped with an external heater, mixer and condenser. Methane sulphonic acid was then added as the acid catalyst (3 % wt., calculated on the amount of glycol). When the temperature of the mixture reached 150 °C, cotton linters were charged during the mixing. After the cotton linters were added to the preheated reaction mixture, the liquefaction process was carried out for 180 min and the reaction temperature was maintained at 150 °C. After that, the reaction product was cooled to room temperature and diluted with a mixture of diethylene glycol and water (ratio 1:1) and centrifuged at 8000 RCF (relative centrifugal force) for 45 min. The sediment was dispersed in a fresh solvent mixture using ultrasound (Hielscher UIP 1500hd). The sonification lasted 20 min and the suspension was centrifuged under the same conditions. This process was repeated three times.

In the next step, the sediment was dispersed in water and sodium hypochlorite was added. The mixture was sonicated for 30 min. After the sonification, the mixture was centrifuged at 8000 RCF for 30 min. The sediment was dispersed in water using ultrasound and again centrifuged under the same conditions. This process was repeated three times. The final sediment was pure nanocrystalline cellulose in water, with a concentration of 7–15 % (Kunaver et al., 2016).

3.1.2. Goal and scope of the LCA

The aim of this study was to evaluate the environmental performance of nanocrystalline cellulose produced on a pilot scale. The results of the novel production process were compared with the results of other available LCA studies on the environmental performance of nanocrystalline cellulose to evaluate the environmental sustainability of the novel process.

The **declared unit** is the production of 1 kg of dry nanocrystalline cellulose. The nanocellulose content in the produced solution is around 7 %, meaning that 1 kg of dry nanocellulose is obtained from 14.44 kg of solution. The declared unit is chosen over a functional unit due to the versatility of the product, which finds applications in many industries. Given the multifaceted uses of the product, a definitive functional unit is challenging to ascertain. Typically, the declared unit pertains to the product at the point of exit from the production chain, within the cradle-to-gate system boundaries (PCR, 2020).

The function of the system refers to the production of nanocrystalline cellulose, which can be applied in various industrial sectors, including the coatings industry, paper mills (for paper coatings), packaging industry (for food-packaging foils), polymer manufacturing (for polymer composites), pharmaceuticals (for drug carriers), and other sectors. Serving as an alternative material in these industries, nanocrystalline cellulose has the potential to substitute raw materials derived from fossil-fuel feedstocks such as crude oil.

3.1.3. System boundaries

In this study the cradle-to-gate LCA approach was employed, meaning that only the product stage is considered. In such a case the system boundaries include (i) the extraction and processing of the raw material, (ii) the production or synthesis of the ancillary materials and chemicals, (iii) the transport of all the materials/chemicals to the production site, (iv) the production and supply of the energy and water, and (v) the production of the nanocrystalline cellulose on a pilot scale. A schematic of the system boundaries is shown in Fig. 1.

The main reason for employing the cradle-to-gate LCA instead of a cradle-to-grave LCA is the lack of inventory data related to the downstream processes (e.g., the integration of the nanocrystalline cellulose into an end-product and the end-of-life treatment of such a product). The cradle-to-gate LCA approach is common practice in the case of building blocks and building materials with different application possibilities. Downstream processes differ depending on the specific application. As such, different scenarios need to be considered in the LCA. However, due to the lack of relevant data and the related large uncertainty, downstream processes can be omitted from the LCA, as is the case in the majority of other available LCA studies of nanocrystalline cellulose (cf. de Figueirêdo et al., 2012; de Nascimento et al., 2016).

3.1.4. Life-cycle inventory

Both primary and secondary data were employed to construct the life-cycle inventories. The majority of the data used were primary, obtained through direct measurements and collected by the producer of the nanocrystalline cellulose. These primary data include input requirements (such as raw materials, chemicals, and electrical energy) with specified quantities, as well as outputs (including intermediate products, amounts of recovered solvent, and volumes of wastewater). In contrast, secondary data cover the emissions associated with the incineration of diethylene glycol in a gas-turbine power plant. These emissions were not directly measured at the production site but were obtained from literature sources (Seljak et al., 2012, 2014).

The Ecoinvent 3.5 database was used to assess the environmental impacts associated with the production and processing of raw material (i.e., cotton fibers) and the production and synthesis of ancillary chemicals (i.e., glycerin, diethylene glycol, methane sulfonic acid and sodium hypochlorite) (Table 1). In addition, the Ecoinvent database was used to evaluate the impacts associated with the production and supply of electricity, water and wastewater treatment. All the applied Ecoinvent datasets are derived from a cut-off system model, with consideration given to aggregated (e.g., system process) datasets. The datasets are technically representative.

Moreover, market datasets that consider all the activities associated with the reference product in a particular geographical region, including average transports of that product within the geography and inputs of the product itself to cover losses in trade and transport (Ecoinvent, 2019), were taken into account with the Ecoinvent database. The advantage of the market datasets is that they provide a good approximation for a certain product within a specific region, which is

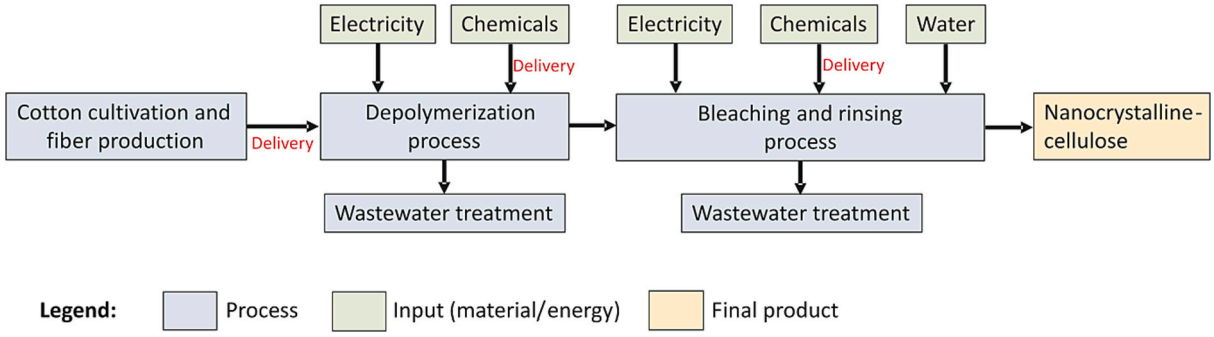

Fig. 1. System boundaries for the production of nanocrystalline cellulose.

particularly beneficial when there is a lack of information about the producer or supplier of the considered product.

3.1.4.1. Inventory data related to the reuse and combustion of diethylene glycol. Ancillary chemicals in the form of glycerin and diethylene glycol are crucial for supporting the reactions involved in the production of nanocrystalline cellulose (Fig. 2). The diethylene glycol used for rinsing can be partly reused in subsequent production series for both rinsing and depolymerization, effectively reducing the demand for this ancillary chemical from primary sources (Fig. 2). However, there are two potential procedures with distinct approaches to reusing diethylene glycol.

In the case of the baseline procedure, diethylene glycol from the initial rinsing cycle is reused in the depolymerization of cotton fibers. In contrast, the alternative procedure treats the diethylene glycol from the first rinsing cycle as a secondary fuel, combusting it on-site in a gas-turbine power plant that generates electrical energy (Fig. 2). For both the procedures the diethylene glycol from subsequent cycles can be reused for rinsing in the following production series.

According to the data in Table 1, the depolymerization process typically necessitates 3.42 kg of diethylene glycol per production series. Additionally, each rinsing cycle uses 22.0 kg of diethylene glycol and 32.05 kg of water, with three rinsing cycles required per series. Through the optimal reuse of the diethylene glycol over multiple production series, an average of 2.44 kg of diethylene glycol can be reused for depolymerization, thereby reducing the need to use diethylene glycol from primary sources (baseline procedure). Moreover, approximately 9.8 kg of the diethylene glycol from the second and third rinsing cycles can be reused for subsequent rinsing in multiple production series, leading to an average requirement of 12.24 kg of diethylene glycol from the primary source for rinsing in successive series.

The diethylene glycol reused in the depolymerization process is entirely consumed, while the diethylene glycol reused for rinsing exits the system as a water mixture that requires further treatment. Recovering the diethylene glycol from this mixture involves distilling the water, which consumes electricity. The recovered diethylene glycol characterized by a high calorific value (22 MJ/kg), serves as a secondary fuel that can be used in a commercial micro gas turbine with an assumed energy-conversion efficiency of 30 %. The electricity generated by the turbine is employed on-site, thereby reducing the reliance on grid electricity (Fig. 2).

3.1.5. Life-cycle impact assessment

The ReCiPe 2016 method (version 1.1) was applied to evaluate the environmental impacts associated with the production of nanocrystalline cellulose. ReCiPe provides harmonized characterization factors at the midpoint and endpoint levels (Huijbregts et al., 2017). In this study, only the results at the mid-point level are shown, as they are more reliable. A list of 19 impact categories is used in ReCiPe 2016. The methodologies and data used in the ReCiPe models are up to date with the latest scientific knowledge (Goedkoop et al., 2009). The Hierarchist perspective, which is based on the most common policy principles regarding the time frame, was applied in this study.

4. Results and discussion

Fig. 3 shows the relative contribution of the raw material, ancillary chemicals, electricity requirements and water consumption to produce nanocrystalline cellulose by means of the depolymerization, rinsing and bleaching of cotton fibers. Wastewater treatment and the combustion of secondary fuel in the form of diethylene glycol in a gas-turbine power plant (for electricity production) are also considered. The baseline procedure related to the reuse of diethylene glycol is taken into account (Fig. 2). Fig. 3 shows how the electricity requirements and glycerin contribute the most to the total environmental impact of the nanocrystalline cellulose, when considering the cradle-to-gate part of the lifecycle.

Electricity contributes more than 50 % of the burden to the impact categories associated with ionizing radiation, the pollution of aquatic ecosystems (freshwater ecotoxicity, freshwater eutrophication, marine ecotoxicity) and human toxicity related to cancer. Electricity also makes the largest contributions to the impacts affecting terrestrial acidification (48 % of the impact), the formation of fine particulate matter (PM) (46 % of the impact) and human toxicity related to non-cancer diseases (37 % of the impact) (Fig. 3). Electricity consumption is an environmental hotspot in the life cycle of nanocellulose; however, the share of electricity deriving from the different sources directly affects the results. The electricity mix of the Slovenian grid was considered, as the pilot production of the nanocrystalline cellulose takes place in Slovenia. The Slovenian grid consists of electricity that is produced in a nuclear power plant (26 %), followed by electricity produced in hydropower plants (20 %) and in a thermal power plant (15 %). About one-third of the electricity is imported from neighboring countries. As a relatively large share of this electricity derives from thermal power plants (in this case run on lignite), the consumption of electricity is associated with a relatively large share of greenhouse-gas emissions, sulfur dioxide emissions and PM emissions that have an effect on fine PM formation, human toxicity (cancer), terrestrial acidification and presumably also on global warming. The combustion of lignite in thermal power plants is directly associated with the depletion of fossil-fuel reserves.

Glycerin, which is required for the depolymerization process, is an environmental hotspot in terms of the marine-eutrophication potential (77 % of total impact), the stratospheric ozone-depletion potential (67 %), land-use potential (53 %), photochemical ozone-formation potential, affecting both human health and ecosystems, (a contribution of around 50 % of the total impact), metal-depletion potential (31 % of total impact) and terrestrial ecotoxicity potential (30 % of total impact). Glycerin also has an impact in terms of human toxicity (non-cancer) potential (30 % of total impact) and terrestrial acidification potential (24 % of total impact), representing secondary environmental hotspots (Fig. 3). Glycerin can be produced in two ways: either as a by-product of saponification and hydrolysis reactions in oleochemical plants or as transesterification reaction in biodiesel plants (Tan et al., 2013). In this

Table 1
Inventory data related to the production of 1 kg of dry nanocrystalline cellulose by depolymerizing cotton fibers, and then rinsing and bleaching of the reaction mass. For the mass balance, see Fig. 2.

Process	Unit	Quantity	Description	Source
Depolymerization				
Input				
- Cotton	kg	2.87	market for cotton fiber (global), system process dataset	Ecoinvent 3.5
- Glycerin	kg	8.0	market for glycerin (Europe), system process dataset	Ecoinvent 3.5
- Diethylene glycol (virgin and reused*)	kg	3.42	market for diethylene glycol (global), system process dataset	Ecoinvent 3.5
- Methane sulfonic acid	kg	0.33	market for methane sulfonic acid (global), system process dataset	Ecoinvent 3.5
- Electrical energy	kWh	11.42	market for electricity, low voltage (Slovenia), system process dataset	Ecoinvent 3.5
Output				
- Wastewater	kg	1.29	market for wastewater (Europe), system process dataset	Ecoinvent 3.5
- Reaction mass	kg	13.33	Intermediate product	/
Rinsing and bleaching				
Input				
- Reaction mass	kg	13.33	Intermediate product from depolymerization	/
- Diethylene glycol (virgin and reused* and **)	kg	22.00	market for diethylene glycol (global), system process dataset	Ecoinvent 3.5
- Sodium hypochlorite	kg	1.11	market for sodium hypochlorite, without water, in 15 % solution state (Europe), system process dataset	Ecoinvent 3.5
- Water for rinsing and bleaching	kg	65.38	market for tap water (global), system process dataset	Ecoinvent 3.5
- Electrical energy	kWh	80	market for electricity (low voltage, Slovenia), 53* kWh and 49.7 kWh** from the grid (system process dataset)	Ecoinvent 3.5
Output				
- Wastewater	L	33.33	market for wastewater (Europe), system process dataset	Ecoinvent 3.5
- Water evaporated into the atmosphere	L	32.05	/	/
- Diethylene glycol for reuse	kg	12.2*/ 11.0**	Reuse in subsequent production series	Ecoinvent 3.5
- Secondary fuel in form of diethylene glycol as 1 kg of dry nanocrystalline cellulose	kg	9.8*/ 11.0**	Combustion in gas-turbine power plant	Literature data: (Seljak et al., 2012, 2014).

* baseline procedure.
** alternative procedure.

study the environmental footprint of glycerin refers to the production process from epichlorohydrin, which is an organic compound. The use of pesticides to produce bio-based raw materials in upstream processes is most likely the reason for the dominant contribution of glycerin in several impact categories, considering the production stage of the nanocrystalline cellulose.

The other ancillary chemicals that have an impact on the overall environmental footprint of the nanocellulose are the diethylene glycol and the cotton fibers. **Diethylene glycol** contributes 48 % of the burden on the total impact regarding the GWP and 43 % of the burden on the total impact regarding fossil-fuel depletion potential, thus representing an environmental hotspot for these two impact categories. Its contribution to metal-depletion and terrestrial ecotoxicity potentials are also relatively high, i.e., 28 % and 30 %, respectively (Fig. 3). Diethylene glycol can be produced from fossil fuels (petroleum, natural gas, coal) or from biomass-based resources (Yue et al., 2012). In this study, diethylene glycol derived from fossil-fuel-based resources was considered, which explains its large contribution to the GWP and the fossil-fuel depletion potential, considering the production stage of the nanocrystalline cellulose.

The **cotton fibers** represent 77 % of the total impact in terms of water-consumption potential and 45 % of the total impact in terms of land-occupation potential. The high impact on water consumption comes from the fact that intensive cotton-crop farming requires irrigation water in the field. Cotton-crop farming is an intensive process anyway, as it requires inputs of seeds, mineral fertilizers, and pesticides. In this sense, the yield of cotton fibers is also a significant contribution to some other impact categories in the life cycle of the nanocellulose, such as ozone depletion (25 % of the total impact), metal depletion (18 % of the total impact) and marine eutrophication (16 % of the total impact) (Fig. 3).

The requirements for the **methane sulfonic acid** and **sodium hypochlorite** (ancillary chemicals) are relatively low compared to the consumption of glycerin and diethylene glycol or biomass. For this reason, the contribution of methane sulfonic acid and sodium hypochlorite to the environmental footprint of the nanocellulose is minor. Sodium hypochlorite only has a contribution to the photochemical ozone-formation potentials (POCPs), considering the discussed life cycle of the nanocellulose, contributing 13 % to the POCPs affecting ecosystems and 15 % to the total POCPs affecting human health (Fig. 3).

4.1. Scenario analysis: evaluation of the two procedures that differ in the reuse of diethylene glycol

The baseline procedure, as detailed earlier, differs from the alternative procedure in terms of diethylene glycol reuse. In the case of the alternative procedure, only primary-source diethylene glycol is used for depolymerization (Fig. 2), resulting in a 9 % increase in the use compared to the basic procedure.

This increased reliance on primary diethylene glycol is a notable drawback of the alternative procedure. However, the alternative procedure yields a larger quantity of recovered secondary fuel in the form of diethylene glycol during the manufacturing process (11.0 kg in the alternative procedure compared to 9.8 kg in the baseline procedure). This reclaimed diethylene glycol can be used in an on-site gas-turbine power plant, allowing for increased electricity generation, a major benefit compared to the baseline procedure.

The electricity produced within the gas-turbine power plant is subsequently used in the bleaching and rinsing processes of the cotton fibers, resulting in reduced electricity demand (from 53.1 kWh in the baseline procedure to 49.7 kWh in the alternative procedure) (Fig. 2).

Detailed comparative insights between the baseline and alternative procedures are outlined in Table 2. Notably, the discrepancies between the two procedures are relatively marginal, with variances of up to 3 % observed. The most significant disparity arises in terms of the GWP. This variance is primarily influenced by the differential demand for diethylene glycol from a primary source as this ancillary chemical notably impacts the GWP. The environmental burdens associated with the alternative procedure exhibit relatively higher impacts in certain categories, including the GWP, while showcasing relatively lower impacts in other categories, in contrast to the to the baseline procedure (Table 2).

Benchmarking the two procedures, a preference can be given to the

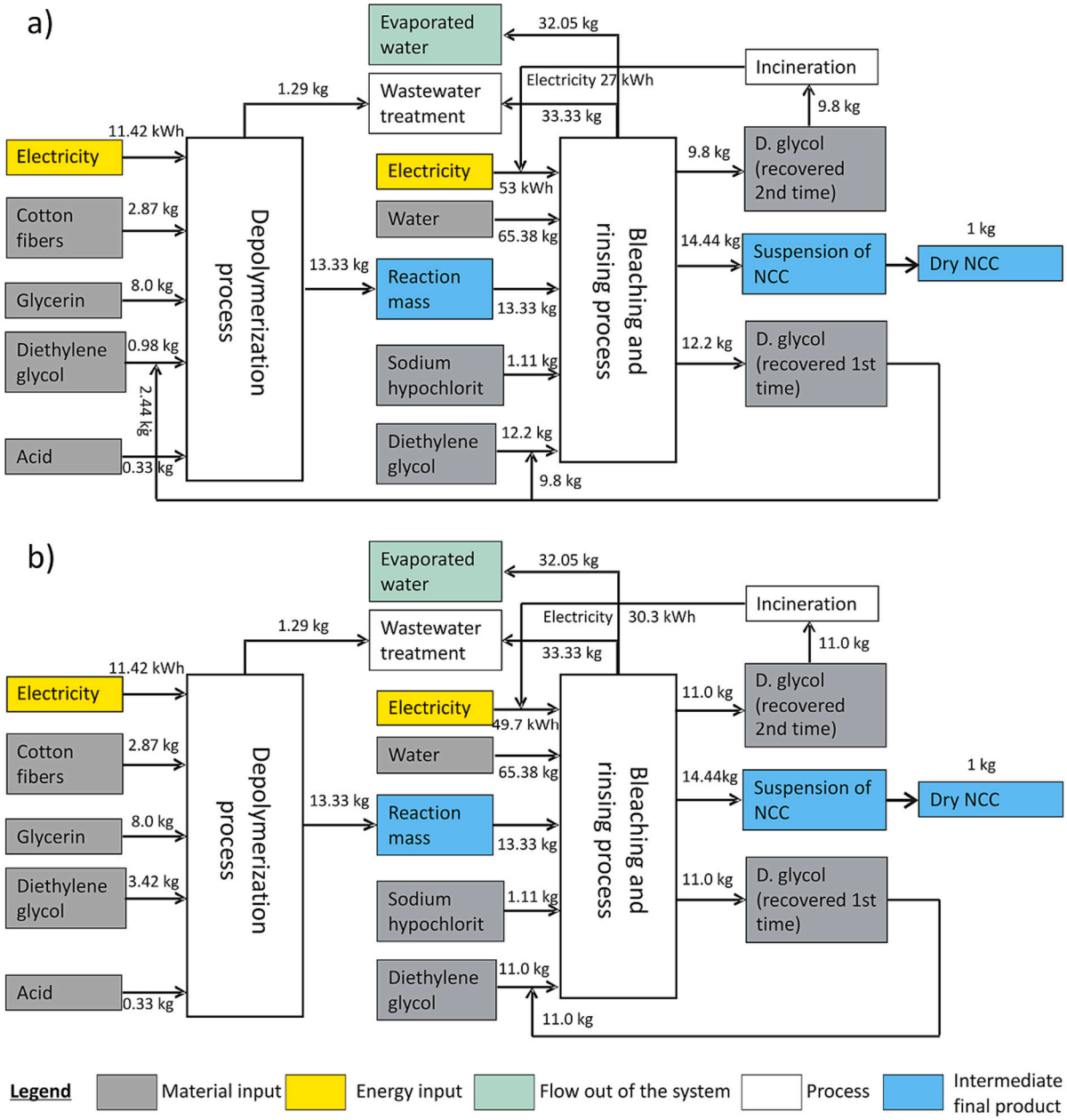

Fig. 2. Process flow chart for the two procedures with indicated mass and energy flows: baseline procedure (a) and alternative procedure (b). The procedures differ only in the reuse of the diethylene glycol recovered from the first rinsing cycle.

baseline procedure that exhibits fewer impacts in most of the categories, including the GWP (63.7 versus 65.9 kg CO_2 equivalent).

4.2. *Sensitivity analysis*

The purpose of a sensitivity analysis is to study the robustness of the results and their sensitivity to data (Wei et al., 2015). In this study, a sensitivity analysis was used to evaluate how the perturbation of selected inputs around a reference input value impacts the LCA results. Moreover, the sensitivity of the process yield to LCA results was also evaluated. A minimal process yield (e.g., 7 %) was considered in the evaluation of the nanocrystalline cellulose's environmental performance. However, the process yield can range up to 15 %, as proven during pilot-scale production.

As indicated in Fig. 3, diethylene glycol, glycerine, and electricity are the parameters that have the most impact on the results of the LCA. When considering the upscaling of the production process from pilot to industrial scale, the values of input-material and energy flows presented in this paper can be regarded as uncertain, to some extent. For this reason, selected parameters were considered to vary ±10 % from the central value measured by the manufacturer (pilot-scale production). The influence on the LCA results is shown in Table 3. Related to the hotspot analysis and the subsequent discussion, a 10 % change in electricity consumption has the highest impact on the LCA results. The impact category that is affected the most (e.g., 6.4 % change) is ionizing radiation, because around 26 % of the electricity in the Slovenian mix derives from its nuclear plant. The impact categories related to aquatic ecotoxicity and freshwater eutrophication are affected by 5–6 %. The formation of fine PM changes by 4 %, and the GWP (incl. biogenic carbon) by 3 %, mostly due to the electricity generation in thermal power plants (Table 3).

A 10 % change in glycerine requirements has the largest impact on marine eutrophication (7.7 % change) and stratospheric ozone depletion (6.7 % change). Land use and photochemical ozone formation

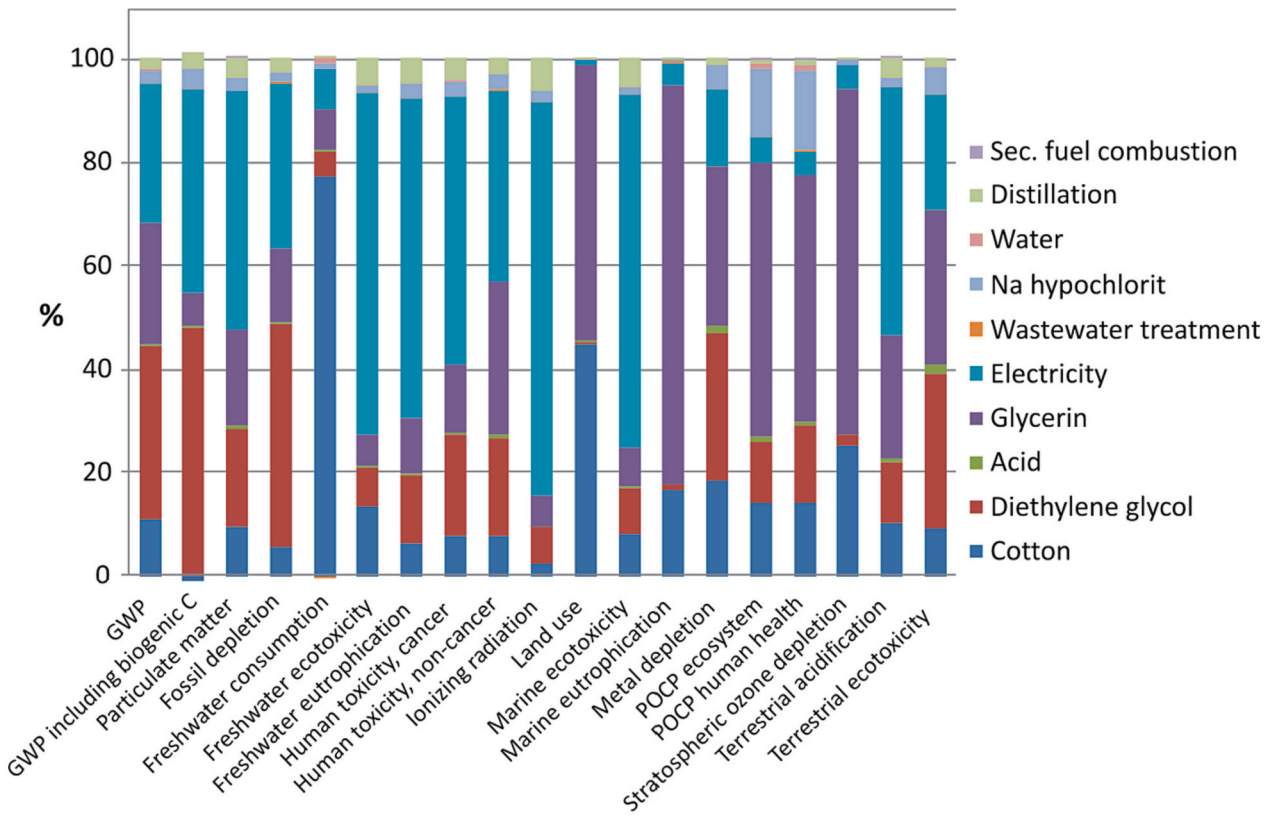

Fig. 3. Relative contributions of materials/chemicals and processes included in nanocrystalline-cellulose production to the environmental footprint. Contributions refer to baseline procedure.

(ecosystems and human health) are affected by about 5 %. While a 10 % alteration in the consumption of diethylene glycol from primary sources mostly affects global warming (incl. biogenic carbon) by 4.8 % and fossil-fuel depletion by 4.3 % (Table 3).

Considering the highest possible process yield achieved during pilot-scale production (e.g., an increase from 7 % to 15 %), the environmental impacts would reduce by a factor of 2.14 (a 214 % reduction) (Table 3). Thus, the process yield is the parameter that has by far the greatest influence on the results.

4.3. Comparative analysis

The environmental performance of the nanocrystalline cellulose produced by the depolymerization, rinsing and bleaching of cotton fibers was compared with the environmental performance of other nanocrystalline-cellulose manufacturing reported in the literature (Fig. 4 and Table 4). The results from the other studies were recalculated to the same functional unit, e.g., the production of 1 kg of dry nanocrystalline cellulose. In addition, the LCA results from the literature studies considered in the comparative analysis were calculated using the ReCiPe impact-assessment method (Hierarchist perspective). Exceptions are the study of Leao et al. (2017), who used the CML 2001 method to calculate the GWP; the study of Gu et al. (2015), who used the TRACI method; the study of Zhang et al. (2022), who used the IMPACT2002+ method; and the study of Teh et al. (2019), who calculated the impacts on the GWP and the human-toxicity potential based on characterization factors published in the work of Wenzel et al. (2000). Conversion factors from Dong et al. (2021) were taken into account to harmonise the LCA results from these studies with ReCiPe.

Fig. 4 illustrates the environmental impacts of 1 kg of nanocrystalline cellulose derived from various raw materials and produced using different processes. The results could only be compared in terms of up to five impact categories, as indicated in the figure. The considered studies covered only a very limited set of impact categories; only Gu et al. (2015) reported ten impact categories. de Figueirêdo et al. (2012) and de Nascimento et al. (2016) reported five impact categories, all other studies even fewer. Some studies reported only the impact on the GWP (see Fig. 4). In terms of the latter impact category, the differences between the studies are significant. The outlier (GWP is 1086 kg CO_2 equiv.) is nanocrystalline cellulose extracted from coconut fibers (de Figueirêdo et al., 2012), which is not indicated in Fig. 4. The energy intensity of the extraction process from coconut fibers directly corresponds to the source materials and it reflects in the environmental (GWP) footprint of the nanocrystalline cellulose. Various source materials (feedstocks) necessitate different cellulose-nanocrystal extraction methods, leading to disparities in electrical energy and water consumption. Additionally, process yield impacts the environmental footprint of the nanocrystalline cellulose; the yields vary among the studies. Moreover, the scale of the production (e.g., laboratory, pilot, industrial) influences the LCA results, with pilot and especially industrial productions being better optimized than laboratory production. In all the cited studies, the energy and material requirements are based on laboratory-scale processes. The exceptions are the studies of Gu et al. (2015), Carneiro and Rudrigues (2022) and Zhang et al. (2022), which refer to pilot production (Table 4).

In this study, the GWP (with biogenic carbon included) of the nanocrystalline cellulose is marginally lower (cca 64 kg CO_2 equiv.) than the average parameter value (cca 68 kg CO_2 equiv.) of the nanocrystalline celluloses indicated in Fig. 4 (outlier value 1086 kg CO_2 equiv., which is not shown in the figure, is excluded from the calculation of the average value). The inclusion of biogenic carbon in the LCA accounts for the carbon dioxide temporarily absorbed by plants during their growth, thereby incorporating the CO_2 sequestration in the biomass resulting from photosynthesis (Guest et al., 2013). The ReCiPe 2016 impact-assessment method reports GWP in two variants: with and without biogenic carbon, as shown in Table 2. Based on the available

Table 2
Comparison of environmental impacts for baseline and alternative procedures.

Impact category	Unit	Baseline procedure	Alternative procedure	Difference
Global warming, excl. biogenic carbon	kg CO_2 eq. to air	9.13E × 10^1	9.33 × 10^1	+2.1
Global warming, incl. biogenic carbon	kg CO_2 eq. to air	6.37 × 10^1	6.59 × 10^1	+3.3
Fine particulate matter formation	kg $PM_{2.5}$ eq. to air	2.33 × 10^{-1}	2.34 × 10^{-1}	+0.3
Fossil-fuel depletion	kg oil eq.	4.25E × 10^1	4.28 × 10^1	+0.8
Freshwater consumption	m^3	7.46	7.49	+0.4
Freshwater ecotoxicity	kg 1.4-DCB eq. to freshwater	2.98	2.94	−1.3
Freshwater eutrophication	kg P eq.	7.06 × 10^{-2}	7.01 × 10^{-2}	−0.7
Human toxicity, cancer	kg 1.4-DCB eq.	4.90	4.92	+0.3
Human toxicity, non-cancer	kg 1.4-DCB eq.	1.62 × 10^2	1.63 × 10^2	+0.6
Ionizing radiation	kg C-60 eq. to air	2.22 × 10^1	2.19 × 10^1	−1.5
Land use	m^2 × year annual cropland eq.	5.43 × 10^1	5.43 × 10^1	0.0
Marine ecotoxicity	kg 1.4-DCB eq. to marine water	3.50	3.45	−1.3
Marine eutrophication	kg N eq.	7.06 × 10^{-2}	7.06 × 10^{-2}	0.0
Metal depletion	kg Cu eq.	2.20 × 10^{-1}	2.24 × 10^{-1}	+2.2
Photochemical ozone formation, ecosystems	kg NOx eq. to air	5.46 × 10^{-1}	5.51 × 10^{-1}	+0.8
Photochemical ozone formation, human health	kg NO_x eq.	4.18 × 10^{-1}	4.22 × 10^{-1}	+1.0
Stratospheric ozone depletion	kg NO_x eq.	2.79 × 10^{-4}	2.79 × 10^{-4}	+0.1
Terrestrial acidification	kg SO_2 eq. to air	7.22 × 10^{-1}	7.19 × 10^{-1}	−0.4
Terrestrial ecotoxicity	kg 1.4-DCB eq.	1.96 × 10^2	2.00 × 10^2	+2.1

Table 3
Sensitivity of LCA results to a ± 10 % variation in selected input parameters (diethylene glycol, glycerine, and electricity) and an increase in process yield from the smallest measured value (7 %) to the largest measured value (15 %).

Impact category	Diethylene glycol ± 10 %	Glycerine ± 10 %	Electricity ± 10 %	Process yield change from 7 % to 15 %
Global warming, excl. Biogenic carbon	±3.4 %	±2.3 %	±2.3 %	−214 %
Global warming, incl. biogenic carbon	±4.8 %	±0.6 %	±3.3 %	−214 %
Fine particulate matter formation	±1.9 %	±1.8 %	±3.9 %	−214 %
Fossil-fuel depletion	±4.3 %	±1.4 %	±2.7 %	−214 %
Freshwater consumption	±0.5 %	±0.8 %	±0.7 %	−214 %
Freshwater ecotoxicity	±0.8 %	±0.6 %	±5.6 %	−214 %
Freshwater eutrophication	±1.3 %	±1.1 %	±5.2 %	−214 %
Human toxicity, cancer	±2.0 %	±1.3 %	±4.4 %	−214 %
Human toxicity, non-cancer	±1.9 %	±3.0 %	±3.1 %	−214 %
Ionizing radiation	±0.7 %	±0.6 %	±6.4 %	−214 %
Land use	±0.1 %	±5.4 %	±0.1 %	−214 %
Marine ecotoxicity	±0.9 %	±0.8 %	±5.8 %	−214 %
Marine eutrophication	±0.1 %	±7.7 %	±0.4 %	−214 %
Metal depletion	±2.8 %	±3.1 %	±1.3 %	−214 %
Photochemical ozone formation, ecosystems	±1.2 %	±5.3 %	±0.9 %	−214 %
Photochemical ozone formation, human health	±1.5 %	±4.8 %	±1.1 %	−214 %
Stratospheric ozone depletion	±0.2 %	±6.7 %	±0.4 %	−214 %
Terrestrial acidification	±1.2 %	±2.4 %	±4.1 %	−214 %
Terrestrial ecotoxicity	±3.0 %	±3.0 %	±1.9 %	−214 %

literature, the GWP (with biogenic carbon included) of nanocrystalline cellulose ranges from 4.7 (Husgafvel et al., 2016) to 207 (de Figueirêdo et al., 2012) kg CO_2 equiv. per kilogram of the nanocrystalline cellulose.

Comparing the LCA results from this study with other studies based on pilot-scale production, the nanocrystalline cellulose produced from cotton fibers via biomass liquefaction with glycols exhibits 130–215 % higher GWP impacts than the nanocrystalline cellulose produced from the same raw material via acid hydrolysis (Zhang et al., 2022). The difference can be attributed to the process yield, which was approximately 53 % in Zhang et al. (2022), but much lower (7 %) in this study. Moreover, production optimization influences the electricity consumption and the use of solvents and water. Electricity consumption, solvent usage (e.g., diethylene glycol), and other ancillary chemicals (e.g., glycerin) are the main contributors to the GWP of the novel nanocrystalline-cellulose production process. Upscaling both production processes to an industrial scale could potentially make the GWP footprints more similar.

The GWP of the nanocrystalline cellulose discussed in Gu et al. (2015) shows roughly half the impact of the nanocrystalline cellulose in our study and a similar impact to that in Zhang et al. (2022). Gu et al. (2015) used wood chips as source materials. Sodium hydroxide consumption accounted for most of the environmental impacts (contributing 55 % to GWP), with electricity consumption, sulfuric acid consumption, and water use also playing significant roles. The study by Gu et al. (2015) documented a process yield of 50 %. In contrast to other studies, Gu et al. (2015) highlighted impacts on the depletion of the ozone layer, photochemical ozone formation, the formation of fine PM, and the depletion of fossil fuels. Our study found these impacts to be higher than the study of Gu et al. (2015), with only the depletion of fossil fuels showing a lower parameter value. Ozone-layer depletion and photochemical ozone formation are predominantly influenced by glycerine (Fig. 3), leaving limited options for reducing these impacts, except through an enhancement of the process yield.

The impacts on human-toxicity potential, freshwater-eutrophication potential, and terrestrial acidification potential fall within a similar range, considering the different studies reporting these impact categories, although notable differences exist. It should be emphasized that only a few studies reported all these impact categories. The novel nanocrystalline-cellulose production process has a higher impact on the human-toxicity potential (166.4 kg 1.4-DCB equiv.) than other available studies (the average parameter value of the four available studies is 98.9 kg 1.4-DCB equiv.). Furthermore, the impacts on the eutrophication and acidification potentials are roughly twice the average parameter values of the other five available studies (e.g., 0.071 kg N equiv. versus 0.037 kg N equiv. for eutrophication and 0.72 kg SO_2 equiv. versus 0.32 kg SO_2 equiv. for acidification). The relatively high impacts on the human toxicity potential, eutrophication potential, and acidification potential of the nanocrystalline cellulose discussed in our study can be attributed

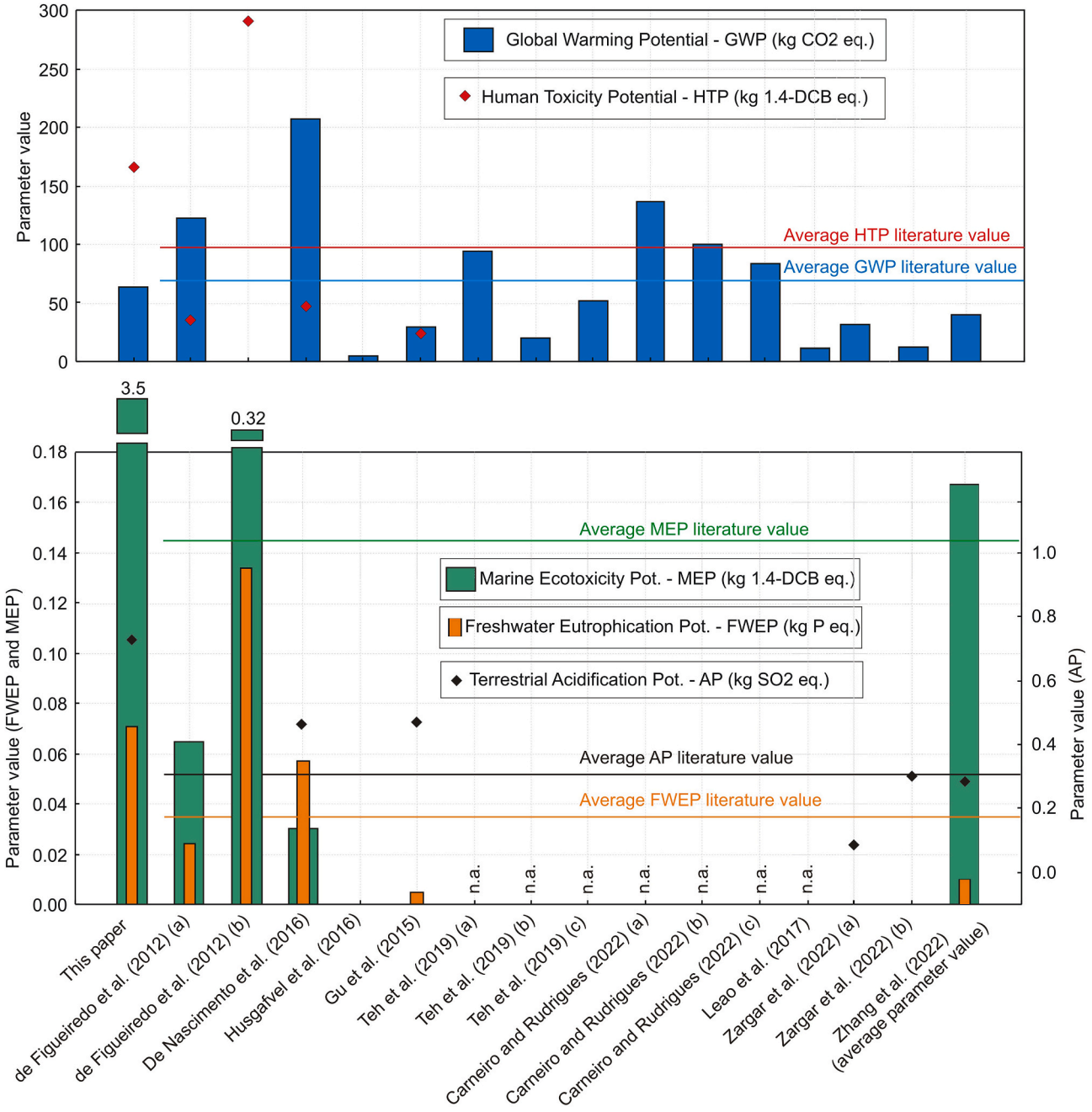

Fig. 4. Comparison of the environmental footprints of nanocrystalline celluloses in literature. The results are for 1 kg of nanocrystalline cellulose.

to the energy intensity of the production process and the relatively low process yield, ranging from 7 % to 15 %. Considering the conservative approach, we used the lowest-possible yield of 7 %. If the highest-possible yield of the pilot-scale process (15 %) were to be considered, the environmental footprints of the nanocrystalline cellulose would be halved (Table 3), aligning more closely with the footprints of nanocrystalline celluloses from studies conducted by Gu et al. (2015) and Zhang et al. (2022).

Regarding the impact on the marine-ecotoxicity potential, the novel nanocrystalline-cellulose production process has a high impact (e.g., 3.5 kg 1.4-DCB equiv.). Around two-thirds of this impact is linked to the electricity requirements in the production process. The average value of this parameter, considering five other available studies, is 0.18 kg 1.4-DCB equiv. The impact on marine-ecotoxicity potential appears to be the primary weakness of the novel production of nanocrystalline cellulose. Mitigation measures can only reduce this impact to a limited extent. Improving the process yield, potentially achievable at an industrial level of production, would be the most effective way to reduce the marine-ecotoxicity potential and all the other impacts.

5. Conclusions

The key findings and recommendations from this study provide valuable insights for key stakeholders and future research in the field: (1) Environmental Impact of 1 kg of dry nanocrystalline-cellulose production based on the novel process results in 63.7 kg CO_2 equivalent emissions, which is lower than the average value reported in previous studies. Solvent use, particularly diethylene glycol, was identified as the major contributor to the GWP and fossil-fuel-depletion potential. (2) Electricity requirements and glycerin production were identified as

Table 4
Raw materials used for production of nanocrystalline cellulose, production processes and scales of production processes in literature sources.

Study	Raw material	Production process	Scale of production
This paper	Cotton fibers	Depolymerization, rinsing and bleaching	Pilot
de Figueirêdo et al. (2012) (a)	Cotton fibers	Acid hydrolysis	Laboratory
de Figueirêdo et al. (2012) (b)	Coconut fibers	Acid hydrolysis	Laboratory
de Nascimento et al. (2016)	Coconut fibers	Acid hydrolysis	Laboratory
Husgafvel et al., 2016	Chemical pulp	Hydrolysis	Laboratory
Leao et al. (2017)	Sugarcane bagasse	Acid hydrolysis	Laboratory
Gu et al., 2015	Wood chips	Chemical acid hydrolysis	Pilot
Teh et al., 2019 (a)	Empty fruit bunch	Acid hydrolysis (chlorine bleaching)	Laboratory
Teh et al., 2019 (b)	Empty fruit bunch	Acid hydrolysis (chlorine-free bleaching)	Laboratory
Teh et al., 2019 (c)	Empty fruit bunch	TEMPO oxidation	Laboratory
Carneiro and Rudrigues, 2022 (a)	Mango shells	Process calculations method based on lab experiment	Laboratory
Carneiro and Rudrigues, 2022 (b)	Wood pulp	Adapted pilot-plant data	Pilot
Carneiro and Rudrigues, 2022 (c)	Mango shells	Adapted data from techno-economic analysis	n.a.
Zargar et al. (2022) (a)	Thermomechanical pulp	Acidic DES pretreatment	Laboratory
Zargar et al. (2022) (b)	Dissolving pulp	Acid hydrolysis	Laboratory
Zhang et al. (2022)	Cotton	Acid hydrolysis	Pilot

environmental hotspots in fifteen of the nineteen impact categories analyzed. The water-consumption potential was a hotspot in the production of cotton fibers as raw material. (3) A scenario analysis related to the evaluation of two procedures, which differ in the reuse of a solvent, showed no significant differences in terms of environmental impacts. However, a slight preference can be given to the baseline procedure with fewer impacts in most categories, including the GWP.

Based on our research work, we have the following future recommendations: (1) Key stakeholders should consider employing on-site renewable sources of electrical energy in the nanocrystalline-cellulose production process. This would help reduce the GWP associated with electricity consumption. (2) Shifting from fossil-fuel feedstock to biomass-based resources, such as diethylene glycol, would reduce both the GWP and the fossil-fuel-depletion potential of nanocrystalline cellulose. This recommendation highlights the importance of using sustainable and renewable resources in the production process. (3) Related to future research, our recommendations concern upscaling the production and a comprehensive LCA methodology. Further research should focus on upscaling the nanocrystalline-cellulose production process from pilot to industrial scale. This shift could reduce electricity consumption and enhance the process yield, both of which strongly influence the outcomes of the LCA. (4) Future studies could also expand the scope of the LCA to include a more comprehensive analysis of the nanocrystalline cellulose's life cycle. This would involve considering additional stages, such as the use stage and end-of-life scenarios, to gain a holistic understanding of the environmental performance.

A limitation of a comparative LCA analysis is the potential variance in the characteristics of cellulose nanocrystals extracted from diverse raw materials using different methods. Characteristics such as crystallinity, purity, particle size, and thermal properties can differ a lot. The downstream application of nanocrystalline cellulose can be contingent upon these specific characteristics. Consequently, comparing the environmental performance of nanocrystalline cellulose across different LCA studies becomes problematic without a comprehensive knowledge of their unique attributes and intended applications. To improve the transparency and relevance of LCA studies, future LCAs should be conducted together with the chemical structure, crystallinity, and morphology characterization of the extracted nanocrystalline cellulose, considering its specific field of application.

In conclusion, the LCA provided valuable insights into the environmental performance of nanocrystalline-cellulose production. The key findings underscore the importance of optimizing the production process to improve process yield and reduce electricity consumption, employing renewable-energy sources, and using bio-based solvents. These recommendations can guide key stakeholders in improving the environmental sustainability of nanocrystalline-cellulose production and lay the groundwork for future research.

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CRediT authorship contribution statement

Katja Malovrh Rebec: Methodology, Formal analysis, Visualization, Writing – original draft, Writing – review & editing. **Janez Turk:** Methodology, Formal analysis, Visualization, Writing – original draft. **Matjaž Kunaver:** Data curation, Investigation, Supervision, Writing – original draft.

Declaration of competing interest

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

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