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Life Cycle Assessment of Novel Antibacterial Polylactide-Hemp-Nanosilver-Biocomposites

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Abstract
Climate change is one of the most complex and time-sensitive crises humanity is facing, thereby driving a quest for more sustainable product alternatives across all sectors. In hygiene-critical sectors like medicine, the development of sustainable materials is imperative because of the predominant use of single use plastics. Polylactic acid (PLA) has emerged as a bio-based alternative for clinical applications because of its degradation profile. To further enhance antibacterial properties and sustainability performance, a novel biocomposite based on PLA with hemp hurd filled with silver nanoparticles (AgNP) was developed. Here we present a life cycle assessment (LCA) based on ISO 14040 and 14044 to evaluate the added value of the antibacterial biocomposite compared to conventional antibacterial materials from an environmental perspective. The environmental impacts are analyzed in comparison to virgin PLA, fossil-based polypropylene (PP) and high-density polyethylene (HDPE) combined in a disinfecting wipes and sterilized application. Our results show that the novel biocomposite has in general higher environmental impacts than its counterparts. Notably, our findings suggest a significant impact derived from high energy demand required for AgNP synthesis. To be able to compete with other materials, the energy demand needs to be minimized, which should be the focus of further research.

Keywords Life cycle assessment · Polylactide · Hemp · Silver nanoparticles · Antibacterial · Biocomposites

Introduction

Today, the world is facing natural disasters, diseases, and climate change due to the elevated ambient temperature leading to extreme weather conditions causing a threat to biodiversity and human habitat. Furthermore, climate change affects health by endangering clean air, safe drinking water, sufficient food, and secure shelter [1]. These triple planetary crises of climate change, biodiversity loss, and pollution stem from unsustainable patterns of consumption and production [2], with consumers and industry being equally responsible for engendering the underlying environmental impact. In addition to increasing scarcity and dependence on natural resources, the unsustainable usage patterns lead to amassing enormous levels of waste [2]. The greenhouse gas emissions generated from solid waste treatment and disposal are projected to increase from 2016 level of 1.6 billion tons (5% of global emissions) to 2.6 billion tons in 2050 if no corrective actions are implemented. While 44% of the global waste is constituted by food and green waste, 17% is comprised of paper and cardboard, and 12% by plastic [3]. Resource scarcity, the greenhouse effect, population growth and the quest for sustainable development have rekindled interest in renewable and biodegradable raw materials for energy and material use.

The COVID-19 pandemic demonstrated the vulnerabilities of modern society and despite existing commitments to reduce overall environmental footprints, the pandemic aggravated the global pollution crisis, in particular plastics pollution [2] because of the increased waste generated by disposable, single-use gloves and masks and disinfectants.

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Single-use products can be made reusable if an antibacterial property is introduced at the surface to prevent the spread of infection, e.g., methicillin-resistant staphylococcus aureus (MRSA) [4]. The use of novel bio-based antibacterial plastics based on PLA can mitigate the dependence on fossil resources and simultaneously the antibacterial property minimizing the transmission of diseases. The motivation of this work is to analyze if there is an added environmental value compared to conventional materials, which do not have an inherent antibacterial property, hence necessitating a separate treatment. Embedding silver nanoparticles (AgNP) in biopolymers inhibits microbial growth and enhances water vapor permeability, extending product shelf life. These AgNP improve the barrier capabilities, mechanical properties, and UV resistance of biopolymers such as PLA, whereas their antimicrobial activity is dictated by their shape, size, and surface charge. Thermal extrusion is an appropriate method of compounding AgNP with PLA [5]. For example, Momeni et al. analyzed the effect of inclusion of 1.5 wt.% AgNP on surface, chemical, thermal, and biocompatibility aspects of PLA bionanocomposites produced via melt mixing. They demonstrated enhanced stability and antimicrobial properties in resultant bionanocomposites [6].

Here we consider an antibacterial polylactide-hemp-nanosilver-biocomposite (AgNP-HH-PLA) to address the aforementioned challenge. We endeavor to bridge the data in the sustainability by assessing the environmental impact of this PLA-based antibacterial biocomposite. We compare the antibacterial material with a non-antibacterial control and perform a life cycle assessment (LCA) based on ISO 14040 [7] and ISO 14044 [8]. Based on the results, we infer the possible directions for further development of the AgNP-HH-PLA biocomposite.

While the technology and its development on how to produce the novel biocomposite has been developed by Khan et al., 2016 [9], this study focuses on the environmental impact of the newly developed material. While the environmental assessment of nanocomposites as well as connected antibacterial properties has been conducted before [10, 11], currently no LCA for AgNP-HH-PLA is available, thereby this research is closing a gap with regards to the environmental impact of the novel biocomposite also in comparison to other fossil-based and bio-based alternatives.

There are different ways to obtain antibacterial activity which also result in different degrees of hygiene. To reduce the microbial load on objects or the skin and mucous membrane, one can choose between disinfection or sterilization. Disinfection is used when a low degree of hygiene is sufficient. Sterilization is used for a high degree of hygiene [12, 13]. Disinfection is an infection prophylactic measure to reduce germs or to kill a significant portion of the microorganism population on objects so that there is no risk of infection. Physical and chemical methods are used for disinfection, with physical ones are more preferred because they are safer and more reliable, as well as environmentally and toxicologically safer than chemical methods. However, material compatibility must also be considered when choosing a process [12]. Other ways of removing microorganisms are sterilization processes. These are mainly used in the medical sector for critical products. A sterile medical device (MD) is completely free of viable microorganisms, including bacterial spores. Depending on the requirements, sterilization can be physical (heat: thermal sterilization; ionizing radiation: radiation sterilization) or chemical (use of gases) [12]. Depending on the type of sterilization, the packaging of the sterilized product has to be suitable for the sterilization process, e.g., permeable to the sterilization method but germ-tight to prevent recontamination [12, 13]. Next to these methods there are also materials which already have an antibacterial activity. In this work, we use AgNP. The antimicrobial efficacy of silver is enhanced at nanoscale, i.e., when the dimension of silver is <100 nm [14]. The Nanotechnology Products Database [15], lists the current use of silver in products by relative market share for each industrial division. The largest share falls to the medical applications (24%) which include bandages, catheters, and creams. Textiles are in second place (17.6%), followed by cosmetics (13.4%), and home appliances (9.4%) [15]. Hicks & Temizel-Sekeryan, 2019 divided nanosilver products into three categories of potential environmental benefits which are (1) human behavioral benefits: reductions in environmental impact as a result of changes in human behavior (e.g. textiles are laundered less frequently), (2) passive benefits: no reductions of the environmental impact of the product itself, but other environmental benefits (e.g. food storage containers reducing food losses and thus the environmental impact of food production due to prolongation of the edible lifetime of the food), (3) replacement benefits: replacing of another component (e.g. AgNP enabled bandages replace additional ointment needed for conventional bandages in medicine) [14].

Because of the negative impacts on the human body, the use of nanosilver in products which come in contact with food or the human body in a critical way is restricted and led to the establishment of the European Union Directive 2002/72/EC which governs the migration and release rates of silver nanoparticles (AgNP) into food and water. The migration testing of materials which may contact food is governed by the European Council Directive 82/711/EEC [9]. According to the EPA (US Environmental Protection Agency), a daily intake of silver (based on body weight) of 5 µg/kg is acceptable and does not lead to poisoning [16]. There are several ways to synthesize AgNPs. Either physical routes can be used, wet chemistry or bio-based syntheses.

The studies from Pourzahedi & Eckelman, 2015 a) and b) [17, 18], Bafana et al. [19] and Temizel-Sekeryan & Hicks [20] provide life cycle inventory data and assessment results for several synthesis routes. An overview of this data can be found in SI1. AgNPs are only an additive to gain antibacterial activity, while the main structure of the novel material is a biocomposite. Biocomposites are compounds made from biopolymers and a natural fiber reinforcing the biopolymer. The biopolymers can be classified into three groups, based on their origin (=feedstock) and their degradability (=end-of-life): (1) Bio-based and non-biodegradable: as long-lasting as possible; e.g. bio-polyethylene (bio-PE), bio-propylene (bio-PP); (2) Bio-based and biodegradable: synthesized from renewable raw materials and degradable; e.g. PLA; within biodegradable materials, compostable materials can be differentiated: some materials, like PLA, are degradable in industrial plants but they cannot rot in the compost. (3) fossil-based and biodegradable: conventional polymers, e.g., polycaprolactone (PCL).

For bio-based plastics, renewable resources such as sugar, starch, vegetable oils or cellulose are the source, as well as substances of animal origin (e.g., chitin) [21, 22]. Like conventional plastics, bioplastics are also used mostly in the packaging industry. By using renewable raw materials, finite resources can be conserved, CO_2 emissions can be reduced, and disposal options can be improved [22]. A major point of criticism is the required use of agricultural land and the use of the yields for plastic production instead for food, feed, or plants for material use. However, only 0.017% (in 2020) of agricultural land was used for bioplastic production [22, 23]. There are several natural fibers which can be used for a biocomposite. Hemp, e.g., belongs, just like flax, to the stem fibers, which are bast fibers. Other bast fibers are leaf fibers, e.g., sisal and abaca or fruit fibers, e.g., coconut and kapok. Hemp hurd (HH) is a residue which is left at the industrial hemp plant after the bast fibers and other commercial bio-products have been extracted. 70–80% of a hemp stem is made up of the hemp hurd, which has a porous structure [9]. In comparison to flax, hemp is coarser and stiffer and contains approx. 3% lignin while flax has only 2%. Like flax, hemp has a low elongation at break of up to 4%, a high strength of 40–80 cN/tex and moisture absorption of up to 30% [21]. Even though it is sometimes used as bio-based filler in construction and animal bedding the majority of the HH is disposed of by incineration or landfilling. When achieving a high yield per kg it has been used successfully in ethanol production. Hemp hurd exhibits antibacterial activity against *Escherichia coli (E. coli)* and has potential to be compounded with PLA, when using glycidyl methacrylate (GMA) as compatibilizer [9].

Although LCAs of biocomposites have been performed, to the authors' knowledge, their antibacterial properties as a function have not been discussed in the literature in detail (see section "Life Cycle Assessment"). Therefore, in this work we shall address the question of whether the novel biocomposite with its antibacterial properties is a more environmentally friendly solution compared to current conventional materials. We address this question by conducting a literature review and conducting a LCA of the biocomposite with the help of LCA for Experts (formerly known as GaBi) software [24], thereby assessing their environmental impacts. The results of LCA are then analyzed, to define the parameters, which need to be adjusted so that the novel biocomposite can compete with conventional materials.

Table 1 Overview of scenarios

Product system No	Material composition	Scenario 1	Scenario 2	Scenario 3
1	AgNP-HH-PLA (versions a/b/c)	Granulate	Ointment cap + antibacterial activity	Ointment cap + antibacterial activity
2	PLA	Granulate	Ointment cap + wipe & disinfecting spray	Ointment cap + EtO sterilization
3	PP	Granulate	Ointment cap + wipe & disinfecting spray	Ointment cap + EtO sterilization
4	HDPE	Granulate	Ointment cap + wipe & disinfecting spray	ointment cap + EtO sterilization

Materials and Methods

On basis of the results of the literature review, three different scenarios (S) have been analyzed. The modeled three scenarios are the following, listed in Table 1. Within each scenario four different product systems (P) are assessed. The antibacterial AgNP-HH-PLA is "product system 1" and supposed to be used for packaging in the medical sector. Additionally, product system 1 is assessed in 3 different variations of AgNP-content (a/b/c). Since the new material referred to as "product system 1" includes PLA, virgin PLA was selected as "product system 2", in order to compare how the additives change the environmental impacts. Furthermore, the fossil-based polymers polypropylene (PP) referred to as "product system 3" and high-density polyethylene (HDPE) as "product system 4" have been chosen. The plastics from product systems 3 and 4 have been chosen on basis of conventional packaging material on the market.

Scenario 1: Granulate

For scenario 1, an assessment is conducted on granulate level (1kg of material), without considering an application and the antibacterial properties. For scenario 2 a potential application where the antibacterial aspects are considered (e.g., as a cap or spout for ointment tubes or pharmaceutical spray (e.g., nasal spray)) with multiple use is analyzed. Therefore, the antibacterial activity for products system 1 and the use of wipes and disinfection spray for product systems 2–4 is considered. For scenario 3 a potential product application in the medical sector with single sterilization is considered. In comparison to scenario 2 for the product systems 2–4 an Ethylene oxide (EtO) sterilization process is considered, while the antibacterial activity is also considered for product system 1. The EtO sterilization is a low temperature sterilization method. Therefore, it is used for temperature-sensitive and moisture-sensitive materials and devices. The sterilization can be conducted with 100% EtO or 10% EtO and 90% hydrochlorofluorocarbon (HCFC) or with 8.6% EtO diluted in 91.4% carbon dioxide (CO_2). [25] For scenario 3, 100% EtO was chosen based on available data sets. The considered product application of an ointment cap was chosen as one potential representative of a product application for such materials. However, this product does not represent a certain market share or preferred application. The main purpose of inclusion of product level is to also include the use phase where the antibacterial properties can be considered.

System 1 must be able to compete with these in order to substitute the materials, i.e., which benefit would put the new system at the top of the market or which flaws may need to be solved, for better competition? It is important to emphasize that the new material has only been produced as granulate in laboratory scale so far. Going further into the development of the material, tests have to show whether the chosen use is applicable in practice and benchmarking should be conducted. In the use phase, the product made from antibacterial material would not require cleaning after use to kill bacteria and maintain an antibacterial activity while the other materials would need a disinfecting agent, as suggested by Pucciarelli et al. [26] For scenario 2 it is assumed that the cap product is used daily for half a year (180 days) after first opening the container. To gain antibacterial activity in the comparison materials, disinfecting wipes could be used after each use. For scenario 3, the EtO sterilization method has also been modeled to compare its environmental impact to the other antibacterial options, even though the practical use is questionable since once used the product would not be sterile anymore after single use and therefore a sterilization after each use and airtight packaging in order to keep it sterile until the next use would be necessary. Furthermore, the embedded antibacterial property of the material by AgNPs does not replace the sterilization process, which is often required in the medical sector.

Table 2 Suitability of sterilization methods for PLA, PP and HDPE (based on [28])

Polymer	Steam	Dry heat	Ethylene oxide	Gamma radiation	e-beam
PLA	Poor	Fair	Good	Good	Good
PP	Good	Fair	Good	Fair	Fair
HDPE	Poor	Poor	Good	Good	Good

Scenario 2: Disinfecting Wipes

Assuming one cap needs ca. 5 g of granulate (varying depending on the density of the polymer), 200 units could be produced from 1 kg. Under the assumption that after opening the ointment or spray container is usable for 6 months (180 days) and it is used daily, a reusable 100% PET wipe (16 cm × 15 cm; 1230g) is used combined with 5400 ml disinfectant spray. All the calculations and used data are shown in SI3.

The recipe for the disinfectant is taken from the WHO report [27] which has published a simple way for disinfectant production. For 1l of disinfectant, 830 ml ethanol (95%), 45 ml hydrogen peroxide (3%), 15 ml glycerin (98%) and 110 ml boiled water are used. The energy demand for boiling the water has been calculated by multiplying the temperature difference (ΔT) of 80 °C (from 20 °C to 100 °C) with the mass (m) of 0.11 kg and the heating capacity (c) of 4.18 kJ which is characteristic for water. As a result, an energy (E) of 36.784 kJ is needed.

Scenario 3: EtO Sterilization

In the case of sterilization, the EtO sterilization has been chosen since this type of sterilization is appropriate for all three polymers (PLA, PP and HDPE) according to Sastri [28] (see Table 2). To have the same function for all systems, i.e., protection against bacteria, the EtO sterilization would need to be conducted after or before each use phase which sums up to 180 sterilization processes. Using the sterilizer before the next use would erase the need for extra packaging which would be needed to keep the cap/spout sterile (and therefore antibacterial) until the next use. Even this situation does not reflect the reality, which would allow for an EtO sterilization only once before the product is used for the first time (single use).

The EtO sterilization is a low temperature sterilization method, used for temperature-sensitive and moisture-sensitive materials and devices. The sterilization can be conducted with 100% EtO or 10% EtO and 90% hydrochlorofluorocarbon (HCFC) or with 8.6% EtO diluted in

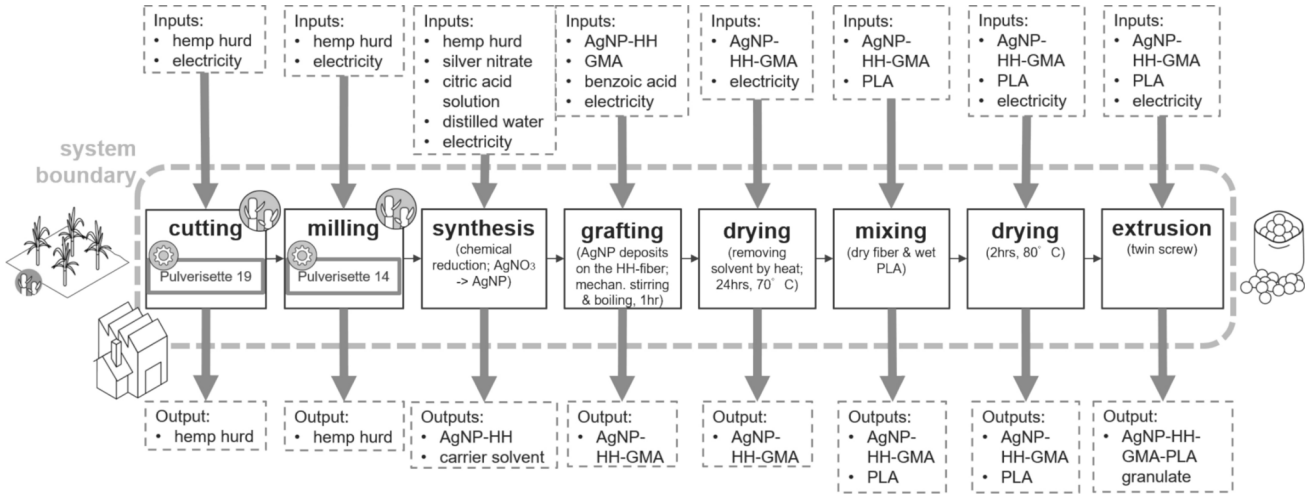

Fig. 1 Process flow diagram of AgNP-HH-PLA granulate production

Table 3 Overview of varied ratios of nanosilver particles

Alternative	a)		b)		c)	
	wt	wt%	wt	wt%	wt	wt%
Hemp hurd	10 g	99.50%	10 g	97.56%	10 g	95.24%
$AgNO_3$	0.016 g	0.16%	0.08 g	0.78%	0.16 g	1.52%
Citric acid	0.034 g	0.34%	0.17 g	1.66%	0.34 g	3.24%
Resulting AgNP-HH	10.05 g	100%	10.25 g	100%	10.5 g	100%

91.4% carbon dioxide (CO_2). Due to the available datasets in the modelling software, the 100% EtO option has been chosen for the modelling. Ethylene gas itself is flammable, explosive and an alkylating agent, and in addition, toxic and carcinogenic. When proteins react with EtO, the proteins can be denaturized [28]. For the EtO sterilization, the products have to be preconditioned and then inserted into the sterilization chamber, which is evacuated and then heated to 50–60°C. Moisture and EtO are introduced afterwards. The concentration of the gas ranges at 200–800 mg/l. The chamber pressure is maintained at sub-atmospheric pressure to preclude the diffusion or leakage of EtO. When the sterilization is finished, the EtO is removed and filtered sterile air is led into the chamber to aerate the product removing the EtO. The products are evaluated for sterility and acceptable levels of residual EtO [28].

Product System 1: Polylactide-hemp-nanosilver-biocomposite (AgNP-HH-PLA)

The fundamentals to this work were already established by Khan et al. [9]. The data collected in 2015 has been used to review the environmental performance of this material. The production process of the novel AgNP-HH-PLA granulate at a laboratory scale is shown in the flow diagram in Fig. 1. It starts with cutting the delivered HH into 0.75 mm pieces with a milling machine (Pulverisette 19) for which electricity is needed. The small HH chips are then ground in the electrical milling machine (Pulverisette 14) into smaller pieces of 0.08 mm mean size. In the synthesis, the HH powder is mixed with silver nitrate, citric acid solution and distilled water using electricity. This chemical reduction creates the AgNP-HH by converting $AgNO_3$ to AgNP and leaves the carrier solvent as residue. Afterwards the AgNP gets deposited on the HH-fiber in the grafting stage, where GMA and benzoic acid are added. For the mechanical stirring and boiling, 1 h of electricity is used resulting in AgNP-HH-GMA product. To remove the solvent, a 24-h drying process at 70 °C is needed using heat produced by electricity. Then, wet PLA is mixed by hand to the dry AgNP-loaded HH-fiber, leading to another drying step at 80 °C for 2 h. The extrusion of the AgNP-HH-PLA with a twin screw (Thermo Scientific Process 11) is the last step of producing the granulate. A general level of 5% loss has been assumed, which results in a 95% yield.

The AgNP-HH-PLA granulate was produced with three different ratios of nanosilver particles and therefore also a changing amount of PLA has been used. The different component proportions are listed in Tables 3 and 4 for the variants a, b, and c.

The amount of AgNP-HH has been scaled up to produce 1 kg of granulate.

The nanosilver particle synthesis used silver nitrate and a chemical reduction method with citric acid and HH. In

Table 4 Overview of varied ratios of AgNP-HH and PLA

Alternative	a)		b)		c)	
	wt	wt%	wt	wt%	wt	wt%
AgNP-HH	100 g	10%	200 g	20%	300 g	30%
GMA	10 g	1%	10 g	1%	10 g	1%
Benzoic Acid	15 g	1.5%	15 g	1.5%	15 g	1.5%
PLA	875 g	87.5%	775 g	77.5%	675 g	67.5%
Resulting AgNP-HH-PLA	1000 g	100%	1000 g	100%	1000 g	100%

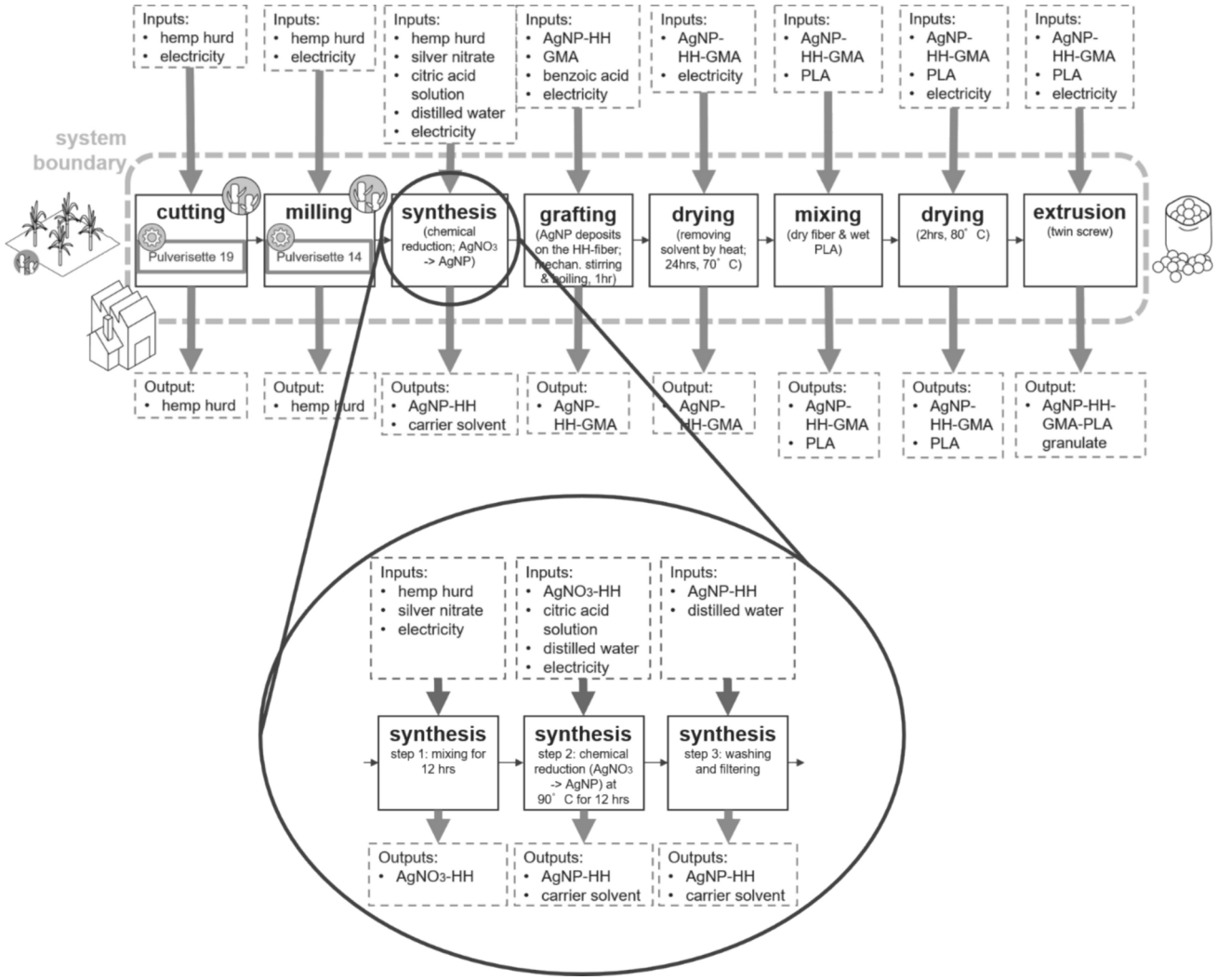

Fig. 2 Detailed description of the various steps for the AgNP synthesis

the first step, HH and silver nitrate were mixed for 12 h on a hot plate stirrer. Afterwards, this mixture was added in citric acid and water solution and stirred and heated for another 12 h at 90 °C. The output was HH with nanosilver particles. In the third step, the mixture has been washed with distilled water and filtered (see Fig. 2). The synthesis of the nanosilver particles has been conducted in-house at a laboratory. To check to what extent the selected synthesis route has an influence on the environmental impact and whether another route would be more environmentally friendly, it was compared with the LCAs of other routes from the studies Pourzahedi & Eckelman, 2015 a) [17], supplemented by b) [18], Temizel-Sekeryan & Hicks, 2019 [20] and Bafana et al., 2018 [19]. A summary of the analyzed synthesis routes and the results can be found in SI1.

Product System 2: Polylactic Acid (PLA)

For the System 2 the Ingeo.™ PLA by NatureWorks was selected. This PLA is made from corn and has been used in

system 1 as well. After the starch is extracted, hydrolysis is needed to get glucose, which is a monosaccharide. Along with water and microorganisms, lactic acid is produced with CO_2 as by-product. Dehydration splits the water from the lactic acid and lactide is formed. Together with a catalyst, the polymerization step generates PLA [29].

Product System 3: Polypropylene (PP)

After PE, PP has the second largest market share, mainly used in the packaging and fiber sectors, and possesses low density, high melting point and good processability. The PP property profile can be adjusted as required by appropriate measures and can be tailored as robustly rigid (e.g., garden furniture) to soft flexible fibers (e.g., baby diapers), and applications such as heat-resistant (e.g., microwave containers) or melting for heat-sealed food packaging. PP grades are both physiologically inert (i.e., applicable in medicine) and food safe. Low-emission synthesis processes have been developed and material recycling is well developed under ideal conditions, although color and odor have quality implications [21]. Crude oil is extracted and transported to the oil refinery where it is converted into Propene gas. By steam cracking, olefines are formed which are transformed into monomers by chemical reactions. The polymerization process turns the monomers into PP.

Product System 4: High Density Polyethylene (HDPE)

PE is the most consumed of all thermoplastics, and preferred option for household goods, storage, and transport containers, but also packaging films and bags. PE is available in a wide variety of grades and therefore also in a wide variety of shaft profiles. PE has a low density compared to other plastics, very good electrical properties, low water absorption and water vapor permeability, and high chemical resistance. HDPE is synthesized either by the Ziegler process using titanium halides, titanium esters and aluminum alkyls as catalysts or by the Phillips process using a chromium oxide catalyst at low process pressures (20–40 bar at 85–180 °C or 1–50 bar at 20–150°C). The ethylene molecules are joined in an insert polymerization to form linear macromolecules. The low degree of branching increases the density (0.942–0.965 g/cm.^{3}) and the degree of crystallinity (60–80%). PE can be sterilized by steam, ethylene oxide, or ionizing radiation. There are no food law or pharmaceutical concerns with the use of PE [21].

Life Cycle Assessment

The LCA is performed based on ISO 14040 [7] and 14044 [8] with the Sphera LCA for experts (GaBi) software by Sphera Solutions GmbH (version 10.7.0.183) [24]. The used datasets were taken from the Sphera (GaBi) database (GaBi Professional Database; content version 2023.1) [30] and supplemented by the Ecoinvent database (Ecoinvent 3.8; content version 2022.1) [31] in case a suitable dataset was not available in the Sphera (GaBi) database. For this study, the Environmental Footprint (EF) 3.0 was chosen as the main LCIA method [32]. The intended applications of the results are to evaluate the potential environmental impact of the novel antibacterial polylactide-hemp-nanosilver biocomposite (AgNP-HH-PLA) and perform a comparison with virgin PLA, fossil-based PP, and HDPE which need an additive to gain the antibacterial property. The reasons for conducting the study is to provide information on the life cycle environmental performance of the material to researchers for further development of the plastic-blend. The production process of the antibacterial AgNP-HH-PLA biocomposite is yet to be optimized, since the granulate is only produced on a laboratory scale. Whenever an assumption has been made, its limitations have been discussed.

Declared Unit and Functional Unit

A literature review was conducted to find out how antibacterial properties were already considered in existing LCAs. The focus was on the used functional units (FU). The proposed application of the novel biocomposite in the food packaging sector from Khan et al., 2016 was switched to the medical sector because of its increasing importance, showed by the COVID-19 pandemic. The detailed process steps of the literature review as well as the found studies can be found in SI2.

The Functional Unit (FU) is used to clearly specify and quantify the performance characteristics of a product system. “Comparisons between systems shall be made on the basis of the same function(s), quantified by the same FU(s) in the form of their reference flows” [8]. In case of intermediate products (e.g., polymer granulate), a Declared Unit (DU) shall be used, since the intended use and therefore the functions have not been specified [33].

After analyzing the studies, the following approaches to define a FU for an antibacterial product and a DU for intermediate products, were applied for the different scenarios:

- Scenario 1: DU: Production of 1 kg of plastic granulates in Australia (without considering the antibacterial activity)

- Scenario 2: FU: Production of 200 caps (5g of granulate per ointment cap) in Australia. For product system 1 antibacterial activity and for product systems 2–4 wipe & disinfecting spray (multi use) are considered (5400 ml disinfectant and 1230 g wipes).
- Scenario 3: FU: Production of 200 caps (5g of granulate per ointment cap) in Australia. For product system 1 antibacterial activity and for product systems 2–4 + a EtO sterilization (single use) are considered.

As Miseljic and Olsen [34] emphasized, the FU should include the antibacterial activity since this is the special function. Without it, there would be no need to add such a rare and environmentally damaging material like silver to the extraction process. The mass-based approach could be used, when not a product but only the production steps up to the, e.g., polymer granulate are considered, which would end up in using a DU of 1 kg granulate of the antibacterial and the non-antibacterial polymer. "Declared units are useful in research as the data can be readily transferable to other research efforts" [19]. This transfer opportunity has been used for the comparison of the AgNP synthesis routes.

Modelling Framework and System Boundaries

The system boundaries for the assessment are shown in Fig. 3. The life cycle phases manufacturing and distribution (indicated with dotted lines) have been excluded from the assessment. This also includes the (drying and) injection molding process step to produce the ointment caps in scenarios 2 and 3. From the preliminary assessment [35], it was found out the processing phase that includes drying and injection molding for PLA-based variants and injection moulding for PP- and HDPE-based variants, has a share of up to 5% of the total environmental impacts for all the product systems. Therefore, in accordance to the ISO 14044 and methodologies for the cut-off criteria developed by the European Union [36], it was decided that the processing phase was not considered for this study. However, the absolute environmental impacts of the processing phase, along with their individual share towards environmental impacts are shown in SI5, using the secondary data [37]. From the values, it can be seen that the share of processing of Silver Nano Composites, on an average contributes to less than 1% of the total impacts and for processing of PLA, PP and HDPE, it is at a max up to 5% of the total impact across all the impact categories. These results are in agreement to our initial assumption of not considering the processing phase in our study. With the availability of primary data, the impacts of the processing phase can be calculated better in the future. The foreground system, and therefore the processes specific to the analyzed system, is only the production of the AgNP-HH-PLA granulate and the following is the manufacturing of the product. The pre-processed raw materials and chemicals used as well as the PLA, PP and HDPE granulate have been purchased from the supplier. Even though 84% of plastic waste is sent to landfill and only 13% is recycled in Australia [38], the application in the medical sector would lead to incineration at the end-of-life stage, along with the other medical waste. Hence, only one end-of-life option has been modeled, and no credits have been assigned. The incineration process for the different materials has been modeled in scenarios 2 and 3. All limitations which result from the assumptions for the modelling are discussed in chapter results and discussions.

Each life cycle of the three scenarios with each of the four product systems was modeled in the software, showing the input and output flows into and out of the assessed system

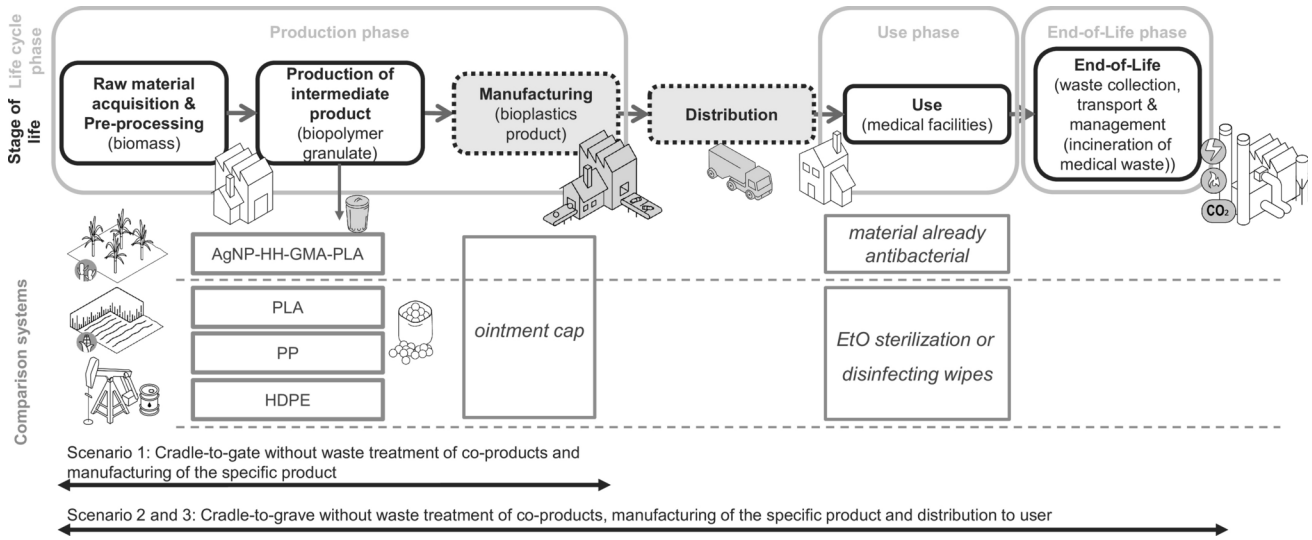

Fig. 3 System boundaries for scenarios 1, 2 and 3

boundary, as well as the considered life cycle stages. The manufacturing process of the product has been excluded from the system boundary because the polymers have similar melting temperatures and therefore exhibit similar energy consumption during the manufacturing process. Also, the variations in distribution were excluded since similar transportation routes have been assumed. Hereby it has been omitted that the additional materials, i.e., disinfectant and wipes, would need to be transported as well and therefore lead to additional emissions. The impacts caused by the infrastructure and buildings are not included in the assessment. This includes the EtO sterilizer, which would be an additional burden during the use phase for scenario 3. The waste treatment of the co-products has not been included into the system boundary because of lack of data and early phase of development do not allow specifying this part of the life cycle for the new material. Therefore, it has been excluded from all the materials to maintain comparability.

LCI Model and Data

The data collection can be found in SI3, which lists all the collected data along with the unit process, input or output type, material type, name, value and unit, the source, and the type of data, i.e., measured, calculated, or estimated. Furthermore, the region where this data is applicable has been documented, along with any assumptions and discrepancies arising from the modelling of the systems.

Some primary data has been derived from the previous work of the co-authors during the production process of the AgNP-HH-PLA granulate, but only 28% of this data can be classified as primary data. Further information has been collected as secondary data using calculations on literature/ estimated/ measured parameters by applying suitable equations. Some estimations were made based on general industry data or machine data, taken from the manufacturer's website. The transportation routes have been calculated with the online platform [39] and are listed in SI4. The inventory data for LCA is obtained from the Sphera (GaBi) and Ecoinvent databases.

Results and Discussions

LCA Results of Scenario 1

The modelling of the production of granulates, including the transportation to the laboratory in Australia, have led to the following results, as shown in Fig. 4 (relative values) and Table 5 (absolute values). The results show that in general the impacts of product system 1 (AgNP-HH-PLA) are significantly higher in comparison to the environmental impacts of the other product systems (PLA, PP and HDPE) across different impact categories. The impacts of product system 1 with its three different variants (ratios of AgNP-HH) increase with a decreasing fraction of PLA and increase of AgNP-HH. Since the impact is driven by the energy demand which comes mainly from the AgNP synthesis, a higher fraction of AgNP signifies larger fraction of material to be synthesized and therefore requiring higher energy input. The distinction of the origin of the impacts, i.e., generated by energy, material, transportation, or discharge, shows that in general the energy demand has the biggest influence on these results. The environmental impacts of the product system 1 increases with a higher ratio of AgNPs. In the case of the granulates made of PLA, PP and HDPE, the use of bio-based feedstocks in PLA results in lower environmental impacts in indicators like human toxicity (cancer inorganics, non-cancer metals) and resource use (fossils and metals) in comparison to the PP and HDPE granulates. However, the use of fertilizers and pesticides during the cultivation of biomass to produce PLA granulates results in the higher environmental impacts across all the other impact categories unlike PP and HDPE, which are produced from fossil-based feedstocks.

Apart from calculating the potential environmental impacts of synthesis of AgNP-HH-PLA product system, an assessment of the raw material influence has been conducted due to the high energy consumption during the synthesis of product system 1 (AgNP-HH-PLA). This analysis ensured the results were independent from the location of the process and therefore also from the energy source. This analysis shall help to focus objectively on the environmental performance of the raw materials, and thereby help us in the selection of material compositions in the future. This approach is critical as renewable energy sources are researched intensively and therefore changes in the energy supply are likely in the near future.

While the product systems 2, 3 and 4 (PLA, PP and HDPE) have a main contribution by the used polymers, for product system 1 (AgNP-HH-PLA) various chemicals used for the AgNP synthesis as well as the HH are included. The environmental impact for AgNP-HH-PLA considering the different raw material is shown in Fig. 5. Depending on the AgNP-HH: PLA ratio (variations a, b, and c) the silver nitrate impact rises. In general, the PLA causes the highest impacts but the impact decreases when using lower PLA content, as expected. Compared to the other materials, HH exhibits the highest impact in climate change—biogenic and eutrophication, freshwater. GMA is the highest contributor to ozone depletion. The silver nitrate impact rises from variation a to c, which was expected since the used amount of AgNPs increases from a to c. Also, the effect of silver

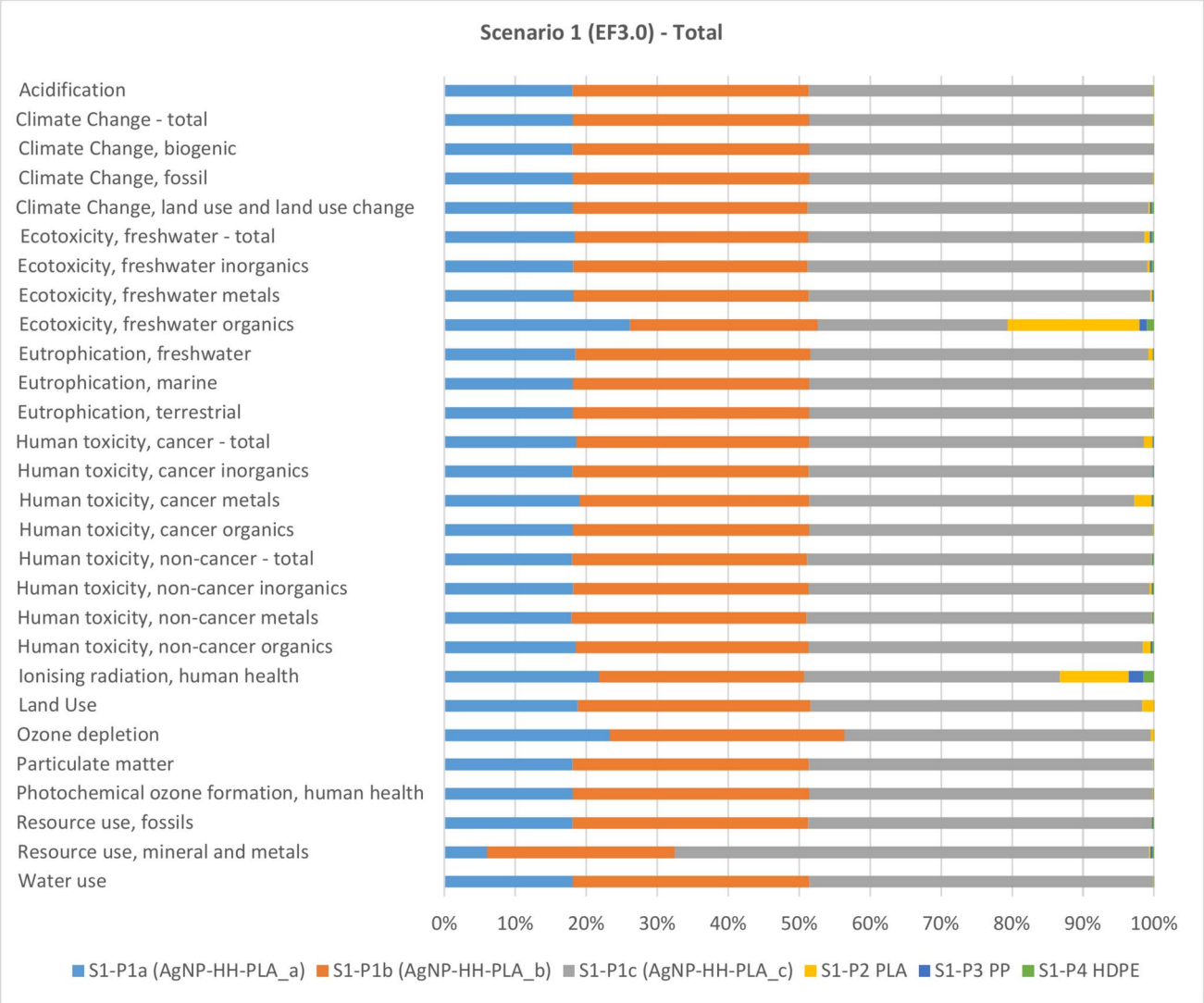

Fig. 4 LCA-results for Scenario 1–relative values

nitrate on the resource use, minerals and materials singularly was anticipated.

LCA Results of Scenarios 2 and 3

The modelling of scenarios 2 and 3 has been performed using separate models. The results are calculated by adding the impact of the granulate including the transportation of the materials to the laboratory, and the additives (disinfecting wipes or EtO sterilization) needed in the use phase and the end-of-life scenario, as shown in Fig. 6 (relative values) and Table 6 (absolute values). The diagrams show that in general the impacts of system 1 are consistently and significantly high. The distinction of the origin of the impacts shows that the energy demand bears the biggest effect on these results. Only the threat for human health by the ionizing radiation from scenario 2 from PLA, PP and HDPE is almost as high as the impact from the AgNP-HH-PLA. Also, land use shows a difference between the bio-based materials and the fossil-based ones. Additional results can be found in SI5.

Limitations and Data Gaps

LCI Data Quality and Data Availability

Even though the AgNP-HH-PLA (product system 1) was produced in the laboratory, unfortunately only few data values were measured and therefore limited primary inventory data exists. The missing data was sourced from general processes from the literature, exemplary technical machine data, assumptions, or calculations. Therefore, the data basis on which the modelling is conducted is not as reliable as

Table 5 LCA results for Scenario 1–absolute values

Impact category	S1-P1a AgNP-HH-PLA_a	S1-P1b AgNP-HH-PLA_b	S1-P1c AgNP-HH-PLA_c	S1-P2 PLA	S1-P3 PP	S1-P4 HDPE	Unit/DU
Acidification	3.75×10^{0}	6.90×10^{0}	1.01×10^{0}	1.49×10^{-2}	9.26×10^{-3}	9.16×10^{-3}	Mole of H + eq.
Climate Change—total	7.42×10^{2}	1.36×10^{3}	1.99×10^{3}	2.80×10^{0}	2.35×10^{0}	2.18×10^{0}	kg CO_2 eq.
Climate Change, biogenic	9.31×10^{-1}	1.72×10^{0}	2.50×10^{0}	8.07×10^{-5}	2.43×10^{-3}	2.39×10^{-3}	kg CO_2 eq.
Climate Change, fossil	7.41×10^{2}	1.36×10^{3}	1.98×10^{3}	2.80×10^{0}	2.35×10^{0}	2.18×10^{0}	kg CO_2 eq.
Climate Change, land use and land use change	1.50×10^{-2}	2.74×10^{-2}	3.99×10^{-2}	1.72×10^{-4}	2.36×10^{-4}	2.31×10^{-4}	kg CO_2 eq.
Ecotoxicity, freshwater—total	1.50×10^{3}	2.69×10^{3}	3.88×10^{3}	6.12×10^{1}	2.46×10^{1}	2.42×10^{1}	CTUe
Ecotoxicity, freshwater inorganics	1.30×10^{3}	2.37×10^{3}	3.44×10^{3}	2.81×10^{1}	2.18×10^{1}	2.16×10^{1}	CTUe
Ecotoxicity, freshwater metals	1.52×10^{2}	2.77×10^{2}	4.02×10^{2}	2.52×10^{0}	1.16×10^{0}	9.77×10^{-1}	CTUe
Ecotoxicity, freshwater organics	4.29×10^{1}	4.34×10^{1}	4.40×10^{1}	3.05×10^{1}	1.68×10^{0}	1.68×10^{0}	CTUe
Eutrophication, freshwater	4.39×10^{-4}	7.85×10^{-4}	1.13×10^{-3}	1.53×10^{-5}	1.93×10^{-6}	1.89×10^{-6}	kg P eq
Eutrophication, marine	8.07×10^{-1}	1.48×10^{0}	2.15×10^{0}	4.81×10^{-3}	3.00×10^{-3}	2.99×10^{-3}	kg N eq
Eutrophication, terrestrial	8.80×10^{0}	1.62×10^{1}	2.35×10^{1}	4.81×10^{-2}	3.29×10^{-2}	3.27×10^{-2}	Mole of N eq.
Human toxicity, cancer—total	8.01×10^{-8}	1.42×10^{-7}	2.03×10^{-7}	5.31×10^{-9}	5.07×10^{-10}	4.96×10^{-10}	CTUh
Human toxicity, cancer inorganics	2.04×10^{-19}	3.76×10^{-19}	5.48×10^{-19}	4.56×10^{-23}	9.91×10^{-22}	9.72×10^{-22}	CTUh
Human toxicity, cancer metals	4.07×10^{-8}	6.91×10^{-8}	9.77×10^{-8}	5.21×10^{-9}	3.78×10^{-10}	3.75×10^{-10}	CTUh
Human toxicity, cancer organics	3.95×10^{-8}	7.26×10^{-8}	1.06×10^{-7}	9.91×10^{-11}	1.30×10^{-10}	1.21×10^{-10}	CTUh
Human toxicity, non-cancer—total	2.06×10^{-6}	3.79×10^{-6}	5.55×10^{-6}	1.72×10^{-8}	1.64×10^{-8}	1.62×10^{-8}	CTUh
Human toxicity, non-cancer inorganics	4.97×10^{-7}	9.07×10^{-7}	1.32×10^{-6}	8.11×10^{-9}	4.92×10^{-9}	4.81×10^{-9}	CTUh
Human toxicity, non-cancer metals	1.55×10^{-6}	2.88×10^{-6}	4.22×10^{-6}	6.37×10^{-9}	1.14×10^{-8}	1.12×10^{-8}	CTUh
Human toxicity, non-cancer organics	5.13×10^{-8}	9.10×10^{-8}	1.31×10^{-7}	3.03×10^{-9}	6.81×10^{-10}	6.93×10^{-10}	CTUh
Ionising radiation, human health	4.01×10^{-1}	5.32×10^{-1}	6.65×10^{-1}	1.78×10^{-1}	3.78×10^{-2}	2.77×10^{-2}	kBq U235 eq.
Land Use	1.01×10^{3}	1.77×10^{3}	2.52×10^{3}	8.74×10^{1}	9.46×10^{-1}	8.22×10^{-1}	Pt
Ozone depletion	9.71×10^{-9}	1.38×10^{-8}	1.80×10^{-8}	2.12×10^{-10}	1.13×10^{-12}	8.24×10^{-13}	kg CFC-11 eq.
Particulate matter	3.66×10^{-5}	6.74×10^{-5}	9.81×10^{-5}	1.61×10^{-7}	1.17×10^{-7}	1.25×10^{-7}	Disease incidences
Photochemical ozone formation, human health	2.24×10^{0}	4.11×10^{0}	5.98×10^{0}	1.22×10^{-2}	8.85×10^{-3}	9.13×10^{-3}	kg NMVOC eq.
Resource use, fossils	8.13×10^{3}	1.49×10^{4}	2.17×10^{4}	4.03×10^{1}	7.39×10^{1}	7.27×10^{1}	MJ
Resource use, mineral and metals	3.71×10^{-5}	1.63×10^{-4}	4.14×10^{-4}	7.91×10^{-7}	1.71×10^{-6}	1.69×10^{-6}	kg Sb eq.
Water use	2.91×10^{2}	5.35×10^{2}	7.78×10^{2}	1.54×10^{0}	3.58×10^{-1}	3.41×10^{-1}	m^3 world equiv.

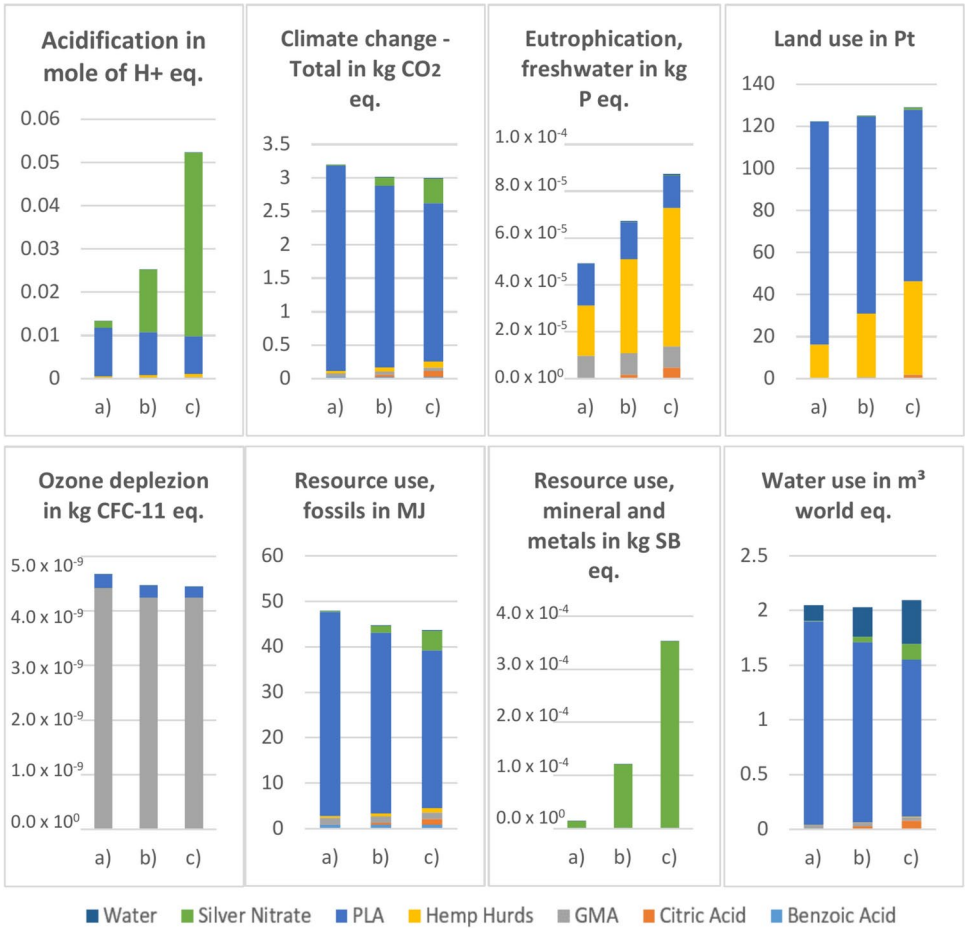

Fig. 5 LCA results for scenario 1–product system 1 a,b,c for the comparison of the influences of the various components of AgNP-HH-PLA a,b,c

measured primary data utilized for all aspects. Measuring the energy, material, and waste flows during the production of the novel biocomposite would lead to a more accurate assessment of the scenarios.

While using the Sphera (GaBi) database, specific datasets were not always available. For the incineration of the AgNP-HH-PLA a starch-PLA blend dataset has been used instead of a cellulose dataset. This substitution for the hemp hurd-based material could affect the results because of the calorific value of the material which is important for the incineration process. The calorific value of starch is 17.61 kJ/g of cellulose 17.3 kJ/g and of hemp 17 kJ/g. [40] These differences of the calorific values are small (not more than 3.5%), which is why the chosen dataset was considered sufficient. The dataset used for GMA was the Ecoinvent dataset market for chemical organic–global because no other specific data was available. This average inventory related to the production of an unspecified organic chemical for GMA has been based on a case study conducted by Nessi et al. [33, 41].

While the comparison materials were all modeled by choosing the respective granulate and the transportation route, the production of the novel granulate has been modeled in more detail. Furthermore, reverting to datasets which do not fit exactly, but represent a similar matter poses an inconsistency. Regional and temporal differences were considered when choosing the transportation route, type of vehicle and origin of the fuel. When no dataset was available for the modelled region, a dataset from another region was used, e.g., for incineration processes due to limitations in availability of datasets for Australia. The end-of-life datasets for the incineration of the different polymers were not available for Australia. However, to be able to conduct a comparison of the different materials, the same region was used in all systems. This way, they all have the same deviation and no further differences because various localities influence the results. The system boundary is consistent for the product systems. However, in scenario 2, the end-of-life of the disinfecting agent and the wipes have not been included since the disinfectant dissolves into air after use and the reusable PET wipe can be used for a longer period of time and therefore would not be disposed as trash along with the ointment tube or spray bottle, similar to using cleaning spray for glasses with a reusable wipe. And, from these packaging containers only the cap or spout, which is made of the antibacterial material and compared to the other

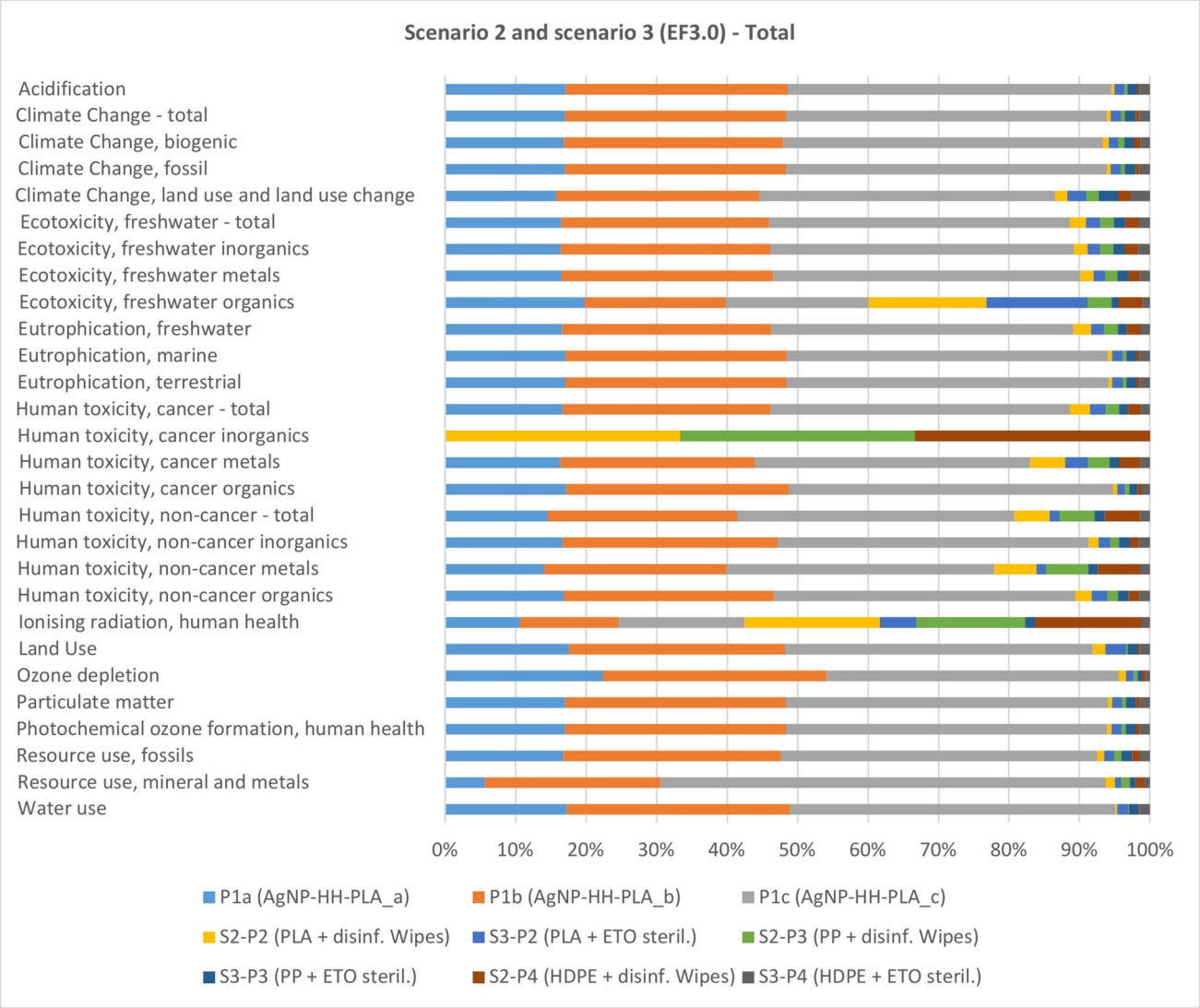

Fig. 6 LCA results for scenarios 2 and 3–relative values

materials, are included in the end-of-life. The other parts of the containers are not considered since they are assumed to be the same. All the listed inconsistencies and limitations may lead to potential errors which is why a reliable data basis is important as well as a well-planned approach when developing a new material or product. Furthermore, the setting of the system boundary without manufacturing, distribution and waste treatment limits the significance of the LCA when evaluating scenarios 2 and 3.

Consideration of Co-products

There were no credits assigned for the potential use of co-products from incineration. An assignment of credits is conducted especially for the waste flows, to show their potential use in another form than as waste from the current process. This allocation is useful especially when different scenarios are compared, such as incineration, landfill, or recycling at the end-of-life. In the present work, no comparison of different end-of-life strategies has been conducted.

For the waste treatment of co-products neither emissions nor benefits are considered. The waste treatment, similar to the loss of hemp hurd during the cutting processes or the release of carrier solvent but also the outputs of the incineration processes were not included in the model. The option to let the general loss during the production phase re-enter the production phase when the next batch is produced could lower this waste. The reason for this is the lack of data for the waste treatment due to the early stage of the life cycle for this new material. It has not yet been discussed whether some outputs could be reentered into the production process or used in another way. Using incineration as end-of-life option may have positive benefits like energy recovery, but also negative aspects like emissions. Furthermore, the waste

Table 6 LCA results for scenarios 2 and 3–absolute values

Impact category	P1a (AgNP-HH-PLA_a)	P1b (AgNP-HH-PLA_b)	P1c (AgNP-HH-PLA_c)	S2-P2 (PLA+dis-inf. Wipes)	S3-P2 (PLA+ETO steril.)	S2-P3 (PP+disinf. Wipes)	S3-P3 (PP+ETO steril.)	S2-P4 (HDPE+dis-inf. Wipes)	S3-p4 (HDPE+ETO steril.)	Unit/FU
Acidification	3.75×10^{0}	6.90×10^{0}	1.01×10^{1}	9.17×10^{-2}	3.15×10^{-1}	8.60×10^{-2}	3.09×10^{-1}	8.59×10^{-2}	3.09×10^{-1}	Mole of H+eq.
Climate Change—total	7.42×10^{2}	1.36×10^{3}	1.99×10^{3}	2.27×10^{1}	6.24×10^{1}	2.54×10^{1}	6.50×10^{1}	2.52×10^{1}	6.49×10^{1}	kg CO2 eq.
Climate Change, biogenic	9.31×10^{-1}	1.72×10^{0}	2.50×10^{0}	4.74×10^{-2}	7.24×10^{-2}	5.01×10^{-2}	7.51×10^{-2}	5.01×10^{-2}	7.51×10^{-2}	kg CO2 eq.
Climate Change, fossil	7.41×10^{2}	1.36×10^{3}	1.98×10^{3}	2.27×10^{1}	6.23×10^{1}	2.53×10^{1}	6.49×10^{1}	2.52×10^{1}	6.48×10^{1}	kg CO2 eq.
Climate Change, land use and land use change	1.50×10^{-2}	2.74×10^{-2}	3.99×10^{-2}	1.62×10^{-3}	2.54×10^{-3}	1.70×10^{-3}	2.61×10^{-3}	1.69×10^{-3}	2.61×10^{-3}	kg CO2 eq.
Ecotoxicity, freshwater—total	1.50×10^{3}	2.69×10^{3}	3.88×10^{3}	2.11×10^{2}	1.8×10^{2}	1.74×10^{2}	1.48×10^{2}	1.74×10^{2}	1.48×10^{2}	CTUe
Ecotoxicity, freshwater inorganics	1.30×10^{3}	2.37×10^{3}	3.44×10^{3}	1.57×10^{2}	1.39×10^{2}	1.51×10^{2}	1.32×10^{2}	1.51×10^{2}	1.32×10^{2}	CTUe
Ecotoxicity, freshwater metals	1.52×10^{2}	2.77×10^{2}	4.02×10^{2}	1.76×10^{1}	1.48×10^{1}	1.62×10^{1}	1.34×10^{1}	1.60×10^{1}	1.33×10^{1}	CTUe
Ecotoxicity, freshwater organics	4.29×10^{1}	4.34×10^{1}	4.40×10^{1}	3.62×10^{1}	3.11×10^{1}	7.37×10^{0}	2.25×10^{0}	7.36×10^{0}	2.25×10^{0}	CTUe
Eutrophication, freshwater	4.39×10^{-4}	7.85×10^{-4}	1.13×10^{-3}	6.57×10^{-5}	4.80×10^{-5}	5.22×10^{-5}	3.45×10^{-5}	5.22×10^{-5}	3.45×10^{-5}	kg P eq.
Eutrophication, marine	8.07×10^{-1}	1.48×10^{0}	2.15×10^{0}	2.68×10^{-2}	6.91×10^{-2}	2.49×10^{-2}	6.72×10^{-2}	2.49×10^{-2}	6.72×10^{-2}	kg N eq.
Eutrophication, terrestrial	8.80×10^{0}	1.62×10^{1}	2.35×10^{1}	2.87×10^{-1}	7.50×10^{-1}	2.72×10^{-1}	7.35×10^{-1}	2.72×10^{-1}	7.34×10^{-1}	Mole of N eq.
Human toxicity, cancer—total	8.01×10^{-8}	1.42×10^{-7}	2.03×10^{-7}	1.37×10^{-8}	1.08×10^{-8}	8.96×10^{-9}	6.05×10^{-9}	8.94×10^{-9}	6.04×10^{-9}	CTUh
Human toxicity, cancer inorganics	1.97×10^{-19}	3.70×10^{-19}	5.55×10^{-19}	2.85×10^{-15}	1.55×10^{-20}	2.85×10^{-15}	1.65×10^{-20}	2.85×10^{-15}	1.64×10^{-20}	CTUh
Human toxicity, cancer metals	4.07×10^{-8}	6.91×10^{-8}	9.77×10^{-8}	1.24×10^{-8}	8.23×10^{-9}	7.53×10^{-9}	3.41×10^{-9}	7.52×10^{-9}	3.40×10^{-9}	CTUh
Human toxicity, cancer organics	3.94×10^{-8}	7.26×10^{-8}	1.06×10^{-7}	1.38×10^{-9}	2.60×10^{-9}	1.43×10^{-9}	2.65×10^{-9}	1.42×10^{-9}	2.63×10^{-9}	CTUh
Human toxicity, non-cancer—total	2.06×10^{-6}	3.79×10^{-6}	5.55×10^{-6}	7.05×10^{-7}	2.01×10^{-7}	7.05×10^{-7}	2.02×10^{-7}	7.05×10^{-7}	2.01×10^{-7}	CTUh
Human toxicity, non-cancer inorganics	4.97×10^{-7}	9.07×10^{-7}	1.32×10^{-6}	4.01×10^{-8}	5.02×10^{-8}	3.70×10^{-8}	4.72×10^{-8}	3.69×10^{-8}	4.71×10^{-8}	CTUh
Human toxicity, non-cancer metals	1.55×10^{-6}	2.88×10^{-6}	4.22×10^{-6}	6.61×10^{-7}	1.48×10^{-7}	6.67×10^{-7}	1.54×10^{-7}	6.67×10^{-7}	1.54×10^{-7}	CTUh
Human toxicity, non-cancer organics	5.13×10^{-8}	9.10×10^{-8}	1.31×10^{-7}	6.97×10^{-9}	6.85×10^{-9}	4.66×10^{-9}	4.54×10^{-9}	4.67×10^{-9}	4.55×10^{-9}	CTUh
Ionising radiation, human health	3.97×10^{-1}	5.27×10^{-1}	6.69×10^{-1}	7.17×10^{-1}	1.93×10^{-1}	5.78×10^{-1}	5.35×10^{-2}	5.67×10^{-1}	4.33×10^{-2}	kBq U235 eq.
Land Use	1.01×10^{3}	1.77×10^{3}	2.52×10^{3}	1.01×10^{2}	1.70×10^{2}	1.51×10^{1}	8.32×10^{1}	1.49×10^{1}	8.31×10^{1}	Pt

Table 6 (continued)

Impact category	P1a (AgNP-HH-PLA_a)	P1b (AgNP-HH-PLA_b)	P1c (AgNP-HH-PLA_c)	S2-P2 (PLA+dis-inf. Wipes)	S3-P2 (PLA+ETO steril.)	S2-P3 (PP+disinf. Wipes)	S3-P3 (PP+ETO steril.)	S2-P4 (HDPE+dis-inf. Wipes)	S3-p4 (HDPE+ETO steril.)	Unit/FU
Ozone depletion	9.71×10^{-9}	1.38×10^{-8}	1.80×10^{-8}	4.47×10^{-10}	4.77×10^{-10}	2.36×10^{-10}	2.66×10^{-10}	2.35×10^{-10}	2.65×10^{-10}	kg CFC-11 eq.
Particulate matter	3.66×10^{-5}	6.74×10^{-5}	9.81×10^{-5}	1.24×10^{-6}	3.09×10^{-6}	1.20×10^{-6}	3.05×10^{-6}	1.21×10^{-6}	3.06×10^{-6}	Disease incidences
Photochemical ozone formation, human health	2.24×10^{0}	4.11×10^{0}	5.98×10^{0}	8.02×10^{-2}	1.90×10^{-1}	7.68×10^{-2}	1.87×10^{-1}	7.71×10^{-2}	1.87×10^{-1}	kg NMVOC eq.
Resource use, fossils	8.13×10^{3}	1.49×10^{4}	2.17×10^{4}	4.81×10^{2}	6.83×10^{2}	5.16×10^{2}	7.19×10^{2}	5.15×10^{2}	7.17×10^{2}	MJ
Resource use, mineral and metals	3.72×10^{-5}	1.63×10^{-4}	4.14×10^{-4}	8.62×10^{-6}	5.72×10^{-6}	8.20×10^{-6}	5.30×10^{-6}	8.20×10^{-6}	5.30×10^{-6}	kg Sb eq.
Water use	2.91×10^{2}	5.34×10^{2}	7.78×10^{2}	3.90×10^{0}	2.50×10^{1}	2.82×10^{0}	2.39×10^{1}	2.81×10^{0}	2.39×10^{1}	m^3 world equiv.

of using wipes has not been considered, since the reusable wipes can last longer than the ointment or spray considered in the intended use phase. Therefore, the end-of-life of the wipes can be excluded.

To address the potential use of co-products or material in output flows a more detailed plan on how the novel granulate shall be used and therefore manufactured someday is necessary. After specifying these points, a reassessment of the end-of-life option should be conducted.

Assumptions for Modelling

The intended use as medical packaging may be changed to an actual product. The assignment of a definite intended use of medical or pharmaceutical packaging was challenging because it is difficult to find a reasonable comparison material. General packaging made from films, e.g., blister packs for tablets or bags for surgical instruments or catheters would not require antibacterial property, since medical devices usually go through a sterilization process which makes the packed products free from any pathogens and more than one use is not intended. Even for reusable medical devices, re-wrapping and re-sterilization are required, which leads to the same scenario. Therefore, antibacterial material would not have any applicability. Choosing the cap as intended use allowed us to find a scenario where the function of the antibacterial material could be included. But only scenario 2 with the disinfecting wipes is a practicable approach. Scenario 3 should not be further considered with this type of intended use. It could help if the goal were to produce an actual product, e.g., a catheter. Since this medical device may be used for a longer period, the antibacterial activity of the material could prevent infections of the patient, which may occur due to bacteria on the product's surface. If the use is a medical device itself, of course conformity with the current medical device regulations has to be assured but this also applies for the medical packaging. Also surfaces in general could use this antibacterial activity, as they host the microorganisms in short term, and frequent sanitization using antibacterial material for high contact surfaces such as door handles, switches, buttons, or handrails could reduce the infection rate. Comparing this with conventional materials could also lead to a comparison with cleaning personnel using disinfectant several times a day to kill bacteria. If the intended use would be a medical device, like a catheter, antibacterial material could also extend the product's life span. Depending on the type of catheter it may be used for only a few days, for up to 30 days or long-term catheters for up to 3 months. These life spans could maybe be extended, which would lead to less catheter changes for the patient and thereby less waste. Even though the last pandemic let researchers focus on medical applications, the

food sector is likewise important as stated in the introduction of this work. And since the originally published paper on the approach of this new material also intended a use in this sector, it should not be dismissed lightly. The current definition of the intended use does not seem to be the best solution and other applications should be determined.

The incineration option for end-of-life has been chosen since the use in the medical sector leads to the assumption that the packaging will be discharged together with other medical waste which is hazardous waste and therefore usually incinerated. Depending on the user, i.e., not medical personnel and patients in hospitals but use at home would lead to disposal in the household trash. This scenario would lead to an end-of-life in landfill or recycling, which would influence the outcome of the assessment.

Further Development

One of the most discussed data points in this work is the energy demand. A scenario where renewable energy is used should lead to less environmental impact of the novel biocomposite and especially the AgNP synthesis but since it would only be fair to use renewable energy for all systems, the compared systems should also become more environmentally friendly. Therefore, this adjustment should lead to a better LCA result in general but would not influence the comparison of the materials. The optimization of the energy demand and source is the most pressing adjustment. The influence of the energy demand has been made clear in the analysis of the results. Therefore, the use of renewable energy sources and a more energy efficient production process should be assessed, especially because a direct proportion of the energy reduction and the decrease of the environmental impact has been observed.

Using bio-based fuel would have similar influence as using renewable energy (see above), i.e., a general LCA result would change but not the comparison results with the other systems, since the change from fossil-based fuel to bio-based fuel would occur for all systems in the same way. The significant issues, which generate more than 80% of the environmental impacts are the polymer granulates, the transportation and the incineration for comparison materials. Regarding Australia as the geographical scope of this LCA, choosing supplier closer or in Australia or other ways of transportation could reduce the impact. But this depends on the availability of "cleaner" transportation options and the locality of suppliers. The AgNP-HH-PLA is mainly influenced by the energy used for the AgNP-HH-PLA synthesis which has been addressed above.

It has to be kept in mind, that the assessment of the novel material is based on data for a laboratory scale, while the granulates of the comparison materials have already been produced in industrial scale and therefore may have a higher energy efficiency and reduced waste, since producing a bigger batch of the materials would reduce the percentage of lost material, which may be caused by residue staying in the production machine or loss due to the transfer of intermediate products into another machine for the next production step. In general, scaling-up production leads to a reduction of environmental impacts because energy and material can be saved. Up to 90% of environmental emissions may be reduced. [20, 42] As soon as the laboratory scale production of the novel biocomposite is optimized, it should be assessed, how the production in industrial scale may impact the current results. When a scaling-up from laboratory scale to industrial scale has been conducted, the used machinery could be included as well, or an assessment, whether the conventional machine for other plastic granulates is sufficient or whether any other equipment is specially needed.

As described in Pourzahedi & Eckelman, 2015 a) [17], the larger the diameter, the smaller surface-to-volume ratio and therefore, larger particles are needed for the same antimicrobial functionality as smaller particles. Comparing the particle size of the in-house made AgNPs with the sizes of the other AgNPs in SI1, the diameter of 75 ± 21 nm is quite large compared to the diameters of ca. 10 or less nm of the GS, CR-Starch, CR-EG or FSP syntheses. Therefore, an adjustment of the chemical reduction to gain smaller AgNPs could lead to a better antibacterial activity of the material or to less AgNP need to maintain the same antibacterial activity, which reduces the material need.

Summary and Conclusion

The present work was conducted to examine the environmental impacts of the novel biocomposite by applying LCA methodology based on ISO 14040/44. As comparison materials PLA, PP and HDPE have been chosen.

The results of this LCA study give an overview on the potential environmental impact of the assessed materials. Although absolute values for the impacts are provided, these results should not directly be compared to other LCA studies, unless the same methodology for LCA (e.g. system boundaries, functional unit) has been applied.

The fundamental concept was that while the AgNP-HH-PLA can be used without antibacterial additives, the other systems gain their antibacterial property by using either EtO sterilization or disinfecting wipes. The LCIA showed that the energy demand for the novel biocomposite is very high and poses the highest environmental impact. Furthermore, AgNP-HH-PLA cannot yet compete with the compared materials from an environmental perspective. The environmental impact of only the material has been assessed

since the impact from the energy varies with the location and used electricity mix. It showed that the use of the novel antibacterial material has less environmental impact than using conventional material but only when the disinfecting wipes are used to gain the same function, which is the antibacterial activity. The discussion shows that there are various aspects which should be adjusted, like the quality of the inventory data, the inclusion of the waste treatment and the available datasets. Also, additionally approaches may be considered, like renewable energy use, scaling-up from laboratory scale, other end-of-life options, and further research on the AgNP impact. Since the LCA has been conducted in the early stages of the development of the new material, the mentioned considerations can still be assessed and implemented, if suitable. This work showed that there is room for improvement, but still, it is important to develop materials with antimicrobial properties. They can repel pathogens and thus help control infection without the quality of the defense being dependent on the disinfectant, the time interval between cleanings, or the performance of the cleaning personnel. Therefore, further development of such materials should be pursued while their integration into the circular economy should be considered. A comparison of a biocomposite with a bio-based antimicrobial compound, like chitosan could be evaluated in terms of antibacterial performance and environmental impact in future studies as alternative to nanosilver. With the availability of primary data in the future, the environmental performance of the product system can be better quantified and made comparable with the other benchmark materials.

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Declarations

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

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