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Reference:
Nadeem Humayun, Nimmegeers Philippe, Batchelor Warren, Billen Pieter.- Cellulose nanofibre films as a substitute for plastic packaging : a comparative environmental life cycle assessment
Food and bioproducts processing - ISSN 1744-3571 - 145(2024), p. 175-186
Full text (Publisher's DOI): https://doi.org/10.1016/J.FBP.2024.03.005
To cite this reference: https://hdl.handle.net/10067/2050150151162165141

Cellulose nanofibre films as a substitute for plastic packaging: a comparative environmental life cycle assessment

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ABSTRACT

Accumulation of synthetic food packaging in the natural environment has increased considerably in last few decades, posing serious concerns to human and aquatic life. Hence, substituting synthetic packaging with biobased packaging is one viable option. Cellulose nanofibres can easily be transformed into films, which could be a suitable alternative for petroleum-derived packaging. However, production of these films consumes a significant amount of energy as compared with the synthetic packaging. The main contributors for this high energy consumption are associated with the treatment of pulp, production of fibres and drying of films. This study analyzes the environmental impacts (in terms of energy and water) of spray deposited cellulose nanofibre films and its key drivers through an attributional cradle-to-gate life cycle assessment. Different scenarios were formulated and studied in terms of waste feedstock choice, solid content, forming composite of cellulose nanofibres, and location

(Australia, China, Germany, Great Britain and United States of America). The results indicate that the environmental impacts of spray deposited cellulose nanofibre films based on a baseline scenario for a large-scale production were still higher as compared with petroleum-based synthetic packaging. However, establishing a recommended scenario from the studied scenarios could lower the environmental impacts significantly (two to five times) in comparison to synthetic food packaging. This life cycle assessment model has the potential to support the reduction of the environmental impact of the studied green chemistry option and to accelerate the transition towards more sustainable packaging.

Keywords:

Cellulose nanofiber films; Large-scale production; Life cycle assessment; Spray deposition; Synthetic packaging

1. Introduction

The use of food packaging has risen significantly in recent decades and with a 5% annual growth, the global packaging food market is estimated to reach $3.4 trillion by 2030 [1]. The use of synthetic plastics in food packaging is currently considered as an essential part of our lives owing to their remarkable features such as their resistance to erosion, durability, low cost and ease of manufacturing [2]. Unfortunately, owing to their extended durability, these petroleum-derived packaging materials remain in the environment as waste, resulting in water, air, and soil pollution [3]. Recycling is a viable option, but being a mixture of different synthetic polymers, causing a reduction of properties and quality of the recycled material [4]. Therefore, the biobased polymers have gained considerable attention in last few decades owing to their sustainability aspects and development and diffusion of new green technologies for a biobased economy [5]; however, they possess limited mechanical and barrier properties [6]. One type of biobased polymers is cellulose, which is the most abundant organic polymer on earth. Cellulose-based packaging is biodegradable, renewable, and recyclable, but has poor moisture and oxygen barrier properties [7]. Therefore, cellulose is fibrillated into nanomaterials referred to as nanocellulose, possessing distinguishing characteristics such as non-toxicity, high surface to volume ratio, strength, and recyclability [8, 9]. Nanocelluloses are generally divided into three main types such as cellulose nanofibres (CNF), cellulose nanocrystals (CNC) and bacterial nanocellulose (BNC) [10]. CNF can be produced via mechanical defibrillation, while CNC and BNC types can commonly be synthesized via acid hydrolysis of cellulose and the use of bacteria or enzymes respectively [11, 12]. However, the scope of this study is limited to CNF as among all nanocellulose, only this type could be produced at a competitive cost as compared with the synthetic polymers [13].

The CNFs can easily be transformed into films: exhibiting translucency, strength, excellent oxygen barrier permeability and reasonable moisture barrier performance [7, 14, 15]. Casting and vacuum filtration are the conventional laboratory methods to produce CNF films; however, these processes have serious limitations [7]. The CNF films produced via casting usually require long drying times, while lacking uniformity in basis weight and thickness [16]. On the other hand, films fabricated via vacuum filtration suffer from separation constraints as removing the films from the filter papers is a tedious task [17]. Spray deposition has emerged as a potential alternative to conventional methods owing to the speed, simplicity, and continuity of the process [7, 18]. The spray system works by spraying a suspension using a spray pump onto a base surface placed on a moving conveyor belt. The basis weight of the films can easily be controlled either via altering the velocity of the conveyor or changing the suspension concentrations [7, 19].

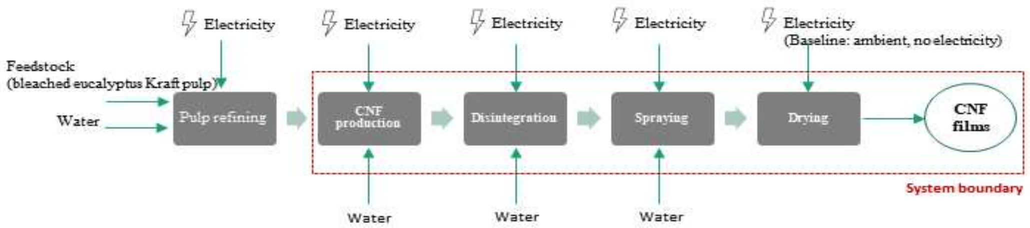
The CNF films require high energy consumption mainly associated with the production of fibres needing up to 75,000 kWh/tonne to fibrillate the cellulose via mechanical treatment [13, 18, 20]. Subjecting CNF fibres to pre-treatment via different methods such as carboxymethylation, enzymatic hydrolysis, catalytic oxidation using 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) is useful in reducing the energy requirements to a significant extent depending on the treatment employed, but all methods have certain drawbacks such as cost, toxicity and corrosiveness [21, 22]. For example, TEMPO oxidation resulted in a reduction of energy consumption up to 100 times during the mechanical disintegration process; however, at an expense of high cost and toxicity [23]. Similarly, the enzymes are expensive, while the use of acids imparts corrosiveness to the process . Presently, there are a limited number of LCA studies available in the literature on the production of CNF fibres [24-27]. According to these studies, the environmental impacts of the CNF fibres are high compared to synthetic polymers. Surprisingly, to date, only one LCA study exists in the literature highlighting the production of CNF films [13]; however, the focus of the research is on laboratory scale production of these films. Considering the ongoing research interests towards CNF films as substitute of synthetic packaging, LCA is an important tool to analyse the performance of this plant-based films compared to other alternatives currently in use in the food packaging. Additionally, to the best of our knowledge, there is no LCA study available in the literature for simulated large-scale production of these films that can be used as a comparison with synthetic polymers used in packaging applications.

The main aim of this study is to evaluate the life cycle environmental impacts (in terms of energy and water) of CNF films for a batch of 1 tonne production. The secondary aim is to investigate the effect of feedstock, electricity mix, increasing solid content, forming composites, commercial heat drying and recycling of fibres to produce these films through a scenario analysis. Additionally, based on the LCA results, a recommended scenario is also suggested for future commercial production of CNF films. Heat losses for the overall process are assumed to be negligible. The reproducible LCA study/model that has been developed can serve as a tool to accelerate the transition towards more sustainable biobased food packaging.

2. Materials and Method

2.1. Goal and Scope Definition

LCA is a referred to as a comprehensive, structured, and globally recognized ISO standard method (ISO 14040, ISO 14044) [28, 29]. The main goal of this study is to evaluate the cradle-to-gate environmental impacts of CNF films for a batch of 1 tonne production. The attributional cradle-to-gate LCA starts from the bleached eucalyptus Kraft (BEK) pulp to produce CNF. The unrefined BEK pulp consisted of fibres having length of 1 mm and an average diameter of 10–15 μm was supplied from Australian Paper Maryvale [30]. The baseline scenario constitutes of converting pulp to fibres via refining, followed by disintegration, spraying and drying at ambient conditions to produce the CNF films in a research facility in Australia (AU). The data for the production of refined fibres was obtained from Nadeem et al. [13] and Arvidsson et al. [24]. The fibres' suspensions were disintegrated and transformed into films via spray deposition followed by commercial drying of films. Another objective of this assessment is to highlight those scenarios for the production of films that may have a substantial influence on the total environmental footprints of the product. The impact of feedstock (eucalyptus and waste carrots), drying (ambient and drying via tunnel), five countries in four different continents, increasing the solid content (1.5 wt.% and 4.5 wt.% using stirring via spraying), and forming composites with carboxymethyl cellulose (CMC) on the fabricated films were determined and compared with the baseline scenario (AU) in a scenario analysis. The selected scenarios considered to have a major impact on the production of films and were selected based on experimental work and research being carried out on CNF films. Eucalyptus is chosen as it is native to Australia and categorized as hardwood; however, requiring quite high energy consumption for defibrillation of fibres. Additionally, waste carrots (a representative of vegetable waste) were chosen owing to the reason that they have successfully been converted to fibres, requiring four times less energy compared to the conventional processes [31]. For desired applications, commercial juice bars or food processing plants can be used as sources for carrot pulp at a small scale. Additionally, biorefineries uses organic waste to produce biofuels and bioenergy and thus could also be a source of this waste product. The production protocol was assumed to be the same as used by Varanasi et al. [31]. Briefly, the waste carrots were initially shredded followed by blending to produce a pulp. The pulp mixture was then subjected to blanching via heating at 80 °C for one hour with a continuous stirring to ensure a uniform heating. The mixture was allowed to cool and then vacuum filtered to remove the excess liquid. After filtration, the dried pulp was subjected to refining to produce fibres. The fibres obtained from carrot waste pulp had an average aspect ratio (ratio of length to diameter) of around 288. The films produced from these fibres showed high strength and exhibited translucency. The range of solid contents were selected based on experiments as the suspensions having less than 1.5 wt.% were quite watery and did not make a good film, while the suspensions were not sprayable at higher solid concentrations (>4.5 wt.%) (Appendix B). The LCA conducted by Nadeem et al. was focused on ambient conditions and oven drying of the CNF films; however, none of them are considered feasible for large scale production [13]. Therefore, the impact of commercial drying via a 19 kW Infrared drying tunnel GW-1200H (Dongguan Hoystar, China) having dimensions 5500×1300×1500mm was chosen in current LCA study. The different electricity mixes were chosen to consider the impact across various continents. Increasing the solid content reduced the water content, thus resulting in less energy consumption for drying [32]. Hence, increasing solid content could be a potential tool to reduce the drying time of films and consequently, the energy consumptions associated with it. CMC was chosen to produce the composites with CNF in this study as CNF/CMC films already showed good results in terms of mechanical and barrier properties. Additionally, the resultant films indicated decrease in embodied energy values owing to approximately three times lower environmental impacts associated with CMC compared to CNF [14]. Figure 1 shows the system boundary for the overall LCA used in this research. It includes a cradle-to-gate analysis for the conversion of CNF fibres into films. According to the system boundary for the fabrication of CNF films, the first step includes mechanical disintegration of the fibres' suspension, followed by spraying via a spray system and commercial drying in a tunnel dryer respectively. The waste types generated during the whole process were the sprayed suspension and wastewater for cleaning from spraying; however, they could easily be collected and recycled to films again via disintegration and spraying [6, 33]. The following midpoint environmental impact indicators have been calculated to cover a broad range, using the ReCiPe 2016 method and 1 tonne dry CNF films as functional unit: global warming potential (GWP), water depletion (WD), ozone formation (OF), terrestrial acidification (TA) and respiratory effects in terms of fine particulate matter (PM).



No.	Feedstock	Drying	Country	Solid content (wt. %)	Recycling	Composite
1 (Baseline)	BEK	Ambient	Australia	1.5	No	No
2	Waste Carrot	Ambient	Australia	1.5	No	No
3	BEK	Drying Tunnel (Infrared)	Australia	1.5	No	No
4	BEK	Ambient	China	1.5	No	No
5	BEK	Ambient	Germany	1.5	No	No
6	BEK	Ambient	Great Britain	1.5	No	No
7	BEK	Ambient	United States	1.5	No	No
8	BEK	Ambient	Australia	4.5	No	No
9	BEK	Ambient	Australia	1.5	Yes	No
10	BEK	Ambient	Australia	1.5	No	Yes

Figure 1: Cradle-to-cradle LCA system boundary (represented by the dotted line box) for large-scale CNF film production plant with studied scenarios.

2.2. CNF Films Production Processes

The methodology for the production of refined CNF fibres in this analysis was the same as adopted by Nadeem et al. [13]. A flowchart describing the studied production scenarios is shown in Figure 2. Briefly, 10 wt. % unrefined BEK pulp was treated via an industrial disc refiner, LC Aikawa Canada (112 kW). The refined CNF fibres were suspended in water (1.5 wt.%), followed by disintegrating in a 45 kW industrial disintegrator having a Buster model (Sukimar Engineering, India). The disintegrated suspension was then introduced to a spray system constituting of a Wagner spray pump (0.56 kW) Model F230 (Germany), a 0.38 mm orifice Wagner spray nozzle Type 315 (Australia) and a Schroder custom conveyor (Australia) having dimensions of 3198 mm×728 mm×839 mm (power consumption: 0.75 kW). [7, 19]. The suspensions were sprayed onto stainless steel plates (roughly 159 mm diameter) moving on a conveyor belt. The conveyor belt was set at 0.65 cm/s and the height of the nozzle tip from the top of steel plates was around 30 cm. Finally, the suspensions sprayed on plates were subjected to drying at ambient conditions for 24 hours, resulting in CNF films. Based on experiments, it is suggested that around 6 kg of CNF films could be produced for a batch of 1 tonnes.

2.3. Function and Allocations

CNF is an emerging material and therefore, a number of uncertainties exists regarding its technical aspects. Assessing emerging and new materials in terms of LCA is quite challenging owing to many reasons such as data unavailability, limited literature comparison with existing materials, and inadequate knowledge regarding the impacting factors for scale-up etc. [34, 35]. For these concerns, a scenario analysis of such materials would be helpful in analyzing their future environmental impacts. Additionally, analyzing and using literature data, conducting interviews with experts in the field and appropriate modelling techniques could also be helpful in this regard [36]. Table 1 displays the energy inputs required for conducting the LCA in a baseline scenario and their respective sources.

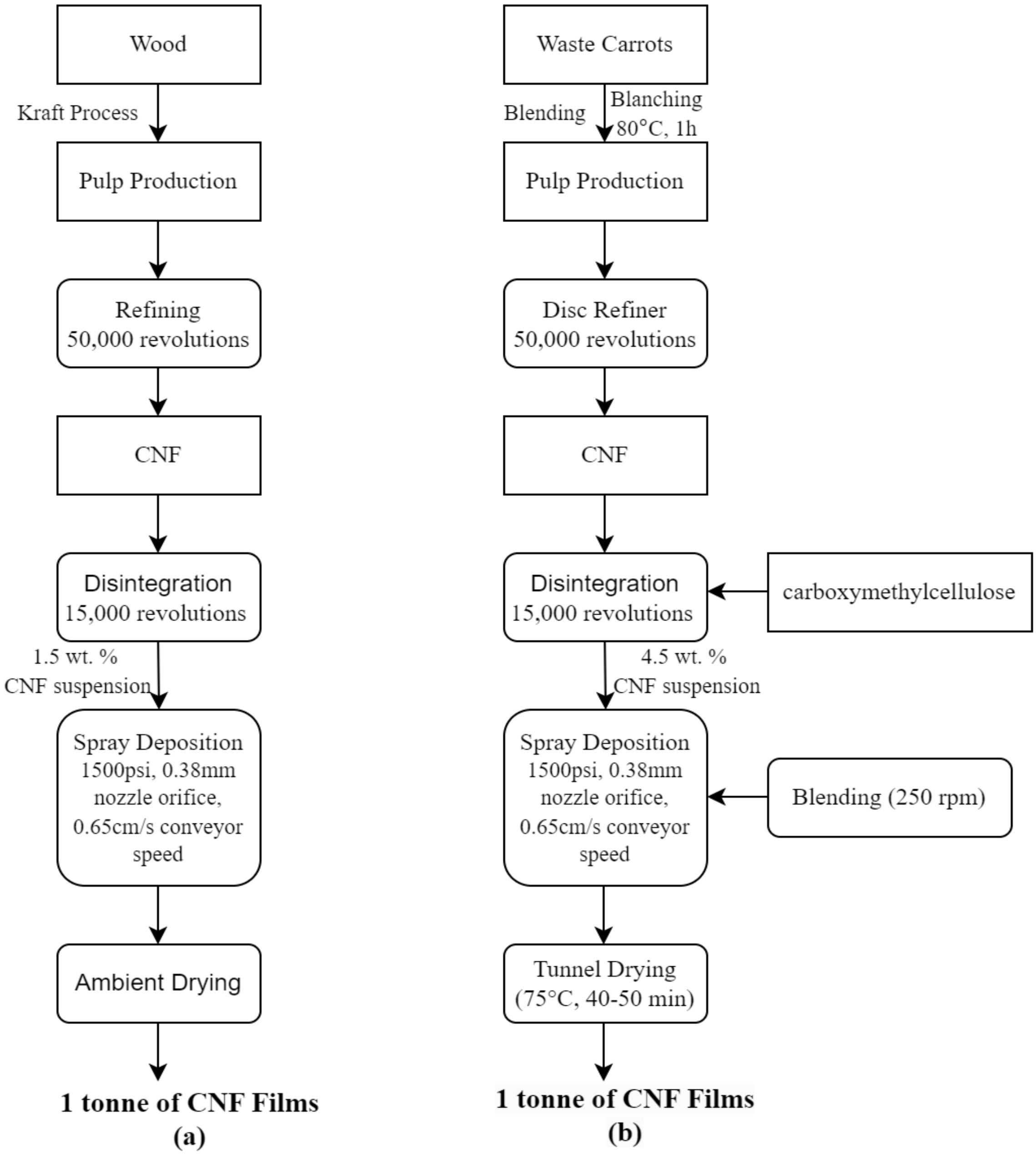


Figure 2. Flowchart of the studied system, showing the two studied production scenarios (a) Baseline scenario, (b) Recommended scenario

Table 1: Energy Inventory for 1 tonne of spray deposited CNF films in a Baseline Scenario

Spray Deposition Per Film		
Activity	**Baseline Scenario**	**Reference**
Pulp	21500 MJ	[24]
Pulp Treatment (Electricity)	4000 MJ	[24]
Refining	7485 MJ	[23]
Disintegration	1728 MJ	[13]
Spray Pump	400 MJ	[12]
Belt Conveyor	600 MJ	[12]
Ambient Drying/Heat Drying	14000 MJ	[12]

2.4. Life cycle Inventory

The impacts per unit (kg, L, etc.) for the fabrication of films were acquired from Ecoinvent (version 3.6) and finally being modelled via openLCA 2022. The main sources of attaining the data for this study are Nadeem et al. [13], Arvidsson et al. [24], Varanasi et al. [31], Ang et al. [30], from estimations and experimentally measured information and unpublished work as part of this project. The life cycle inventory of this study is supplemented in the supporting information as Appendix A.

2.4.1. Foreground Data

Foreground data are measured from the equipment at the production facility and used directly for this study. For instance, the mechanical processing is directly measured in terms of electricity and being calculated as a product of operation time and power ratings of the equipment.

2.4.2. Background Data

The modelling of the electricity mix (in kWh) is done within Ecoinvent database v.3.9.1 (2022) (AU). Water used in this study is also modelled using the inventory database via Ecoinvent database v.3.9.1 (AU) in terms of litre. The production of kraft pulp and its treatment is based on the data used by Arvidsson et al. [24]. The consistency of the refined pulp was the same in this study as used by Ang et al. [30]. The production of refined CNF fibres is considered the same as reported by Nadeem et al. [13] with few modifications for large scale production of the films. For instance, a low consistency (accommodating up to 6 wt.% solids) pilot scale disc refiner (120 kW) has been considered for analysis in this study [37]. The disc refiner is powered by a 112-kW motor and uses a 30-kW centrifugal pump. The specific edge load was kept at 0.6 J/m by using a plate of bar length 2.74 km/rev rotating at a horizontal speed of 1200 rpm. Similarly, the disintegration is considered to be conducted in a large-scale disintegrator (45 kW) from Sukumar Engineering. The production of CNF films is assumed to be carried out in a spray system used by Nadeem et al. [13].

2.4.3. Interpretation

The uncertainty in the production of pulp from wood in terms of yield was estimated to be approximately 5% as suggested by Arvidsson et al. [24]. Similarly, water used for the washing an equipment was also estimated based on actual observation and the considered uncertainty was approximately 10%. Considering the drying of films at ambient conditions, it is assumed that films were kept in a covered area built by green tiles [13, 38]. Additionally, an assumption was made based on experimental data i.e., high solid contents are sprayable via subjecting the suspensions to continuous stirring in a hopper during the spraying process. Increasing the solid contents results in decreasing the amount of water to be dried and consequently, reducing the drying time of the films [32].

2.5. Impact Scenarios

Six impact scenarios that could possibly influence environmental impact of the production of CNF films are considered in this study.

2.5.1. Feedstock

Cellulose is regarded as the most abundant organic polymer on earth that accounts approximately 40% of a lignocellulosic biomass [39]. Cellulose was fibrillated mechanically resulting in reducing the diameter of fibres and breaking the strong hydrogen bonds between its chains to produce a biopolymer referred to as CNF [40]. Briefly, a pulp was produced from wood via pre-treatment and then resulting in the production of CNF fibres via mechanical treatment. A number of studies has highlighted the use of high energy consumption in producing CNF (5-250 MJ/kg) depending on the quality and properties of resulting fibres [18, 20, 24, 25]. Therefore, the environmental impacts of CNF films were mainly associated with the production of nanofibres [13, 14]. Recently, Varanasi et al. suggested that the use of waste feedstock for the production of fibres has resulted in approximately four times less energy consumption compared to conventional mechanical defibrillation processes [31]. Considering this aspect, the impacts of two different feedstocks have been analysed in this study. These include the use of Eucalyptus as highlighted by Nadeem et al. [13] and using waste carrots as suggested by Varanasi et al. [31] to produce refined CNF fibres.

2.5.2. Transportation

The impacts of transportation are mostly considered as negligible when the materials are transported in bulk. It is assumed that all the materials (waste carrots, pulp, CMC) are transported within the range of 80-90 km from the production facility by light commercial vehicle EURO 2006.

2.5.3. Electricity Mix

Given the high electricity input of the process, the type of electricity mix could significantly influence the environmental impacts for a large-scale production of films. To cover a broader range, environmental impacts of four different continents have been analyzed in this assessment. The five countries chosen in this LCA are China (CN), United States (USA), AU, Germany (DE) and Great Britain (GBR). The data used in this analysis was obtained from the Ecoinvent version 3.9.1 (2022) [41].

2.5.4. Drying

It is experimentally observed that the dimensional stability of the CNF films could be compromised when subjected to heat drying [42, 43] and hence, they are usually dried at ambient conditions requiring at least 24 hours [7]. Additionally, drying at ambient conditions requires a considerable space and thus contributes to the environmental impacts [7, 19]. However, these limitations are a huge constraint regarding the large-scale production of these films. Recently, it is revealed that CNF films could be heat-dried without compromising the dimensional stability [15]. The LCA conducted by Nadeem et al. [13] highlighted that drying of the films in an oven resulted in a 7-8 times higher energy consumption and global warming potential as compared with the ambient dried films. However, using a fast and continuous drying process with high production capacity could potentially reduce the environmental impacts associated with these films. Considering this aspect, two scenarios are considered for this LCA study i.e., drying at ambient conditions and using a commercial drying tunnel that would dry around 6 kg of films in 2 hours.

2.5.5. Solid Content

Percentage of solids in suspension could significantly influence the environmental impacts for large-scale production as increasing the solids concentration in a suspension could potentially result in less reaction water and consequently faster drying time [32]. However, spraying of the CNF fibres at higher percentage of solids is quite a challenging task owing to its increasing viscosity [44]. To make the suspension sprayable at a high solid content, a Eurostar mixer (Germany) was added with the hopper (Appendix B).

2.5.6. Composites Production

Substituting the portion of CNF with comparatively low impact materials and forming composites could be advantageous in terms of their life cycle impacts, considering the high energy consumption of CNF production [22]. One popular choice regarding forming composites is the use of additives such as CMC, that can be produced via subjecting cellulose dispersion in alkali followed by acid treatment, resulting in substitute hydroxyl groups of glucose [45]. CMC was chosen in this analysis owing to its characteristics such as heat resistance, rheology modifier and low environmental impact [14, 46]. Additionally, the CNF/CMC composite produced by Nadeem et al. showed mechanical and barrier properties comparable with synthetic packaging [14].

3. Results and Discussion

3.1. Baseline Scenario

The results for the baseline scenario for cradle-to-gate LCA of CNF films and their corresponding life cycle inventory are shown in Table 2 and Figure 3 respectively. The environmental impacts were calculated based on the published work by Nadeem et. al with few exceptions for large-scale production [13]. As seen from Figure 3, the environmental impacts for a functional unit of 1 tonnes of CNF films are as follows: global warming potential (GWP) of 5067.0 kg CO_2 eq., water depletion (WD) of approximately 294.0 m^3, ozone formation (OF) of 2.4 kg NOx eq., terrestrial acidity (TA) of 0.9 kg SO_2 eq. and particular matter (PM) of 0.3 kgPM2.5 eq. in a baseline scenario. The environmental impacts in this case study are only driven by energy and water use. Therefore, the GWP, OF, TA and PM have the same fractional distribution over the different stages. However, for water use there is a slight difference. This is also clear from Figure 3 and more details are summarized in Table S1. It is observed that the mechanical pretreatment and fibers' defibrillation consume a considerable electricity and are mainly contributing to high environmental impacts associated with CNF films [13]. Additionally, the environmental impacts per tonne of CNF films calculated in this LCA study for a large-scale production in terms of GWP were approximately 70% lower than the values reported in the literature [13]. Considering the energy inventory for the fabrication of films, the production of pulp and its pretreatment is the most energy intensive step accounting for more than 50% of the total energy consumed, followed by drying of films (28%) and defibrillation of fibres (18%) respectively. As expected, the production of CNF fibres predominately contributing to the energy consumption (approximately 70%) in producing films [13, 14]. Nevertheless, the environmental impacts for these refined CNF films were still 0.5-1.5 times higher than conventional synthetic packaging films [47].

Table 2: Life cycle inventory for the baseline scenario

	Category	From	Database	Quantity	Unit	Reference
Material	Wood	-	-	4	ton	[48]
	Pulp	-	-	10	ton	[13, 30]
	Water (Reaction)	-	-	275000	L	-
	Water (Washing)	-	-	110	L	[13]
	Water (Refining)	-	-	19000	L	-
Transportation	Pulp		Ecoinvent	3.44×10^{-1}	MJ	[13, 49]
Electricity	Electricity (production) mix	AU				[50]
	Pulp Treatment	AU	Ecoinvent	25500	MJ	[24]
	Refiner	AU	Ecoinvent	7485	MJ	
	Disintegrator	AU	Ecoinvent	1728	MJ	
	Spray Pump	AU	Ecoinvent	400	MJ	
	Belt Conveyor	AU	Ecoinvent	600	MJ	
	Drying	AU	Ecoinvent	14000	MJ	[38]
Product	Nanocellulose Film	-		1	ton	

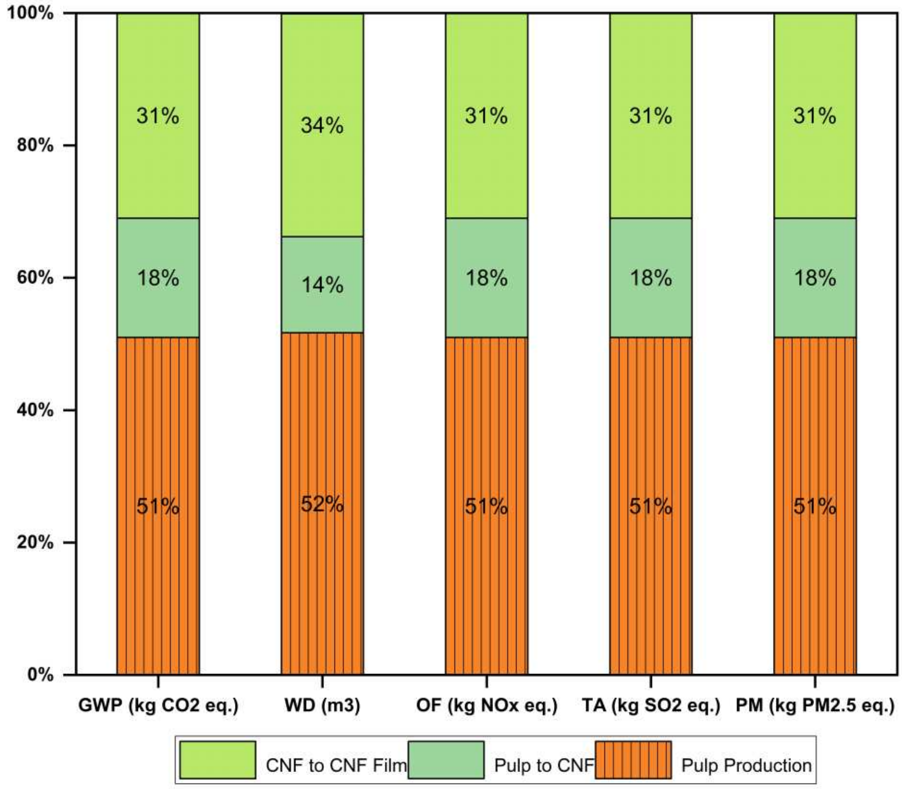


Figure 3: Environmental impacts per tonne of CNF films in a baseline scenario. Established based on BEK pulp as feedstock, 1.5% solid content, ambient drying and AU electricity mix

3.2. Scenario Analysis

There are six scenarios identified to have a major impact on the large-scale production of CNF films: I. Feedstock, II. Drying, III. Electricity mix, IV. Solid content, V. Producing composites. In this LCA study, what if scenario is being used as an analysis method [51] and results of the scenario analysis based on above-mentioned scenarios are summarized in Figure 4. As seen from Figure 4, to produce one tonne of CNF films, the highest environmental impact in terms of GWP was observed in AU electricity mix (baseline scenario) followed by composites production and commercial drying, while using waste carrot as feedstock showed the least. The environmental impacts in terms of GWP for composite production and commercial drying were approximately 9% and 12% lower as compared with the baseline scenario. Interestingly, producing composites of CNF did not contribute significantly to reducing the environmental impacts, as the values were around 10-15% lower than the baseline scenario owing to the additional stirring required to produce CMC suspensions. Contrarily, changing the feedstock with a waste stream showed the least environmental impact in terms of GWP accounting approximately 55% lower than the baseline scenario. The life cycle inventory for the studied scenarios is incorporated in Table 3.

Considering other environmental impacts such as OF, TA, and PM, Chinese (CN) electricity mix has resulted in highest footprints, while increasing solid content showed the least. The environmental impacts in terms of OF, TA and PM for CN were increased up 587%, 1992%, and 592% comparing with baseline scenario and 19, 70 and 19 times than increasing solid contents. It should be noted that the location showed a significant influence on OF, TA and PM of CNF films. For instance, the environmental footprints in terms of TA in CN mix has increased to 64, 64 and 68 times than in DE, GBR and USA. The significantly high environmental footprints for CN electricity mix attributed to their major dependance on coal as source of electricity generation. It is also noteworthy that environmental impacts of GBR electricity mix were increased up to 70% for OF, 3% for TA and 190% for PM comparing to scenario of increasing solid content. The WD was almost the same for all scenarios except when increasing the solid content. The WD was mainly associated with the water used in the reaction and via increasing the solid contents (4.5 wt.%), the water usage approximately 62% lower than all other scenarios in this study.

Table 3: Life cycle inventory for the studied scenarios. Data acquired from the Ecoinvent database for 1 ton of nanocellullose films.

	Category	Feedstock impact	Commercial drying	Electricity mix	Increasing solid content (4.5 wt%)	CNF composites production	Unit	Reference
Material	Wood	4	4	4	4	4	ton	[48]
	Pulp (solid content)	10 (1.5 wt%)	10 (1.5 wt%)	10 (1.5 wt%)	10 (4.5 wt%)	10 (1.5 wt%)	ton	[13, 30]
	Water (Reaction)	275000	275000	275000	91670	275000	L	-
	Water (Washing)	110	110	110	110	110	L	[13]
	Water (Refining)	19000	19000	19000	19000	19000	L	-
Transportation	Pulp	3.44×10^{-1}	3.44×10^{-1}	3.44×10^{-1}	3.44×10^{-1}	3.44×10^{-1}	MJ	[13, 49]
Electricity (country)	Pulp Treatment	25500 (AU)	25500 (AU)	20910 (CN); 12750 (DE); 12495 (GBR); 12648 (USA)	25500 (AU)	12750 (AU)	MJ	[24]
	Blender	764 (AU)	-	-	369 (AU)	-		
	Refiner	4855 (AU)	4855 (AU)	6138 (CN); 3743 (DE); 3668 (GBR); 3712 (USA)	4855 (AU)	7485 (AU)	MJ	
	CMC	-	-			8056 (AU)		
	Disintegrator	1728 (AU)	1728 (AU)	1417 (CN); 864 (DE); 847 (GBR); 857 (USA)	1728 (AU)	1728 (AU)	MJ	
	Spray Pump	400 (AU)	400 (AU)	328 (CN); 200 (DE); 196 (GBR); 198 (USA)	400 (AU)	400 (AU)	MJ	
	Belt Conveyor	600 (AU)	600 (AU)	492 (CN); 300 (DE); 294 (GBR); 298 (USA)	600 (AU)	600 (AU)	MJ	
	Drying	14000 (AU)	8136 (AU)	11480 (CN); 7000 (DE); 6860 (GBR); 6944 (USA)	4068 (AU)	8056 (AU)	MJ	[38]

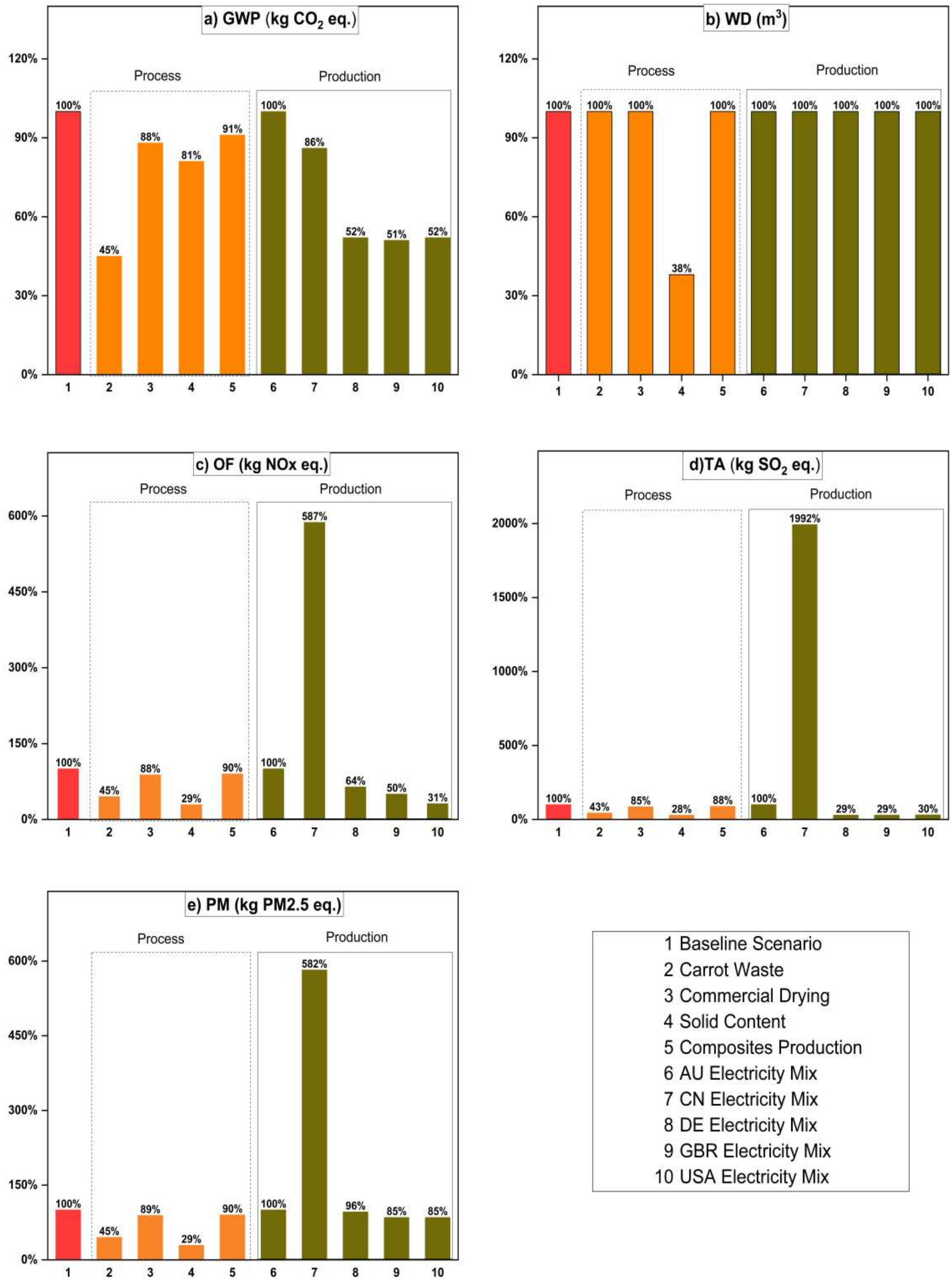


Figure 4: Scenario analysis for 1 tonne of CNF films production, expressed relative to the base case scenario. Scales are different for different impacts. (Red represents "Baseline Scenario", Orange represents process categories such as feedstock, commercial drying, solid content and composites production, Green presents locations such as AU, DE, CN, GBR and USA.

3.3. Recommended Scenario

Based on the scenario analysis and prior LCA studies conducted, it is observed that the high energy consumption was mainly associated with the pre-treatment of pulp and production of fibres [13, 24-26]. The energy consumed in pre-treatment of pulp could be reduced via substituting the feedstock, while waste feedstock is an ideal candidate for this purpose. However, the properties of the resultant fibres play a crucial role in this decision. For instance, the films produced via treatment of waste carrot fibres showed quite a good strength and optical performance as observed by Varanasi et al. [31]. Therefore, this aspect of using a waste-based feedstock could be quite effective to reducing the energy consumption of the films on a commercial scale. Considering the production of fibres, high pressure homogenization consumes a lot of energy as compared with the laboratory and pilot-scale refining [37]. Additionally, the properties of the produced fibres via each method are important. According to different studies, the mechanical, chemical, barrier, and thermal properties of the resultant films do not change considerably, whether the fibres are processed either via refining or in combination of refining and homogenization [30, 52]. High pressure homogenization resulted in producing quite fine fibres and showed slightly better mechanical and barrier performance when transformed into films; however, at the expense of approximately a 500 times increase in energy consumptions as compared with the pilot scale refining used in this study [13]. There is always a trade-off between the soaring energy consumption and properties of the CNF films, but as observed, the properties of CNF films could easily be improved by changing the chemistry of the fibres and producing composites [14, 32].

Considering the baseline scenario, the films were assumed to be dried at ambient conditions and thus requiring considerable space and time. The additional energy consumption and costs make this method infeasible for production on a commercial scale [13, 18]. Hence, drying the films at a continuous and energy efficient way is desirable and for this purpose; thus, infrared-based tunnel drying could be a viable option. Moreover, increasing the solid contents in the CNF suspension could significantly reduce the energy consumption associated with the drying of these films via reducing the amount of reaction water [32, 53].

Producing composites of CNF itself has resulted in an increased environmental impacts of the resultant films as observed in the scenario analysis; however, these composite films have shown excellent properties [8, 14, 32, 54, 55]. Additionally, incorporating a rheology modifier such as CMC with CNF could potentially increase the sprayable limit of the suspensions and thus reducing the water needed to dry and consequently reducing the energy cost associated with the drying of these films. The type of electricity mix at different locations also plays a significant role regarding the environmental impacts of the final product. For instance, the type of electricity mix in GBR, DE and USA are mainly based on natural gas have almost the same environmental impacts. Australia and China are mainly dependent on hard coal for their electricity generation and thus, the environmental impacts per kWh were on the higher side. As electricity consumption is significantly contributing to the environmental impacts, therefore, a slight change in their values could contribute tremendously to the energy consumption on a commercial scale.

Based on all these factors and considerations, a recommended scenario is established in this LCA study and the results are displayed in Figure 4. Based on this scenario, waste carrot as feedstock is subjected to bleaching and refining to produce CNF fibres. These CNF fibres were incorporated with CMC suspensions (1:1) and disintegrated in an industrial disintegrator at a 9 wt.% solid content without compromising the mechanical and barrier properties (based on experimental work). The suspensions were then sprayed via a spray system and finally, the composite films were dried in a drying tunnel at approximately 75 °C for 40-50 minutes (based on experimental work).

As shown in Figure 5, considering the recommended scenario, the environmental impacts for the large-scale production of CNF films have reduced significantly. For instance, the GWP of the resultant films in this scenario was approximately 700 kg CO2 eq. The overall environmental impacts (except WD) of the CNF-based films in a recommended scenario were approximately 86% and 69% lower as compared with the baseline and waste carrot as feedstock respectively. Additionally, the WD in the recommended scenario has reduced to 84% and 62% as compared with the baseline and increasing solid content scenarios respectively.

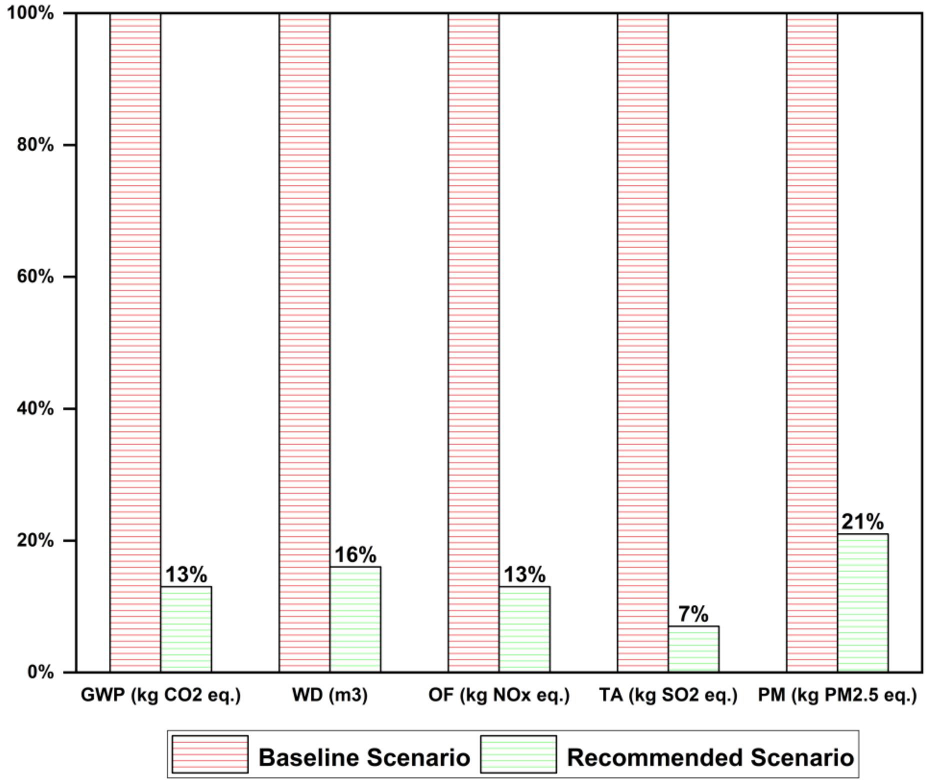


Figure 5: Recommended scenario versus baseline scenario for the large-scale production of 1 tonne of CNF films. Established based on waste carrot as feedstock, producing composites of CMC with CNF at 1:1 composition, commercial drying in a tunnel (fabricating 6 kg of films at 75 °C in 40 minutes), Great Britain electricity mix requiring 0.33 kg CO2 eq. in generating 1 kWh, Increased solid content (9 wt.%).

3.4. Impact comparison with synthetic and biobased food packaging

Figure 6 displays the comparison of environmental impacts of 1 tonne of synthetic, biobased packaging, and CNF films. Considering synthetic packaging, the values for GWP was obtained from a thesis by Hottle et al. [47], while other environmental impacts were calculated via using an Ecoivent v.3.9 (2022) based on energy data in a published chapter by Parker [56]. Similarly, for polylactic acid (PLA) and polyhydroxyalkanoates (PHA) films, the GWP data was acquired from published studies by Alba et al. [57] and Kim et al. [58] respectively. Thus, the data is an indication of general trends and should not be considered for specific cases. The WD results are not included in the comparison of results as water could be recycled and transformed into films again in producing CNF-based films. The environmental impacts of CNF films in a recommended scenario were approximately 76%, 68% and 81% lower compared to the polyethylene terephthalate (PET), low density polyethylene film (LDPE) and polystyrene (PS) food packaging respectively (Figure 6). Similarly, considering a comparison with biobased films, the environmental impacts were around 66% and 76% lower than PLA and PHA films respectively. The lower environmental impacts attribute to a number of process improvements conducted in this research work. These include changing the feedstock from Eucalyptus to waste carrots, conducting only refining of pulp and skipping the homogenization, increasing the solid contents for spraying (1.5 wt.% to 4.5 wt.%), using an environmentally friendly electricity mix (GBR), producing composites of CNF with environmentally friendly and compatible additives (CMC in this case), and switching from ambient or oven drying to commercial drying in a tunnel. Unlike the CNF films, where the environmental impacts were mainly associated with the production of fibres, the manufacturing stages contributing mainly to the energy consumptions of the synthetic packaging [59]. Comparing with the LCA study published by Nadeem et al. [13], the values of GWP were around 92% lower than the calculated values in this work. Similarly, the environmental impacts reported in this study decreased by

83% in GWP compared with the chitosan-CNF composite as reported by Ponnusamy and Mani [60].

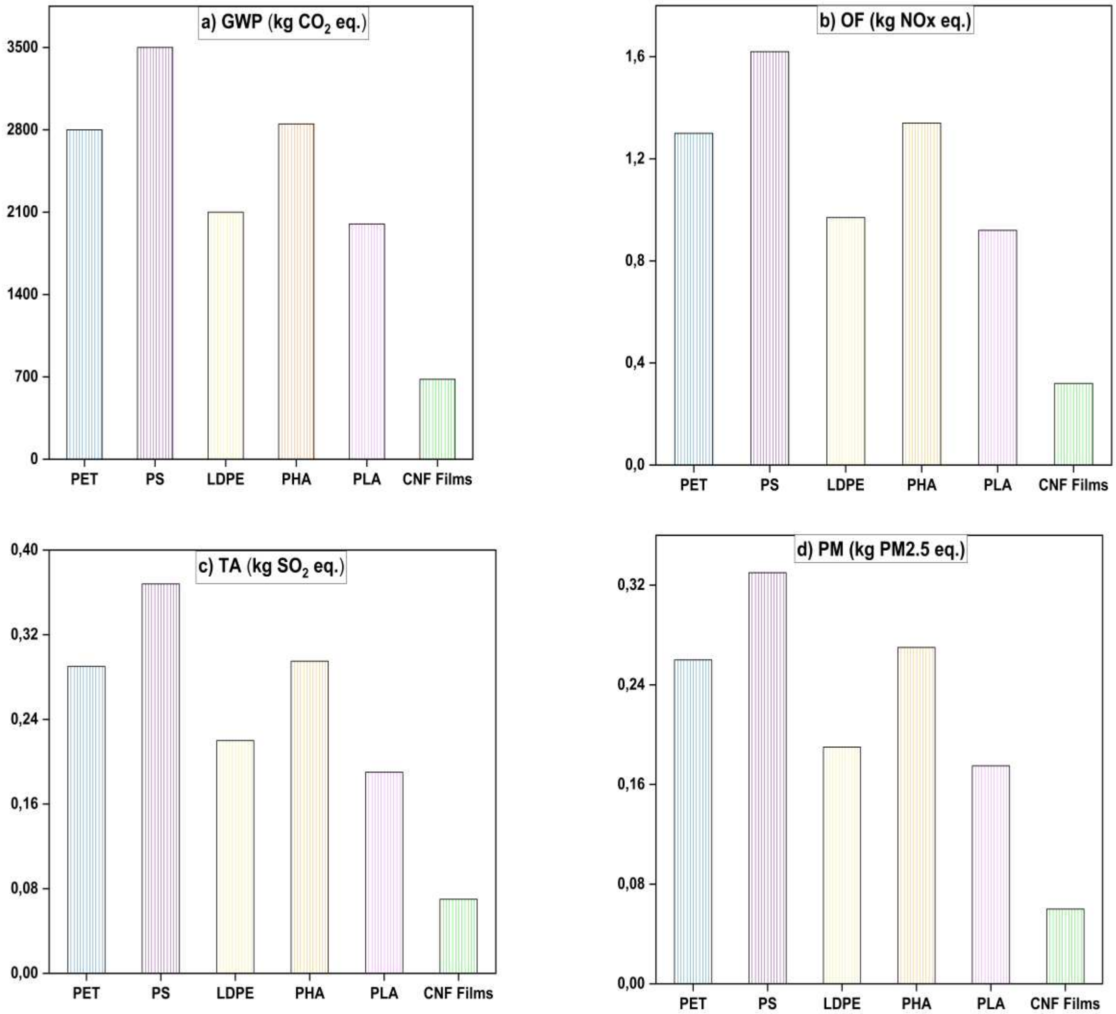


Figure 6: Impact comparison of CNF films and conventional packaging

Discussion

Based on scenario analysis, a recommended scenario is established in this study that results in significantly lower environmental footprints as compared with the synthetic packaging. However, it should be the noted that the studied CNF films in this work are still hydrophilic and might need further modifications to impart hydrophobicity to them. These modifications might contribute significantly to the environmental impacts of the existing results achieved in current study. Unfortunately, there are no available literature reports highlighting the environmental impacts of modification of CNF films and hence, there is no data for comparison. Cellulose-based materials are generally biodegradable i.e., they have the ability to be broken down naturally by the organisms in an ecosystem. It is also worth noting that modifications (especially chemical modifications) have negative effective on the biodegradability of the CNF films. The modified CNF films are expected to degrade at a lower rate than an unmodified ones. For instance, Surya et al., observed that the silane modified CNF composite films resulted in a lower degradation rate compared to the unmodified films [61]. It was also found that the unmodified and modified CNF composite films showed a weight loss of 75.1% and 52.6%, respectively, indicating a lower rate of degradation for the composite films. Although requiring more time for their degradation, the hydrophobic CNF films will still be biodegradable; however, expected to have slightly higher environmental impacts.

Assuming the significant environmental impacts for modification of CNF films, further steps needs to be taken to reducing the energy consumptions associated with fibres and the process. As increasing the production capacity to one tonne of CNF films per batch has significantly reduced the environmental impacts, further enhancing the scale of production possibly results in reducing the environmental impacts to a substantial extent. It is also observed that the location (electricity mix) has also shown a considerable impact on the environmental footprints of CNF films. Switching to the use of carbon neutral source of electricity and reusing the heat consumed in the drying process for other cycles of the process could possibly further lower the environmental impacts [13].

Considering the end-of-life (EOL) options, there is limited LCA data available for bio-based materials apart from PLA. Although there are a number of EOL options that could be analyzed for CNF films; however, owing to unavailability of data and limited scope of this research study, three scenarios are considered i.e., recycling, incineration and landfilling. The data for recycling was based on experimental work conducted [33], while the data for incineration was obtained from the study by Daniele et al. on the reuse options of textile fibres [62]. Additionally, the data for landfilling was assumed to be same as for waste paperboard (degradability of 34.5% for 100 years) and acquired from Econivent. Figure 7 illustrates the GWP comparison of EOL options for 100 tonnes of CNF films.

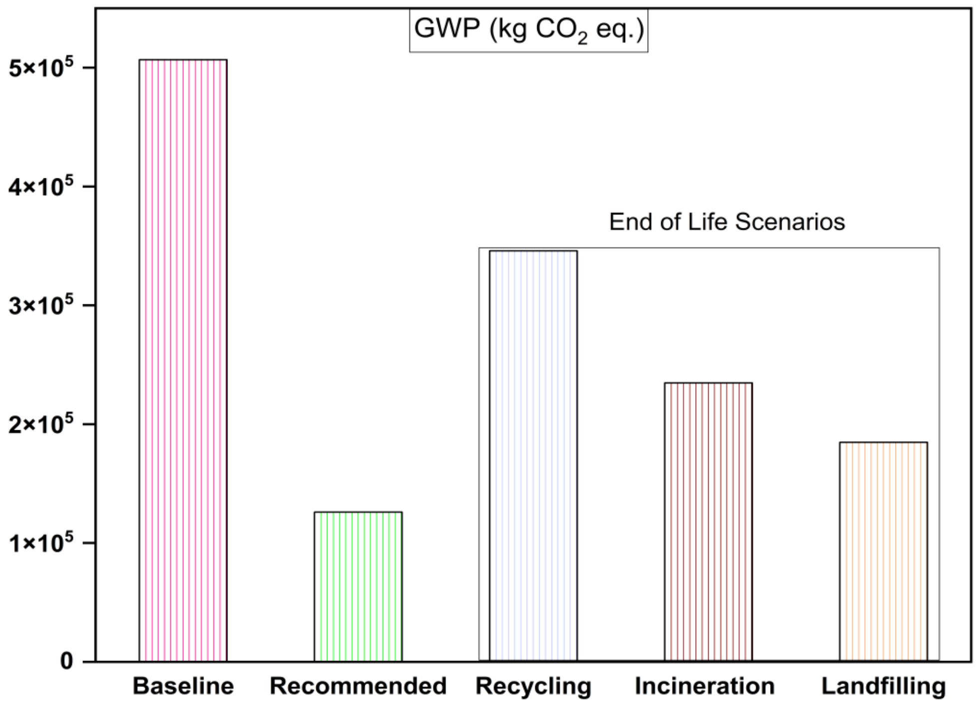


Figure 7: GWP comparison of end of life scenarios for 100 tonnes of CNF films

With the current technology, recycling of CNF films seem to be an expensive option owing to high processing energy associated with disintegration to breakdown the agglomeration of fibres [33]. The GWP potential for recycling has increased up to 45% and 84% as compared with incineration and landfilling respectively. The GWP for incineration can further be reduced via recovering energy and heat from the process and thus can further reduce the environmental impacts associated with it. Although a biodegradable polymer, cellulosic materials resulting in a breakdown to CH_4 and CO_2 [63], while CH_4 emissions have 34 times more GWP than CO_2 [57]. However, 99% of CH_4 can be captured but needing an additional energy for this purpose and should be taken into consideration.

The recycling of films is one area that should be taken into account seriously as recycled fibres could be transformed into films again, resulting in waste minimization and added energy and cost benefits. However, one concern is the excessive disintegration; contributing considerably in higher environmental impacts for recycling of fibres and needs to be reduced. Based on the TAPPI standard for pulp recycling (T205 SP-02) and a published work by Shanmugam et al. [6], these films could be recycled at 75×10^3 number of revolutions in a disintegrator. However, the mechanical and barrier properties of the films were significantly reduced to 30-40% as compared with the non-recycled ones. Based on published work by Nadeem et al., these films were recycled at 300×10^3 revolutions without compromising their dimensional stability, mechanical and barrier properties [33]. Unfortunately, recycling the films at 20 times higher number of revolutions could contribute to environmental impacts to a great extent. The increased energy consumption has resulted in higher environmental impacts. For instance, considering GBR as location, using 9% solid contents, one time recycling and recycling efficiency of 80%, the GWP was approximately 5500 kg CO_2 eq. to produce 1.8 tonnes of CNF films.

As seen from Figure 7, to produce 100 tonnes of CNF films via recycling, the GWP has decreased to 45% as compared to baseline scenario (AU) and increased to approximately 175% than a recommended scenario suggested in this study. Considering these results, it is conclusive that producing the CNF films via using a waste feedstock (without recycling) is more environmentally friendly than choosing a wood as feedstock and subjecting it to recycling. However, this will result in soaring waste generation. For this reason, the number of revolutions to disintegrate the fibres needs to be reduced. At present, number of revolutions lower than 300×10^3 has resulted in significant decrease in mechanical and barrier properties [6], which is not desirable. Forming composites could be one viable option in this regard as they tend to reduce the self-aggregation of fibres and might result in requiring lower number of disintegration levels on recycling [64].

Conclusion

This LCA study investigates the cradle-to-gate environmental impacts for a large-scale production of spray deposited CNF films. The spray deposited CNF films in a baseline scenario showed approximately 1.5-2.5 times higher environmental impacts in terms of GWP as compared with synthetic packaging [47]. However, via thoroughly assessing the key influencing categories such as changing the feedstock, increasing the solid content, commercial drying and location, the environmental impacts were significantly lowered. For instance, increasing the solid contents contributed substantially to reducing the environmental impacts i.e., 19% for GWP, 62% for WD, 71% for OF, 72% for TA, and 71% for PM as compared with baseline respectively.

Considering the various impacts categories in a scenario analysis, a recommended scenario was established in this study. Increasing the solid contents, using waste as feedstock, incorporating CMC, and changing the type of electricity mix have reduced the environmental impacts to a substantial extent. The environmental impacts of a recommended scenario were tremendously decreased (70-80%) as compared with the synthetic packaging.

Acknowledgements

Philippe Nimmegeers holds a FWO senior postdoctoral fellowship (grant number: 1215523N) granted by FWO Vlaanderen/Research Foundation Flanders.

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

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