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Life cycle assessment of advanced grade PLA product with novel end-of-life treatment through depolymerization

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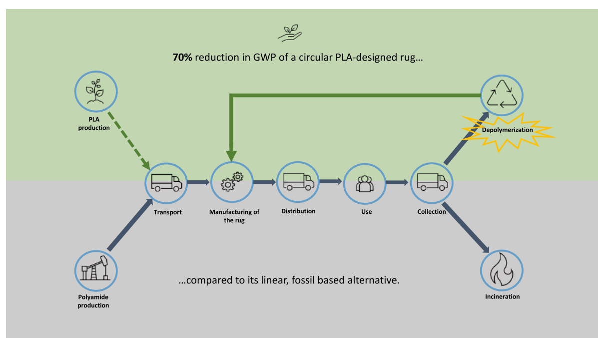
HIGHLIGHTS

- A novel depolymerization method for PLA enables circular and biobased products.
- The LCA shows environmental benefits for a circular rug made fully from PLA.
- Collection rate of PLA products for recycling has high sensitivity and uncertainty.
- A circular design of biobased products is crucial for environmental performance.

GRAPHICAL ABSTRACT



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ABSTRACT

Using biobased plastics has the potential to avoid fossil resource depletion and fossil CO_2 emissions. Polylactic acid (PLA) is a fast-growing bio-based plastic made from fermented sugars. Nowadays, PLA is used to replace fossil-based polymers in healthcare and single-use applications, such as for packaging applications. However, PLA offers a much broader application range with the targeted use of a combination of its stereoisomers; PL(*L*)A and PL(*D*)A. A variety of these advanced grades of PLA can be used for multiple purposes in durable consumer products such as furniture. Recycling complex, mixed material and advanced grades of PLA is currently limited, as mechanical recycling has limitations in recycling mixed PLA grades. Using a depolymerization technology, products of such advanced grades of PLA can be recycled to form high-quality recycled PLA. A cradle-to-grave life cycle assessment study was executed to evaluate the sustainability of high-end durable product (a rug) with mixtures of PLA grade and the novel depolymerization technology. The findings of the study showed a 70 % reduction in CO_2-eq. emissions compared to a conventionally designed rug. However, an increase is indicated in the following environmental impact categories: land use, eutrophication, and environmental toxicity. Sensitivity analyses for collection rates showcased that design for collection and recycling are key to obtaining a more sustainable biobased products. Additionally, scenario analysis supported depolymerization for PLA as recycling technology with low CO_2-eq. emissions. Based on the results of the LCA and additional scenario analysis, the use of PLA is encouraged to be used in more durable and lasting products, such as furniture, from an environmental

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perspective, provided that the products are designed for collection and high-quality recycling to ensure material circularity.

1. Introduction

In the last years, annual plastic production increased considerably from 1.5 million tonnes in 1950 to 391 million tonnes in 2022 (Plastics Europe, 2022). Such intense use of fossil-based resources for plastics production contributes to multiple environmental impacts, including global warming impact through greenhouse gas (GHG) emissions. In 2019, the global carbon footprint of plastics along the entire value chain equaled 1.8 billion tons of CO_2 equivalent annually (OECD, 2022). To achieve carbon neutrality by 2050, significant GHG emission reductions are required across all sectors. This decarbonization urgency has resulted in explorations of the plastics industry to bio-based options for plastics production (Negri and Ligthart, 2021). Biobased plastics are materials that are partly or entirely derived from renewable, bio-based resources (Ferreira-Filipe et al., 2021). Therefore, bioplastics can decrease the dependency on fossil resources and potentially reduce GHG emissions and other impacts in the production stage, such as plastic pollution (MacLeod et al., 2021). Furthermore, using biobased sources for plastics can result in additional GHG emissions savings by storing biogenic carbon (Benavides et al., 2020; Rezvani Ghomi et al., 2021).

At the moment, the biobased plastic with the highest market share is polylactic acid (PLA) (European Bioplastics, 2020). PLA is produced through the fermentation of sugars to form lactic acid. Next, lactic acid is treated to derive lactide which is submitted to a ring-opening polymerization acid lactic acid has two stereoisomers, L-lactic acid and D-lactic acid, where the L-lactic acid is more common in nature and most widely used in PLA production (Groot and Borén, 2010). Due to its position as a biobased and compostable polymer, poly (L) lactic acid (PLLA) is used in healthcare, 3D printing, textiles and mostly, as an alternative to fossil-based polymers in single-use plastic packaging. (Auras et al., 2004). Furthermore, stereo complexity of lactic acid provides enhanced properties and consequently enhanced performance of PLA, such as longer and better durability, strength and different temperature resistance (Groot and Borén, 2010). This offers a broad application range such as in furniture and composites (Farah et al., 2016; Groot and Borén, 2010; Tsuji, 2005). Major producers of PLA such as TotalEnergies Corbion PLA (TotalEnergies Corbion, 2022), NatureWorks (NatureWorks, 2022) and Futerro, together with Galactic (Galactic Manufacturer of Lactic Acid, 2022) are now starting to produce advanced PLA grades to be used in complex, high-end products, such as furniture, leisure products and durable textiles.

Life cycle assessment (LCA) can be used to quantify the environmental impacts associated with the production of materials along the value chain. The majority of LCA studies focus on PLA material used in single-use packaging applications (Bishop et al., 2021; Gironi and Piemonte, 2011; Moretti et al., 2021). For instance, Gironi and Piemonte (2011) compared the environmental impacts of PLA bottles for drinking water with Polyethylene terephthalate (PET) bottles for the same use. They emphasized that PLA bottles indeed have an advantage due to the usage of renewable resources. However, they have a higher impact in respect to human health and ecosystem quality, due to the use of pesticides, fertilizer as well as land and water consumption in production. A different study resulted in similar conclusions when studying the environmental impacts of PLA and PET drinking cups (Moretti et al., 2021). In their study, PLA cups showcase worse environmental performance to their fossil alternatives for many impacts other than climate change and fossil fuel depletion.

Despite the observed GHG emission reductions through the use of bioplastics in the production phase, there are still concerns related to their end-of-life treatment (Van Roijen and Miller, 2022). Typical global end-of-life practices used for bioplastics include landfilling, incineration and (industrial) composting (Bishop et al., 2021; Rezvani Ghomi et al., 2021; Rossi et al., 2015). These end-of-life options fail to provide improvement in environmental impact when compared to using fossil plastics. For example, composting leads to avoidance of peat, which has low environmental benefits (Farrell and Jones, 2010). Incineration of biobased plastics will results in direct CO_2 emissions (Moretti et al., 2021; Rossi et al., 2015). Improvements in different end-of-life treatments can increase the overall environmental performance of bioplastics. Recyclate can be used to avoid the use of virgin plastic material (Maga et al., 2019). Hence, recycling materials contributes to the circularity of biobased plastics and reduction in resource use.

Additionally, recycling options for complex, high-end products are limited. The quality of recyclate through mechanical recycling is reduced due to the presence of additives, color, and multiple compositions or forms within one product (such as fibres, film, and composites). As the presence of these aspects increases with higher complexity of products, the quality of mechanical recycling output is reduced. Similar problems will occur for recycling complex, high-end PLA products. During the mechanical recycling of PLA, hydrolysis can occur, resulting in a significant reduction of the length of the molecular chain. The shorter molecular chains lead to a drop in mechanical properties (Budin et al., 2019). Due to quality reduction with mechanical recycling, recyclers need to mix virgin PLA into recycled PLA to obtain a sellable product (Wolberg, 2021). As described earlier, recycling options such as composting or digestion are not preferable as the material is lost or downcycled, which leads to a poorer environmental performance (Maga et al., 2019; Rossi et al., 2015). Fortunately, the structure of PLA, being in principle of the polycondensation family, allows its monomers to be recycled back into lactide, dilactide or lactic acid (Maga et al., 2019; Piemonte et al., 2013). To close the recycling gap of complex, high-end PLA products, Arapaha developed a new proprietary chemical recycling technology with the aim to recycle complex, high-end PLA products using advanced PLA structures through depolymerization. Through this recycling technology, complex, high-end products can be developed, using PLA as a biobased material, in a potentially fully circular approach. However, to ensure environmental impact reductions of the end-of-life treatment, these still needs to be assessed and compared with other state-of-the-art technologies for PLA. Additionally, there is limited information on the environmental performance of complex, high end PLA products, and a comparison with the conventional alternative product is required as well.

Therefore, the objective of this study is to conduct a comparative cradle-to-grave LCA on a PLA product containing advanced grade PLA on multiple environmental impact categories, but with focus on the global warming potential (kg CO_2-eq). This product will be recycled through a novel depolymerization recycling method developed by Arapaha (Kunst et al., 2023). To compare the environmental impacts of the production, use, collection and recycling of advanced grade PLA products, also the conventional alternative product is assessed which is largely made from fossil-based plastics. Two sensitivity analysis will be performed on the collection rate and to the effect of carbon sequestration. Furthermore, additional scenarios are developed to increase the robustness of the study. The first scenario will include the environmental impact comparison of the novel depolymerization recycling method to other end-of-life options for PLA. The second scenario includes the environmental impact of the PLA rug over multiple use cycles. The global warming potential is assessed for the scenarios and carbon sequestration, while all impacts for collection rate sensitivity are assessed.

2. Methodology

This study was conducted based on the LCA methodology guidelines which are included in the International Organization for Standardization (ISO) 14040 and ISO 14044 (ISO, 2006). ISO 14040 provides the 'principles and framework' of the standard, while ISO 14044 provides an outline of the 'requirements and guidelines' (ISO, 2006). The LCA method was performed according to four main steps: goal and scope definition, inventory analysis, impact assessment, and interpretation.

2.1. Cradle-to-grave analysis

2.1.1. Goal and scope

The main goal and aim of this study is to evaluate the environmental impacts of a high-end rug specifically designed for recycling, made primarily from advanced grade PLA. The rug is produced by Arapaha B. V. (2022), to be used to cover floors in hotels, offices, and houses. The product is designed with the intention to be fully circular by having all components PLA based, starting with the face fibre, the primary backing, the hot melt as well as the secondary backing, creating a unique first in the world fully biobased and circular rug. It is distributed among consumers and recollected after use, to be recycled and reused in new products by Arapaha. The PLA rug is compared to a rug with a conventional design, typically made from polyamide 6 face fibre, a polypropylene primary backing and a latex-based glue. The functional unit used for the product comparison is the following: "one piece of rug (2.4 m × 1.7 m; 4.08 m^2) used for insulation of a floor". The conventional designed product will also be distributed among consumers but will be collected by waste disposal instead, as its complex material mix renders it unrecyclable (Deutsche Umwelthilfe E.V, 2017). Due to a lack of recollection infrastructure and without design for recycling, closed-loop recycling of this product is assumed not possible.

2.1.2. System boundaries

The comparative LCA for the rug is a cradle-to-grave study with defined system boundaries (Fig. 1). The system includes seven sections; (i) extraction and production of the material, (ii) transport of the materials to the product manufacturing facility, (iii) product manufacturing, (iv) distribution of the products to the customers, (v) the use of the product over its life span, (vi) recollection for recycling or the waste collection and (vii) end-of-life of the product. The production takes place in various locations, specified per section. The use, collection and end-of-life are assumed to take place in Western Europe, and the production and depolymerization are set in The Netherlands. The temporal scope is set in the year 2022. Recycled content created within the cradle-to-grave assessment are included by allocating recyclate and other potentially avoided materials (heat, electricity) as avoided burdens. System expansion was also used to model PLA production in Morão and de Bie, 2019 (Morão and de Bie, 2019). With this approach, the benefits of recycling and energy recovery through incineration have been considered as avoided impacts.

2.2. Life cycle inventory

The LCA uses primary data and literature data. For the background processes ecoinvent database v3.7.1 and Industry data 2.0 was used.

2.2.1. Production

The rough composition of the circular and conventional design of the rug is described in Table 1. PLA in the rug is largely supplied by TotalEnergies Corbion PLA under the brand Luminy®, which produces PLA from sugar cane in Thailand. Data on the production of PLA in Thailand is made available by Corbion (Morão and de Bie, 2019). As the origin of the material supplies in conventional design product was not well known, the 'market' background datasets of the ecoinvent database have been used, representing production average technologies and geographical origins available on the global market. The origin of the raw material, production locations and ecoinvent profiles of the conventional rug parts are supplied in Tables S2 and S4. For the PLA designed rug, it was assumed that the manufacturing stage takes place in Maastricht, The Netherlands. Data on the energy consumption and efficiency of the manufacturing operations for both the PLA designed rug and the conventional rug were extracted from the literature and presented in Table S3 and Table S6. The PLA designed rug is manufactured using a biobased PLA based hotmelt, which required less heating during the manufacturing of the rug. The conventional design product is

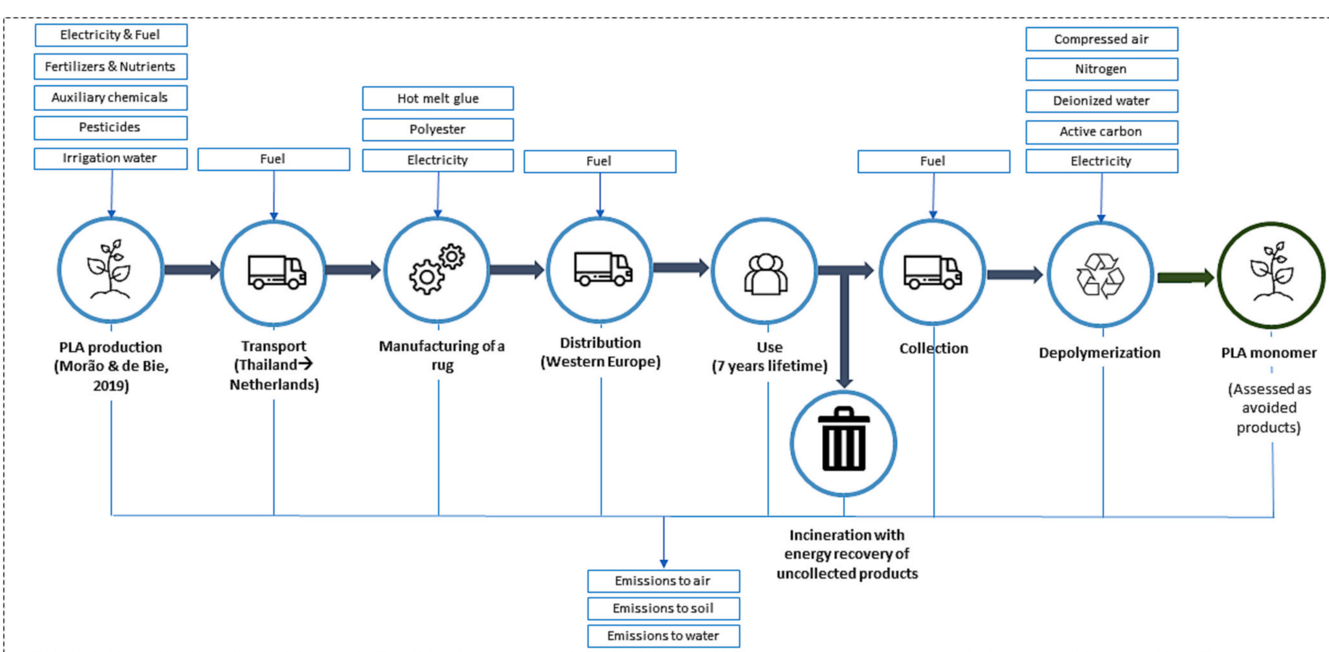

Fig. 1. System boundaries of the cradle-to-gate life cycle of the PLA designed rug. The thick blue arrow represent the cradle-to-grave value chain. The thick green arrow and color depicts the avoided PLA monomer, which can be re-used from the manufacturing stage. The small blue arrows depict inputs and outputs of the cradle-to-grave system.

Table 1
Summarized Inventory data for PLA design rug and conventional design rug.

Product	PLA design					Conventional design	
	Size (m^2)	Mass (kg)	PL(*L*)A (%)	PL(*D*)A (%)	Other components (%)	Mass (kg)	Main material
Rug/carpet	4.08	12.08	91.5	1.6	6.9	12.18	Polyamide

typically manufactured through coating the rug with latex which is then consolidated in a gas fired oven, which is the state-of-the-art production method for rugs.

2.2.2. *Distribution and use*

The distribution area for the PLA designed rug is set in Northwestern Europe with an average distribution distance of 200 km. For the conventional design, similar distribution is used as the PLA designed rug. As the manufacturing locations of the conventional design rug are unknown, this can potentially be an underestimation, especially since rugs and furniture are transported globally. For both product alternatives, any packaging requirements during distribution are left out of scope. Once the products are distributed to the customers, the essential factor to consider in the use phase is the product's lifetime. According to European carpet and rug association (ECRA) the lifespan of rugs is set from 3 (minimal) to 10 (maximal) years (Donkersloot, 2022; ECRA, 2022). The lifetime of both rugs in this study is assumed to be 7 years on average, which is similar to lifetimes estimated in literature for comparable products (Donkersloot, 2022). The impacts that are related to the products lifetime, such as cleaning or potential microplastic formation, are left out of scope.

2.2.3. *Collection*

After use, the PLA designed rugs are recollected for recycling. It was assumed that 50 % of the rugs will be collected at the customer by lorry for recycling. This estimate is based on data from ECRA (Donkersloot, 2022, ECRA, 2022) for similar products and expert knowledge. This value is chosen as the collection system infrastructure of these products is relatively new and therefore lack efficiency, especially in consumer awareness. However, these values do contain a high level of uncertainty. The collection and recycling location for the PLA designed rugs is assumed to be in Emmen, and similar traveling distances are taken as used in the distribution step. The PLA designed rugs that are not collected are assumed to be incinerated, with energy recovery, see chapter 2 of the Supplementary information on the energy recovery modelling.

2.2.4. *End-of-life*

The end-of-life treatment considered for the PLA designed rug is the new depolymerization technology (Kunst et al., 2023). The technology includes two recycling variations. In the first variation, PLA is recycled to its oligomers which can then be fed into an existing PLA production plant, while in the second variation, PLA is recycled to lactide, which again van be fed into an existing PLA production plant or used in any other application of lactide. The mass and energy balance of the depolymerization technology and the corresponding ecoinvent inventories are reported in Table 2. Energy requirements are calculated based on temperatures, heat capacity, and efficiency. The oligomer process is assumed to be fully electrified, so all energy consumption is in the form of electricity. The lactide process uses both electricity and steam as heat sources. The ring depolymerization and distillation steps are assumed to use steam. For both the oligomer and the lactide route, a relatively high efficiency of 95 % of recovery of PLA material was assumed, resulting in a 5 % PLA/LA loss estimate during the process. It was assumed these losses take place during the main process, which includes depolymerization, purification, and chiral separation. All additional materials, such as additives and contamination, are lost as well. The waste materials during the depolymerization and non-collected rugs, are treated

Table 2
Mass and energy balance of the depolymerization of one tonne of PLA input.

Name	Process name (ecoinvent)	Amount	Unit
Inputs			
PLA input	Inventory data from Morão and de Bie, 2019	1	t
Water (deionised)	Water, deionised {Europe without Switzerland}| market for water, deionised | Cut-off, U	100	kg
(new) active carbon	Activated carbon, granular {RER}| activated carbon production, granular from hard coal | Cut-off, U	5.1	kg
Reactivated carbon	Activated carbon, granular {RER}| treatment of spent activated carbon, granular from hard coal, reactivation | Cut-off, U	46.2	kg
Nitrogen	Nitrogen, liquid {RER}| market for | Cut-off, U	60	kg
Compressed air	Compressed air, 1000 kPa gauge {RER}| compressed air production, 1000 kPa gauge, <30 kW, average generation | Cut-off, U	42.4	m3
Outputs			
Lost lactide	PLA to municipal incineration	50	kg
Wastewater	Wastewater, average {Europe without Switzerland}| market for wastewater, average | Cut-off, U	0.003	T
Output	Polylactic acid (Groot and Borén, 2010)	950	kg
Energy requirements			
Electricity for shredding	Electricity, medium voltage {NL}| market for | Cut-off, U	39	kWh
Oligomer: electricity	Electricity, medium voltage {NL}| market for | Cut-off, U	537	kWh
Lactide: electricity	Electricity, medium voltage {NL}| market for | Cut-off, U	309	kWh
Lactide: steam	Heat, from steam, in chemical industry {RER}| market for heat, from steam, in chemical industry | Cut-off, U	866	MJ

with incineration with energy recovery.

The conventional designed rug is assumed to be collected through the main waste collection and will be incinerated with energy recovery. The incineration process used is representative of an installation in the Netherlands which is used as a proxy for Northwestern Europe. For incineration, an average transport distance of 100 km is taken.

2.3. *Cradle-to-grave sensitivity analysis*

For the cradle to grave assessments, two sensitivity analyses are included. The first sensitivity analysis will address the effects on the LCA of CO_2 extraction from the air, which is sequestered in products when assessing a biobased material (Vogtländer et al., 2014). In the cradle-to-grave assessment, the CO_2 sequestration is not included, whereas in the sensitivity it is included in the production phase, which additionally affects the CO_2 emissions budgets in end-of-life. In the second sensitivity analysis, the impacts of different collection rates during the transition of use to end-of-life are assessed.

2.3.1. *CO_2 sequestration*

In the cradle-to-gate LCAs that are published for PLA production, the

LCA accounts for carbon sequestration in the PLA itself. This means that CO_2 is captured and stored during the growth of the crops. However, the sequestration of CO_2 in soils is excluded. Hence, the global warming potential effects of land transformation are already included in the ecoinvent processes in relevant cases. However, the CO_2 sequestration results in a lower global warming potential compared to fossil-based polymers, as take-up results in a negative carbon footprint at the start. The amount of sequestered carbon is set at 1.833 kg CO_2 eq./kg PLA based on the molecular weight of sugar (Groot and Borén, 2010; Morão and de Bie, 2019; NatureWorks, 2022). At the end-of-life, when the carbon that is incinerated from the PLA is either classified as biogenic (when uptake is included) or fossil (when uptake is not included). This is done as the global warming potential in the ReCiPe2016 methodology does not assess biogenic CO_2 emissions. Therefore, if no CO_2 emissions were allocated with sequestration, the carbon balance would be incorrect as CO_2 would conveniently 'disappear' from the system. Through this sensitivity analysis, the effect of taking sequestration into account is assessed. In literature, it is discussed whether short-lived products are allowed to sequester carbon, as their lifetime, or the carbons' time 'in stock', is limited (Vogtländer et al., 2014). On the other hand, for advanced PLA grades, the lifetime of the product and hence 'stock-time' of carbon is longer and can be compared to products such as wood. Still, the removal of carbon in stock in the biosphere is transferred to stock in the technosphere; hence this carbon is unavailable to nature. In addition, Arapaha has a strict closed loop regime (Arapaha B.V., 2022) in which the return rates of the products are maximized. Therefore, the effect of inclusion of sequestration of CO_2 used as an alternative method to account for carbon sequestration in LCA, and with that, there are many more methods to take carbon storage into account (Vogtländer et al., 2014).

2.3.2. Collection rates

When recycling the PLA design rug, it should be noted that the materials need to be separately collected. Since the product that is assessed is not yet on the market, no available data on collection rates was available. By adding a QR code physically attached to the rug this process is made as easy as possible for consumers, to maximise the return rates. Therefore, internal market studies by Arapaha been used to obtain an average of 50 % collection and 50 % non-collected rugs. The sensitivity analysis was performed to measure the sensitivity of the collection rate for the rug within the multiple use cycle scenario. This parameter is an estimate and presents high uncertainty. The collection rate might vary in the range of low response values (30 %) to highest response values (80 %), around the average of 50 %.

2.4. Scenario analysis — comparison of different end-of-life pathways for PLA

For robustness of the comparative cradle-to-grave LCA, the environmental impact of chemical recycling practices for PLA is evaluated. A comparison of the environmental impacts of the new recycling method with other end-of-life options for PLA was executed. These include incineration with energy recovery, composting, digestion and mechanical recycling. Since the advanced grade PLA rug is a new product and has not been tested in other recycling technology, the decision was made to broaden the functional unit to all PLA products. Therefore, the functional unit will be as follows: "Recycling of 1 tonne of PLA waste". The detailed descriptions of each end-of-life pathway is described in chapter 2 of Supplementary information file.

2.5. Scenario analysis — multiple use cycle analysis

As a final scenario, environmental impact over multiple use cycles is assessed. This was done by comparing the PLA designed rug with its conventional alternative, where floor covering over a longer timeframe is assessed. In order to cover the larger timeframe, the functional unit will be as follows: "Providing floor cover for insulation of 4.08 m^2 until 2050".

Recycled materials from a product can be re-used in the following product life cycle. Recycled PLA is re-used in the rug, thereby it is assumed less virgin PLA is required when producing the recycled product. When no quality decrease of the material takes place, this can be done for multiple products over time, contributing to the circularity of the product. In the multiple use cycles, the temporal scope is kept into account depending on the product's lifespan. It is assumed that the new potential production of the product is taking place the same year as the recycling process of the old product. The temporal scope of the rug is set until 2049, covering 28 years. Until then the rug with the average lifespan of 7 years needs to be replaced four times. This is an adequate time period to showcase the impact of the environmental emissions of the multicycle usage throughout the cycles. In addition, the model considers also that the electricity grid mix will become more sustainable with increasing renewable sources. The KEV 2022 predictions for the electricity sources in 2025, 2030 and 2040 are used to calculate the impact of electricity use in this multiple use cycle analysis (PBL, 2020). The sources of electricity from the grid are presented in the tables in the KEV report. This adaptation overtime is only considered for energy use in recycling; production takes place in different countries with different goals and electricity sources. This is kept out of scope. It also affects the avoided impacts associated with energy recovery. Moreover, the collection rate of the PLA design rugs is kept at 50 % for the first three cycles. In 2042, a system that can recollect 80 % of the rugs in place is assumed. CO_2 sequestration is included, due to the assumed longevity of the carbon used in various cycles.

2.6. Life cycle impact assessment

LCA modelling was done using Simapro software version 9.3.03, provided by Pre-Consultancy in the Netherlands (PRé, 2023). ReCiPe 2016 Midpoint (H) impact assessment method was chosen to calculate the environmental impacts in all 16 impact categories. The focus is on the Global warming potential; (GWP; kg CO_2 eq.). Additionally, also the impacts of stratospheric ozone depletion; (SOD; kg CFC11 eq.), Ionizing radiation (IR; kBq Co-60 eq.); Human health (HH; kg NOx eq.) Fine particulate matter formation (FPM; kg PM2.5 eq.), Ozone formation, Terrestrial ecosystems; (TE; kg NOx eq.) Terrestrial acidification (TA; kg SO_2 eq.); Freshwater eutrophication (FE; kg P eq.), Marine eutrophication (ME; kg N eq.), Terrestrial ecotoxicity (TE; kg 1.4-DCB) Freshwater ecotoxicity (FWE; kg 1.4-DCB) Marine ecotoxicity (ME; kg 1.4-DCB) Human carcinogenic toxicity (HT- cancer; kg 1.4-DCB) Human non-carcinogenic toxicity (HT- non-cancer; kg 1.4-DCB) Land use (LU; m^2a crop eq.), Mineral resource scarcity (MRS; kg Cu eq.), Fossil resource scarcity (FRS; kg oil eq.), Water consumption (WC; m3) are included in the cradle-to-grave analysis.

3. Results

In this section, the LCA results are presented and discussed. The main case investigated is (i) cradle-to-gate analysis of LCA of a rug, compared to the conventionally designed rug followed by (ii) sensitivity analysis of CO_2 sequestration in the life cycle (iii) sensitivity analysis on collection rates, (iv) scenario on comparison of different end-of-life routes for PLA, and (v) scenario on the multiple use cycle analysis of a rug.

3.1. Environmental impact cradle-to-grave analysis

The global warming impacts of the cradle-to-grave analysis of the two rug designs per step are shown in Fig. 2. The global warming impacts of the PLA designed rug is 27.7 kg CO_2 eq., which is almost 70 % lower compared to the conventional designed rug. The conventional rug emits 93 kg CO_2 eq. over its lifetime. The biggest contributors for both designs are production (raw materials) and the end-of-life incineration

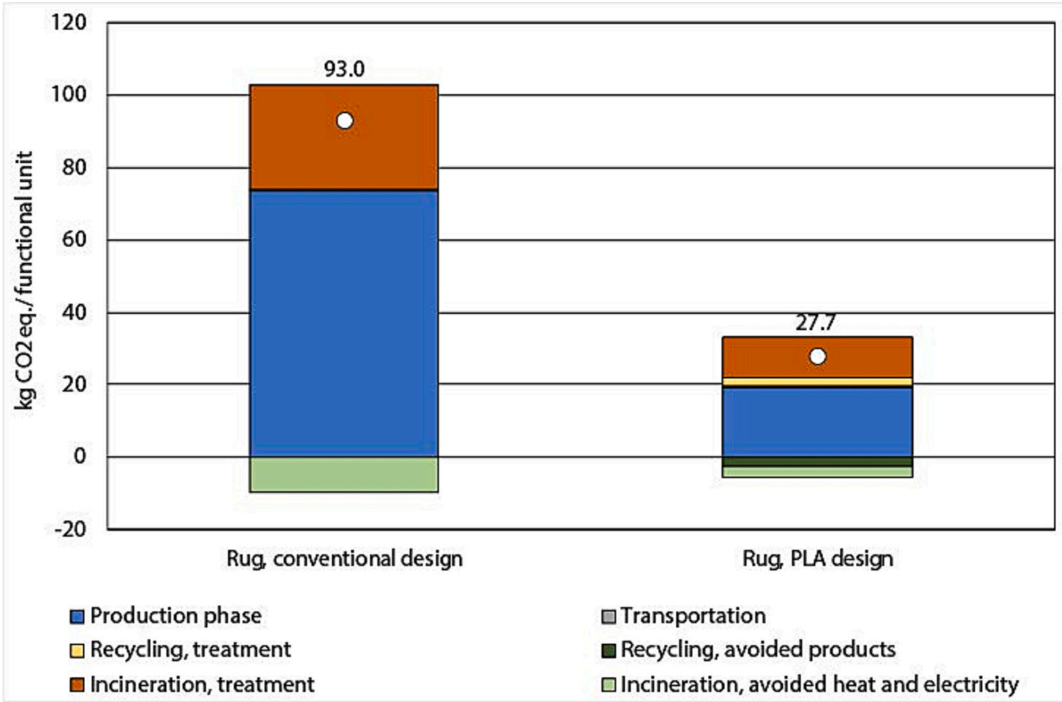

Fig. 2. Cradle-to-grave LCA results on global warming potential for the PLA designed rug and the conventional rug, both of 4.8 m^2.

of the materials, where polyamide contributes most of the impact for the conventional rug and PLA production for the PLA designed rug. Transport has a negligible contribution to the total carbon footprint. In Fig. 3, all ReCiPe impact categories are presented on a percentage scale, where the impact of the conventional design is scaled to 100 %. Two categories are displayed separately in Fig. 3b due to the large difference in scale with the other factors in Fig. 3a. For 9 out of 18 categories, the PLA design rug has a lower impact than the conventional design. The highest impact reduction is seen for global warming, stratospheric ozone depletion, and ionizing radiation (respectively 70 %, 89 %, and 88 %). These impacts can be fully allocated to the fossil raw material use (polyamide) in the classic rug. On the other hand, PLA designed rugs have a significantly higher impact on two impact categories, especially for land use and marine eutrophication (2116 %). Water consumption and ecotoxicity also have a higher impact for the PLA designed rug. A negative water consumption is registered during polyamide production, the main component of the conventional rug. Unfortunately, due to the aggregated nature of the industry 2.0 database, the source of this water savings is not found. It is assumed that the polycondensation step required for polyamide production results in a water 'surplus' with the system boundaries of the database. The impacts that are higher for the PLA designed rug, are a result of the growth of agricultural products required for PLA, which comes with the use of land, pesticides, and fertilizers. The main component of the conventional rug is nylon, which is a fossil-based plastic that does not require agricultural activities.

3.2. Sensitivity analysis of collection rates

The sensitivity analysis on the collection rates of the PLA designed rug highlights the significant effect of this phase (Fig. 4). In the cradle-to-grave LCA, an average collection rate of 50 % is used. With a low assumed collection rate (30 %), there is an increase of 12 % in the total GWP emissions, a result of lower recycling rate and hence material recovery rates, and increased incineration emissions. The higher collection rates (80 %) result in 18 % decrease in global warming impact. For other impacts results the improvements vary strongly. Ionizing radiation has little improvement with a higher collection. On the other hand, ecotoxicity and eutrophication impact categories result in the highest decrease of impacts (up to 50 % for freshwater eutrophication). Human carcinogenic toxicity and mineral scarcity are two impacts that increase in impact. Likely this is a result of increased transport distances by lorry and resource use during recycling. This analysis highlights the prerequisite of a proper collection system or return scheme, which should result in high response rates to decrease total environmental impacts.

3.3. Sensitivity analysis of CO_2 sequestration

To assess the effect on the LCA of including CO_2 sequestration during crop growth, the full cradle-to-grave for the PLA designed rug is executed without sequestration. No CO_2 sequestration is included in the production, while at end of life, emissions are included as biogenic CO_2 emissions. These do not add to the total global warming impact budget when using the IPCC methodology which is used in ReCiPe 2016. The global warming impacts of the PLA designed rug are very similar when regarding the full cradle-to-grave life cycle, only emissions are allocated to different phases (Fig. S1). Therefore, it is concluded that using CO_2 sequestration in the cradle-to-grave LCA does not significantly impact the results. Nevertheless, it is important to report whether sequestration is used as the impacts are allocated to different steps in the life cycle, making it important when LCAs are limited to production only (cradle-to-gate) or end-of-life only (gate-to-grave).

3.4. Scenario analysis; end-of-life alternatives for PLA

The global warming impacts of recycling of 1 tonne of PLA is compared to the state-of-the-art PLA recycling options is summarized in Fig. 5. Mechanical recycling performs the best with −1.75 tonnes CO_2-eq/tonne recycled PLA, where 10 % reduction in quality is considered due to this process. The GWP for the lactide and oligomer produced with the new recycling technology are similar to mechanical recycling, with both −1.67 tonnes CO_2-eq/tonne recycled PLA, respectively. Above three technologies score significantly better than other end-of-life treatments; digestion, composting and incineration. Composting performs the worst with a 125 kg CO_2-eq per tonne PLA. Digestion and

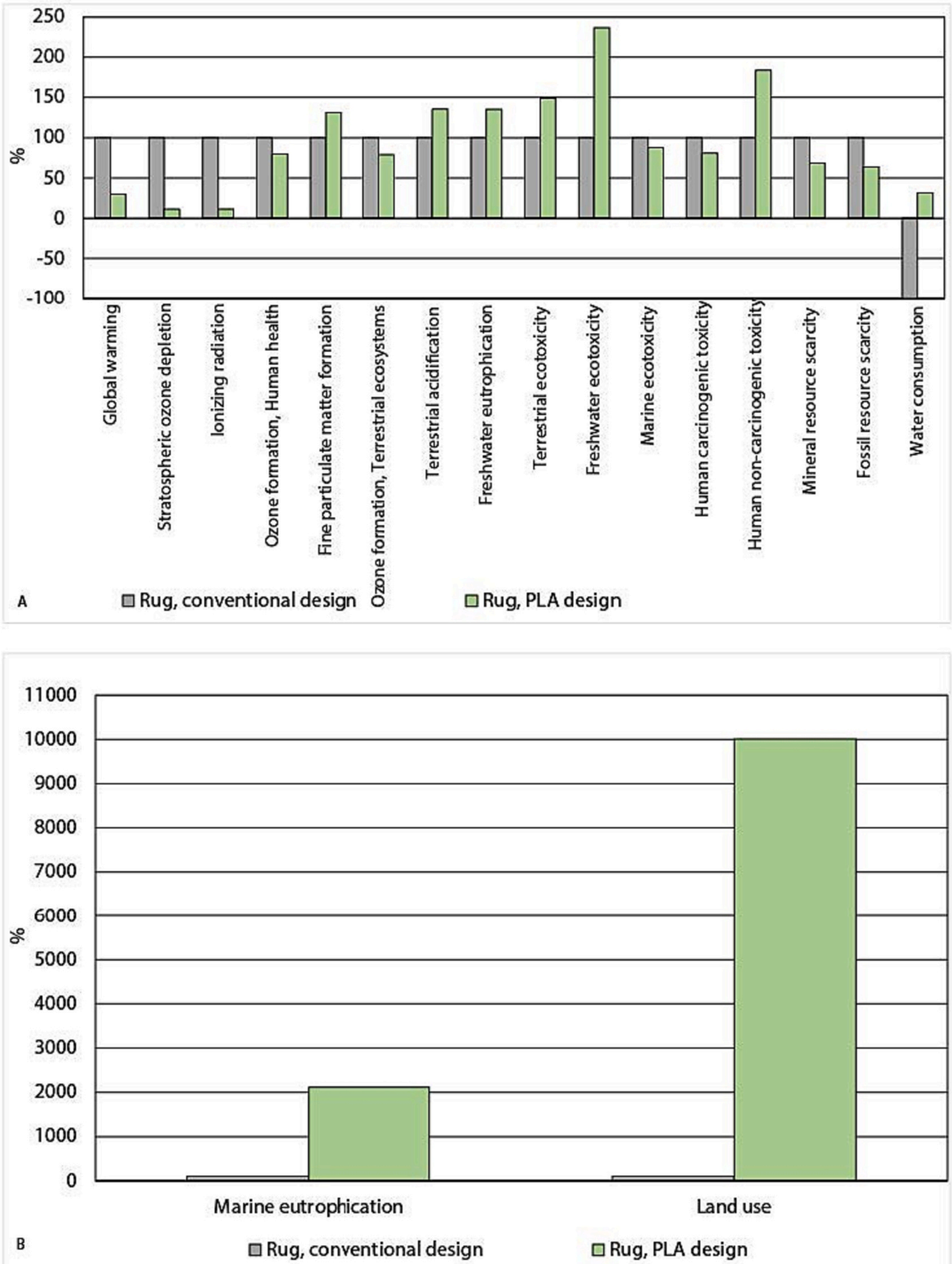

Fig. 3. 100 % graph for all ReCiPe impact categories from the cradle-to-grave analysis of the two rugs of 4.8 m^2, where the conventional design is set to 100 % of impacts and PLA designed rug scaled towards the conventional rug. A) All impact categories, without marine eutrophication and land use. B) For marine eutrophication and land use (adapted scale).

incineration with energy recovery showcase −509 and −451 kg CO_2-eq., respectively. Due to larger shares of renewables in the electricity mix in the future, there will be fewer and fewer savings with energy recovery in the incineration plant. Hence the carbon footprint of the incineration in 2030 and 2040 is more significant than right now. In the PLA designed rug, mechanical recycling is a non-preferred route, as it is likely that a low quality recyclate is obtained due to the complexity of the product with additives and presence of a mix of PLA grades. Therefore, we conclude that the new recycling technology performs very well environmentally and fills a gap in recycling high end products with advanced PLA.

3.5. Scenario analysis; multiple use cycle analysis

After multiple cycles of recycling and use, the PLA designed rug is observed to reduce global warming impacts (Fig. 6). From cycle 1 to cycle 2, the second PLA designed rug has 12 % lower impacts. Due to collection rates set on 50 %, impact reduction is limited. Additional

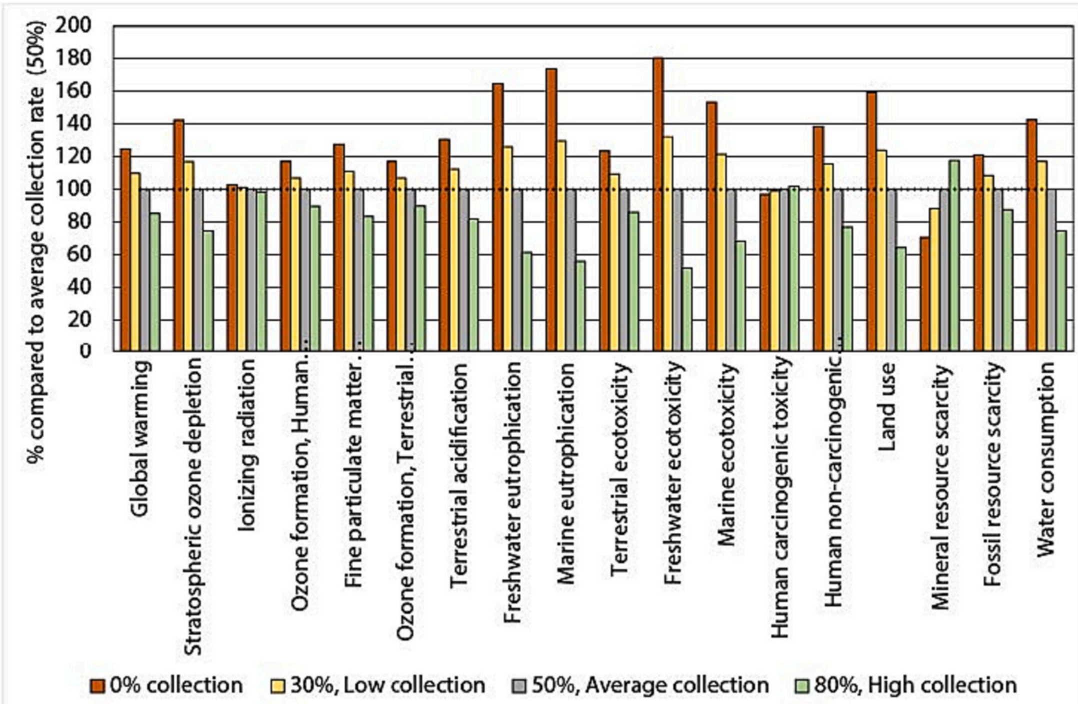

Fig. 4. Results of the sensitivity analysis for cradle-to-grave PLA-designed rug of 4.8 m^2 where collection rate of materials is adapted. The average collection rate (50 %) is scaled to 100 % of impacts and highlighted with the grey line.

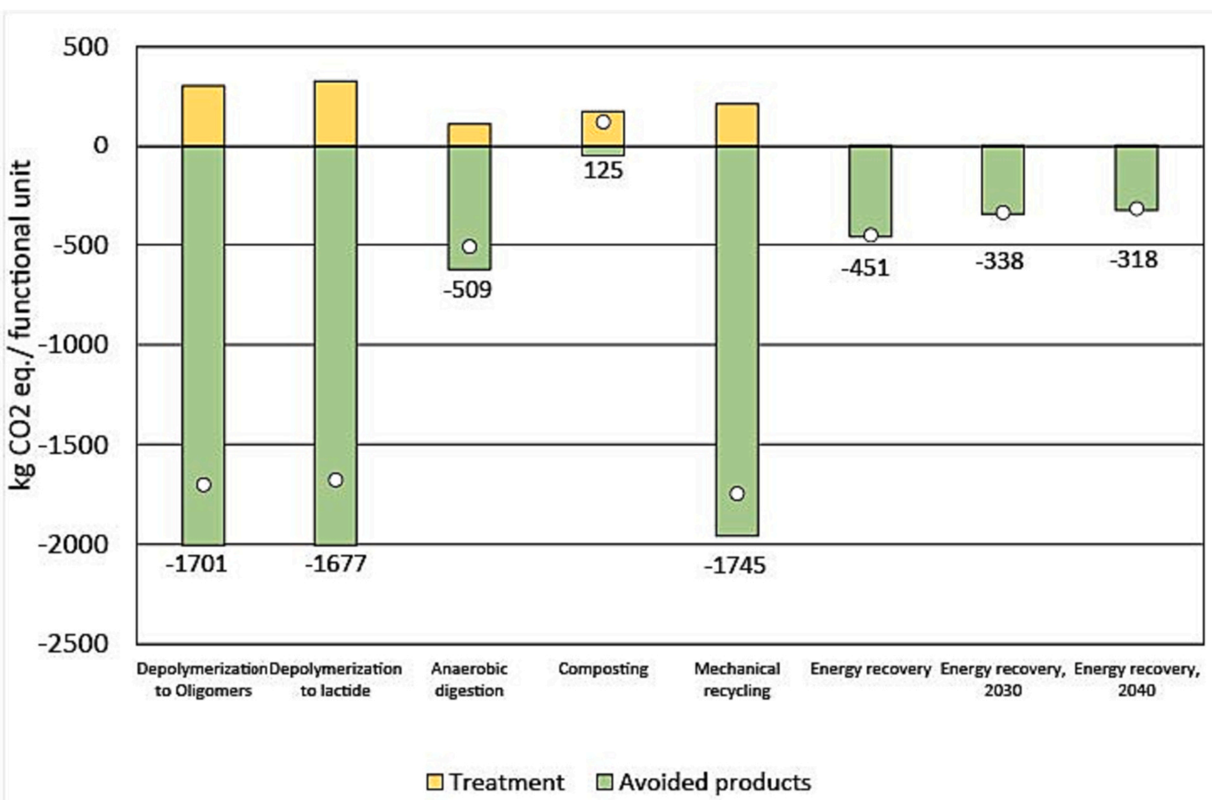

Fig. 5. Global warming impact results for the analysis of the end of life for 1 tonne of waste PLA.

materials and processes for production and manufacturing were not avoided and impacts are still significant. Impacts of renewable electricity mix results in limited savings (cycle 2–cycle 3). A higher collection rate, as assumed in cycle 4, decreases the GWP significantly (40 % compared to cycle 1). Overall, a 72 % CO_2 reduction is achieved of PLA designed rug compared to a conventional design over four cycles.

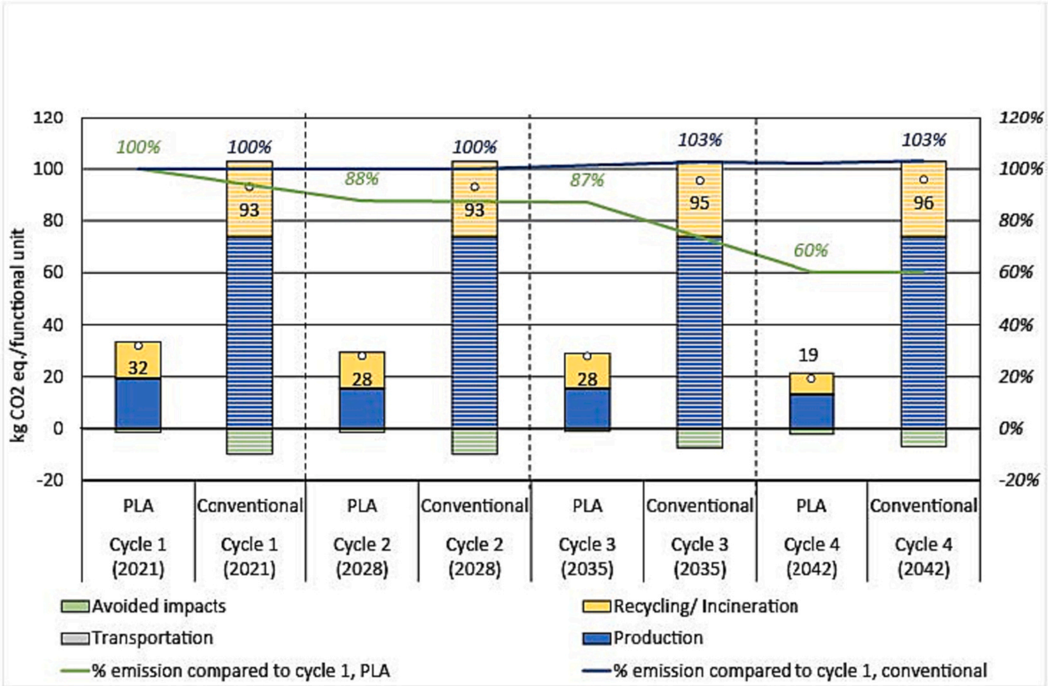

Fig. 6. Global warming impact results from the multiple use cycle analysis for providing floor cover for insulation of 4.081 m2 until 2050′. This covers 4 use cycles from 2022 to 2050.

The impact of 1 cycle for conventional design is almost as high as the total impact for the PLA designed rug over four cycles (107 vs 93 kg CO_2-eq.). Hence, also for multiple use cycles, most environmental impact improvements are a result of a proper collection system or return scheme.

4. Discussion

A 70 % reduction in global warming potential with the PLA designed rug can be achieved, compared to the conventional rug. However, an increase is indicated in the following environmental impact categories: land use, eutrophication, and environmental toxicity. As PLA originates from an agricultural production chain, impacts mainly originate from the production phase. The use of fertilizers increases the concentration of phosphor and nitrogen in the environment while the use of pesticides and herbicides increases the toxicity indicators. Alternative agricultural activities might change the outcome of the LCA when sugarcane or maize is cultivated without the use of these products. Additionally, organic agriculture can also decrease GHG emissions (Meisterling et al., 2008). However, it is difficult to compare the environmental impact of 'standard' cultivation to alternative or organic agricultural practices, as quantification of the latter is still under development in LCA (Ferreira-Filipe et al., 2021) The results of the current study are similar to other LCA studies, where PLA products perform better in the climate change category, whereas in other impact categories the impact is higher (Bishop et al., 2021; Gironi and Piemonte, 2011; Morett et al., 2021). In the cradle-to-grave study, a new depolymerization technology is incorporated for a PLA rug, which results in decreased environmental impact significantly compared to the conventional design with state-of-the art end of life treatments. However, the scenario study to environmental impacts of the end-of-life for PLA highlight the importance of the end-of-life treatment choice. In other LCA studies, PLA products are usually composted or incinerated. Indeed, without proper recycling options available for PLA, this is the most likely end-of-life scenario. For example, Tamburini et al. compared biobased PLA water bottles with PET (fossil-based) water bottle. In this study, PLA water bottles have a higher GWP impact than the PET alternative (Tamburini et al., 2021). Here, the main difference between GWP impact of the products is related to the end-of-life scenarios. In the study, the PET water bottles are recycled, whereas the PLA bottles are either incinerated or composted. For all types of products, recycling and circularity are key in reducing the environmental impact of the product. Therefore, it can be concluded that substituting biobased plastics for fossil plastics alone does not improve the environmental performance of a product. Without proper end-of-life where recyclate is obtained, biobased products will likely have similar, if not higher, impacts on the environment, as also shown in Fig. 5 of the results. The current study highlights that the new chemical recycling method can be a key technology for a circular and biobased economy. Chemical recycling enables recycling of complex, high-end PLA products. Mechanical recycling will not be sufficient to obtain high quality recyclate for this type of products, due to a mixture of materials and PLA grades within the product. Design for recycling remains a key factor for proper, low-impact recycling also for depolymerizations. Hence, non-removable additives or combining PLA with other materials should be limited in the design of the product as it affects the performance of the depolymerization process. Recycling through depolymerization can also become environmentally competitive with mechanical recycling for less complex, high-end products, as the environmental impacts are relatively similar, especially considering quality reduction after several rounds of mechanical recycling (Budin et al., 2019). Additionally, a Techno-Economic Assessment (TEA) can be conducted to compare the depolymerization and mechanical recycling technologies based on economic and technological performance.

The current study can support developing policy frameworks within the European Green Deal (EDG) (European Commission, 2019) and the Circular Economy Action Plan (CEAP) (European Commission, 2022). The framework will be implemented in all sectors including plastics on targeted measures to tackle plastic pollution and other sustainability challenges relating to the use of bioplastics. One of the focus points of the roadmap is developing criteria of the usage of bioplastic solely based on the environmental assessment of the application where the use of bioplastic is beneficial to the environment. This kind of assessment has been made in this study on the case of the rug. Due to the possibilities with advanced PLA, this material can likely function as substitute material in many more products (Tsuji, 2005). These include a substitution of metals and even composites. Depending on the conventional design, environmental benefits or detriments may be very significant for multiple indicators.

Due to the novelty of the current study, several assumptions were required due to data limitations. First, it has been assumed that the rug containing advanced PLA has the same lifespan as the conventional rug, which is based on the previous assumption that advanced grades of PLA have similar material properties as fossil-based plastics. Use of the products must show similar lifetimes and experience and research will be needed in the future to support this assumption. Secondly, the collection rates of the PLA rugs are highly uncertain because it was impossible to estimate the actual collection rate of the product, since large-scale production and distribution are not taking place yet. Therefore, the sensitivity analysis was made which showed that including an effective waste collection system is a crucial factor in environmental performance, especially in the multiple use cycle analysis. With higher collection rates and proper collection system, less virgin material is required to replenish losses which was a main environmental impact. The transport that is required in the collection phase of the advanced grade PLA products is observed to be negligible compared to virgin PLA production in Thailand and transport to Europe in this LCA and few impact categories show increase in impact with higher collection rate. Currently, there is a lack of effective product recollection systems. Policy frameworks for circular economy should encourage investments designing effective collections systems with higher return rates. Further studies on the collection systems are required to improve the accuracy of the collection rates. Lastly, the baseline of the new recycling technology assessed is based on pilot plant data extrapolated to the higher scale. It is recommended that another LCA study is conducted once the technology reaches higher TRL.

5. Conclusion

A cradle-to-grave life cycle assessment study was executed to evaluate the environmental impact of complex, high-end product (a rug) with advanced PLA grades, together with a collection system and a new depolymerization technology for end-of-life. Compared to the conventional, fossil-based product, global warming impacts are reduced with a PLA rug. However, these impact reductions are a result of design, collection and recycling which are in place for the PLA rug. Hence, circularity through design for recycling is key to obtaining sustainability for biobased products. Based on the results of the LCA, the use of PLA polymers is encouraged to be used in more durable and lasting products such as furniture from an environmental perspective, as long as the products are designed for collection and high- quality recycling to ensure material circularity.

CRediT authorship contribution statement

Anna Schwarz: Methodology, Writing - original draft, Modelling; Spela Ferjan: Conceptualization, Writing - original draft, Visualization; Josse Kunst: Resources, Funding acquisition, Writing - review & editing.

Declaration of competing interest

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

Data availability

See supplementary info

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.167020.

References

Arapaha B.V, 2022. WWW document. URL. https://www.arapaha.com/ill-be-back/ (accessed 5.11.22).
Auras, R., Harte, B., Selke, S., 2004. An overview of polylactides as packaging materials. Macromol. Biosci. 4, 835–864. https://doi.org/10.1002/mabi.200400043.
Benavides, P.T., Lee, U., Zarè-Mehrjerdi, O., 2020. Life cycle greenhouse gas emissions and energy use of polylactic acid, bio-derived polyethylene, and fossil-derived polyethylene. J. Clean. Prod. 277, 124010. https://doi.org/10.1016/J.JCLEPRO.2020.124010.
Bishop, G., Styles, D., Lens, P.N.L., 2021. Environmental performance of bioplastic packaging on fresh food produce: a consequential life cycle assessment. J. Clean. Prod. 317, 128377. https://doi.org/10.1016/J.JCLEPRO.2021.128377.
Budin, S., Maideen, N.C., Koay, M.H., Ibrahim, D., Yusoff, H., 2019. A comparison study on mechanical properties of virgin and recycled polylactic acid (PLA). J. Phys. Conf. Ser. 1349 https://doi.org/10.1088/1742-6596/1349/1/012002.
Corbion, 2022. WWW document. URL. https://www.corbion.com/en/Products/Bioplastic-products (accessed 8.2.22).
Deutsche Umwelthilfe E.V, 2017. Swept under the Carpet: The Big Waste Problem of the Carpet Industry in Germany.
Donkersloot, 2022. WWW document. URL. https://www.donkersloot-tapijt.nl/ (accessed 5.2.22).
ECRA, 2022. WWW document. URL. https://www.ecra.eu/ (accessed 5.2.22).
European Bioplastics, 2020. European Bioplastics e.V. [WWW document]. Eur. Bioplastics. URL. https://www.european-bioplastics.org/market/.
European Commission, 2019. A European green deal | European Commission. Eur. Comm. 24.
European Commission, 2022. Circular economy action plan [WWW Document]. Eur. Comm. URL. https://environment.ec.europa.eu/strategy/circular-economy-action-plan_en (accessed 11.2.22).
Farah, S., Anderson, D.G., Langer, R., 2016. Physical and mechanical properties of PLA, and their functions in widespread applications—a comprehensive review. Adv. Drug Deliv. Rev. 107, 367–392. https://doi.org/10.1016/J.ADDR.2016.06.012.
Farrell, M., Jones, D.L., 2010. Food waste composting: its use as a peat replacement. Waste Manag. 30, 1495–1501. https://doi.org/10.1016/J.WASMAN.2010.01.032.
Ferreira-Filipe, D.A., Paço, A., Duarte, A.C., Rocha-Santos, T., Silva, A.L.P., 2021. Are biobased plastics green alternatives?—a critical review. Int. J. Environ. Res. Publ. Health 18, 7729. https://doi.org/10.3390/IJERPH18157729.
Galactic Manufacturer of Lactic Acid, 2022. WWW document. URL. https://www.lactic.com/en (accessed 8.2.22).
Gironi, F., Piemonte, V., 2011. Life cycle assessment of polylactic acid and polyethylene terephthalate bottles for drinking water. Environ. Prog. Sustain. Energy 30, 459–468. https://doi.org/10.1002/EP.10490.
Groot, W.J., Borén, T., 2010. Life cycle assessment of the manufacture of lactide and PLA biopolymers from sugarcane in Thailand. Int. J. Life Cycle Assess. 15, 970–984. https://doi.org/10.1007/S11367-010-0225-Y/FIGURES/11.
ISO, 2006. Environmental Management – Life Cycle Assessment – Requirements and Guidelines (ISO 14044:2006).
Kunst, J.A.F., Behage, J.E., Jager, J., 2023. A Method of Closed Loop Recycling of Polylactic Acid. WO2023/012142 A2.
MacLeod, M., Arp, H.P.H., Tekman, M.B., Jahnke, A., 2021. The global threat from plastic pollution. Science 373, 61–65. https://doi.org/10.1126/SCIENCE.ABG5433.
Maga, D., Hiebel, M., Aryan, V., 2019. A comparative life cycle assessment of meat trays made of various packaging materials. Sustain. 11 https://doi.org/10.3390/SU11195324.
Meisterling, K., Samaras, C., Schweizer, V., 2008. Decisions to Reduce Greenhouse Gases from Agriculture and Product Transport: LCA Case Study of Organic and Conventional Wheat. https://doi.org/10.1016/j.jclepro.2008.04.009.
Morão, A., de Bie, F., 2019. Life cycle impact assessment of polylactic acid (PLA) produced from sugarcane in Thailand. J. Polym. Environ. 27, 2523–2539. https://doi.org/10.1007/S10924-019-01525-9/TABLES/7.
Morett, I.C., Hamelin, L., Jakobsen, L.G., Junginger, M.H., Steingrimsdottir, M.M., Høibye, L., Shen, L., 2021. Cradle-to-grave life cycle assessment of single-use cups made from PLA, PP and PET. Resour. Conserv. Recycl. 169 https://doi.org/10.1016/J.RESCONREC.2021.105508.
Moretti, C., Hamelin, L., Jakobsen, L.G., Junginger, M.H., Steingrimsdottir, M.M., Høibye, L., Shen, L., 2021. Cradle-to-grave life cycle assessment of single-use cups made from PLA, PP and PET. Resour. Conserv. Recycl. 169, 105508. https://doi.org/10.1016/J.RESCONREC.2021.105508.

NatureWorks, 2022. WWW document. URL. https://www.natureworksllc.com/ (accessed 8.2.22).
Negri, A., Ligthart, T., 2021. Decarbonisation Options for the Dutch Polyolefins Industry — Manufacturing Industry Decarbonisation Data Exchange Network Decarbonisation Options for the Dutch Polyolefins Industry.
OECD, 2022. Plastic leakage and greenhouse gas emissions are increasing [WWW Document]. https://www.oecd.org/.URL (web archive link, 10 April 2022). https://www.oecd.org/environment/plastics/increased-plastic-leakage-and-greenhouse-gas-emissions.htm (accessed 10.4.22).
PBL, 2020. Klimaat- en Energieverkenning 2020.
Piemonte, V., Sabatini, S., Gironi, F., 2013. Chemical recycling of PLA: a great opportunity towards the sustainable development? J. Polym. Environ. 21, 640–647. https://doi.org/10.1007/S10924-013-0608-9.
Plastics Europe, 2022. Plastics — the facts 2022 [WWW document]. URL. https://plasticseurope.org/knowledge-hub/plastics-the-facts-2022/ (accessed 9.7.23).
PRé, 2023. Fact-based sustainability [WWW Document]. URL. https://pre-sustainability.com/ (accessed 8.7.23).
Rezvani Ghomi, E., Khosravi, F., Saedi Ardahaei, A., Dai, Y., Neisiany, R.E., Foroughi, F., Wu, M., Das, O., Ramakrishna, S., 2021. The life cycle assessment for polylactic acid (PLA) to make it a low-carbon material. Polymers (Basel) 13. https://doi.org/10.3390/POLYM13111854.
Rossi, V., Cleeve-Edwards, N., Lundquist, L., Schenker, U., Dubois, C., Humbert, S., Jolliet, O., 2015. Life cycle assessment of end-of-life options for two biodegradable packaging materials: sound application of the European waste hierarchy. J. Clean. Prod. 86, 132–145. https://doi.org/10.1016/J.JCLEPRO.2014.08.049.
Tamburini, E., Costa, S., Summa, D., Battistella, L., Fano, E.A., Castaldelli, G., 2021. Plastic (PET) vs bioplastic (PLA) or refillable aluminium bottles — what is the most sustainable choice for drinking water? A life-cycle (LCA) analysis. Environ. Res. 196, 110974. https://doi.org/10.1016/J.ENVRES.2021.110974.
Tsuji, H., 2005. Poly(lactide) stereocomplexes: formation, structure, properties, degradation, and applications. Macromol. Biosci. 5, 569–597. https://doi.org/10.1002/mabi.200500062.
Van Roijen, E.C., Miller, S.A., 2022. A review of bioplastics at end-of-life: linking experimental biodegradation studies and life cycle impact assessments. Resour. Conserv. Recycl. 181, 106236. https://doi.org/10.1016/J.RESCONREC.2022.106236.
Vogtländer, J.G., Van Der Velden, N.M., Van Der Lugt, P., 2014. Carbon sequestration in LCA, a proposal for a new approach based on the global carbon cycle; cases on wood and on bamboo. Int. J. Life Cycle Assess. 19, 13–23. https://doi.org/10.1007/S11367-013-0629-6/TABLES/2.
Wolberg, C., 2021. The Performance of Recycled Plastics Vs Virgin Plastics [WWW Document]. Ocean Work. URL. https://oceanworks.co/blogs/ocean-plastic-news/the-performance-of-recycled-vs-virgin-plastics (accessed 8.2.22).