ORIGINAL PAPER



A cradle-to-gate life cycle assessment of polyamide-starch biocomposites: carbon footprint as an indicator of sustainability

Laura Äkräs¹ · Frans Silvenius² · Hossein Baniasadi¹ · Marjatta Vahvaselkä³ · Hannu Ilvesniemi³ · Jukka Seppälä¹

Received: 29 November 2023 / Accepted: 6 May 2024 / Published online: 29 May 2024 $\ensuremath{\textcircled{O}}$ The Author(s) 2024

Abstract

Accelerating climate change poses an alarming global issue, demanding a range of prompt and effective solutions. In response, bio-based plastics and biocomposites have emerged as extensively researched alternatives to combat the environmental threats posed by a warming climate. In this context, the present paper presents a cradle-to-gate life cycle assessment of a newly developed polyamide-starch biocomposite, with varying content of potato starch as the biofiller (ranging from 0 to 70 wt%). The primary aim was to quantitatively measure the total carbon footprint of the selected biocomposite. The results indicated that the progressive addition of potato starch as the biofiller into the copolyamide matrix significantly reduced the total carbon footprint of the biocomposite, achieving a maximum reduction of 42–43% with the highest starch content of 70 wt%. Moreover, the newly developed polyamide-starch biocomposite demonstrated excellent performance compared to reference fossil-based polyamides of polyamide 6 (PA6), polyamide 12 (PA12), and polyamide 6.6 (PA6.6), as well as composites of PA610/80 wt% polylactic acid modified by reactive extrusion (REX-PLA) and PA40/30 wt% glass fibers, with carbon footprint reductions of 29, 39, 42, 59, and 79%, respectively. Based on these findings, the polyamide-starch biocomposite, especially with the highest content of potato starch (70 wt%), exhibits significant potential as a new material solution to reduce the carbon footprint of several existing fossil- and bio-based polyamides together with polyamide-based composites. In doing so, it contributes to advancing the development of a more climate-friendly future for plastics through reductions in their carbon footprints.

Graphical abstract



A concise illustration, showcasing the main content of the present paper; a cradle-to-gate life cycle assessment of the newly developed polyamide-starch biocomposites, which results indicated substantial reductions in the carbon footprint of these biocomposites

Keywords Copolyamide \cdot Starch \cdot Biocomposite \cdot Life cycle assessment (LCA) \cdot Cradle-to-gate approach \cdot Carbon footprint

Extended author information available on the last page of the article

Introduction

The current world encounters several environmental threats, such as the depletion of fossil resources, typically used as an energy source for different purposes or as building blocks for various plastic products, a challenge of accumulated waste in nature due to inadequate disposal practices and non-biodegradable nature of some of the commercial plastic products, as well as, above all, accelerating climate change with multiple innocuous consequences on the ecosystems and human health. The ways to slow down the development of these challenges include the utilization of several different steps on a complementary basis through, for instance, more excessive use of bio-based feedstocks both for energy generation and for manufacture of plastic products (Yang et al. 2021; Zheng and Suh 2019), along with the development of new, sustainable, bio-based plastic products with comparable techno-economic properties with their fossil-based counterparts (Bishop et al. 2021; van den Oever and Molenveld 2017). These are accompanied with the substitution of fossil-based plastic products with their bio-based alternatives (Zheng and Suh 2019) and, when applicable, increased recycling rates of plastic products (Zheng and Suh 2019), together with appropriate and effective legislation to guide these suggested actions. In addition to the acts mentioned above, quantifying, evaluating, and understanding the environmental impacts and hotspots caused by the manufactured bio- and fossil-based plastic products is becoming more and more widely recognized, appreciated, and required as an aid in sustainable decision-making in multiple different arenas. In this context, for the quantification and evaluation of the environmental impacts, LCA with carbon footprint as the most researched impact category is a well-established technique widely in use (Banerjee and Ray 2022).

Against this background, from the material perspective, blending biofillers with different plastic matrices has been realized to enable a high bio-based content (Mohanty et al. 2018), frequently improving some of the properties of the individual plastics, such as their tensile strength (Shanmugam et al. 2021; Sadasivuni et al. 2020), impact strength (Sadasivuni et al. 2020), tensile modulus (Shanmugam et al. 2021), and stiffness (Mohanty et al. 2018), with simultaneous enhancements in the overall biodegradability of the resulted (bio)composites per se (Shanmugam et al. 2021). On the other hand, from the environmental point of view, bio-based plastics have been reported to often possess a lower carbon footprint in comparison with their fossil-based alternatives (Yang et al. 2021; Zheng and Suh 2019; Broeren et al. 2017); (bio)composites have similarly been found to have generally decreased environmental footprints than fossil-based plastics (Shanmugam et al. 2021); and the addition of functional (bio) fillers in plastics have also been shown to reduce the carbon footprint of these plastics when compared to their plain counterparts (Civancik-Uslu et al. 2018). In addition to these findings, the inclusion of biofillers into the bio-based plastic matrices can decrease their production costs (Shanmugam et al. 2021), therefore making them more cost-efficient. Hence, (bio)composites undoubtedly possess some proven potential to improve the performance of individual plastics in multiple different ways.

Stemming from this, recently, fabrication methods for different types of (bio)composites have been researched and analyzed in the literature (Sadasivuni et al. 2020; Andrew and Dhakal 2022; Kostag and El Seoud 2021; Baniasadi et al. 2021), among which polyamide-based (bio)composites have shown promising results, mostly regarding their enhanced mechanical properties (Baniasadi et al. 2023a, b; Oliver-Ortega et al. 2016) but also improved viscoelastic performances and water vapor transmission (Baniasadi et al. 2021), when compared to the plain polyamide matrices (Baniasadi et al. 2021; Baniasadi et al. 2023a, b; Oliver-Ortega et al. 2016). Despite this, according to the literature survey conducted by the authors for the purposes of this paper, the number of LCA studies regarding (bio)composites with polyamides as one of the constituents seems scarce. In this respect, only a few studies regarding the matter were able to be found, namely the comparison of PA11/stone ground wood biocomposite with the composite of polypropylene (PP)/carbon fibers with cradle-to-grave approach (Oliver-Ortega et al. 2019), analysis of the commercial composites of PA610/glass fibers and PA410/glass fibers (in more detail, RTP 2099 X 115387 B and DSM EcoPaXXTM Q-KGS6, respectively) from cradle to gate (Petersson et al. 2013), together with the cradle-to-gate LCA of the composites of REX-PLA/acrylonitrile butadiene styrene (ABS), REX-PLA/PA610, and REX-PLA/PA1010 (Torrell Gines 2016).

In this discourse, up to date, no LCA studies specifically about polyamide-starch biocomposites were able to be detected by the authors, while studies focusing on other types of (bio)composites with starch as a matrix or biofiller were more readily available. As an example, a cradle-tofactory gate LCA about six starch plastics was conducted, consisting of the selected bio- or fossil-based polymers of polybutyrate adipate-co-terephthalate (PBAT), PLA, postindustrial recyclate PLA (re-PLA), polybutylene succinate (PBS), and polyhydroxybutyrate (PHB), virgin or reclaimed starch and/or natural fibers as the biofillers, combined with various additives (Broeren et al. 2017). On the other hand, two biocomposites from cradle to gate were also studied, and these biocomposites consisted of either PLA or PLA and thermoplastic starch (TPS) as the matrix and wood fibers as the biofillers (Mahalle et al. 2014). Similarly, the environmental impacts of boxes made of fossil-derived polystyrene (PS), corn-derived PLA, and PLA-cassava starch biocomposite were researched from cradle to consumer gate (Suwanmanee et al. 2013), while elsewhere a cradle-to-grave LCA about wheat-polyvinyl alcohol (PVOH), potato starch-PVOH, and maize starch-PVOH, also including minor additives, was performed (Guo and Murphy 2012). In general, many of these reviewed studies pointed out that the increase in the bio-based content of the (bio)composites, in the form of a matrix or biofiller, can reduce some of the environmental impacts caused by these (bio)composites, such as carbon footprint, non-renewable energy use (NREU), and ozone depletion potential (ODP) (Broeren et al. 2017; Oliver-Ortega et al. 2019; Torrell Gines 2016; Mahalle et al. 2014; Guo and Murphy 2012), in comparison with a neat polymer or fossil-based alternatives. Despite this, the reduction in these specific categories tends to occur with a simultaneous increase in the agricultural-related impacts, such as eutrophication, land use, as well as freshwater and terrestrial ecotoxicity (Broeren et al. 2017; Mahalle et al. 2014; Guo and Murphy 2012). This reported reduction in the carbon footprint, nonetheless, may require more emission-intensive manufacture of the monomers for the polymerization of the selected plastics when compared to the cultivation and processing of the most often locally produced biofillers (Oliver-Ortega et al. 2019; Mahalle et al. 2014), such as starch. Additionally, in most of the reviewed studies, transportation contributed only minorly to the environmental impacts, for example, the carbon footprint (Oliver-Ortega et al. 2019; Suwanmanee et al. 2013; Guo and Murphy 2012).

Against this background, there evidently is a need for research considering the environmental impacts of polyamide-based biocomposites. Analysis of the environmental impacts would also be interesting due to the composition of these biocomposites; for example, due to the spherical morphology of the starch particles and their plasticizing effect, the latter one being facilitated by the applied compatibilization chemistry that enabled the starch content of the biocomposite to be elevated to an impressive 70 wt%. This would not have been feasible with alternative biofillers, e.g., cellulose or lignin-based materials, owing to the unique attributes of starch and the implementation of effective chemical compatibility measures. Furthermore, the newly developed polyamide-starch biocomposite has been reported to exhibit other promising characteristics, such as exceptionally low melting point, enhanced adhesion between the polymer and biofiller together with improved dispersion of the biofiller within the polymer matrix, combined with excellent rheological properties, and a commendable balance between stiffness and toughness (Baniasadi et al. 2023b). Yet its environmental impacts still await to be researched. Consequently, the goal of the present paper was to quantify the total carbon footprint and further identify the hotspots of the cradle-to-gate manufacture of the newly developed polyamide-starch biocomposite on the laboratory scale to investigate any latent potential and bottlenecks for its further development from an environmental point of view. The ultimate goal was, therefore, to find out whether the sustainability aspect of the polyamide-starch biocomposite in terms of its carbon footprint was in line with its previously presented promising properties.

As the reasoning, identification of the hotspots and bottlenecks of a new material alternative already on the laboratory scale can be argued to enable a more straightforward and affordable adjustment of the processes in comparison with a more elevated process setup. Moreover, the selected approach can be justified by Pini et al. (2020), who emphasized that the use of LCA also for the laboratory-scale research of chemical compounds should be highly recommended or, depending on the case, even mandatory. This statement is supported by the good availability of previously conducted laboratory-scale LCAs about the synthesis of bio-based plastics (Pini et al. 2020; Ang et al. 2021; Nitkiewicz et al. 2020). Intriguingly, a framework for scaling up the laboratory-scale chemical production processes for LCA was found by the authors (Piccinno et al. 2016), yet it was excluded from the study. This is because the framework was found not to be applicable to the laboratory-scale setup of the present paper, since it was limited to wet chemistry (Piccinno et al. 2016), excluding solid-state chemistry, consequently not representing the laboratory processes of the present study.

Lastly, it is claimed that the carbon footprint of the newly developed polyamide-starch biocomposite will be reduced with the increased content of potato starch, outperforming the neat copolyamide and, partially, the fossil-based plastics and composites used as the references. In this regard, LDPE and PP were selected for the reference fossil-based plastics due to their reported high comparability to starchbased plastics (Broeren et al. 2017), whereas HDPE, PA6, and PA6.6 were added to the list due to the contemplated advantage to compare the performance of the polyamidestarch biocomposite to other plain, fossil-based polyamides and plastics. In addition to this, a few polyamide-based composites were used as the reference, namely PA610/80 wt% REX-PLA and PA40/30 wt% glass fibers, to compare the carbon footprint of the polyamide-starch biocomposite also to some utterly commercial composites or the ones including a commercial polyamide.

Materials and methodology

Goal and scope definition

The aim of the present study was to quantify the carbon footprint and identify the hotspots of the laboratory-scale manufacture of the novel polyamide-starch biocomposite, with the potato starch contents of 0, 10, 30, 50, and 70 wt%, by utilizing the widely used technique of LCA according to the guidelines set by ISO 14040 and 14044 standards (Internal Organization for Standardization (ISO) 2006a; International Organization for Standardization (ISO) 2006b). For this purpose, the focus of the present study was set on a cradle-to-gate approach, in which the first two life cycle stages of the polyamide-starch biocomposite were considered for the LCA. The system boundaries, therefore, consisted of the raw material extraction and acquisition (cultivation and/ or preparation of the substances for the manufacture of the biocomposite) and, further, manufacture of the polyamidestarch biocomposite per se (see Fig. 1 for the details). The steps of use and grave (End-of-Life (EoL) of the biocomposite) were, nonetheless, excluded from the present study. The regionalization of the study, in turn, was set to be Finland and either Finland, Germany, or France for the manufacture of the polyamide-starch biocomposite and the different substances, respectively, while the functional unit was selected to be 1 kg of biocomposite. The selected functional unit can be stated to be comparable to the mass-based functional units applied for the recent LCA studies, in which the (bio) composites per study possessed differences in the matrix types (Broeren et al. 2017; Kane et al. 2022), filler contents (Broeren et al. 2017; Kane et al. 2022), technical properties of the resulted (bio)composites (Broeren et al. 2017; Kane et al. 2022), and/or different target applications (Broeren et al. 2017; Kane et al. 2022). Finally, the target audience for the present study consisted of, for instance, scholars and industry representatives with a keen interest in bio-based composites, particularly polyamide-based ones, and the quantified carbon footprint they possess.

Life cycle inventory (LCI)

Manufacture of substances and their LCI

Cultivation of potatoes and production of potato starch The step-by-step progression of the cultivation of potatoes, in general, included the actions of liming and tillage to maintain the basic growth conditions, together with the plowing of the field. These were followed by planting the potato seeds, shaping the potato bed, caring for the crops through sufficient fertilization, efficient use of fungicides, herbicides, and insecticides, as well as adjusted sprinkler irrigation (Aaltonen et al. 2016). In the end, the potatoes were



Fig. 1 A simplified scheme about the system boundaries of the manufacture of polyamide-starch biocomposite from cradle to gate. The gray dashed line represents the overall system and, in more detail, the manufacture of the biocomposite, while the color-coded blocks stand for the different unit processes, respectively. Additionally, the dashed line, red in color, indicates substances that have been excluded from the study (clarified in more detail in subSect. "Manufacture of substances and their LCI"). Noteworthy, pelletizing has been included in the processes of copolymerization and melt blending, even if it has been excluded from the scheme, whereas transportation has not been depicted in this scheme harvested, tentatively cleaned, and placed in the intermediate storage when required (Finnamyl and Lapuan Peruna 2017).

After the successful cultivation, potato starch was extracted from the potatoes by utilizing either a filtration or a decanter process, which was largely based on the same steps, apart from the treatment of potato juice. Namely, in the decanter process, as much potato juice as possible was extracted after the crushing of potatoes, whereas in the filtration process, only part of the potato juice was extracted, followed by utter removal later through fine sizing. In general, the process steps for both the filtration and the decanter process (the exact order of the filtering of potato mush and extracting of potato juice depending on the type of process) included obtaining the potatoes, removing the dry soil, storing the potatoes, floating the potatoes to remove more soil, and extracting the stones and other light substances. This was followed by further washing the potatoes and crushing them, filtering the resulting potato mush to remove the fibers, extracting the potato juice completely or partially, concentrating and washing the potato juice, as well as fine sizing the mush to remove the rest of the fibers and potato juice. The processes ended up with pre-drying, final drying, as well as temporary storage and packing of the resulting potato starch. (Pääkkönen et al. 2004; Ahokas et al. 2012).

Consequently, the potato starch processing resulted in the by-products of potato juice and pulp, further containing protein and fiber, whereas the waste streams consisted of dry soil, stones and other light substances, sludge, as well as wastewater (Pääkkönen et al. 2004; Ahokas et al. 2012). In more detail, wastewater included, for instance, potato juice and sludge as a result of the washing steps (Välimaa et al. 2017). Potato juice was mainly utilized as a fertilizer, and potato pulp was used as a feed, whereas soil and sludge were further able to be used, for example, for landscaping, backfilling, and as soil on the fields, excluding potato and beet fields. Recovery of protein from potato juice by coagulation was also possible; nonetheless, it is not yet applicable in Finland. Wastewater, in turn, was treated in the activated sludge plants within the potato starch processing factories (Pääkkönen et al. 2004).

Moreover, the data of Natural Resources Institute Finland's case study for the cultivation of potatoes was solely gathered from Finland, whereas the Ecoinvent data (obtained from Ecoinvent v3.8, available in the PRé Sustainability's LCA software of SimaPro) for the processing and modification of potato starch was collected from the US, Europe in general, Germany, and Finland. Nonetheless, for the purposes of the present study, both processes (cultivation of potatoes and production of potato starch) were assumed to occur in Finland. Finnish Finnamyl Oy was further selected as the potato starch processing company for the present study due to its highest production quantities of potato starch (100 000–105 000 t/a) (Pääkkönen et al. 2004), combined with the shortest distance to the facilities of Aalto University, where the manufacture of polyamide-starch biocomposite was set to be occurred.

For the potato starch processing, dry soil, stones, and other light substances were excluded from the LCI of the present study due to their assumed lower value as coproducts and, therefore, fewer requirements for further processing. Potato protein, in turn, was included as an output flow due to the utilization of German data. Additionally, as defined in ISO 14040 standard (International Organization for Standardization (ISO) 2006b), the allocation was not able to be avoided due to the difficulty of dividing the multi-output process of potato starch processing into subprocesses or conducting a system expansion due to lack of data. Therefore, the allocation was based on the quantitative relationship between the coproducts (International Organization for Standardization (ISO) 2006b); as the share of potato starch was over half of the coproducts, all the burdens were allocated to potato starch. Lastly, wastewater was excluded from this allocation, while allocation was not considered for the process of cultivation of potatoes.

Regarding the sources of input flows for the processing of potato starch, integrated steam and heat production (CHP) has been reported to be widely used in Finland with an efficiency of 85% or even 90% (Silvonen and Mäkelä 1996). Based on this fact, in Sphera's software of LCA for Experts (LCA FE), the process steam from natural gas for the processing of potato starch was selected to be the Finnish one with an efficiency of 85%, while the EU-28 tap water from groundwater at the plant served as a source of tap water. The activated sludge plant for the treatment of wastewater of potato starch processing, in turn, was selected to be EU-28 municipal wastewater treatment (with agricultural sludge application) due to the fact that, in Finland, the sludge resulted from the treatment of wastewater is used as organic fertilizers in the fields (Pääkkönen et al. 2004). The input flow for this process was further selected to be organic, contaminated, and untreated industrial wastewater, treated according to Directive 91/271/EEC concerning urban wastewater treatment.

Finally, the collected LCI from the Ecoinvent database and case study of Natural Resources Institute of Finland is not more detailly showcased in the present paper due to the commercial origin and general confidentiality of the data, respectively. However, the auxiliary processes used to model the production of potato starch in LCA FE are presented in Supplementary Information's (SI) Table S1, excluding the cultivation of potatoes as no auxiliary processes were utilized for its modeling.

Manufacture of monomers and compatibilizer Altogether, three monomers were considered for the present LCA study,

namely 11-aminoundecanoic acid, 1,18-octadecanedioic acid, and 1,12-diaminododecane. To start with, the manufacture of 11-aminoundecanoic acid was initiated by the cultivation of castor beans within the castor plant (Ricinus communis L.) in low humidity, adequately high temperatures, and deep sandy loams with required fertility, acidity, and drainage, which was followed by the extraction of castor oil from the cultivated castor beans, for example, through mechanical pressing and/or solvent extraction (Mubofu 2016). The cultivation of castor beans and extraction of castor oil occurred in India, which currently dominates both processes globally (Mubofu 2016). In the next step, at the manufacturing plant located in Germany to which the produced castor oil was transported, transesterification of castor oil in the presence of methanol and a catalyst was applied to yield methyl ricinoleate and glycerol, after which pyrolysis in the presence of steam at the elevated temperatures between 500-600 °C was further utilized to gain methyl 10-undecenate and heptanal as the by-product. This was followed by bromination, which converted the methyl 10-undecenate into 11-bromoundecanoic acid, and amination, resulting in the final intermediate product, 11-aminoundecanoic acid (Dimian et al. 2019).

The second monomer, 1,18-octadecanedioic acid was manufactured by olefin metathesis (for example, alkenolysis) of purified plant oil (such as rapeseed oil) with 1-butene, followed by separation of the resulting olefins and esters through distillation. Next, the subsequent transesterification of these esters into monoesters, typically fatty acid methyl esters (FAME), such as methyl oleate, occurred in the ester stream. Finally, the monoesters and the resulting glycerol were separated to obtain a range of products, including α,ω dicarboxylic acids, such as 1,18-octadecanedioic acid (Chikkali and Mecking 2012).

Lastly, the manufacture of the third monomer, 1,12-diaminododecane (DMDA), started with the trimerization of butadiene, having been derived from fossil-based sources (Yang et al. 2010), to 1,5,9-cyclododecatriene (CDT) (Gaide et al. 2016), catalyzed by nickel (Yang et al. 2010), which was followed by either two- or three-step reaction route (Gaide et al. 2016). The former included the hydrogenation of CDT to cyclododecane and oxidation of cyclododecane with air or oxygen to obtain a mixture of cyclododecanol and cyclododecanone, followed by the oxidation step with nitric acid to eventually yield 1,12-dodecanedioic acid (Yang et al. 2010; Gaide et al. 2016). On the other hand, the latter route consisted of the partial hydrogenation of CDT to cyclododecene with the subsequent oxidative ozonolytic cleavage to obtain 1,12-dodecanedioic acid (Gaide et al. 2016). Finally, these reaction steps were followed by the hydrogenation of 1,12-dodecanedioic acid, together with the subsequent amination of 1,12-dodecanediol, in the presence of liquid ammonia and catalyst of Ru/triphos system to obtain the desired 1,12-diaminododecane (Shi et al. 2017).

In addition to these monomers, the compatibilizer of ODI was required for the manufacture of polyamide-starch biocomposite, which consequently was also considered for the present LCA. In this regard, the ODI was synthesized, as most of the isocyanates, through a reaction between an amine or its salt and phosgene (Ozaki 1972; Saunders and Slocombe 1948), which can either be conducted in a solid-, liquid-, or vapor-phase, the need of a catalyst depending on the selected synthesizing method, and the reaction also resulting in the formation of urea as the side-product (Saunders and Slocombe 1948).

The required LCI data were purchased from the private company of Sphera Solutions GmbH (in the case of monomers) or collected from the MLC database (in terms of the compatibilizer). Nonetheless, the stearic acid (octadecanoic acid) dataset, purchased from Sphera Solutions GmbH, and a readily available process in LCA FE about aliphatic isocyanates were used as a proxy for the monomer of 1,18-octadecanedioic acid and compatibilizer of ODI, respectively, due to the lack of precise data for the original compatibilizer and monomer. These actions were justified by the reported possibility of replacing the materials, which are absent in the LCA databases, with their closely related analogue (Pini et al. 2020). The exact quantities of the input and output flows as well as other assumptions of the monomers and compatibilizer have, nonetheless, not been reported in the present paper due to the commercial origin of the data as well as the guidelines set by Sphera Solutions GmbH for the datasets developed by them.

Manufacture of polyamide-starch biocomposite The manufacture of the novel polyamide-starch biocomposite consisted of four basic steps, namely, synthesizing of the low-melting-point polyamide through copolymerization, surface modification of potato starch to ease its compatibilization with the copolyamide matrix, and melt blending of these two resulted constituents to obtain the polyamidestarch biocomposite. In more detail, firstly, the monomers of 11-aminoundecanoic acid, 1,18-octadecanedioic acid, and 1,12-diaminododecane were copolymerized through a polycondensation reaction for 4 h at the temperature of 200-240 C° with the aid of a catalyst of sodium hypophosphite monohydrate under nitrogen flow to obtain the copolymer of PA11 and PA1218 (in short, PA11coPA1218) and water as a by-product. In the end, the synthesized PA11coPA1218 was cooled and pelletized, resulting in plastic granules (Baniasadi et al. 2023b).

This was followed by the surface modification of potato starch with the compatibilizer of ODI, consisting of thorough drying of the starch at 70 °C for 48 h as well as heating of the mixture of starch and ODI at 100 °C for 24 h to obtain Table 1LCI for thecopolymerization of monomersto obtain PA11coPA1218

Input or output flow	Type of constituent	Quantity of the constituent (per 1 kg of biocomposite)
Input	11-aminoundecanoic acid	0.0705 kg
Input	1,18-octadecanedioic acid	0.11 kg
Input	1,12-diaminododecane	0.0701 kg
Input	Sodium hypophosphite monohydrate	0.0002 kg
Input	Electricity	Confidential information (CI)
Input	Nitrogen	0.0006 kg
Input	Water	40 kg
Output	PA11coPA1218	0.23 kg
Output	Wastewater (untreated)	40 kg

 Table 2
 LCI for the surface modification of starch to obtain ODI-g-starch

Input or output flow	Type of constituent	Quantity of the constituent (per 1 kg of biocomposite)
Input	ODI	0.01 kg
Input	Potato starch	0.1 kg
Input	Electricity	CI
Output	ODI-g-starch	0.11 kg

 Table 3
 LCI for the melt blending of PA11coPA1218 and ODI-gstarch to obtain polyamide-starch biocomposite

Input or output flow	Type of constituent	Quantity of the constituent (per 1 kg of polyamide-starch biocomposite)
Input	PA11coPA1218 granules	0.9, 0.7, 0.5, or 0.3 kg*
Input	ODI-g-starch	0.1, 0.3, 0.5, or 0.7 kg*
Input	Electricity	CI
Input	Water	50 kg
Output	Polyamide-starch bio- composite granules	1 kg

*Depending on the content of potato starch used as a biofiller in the copolyamide matrix, namely, 10, 30, 50, or 70 wt%, respectively

the surface-modified starch (in short, ODI-*g*-starch). Finally, the resulting PA11coPA1218 and ODI-*g*-starch were melt blended at 200 °C for 1 h by using a twin-screw extruder, followed by cooling and pelletizing to obtain the biocomposite granules. After the experiments, the cooling water was poured into the sewer (Baniasadi et al. 2023a, b).

The LCI data required for the manufacture of polyamidestarch biocomposite was directly derived from these laboratory-scale experiments conducted at the facilities of Aalto University School of Chemical Engineering, except of the value for electricity, which was collected from the Ecoinvent database. All the input and output flows together with the quantities of these unit processes of copolymerization, surface modification, and melt blending are further listed in Tables 1, 2, and 3, respectively. Additionally, the comprehensive description of all the auxiliary processes, flows, and data required to model these unit processes in LCA FE is provided in SI's Tables S2–S4. Noteworthy, the LCI data for electricity is not showcased in the present paper due to its confidentiality.

The manufacture of the catalyst of sodium hypophosphite monohydrate for the copolymerization of PA11coPA1218 was, nonetheless, excluded from the present study since it was used in rather small quantities (less than 1% from the functional unit of 1 kg of biocomposite), and some polymerization reactions can run without a specific catalyst. On the other hand, the manufacture of polyamide-starch biocomposite on the laboratory scale was assumed not to produce considerable solid waste streams or losses of substances, which is why these streams and losses were excluded from the present study. The utilized cooling water served as an exception, resulting in the wastewater being assumed to be treated in the EU-28 municipal wastewater facilities.

Similarly, only the copolymerization of 11-aminoundecanoic acid, 1,18-octadecanedioic acid, and 1,12-diaminododecane, resulting in PA11coPA1218, was estimated to yield a vanishingly small amount of water as a by-product with no explicit end-users due to its collection as part of the nitrogen flow, which is why also this water was excluded. Consequently, due to the exclusion of solid waste streams and by-products, the allocation was not essential to use in this manufacturing step of polyamide-starch biocomposite. In addition to these, due to its assumably low impacts, pelletizing was included in the unit processes of copolymerization of PA11coPA1218 and its subsequent melt blending with surface-modified potato starch without separate modeling of this unit process in LCA FE. In other words, pelletizing was excluded from the present study.

Additionally, the European electricity grid mix with the distribution of energy sources in the European electricity

grid was utilized as the energy source for all the unit processes of this manufacturing stage. The source of nitrogen, in turn, for the unit process of copolymerization of monomers was selected to be EU-28 nitrogen (gaseous), which can also be found in the Sphera Managed LCA Content (MLC) 2023.1 database, available in LCA FE. The value for nitrogen flow was estimated based on the experimental setup by calculating the amount of gaseous nitrogen being consumed during the four hours of copolymerization with the flow rate of 2 ml/min. The calculations for the exact amount of utilized nitrogen are thoroughly explained in SI's Table S5.

Further, since the overall share of groundwater as a process water to produce plastics in Finland can be assumed to be slightly greater than that of surface water (Salminen et al. 2017), desalinated and deionized EU-28 process water from groundwater, being available in the MLC 2023.1 database, was exploited as a source of cooling water for the unit processes of copolymerization of monomers and melt blending of constituents. As in the case of nitrogen flow, the value for cooling water was assessed based on the laboratory experiments by estimating the amount of consumed water for four hours and one hour for the unit processes of copolymerization and melt blending, respectively.

Transportation of substances The processed potato starch was assumed to be transported from its processor of Finnamyl Oy, located in Kokemäki in Finland, monomers from their selected manufacturing plant of abcr Gute Chemie, nestled in Bruchsal in Germany, and the compatibilizer of ODI from its manufacturer of Vencorex in France, respectively. Germany was selected as the manufacturing country for the monomers due to its relative proximity to Finland in comparison with some other European countries and the fact that a multitude of fine chemical manufacturers and/or suppliers operate in Germany. Noteworthy, for the potato starch, monomers, and compatibilizer, the EU-28 diesel mix at the refinery, manufactured both from crude oil and from biocomponents, together with EU-28 heavy fuel oil at refinery (1.0 wt% S) was selected as the energy sources for transportation in LCA FE, while transportation was excluded from the laboratory-scale manufacturing processes of the polyamide-starch biocomposite due to the permanent location of the experiments with no need for transportation. Lastly, the LCI data for the transport of substances was gathered from the MLC database, the places of departure and arrival, as well as the distances, having been estimated with a basis on the selected websites. An in-depth report about the transport of these substances can also be found in SI, with all the transportation details being concisely presented in SI's Table S6.

Sequestered CO₂ in potato starch The CO₂ sequestration into the biomass was considered by calculating the CO₂ content of the processed and modified potato starch per 1 kg of biocomposite according to Eq. (1). For these calculations, potato starch was selected as the carbon sink instead of potatoes due to the better consideration of its main output flows in the LCA model, namely, all the burdens of this production step were allocated to potato starch. Finally, these calculations of sequestered CO₂ in potato starch are explained in more detail in SI's Table S7.

$$CDR(CO_2) = \left(\frac{\left(\frac{M(C)_{ST}}{M(ST)}\right) \cdot m(ST)_{biocomposite}}{\left(\frac{M(C)_{CO2}}{M(CO_2)}\right)}\right) \cdot Cf(CO_2) \quad (1)$$

where CDR(CO₂) stands for the amount of sequestered CO₂ in potato starch, $M(C)_{ST}$ for the molecular weight of carbon in potato starch, M(ST) for the molecular weight of potato starch, $m(ST)_{biocomposite}$ for the amount of potato starch required for 1 kg of biocomposite, $M(C)_{CO2}$ for the molecular weight of carbon in CO₂, $M(CO_2)$ for the molecular weight of CO₂, and Cf(CO₂) for the characterization factor of CO₂.

Life cycle impact assessment (LCIA)

Software, databases, and impact category

Sphera's LCA FE with MLC 2023.1 databases, including the impact assessment method EF 3.1, was used to execute the LCIA of the present paper. Further, climate impact (expressed as kg CO₂ eq./kg of biocomposite) played the role of the principal impact category for the present LCA study to quantify the carbon footprint of the biocomposite under analysis. In more detail, climate impact was selected for the study since it is the most researched impact category, which has successfully been used to quantify the carbon footprint reductions of different types of (bio)composites (Broeren et al. 2017; Oliver-Ortega et al. 2019; Torrell Gines 2016; Mahalle et al. 2014; Guo and Murphy 2012; Kane et al. 2022), therefore possessing potential in aiding the development of more climate-friendly plastics.

Sensitivity analysis

Sensitivity analysis was applied in the present paper to see how the alteration of data and model details will affect the LCIA results, with an aim to both tentatively simulate industrial-scale production and preliminary explore the influence of a decarbonized electricity grid mix in the future. This was done by adding internal circulation of cooling water in the manufacture of biocomposite on the laboratory scale and switching the European electricity grid mix to the Finnish alternative to accomplish the first and second aim, respectively. Nonetheless, the electricity grid mix used for the datasets of monomers and compatibilizer was left intact due to their commercial origin and, thus, non-modifiable characteristics. Stemming from this overview, in the following results and conclusion Sects. "Results and discussion" and "Conclusions," respectively, the two types of sensitivity analyses are referred to as sensitivity analysis 1 and sensitivity analysis 2 (in short, SA1 and SA2), respectively, the former representing the case with the Finnish electricity grid mix and internal circulation of cooling water and the latter one with Finnish electricity grid mix and without an internal circulation of cooling water.

Results and discussion

Carbon footprint and sensitivity analysis of polyamide-starch biocomposite

The core aim of the conducted LCA of the present paper, as previously defined in the introduction and subSect. "Goal and scope definition," was to quantify the total carbon footprint and identify the hotspots of the manufacture of polyamide-starch biocomposite, closely followed by exploring the effect of applied sensitivity analyses of the selected processes on the final LCIA results. Ergo, in the present subsection, an in-depth analysis of the obtained LCIA results has been conducted with a joint and, when required, separate focus on each of the cases (baseline case and the two types of sensitivity analyses SA1 and SA2, respectively), consisting of the analysis of the general phenomenon, caused by the addition of potato starch as the biofiller into the copolyamide matrix, together with the identification and evaluation of the hotspots within the defined system boundaries. The core focus of the present paper was on a carbon footprint; however, a primary analysis about acidification, eutrophication (terrestrial, freshwater, and marine), as well as land use impacts of polyamide-starch biocomposite was also conducted and is provided in SI's Figures S1–S3.

Having said this, all the total carbon footprints and hotspots per baseline case as well as SA1 and SA2, respectively, are presented briefly in Fig. 2. Noteworthy, the amount of sequestered CO₂ in potato starch was calculated to be as low as -0.16 kg CO₂ eq./kg of biocomposite with a minor effect on the total LCIA results, which is why the sequestered CO₂ is excluded from the analysis of the present subsection and Fig. 2, subSect. "Comparison of the LCIA results to the literature and reference plastics" and Fig. 3 being an exception. Additionally, the production processes of monomers and compatibilizer were combined for the LCA calculations as well as presenting the LCIA results to meet the guidelines set by Sphera Solutions GmbH for the purchased datasets developed by them and the ones available in their LCA FE software. Lastly, to enhance the reportability and understandability of the LCIA results, the carbon footprints caused by the unit processes of copolymerization, surface modification, and melt blending—their values being vanishingly low on their own—were calculated, presented, and analyzed under the umbrella term of manufacture of polyamide-starch biocomposite.

To start with, the general phenomenon that resulted in the addition of potato starch into the copolyamide matrix was evident and visible in terms of all the studied cases; this addition progressively reduced the total carbon footprint of polyamide-starch biocomposite, even up to 42-43% with the highest potato starch content of 70 wt% in comparison with the plain copolyamide. This occurrence can be attributed to the production processes of potato starch per se, in more detail, the cultivation of potatoes, as well as the processing and modification of potato starch. Namely, as can also be seen from Fig. 2, the carbon footprint of these two processes, altogether, was constantly considerably lower than the one caused by the production of monomers and compatibilizer (the energy-intensive monomers being the major contributor to the total carbon footprint of polyamide-starch biocomposite). The contribution of these monomers and compatibilizer, both requiring long-distance transportation from their manufacturers, was replaced step by step to a greater extent upon the addition of locally produced potato starch (Oliver-Ortega et al. 2019; Guo and Murphy 2012). In more detail, on one hand, the share of the production of monomers and compatibilizer from the total carbon footprint of polyamidestarch biocomposite was decreased by 14, 12.7, and 12.4% for the baseline case, SA1, and SA2, respectively. On the other hand, the share of cultivation of potatoes from the total carbon footprint was increased by 7.4, 8, and 7.5% for the baseline case, SA1, and SA2, respectively, and simultaneously, the share of processing and modification of potato starch from the same total carbon footprint was increased by 5.1, 3.8, and 3.6% for the baseline case, SA1, and SA2, respectively.

Nonetheless, despite this unquestionable role of the cultivation of potatoes together with processing and modification of potato starch in reducing the total carbon footprint of polyamide-starch biocomposite, also other processes within the system boundaries, required to manufacture the biocomposite, participated in this job, although to a lesser extent. *Ipso facto*, upon the increased addition of potato starch into the copolyamide matrix, the carbon footprint caused by the transport of substances was increased by 0.32, 0.38, and 0.35%, and the manufacture of polyamide-starch biocomposite by 1.2, 0.54, and 0.9% for the baseline case, SA1, and SA2, respectively. These can be stated to be caused by



Fig. 2 A set of graphs showcasing the total LCIA results of the polyamide-starch biocomposite with varying content of potato starch, combined with the contribution of each production step, per baseline case and applied sensitivity analyses SA1 and SA2, respectively. Further, the abbreviations BioCo1, BioCo2, BioCo3, BioCo4, and BioCo5 stand for the polyamide-starch biocomposite with the starch

contents of 0, 10, 30, 50, and 70 wt%, respectively. **A.** The baseline case (without an internal circulation of cooling water). **B.** SA1 (with Finnish electricity grid mix and an internal circulation of cooling water). **C** SA2 (with Finnish electricity grid mix and without an internal circulation of cooling water). Noteworthy, the sequestered CO_2 in potato starch has not been considered in the present figure

the slightly increased amount of potato starch transported to Aalto University and further used to manufacture the polyamide-starch biocomposite at the facilities of Aalto University.

Additionally, when having a further look at the differences between the studied cases, the total carbon footprint with the progressively increased content of potato starch was found to be 5.4–7.7% and 0.1–2.1% lower in terms of SA1 and SA2, respectively, when compared to the total carbon footprint of the baseline case. In more detail, considering the former case (SA1), this can mainly be attributed to the simulated internal circulation of cooling water with an utter removal of the impacts caused by the production, circulation, and wastewater treatment of this cooling water, which slightly decreased the total carbon footprint. This is a reasonable outcome because the environmental impacts caused by the laboratory-scale processes are often higher than their industrial-scale counterparts (Piccinno et al. 2016).

On the other hand, in terms of the latter case (SA2), the occurrence can be reasoned by the different sources used for the European and Finnish electricity generation, the European one mainly relying on coal, gas, and other fossil fuels with a share of 39% of the total electricity generation, followed by nuclear as well as combined wind and solar (both having a share of 22%), while Finland primarily utilizes nuclear (close to 40%) with a higher





Fig. 3 A set of graphs comparing the performance of polyamidestarch biocomposite of the baseline case with the lowest and highest content of potato starch (namely, 0 and 70 wt%, respectively) to the selected fossil-based plastics and polyamide-based composites. Further, the abbreviations of BioCo1, BioCo5, Co1, and Co2 stand for the plain copolyamide, polyamide-starch biocomposite with a starch content of 70 wt%, PA610/80 wt% REX-PLA, and PA40/30 wt%

share of hydro- and bioenergy (both close to 20%) (Jones et al. 2023). As the reasoning, the burning of fossil fuels has been reported to emit higher CO_2 emissions than, for example, the use of nuclear and hydro as the sources for electricity generation (Yadav et al. 2021), which consequently decreases the impact caused by the processing and modification of potato starch together with the manufacture of polyamide-starch biocomposite of the present study upon the application of Finnish electricity.

After evaluating the bigger picture, it is time to cap off this subsection by having a detailed look at the hotspots. Regarding the matter, Fig. 2 clearly showcases that the production step of monomers and compatibilizer (and primarily the monomers) evidently was the major hotspot for all the cases, and its share ranged between 79-93, 86-99, and 81-93% of the total carbon footprint for the baseline case, SA1, and SA2, respectively. Apart from this, the otherwise slight variation in the order of hotspots was found to be dependent both on the content of potato starch and on different cases, namely, in terms of the baseline case and SA2, excluding the highest content of potato starch, while the second most contributing production step was the manufacture of polyamide-starch biocomposite with the share of total carbon footprint ranging between 5.6-6.8% and 5.5-6.4% for the baseline case and SA2, respectively. Yet, SA1 served

glass fibers, respectively. **A**, Polyamide-starch biocomposite vs. the selected fossil-based plastics. **B**, Polyamide-starch biocomposite vs. the selected polyamide-based composites. Noteworthy, in the figure, the sequestered CO_2 is included in the carbon footprint of polyamide-starch biocomposite and the plain copolyamide. Additionally, the sequestered CO_2 has been assumed to be considered in the carbon footprint of all the reference polyamide-based composites

as the exception in this observation, with the biocomposite's manufacturing step having been the minor contributor throughout the varying content of potato starch with a share of 0.2-0.7% of the total carbon footprint.

In the case of the rest of the production steps, transportation contributed, most of the time, the least to the total carbon footprint in the baseline case as well as SA2 and the second/third least in SA1 with the share of 1.24-1.69% of the total carbon footprint. Finally, the contribution of the production steps of potatoes and potato starch progressively increased upon the increased addition of potato starch, with these two steps ultimately having been the fourth and second/third most contributing steps with the share of 7.4–8 and 3.6-5.1%, respectively, in the case of the highest content of potato starch in all the cases under analysis.

Comparison of the LCIA results to the literature and reference plastics

As previously stated, the available LCA studies about the polyamide-based or starch-including (bio)composites indicated that the increase in the bio-based content of the (bio) composites could reduce their environmental impacts in certain impact categories (such as in terms of their carbon footprint) (Oliver-Ortega et al. 2019; Torrell Gines 2016; Mahalle et al. 2014), making them outperform some of the fossil-based plastics in these categories (Broeren et al. 2017; Mahalle et al. 2014; Guo and Murphy 2012). Despite the reductions in the carbon footprint, upon the increased bio-based content, the agricultural-related impacts of these (bio)composites are simultaneously worsened (Broeren et al. 2017; Mahalle et al. 2014; Guo and Murphy 2012).

Regarding the hotspots, the process with the highest carbon footprint, irrespective of the polymer matrix or biofiller, seems to be, in some cases, the manufacture of monomers for the plastic matrix, whereas transportation is the least polluting one (Oliver-Ortega et al. 2019; Suwanmanee et al. 2013; Guo and Murphy 2012), followed by the manufacture of biofiller and (bio)composite (Correa et al. 2019; Oliver-Ortega et al. 2019). Stemming from this, the cultivation and processing of the biofiller can be stated to act as a minor contributor to the total carbon footprint, for example, in the case of virgin potato starch (Broeren et al. 2017), for which the carbon footprint per se is rather low (e.g., 0.88 CO₂ eq./ kg of dry native starch produced from wheat, maize, or potatoes (An et al. 2012)). Despite these outcomes, it is good to bear in mind that the results in the form of their absolute values cannot directly be compared with each other due to the methodological differences of various LCA studies (Walker and Rothman 2020); ergo, the direct comparison of these literature findings with the LCIA results acquired from the present study was excluded from the discussion. However, it can be concluded that the general phenomenon in the outline is similar between the LCA studies of the analyzed literature and the present paper.

Additionally, Fig. 3 showcases the performance of the polyamide-starch biocomposite vs. the selected reference fossil-based plastics and composites, from which the data for PP, HDPE, LDPE, and PA6.6 were collected from the reports of PlasticsEurope (PlasticsEurope 2014a, 2014b, 2014c), while the data for PA12, PA6, PA40/30 wt% glass fibers, and PA610/80 wt% REX-PLA were found from the literature (Petersson et al. 2013; Torrell Gines 2016; London 2020; Brehmer 2014). Noteworthy, the carbon footprints of these reference plastics and composites have not been quantified along with the ones of polyamide-starch biocomposite but extracted from the reports or literature, which is a viable approach previously used by Broeren et al. (2017). Regarding the matter, the total carbon footprint caused by the baseline case's polyamide-starch biocomposite with the highest content of potato starch (70 wt%) was 64, 49, and 43% higher in comparison with the fossil-based plastics of PP, HDPE, and LDPE, respectively, and this specific biocomposite simultaneously had 29, 39, and 42% lower carbon footprint than the ones of fossil-based polyamides, PA6, PA12, and PA6.6, respectively. In a similar manner, the carbon footprint of the baseline case's polyamide-starch biocomposite was decreased by 59 and 79% in comparison with the ones of the composites, PA610/80 wt% REX-PLA and PA40/30 wt% glass fibers, respectively.

Noteworthy, despite the fact that the functional units and system boundaries of these reference plastics and composites are in outline comparable to the one of the present paper, their LCIA results can be slightly affected by the applied LCIA impact assessment methods other than EF 3.1 (difference 1) (Torrell Gines 2016; PlasticsEurope 2014a, 2014b, 2014c), the separate inclusion of the electricity-requiring pelletizer in the LCA (difference 2) (Torrell Gines 2016; PlasticsEurope 2014a, 2014b, 2014c), together with the addition of product manufacturing step (difference 3), for example, through injection molding (Petersson et al. 2013). These differences might either slightly underestimate or overestimate the final LCIA results in the case of difference 1 and 2/3, respectively, in comparison with the present study with, although assumably, no remarkable effect on the final conclusions presented in this subsection.

Based on this analysis, the specific polyamide-starch biocomposite of the present study with previously reported excellent properties can be envisaged to possess a better environmental performance from the global warming potential point of view in comparison with the commercial, energy-intensive fossil-based polyamides and, partially, some of the polyamide-based composites, consequently advancing the more climate-friendly manufacture of plastics through blatant reductions in their total carbon footprint. Despite this outcome, advancements in the manufacture of monomers, combined with the minimization of the amount of compatibilizer (Broeren et al. 2017), are required to ensure the polyamide-starch biocomposite's ability to better compete with the fossil-based plastics of PP, HDPE, and LDPE with a relatively low climate impact, for instance through decarbonization of energy by increasing the share of utterly biosourced electricity (Zheng and Suh 2019). Alternatively, evaluation of other possible bio-based plastic matrices for biocomposites with initially lower carbon footprint or application of agricultural wastes as biofillers are also options worth considering in reducing the total carbon footprint of biocomposites more remarkably. One promising option would also be to blend biofillers with fossil-based polyolefins to increase their bio-based content and, potentially, lower their carbon footprint to some extent.

Limitations of the study

Despite the great efforts, every LCA study possesses its own limitations as well as requirements for enhancement and future research, and the study in question is no exception. Consequently, calculations of the amount of nitrogen and water for the unit processes of copolymerization and melt blending contain some uncertainties due to the difficulty of estimating the exact quantity for both the initial nitrogen and the water flows as well as the timescale for the exact utilization of these resources during the experimental work. Therefore, it would have been ideal to utilize more accurate data for these purposes, which had required, instead of assessing the matter through calculations and/or assumptions afterward, the direct and precise measurement of water and nitrogen flows already during the experimental work. In terms of other estimations, the places of arrival and departure, as well as approximated distances of land and ocean routes for the transportation of substances, primarily had grounded on assumptions, which, however, can be argued to have only a marginal effect on the acquired LCIA results.

In addition to the previous aspects, the use of stearic acid and a mix of aliphatic isocyanates as a proxy for 1,18-octadecanedioic acid (the monomer) and ODI (the compatibilizer), respectively, as the input flows for the unit processes of copolymerization or melt blending has, to some extent, influenced the LCIA results. In more detail, the application of stearic acid has most probably underestimated the real carbon footprint of 1,18-octadecanedioic acid since some of its process steps are missing in the proxy, while the utilization of aliphatic isocyanates has, most likely, overestimated the actual carbon footprint of ODI due to the higher number of isocyanates included in this proxy dataset. In this context, the exclusion of pelletizing step may also have slightly underestimated the carbon footprint caused by the manufacture of biocomposite. Finally, the comparison of the LCIA results of different technological scales (laboratory scale vs. the industrial one) can also be considered as one of the limitations of the present study.

Development points and suggestions for further research

A full scaling up of the present laboratory-scale experiments could have been worth considering because, on the industrial scale, the equipment of chemical processes is more complex with linked process steps (Piccinno et al. 2016), which affects the final LCIA results by usually lowering the total impacts in comparison with the ones derived from the laboratory conditions (Piccinno et al. 2016). This scaling up would also have improved the comparison of polyamidestarch biocomposite, manufactured in the laboratory, with the commercial alternatives (Piccinno et al. 2016). Despite this, the acquired LCIA results of the present paper, as they are, already possess societal relevance. Lastly, the inclusion of a wider set of impact categories could have derived a broader picture of the environmental impacts caused by the manufacture of polyamide-starch biocomposite of the present paper.

Overall, in the future, a discreet enhancement of the LCI data and the related assumptions, a full scaling up of the laboratory-scale experiments of the present study into an industrial scale, together with an inclusion of a broader set of impact categories, would be beneficial in obtaining a more relevant and comprehensive understanding of the environmental impacts caused by the manufacture of polyamide-starch biocomposite of the present paper. The proposed approach could also better facilitate the development and promotion of the newly developed polyamide-starch biocomposite (Piccinno et al. 2016), potentially also on the industrial scale.

Conclusions

In the present paper, a comprehensive cradle-to-gate LCA of a newly developed polyamide-starch biocomposite was conducted, varying the content of potato starch from 0 to 70 wt%, with a specific focus on its carbon footprint. The results of the LCA are promising and provide valuable insights about the early-stage environmental burdens of the biocomposite with a possibility to forecast the impacts of its industrial-scale production; nonetheless, there is still potential for further carbon footprint reductions. One approach would, therefore, be to enhance the production of energyintensive monomers, while also reducing the required amount of compatibilizer. These improvements would enable the developed biocomposite to better compete with the commercial, fossil-based polyolefins that currently have a relatively low carbon footprint. For future LCA studies, in turn, it is recommended to scale up the laboratory setup to facilitate meaningful comparisons with various commercial plastics and (bio)composites. It would also be beneficial to explore a broader set of applied impact categories to gain a comprehensive understanding of the overall environmental impacts caused by the polyamide-starch biocomposite. To end up with, the polyamide-starch biocomposite presented in this paper, especially with the highest content of potato starch (70 wt%), has demonstrated considerable potential for reducing the carbon footprint of plain polyamides.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10098-024-02884-1.

Acknowledgments The authors would like to profusely thank for the received funding No. 327248 (ValueBioMat), having been provided by the Strategic Research Council (SRC), established within the Academy of Finland. Additionally, the authors are very grateful for the excellent service provided by the staff of Sphera Solutions GmbH. Lastly, the authors would like to cordially thank Johan Äkräs for skillfully helping to make the graphical abstract as wonderful as it is today.

Author contributions Laura Äkräs contributed to conceptualization, methodology, software, investigation, data curation, formal analysis, visualization, writing—original draft, and project administration. Frans Silvenius contributed to conceptualization, investigation, data curation, formal analysis, and writing—review and editing. Hossein Baniasadi contributed to conceptualization, investigation, data curation, and writing—review and editing. Marjatta Vahvaselkä contributed to writing—review and editing. Hannu Ilvesniemi contributed to writing—review and editing and funding acquisition. Jukka Seppälä contributed to conceptualization, writing—review and editing, funding acquisition, and supervision.

Funding Open Access funding provided by Aalto University. The research leading to these above-described results received funding from the Strategic Research Council (SRC), established within the Academy of Finland, under Grant Agreement No. 327248 (ValueBioMat).

Data availability The exact LCI and LCIA data not reported in the present paper are either of commercial or of confidential origin and, consequently, cannot be publicly shared. However, the modeling details for the production of potato starch and laboratory-scale experiments, detailed calculations for gaseous nitrogen and sequestered CO_2 in potato starch, transportation details, as well as LCIA results of acidification, eutrophication (terrestrial, freshwater, and marine), and land use are available as Supplementary Information.

Declarations

Conflict of interest The authors have no conflict of interests to declare.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

References

- Aaltonen M, Hannukkala A, Huusela-Veistola E, et al (2016) PERUNA IPM-ohjeet 2016 (POTATO IPM instructions). Luonnonvarakeskus (Natural Resources Institute Finland), Helsinki, Finland
- Ahokas M, Välimaa A-L, Kankaala A, et al (2012) Perunan ja vihannesten sivuvirtojen arvokomponenttien hyötykäyttö (The value component utilization of the sidestreams of potato and vegetables). MTT Jokioinen, Oulu and Ruukki, Finland
- An V, Evelien D, Katrien B (2012) Life Cycle Assessment study of starch products for the European starch industry association (AAF): sector study. The Flemish Institute for Technological Research NV ("VITO"), Boeretang, Belgium
- Andrew JJ, Dhakal HN (2022) Sustainable biobased composites for advanced applications: recent trends and future opportunities—A critical review. Compos Part C Open Access 7:100220. https://doi.org/10.1016/j.jcomc.2021.100220
- Ang P, Mothe SR, Chennamaneni LR et al (2021) Laboratory-scale lifecycle assessment: a comparison of existing and emerging methods of poly(ε-caprolactone) synthesis. ACS Sustain Chem Eng 9:669–683. https://doi.org/10.1021/acssuschemeng.0c06247
- Banerjee R, Ray SS (2022) Sustainability and life cycle assessment of thermoplastic polymers for packaging: a review on fundamental

principles and applications. Macromol Mater Eng 2100794:1–22. https://doi.org/10.1002/mame.202100794

- Baniasadi H, Trifol J, Lipponen S, Seppälä J (2021) Sustainable composites of surface-modified cellulose with low-melting point polyamide. Mater Today Chem 22:100590. https://doi.org/10.1016/j. mtchem.2021.100590
- Baniasadi H, Lipponen S, Asplund M, Seppälä J (2023a) High-concentration lignin biocomposites with low-melting point biopolyamide. Chem Eng J 451:138564. https://doi.org/10.1016/j.cej.2022. 138564
- Baniasadi H, Madani Z, Mohan M et al (2023b) Heat-induced actuator fibers: starch-containing biopolyamide composites for functional textiles. ACS Appl Mater Interfaces. https://doi.org/10.1021/ acsami.3c08774
- Bishop G, Styles D, Lens PNL (2021) Environmental performance comparison of bioplastics and petrochemical plastics: A review of life cycle assessment (LCA) methodological decisions. Resour Conserv Recycl 168:105451. https://doi.org/10.1016/j.resconrec. 2021.105451
- Brehmer B (2014) Polyamides from biomass derived monomers. In:
 Kabasci S (ed) bio-based plastics: materials and applications.
 John Wiley & Sons Inc, First Edit, pp 275–293
- Broeren MLM, Kuling L, Worrell E, Shen L (2017) Environmental impact assessment of six starch plastics focusing on wastewaterderived starch and additives. Resour Conserv Recycl 127:246– 255. https://doi.org/10.1016/j.resconrec.2017.09.001
- Chikkali S, Mecking S (2012) Refining of plant oils to chemicals by olefin metathesis. Angew Chemie Int Ed 51:5802–5808. https://doi.org/10.1002/anie.201107645
- Civancik-Uslu D, Ferrer L, Puig R, Fullana-i-Palmer P (2018) Are functional fillers improving environmental behavior of plastics? A review on LCA studies. Sci Total Environ 626:927–940. https://doi.org/10.1016/j.scitotenv.2018.01.149
- Correa JP, Montalvo-Navarrete JM, Hidalgo-Salazar MA (2019) Carbon footprint considerations for biocomposite materials for sustainable products: a review. J Clean Prod 208:785–794. https://doi.org/10.1016/j.jclepro.2018.10.099
- Dimian AC, Iancu P, Plesu V et al (2019) Castor oil biorefinery: conceptual process design, simulation and economic analysis. Chem Eng Res Des 141:198–219
- Finnamyl, Lapuan Peruna (2017) Tärkkelysperunan viljelyohjeet (Farming guidelines of starch potato). Kokemäki, Finland
- Gaide T, Behr A, Arns A et al (2016) Hydroesterification of methyl 10-undecenoate in thermomorphic multicomponent solvent systems—Process development for the synthesis of sustainable polymer precursors. Chem Eng Process Process Intensif 99:197–204. https://doi.org/10.1016/j.cep.2015.07.009
- Guo M, Murphy RJ (2012) Is there a generic environmental advantage for starch-PVOH biopolymers over petrochemical polymers? J Polym Environ 20:976–990. https://doi.org/10.1007/ s10924-012-0489-3
- International Organization for Standardization (ISO) (2006) ISO 14040:2006. Environmental management—Life cycle assessment—Princicples and framework. https://www.iso.org/stand ard/37456.html. Accessed 25 June 2022
- International Organization for Standardization (ISO) (2006) ISO 14044:2006. Environmental management—Life cycle assessment—Requirements and guidelines. https://www.iso.org/stand ard/38498.html. Accessed 25 June 2022
- Jones D, Brown S, Czyżak P, et al (2023) European Electricity Review 2023. Ember, London, United Kingdom
- Kane S, Van Roijen E, Ryan C, Miller S (2022) Reducing the environmental impacts of plastics while increasing strength: biochar fillers in biodegradable, recycled, and fossil-fuel derived plastics. Compos Part C Open Access 8:100253. https://doi.org/10.1016/j.jcomc.2022.100253

- Kostag M, El Seoud OA (2021) Sustainable biomaterials based on cellulose, chitin and chitosan composites—a review. Carbohydr Polym Technol Appl 2:100079. https://doi.org/10.1016/j.carpta. 2021.100079
- MB London (2020) Cradle-to-Gate Life Cycle Assessment of Multi-Jet Fusion 3D Printing. Master's thesis, University of Michigan
- Mahalle L, Alemdar A, Mihai M, Legros N (2014) A cradle-to-gate life cycle assessment of wood fibre-reinforced polylactic acid (PLA) and polylactic acid/thermoplastic starch (PLA/TPS) biocomposites. Int J Life Cycle Assess 19:1305–1315. https://doi. org/10.1007/s11367-014-0731-4
- Mohanty AK, Vivekanandhan S, Pin J-M, Misra M (2018) Composites from renewable and sustainable resources: challenges and innovations. Science 362:536–542. https://doi.org/10.1126/ science.aat9072
- Mubofu EB (2016) Castor oil as a potential renewable resource for the production of functional materials. Sustain Chem Process 4:1–12. https://doi.org/10.1186/s40508-016-0055-8
- Nitkiewicz T, Wojnarowska M, Sołtysik M et al (2020) How sustainable are biopolymers? Findings from a life cycle assessment of polyhydroxyalkanoate production from rapeseed oil-derivatives. Sci Total Environ 749:141279. https://doi.org/10.1016/j.scitotenv. 2020.141279
- Oliver-Ortega H, Granda LA, Espinach FX et al (2016) Stiffness of biobased polyamide 11 reinforced with softwood stone ground-wood fibres as an alternative to polypropylene-glass fibre composites. Eur Polym J 84:481–489. https://doi.org/10.1016/j.eurpolymj. 2016.09.062
- Oliver-Ortega H, Julian F, Espinach FX et al (2019) Research on the use of lignocellulosic fibers reinforced bio-polyamide 11 with composites for automotive parts: car door handle case study. J Clean Prod 226:64–73. https://doi.org/10.1016/j.jclepro.2019. 04.047
- Ozaki S (1972) Recent advances in isocyanate chemistry. Chem Rev 72:457–496
- J Pääkkönen, S Vuorikoski, K Pirkanniemi, H Hyytiä (2004) Paras käytettävissä oleva tekniikka (BAT) Suomen perunatärkkelysteollisuudessa (The best available technique (BAT) in the Finnish potato starch industry). Suomen ympäristökeskus (Finnish Environment Institute), Helsinki, Finland
- L Petersson, H Martini, M Chiaravalli, et al (2013) Biobased engineering plastics a tool to reduce carbon footprint. In: The 6th International Conference on Life Cycle Management in Gothenburg 2013, Gothenburg, Sweden, pp 1–4
- Piccinno F, Hischier R, Seeger S, Som C (2016) From laboratory to industrial scale: a scale-up framework for chemical processes in life cycle assessment studies. J Clean Prod 135:1085–1097. https://doi.org/10.1016/j.jclepro.2016.06.164
- Pini M, Rosa R, Neri P, Ferrari AM (2020) LCA application to chemical synthesis at laboratory scale. In: Maranghi S, Brondi C (eds) Life cycle assessment in the chemical product chain. Springer Nature Switzerland AG, Italy
- PlasticsEurope (2014c) Eco-profiles and Environmental Product Declarations of the European Plastics Manufacturers: Polyamide 6.6 (PA6.6). Brussels, Belgium
- PlasticsEurope (2014a) Eco-profiles and Environmental Product Declarations of the European Plastics Manufacturers: Polypropylene (PP). Brussels, Belgium
- PlasticsEurope (2014b) Eco-profiles and environmental product declarations of the european plastics manufacturers: high-density polyethylene (HDPE), low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE). Belgium, Brussels

- Sadasivuni KK, Saha P, Adhikari J et al (2020) Recent advances in mechanical properties of biopolymer composites: A review. Polym Compos 41:32–59. https://doi.org/10.1002/pc.25356
- Salminen J, Tikkanen S, Koskiaho J (2017) Kohti vesiviisasta kiertotaloutta (Towards a water-wise circular economy). Suomen ympäristökeskus (Finnish Environment Institute), Helsinki, Finland
- Saunders JH, Slocombe RJ (1948) The chemistry of the organic isocyanates. Chem Rev 43:203–218
- Shanmugam V, Mensah RA, Försth M et al (2021) Circular economy in biocomposite development: state-of-the-art, challenges and emerging trends. Compos Part C Open Access 5:100138. https:// doi.org/10.1016/j.jcomc.2021.100138
- Shi Y, Kamer PCJ, Cole-Hamilton DJ (2017) A new route to α,ω diamines from hydrogenation of dicarboxylic acids and their derivatives in the presence of amines. Green Chem 19:5460–5466. https://doi.org/10.1039/c7gc02838e
- S Silvonen, T Mäkelä (1996) Combined Heat and Power Production in Finland. In: eceee 1997 Summer Study on energy efficiency: sustainable energy opportunities for a greater Europe. Panel 3: Energy Efficiency options for industry pp 1–7
- Suwanmanee U, Varabuntoonvit V, Chaiwutthinan P et al (2013) Life cycle assessment of single use thermoform boxes made from polystyrene (PS), polylactic acid, (PLA), and PLA/starch: cradle to consumer gate. Int J Life Cycle Assess 18:401–417. https://doi. org/10.1007/s11367-012-0479-7
- M Torrell Gines (2016) Environmental Impact of Bioplastic Blends. Bachelor's thesis, Universitat Politécnica de Catalunya
- A-L Välimaa, P Tuunainen, H Siljander-Rasi, E Virtanen (2017) Rehua perunan käsittelyssä ja juustonvalmistuksessa syntyvistä sivujakeista (Feed from the side cuts arising from potato processing and cheese making). Luonnonvarakeskus (Natural Resources Institute Finland), Helsinki
- van den Oever M, Molenveld K (2017) Replacing fossil based plastic performance products by bio-based plastic products—technical feasibility. N Biotechnol 37:48–59. https://doi.org/10.1016/j.nbt. 2016.07.007
- Walker S, Rothman R (2020) Life cycle assessment of bio-based and fossil-based plastic: A review. J Clean Prod 261:121158–121173. https://doi.org/10.1016/j.jclepro.2020.121158
- Yadav P, Ismail N, Essalhi M et al (2021) Assessment of the environmental impact of polymeric membrane production. J Memb Sci 622:118987. https://doi.org/10.1016/j.memsci.2020.118987
- Yang Y, Lu W, Zhang X et al (2010) Two-step biocatalytic route to biobased functional polyesters from ω-carboxy fatty acids and diols. Biomacromol 11:259–268. https://doi.org/10.1021/bm901 112m
- Yang L, Wang X-C, Dai M et al (2021) Shifting from fossil-based economy to bio-based economy: status quo, challenges, and prospects. Energy 228:120533. https://doi.org/10.1016/j.energy.2021.120533
- Zheng J, Suh S (2019) Strategies to reduce the global carbon footprint of plastics. Nat Clim Chang 9:374–378. https://doi.org/10.1038/ s41558-019-0459-z

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Authors and Affiliations

Laura Äkräs¹ · Frans Silvenius² · Hossein Baniasadi¹ · Marjatta Vahvaselkä³ · Hannu Ilvesniemi³ · Jukka Seppälä¹

- ☑ Jukka Seppälä jukka.seppala@aalto.fi
- ¹ Polymer Technology, School of Chemical Engineering, Aalto University, Kemistintie 1, 02150 Espoo, Finland
- ² Bioeconomy and Environment, Natural Resources Institute Finland, Latokartanonkaari 9, 00790 Helsinki, Finland
- ³ Production systems, Natural Resources Institute Finland, Latokartanonkaari 9, 00790 Helsinki, Finland