Sustainability of bio-based plastics in a circular economy

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Sustainability of bio-based plastics in a circular economy

Sustainability of bio-based plastics in a circular economy

Dissertation

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Contents

Summary

Plastics have become indispensable in modern life due to their versatility and affordability. However, their widespread use has resulted in far-reaching environmental damage, including the accumulation of plastic waste, fossil fuel depletion, and significant greenhouse gas emissions. This environmental damage caused by plastics has raised interest in a circular economy for plastics, with a sustainable approach to production, use and disposal. A circular economy is restorative and regenerative by design, with the aim of eliminating waste. Bio-based plastics have been proposed as a sustainable, circular solution to the environmental issues associated with plastics. Bio-based plastics are (at least partially) based on renewable biomass, in contrast with petrochemical-based plastics used today. Plants absorb CO_2 as they grow, and storing this biogenic carbon in a polymer avoids the usage of fossil fuels. However, bio-based plastics are not implicitly sustainable or circular. These aspects are influenced by how a plastic is produced and how it is recovered at end-of-life, implying that careful attention needs to be paid to material development and product design. The research goal of this PhD was therefore: *To explore how material development and product design can enable bio-based plastics to be circular.*

Initially, the PhD research focused on durable products. Durable products constitute a significant fraction of the bio-based plastic market, but there are no resources available for sustainable design of these products. Applying bio-based plastics in durable products requires different considerations compared to applying them in single-use applications. At the time of this study, it was not yet known how value chain actors in durable product value chains perceived bio-based plastics. To understand what challenges these value chain actors face when using bio-based plastics, a workshop involving stakeholders spanning a durable products value chain was conducted. While participants displayed keen interest in bio-based plastics, they perceived the bio-based plastic value chain as severely underdeveloped and they lacked access to dependable information on their usage. The circularity and lower environmental impact of bio-based plastics were identified as strong drivers, but these concepts were considered ill-defined. Participants indicated confusion regarding the recovery of bio-based plastics at end-of-life. Furthermore, participants indicated that the environmental impact of bio-based plastics needed to be better qualified to justify their usage. The lack of reliable information, combined with a lack of knowledge among value chain actors and the public, lead to a risk of greenwashing when using bio-based plastics. The immature value chain of bio-based plastics appeared to be stuck, and would require substantial government stimulation to grow.

After this first study, it became apparent that even base knowledge such as environmental impact and recovery options at end-of-life for bio-based plastics was missing. Therefore, the focus moved away from durable products, towards a holistic understanding of the circularity and environmental impact of bio-based plastics. To achieve this, any available information for all commercially produced bio-based plastics was included in the analyses.

True circularity of bio-based plastics requires efficient material recovery at end-of-life. Recovery is not only determined by material properties, but also by product design. At the time of writing, there were no resources for product designers that facilitate design for recovery using bio-based plastics. To address this gap, a rigorous literature review was conducted. A framework for the recovery of bio-based plastics was developed, with 8 recovery pathways: mechanical recycling, dissolution, solvolysis, enzymatic depolymerisation, thermochemical recycling, anaerobic digestion, aerobic digestion, and incineration. Incineration and biodegradation can be considered circular recovery pathways for bio-based plastics due to their biogenic carbon, whereas they are linear for petrochemical-based plastics. The compatibility of these recovery pathways with commercially available bio-based plastics was studied.

Based on technical characteristics of specific recovery pathways, recommendations for product design were established. For mechanical recycling, polymer blending, multimaterial manufacturing and using certain additives should be avoided. During dissolution, solvolysis and enzymatic depolymerisation, many additives and blends can be separated, but due to the (typically) high energy requirements of these processes, they should be avoided for thick-walled parts. Thermochemical recycling can in theory deal with any (bio-based) plastic, but significant changes in plastic composition may affect the value of thermochemical recycling products. For industrial anaerobic and aerobic digestion, product designers should avoid non-biodegradable additives and ensure that the product will completely biodegrade under the conditions of commercial composting plants. When targeting biodegradation in nature, thick-walled parts should be avoided since they have relatively longer degradation times.

In addition to recovery, the environmental impact of bio-based plastics is essential knowledge for circular product design. A well-established tool for calculating the environmental impact of processes, products, or services is lifecycle assessment (LCA). The outcomes of published LCAs of bio-based plastics vary by several orders of magnitude for the same plastic. This has been primarily attributed to methodological inconsistencies and poor quality data.

In order to quantify the role of methodological inconsistencies and poor data quality and identify other factors affect the environmental impact of bio-based plastics, LCAs based on lifecycle inventory (LCI) data in literature were conducted. A total of 34 scenarios for bio-based high-density polyethylene (HDPE) and polyethylene terephthalate (PET) were compared with petrochemical-based equivalents. Remarkably, a consistent methodology did not decrease the variations in bio-based plastics LCAs. A detailed study of the LCIs revealed that the differences were the results of four factors: biomass type, biomass cultivation location, lack of data, and limited scope of the studied LCAs. The type of biomass on which the plastic was based as well as the location of production were found to affect the environmental impact significantly. The lack of high-quality LCI data regarding biomass cultivation and chemical conversion processes resulted in gaps in the LCIs which were perpetuated in the LCA outcomes. Finally, the limited scope of the studied LCAs meant that LCI data for some impact categories was missing, resulting in potentially misleading outcomes.

The effect of biomass sourcing on the environmental impact of bio-based plastics was further explored in the final study of the PhD. 31 biomass sourcing scenarios for bio-

based HDPE were compared to petrochemical-based HDPE. Additionally, five scenarios for plastic recovery were studied for each biomass sourcing scenario: mechanical recycling, incineration with and without energy recovery, sanitary landfill, and unsanitary landfill. For first generation biomass feedstock (i.e., edible crops), the direct availability of targeted molecules correlated with a lower environmental impact of a bio-based plastic. In this research, the targeted substance was sugar. The crops with high concentrations of simple sugars, such as sugar beet or sugarcane, resulted in a relatively low environmental impact across impact categories. Other biomass types such as maize and potatoes resulted in substantially higher environmental impacts. These biomass types were high in starch instead sugar, and that starch still needed to be broken down into sugars. For maize, the environmental impact of growing them was much higher than that of sugar-based crops, and for potatoes, a relatively large mass of potatoes was required. The production location affected the resources needed for biomass cultivation (such as water and fertiliser requirements and harvesting methods), and the environmental impact of processing due to the energy mix.

Two strategies for accounting for biogenic carbon (carbon absorbed from the atmosphere during plant growth) were compared: accounting for carbon during the plastic production or upon molecular decomposition. The latter strategy led to results that were not in line with circular economy principles. Bio-based plastics could only 'benefit' from biogenic carbon if cradle-to-grave emissions were considered and the plastic was incinerated. This meant that in cradle-to-gate (i.e. only plastic production) CO_2-eq emissions, bio-based plastics were disadvantaged. Furthermore, incineration was the recovery pathway with the lowest CO_2-eq emissions, compared to e.g. mechanical recycling. This is counter-intuitive with circular economy principles, which prioritise keeping the plastic at its highest possible value. When biogenic carbon was accounted for during production, mechanical recycling resulted in the lowest CO_2-eq emissions. Therefore, biogenic carbon should be incorporated in the production stage of bio-based plastics LCAs.

Circular product design with bio-based plastics requires careful consideration of biomass sourcing and recovery. Although much information regarding these aspects is still missing, the research presented in this dissertation provides some guidelines for circular product design with bio-based plastics. In order to reduce environmental impacts, bio-based plastics should be produced with agricultural by-products or with biomass types with a high conversion efficiency. Biomass for bio-based plastics should be cultivated with minimal use of land, water, chemicals and fossil fuels. Environmental impacts can be reduced further by using renewable energy in the production process. Product designers should also consider what recovery pathway they want to target at end-of-life of a product. The plastic composition and product architecture need to reflect the targeted recovery pathway.

Samenvatting

Plastics zijn onmisbaar onze maatschappij vanwege hun veelzijdigheid en betaalbaarheid. Het enorme plastic gebruik zorgt echter voor verstrekkende milieuschade, waaronder de opeenhoping van plastic afval, grondstof gebruik, en de uitstoot van broeikasgassen. De milieuschade die plastics aanrichten creëert groeiende interesse in een circulaire economie voor plastics, met een duurzame benadering voor de productie, het gebruik en de einde levensduur van plastics. Een circulaire economie is van nature herstellend of regeneratief, met als doel verspilling te minimaliseren. Biogebaseerde, 'bio-based', plastics bieden een duurzame, circulaire oplossing voor de milieuproblematiek rond plastics. Bio-based plastics zijn plastics die (in ieder geval gedeeltelijk) gebaseerd zijn op hernieuwbare biomassa, in tegenstelling tot bijna alle plastics die nu worden gebruikt. Planten absorberen CO_2 als ze groeien. Het opslaan van deze zogeheten 'biogene' koolstof in een plastic vermijdt het gebruik van fossiele grondstoffen. Bio-based plastics zijn echter niet impliciet duurzaam of circulair. Deze aspecten worden beïnvloedt door de manier waarop een plastic wordt geproduceerd en hoe het aan het einde van de levensduur wordt teruggewonnen. Dit impliceert een belangrijke rol voor materiaalontwikkeling en productontwerp. Het onderzoeksdoel van dit doctoraat was dan ook: *Onderzoeken hoe materiaalontwikkeling en productontwerp het mogelijk kunnen maken dat bio-based plastics circulair zijn.*

In eerste instantie richtte het promotieonderzoek zich op zogeheten 'durable' producten. Dit zijn producten met een langere levensduur dan wegwerpplastics. Durable producten vormen een aanzienlijk marktdeel voor bio-based plastics, maar er zijn geen hulpmiddelen of richtlijnen voor duurzaam productontwerp van durable producten met deze materialen. Het ontwerpproces van een durable product is anders dan dat van wegwerpproducten. Het was nog niet onderzocht hoe mensen in de waardeketen van durable producten naar bio-based plastics kijken. De uitdagingen van bio-based plastics voor belanghebbenden in de waardeketen van durable producten werden geïdentificeerd door middel van een workshop. Hoewel de deelnemers veel belangstelling hadden voor bio-based plastics, ondervonden ze een onderontwikkelde waardeketen en een gebrek aan betrouwbare informatie. De circulariteit en duurzaamheid van bio-based plastics werden gezien als sterke drijfveren, maar deze aspecten werden ook gezien als slecht onderbouwd. Deelnemers gaven aan dat er verwarring bestaat over hoe bio-based plastics einde levensduur kunnen worden teruggewonnen. Daarnaast gaven deelnemers aan dat de milieu-impact van bio-based plastics nog niet goed genoeg gekwantificeerd is om het gebruik te rechtvaardigen. Het gebrek aan betrouwbare informatie en kennis bij zowel belanghebbenden als in de maatschappij leidt tot een risico op greenwashing bij het gebruik van bio-based plastics. Uit de workshop bleek de waardeketen van bio-based plastics vast te zitten, en deze zou grote stimulatie vanuit overheden nodig hebben om te groeien.

Na dit eerste onderzoek werd het duidelijk dat zelfs basiskennis over de milieueffecten en terugwinningsmogelijkheden bij einde van de levensduur van bio-based plastics ontbrak. Daarom verbreedde de focus van het onderzoek van duurzame producten naar een holistisch begrip van de circulariteit en de milieu-impact van bio-based plastics. Hiervoor werd alle beschikbare informatie over alle commercieel beschikbare bio-based plastics meegenomen in de volgende analyses.

Om bio-based plastics echt circulair te gebruiken is een efficiënte materiaalterugwinning aan het einde van de levensduur nodig. Materiaalterugwinning wordt niet alleen bepaald door de materiaaleigenschappen, maar ook door productontwerp. Toen dit onderzoek begon waren er nog geen hulpmiddelen voor productontwerpers om materiaalterugwinning van bio-based plastics te faciliteren. Om bij te dragen aan zo een hulpmiddel werd een rigoureus literatuuronderzoek uitgevoerd. Er werd een raamwerk voor de terugwinning van bio-based plastics ontwikkeld, met 8 opties voor materiaalterugwinning: mechanische recycling, oplossing, solvolyse, enzymatische depolymerisatie, thermochemische recycling, anaerobe vergisting, aerobe vergisting en verbranding. Verbranding en biologische afbraak zijn circulair voor bio-based plastics, waar ze lineair zijn voor plastics gemaakt van fossiele brandstoffen. De geschiktheid van deze terugwinningsprocessen voor commercieel verkrijgbare bio-based plastics werd verder onderzocht.

Op basis van de technische kenmerken van specifieke materiaalterugwinningsprocessen werden aanbevelingen voor productontwerp opgesteld. Voor mechanische recycling is het belangrijk om blending, multimateriaal fabricage, en het gebruik van specifieke additieven te vermijden. Bij oplossing, solvolyse en enzymatische depolymerisatie kunnen veel additieven en blends wel afgescheiden worden, maar door het meestal hoge energieverbruik van deze processen moeten producten zo dun mogelijk zijn. Thermochemische recycling kan (in theorie) alle bio-based plastics verwerken, maar grote veranderingen in de plastic compositie kunnen de waarde van het product van thermochemisch recyclen aantasten. Voor industriële compostering (aeroob of anaeroob) moeten productontwerpers niet-biodegradeerbare additieven vermijden, en ervoor zorgen dat plastic onderdelen volledig degraderen in een standaard industriële compostcyclus. Wanneer biodegradatie in natuur wordt beoogd, moeten dikwandige onderdelen worden vermeden, omdat deze meer tijd nodig hebben om volledig te degraderen.

Naast materiaalterugwinning is de milieu-impact van bio-based plastics essentiële kennis voor circulair productontwerp. Een levenscyclusanalyse (LCA) is een bekende methode om de milieu-impact van processen, producten of diensten te bestuderen. Er zijn verschillen van meerdere odegroottes tussen de uitkomsten van gepubliceerde LCAs van bio-based plastics. Dit werd voornamelijk toegeschreven aan methodologische inconsistenties en de slechte kwaliteit van beschikbare data.

Om de milieu-impact van bio-based plastics, en de rol van slechte datakwaliteit daarin, te onderzoeken, zijn LCAs uitgevoerd op basis van levenscyclusinventarisatie (LCI) data uit de wetenschappelijke literatuur. In totaal 34 scenario's voor bio-based polyethyleen en polyethyleentereftalaat zijn vergeleken met fossiel-gebaseerde equivalenten. Opvallend genoeg vergrootte een consistente methodologie de variaties tussen LCA uitkomsten. Bij het uitpluizen van de LCIs bleek dat de verschillen het resultaat waren door vier factoren: het soort biomassa, waar deze biomassa gecultiveerd is, het gebrek aan goede gegevens,

en de gelimiteerde omvang van bestaande LCAs. Het soort biomassa waarvan het plastic gemaakt is en de productielocatie bleken een groot effect te hebben op de milieu-impact. Het gebrek aan goede gegevens over de teelt van biomassa en de chemische conversieprocessen zorgde voor grote gaten in de LCIs die tot uiting kwamen in de LCA-uitkomsten. Het gelimiteerde bereik van de bestaande LCAs betekende dat de LCI data voor sommige impact categorieën niet in de analyse waren opgenomen, met potentieel misleidende uitkomsten als resultaat.

Het effect van de inwinning van biomassa op de milieu-impact van bio-based plastics werd verder onderzocht in de laatste studie van het doctoraat. Er werden 31 scenario's voor inkoop (biomassa type en teeltlocatie) voor bio-based hogedichtheidpolyethyleen (HDPE) vergeleken met petrochemisch-gebaseerd HDPE. Daarnaast werden voor elk inwinningsscenario vijf opties voor materiaalterugwinning onderzocht. Voor eerste generatie biomassa (eetbare gewassen) was de directe beschikbaarheid van de doelwitmoleculen een belangrijke voorspeller van de milieu-impact van bio-based plastics. In dit onderzoek was de doel stof suiker en hadden de gewassen met hoge concentraties eenvoudige suikers, zoals suikerbiet of suikerriet, een relatief lage milieubelasting. Andere soorten biomassa, zoals mais en aardappels, hadden substantieel hogere milieu-impacts. In deze soorten biomassa zat de suiker in zetmeelmoleculen, die nog afgebroken moesten worden in suikers voor ze gefermenteerd konden worden. De milieu impact van het groeien van mais was veel hoger dan dat van suiker gebaseerde gewassen. Bij aardappels kwam de hoge milieu impact door de grote hoeveelheden die er nodig waren. De productielocatie beïnvloedde de grondstoffen die nodig zijn voor de teelt van biomassa en de milieu-impact van de verwerking als gevolg van de energiemix.

Er werden twee strategieën voor de verrekening van biogene koolstof vergeleken. De eerste strategie was het meenemen van de biogene koolstof tijdens de productie van het plastic. Bij de tweede strategie werd de biogene koolstof alleen meegenomen bij moleculaire afbraak. Deze laatste strategie leidde tot resultaten die niet overeenstemmen met de principes van de circulaire economie. Bio-based plastics zouden alleen kunnen 'profiteren' van biogene koolstof op het moment dat het plastic verbrandt wordt. Dit betekent dat de productie broeikasgas uitstoot hoger uitvallen en bio-based plastics hierin worden benadeeld. Bovendien had verbranding van bio-based plastics hierdoor een lagere broeikasgas uitstoot dan bijvoorbeeld mechanische recycling. Dit is in strijd met de principes van een circulaire economie, waarin het behoud van waarde in materialen de hoogste prioriteit heeft. Wanneer er in de productie van bio-based plastics rekening gehouden wordt met biogene koolstof, resulteerde mechanisch recyclen in de laagste broeikasgas uitstoot. Daarom is het belangrijk om biogene koolstof in een LCA mee te nemen bij de productie van het plastic.

Circulair productontwerp met bio-based plastics vereist een zorgvuldige overweging van de inwinning en terugwinning van biomassa. Hoewel er nog veel informatie over deze aspecten ontbreekt, biedt het onderzoek in dit proefschrift richtlijnen voor circulair productontwerp met bio-based plastics. Om de milieu-impact te verminderen moeten bio-based plastics worden geproduceerd met agrarische bijproducten of met biomassasoorten met een hoge conversie-efficiëntie: dat wil zeggen dat er zo min mogelijk biomassa nodig is voor de productie van bio-based plastic en dat de impact van deze biomassa zo laag

mogelijk moet zijn. Biomassa voor bio-based plastics moet worden verbouwd met minimaal gebruik van land, water, chemicaliën en fossiele brandstoffen. De milieueffecten kunnen verder worden verminderd door hernieuwbare energie in het productieproces te gebruiken. Productontwerpers moeten ook overwegen welk materiaalterugwinningsproces zij willen nastreven aan het einde van de levensduur van een product. De kunststofsamenstelling en productarchitectuur moeten het beoogde materiaalterugwinningsprocess weerspiegelen.

1

Introduction

1

Plastics are one of the most successful material types of the last century. Plastics are inexpensive and can have a wide range of properties. The main component in plastics is polymers, which are very large molecules consisting of repeating chemical units called monomers which are typically produced from fossil fuels. These polymers can have a wide range of chemical structures and properties, and are compounded with additives to further tailor the properties of the plastic. As a result, plastics are ubiquitous in everyday life: in household items, clothing, electronics and, very often, the packaging they come in. Plastics are also applied in demanding applications, such as protective sports equipment [1], medical devices [2] and aircraft structural components [3]. Plastics provide society with many benefits, such as reduced food waste, and lighter (and, by extension, more fuel efficient) aircraft and cars [4]. Plastics were only commercialised in the 1950's, but between 1950 and 1977 the annual plastics production grew from 1.5 million metric tonnes to 50 million metric tonnes [5]. Today, around 400 million metric tonnes of plastics are produced annually, and their market continues to grow [5].

Despite their many benefits, plastics are causing environmental damage on an enormous scale. The current plastic economy uses finite resources to produce products that become waste that leak into nature or are landfilled or incinerated, and is therefore a highly linear system. In 2015, plastics consumed 8% of all crude oil extracted and caused 4.5% of global greenhouse gas emissions [6]. Plastics are expected to account for 15% of the greenhouse gas emissions budget to limit global temperature rise to 1.5 °C by 2050 [7]. At the same time, only 9% of all plastics ever produced has been recycled, while 79% has accumulated in landfills or the natural environment [8]. Resistance to degradation is an important property for plastics in many applications, but this also means that most plastics do not biodegrade in the natural environment. Instead, plastics degrade slowly over many decades or even centuries into small fragments: micro- and nano-plastics. Plastic fragments have now been discovered in water, animals and human blood [9].

The resource use, environmental impact and pollution of plastics have raised interest in a circular economy for plastics, with a sustainable approach to production, use and disposal. A circular economy is restorative and regenerative by design, with the aim of eliminating waste [10]. In 2019, the European Commission published policy recommendations to achieve a circular economy for plastics [11]. Part of the strategy focuses on prolonging the lifetime of plastic products, through product reuse and repair. When re-use and repair are not an option, the focus shifts to waste management. In a circular economy, materials should be recirculated at their highest possible value [10]. For plastics, this means recycling through mechanical or chemical recycling. However, not all plastics can be mechanically or chemically recycled, and these will then be incinerated. Combined with the growing plastic demand, this still requires the extraction of finite, fossil, resources.

Bio-based plastics have been proposed as a circular solution to the environmental issues related to plastics. The polymers in bio-based plastics are based (at least partially) on biomass, rather than petrochemicals from fossil fuels [12]. Plants absorb CO_2 as they grow. Storing this biogenic carbon in a polymer lowers the greenhouse gas emissions associated with plastic production. By using a renewable feedstock instead of finite fossil fuels, bio-based plastics enable circularity at the polymer production level. Although only

representing 1% of the plastic market today, the market for bio-based plastics is growing at over three times the rate of that of petrochemical-based plastics [13].

Given that improved circularity and reduced environmental impact are the main drivers for a transition to bio-based plastics, one would expect these aspects to be well-defined. Unfortunately, the opposite is the case. The environmental impact and circularity of bio-based plastics production has long been, and continues to be, a debated topic [14–17]. While bio-based plastics have the potential to reduce the environmental impact of the plastics industry, scientific literature has been inconclusive about whether bio-based plastics have a lower environmental impact than petrochemical-based plastics [17]. Moreover, bio-based plastics do not necessarily biodegrade in nature, so they can still contribute to plastic pollution issues if they are not recovered at end-of-life [16].

Bio-based plastics are not implicitly circular. Circularity is influenced by how a plastic is produced and how it is recovered at end-of-life. This means that simply replacing petrochemical-based plastics by bio-based plastics will not automatically reduce the environmental impact of plastics. Instead, careful attention needs to be paid to material development and product design. This poses a challenge to product designers, who need to ensure that the bio-based plastics they use are sustainable and that they can be recovered effectively after use.

This chapter provides background information for the research presented in this dissertation. In section 1.1, the definitions and categorisations of bio-based plastics are given. Section 1.2 elaborates on the role of bio-based plastics in a circular economy. In section 1.3 knowledge gaps that were tackled in this PhD research and the associated research questions are presented. Finally, in section 1.5 the outline of the thesis is presented.

1.1 Bio-based plastics and their applications

1.1.1 Glossary

The field of bio-based plastics is one of confusing terminology. It is therefore important to establish a few definitions before further exploring the topic. Below, the most important terms relating to bio-based plastics are defined.

Polymer: very large molecules consisting of repeating chemical units called monomers.

Monomer: a small molecule that can react together with other monomers to form larger chains or networks.

Plastic: "a material that contains as an essential ingredient one or more organic polymeric substances of large molecular weight, is solid in its finished state, and, at some stages in its manufacture or processing into finished articles, can be shaped by flow" [18].

Bio-based polymer: a polymer in which (part of) the monomers are derived from bio-mass instead of from petrochemicals [12].

Bio-based plastic: a plastic containing a polymer in which (at least part of) the monomers are derived from biomass.

Biodegradable plastic: a plastic that can be degraded by naturally-occurring micro-organisms such as bacteria, fungi and algae [19]. It is a common misconception that all

bio-based plastics are biodegradable [16]. A potential cause for this misconception is the term "*bioplastic*", which refers to a plastic that is either bio-based, biodegradable or both.

Drop-in bio-based polymer: bio-based polymer that is chemically identical to a preexisting petrochemical-based plastic [20].

Dedicated bio-based polymer: bio-based polymer that does not have a petrochemical-based equivalent [20].

1.1.2 The history of bio-based plastics

The very first human-made plastics were bio-based. They contained semi-synthetic polymers based on cellulose developed in the mid-1800's [21]. Cellulose-based plastics replaced expensive materials such as ivory in everyday items like buttons, fashion accessories and knife handles. The first fully synthetic plastic was Bakelite, which was based on fossil fuels [22]. Since its commercialisation in the 1920's, nearly all plastics produced have been based on fossil fuels.

A global oil crisis reignited the interest in bio-based polymers in the 1970's [23, 24]. At the same time, the first scientific studies of the environmental damages caused by plastics were published. The first scientific study of marine plastic debris was published in 1972, reporting small plastic fragments in the Sargasso Sea which could potentially be related to plasticisers found in marine animals [25]. Later that year, the same authors published about fish consuming polystyrene spherules in the coastal waters of New England [26]. The magnitude of the impact of plastic waste on the oceans became apparent in the next decades. In 1985, the plastic concentration in the North Atlantic Subtropical Gyre ocean was 15,000 pieces per square km [27]. The physical impact of plastics in the ocean also became apparent: birds, sea turtles and sea lions got entangled in fishing nets or starved because they mistook plastics for food [28]. This signalled the need to change the plastics industry.

The 21st century saw a surge in the development of bio-based polymers. In 2022, 1.5% of all bio-based plastics produced was bio-based. There are now bio-based alternatives for a wide variety plastics. Some of these alternatives are chemically identical to preexisting petrochemical-based plastics, referred to as drop-in bio-based polymers [20]. Examples of drop-in bio-based polymers are bio-based polyethylene (PE) or polyethylene terephthalate (PET). Alternatively, there are bio-based polymers for which no petrochemical-based equivalent exists, called dedicated bio-based polymers [20]. The most common dedicated bio-based polymers are polylactic acid (PLA), and cellulose acetate [13]. There are also bio-based polymers that have petrochemical-based equivalents with the same group name, but not the same molecular structure. These are often engineering-grade polymers where the name refers to a group of polymers characterised by a repeating bond, for instance polyurethane or polyamide.

Figure 1.1 contains an overview of how bio-based plastics were applied in 2022. As the overview shows, there are now bio-based plastic alternatives for many applications [13]. By far the largest fraction of bio-based plastics was used in packaging, followed by consumer goods and automotive. The range of available bio-based polymers remains, however, much more limited than that of conventional plastics. Therefore, bio-based plastics are not

yet commonly applied in more specialised or high-performance applications, but some high-performance bio-based plastics have been developed in lab quantities [29, 30].

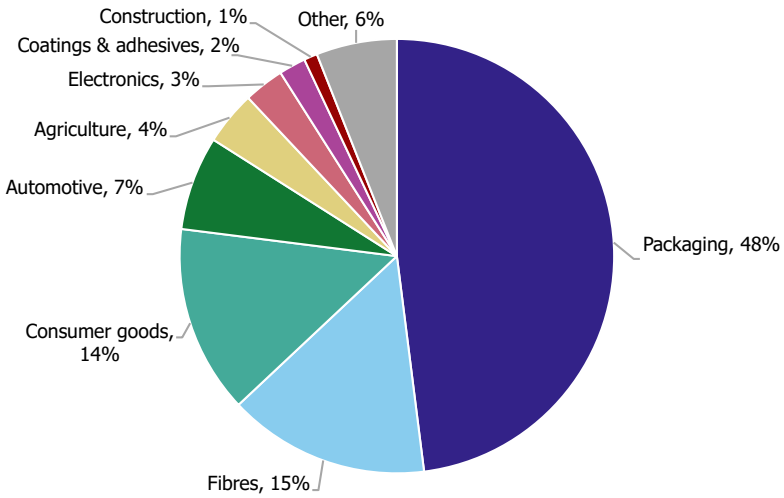

Figure 1.1: Overview of the market segments of bio-based plastics in 2022 [13].

1.1.3 Producing bio-based polymers

There are a number of ways to produce bio-based plastics. By far the most common route to produce bio-based polymers is through the chemical conversion of sugars, proteins or fats from plants [31]. A relatively simple example is the production of bio-based ethylene. Sugars in plants are fermented into ethanol, which is then dehydrated into bio-based ethylene [32, 33]. The bio-based ethylene can then be used to produce for example bio-based PE, bio-based PET or polyethylene furan-2,5-dicarboxylate (PEF). Similarly, for the production of polylactic acid (PLA), sugars are fermented into lactide which is then either condensed directly into PLA or into lactide which is then further processed into PLA. Some bio-based polymers can be derived directly from biomass, such cellulose-based polymers and thermoplastic starch. Finally, bio-based monomers can be produced through the cracking of oils obtained from biomass, in a process resembling the cracking of crude oil [34].

The resources from which bio-based plastics are derived are categorised into four so-called “feedstock generations”, each with important advantages and drawbacks [16, 35]. First generation feedstocks are based on edible crops. Edible biomass has been grown to contain high concentrations of nutrients which are often the basis of bio-based chemicals. Nevertheless, the usage of first generation feedstock has the risk of directly competing with food resources for people, which is an important argument against its usage [16, 36]. Second generation feedstocks are based on non-edible crops and agricultural by-products. These feedstocks reduce food competition, but non-edible crops still require land and water

to grow. Furthermore, second generation feedstocks may require extensive reprocessing with low yield, making them potentially economically unviable [37]. Third generation feedstocks are based on algae, which do not require land and have high yields [16]. However, third generation feedstocks for bio-based polymers are still in development and there are doubts about scalability [38]. Finally, fourth generation feedstocks based on genetically modified plants, algae and microorganisms are also being developed [38]. Genetically modified biomass could result in lower resource uses, but this could also have unintended consequences such as adverse effects on other organisms, new diseases, or antibiotic resistance [39].

1.2 Bio-based plastics in a circular economy

A circular economy aims to minimise waste while maximising resource efficiency. Figure 1.2 shows the circular economy butterfly diagram, which illustrates the flow of materials in a circular economy. The bubterfly diagram distinguishing between biological nutrients and technological nutrients in a separate biocycle and a technocycle. In the technocycle, materials are reintroduced back into the circular economy through "loops" like reuse and mechanical recycling. In the biocycle, materials are eventually cycled back into the biosphere as CO_2 and other simple molecules which are then available for re-absorption by biomass.

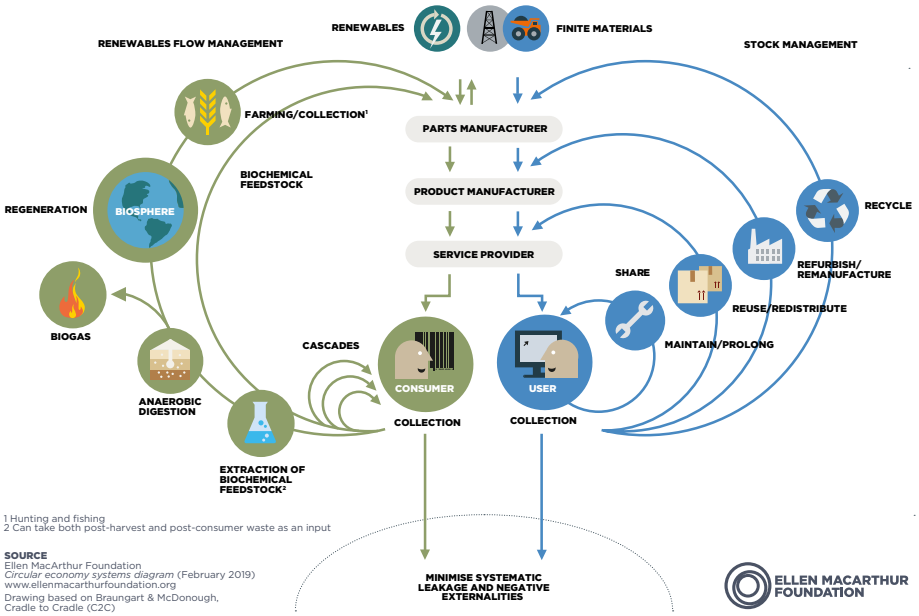

Figure 1.2: The circular economy butterfly diagram, from [10].

Petrochemical-based plastics can only work in the technological cycle of the circular economy. They are produced from non-renewable resources, so after use they need to be recirculated indefinitely in technical loops. However, completely achieving a circular economy with petrochemical-based plastics is unlikely. Some plastics cannot be mechanically recycled because the mechanical recycling processes induces too much damage in the polymer chain or because the plastic becomes too contaminated during use. Those that can be mechanically recycled often suffer a loss in properties and cannot be used in the original application, eventually becoming so damaged that they are unusable [40, 41]. Furthermore, some product types may not be produced with recycled plastics, such as food-contact products [42]. Chemical recycling technologies can recover monomers from some specific types of polymers [43]. However, not all polymers are suitable for chemical recycling and it is currently only industrialised at the pilot level. At the same time, the demand for plastics is still growing [16]. As such, virgin plastics will need to be produced and non-recyclable plastics become waste. In Europe, the most common way to deal with plastics that cannot be recycled is to incinerate them, releasing fossil CO_2 into the atmosphere [11]. As such, a circular economy with petrochemical-based plastics is unrealistic, necessitating a more sustainable approach to plastic production and recovery.

Bio-based plastics are often presented as a key component of a circular plastics economy [11, 41, 44–48]. The carbon in bio-based polymers is biogenic, i.e., it has been derived from the atmosphere rather than from fossil resources. Bio-based plastics therefore enable circularity at the polymer production level. They can also be reused, used in repaired products and many types of bio-based plastics can be mechanically or chemically recycled (depending on chemical composition and available infrastructure). However, when bio-based plastics undergo molecular decomposition (due to incineration or biodegradation), they release biogenic carbon and do therefore not contribute new CO_2 to the atmosphere and this may be considered a circular loop. Bio-based plastics can then be regenerated through the growth of new biomass. This means that bio-based plastics do not fit into the technological and biological nutrient dichotomy. They can be used in both the biocycle and the technocycle. Therefore, Bakker & Balkenende have suggested an alternative diagram that does not make this distinction: the rainbow diagram, seen in figure 1.3 [49].

Although bio-based plastics enable new circular loops through molecular decomposition, it remains important to guarantee that a bio-based plastic enters the intended circular loop. Molecular decomposition can occur by incineration of a plastic or biodegradation. During incineration, a plastic is combusted in a controlled environment. Plastics release heat upon incineration, which may be recovered as electricity. During biodegradation, a plastic is digested by micro-organisms. If oxygen is present during biodegradation, plastics typically decompose into CO_2, water, and inorganic compounds [16]. Most bio-based plastics are not biodegradable at all [46]. Of those that are biodegradable, very few degrade in natural environment [16], but require prolonged periods of elevated temperatures to fully biodegrade. Therefore, it remains necessary to facilitate proper end-of-life management for bio-based plastics.

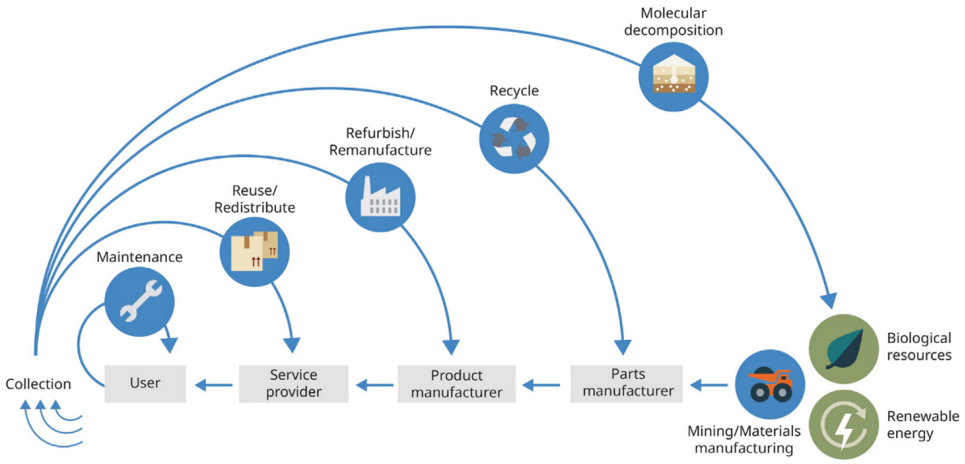

Figure 1.3: Rainbow diagram for the circular economy that removes the dichotomy between biological nutrients and technological nutrients, from [49].

1.3 The environmental impact of bio-based plastics

Producing bio-based plastics producing bio-based plastics involves many steps that all have an environmental impact. It is therefore important to quantify the environmental impact of each step. Lifecycle assessment (LCA) is an established tool to study the environmental impact of a product, process or service. In product design, LCA can be used to compare different design decisions such as material choice in terms of environmental impact. It is therefore a useful tool to compare the environmental impact of bio-based plastics to that of petrochemical-based plastics. The ISO 14040 [50] and ISO 14044 [51] standards contain methodologies for conducting LCAs. An LCA consists of four stages, as illustrated in figure 1.4 The goal and scope definition, inventory analysis, impact assessment and interpretation are carried out iteratively. The starting point of an LCA is the the goal and scope definition, where the aim of the LCA is established as well as the breath and depth of the study. During the inventory analysis, all relevant inputs and outputs are compiled and quantified in into a lifecycle inventory (LCI). In the lifecycle impact assessment (LCIA), this lifecycle inventory is converted into environmental impacts. Finally, in the interpretation, conclusions are drawn from the LCI and lifecycle impact assessment, and the completeness, consistency and sensitivity are reflected upon.

Comparing bio-based plastics and petrochemical-based plastics has proven to be challenging. Petrochemical-based plastics have well-defined resources and processes that are carried out on an enormous scale: hundreds of millions of tonnes per year. In contrast, the market segment of bio-based plastics is still small (3.9 million tonnes [13]) and most bio-based plastics are only produced by one or two companies [1, 52]. As a result, the data used to conduct an LCA of bio-based plastics is scarcely available, it at all [14]. Furthermore, many bio-based plastics are still only produced in lab quantities or at the pilot scale and LCA data is not representative of large scale production. These factors combined have

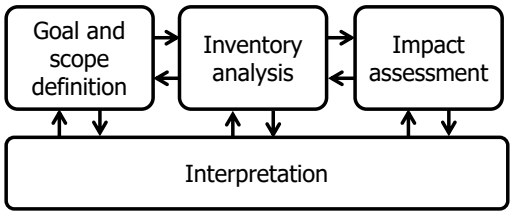

Figure 1.4: Illustration of the different stages of an LCA (based on [50]).

made it difficult to conduct a reliable bio-based plastic LCA. As a result, existing bio-based plastic LCAs can have widely different impacts for the same type of plastic [14, 17]. In existing research, bio-based plastics sometimes yielded a higher environmental impact than petrochemical-based plastics, suggesting that they may not always be a more sustainable alternative today [14, 16, 17, 53].

1.4 Knowledge gaps and research questions

While bio-based plastics hold the potential to lower the environmental impact of the plastics industry, they do not automatically solve the environmental issues related to plastics. The primary research goal of this PhD was therefore:

To explore how material development and product design can enable bio-based plastics to be sustainable and circular.

To contribute to this research goal, we identified four research gaps on which we based four studies that are presented in the next four chapters of this thesis. The knowledge gaps and research questions are elaborated below.

Initially, the PhD research focused on durable products. Bio-based plastics are commonly applied in single-use packaging products and the opportunities and challenges for this market segment are extensively documented [36, 54–58]. Durable products constitute a significant fraction of the bio-based plastic market, but there are no resources available for product designers. At the same time, the use of plastics in single-use applications is limited by environmental legislation in the European Union [59] as well as other countries [60], suggesting that bio-based plastics applied in durable products will become more important. Applying bio-based plastics in durable products requires different considerations and it is not yet known how value chain actors in durable product value chains perceive bio-based plastics. The research question was therefore:
Research question 1: *What challenges do actors throughout a durable product's value chain face when using bio-based plastics?*

After this first study, it became apparent that much of the base knowledge about the sustainable use of bio-based plastics was unknown. Therefore, the focus moved away from durable products, and more towards a holistic understanding of the circularity and environmental impact of bio-based plastics. To achieve this, any available information for all commercially produced bio-based plastics was included.

The second knowledge gap addressed in this PhD research was the recovery of bio-based plastics. The ability to recover a plastic at end-of-life depends on the infrastructure, the material composition, and product design. The importance of product design in the transition to a circular economy with bio-based plastics has already been highlighted [49, 52, 61–64], but there were no overviews of how product design can help the recovery at end-of-life for bio-based plastics. The second research question was:
Research question 2: *How can product designers consider the recovery of bio-based plastics during the design process?*

The outcomes of LCAs of bio-based plastics vary by several orders of magnitude for the same plastic, as highlighted in section 1.3. This makes it impossible to conclude whether bio-based plastics are a more sustainable option compared to petrochemical-based plastics [15, 53]. Studies comparing methodologies and outcomes of published bio-based plastic LCAs have attributed the inconsistencies in LCA outcomes for bio-based plastics to methodological inconsistencies and poor quality data. Methodological inconsistencies concerned many aspects of the LCA, including land-use-change, biogenic carbon accounting, and allocation methods [14, 17]. The poor data quality mainly related to chemical conversion processes used to transform biomass into monomers, for which accurate LCA data is sparsely available [14]. If methodological inconsistencies were removed from LCA studies, this could reveal other factors that affect the environmental impact of bio-based plastics and that can provide useful guidance to plastic developers and product designers that produce and use bio-based plastics. In order to quantify the role of poor quality data and understand if any other factors affect the environmental impact of bio-based plastics, the third research question was set-up.
Research question 3: *How does using a consistent methodology affect the LCA outcomes of bio-based plastics?*

The environmental impact of bio-based plastics depends on many aspects of their production process. Bio-based plastics can be produced from different types of biomass and in different locations. For biofuels, it has been shown that the type of biomass and the cultivation location affect the environmental impact of bio-based chemicals [65]. For bio-based plastics, this effect has been explored in limited comparisons of two or three biomass types and locations [66–69]. As discussed above regarding research question 3, these LCA outcomes cannot be compared directly due to methodological inconsistencies. Therefore, it remains difficult to understand how biomass sourcing (biomass type and production location) affect the environmental impact. Therefore, research question four was set-up.
Research question 4: *Under which biomass sourcing conditions does bio-based polyethylene result in a lower environmental impact than petrochemical-based equivalents?*

Polyethylene was selected for this study since it is a bio-based polymer that is already produced industrially in Brazil with a production capacity of 0.2 megatonnes in 2018 [70]. Bio-based polyethylene is based on ethanol, for which many production facilities already exist since it is also a biofuel. Furthermore, there is a publicly available LCI for the conversion of bio-ethanol into bio-ethylene based on industry data [71].

1.5 Outline

This dissertation presents four studies, each presented as a stand-alone article in chapter 2-5, followed by a discussion and recommendations chapter: chapter 6. In chapter 2-5, the references and labelling of sections, figures and tables have been adjusted to be consistent throughout the dissertation. The content has not been adjusted.

Chapter 2: Drivers and barriers for bio-based plastics in durable products.
Chapter 2 presents the drivers and barriers for bio-based plastic usage in durable products. This study addresses research question 1: *what challenges do actors throughout a durable product's value chain face when using bio-based plastics?* The drivers and barriers were derived from a workshop attended by 46 actors throughout the value chain of a telecommunications company. Although plastics only constitute a relatively small fraction of telecommunications products - which are largely electronics -, the vast majority of these products contain plastics for various functions (for example in electrical insulation and covers). Hence, plastics play an important role in telecommunications products for performance and aesthetics. Furthermore, the entire telecommunications value chain was represented in the study, from material manufacturing to end-of-life management.

Participants were split into three groups and asked to fill out an on-line collaborative whiteboard with drivers and barriers for using bio-based plastics in their durable products. Participants were also able to mark entries as important. In an analysis afterwards, the entries were coded into overarching drivers and barriers as perceived by the participants. The importance of the overarching drivers and barriers was established based on the amount of entries corresponding to them as well as their perceived importance. This exploratory study revealed which knowledge gaps were especially important for durable products.

Chapter 3: Bio-based plastics in a circular economy: review of recovery pathways and implications for product design.
Chapter 3 addresses research question 2: *how can product designers consider the recovery of bio-based plastics during the design process?* A rigorous literature review was conducted about recovery at end-of-life of bio-based plastics. An overview and terminology of 8 recovery pathways was established. For each recovery pathway, the suitability of commercially available bio-based plastics was studied based on existing literature. Combining the technical properties of the recovery pathways and their suitability for specific bio-based plastics allowed us to derive implications for product design.

Chapter 4: Bottlenecks in establishing the environmental impact of bio-based plastics: a case study for bio-based PE and bio-based PET.
Chapter 4 answers research question 3: *how does using a consistent methodology affect the LCA outcomes of bio-based plastics?* To study the effect of consistent methodology on the LCA outcomes of bio-based plastics, LCAs from literature were studied and the lifecycle inventories (i.e., the inputs and outputs of the processes) were compared. Where possible, the LCIs were used to establish a set of methodologically consistent LCAs. Reproducing LCAs from literature with a consistent methodology enabled a more in depth understanding

of the environmental impact of bio-based plastics and allowed us to pinpoint the most critical areas for data quality improvements.

Chapter 5: Sustainability of bio-based polyethylene: the influence of biomass sourcing and end-of-life.
In chapter 5, research question 4 is addressed: *under which biomass sourcing conditions does bio-based polyethylene result in a lower environmental impact than petrochemical-based equivalents?* The environmental impact of bio-based high-density polyethylene (HDPE) in 22 scenarios based on 5 different types of biomass in various locations was compared. The outcomes of the bio-based HDPE scenarios were also compared to petrochemical-based HDPE. Furthermore, we considered three end-of-life options for all the scenarios: mechanical recycling, incineration without energy recovery and incineration with energy recovery. Finally, in this study we also assessed the effect of biogenic carbon. Recently published guidelines dictate that biogenic carbon may not be accounted for upon bio-based plastic production, but can only be subtracted from emissions when this carbon is released back into the atmosphere upon incineration. To study the effect of this rule, we considered two cases for each scenario: one where biogenic carbon was accounted for during biomass growth and one where biogenic carbon was only accounted for upon incineration.

Chapter 6: Discussion and recommendations
In the sixth and final chapter of this dissertation, the main findings of the studies in this thesis are placed in the boarder context of a circular economy for plastics. The chapter also contains reflections on the position of bio-based plastics in the plastics industry and the methods used in this PhD. Furthermore, contributions to science and practice are presented, as well as recommendations for further research.

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2

Drivers and barriers for bio-based plastics in durable applications

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An additional overview of the employment fields of the participants of the study can be found in the supplementary information in table S1 [1]

Abstract

Bio-based plastics are gaining attention as a sustainable, circular alternative to the current, petrochemical-based plastics. The main application of bio-based plastics is in single-use packaging with short lifetimes. Extending the application of bio-based plastics products towards durable consumer products requires the involvement of different value chain actors. An online interactive workshop, with 46 participants representing the entire value chain, produced a list of drivers for using bio-based plastics in durable consumer goods and barriers to overcome. The primary barriers to using bio-based plastics in durable products were related to their underdeveloped value chain and a need for more knowledge. The underdeveloped value chain was associated with high costs and no infrastructure for recovery at end-of-life, reducing potential environmental benefits. Participants indicated that they did not expect the value chain to mature without substantial government stimulation. Participants also noted a lack of knowledge among value chain actors as well as end-users. Value chain actors expressed that they need more clarity about what bio-based plastics are available and how they can be used in a sustainable way. While the market demand for sustainable alternatives is growing and bio-based plastics are a valuable marketing tool, users are poorly informed, and marketing should be thoughtful to avoid greenwashing.

2.1 Introduction

Plastics are vital for modern life, but their environmental impact and damage caused by plastic pollution necessitate a new approach. Plastic production consumes up to 8% of fossil fuels extracted annually [2], while it is estimated that 79% of all plastic ever produced has accumulated in landfills and the natural environment [3]. Bio-based plastics have the potential to enable circularity since they are based (at least in part) on biomass, rather than finite petrochemical resources [4]. The renewable nature of bio-based plastics enables circularity at the plastic production level. While only accounting for 1% of all plastics produced in 2022, the market for bio-based plastics is growing at over three times the rate of that of petrochemical-based plastics [5]. The Circular Economy Action Plan contains plans to stimulate the bio-based sector [6].

Bio-based plastics can be divided into drop-ins and dedicated bio-based plastics [7]. Drop-in bio-based plastics are chemically identical to petrochemical-based plastics of the same name, such as polyethylene (PE). Dedicated bio-based plastics have no petrochemical-based equivalent. Biodegradable plastics are plastics that can be decomposed by living organisms and can be bio- or petrochemical-based. Not all bio-based plastics are biodegradable, although the two are often associated [2].

The main application of bio-based plastics is in single-use packaging with short lifetimes [5]. The application of plastics in single-use products will likely be limited by environmental legislation in the European Union (European Union, 2019) and other countries [8]. The application of bio-based plastics may then shift towards durable products. However, applying bio-based plastics in products with extended lifetimes requires the involvement of value chain actors unfamiliar with these materials.

This study aims to unveil how bio-based plastics are perceived by actors throughout the value chain for durable consumer goods: in this case, the telecommunication sector.

An interactive workshop produced a list of drivers for using bio-based plastics and barriers to overcome in order to extend the lifetime of bio-based plastic products from packaging towards durable consumer goods.

2.2 Methodology

In October 2020, 46 participants representing the entire telecommunications value chain attended an online workshop. Participants were approached through the network of a Dutch telecommunications company and that of the authors. Prior to the workshop, 39 participants filled out a survey about their role in their company and their experience with bio-based plastics. Table 2.1 contains an overview of the participants. Survey participants covered the entire value chain of telecommunications products, in addition to the fields of legislation and research. 26 out of 39 respondents were employed in a sustainability-related role.

Table 2.1: Overview of participants' role. Participants could select multiple answers.

Role	Number of responses
Design and/or development	10
Legislation	4
Management	10
Research	5
Sales and/or customer support	8
Sustainability	26
Other	1

During the workshop, the participants were given a brief introduction to bio-based plastics, followed by an interactive assignment. Participants were asked to fill out an online collaborative whiteboard with drivers and barriers to using bio-based plastics in durable products. Participants could place green dots on entries to mark them as important. After the workshop, all entries were anonymised, and those not phrased clearly were removed. The remaining entries were independently coded by two of the authors and grouped into drivers and barriers. These drivers and barriers were developed into themes that describe the participants' attitudes towards using bio-based plastics in their durable products. To determine the perceived importance of each driver or barrier, the number of post-its corresponding to them was combined with the number of green dots they received.

2.3 Results

2.3.1 Prior knowledge of the participants

Figure 2.1 displays the outcomes of the pre-workshop survey. The majority of respondents rated their knowledge about bio-based plastics as low to very low. Most also had little to no experience working with bio-based plastics. 8% of respondents were already producing products containing bio-based plastics, and 77% of respondents considered it likely to very likely that they would do so in the near future.

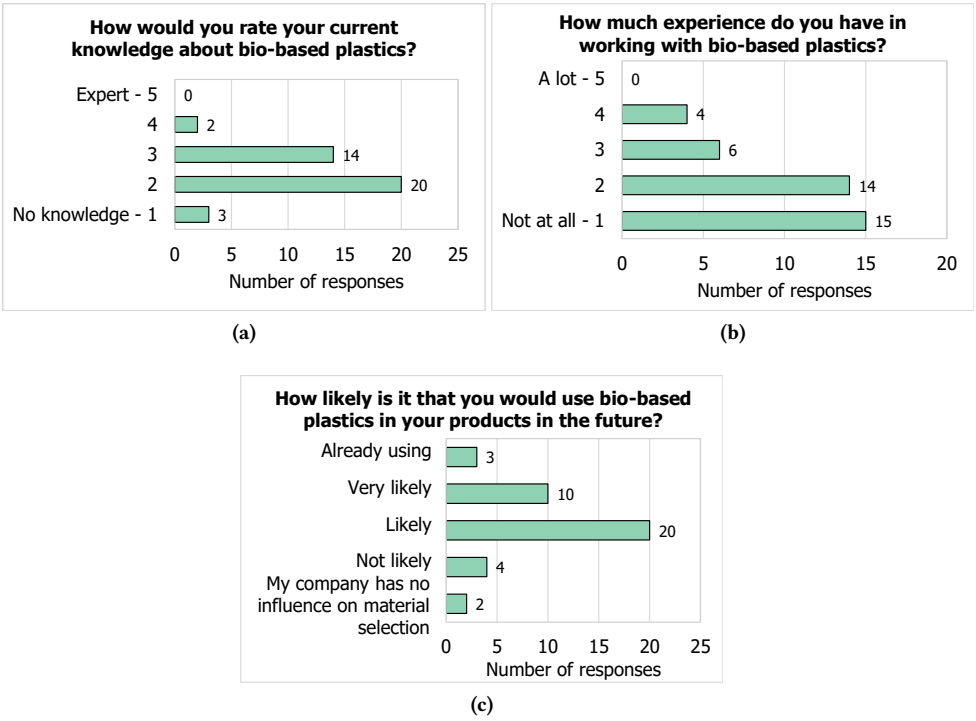

Figure 2.1: Outcomes of the pre-workshop survey about (a) prior knowledge of bio-based plastics, (b) prior experience with bio-based plastics, and (c) likeliness of using bio-based plastics in the near future.

2.3.2 Drivers and barriers for bio-based plastics usage

Drivers for bio-based plastics usage were categorised into the following seven themes: legislation, public perception, sustainability, design opportunities, sourcing, end-of-life, and collaboration. Below, the drivers for each theme are listed in order of perceived importance. It should be noted that the statements represent the participants' views and not necessarily the facts or the authors' views.

Driver theme 1: Legislation
- Existing and future regulations and sustainability targets could incentivise the use of bio-based plastics. For example, the European Green Deal, the Circular Economy Action plan, and CO_2 emission targets.

Driver theme 2: Public perception
- Bio-based plastics can be used as a marketing tool to engage customers who are becoming increasingly environmentally conscientious.
- Being an early adopter of bio-based plastics will reflect well on a company's image and establish them as a frontrunner.
- The interest in bio-based plastics in the corporate world is growing.
- Policymakers are driven by increased public awareness of environmental issues as well as business needs.

Driver theme 3: Sustainability
- Bio-based plastics can help companies to realise a circular business model.
- Bio-based plastics production can have a lower environmental impact than petrochemical-based plastics production.
- Bio-based plastics can be a sustainable solution for the long term due to their renewable resources.

Driver theme 4: Design opportunities
- Bio-based plastics can have new and unique properties that can be exploited in product design to add to performance and user value.
- Drop-in bio-based plastics can directly replace petrochemical-based counterparts, enabling a gradual transition.
- A new material creates the opportunity to experiment and develop new knowledge about its application.

Driver theme 5: Sourcing
- Bio-based plastics can be produced from a wide range of feedstocks, including waste, potentially resulting in a stable and local supply chain that is ultimately less dependent on fossil fuels.

Driver theme 6: End-of-Life
- Biodegradable (i.e. not per se bio-based) plastics can reduce waste and can be used to collect other compostable materials. For instance, biodegradable compost bags to collect home compost.
- Biodegradable plastics can provide a sustainable solution for products that wear or dissipate into the environment, such as tires or shoe soles.

Driver theme 7: Collaboration
- Being a new material, bio-based plastics allow for more interaction, knowledge sharing, and collaboration within value chains.
- Bio-based plastics can create new job opportunities.

Barriers to bio-based plastics usage could be categorised into the following seven themes: costs, lack of knowledge, sourcing, sustainability, end-of-life, an uncertain future and material properties. Below, the barriers for each theme are listed in order of perceived importance by the participants.

Barrier theme 1: Costs

- Bio-based plastics are more expensive than petrochemical-based plastics, increasing the price of a product.
- Users may not be able or willing to pay more.
- The entire value chain must change to accommodate bio-based plastics, which is expensive and time-consuming.

Barrier theme 2: Lack of knowledge

- Not all properties of new bio-based plastics are known. Bio-based plastics may have a lower technical performance than petrochemical-based plastics.
- Adding more variation in plastics adds complexity to proper disposal, making it confusing for end-users.
- It is risky to communicate bio-based with end-users because they do not have much knowledge about the concept, and the environmental benefits are still unclear.
- There are no clear guidelines on how to use bio-based plastics.
- Policy makers are not well informed about bio-based plastics.
- Bio-based plastics are not well known throughout the value chain. There is also insufficient information available.

Barrier theme 3: Sourcing

- Transitioning fully to bio-based plastics may not be possible without competing with food supply.
- The current volumes of available bio-based plastics are too low to cover demand and to enable recovery at end-of-life for dedicated bio-based plastics.
- Pollution from biomass may transfer into the plastic.

Barrier theme 4: Sustainability

- There are no standards for measuring and communicating the environmental impact of bio-based plastics and no policies regarding resource use, potentially leading to greenwashing.
- There is not enough clear information available about the environmental impact of bio-based plastics production and whether it is lower than petrochemical-based plastics.
- Marketing a product as more sustainable may cause end-users to adopt a less critical consumption attitude.
- Company image may suffer if bio-based plastics are derived from biomass that has damaging environmental effects.

Barrier theme 5: End-of-life

- Recovery of bio-based plastics at end-of-life is not yet guaranteed. Especially for dedicated bio-based plastics, production volumes are too small to facilitate reverse value chain infrastructure.
- The degradation levels of bio-based plastics compared to petrochemical-based plastics during recycling are unknown.
- Recyclability still needs to be guaranteed by product design.

Barrier theme 6: Uncertain future

- Certification of bio-based plastics can be complicated, taking years to develop.
- It is unclear how the market will develop, and governments are not taking an active role.
- There is a strong lobby of oil companies.
- Bio-based plastics are a rapidly developing field, which is difficult for companies.

Barrier theme 7: Material properties

- The aesthetics of bio-based plastics may be perceived as less desirable or of lower quality.
- Bio-based plastics properties may not meet material regulations such as fire safety or skin contact.
- Material composition and properties could vary depending on the source.

2.4 Discussion

Figure 2.2 presents an overview of the driver and barrier themes and illustrates the tensions between them. The observations are broadly in-line with pre-existing research. There is a tension between the positive public perception of bio-based plastics and their high costs. Bio-based plastics are more expensive than regular petrochemical-based plastics, which is often seen as a barrier [9–11]. The public perception of bio-based plastics is positive, and consumers state that they would pay an increased price for a bio-based product [12], but not everyone follows through on their stated willingness to pay more for a bio-based products [13, 14]. This value-action gap is a common phenomenon for more sustainable products. Despite their positive perception, the general public's knowledge about bio-based and biodegradable plastics is poorly developed [15]. Using bio-based plastics could therefore be risky, according to the participants. The use must be communicated clearly to the consumer in order to justify an increased cost. When bio-based plastics are applied in durable products, the bio-based aspect is typically mainly reflected in marketing [16]. However, the concept of bio-based plastics is complex, and the sustainability of the plastics is not entirely proven. This puts a company at risk of being accused of greenwashing.

The lack of public knowledge also extends to the recovery of bio-based plastics, combined with a lack of recovery infrastructure. Participants were concerned about proper disposal of bio-based or biodegradable products by end-users, and then by the reverse value chain. After use, drop-in bio-based plastics can easily integrate into existing recovery streams. However, these streams do not exist for novel, dedicated bio-based plastics, and there are no regulations or standards for their recovery at present [17]. Biodegradable plastics are not yet accepted in most industrial composting facilities [9, 11], and rarely fully

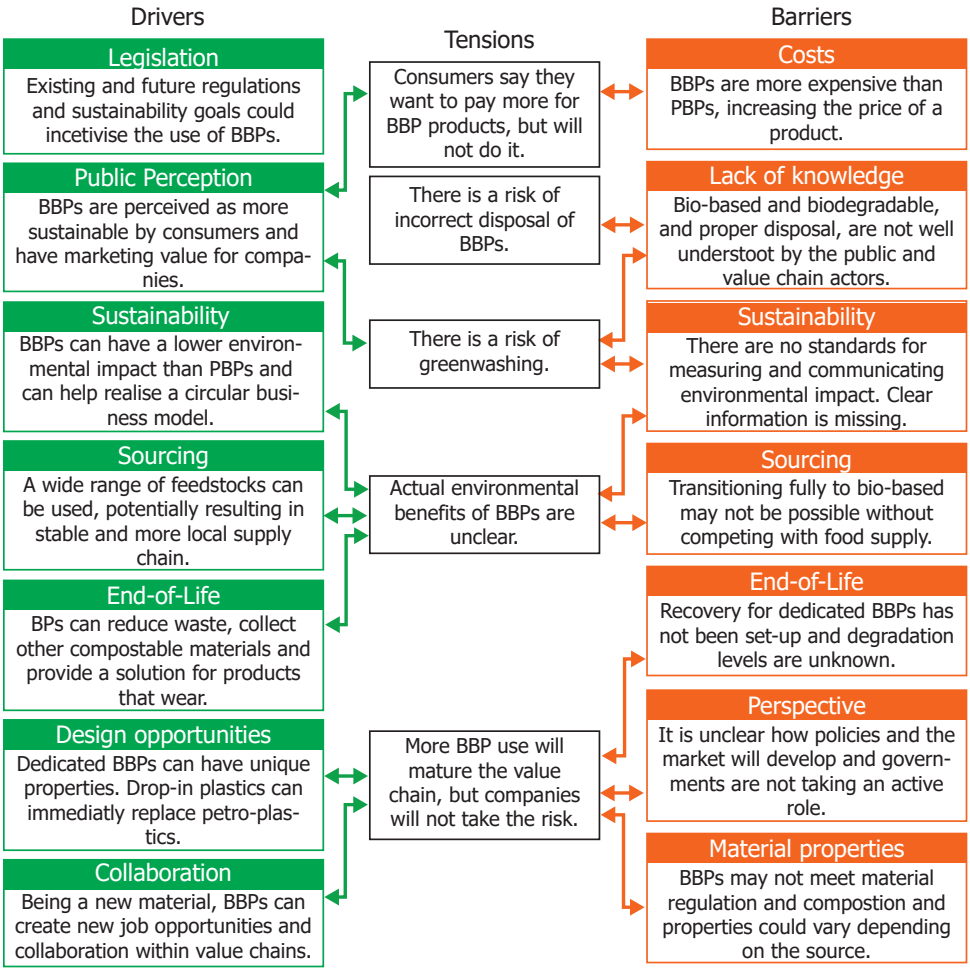

Figure 2.2: Overview of drivers and barriers for using bio-based plastics (BBP). Tensions between the drivers and barriers are highlighted in the middle column.

disintegrate in home compost or nature [2]. This creates the risk of doing more harm than good when using bio-based or biodegradable plastics.

Value chain actors themselves also lack knowledge about bio-based plastics. This already became apparent in the pre-workshop survey. Moreover, biodegradable plastics were often discussed during the workshop as if biodegradability is a property of bio-based plastics. However, biodegradable plastics are not necessarily bio-based, further highlighting the lack of knowledge and confusion. Furthermore, participants were not well informed about alternatives to the plastics used in their products. While bio-based packaging is already readily available, incorporating bio-based plastics in durable products requires the development of new knowledge.

Participants were divided on whether the environmental impact of bio-based plastics would be higher or lower than that of petrochemical-based plastics. Bio-based plastics are perceived to be more sustainable by many of the workshop participants as well as the general public [10], but this is not yet confirmed by lifecycle assessment [18, 19]. Exploiting the sustainable image of bio-based plastics in marketing while the actual environmental impact remains uncertain can lead to greenwashing [20–22].

Most barriers and tensions appeared to originate in the immature value chain of bio-based plastics, which was considered a major barrier. During the workshop, this was labelled as an apparent causality problem, more commonly known as a chicken or egg problem. The immature value chain makes bio-based plastics expensive and poorly understood, resulting in unclear environmental benefits. The value chain cannot develop if bio-based plastics are not used more widely, but it is also a barrier to more widespread usage.

2.5 Conclusions

Although knowledge about and experience with bio-based plastics was low for most participants, they expected that bio-based plastics would be used in their durable products in the near future. Workshop participants reported legislation and public demand for more sustainable products as the main drivers for using bio-based plastics in durable products in the telecommunications sector. Some existing legislation already incentivises the use of bio-based plastics, but participants expected future legislation to further promote bio-based. Bio-based plastics can be valuable in marketing and design, but the lack of knowledge and confusing terminology surrounding them require careful consideration in order to avoid greenwashing.

The circularity and sustainability of bio-based plastics were seen as a driver as well as a barrier. Bio-based plastics are perceived to be more sustainable, but the environmental benefits of bio-based plastics production and upscaling are still debated. Many bio-based plastics cannot be recovered at end-of-life as of yet. Notably, sustainability was not considered as important of a driver as legislation and public perception.

If bio-based plastics are to find widespread usage in durable consumer products rather than single-use packaging, their value chain needs to grow, and information is still missing. The bio-based plastics value chain will not mature by itself but requires government stimulation. Furthermore, bio-based plastic packaging options are readily available, but applying bio-based plastics in durable products requires the generation of new knowledge. There need to be more resources about what bio-based plastics are available and how they can be used in durable products. The sustainability of bio-based plastics needs to be further studied: the environmental impact and the effects of land-use change due to upscaling are not clear at present. Recovery at end-of-life also needs to be guaranteed.

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3

Bio-based plastics in a circular economy: a review of recovery pathways and implications for product design

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Abstract

Bio-based plastics are attracting increasing attention due to their perceived sustainability and circularity. While enabling circularity by using renewable feedstocks, they still contribute to plastic pollution. Furthermore, their rapidly growing market will cause bio-based plastics to constitute significant fractions of plastic waste, necessitating efficient recovery at end-of-life. Technical overviews of potential recovery pathways for bio-based plastics exist, although these have not yet been translated into product design recommendations. In this article, we assess the impact of material composition and product design on the feasibility of eight recovery pathways for bio-based plastics. The ability to recover a plastic not only depends on the plastic composition, but also on the way a product is designed. The alterations made to tailor plastics to be applied in products, and the product architecture, can enable or prohibit some recovery pathways. The outcomes highlight the importance of establishing a wider range of recovery pathways for plastics, and the crucial role of product design in enabling a circular economy for bio-based plastics. We also present a first guidance for product design to enhance the recovery of bio-based plastics.

3.1 Introduction

Plastics have become vital for modern life, owing to their low costs and wide range of properties. In recent years, environmental concerns regarding fossil-fuel consumption and pollution in the linear plastics economy have emerged. Plastic production consumes up to 8% of fossil-fuels extracted annually [2], and it is estimated that 79% of all plastic ever produced has accumulated in landfills and the natural environment [3], causing irreversible harm [4]. A transition to a circular economy has gained traction as a response to these challenges. A circular economy is restorative or regenerative by design, with the aim of eliminating waste [5].

Bio-based plastics are considered a key component of the circular economy [6, 7], since they are based on polymers produced (at least partially) from biomass [8]. The renewable nature of bio-based plastics enables circularity at the polymer production level. Nevertheless, using bio-based plastics does not solve the environmental issues of plastics. The environmental impact of bio-based plastic production and end-of-life is still a debated topic with little data available [9]. Furthermore, bio-based plastics can still contribute to plastic pollution, as the term 'bio-based' only refers to the sourcing of a polymer and not biodegradability in nature. Therefore, recovery at end-of-life will play a vital role in sustainability and circularity for bio-based plastics. In this article, we limit ourselves to the technical feasibility of bio-based plastics recovery.

In a circular economy, products need to be recovered at their highest possible value. As such, recovery strategies are categorised into a waste hierarchy. At the top of this hierarchy are recovery pathways that focus on product integrity (i.e., product-level recovery pathways), such as maintenance, repair, re-use, and remanufacturing [5]. Bio-based plastics do not perform differently from petrochemical-based plastics in these pathways [10]. Lower in the hierarchy are material-level recovery pathways, such as recycling. In material focused recovery pathways, the chemical composition of a plastic affects the feasibility and efficiency of recovery. Here, bio-based plastics may perform differently from petrochemical-based plastics. Material-level recovery includes molecular decomposition

pathways, where bio-based plastics occupy a special position. Since bio-based plastics are based on renewable resources, if the plastic is returned to simple molecules through processes such as incineration and biodegradation, they do not contribute fossil carbon dioxide (CO_2) to the atmosphere. Therefore, molecular decomposition of bio-based plastics can be considered a circular loop [11].

Bio-based plastics are commonly divided into two categories: drop-in and dedicated. Drop-in bio-based polymers are chemically identical to petrochemical-based polymers, whereas dedicated bio-based polymers do not have a petrochemical-based equivalent. Drop-in bio-based polymers can be integrated into existing recycling streams for petrochemical-based equivalents. Dedicated bio-based polymers [12] are currently considered a contaminant in plastic waste streams due to their small volumes [13, 14]. Although bio-based plastics only account for 1% of annual plastics production, their market is growing at twice the rate of petrochemical-based plastics [15]. Dedicated bio-based plastics make up roughly 60% of the bio-based plastics market today, and their share is expected to grow in the near future. New recovery systems need to be established when dedicated bio-based plastics grow into significant fractions of generic plastic waste. The development of the bio-based plastics market further necessitates research into end-of-life management.

Plastic recovery depends not only on recovery infrastructure, but also on product design. Product design covers the entire development process of a product or system to optimise function, value and appearance to benefit users and manufacturers [16]. One of the core principles of the circular economy is that the value of products and the materials they are made of can be preserved by keeping them in the economic system, either by lengthening the life of the products formed from them, or ''looping" them back in the system to be reused [17]. Decisions made during product design and development affect the ability to recover a product at end-of-life. For example, in material-level recovery, product design influences the ability to separate plastic parts from a product, which is important for material-level recovery.

The importance of product design in the transition to a circular economy with bio-based plastics has already been highlighted [10, 11, 14, 18–21]. There are technical overviews of possible recovery pathways for bio-based plastics [10, 14, 19, 20, 22–24], but, to our knowledge, these reviews have not yet been translated into recommendations for product design.

In this article, we assess how existing and future recovery pathways influence product design with bio-based plastics. An overview and terminology of 8 recovery pathways is established, and detailed descriptions of the state-of-the-art of recovery pathways for bio-based plastics are presented. This allows us to highlight the relevant technical characteristics of specific recovery pathways and assess their implications on material selection and product design.

3.2 Methodology

For product design, it is essential to know which recovery pathways are suitable for specific bio-based plastics and also how these recovery pathways can be influenced by product design. The methodology is divided into three steps, displayed in the three boxes in figure 3.1.

3.2.1 Establishing a framework for recovery pathways of bio-based plastics

A rigorous literature review was conducted in Scopus in May 2022 to map the existing recovery pathways for bio-based plastics. The search terms consisted of a combination of the following: (1) synonyms for recovery, namely "end-of-life", "recycling", and "recovery", (2) synonyms for bio-based plastics, including "bio-based polymer", "biopolymer", and "bio-based plastics", yielding 252 articles. Articles presenting an overview of recovery pathways for bio-based plastics were selected, resulting in 7 articles and reports. Snowballing yielded three additional articles. The recovery pathways discussed in these articles were categorised based on their reported definition and combined with ISO standards, resulting in a comprehensive set of 8 recovery pathways and definitions. Finally, a framework was created with these recovery pathways based on how their products feed back into the circular economy.

3.2.2 Scope

The scope of this research is limited to polymers that are either already produced commercially or expected to become commercially available in the near future [15]. This resulted in 13 polymers: high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyethylene terephthalate (PET), Polyamide (PA), polycarbonate (PC), Polypropylene (PP), Polyethylene Furanoate (PEF), Polybutylene succinate (PBS), Polylactic acid (PLA), polyhydroxyalkanoate (PHA) and thermoplastic starch (TPS), polyurethane (PU) and thermoplastic polyurethane (TPU). The recovery pathways were limited to material-level recovery pathways, excluding product-level recovery pathways.

3.2.3 State-of-the-art of recovery of bio-based polymers and plastics

In order to understand how specific recovery pathways are suitable for specific bio-based plastics, a rigorous literature review was conducted in Scopus in June 2022. Search terms were combinations of (1) the recovery pathways and their synonyms, and (2) "bio-based polymer", "bio-based plastic", or any of the 13 bio-based polymers established in section 2.2. Polymer blends and composites were excluded from the results. Commercial plastics (including additives such as stabilisers and colourants) were included. For drop-in bio-based polymers, the results of (chemically identical) petrochemical-based counterparts were also included. For product design, it is essential to know which recovery pathways are suitable for specific bio-based plastics and also how these recovery pathways are influenced by product design. Based on the discussed literature, the suitability of specific bio-based plastics for specific recovery pathways was established. Furthermore, specific technical aspects of recovery pathways were highlighted to define implications for product design.

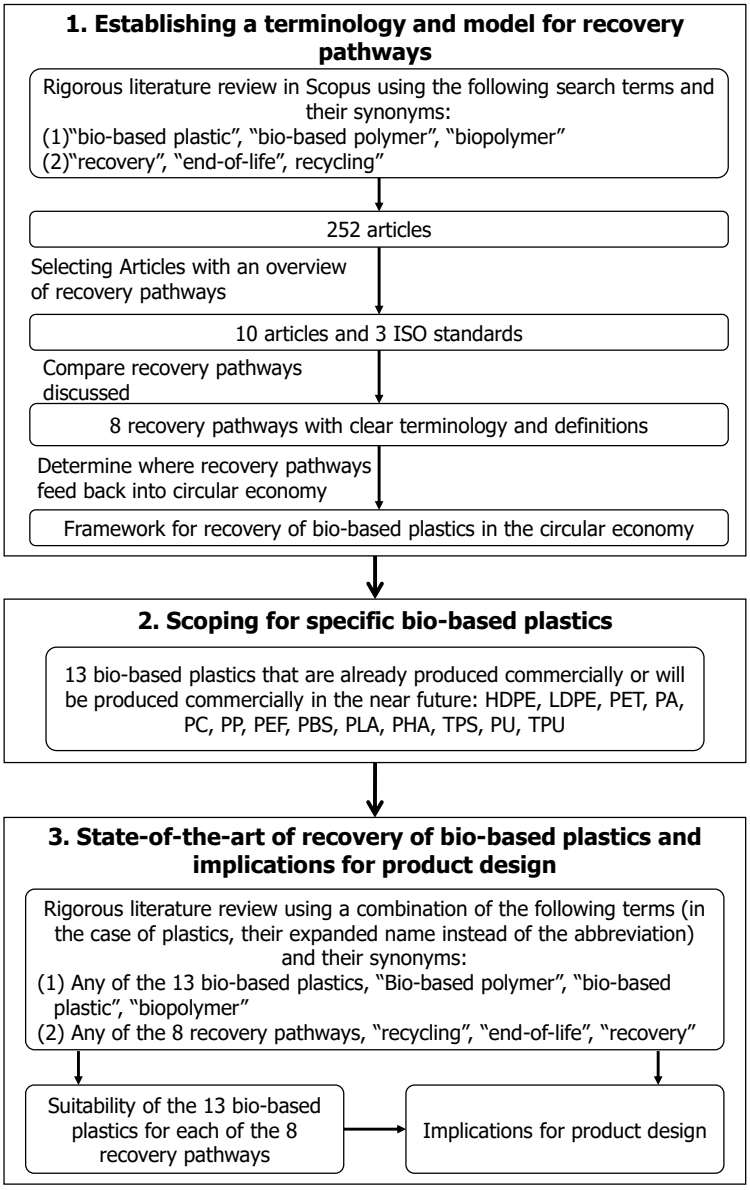

Figure 3.1: Visualisation of the methodology employed in this study.

3.3 Results

3.3.1 An overview of recovery pathways for bio-based plastics

Table 3.1 presents an overview of the terminology used for the recovery of bio-based plastics as gained from existing overviews and frameworks. Terminology in existing literature is often inconsistent for novel recovery pathways, such as chemical recycling and biodegradation. The terminology used throughout this article was established based on the terms used in existing overviews and active ISO standards, as displayed in table 3.1.

Figure 3.2 presents a framework of the circular economy for bio-based plastics incorporated into products, considering the re-entry points of the recovery pathways established in Table 1. Bio-based plastic product manufacturing was divided into six steps. Simple molecules (e.g. CO_2 and water) in the atmosphere are absorbed by plants and converted into biomass. Specific molecules, such as glucose, are isolated and used as feedstock to produce monomers for polymers. The polymer is often compounded with, for instance, additives or other polymers through blending to yield a plastic that is further manufactured into a product. After the use-phase of the product, the product lifetime is extended through product-level recovery. At the material level, recovery can occur through the eight established recovery pathways. The re-entry points of recovery pathways are based on their products. For instance, by definition, the main products of aerobic digestion of polymers are CO_2 and H_2O, which are categorised as simple molecules.

Anaerobic digestion of polymers, by definition, yields large amounts of methane, which can be captured as a feedstock for the production of new plastics in an industrial environment. If anaerobic digestion of bio-based polymers occurs in nature, the product could be considered a “simple molecule”; however, plants do not absorb significant amounts of methane during their growth [25], and therefore, this cannot be considered a circular loop.

Table 3.2 summarises the result of the literature review and provides a first indication of recovery pathways that may be considered when using a specific bio-based polymer in a product. The selected literature has been categorised into the specific recovery pathway and bio-based polymer each article presents. Details can be found in the supplementary information (Table S1-S6 [1]). Recovery pathways for polymers that currently represent a large part of the (petrochemical-based) plastic waste composition, such as bio-HDPE, bio-LDPE, bio-PET and bio-PP, have already been extensively studied. Furthermore, the recovery of commodity-grade dedicated bio-based polymers is well understood, with the exception of PEF.

However, for bio-based polymers classified as engineering-grade (bio-PC, bio-PA, bio-PU and bio-TPU), the opportunities and effects of recovery have not yet been studied. Recovery of these polymers through various technologies is theoretically possible, but the rigorous literature review did not yield any articles in which the possibilities have been reported. It should be noted that although the engineering-grade bio-based polymers presented here share a name with petrochemical-based polymers, they are not chemically identical [26].

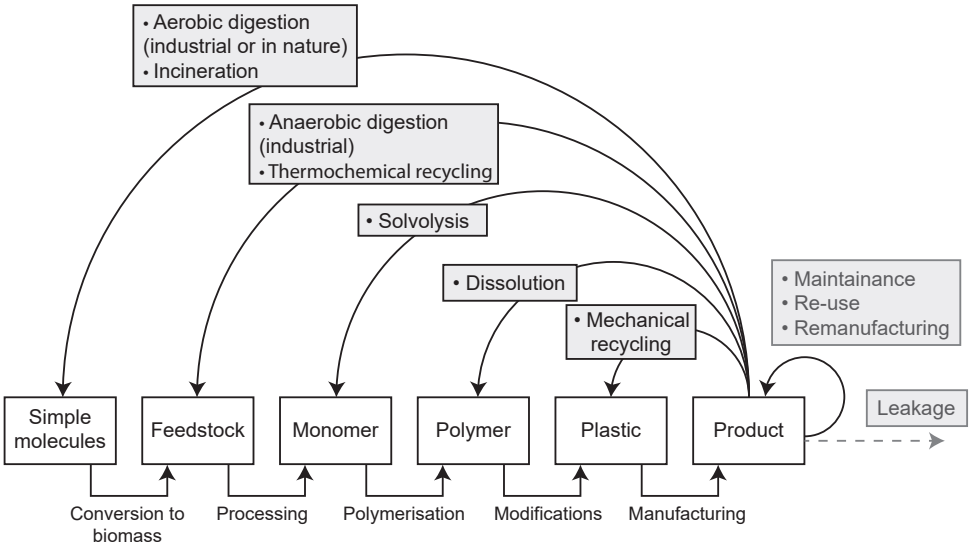

Figure 3.2: Framework for the circular economy of bio-based plastics incorporated into products.

Table 3.1: Terminology for recovery pathways used in this article.

Selected terminology for specific recovery pathways	**Definition**	**Alternative terms**	**Umbrella terms covering multiple recovery pathways**
Mechanical recycling [7, 10, 19, 20, 22–24, 27–29]	"*Processing of plastic waste into secondary raw material or products without significantly changing the chemical structure and composition of the material*" [29].	Recycling [20]	
Dissolution [14]	The recovery of a polymer through its dissolution and precipitation in a suitable solvent/non-solvent, without any alteration to its molecular structure.	Solvent-based purification [7, 28]	Chemical recycling [7, 10, 11, 14, 20, 22, 23, 24, 27, 29, 30] Feedstock recycling [10, 14]
Solvolysis [14, 23]	The cleavage of a polymer by a solvent such as water or alcohol, often in the presence of a catalyst.	Chemolysis [14] Feedstock recycling [10] Depolymerisation [7, 28]	
Thermochemical recycling	The dissociation of polymers through high temperatures.	Feedstock recycling [7, 11, 28] Thermolysis [10] Plastic-to-fuel recycling [11]	
Anaerobic digestion [7, 10, 11, 14, 24, 27, 29]	"*The breakdown of an organic compound by microorganisms in the absence of oxygen to carbon dioxide, methane, water and mineral salts of any other elements present (mineralisation) plus new biomass*" [31].	Anaerobic respiration [10] Anaerobic biodegradation [10, 22]	Biodegradation [7, 10, 20] Composting [20, 22] Organic recycling [14] Microbial degradation [19]
Aerobic digestion [11]	"*The breakdown of an organic compound by microorganisms in the presence of oxygen to carbon dioxide, methane, water and mineral salts of any other element present (mineralisation) plus new biomass*" [32].	Aerobic respiration [10] Aerobic biodegradation [10, 14, 22] Aerobic composting [7, 30] Composting [24, 28]	
Incineration [7, 10, 11, 14, 20, 22, 27, 29]	The combustion of the plastic into mainly carbon dioxide, water and ash.	Energy recovery [20, 23, 30, 33]	

Table 3.2: Summary of possible recovery pathways for commodity-grade, drop-in, dedicated, and engineering-grade, bio-based polymers. Green: research proving that this is a viable option could be found. Yellow: existing research suggests that using this recovery pathway for this specific polymer yields poor results. In the case of aerobic and anaerobic digestion, this means that digestion does not occur in a timeframe that corresponds with current industry practice, or that may cause harm to the natural environment In the case of mechanical recycling, it means a rapid decline in properties. In the case of thermochemical recycling, the products of the process were only suitable as energy resources and not for the production of new polymers. Red: theoretically impossible based on the characteristics of the polymer combined with the targeted recovery pathway. For example, solvolysis of HDPE is theoretically impossible due to the chemical structure of PE. Grey: theoretically possible, but no studies could be found. For example, based on the chemical structure, bio-PA can undergo mechanical recycling, but this has not been demonstrated yet.

Commodity grade, drop-in bio-based polymers					
	Bio-PET	**bio-HDPE**	**Bio-LDPE**	**Bio-PP**	**Bio-PBS**
Mechanical recycling	Viable option [34, 35]	Viable option [36–40]	Viable option [38, 41–43]	Viable option [38, 44–46]	Viable option [36, 47]
Dissolution	Viable option [48, 49]	Viable option [48, 50–53]	Viable option [48, 50–52, 54]	Viable option [48, 50, 52, 55–57]	Theoretically possible, but not researched yet
Solvolysis	Viable option [58–69]	Theoretically impossible	Theoretically impossible	Theoretically impossible	Theoretically possible, but not researched yet
Thermochemical recycling	Viable option [70, 71]	Viable option [50, 71–75]			Theoretically possible, but not researched yet
Anaerobic digestion	Theoretically impossible	Theoretically impossible	Theoretically impossible	Theoretically impossible	Poor outcome [76–78]
Aerobic digestion (industrial)	Theoretically impossible	Theoretically impossible	Theoretically impossible	Theoretically impossible	Poor outcome [79–81]
Aerobic digestion (nature)	Theoretically impossible	Theoretically impossible	Theoretically impossible	Theoretically impossible	Poor outcome [79, 80, 82]
Commodity grade, dedicated bio-based polymers					
	PLA		**PHA**	**TPS**	**PEF**
Mechanical recycling	Poor outcome [83–88]		Poor outcome [89]	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet
Dissolution	Theoretically possible, but not researched yet		Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet

Table 3.2: Continued

Solvolysis	Viable option [64, 68, 90–100]	Viable option [64, 101–104]	Theoretically possible, but not researched yet	Viable option [105–108]
Thermochemical recycling	Poor outcome [109]	Poor outcome [110]	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet
Anaerobic digestion	Poor outcome [78, 111–114]	Poor outcome [77, 78, 115]	Poor outcome [116, 117]	Theoretically possible, but not researched yet
Aerobic digestion (industrial)	Poor outcome [111, 118–123]	Poor outcome [124, 125]	Poor outcome [117, 126]	Theoretically possible, but not researched yet
Aerobic digestion (nature)	Poor outcome [2, 127–130]	Poor outcome [131–136]	Poor outcome [126]	Theoretically possible, but not researched yet
Engineering grade bio-based polymers				
	Bio-PA	**Bio-PC**	**Bio-PU**	**Bio-TPU**
Mechanical recycling	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically impossible	Theoretically possible, but not researched yet
Dissolution	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically impossible	Theoretically impossible
Solvolysis	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet
Thermochemical recycling	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet
Anaerobic digestion	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically impossible	Theoretically impossible
Aerobic digestion (industrial)	Theoretically possible, but not researched yet	Theoretically possible, but not researched yet	Theoretically impossible	Theoretically impossible
Aerobic digestion (nature)	Theoretically impossible	Theoretically impossible	Theoretically impossible	Theoretically impossible

3.3.2 State-of-the-art of recovery pathways for bio-based plastics

Mechanical recycling

Mechanical recycling is the "*processing of plastic waste into secondary raw material or products without significantly changing the chemical structure of the material*" [29]. In order to be suitable for mechanical recycling, a plastic must be melt-processable, i.e. a thermoplastic, and withstand the conditions under which mechanical recycling occurs. Mechanical recycling generally consists of the following steps: sorting, shredding, washing and drying, and reprocessing [137]. During sorting, different types of plastic are separated, and impurities are removed. Next, the plastic is washed and dried before reprocessing, where granulate or new products are produced from recycled plastics using conventional melt-processing techniques. Thermomechanical stresses during reprocessing can change the molecular structure of a polymer through chain scission, oxidation or a reaction with contaminants, additives or dissociated pieces of the polymer itself. These changes can result in a recycled plastic with different properties than virgin plastic, which cannot directly replace virgin plastics [11, 138]. Contaminants and impurities cause degradation in plastic processing during reprocessing, and thus sorting accuracy affects the quality of recycled plastics. Currently, the most used separation techniques are air and float-sink separation, which are inaccurate when different plastics have a similar density; other techniques, such as near-infrared (NIR) sorting, can improve sorting accuracy in the future [139].

Table 3.3 summarises the effects of mechanical recycling, specifically reprocessing, on drop-in and dedicated bio-based polymers. Some drop-in bio-based plastics, namely bio-HDPE, bio-LDPE, bio-PET and bio-PP, are known to be good candidates for mechanical recycling due to extensive research into their petrochemical-based counterparts. Since bio-HDPE, bio-LDPE, bio-Pet and bio-PP are chemically identical to their petrochemical-based counterparts, the results of this research also apply to the bio-based versions. Mechanical recycling of PLA has been investigated extensively and is applied on a small scale through at-home recyclers for 3D printed parts [87]. The thermal properties and processing window of recycled PLA do not differ from its virgin form, but barrier, tensile and impact properties degrade rapidly upon reprocessing [84, 88]. PHAs show a sharp decline in many properties after mechanical recycling [89]. (Bio-)PBS withstands mechanical recycling without a significant change in the molecular structure [47] in bending strength or bending modulus [36], although the effect on impact properties has not yet been reported. The degree of degradation due to mechanical recycling is affected by the processing conditions. The processing conditions used to study the effects of mechanical recycling on a plastic in the studies in table 3.3 varied, which also affected the outcomes. A detailed overview of these results and the processing conditions can be found in the supplementary information (Table S1 [1]).

Table 3.3: Summary of mechanical recycling of bio-based polymers (without additives).

	Polymer	Thermal and processing properties	Mechanical properties	Impact properties	References
Drop-in	**Bio-HDPE**	Reduced melt flow index after 5 cycles	Yield stress, tensile stress and elastic modulus remain constant for 10 cycles, then decrease	Slight increase in impact strength after one cycle	[36–40]
	Bio-LDPE	Unchanged until 40 cycles			[38, 41–43]
	Bio-PP	Reduced melt viscosity after 4 cycles	Strength and stiffness increase; strain at break decreases after 4 cycles	Impact strength remains unchanged for up to 6 cycles, then decreases	[38, 43–46]
	Bio-PET	Reduced melt viscosity after one cycle	Yield stress, tensile stress and elastic modulus reduced after one cycle	Impact strength reduced after one cycle	[34, 35]
	Bio-PBS	Reduced melt flow index after one cycle	Unknown	Unknown	[36, 47]
Dedicated	**PLA**	Unchanged after one cycle	Young's modulus and hardness decreased after one cycle	Impact strength decreased after one cycle	[83–88]
	PHA	Viscosity reduced after one cycle	Tensile strength, tensile modulus remain unchanged until 6 cycles	Impact strength unchanged until 6 cycles	[89]

Dissolution

Dissolution is the recovery of a polymer by dissolving it in a solvent, followed by precipitation in a non-solvent, without any alteration to its molecular structure. Dissolution is applied in numerous chemical processes, such as coatings, and its molecular transport phenomena have been studied extensively [140]. During dissolution, any additives or contaminants in a plastic can be removed, achieving high recovery rates with homogeneous products while being able to selectively recover specific polymers from mixed plastic waste [51, 52]. However, dissolution requires large amounts of solvents and non-solvents, which are currently not bio-based. The solvent-to-polymer volume ratio is usually above

7:1, while solvent recovery is never 100%. Dissolution is also energy intensive as it often requires temperatures above 100 °C.

Dissolution for polymer recovery is currently only applied at a pilot scale for polystyrene [141], and no research has been reported for bio-based polymers. Nevertheless, the dissolution of commodity polymers has been studied with good results. These results can be extended to chemically-identical drop-ins: specifically bio-LDPE, bio-HDPE, bio-PET and bio-PP [48, 51, 52]. A detailed overview of articles reporting the dissolution of polymers for which bio-based drop-ins exist can be found in the supplementary information (Table S2 [1]).

Solvolysis

Solvolysis is the cleavage of a polymer by a solvent, often in the presence of a catalyst. Polymers containing ether, ester and amine bonds (synthesised through condensation polymerisation) can be recovered through solvolysis [11]. In contrast to dissolution and mechanical recycling, solvolysis can also recover thermoset polymers. Solvolysis processes are subdivided based on the solvent used, i.e. hydrolysis for water or alcoholysis for alcohols. The products of solvolysis may be fuels, useful chemicals or molecules that can be used directly in the production of new polymers. In most cases, solvolysis products require extra conversion steps to be useful for polymers. Additives can be filtered out. Polymer-to-solvent ratios during solvolysis are typically low, requiring elevated temperatures and catalysts.

In theory, (bio-based) polymers and plastics suitable for solvolysis include bio-PET, PLA, PHA, TPS, bio-PBS, bio-PA and PEF. Solvolysis of (bio-)PET has been demonstrated through hydrolysis [61], glycolysis [59, 68], aminolysis [60] and alcoholysis [67]. Solvolysis of PLA can occur through alcoholysis [91, 93, 142], hydrolysis [99] or alcoholised. Hydrolysis [104] and alcoholysis [102, 103] of PHA have been demonstrated to yield valuable chemicals.

Enzymatic depolymerisation is a specific type of solvolysis that uses enzymes as biological catalysts. Enzymatic depolymerisation occurs at temperatures between 50 °C and 75 °C, which is lower than those used with synthetic catalysts. However, the polymer chains need to have a certain mobility at the process temperature. For example, consumer-grade PET has a high crystallinity that reduces polymer mobility, making it difficult to degrade by enzymatic depolymerisation [65, 66]. Enzymatic depolymerisation of polymers into monomers has been successfully demonstrated for PLA [92], PEF [106, 107] and PET [66, 69]. Depending on the enzymes present and the reaction conditions, enzymatic depolymerisation can also yield chemical intermediates that can be processed into monomers or used in other applications [58, 62, 65]. A detailed overview of different solvolysis processes, including enzymatic depolymerisation and its products for bio-based polymers be found in the supplementary information (table S3 [1]).

Thermochemical recycling

Thermochemical recycling is the dissociation of polymers through high temperatures. Polymers are dissociated into gases (e.g. CO_2, methane), liquids (longer hydrocarbons) and solids (tar). Thermochemical recycling processes are differentiated by process temperature, pressure and atmosphere [143]. The most common thermochemical recycling processes are listed below.

- Pyrolysis occurs in an inert atmosphere at around 500 °C and 1-2 standard atmosphere (atm), with or without a catalyst, resulting in either depolymerisation or random fragmentation, depending on the plastic composition and other materials present. The products of pyrolysis can be gases, liquids, solids or any mixture of these [71, 144].
- Gasification occurs in an atmosphere of air or pure oxygen at 700 – 1200 °C at standard pressure. Plastics break down into so-called syngas (consisting primarily of nitrogen, carbon monoxide, hydrogen and CO_2) [144].
- Hydrogenation occurs in the presence of hydrogen (H_2) and a catalyst at 350 – 400 °C at roughly 70 atm. The plastic is liquefied during the process and can be filtered to yield naphtha or oil. The presence of hydrogen improves the quality of the resulting feedstock [144].

The decomposition behaviour of plastics in thermochemical recycling is complex, yielding a wide range of compounds [144]. Plastics can be studied in isolation, but thermochemical recycling usually uses mixtures, influencing the reactions that occur. For example, pyrolysis of PP in isolation at 760 °C produces benzene [71], but under different conditions, no benzene is produced [72, 145]. Products of thermochemical recycling are primarily used as fuels due to the wide and poorly defined range of feedstocks produced [144]. Specific chemicals could be removed from the mixture and used to produce new polymers (either directly or after conversion) [146].

Anaerobic digestion

Anaerobic digestion is "*the breakdown of an organic compound by microorganisms in the absence of oxygen to carbon dioxide, methane, water and mineral salts of any other elements present (mineralisation) plus new biomass*" [31]. Anaerobic digestion can occur in industrial composting facilities and landfills (with methane capturing), but also in uncontrolled environments, such as underground or in the bottom layers of a home compost bin [147]. Industrial anaerobic digestion focuses on producing biogas (methane) as an energy source, which can also be a precursor for polymers [148]. Another product of anaerobic digestion is the so-called digestate consisting of residual materials. This digestate is often used as a fertiliser [149]. However, synthetic biodegradable polymers degrade fully into methane, water and other gases and do therefore not add to this fertiliser. When anaerobic digestion occurs in natural environments, the highly potent greenhouse gas methane leaks into the atmosphere [150].

Before industrial anaerobic digestion, physical contaminants (e.g. glass or metals) are removed from bio-waste and its composition is optimised, commonly by adding carbon-rich waste, since bio-waste is often too high in nitrogen [151]. Since biodegradable plastics are relatively high in carbon, they could be used to optimise the waste composition. To be compatible with industrial anerobic digestion, the degradation time for plastics under typical industrial anaerobic digestion conditions needs to be similar to that of the bio-waste. A typical industrial anerobic digestion process occurs at 30-60 °C for up to 60 days [149, 152]. For example, PLA can degrade up to 95% in 40 days at 55 °C [113] but only reached 20% biodegradation after 65 days at 37 °C[114]. PHA degrades fully within 42 days at 37 °C [115]. TPS degraded by 23% after 28 days at 35 °C [116]. This implies that bio-based plastics

are only candidates for anaerobic digestion if additional heat is applied. An overview of experiments with biodegradable plastics under anaerobic conditions is provided in the supplementary information (table S4 [1]).

Aerobic digestion

Aerobic digestion is "*the breakdown of an organic compound by microorganisms in the presence of oxygen to carbon dioxide, methane, water and mineral salts of any other element present (mineralisation) plus new biomass*" [33]. Composting facilities utilise industrial aerobic digestion to produce compost from organic waste [14]. Most polymers degrade into CO_2 and water under aerobic digestion conditions and do not contribute significant mass to the compost [2, 153]. Similar to anaerobic digestion, aerobic digestion occurs at an optimal ratio of carbon and nitrogen atoms to which plastics can contribute [14]. Aerobic digestion also occurs in home composting bins and in the top layers of soil and agricultural fields [154].

Bio-based plastics should be compatible with the industrial composting cycle, which ISO standards describe as taking up to 56 days at temperatures above 50 °C [155]. table 3.4 contains a summary of experiments on aerobic digestion of pure polymers of different shapes under specified composting conditions, demonstrating the differences in the degree of degradation due to different product thicknesses and composting conditions. This highlights the importance of understanding the degradation of not just the material, but also the product in the targeted composting conditions. A table with further details of aerobic digestion experiments, including blends and additives, can be found in table S5 in the supplementary information [1]. If biodegradation of a plastic is not completed during industrial composting, the partially decomposed plastic will end up as fragments in the compost that is often used as a fertiliser. Aerobic digestion conditions in nature are different from industrial composting, and the plastic fragments may not fully decompose [156]. This may result in the introduction of more micro- and nanoplastics formation but the effects of this are still unknown [2].

Aerobic digestion in nature could be an attractive recovery pathway for plastic products, as it can prevent plastic pollution. Understanding biodegradation in nature remains challenging, with little research in realistic conditions. Moreover, soil microbiomes vary per region and soil type, influencing the effectiveness of biodegradation [131]. (Bio-)PBS only degrades slowly (<5% weight loss in 80 days) in natural soil and requires additives to enhance aerobic digestion under natural conditions [79]. PLA does not biodegrade under natural conditions [2]. Biodegradation of PHAs depends mainly on the molecular structure of the type of PHA and the soil microbiome [132]. PHAs are often blended with other polymers in order to enhance their properties, but how this affects biodegradation is not yet understood [132]. TPS showed aerobic digestion under simulated natural conditions but showed little deterioration in actual natural conditions [126]. A more detailed overview of these experiments can be found in table S6 in the supplementary information (Table S6 [1]).

Table 3.4: Summary of industrial aerobic digestion experiments with pure polymers.

Polymer	Inoculum type, weight ratio plastic:inoculum	Product/shape	Time (days)	Temperature (°C)	Degradation degree	Reference
PLA	Compost, unknown ratio	Pellets	60	58 °C	34% – 45%	[118]
		Film			35% – 100%	
	Municipal organic solid waste, unknown ratio	Bottle	58	65 °C	78% – 84%	[119]
	Agricultural and tree waste, unknown ratio	Powder (0.5 mm)	45	58 °C	55% – 75%	[120]
PLLA	Vegetable waste, 1:500	Film	20	70 °C	99%	[111]
		Fabric	40		73%	
	Inoculated compost, 1:500	Powder (0.5 mm)	90	58 °C	90%	[122]
PHA	Synthetic compost, unknown ratio	Film	39	58 °C	100%	[124]
	Mature compost, unknown ratio	Film	110	58 °C	80% – 91%	[125]
PBS	Mature compost, 1:6	Pellets (3 mm)	90	58 °C	14.1%	[81]
		Powder (0.042 mm)			71.9%	
		Film			71.9%	
TPS	Municipal biowaste, 1:100	Carrier bags	72	60 °C	100%	[117]

Incineration

Incineration, or energy recovery, is the "production of useful energy through direct and controlled combustion" [29]. During incineration, the plastic is returned to simple molecules and bottom ash. Because bio-based plastics contain carbon originating from CO_2 from the atmosphere, their incineration could be considered carbon neutral [11]. The amount of energy released during the incineration of specific polymers can be estimated according to their chemical structure [157]. Incineration is currently the most viable recovery pathway for bio-based plastics without dedicated other dedicated recovery pathways. Incineration of plastics is accepted as a total conversion of plastic waste [3]. However, the incineration of plastics can produce toxic by-products [158]. Furthermore, microplastics have been found in bottom ash from municipal waste incinerators: 1.9-565 pieces of microplastic per kilogram of bottom ash [159].

3.4 Discussion - defining implications for product design

We presented a framework for the material-level recovery of bio-based plastics in a circular economy. This demonstrated that bio-based polymers operate differently in the circular economy compared to petrochemical-based polymers when material-level recovery is considered. The circular economy typically distinguishes between a biocycle and a technocycle, where recovery of petrochemical-based plastics occurs only in the technocycle. For petrochemical-based plastics, recovery pathways like incineration and aerobic digestion cannot be considered circular loops, as they introduce fossil greenhouse gas emissions to the atmosphere. Bio-based plastics, on the other hand, can also flow through the biocycle because they are produced from biomass [11].

Section 3.3 has provided an overview of the state-of-the-art of different recovery pathways for bio-based plastics from a technical perspective, indicating which recovery pathways can be considered for specific bio-based polymers. Merely choosing a suitable polymer and recovery pathway is insufficient to guarantee efficient recovery. Additives and blending need to be considered, as well as how a product is constructed and manufactured since these aspects can make a product unsuitable for specific recovery pathways. This implies that the recovery of bio-based plastics has implications for product design. In this section, we describe some of these implications based on the technical characteristics of recovery pathways. The product design implications are summarised in table 3.5. Figure 3.3 displays how these product design implications may be applied in the design process. Note: in a CE, higher-value recovery options are preferred - figure 3.3 and table 3.5 focus solely on material-level recovery.

In the hierarchy of material-level recovery pathways, mechanical recycling is usually at the top [14]. However, mechanical recycling is not always preferred or possible as it may result in reduced properties. [160] developed some design for mechanical recycling recommendations, focussing on material composition and ease of separation of plastic components in the product. Since its publication in 1995, separation and sorting technologies have improved, and they will become more accurate [139]. This renders some recommendations obsolete, for example, avoiding different plastics in a single product or not using

certain additives that change the density of the plastic. Multi-material manufacturing, where different plastics are fused and cannot be mechanically separated, should still be avoided when designing for mechanical recycling. Furthermore, plastics with different molecular weights and additives are not separated. Therefore, additives that reduce the value of recycled plastics should be avoided, such as colourants and additives that induce molecular damage during reprocessing. Blends of different polymers are typically not sorted and should also be avoided.

Dissolution and solvolysis occur at the surface of a plastic, and their reaction rate is affected by the surface area to volume ratio [140]. Both dissolution and solvolysis enable the separation of additives and blends [51, 52], implying that products can contain additives and blends without compromising the value of recovered polymers and monomers: an advantage over mechanical recycling. Thermochemical recycling can process essentially any plastic, but thermochemical recycling processes are sensitive to the input composition [144]. The entire plastic waste composition will likely not be affected significantly by a single product range, but plastics can yield harmful products in combination with the current plastic waste composition.

Biodegradation processes (aerobic and anaerobic) occur at the surface of a plastic, and the surface-to-volume ratio should be optimised to increase the reaction rate [118]. Any components not fully degraded during industrial biodegradation may be used as fertiliser, and the product used should either fully degrade within the industrial process or be able to degrade in soil. This also applies to any contaminants from the use-phase. In aerobic conditions, polymers biodegrade into CO_2 and H_2O, not adding mass or nutrients to compost [2, 153] and any functional value in the plastic is completely lost. Therefore, aerobic biodegradation should primarily be considered as a recovery pathway to prevent plastic waste; if the product is bound to end up in nature and if the plastic is shown to disintegrate completely. In uncontrolled anaerobic conditions, polymers produce methane, which can be released into the atmosphere, where it is a potent greenhouse gas [150]. Anaerobic biodegradation should therefore be avoided in natural environments.

Table 3.5: Identification of product design implications when targeting specific recovery pathways, based on plastic suitability and technical properties.

Recovery pathway	Technical characteristic	Product design implication(s)
Mechanical recycling	Recycled plastic becomes a mix of the input plastics. Additives, blends and different molecular weights are not separated.	Avoid or minimise using additives that reduce the value of recycled plastics.
		Avoid blending different polymers unless a closed-loop system can be set-up for a product to recycle the blend directly.
		Ensure that different plastics in a product can be mechanically separated, for instance, during shredding. Products should not be produced using multi-material manufacturing, such as 2K or 3K, and products should not have coatings.
	Sorting effectivity determines the quality of recycled plastics.	Avoid using additives that hinder sorting.
		Avoid mechanical recycling for products that can easily be contaminated by substances that cannot be removed in the targeted mechanical recycling process.
Dissolution and Solvolysis	Dissolution and solvolysis occur at the surface of a plastic.	Optimise surface area to volume ratio.
	Dissolution and solvolysis can sort out additives and separate different molecular weights and blends.	Allows for the incorporation of additives and different molecular weights and blends.
Solvolysis (enzymatic depolymerisation)	In the case of enzymatic depolymerisation: the process occurs at a lower temperature (below 60 °C).	Ensure solvolysis can occur at moderate temperatures.
Thermochemical recycling	A change in waste composition due to a growing market share of bio-based plastics may affect the feedstock produced from thermochemical recycling.	Consider the current plastic waste composition and if introducing the targeted plastic will not yield harmful products.

Table 3.5 continued.

Recovery pathway	Technical characteristic	Product design implication(s)
Industrial biodegradation (anaerobic digestion and aerobic digestion)	Anaerobic and aerobic digestion occur at the surface of a plastic.	Optimise for surface area to volume ratio.
	After the industrial process, anything that does not degrade may be used as fertiliser, eventually ending up in nature.	Ensure all components (such as the polymer and additives) will degrade fully during industrial biodegradation. If they do not, they should degrade aerobically in natural conditions.
		Avoid for products that get contaminated with non-biodegradable substances that are harmful to nature.
Biodegradation in nature (only aerobic)	Any value of the plastic is completely lost as it returns to CO_2 and H_2O while no energy is recovered.	Use for products that inevitably end up in nature (e.g. products that wear such as shoe soles or agricultural films).
	Anaerobic digestion in uncontrolled environments is highly undesirable as the methane produced will leak into the atmosphere.	Avoid anaerobic digestion for products that will end up in anaerobic environments, such as the bottoms of compost bins.

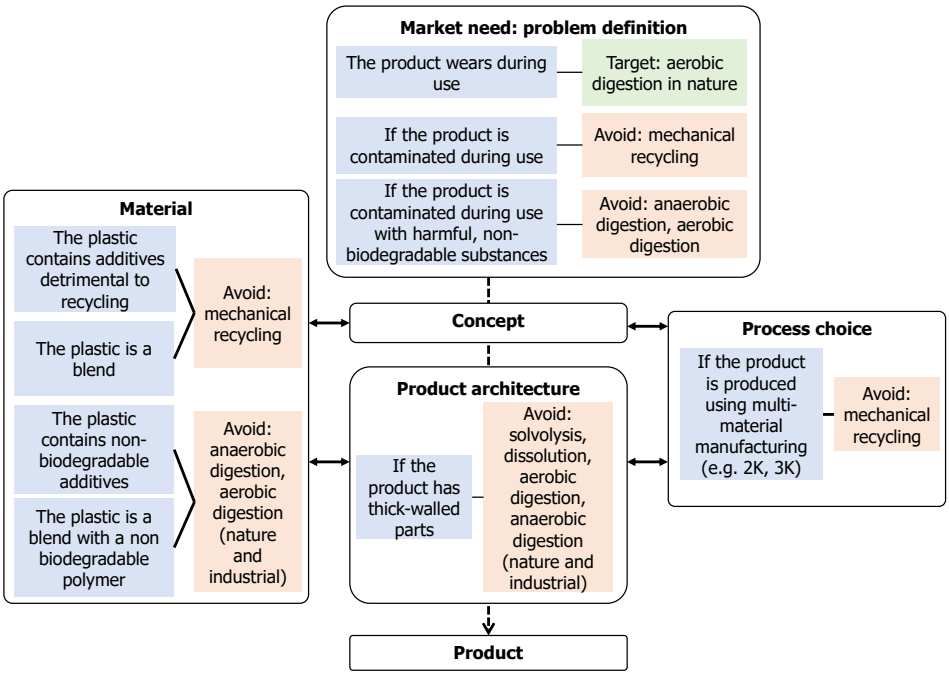

Figure 3.3: First iteration of the implementation of material-level recovery in a product design process using bio-based plastics. Figure adapted from Ashby et al. [161].

3.5 Conclusions

Bio-based plastics have attracted attention due to their perceived sustainability and circularity, evidenced by a rapidly growing market share. In order to avoid contributing to plastic pollution, efficient recovery of bio-based plastics at end-of-life needs to be facilitated. Understanding and enabling the recovery will become increasingly relevant as bio-based plastics grow into a larger fraction of plastic waste. Although bio-based plastics do not necessarily perform differently from petrochemical-based plastics in higher levels of the waste hierarchy (product-focused recovery pathways), they do perform differently in material-level recovery. The existing body of scientific knowledge does not sufficiently support circularity for bio-based plastics. The potential recovery pathways for many bio-based plastics have not yet been studied. This article contributes to this body of research by describing the available recovery pathways, how they work for specific bio-based polymers and addressing the role of product design in improving the circularity of bio-based plastics.

Dissolution, solvolysis and thermochemical recycling can deal with plastics containing additives and blends of different polymers, but often at a high environmental and economic cost. Moreover, the application of these novel recovery pathways to most bio-based plastics is not yet understood, especially in the case of dedicated bio-based plastics. Therefore,

further development of novel recovery pathways will be required, as well as further development of waste collection and sorting systems.

Biodegradation in nature is often seen as a recovery pathway to reduce plastic pollution. However, there is insufficient evidence that most of these biodegradable bio-based plastics fully degrade in nature into CO_2 and water, avoiding methane emissions. More research in realistic natural conditions over longer periods is needed to justify the use of biodegradable plastics in nature. It is also unknown if partially biodegraded plastics may become a source of micro- and nanoplastics. However, some plastic products will inevitably end up in nature, specifically products that wear during use (such as car tires or elastomer shoe soles). For these applications, biodegradation in nature may be a valid choice.

Product design plays an essential but often overlooked role in improving the circularity of bio-based plastics. Decisions made during the product design process determine the range of recovery pathways for a product, along with the presence of associated services. Therefore, the recovery must be considered starting early in the design process. The implications discussed in this paper can be expanded by including different perspectives, such as those of legislation, business development and economy. The environmental impact of different recovery pathways has not yet been sufficiently quantified and presents an important area for future research.

Bio-based plastics offer an opportunity to accelerate the transition to a circular economy, but this requires a concerted effort to consider recovery at end-of-life carefully. The results presented in this article can be used by product designers, recyclers, and plastic producers. Product designers may use the outcomes when selecting a bio-based plastic and a targeted recovery pathway while ensuring that this recovery pathway is encouraged though the product design. The results should help recyclers and plastic manufacturers facilitate efficient recovery of bio-based plastics at end-of-life. Recyclers are encouraged to consider which recovery infrastructure may become relevant for the future end-of-life plastics composition. Finally, plastic producers are stimulated to consider the potential recovery of plastics under development.

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4

Bottlenecks in establishing the environmental impact of bio-based plastics: a case study of bio-based HDPE and bio-based PET

Supplementary information can be retrieved from: Ritzen, L., 2023. Supplementary information: Sustainable design with bio-based plastics in a circular economy. doi:10.4121/ba9bc787-9613-4bff-9209-00bc39ed9150 [1].

Abstract

Bio-based plastics hold the potential to reduce the environmental impact of the plastics industry because they are based on renewable resources instead of petrochemicals. However, existing lifecycle assessment (LCA) studies of bio-based plastics have shown large discrepancies in outcomes for the same plastic. Methodological inconsistencies between LCAs have made it impossible to compare LCA outcomes directly. We conducted a series of methodologically consistent LCAs of bio-based high-density polyethylene (HDPE) and bio-based polyethylene terephthalate (PET) based on lifecycle inventories from literature to further study the factors that affect the environmental impact of bio-based plastics. Even with methodologically consistent LCAs, bio-based HDPE and bio-based PET both yielded significantly varying environmental impacts, in some cases higher than their petrochemical-based counterparts. Four key factors that contribute to variations were identified. Two could be attributed to different processes in practice: biomass type and processing. The other two were limited information and limited scoping of the studied LCAs. Understanding the factors from practice provides additional information about how bio-based plastics should be developed and how their LCAs should be interpreted. Interpreting bio-based plastic LCA outcomes should therefore not just consider the type of polymer, but also the sourcing. The process of conducting a bio-based plastic LCA should become more transparent and standardised in order to make LCA outcomes comparable and reliable.

4.1 Introduction

Plastics are indispensable materials in everyday life. Plastics are cheap, easy to manufacture and can have a wide range of properties, making them an attractive material for a many products. Today, most plastics are based on petrochemicals derived from fossil fuels, accounting for 8% of all crude oil extracted in 2015 [2]. At the same time, plastics are rarely recycled and an estimated 79% of all plastics ever produced sit in landfills or the natural environment [3]. The current, linear, plastic economy is causing irreversible damage to the environment through carbon dioxide (CO_2) emissions [4], fossil fuel depletion [4], plastic pollution [5] and the formation of microplastics [6].

Bio-based plastics are considered a sustainable alternative to petrochemical-based plastics because they are based on polymers produced (at least partially) from biomass [7]. Biomass is a renewable resource that absorbs CO_2 during its growth, and could therefore reduce the environmental impact of plastics and enhance their circularity [8–10]. A wide range of bio-based polymers has been developed, several of which are commercially available [11, 12]. Some bio-based polymers – the so-called "drop-in" bio-based polymers – are chemically identical to petrochemical-based polymers. Examples of drop-in bio-based polymers are polyethylene (PE) and polyethylene terephthalate (PET). Other, "dedicated" bio-based polymers do not have a petrochemical-based equivalent, such as polylactic acid (PLA) [13]. In 2022, bio-based plastics only accounted for 1% of the plastic market. In that year, the bio-based plastic market grew by 14%, compared to 4% for petrochemical-based plastics [14].

Biomass is a renewable resource, but the associated agricultural and conversion processes require substantial resources and energy. It is therefore essential to quantify the environmental impact of bio-based plastics by sustainability assessments such as lifecycle assessment (LCA). LCA is an established method for quantifying the environmental impact of a product, process or service [15]. An LCA is preferably performed on an existing industrial process, but it may also be used to guide the development of technologies that do not yet exist yet [16]. In such an ex-ante LCA, lifecycle inventory (LCI) data can be based on secondary data such as lab experiments [17], patents [18], interviews, process simulations, or scientific articles [17–19]. Many biobased plastics are not yet produced commercially or are in pilot-stages [11], and thus LCAs in scientific literature typically rely on secondary information.

The scientific literature is inconclusive whether bio-based plastics yield a lower environmental impact [20–23]. Spierling et al. [22], compared LCA outcomes for 10 bio-based polymers and only found sufficient data to compare greenhouse gas emissions and no other impact categories. A lack of data prevented them from concluding whether bio-based plastics were more sustainable. The large variations in bio-based plastic LCA outcomes have been attributed to both methodological inconsistencies and insufficient data. Walker & Rothman [23] compared the outcomes of 50 bio-based plastic LCAs, finding variations of over 1000% in some impact categories for the same polymer. The source of these variations could not be identified as the methodologies employed in the LCAs varied substantially. Bishop et al. [20] compared methodological decisions in 44 studies, finding inconsistencies in how studies handled aspects such as land-use-change, biogenic carbon accounting and allocation methods. They also highlighted the poor transparency of LCI data in most LCAs studied.

In this article, we take the next step in understanding the causes of the discrepancies in bio-based plastic LCAs to establish whether bio-based polymers have a lower environmental impact than petrochemical based polymers if methodological inconsistencies are removed. Furthermore, this allows us to study the sources of these discrepancies, as they may indicate specific processes with a lower environmental impact. This was achieved by studying LCIs in existing literature and replicating them with a consistent methodology. Instead of replicating LCAs from literature entirely, the production process of bio-based plastics was split up into several stages. Each stage was varied individually, keeping the others at a so-called 'base' process. This approach had three advantages. It allowed us to use a larger selection of literature, study the effect of individual stages, and to pinpoint critical areas for data quality improvements.

4.2 Methodology

4.2.1 Finding existing LCAs of bio-based plastics

Existing LCAs of bio-based polymers were found through a literature review in Scopus, using combinations of the following terms: "lifecycle assessment", "LCA", "bio-based", "polymer", "plastic", limited to journal articles published since 2010. Biofuel LCAs were not included in the analysis. The search was conducted in April 2023 and yielded 146 documents, from which 32 articles containing an LCA of bulk bio-based polymers were selected. Next, the LCAs in the articles were categorised as (partially) reproducible or non-reproducible. Polymers with only a single one reproducible LCA were not further considered, because comparisons would be impossible. LCAs with an LCI copied from previously published LCAs were also removed, because this would result in two replications of the same LCA. This resulted in 10 articles out of 32. Sufficient information to evaluate the effects of methodological differences was only available for two bio-based polymers: bio-PE and bio-PET. There are different types of PE with different molecular structures. These different molecular structures are achieved by controlling the conditions during polymerisation. The most common types of PE are high-density polyethylene (HDPE) and low-density polyethylene (LDPE). Within this work, we considered HDPE. The difference in greenhouse gas emissions between petrochemical-based HDPE and LDPE is less than 0.5% [24].

4.2.2 Methodologically consistent LCAs for bio-based plastics

Goal and scope

We conducted a series of methodologically consistent LCAs based on LCI data from scientific literature, comparing the cradle-to-gate environmental impact of producing 1 kg of polymer. Figure 4.1 shows the system diagrams of bio-HDPE and bio-PET, including three variables and the corresponding scenarios. A base scenario was established which allowed us to change one step of the polymer production process, i.e. one variable, at a time while using this base scenario for the other steps. This allowed us to use incomplete or partially confidential LCIs, and to analyse the effect of LCI data in each step in isolation. The LCAs were conducted using the LCA softwaree Activity Browser [25] with ReCiPe Midpoint V1.13 for the lifecycle impact assessment (LCIA) [26]. The background database was Ecoinvent V3.8 [24].

Both bio-HDPE and bio-PET are based on ethylene from ethanol. Ethanol is derived from carbohydrates which are abundant in many types of biomass. 11 different LCIs for ethanol production from biomass and four LCIs for the ethylene production process were found. Bio-PET is a copolymer of ethylene-based monoethylene glycol (MEG) and terephthalic acid (TA). Four LCIs for the production of ethylene from ethanol were found. Additionally, TA can be either bio-based or petrochemical-based. Petrochemical-based TA was used for partially bio-based PET as the base scenario, because fully bio-based PET is not yet commercially available [27]. Three replicable LCIs for bio-based TA yielded three fully bio-based PET scenarios. For petrochemical-based HDPE and PET, scenarios inside Europe (RER) and outside Europe (RoW) were included.

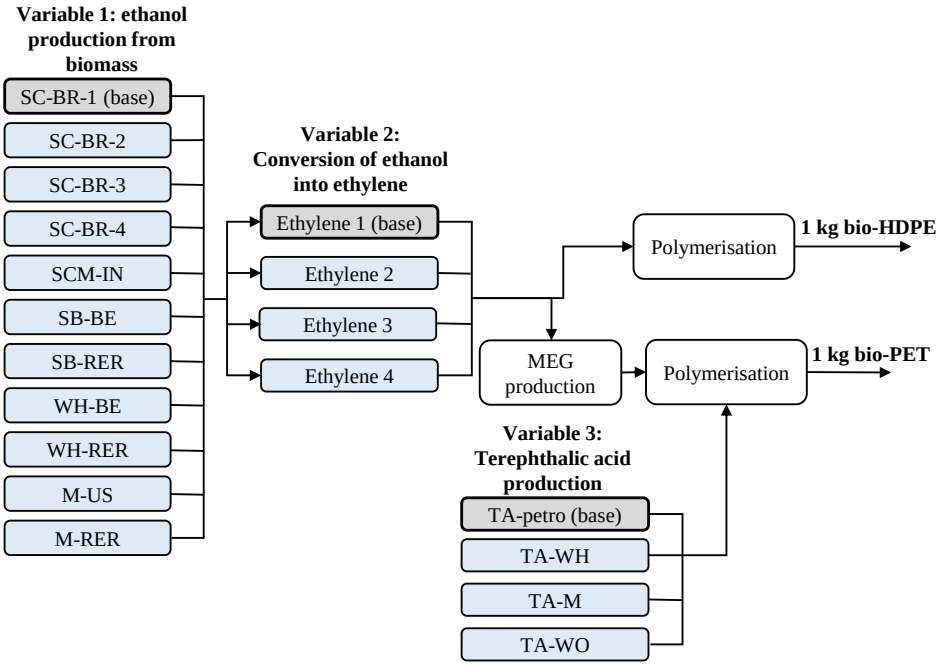


Figure 4.1: System diagram for bio-HDPE and bio-PET, marking the three variables and the base scenario considered in this study. The abbreviations and scenarios are further explained in table 4.1. SC: Sugarcane; SCM: sugarcane molasses; SB: sugar beet; WH: wheat; M: Maize, WO; Wood; BR: Brazil; IN: India; BE: Belgium; RER: the European market; US: the United States of America; TA: Terephthalic acid; MEG: monoethylene glycol.

Given the limited scope and information availability of preexisting LCA studies it is difficult to guarantee that the LCAs presented in this study are fully methodologically consistent. While the approach to aspects such as biogenic carbon accounting, LCIA, and scope are methodologically consistent, methodological differences concerning allocation and multifunctionality could not be entirely mitigated. This is because it was not always clear if a multifunctional process in the LCI was the result of a practical difference (for instance, producing bagasse as a by-product of sugarcane fermentation which can be incinerated for electricity), or a methodological decision. Multifunctionality was relevant in 4 out of the 11 ethanol production LCIs (SC-BR-2, SCM-IN, SB-BE, and WH-BE). In the SC-BR-2 scenario, bagasse was incinerated and electricity was returned to the Brazilean grid. This was approached by system expansion. In the other 3 scenarios including allocation, system expansion was not practical and economic allocation was applied.

In this article, we present the results of four ReCiPe midpoint environmental impact categories in depth. These impact categories were selected based on an endpoint LCA of the bio-HDPE base scenario breaking down the total impact in scores for each impact category to determine their relative contributions to total damage.

The outcomes of this analysis can be found in the supplementary information in table S1 [1]. Based on this end-point impact assessment, the most important impact categories were GWP100 (23% of the total impact), agricultural land occupation (25% of the total impact), and particulate matter formation (31% of the total impact). The last impact category we discuss, water depletion, does not have a midpoint to endpoint characterisation factor [26]. The effects of water depletion are reflected in other damage pathways such as freshwater and marine ecotoxicity and human health-related categories. As such, its contribution to total environmental impact could not directly be derived. Nevertheless, it was included because water consumption is an important aspect of the agriculture needed to produce bio-based polymers [28]. While contributions to ReCiPe endpoint impact categories were used to identify the most important environmental impact categories to discuss, the outcomes of these impact categories are less accurate compared to ReCiPe midpoint impact categories [26]. Therefore, we discuss the midpoint results of the impact categories.

Scenario overview

Table 4.1 contains an overview of the scenarios as well as the literature from which the LCIs in each scenario was derived. In this section, we provide a brief overview of the scenarios in this study. The detailed LCIs of all scenarios can be found in the supplementary information (table S2 - S23 [1]).

Base scenarios

The base scenario for ethanol production from biomass was sugarcane-based ethanol in Brazil. The conversion of ethanol into ethylene was based on industry LCI data published in [29]. For bio-PET, ethylene conversion into MEG was adapted from [24], replacing petrochemical-based ethylene with bio-based ethylene. The base scenario for TA production was petrochemical-based TA from the background database, because bio-PET using bio-based TA was not commercially available at the time of writing. Both bio-HDPE and bio-PET polymerisation was assumed to be equivalent to petrochemical-based production, replacing petrochemical-based ethylene and MEG with the bio-based equivalents.

Variable 1: biomass and ethanol production

The first variable studied was biomass cultivation and its conversion into ethanol. These two production stages were combined because biomass conversion into ethanol is tailored for the biomass type. For example, producing ethanol from sugarcane requires different resources than producing ethanol from maize, so the maize ethanol process cannot be applied to sugarcane.

Table 4.1: Overview of the scenarios corresponding to the three variables considered in the LCA.

Variable 1: ethanol production from biomass		
Scenario abbreviation	**Scenario description**	**Reference(s) for LCI data**
SC-BR-1 (base)	Base scenario, using ethanol based on sugarcane from Brazil using the process from the background database.	[24]
SC-BR-2	Ethanol based on sugarcane in Brazil, based on [30].	[31, 32]
SC-BR-3	Ethanol based on sugarcane in Brazil, using ecoinvent data [24].	[29]
SC-BR-4	Ethanol based on sugarcane in Brazil, based on [33].	[33, 34]
SCM-IN	Ethanol based on sugarcane molasses produced in India, using ecoinvent [35], scientific literature [36], and industry data [37].	[31, 32]
SB-BE	Ethanol based on sugar beets from Belgium, based on Belgian industry data with the ecoinvent database [35] and averages of peer reviewed and grey literature data [38–44].	[45]
SB-RER	Ethanol based on sugar beets from the total European market using the AgriFootprint database [46].	[47]
WH-BE	Ethanol based on wheat from Belgium, based on Belgian industry data with the ecoinvent database [35].	[45]
WH-RER	Ethanol based on wheat from the total European market, using the AgriFootprint database [46] and previously published industry data [48].	[47]
M-US	Ethanol based on maize from the United States of America using ecoinvent data [35].	[49]
M-RER	Ethanol based on maize from the total European market using the AgriFootprint database [46] and scientific literature [50].	[47]
Variable 2: conversion of ethanol into ethylene		
Ethylene 1 (base)	Base scenario of ethylene production. Based on industry data.	[29]
Ethylene 2	Ethylene produced through the dehydration of ethanol, based on literature data [34, 51].	[45]
Ethylene 3	Ethylene produced through the dehydration of ethanol, based on simulations in Aspen Hysys®of a process described in a patent [52].	[34]
Ethylene 4	Ethylene produced through the dehydration of ethanol, based on the GaBi database [53].	[54]
Variable 3: Terephthalic acid production from biomass		
TA-petro (base)	Petrochemical-based TA from the background database.	[24]
TA-WH	TA based on wheat through the muconic acid pathway.	[49]
TA-M	TA based on maize through the isobutanol pathway.	[49]
TA-WO	TA based on wood through the isobutanol pathway.	[54]

Variable 2: ethylene production

In addition to the base scenario for ethylene production from ethanol, three LCIs could be reproduced from literature. These three LCIs correspond to the ethylene 2, 3 and 4 scenarios. These scenarios all used different data sources. In ethylene 2, the data was based on literature data, notably including that used in [34] which ethylene 3 was derived from. In ethylene 3, the LCI was calculated based on thermodynamic simulations of patent data. In ethylene 4, the data was based on industry data from the GaBi database [53].

Variable 3: terephthalic acid production

TA is a monomer used in PET production but not in PE production. In the base scenario, petrochemical-based TA from the background database was used. Three LCIs for bio-based TA production could be reproduced from literature, each using a different production process as shown in the system diagrams in figure 4.2. In the TA-WH scenario in figure figure 4.2a, muconic acid is derived from the lignin in wheat [49]. This muconic acid is converted into cyclohexa-2,5-diene-1,4-dicarboxilate in a Diels-Alder process, which is dehydrogenated into TA. There was no exact data for each of these steps, and all were based on similar processes from literature [49]. The TA-M and TA-WO scenarios both converted biomass into TA using an isobutanol pathway. In this pathway, glucose in biomass is converted into butanol by modified yeasts. Isobutanol is then converted into isobutylene, further converted into isooctane, isooctane and into paraxylene. For all of these steps, no LCA data was available, and they were estimated based on similar processes from literature [49]. For the final step, the conversion of paraxylene into TA by oxidation, ecoinvent data was available. In the LCA on which the TA-WO scenario was based, the processes for the conversion of isobutanol into paraxylene were combined, although the underlying process was the same [54].

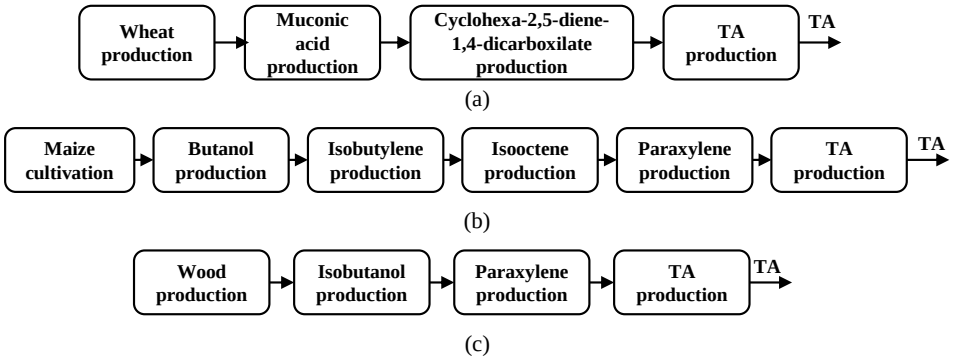


Figure 4.2: System diagrams of the three bio-based TA production routes. (a) TA from wheat using the muconic acid route (TA-WH), (b) TA from maize using the isobutanol route (TA-M), and (c) TA from wood using the isobutanol route (TA-WO) ((a) and (b) are based on [49], (c) is based on [54])

Biogenic carbon

Biomass captures CO_2 from the atmosphere as it grows. This "biogenic" carbon is then stored in the biomass and consequently stored in a bio-based plastic. All but one ([34]) of the studied LCAs removed biogenic carbon from production emissions. However, the European Union Joint Research Commission published a guideline for LCA of bio-based plastics in 2021 [28], stating that biogenic carbon should not be subtracted from production emissions but only when the biogenic carbon is reintroduced into the atmosphere (e.g. upon incineration). Since the scope of this study is cradle-to-gate, biogenic carbon would not be accounted for if this methodology was used. Furthermore, we recently showed that following the JRC procedure leads to a considerable apparent disadvantage for durable products containing bio-based plastics [55] (chapter 5). To include the effect of biogenic carbon, we accounted for it in production emissions, in the biomass cultivation stage. Only the biogenic carbon that was converted into ethanol or TA was taken into account. Biogenic carbon calculations can be found in the supplementary information [1]. They resulted in 3.14 kg for bio-HDPE, 0.46 kg for partially bio-based PET and 2.29 kg for fully bio-based PET. These values are in agreement with previously published values [32, 45].

4.3 Results

4.3.1 Global warming potential (GWP100)

In this section, we explore four impact categories in detail: global warming potential (GWP100), agricultural land occupation, particulate matter formation, and water depletion. The results of the LCIA of other impact categories can be found in the supplementary information (figure S2 [1]).

The effect of biomass cultivation and ethanol production

Figure 4.3 shows the GWP100 of the bio-based and petrochemical-based HDPE and PET. Most scenarios for bio-HDPE resulted in a lower GWP than petro-HDPE, with the exception of wheat in the total European market (RER) and the two maize scenarios. For bio-PET, most of the studied scenarios also resulted in a lower GWP100 compared to petro-PET produced in Europe (RER), with the exception of the two maize scenarios. Compared to petro-PET produced outside of Europe (RoW), only wheat in the European market and maize in the US resulted in a higher GWP100. Accounting for biogenic carbon yielded net negative emissions in nearly all bio-HDPE scenarios. The scenarios based on sugarcane molasses in India also yielded negative emissions during the biomass cultivation stage, since a by-product of sugarcane molasses production is electricity. This electricity replaces electricity in the Indian grid resulting in avoided emissions.

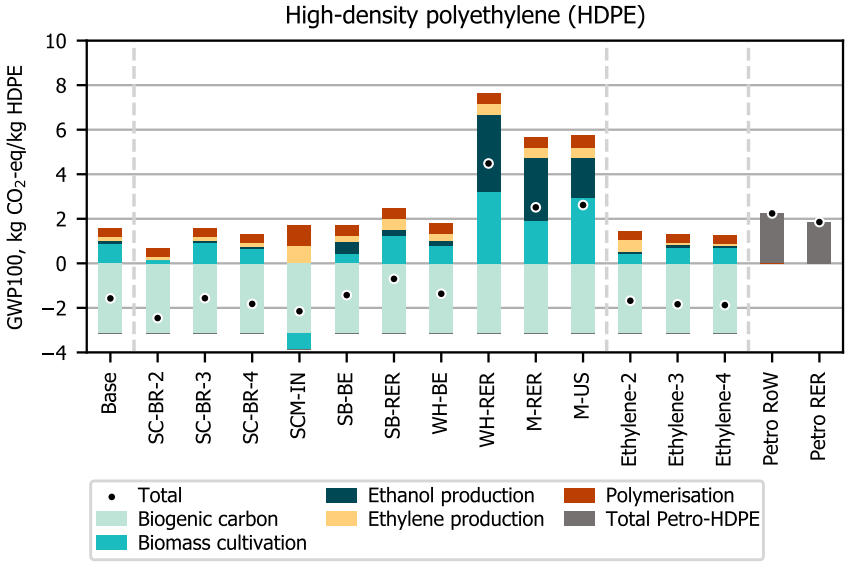
GWP100 ranged from -2.46 to 4.48 kg CO_2-eq for bio-HDPE and from 1.93 to 3.18 kg CO_2-eq for bio-PET. The variations for bio-PET were smaller due to a lower amount of ethanol used to produce this polymer (0.41 kg in comparison to 2.08 kg for bio-HDPE). Furthermore, TA production always constituted the largest share of the GWP100 for bio-PET. The biomass type with the largest variation between scenarios was wheat, with 1.99 kg CO_2-eq for the Belgian scenario and 3.16 kg CO_2-eq for the generic European scenario.

The LCI data for the processes was different for similar biomass sourcing scenarios. This could either reflect actual differences in the processes or different quality of data sources. Two examples of similar scenarios with significantly different LCIs and resulting GWP100 were the two wheat scenarios. Table 4.2 shows simplified LCIs for the production of 1 kg of ethanol from wheat. The environmental impacts associated with wheat cultivation in Belgium were much lower than those in the generic European scenario. Part of the difference may be the result of economic allocation in which 67% of impacts were allocated to ethanol production from wheat, because co-products from the process were used as fuel [45]. Comparing the LCIs, the land-use for the Belgian scenario was 9% of that assumed in the generic European scenario and fertiliser use was 55% that of the generic European scenario. Resource use for the conversion of wheat into ethanol was also several factors lower in the Belgian scenario. This generic European scenario was constituted from data from France and Germany, two of Belgium's neighbours, and the difference was therefore unexpected.

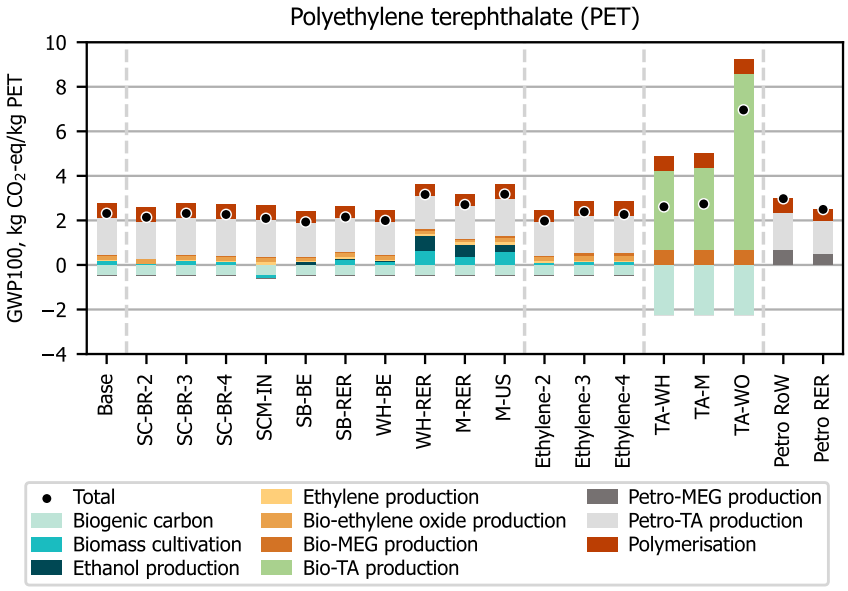
There were two explanations for variations in GWP100: sourcing and different processing. The sourcing of the bio-based plastic encompassed the biomass type and cultivation location. Biomass type in particular affected the environmental impact of bio-based plastics, as different crops require different resources to grow and to be processed into ethanol. For instance, the GWP100 associated with bio-HDPE and bio-PET using maize as biomass were always significantly higher than using sugarcane or sugar beet as biomass. The location of cultivating biomass also affected its environmental impact due to local climate and agricultural practice (e.g. harvesting methods, type of fuels in agricultural machinery). Cultivating sugar beets and wheat in Belgium resulted in lower GWP100 compared to the European market scenarios. Both maize-based cases had nearly identical GWP100 with a 4.1% difference for bio-HDPE. Yet, stage-specific emissions varied; European market saw lower biomass cultivation emissions and higher ethanol production emissions than the US scenario.

The effect of ethylene production

For the ethylene scenarios in figure 4.3, the processes for biomass cultivation and ethanol production were taken from the base scenario while the conversion into ethylene was varied based on LCIs found in literature. The LCA that the base scenario was based on used industry data, whereas the other scenarios were based on data from thermodynamical simulations. All the reproducible LCAs from literature modelled the same process for the dehydration of ethanol into ethylene, which has been patented in 1981 [51]. Incorporating the different ethylene production processes in the LCAs for polymer production resulted in a GWP100 ranging from -1.87 kg CO_2-eq to -1.57 kg CO_2-eq for bio-HDPE and from 1.97 to 2.40 kg CO_2-eq for bio-PET. These variations are relatively low compared to those found when varying biomass cultivation and ethanol production. Moreover, in all scenarios but ethylene 2, ethylene production accounted for less than 10% of the GWP100 of the production processes (excluding the biogenic carbon). In the ethylene-2 scenario, emissions due to ethylene production were relatively high (38% of total production GWP100).



(a)



(b)

Figure 4.3: GWP100 for all scenarios for the production of (a) bio-HDPE and petro-HDPE, and (b) bio-PET and petro-PET. Emissions are split up into different production stages.

Table 4.2: Simplified LCIs for the production of 1 kg of ethanol from wheat.

Producing wheat for 1 kg bio-ethanol		
	WH-BE	**WH-RER**
Total wheat needed for 1 kg ethanol	3.9 kg	3.43 kg
Agricultural land	0.45 m^2a	4.85 m^2a
Lime		2.67 g
Fertilisers	114.3 g	208 g
Pesticides	1.40 g	0.65 g
Herbicides		0.19 g
Agricultural operations		0.007 ha
Transport		0.011 t km
Packaging		282 kg
Water		463 L
Seeds		48 g
Organic chemicals		0.18 g
Other resources for producing 1 kg bio-ethanol		
	WH-BE	**WH-RER**
Heat	1.80 MJ	13.58 MJ
Organic chemicals	0.23 g	0.64 g
Inorganic chemicals	12.14 g	34.39 g

Table 4.3 contains the LCIs of the four processes. The conversion efficiency directly influenced ethanol and biomass requirements. The ethylene-2 scenario made the most optimistic assumption regarding conversion efficiency, with 1.06 kg of ethanol needed to produce 1 kg of ethylene. In contrast, the ethylene-1 scenario assumed 2.08 kg of ethanol, almost twice as high. Consequently, the environmental impact of biomass cultivation and ethanol production in ethylene-1 scenario doubled that of ethylene-2. However, the ethylene-2 scenario did have a higher environmental impact due to the ethylene production process itself, reflecting assumptions regarding resource use. The ethylene-2 scenario assumed 1.5 kg of steam would be needed to produce 1 kg of ethylene, which increased the GWP100 significantly. Steam was not included in any other ethylene production process considered in this work.

The effect of terephthalic acid production

The bio-based TA production processes in figure 4.3 resulted in a higher environmental impact compared to petrochemical-based TA. GWP100 of the fully bio-based PET scenarios ranged from 3.1 – 7.4 kg CO_2-eq. TA produced from wood through the isobutanol pathway resulted in the highest GWP100, attributed to the energy consumed during processing. TA produced through the isobutanol pathway in the TA-M scenario had a comparatively low environmental impact with the primary contributor being maize cultivation. TA from wheat resulted in slightly higher GWP100 than TA-M due to higher electricity requirements.

Table 4.3: LCIs of the four scenarios for the conversion of ethanol into ethylene.

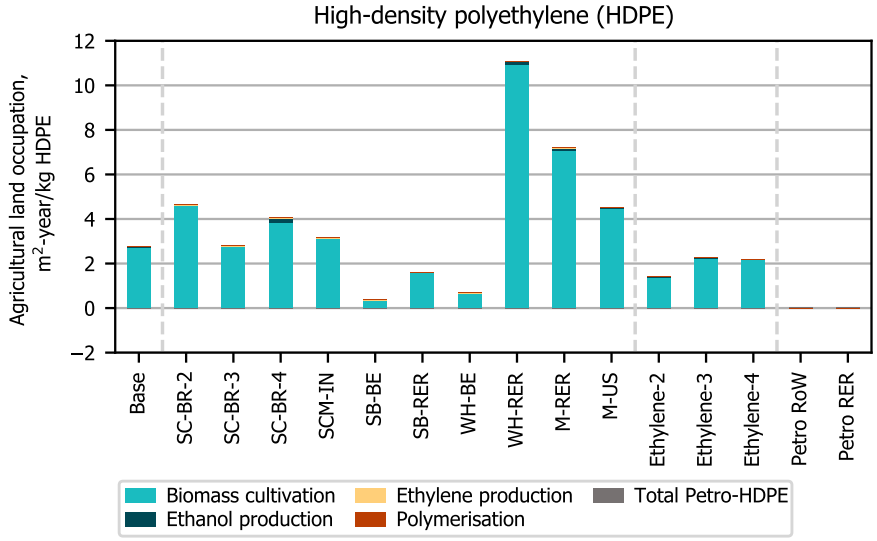
	Ethylene-1	Ethylene-2	Ethylene-3	Ethylene-4
Ethanol	2.08 kg	1.06 kg	1.70 kg	1.65 kg
Electricity	0.47 kWh	0.43 kWh	0.50 kWh	0.32 kWh
Heat, from bagasse	4.84 MJ		5.6 MJ	
Heat, from natural gas		0.26 MJ		
Liquid nitrogen	0.1044 kg			
Zeolite	0.11 g			
Sodium bicarbonate	0.0266 kg			
Water	2.57 kg			
Propylene	0.0035 kg			
Steam		1.55 kg		

4.3.2 Agricultural land occupation

In contrast with petrochemical-based polymers, the biomass for bio-based polymers needs land. The impact category "agricultural land occupation" quantifies the area of land that is occupied annually for the production of these polymers. Figure 4.4 shows the LCIA results for agricultural land occupation of bio-HDPE and bio-PET for the scenarios. Agricultural land occupation was consistently higher for the bio-based scenarios, whereas agricultural land occupation for petrochemical-based HDPE and PET was close to 0. Agricultural land occupation could mainly be attributed to biomass cultivation for both bio-HDPE and bio-PET. The results for bio-HDPE and bio-PET showed a similar pattern, but the values for bio-PET are lower because less ethanol is used in bio-PET production. Overall, agricultural land occupation ranged from 0.39 – 11.10 m^2-year for bio-HDPE and 0.12 – 2.24 m^2-year for bio-PET.

The effect of biomass cultivation and ethanol production

Similar to the variations in GWP100, the variations in agricultural land-use can partially be attributed to differences in biomass feedstock type and cultivation location. An example of the effect of feedstock type is that sugar-beet-based polymers had a consistently and considerably lower agricultural land-use than sugarcane- or maize-based equivalents. Table 4.4 shows the agricultural land occupation of 1 kg of biomass, and the amount of biomass needed to produce 1 kg bio-HDPE. For sugarcane, the difference between agricultural land occupation for 1 kg sugarcane was only 3%. The differences in the LCIA outcome were therefore a result of different conversion efficiencies for the conversion of biomass into ethanol, as already discussed in section 4.3.1. For sugar beets, wheat, and maize, the assumed biomass yield did vary significantly. The largest variation occurred between the wheat-based scenario, both in assumed biomass yield and total agricultural land-occupation.



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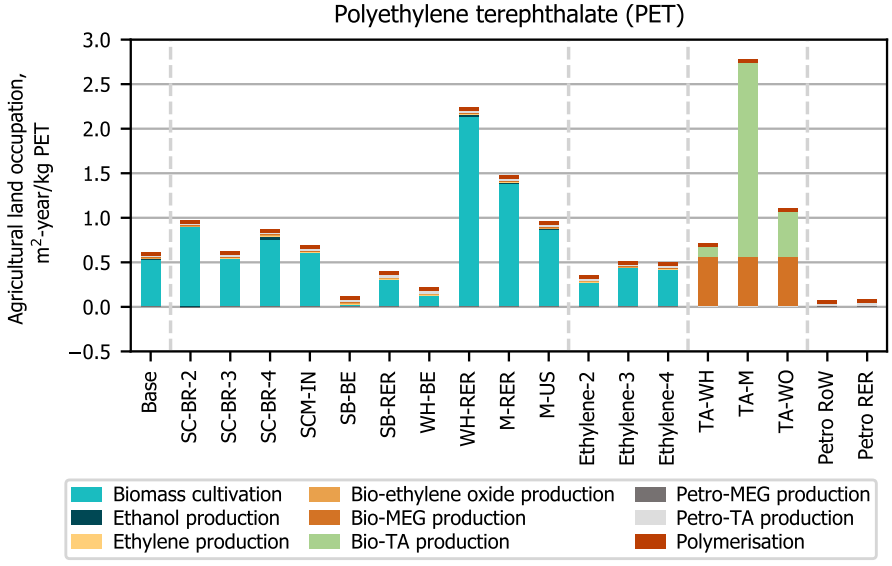


(b)

Figure 4.4: Agricultural land occupation for all scenarios for the production of (a) bio-HDPE and petro-HDPE, and (b) bio-PET and petro-PET. Land occupation is split up into different production stages.

Table 4.4: Overview of the amount of biomass required to produce 1 kg bio-HDPE and the agricultural land occupation of 1 kg biomass in each scenario.

Scenario	Amount of biomass to produce 1kg bio-HDPE	Agricultural land occupation of 1 kg biomass
SC-BR-1	18.6 kg	0.147 m^2a
SC-BR-2	31.2 kg	0.147 m^2a
SC-BR-3	18.8 kg	0.147 m^2a
SC-BR-4	25.2 kg	0.152 m^2a
SCM-IN	18.4 kg	0.169 m^2a
SB-BE	22.4 kg	0.014 m^2a
SB-RER	23.3 kg	0.068 m^2a
WH-BE	5.2 kg	0.121 m^2a
WH-RER	7.1 kg	1.531 m^2a
M-RER	5.3 kg	1.332 m^2a
M-US	6.7 kg	0.662 m^2a

Belboom & Léonard [45] stated that Belgian sugar beet and wheat production had one of the highest yields per hectare in the world. It reported a land occupation of 0.121 m^2 annually per kg wheat (with 67% impact allocation to bio-based ethanol as discussed in section 4.3.1), whereas the other wheat scenario assumed a land occupation of 1.5 m^2 annually [47]. Hence, more efficient agriculture and allocation effects reduced the agricultural land-occupation of the WH-BE scenario compared to the WH-RER scenario.

The effect of ethylene production

All bio-based scenarios for ethylene production resulted in higher agricultural land occupation than petrochemical-based scenarios in figure 4.4. The outcomes for bio-HDPE and bio-PET showed similar trends. Biomass cultivation was always the largest contributor to agricultural land occupation. The base scenario always had the highest occupation, followed by ethylene 3, ethylene 4 and finally ethylene 2. These outcomes correlated inversely with the assumed conversion efficiency, which was lowest for the base scenario and highest for the ethylene 2 scenario.

The effect of terephthalic acid production

Similar to partially bio-based PET, the agricultural land occupation of fully bio-based PET figure 4.4b) was always higher than its petrochemical-based equivalent. Agricultural land occupation ranged from 0.72 – 2.78 m^2 annually. Bio-PET using maize-based TA had the highest agricultural land occupation. However, the other two replicated LCAs lacked a land-use analysis [49, 54]. For the wood-based TA scenario, an estimation using the Ecoinvent process "Wood chips production, softwood, at sawmill [RoW]" for the amount of wood needed for 1kg of bio-PET yielded 4.7 m^2 annually per kg. This would place TA-WO above the TA-M scenario. The LCA for the production of TA from wheat did not include information about the amount of wheat needed to produce 1 kg bio-PET, so no estimation could be made.

4.3.3 Water depletion

Figure 4.5 shows the LCIA results for water depletion of bio-HDPE and PET for the scenarios where only biomass cultivation and its conversion into ethanol were varied. Water depletion refers to the volume of fresh- and groundwater consumed, and that is no longer available in reservoirs [26]. It does not include rainwater. water depletion for bio-HDPE ranged from 0.005 m3 to 1.68 m3. Water depletion for bio-PET ranged from 6.9 l to 360 l, showing similar trends as the results for bio-HDPE.

The effect of biomass cultivation and ethanol production

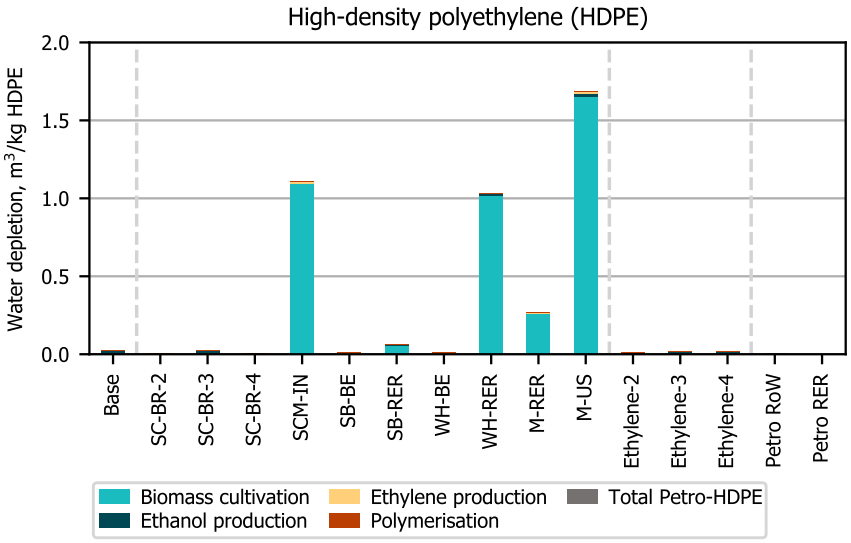
In the scenarios with a relatively high water depletion in figure 4.5, this could be primarily attributed to biomass cultivation. The water depletion depended on both the needs of the biomass type and the local climate. An example of how local climate influences water depletion were the maize-based scenarios: cultivating maize in the USA resulted in a higher water depletion than cultivating it in Europe [56]. Sugarcane cultivation in Brazil requires no or little irrigation due to the wet climate [31]. The scope of the studied LCAs was another cause for the large discrepancies. For example, the LCA from which the LCIs for sugar beets and wheat in Belgium were derived did not study water depletion, and did not include data on irrigation [45].

The effect of ethylene production

Since the base scenario for biomass cultivation and ethanol production had a relatively low water depletion in figure 4.5, the effect of ethylene production was relatively low. Similar to agricultural land occupation, water depletion was mainly the result of biomass cultivation. Therefore, the conversion efficiency, i.e. the amount of ethanol needed to produce ethylene, largely determined the resulting water depletion of the scenario.

The effect of terephthalic acid production

The results for water depletion in figure 4.5 showed a similar trend to the results for agricultural land occupation, ranging between 22 – 815 l. Maize-based bio-PET appeared to result in a relatively high water depletion compared to wheat and wood. For maize-based bio-PET, the water depletion could be attributed to maize cultivation. However, the outcomes were skewed because the LCIs of TA-WO and TA-WH did not include data on irrigation. Because wood is often not irrigated, the low water depletion due to wood seems realistic. For wheat, a higher water depletion would have been expected, especially considering that the water depletion of bio-based ethylene based on wheat resulted in a relatively high water depletion.



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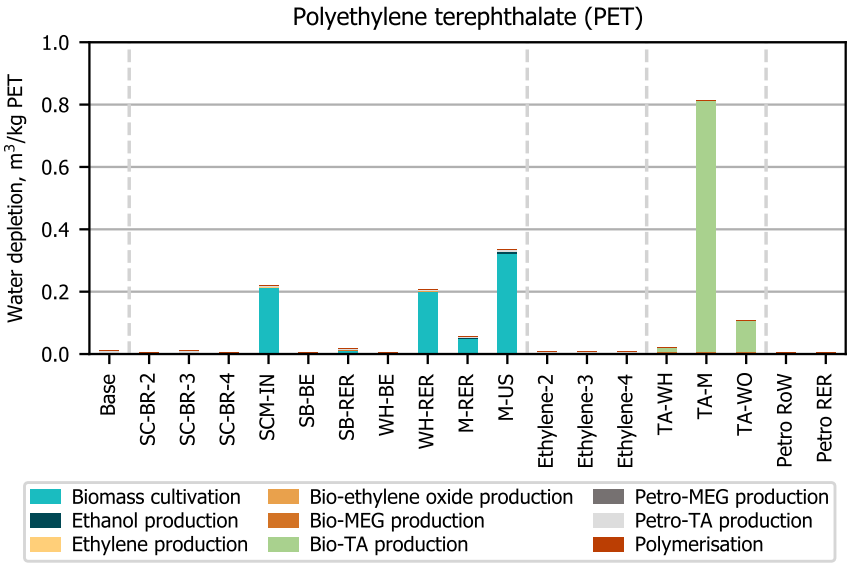


(b)

Figure 4.5: Water depletion for all scenarios for the production of (a) bio-HDPE and petro-HDPE, and (b) bio-PET and petro-PET. Water depletion is split up into different production stages.

4.3.4 Particulate matter formation

Particulate matter formation is a measure of the amount of chemicals released into the atmosphere that result in toxic effects in humans when inhaled. The particulate matter formation resulting from the different scenarios for biomass cultivation and ethanol production can be seen in figure 4.6. Outcomes in this impact categories ranged from 1.92 – 22.84 g PM10-eq for bio-HDPE and 2.72 – 7.5 g PM10-eq for bio-PET. Biomass cultivation and ethanol production were the major contributors to particulate matter formation in bio-HDPE scenarios. For bio-PET, TA production and polymerisation were also significant contributors. The particulate matter formation due to sugarcane molasses production was negative due to the avoided emissions from electricity generated during the conversion of sugarcane into molasses.

The effect of biomass cultivation and ethanol production

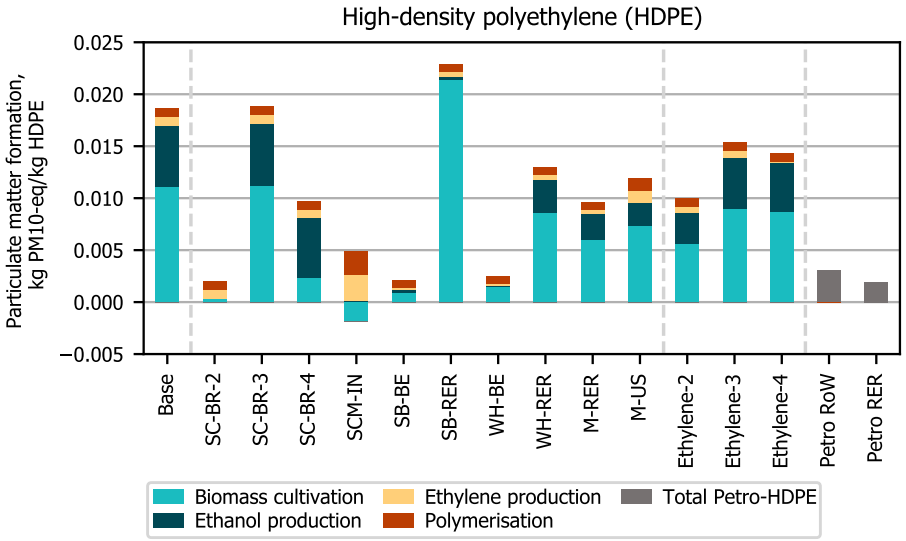
The main contributors to particulate matter formation in agricultural operations were diesel and fertilisers. The use of diesel depended on the location as different harvesting methods or agricultural machines were used. The use of fertilisers depended on the biomass type and location. For example, the largest difference was observed when comparing the two scenarios for bio-HDPE based on sugar beets: 2.11 g PM10-eq for the Belgian scenario compared to 22.84 for the generic European market scenario. In both cases, the particulate matter formation could be attributed to fertiliser use, but the assumptions for the amount and type of fertiliser used were vastly different. Producing 1 kg of sugar beets in Belgium assumed 5 grams of fertiliser, whereas in the European scenario a total of 138 grams was used. The Global Feed LCA database [56] states that Belgian sugar beet has a higher particulate matter production per kg than the countries in the European scenario (France and Germany). Furthermore, prior research suggests that Belgian sugar beet cultivation requires roughly 70 g per kg sugar beet [57]. This suggests that the fertiliser use in the literature of the Belgian scenario may be underestimated.

The effect of ethylene production

Particulate matter formation of bio-HDPE could largely be attributed to biomass cultivation and ethanol production, whereas for partially bio-based PET, it was also the result of petro-TA production. For bio-HDPE, the cause of variations was primarily the conversion efficiency of the ehtylene production process, similar to agricultural land occupation and water depletion. For partially bio-PET, the effect of this conversion efficiency was less pronounced, because the particulate matter formation due to petro-TA production did not change for the ethylene scenarios.

The effect of terephthalic acid production

Particulate matter formation for the TA scenarios in figure 4.6b followed a similar pattern as the GWP100, ranging from 13 – 22 g PM10-eq. Bio-TA always resulted in particulate matter formation at least three times as high as that of petro-TA. This could primarily be attributed to the extensive conversion steps to convert biomass to bio-TA. Bio-TA based on maize through the muconic acid pathway resulted in the lowest particulate matter formation, followed by bio-TA based on wheat. Bio-TA based on wood had the highest particulate matter formation.



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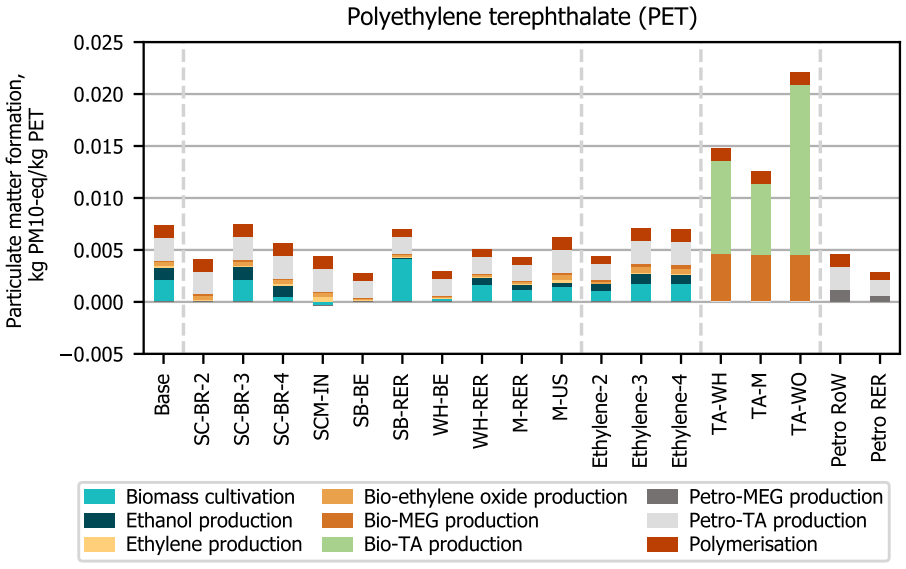

(b)

Figure 4.6: Particulate matter formation for all scenarios for the production of (a) bio-HDPE and petro-HDPE, and (b) bio-PET and petro-PET. Particulate matter formation is split up into different production stages.

4.4 Discussion

The large variations observed in bio-based plastic LCA outcomes have largely been attributed to methodological inconsistencies between studies [20, 21, 23]. We conducted methodologically consistent replications of existing bio-based polymer LCAs to establish whether bio-based polymers have a lower environmental impact than petrochemical based polymers if methodological inconsistencies are removed. In some impact categories, such as land-use and water depletion, the bio-based plastics always yielded higher outcomes than petrochemical-based equivalents. In the other impact categories discussed (GWP100 and particulate matter formation), it depended on the scenario. Variations remained high: GWP100 varied by 282% for bio-HDPE and 284% for bio-PET. Four causes were identified for these variations. Two of them can be attributed to different processes in practice: biomass type and different chemical processing. Understanding the factors from practice provides additional information about how bio-based plastics should be developed and how their LCAs should be interpreted. The other two causes were a lack of information and limited scoping of the studied LCAs. These causes indicate areas in which LCA data availability and LCA methodology for bio-based plastics should be improved. In this section, we will further elaborate on these causes for variations.

The two practical factors that affected the environmental impact of bio-HDPE and bio-PET were biomass sourcing and chemical processes. Bio-based polymers can be produced from a wide range of biomass types produced in different locations that yield varying environmental impacts, which is already well-known from biofuel research [58, 59]. As such, bio-based polymers such as bio-HDPE or bio-PET do not have an unambiguously defined environmental impact. Differences in chemical processing also affected the environmental impact of bio-HDPE and bio-PET. A higher conversion efficiency or a more energy efficient process resulted in a lower environmental impact. In the TA production processes, we compared 3 processes which resulted in significantly different environmental impacts. Since publicly available data was limited (only one LCI per scenario for bio-TA) and often derived from other processes, it was not possible to establish whether processes were real or estimated/assumed. Some other differences in processing were the use or production of by-products. Some processes considered yielded by-products, such as electricity or fuel, which replaced the local energy mix. These by-products do not reduce the environmental impact of the production process itself. However, in the LCA, they either resulted in avoided emissions or in reduced production emissions.

The transparency of LCA data for bio-based chemicals production is notoriously poor [20]. Hence, LCAs in literature made estimations based on simulations of secondary information [18], or secondary data (e.g. agricultural reports). These data have high uncertainties which can significantly affect the environmental impact. For instance, the four scenarios for the production of ethylene from ethanol (which all modelled the same process [51]), yielded variations as large as 95% for bio-HDPE in some impact categories. Factors such as conversion efficiency and energy requirements are therefore important in bio-based plastic LCAs. Sensitivity studies should be employed to quantify their effect. Improving processes may lead to significantly lower environmental impacts.

Finally, we note the limited scope of some published bio-based plastic LCAs as a cause for variations. Previous research also noted that the limited scope of bio-based plastic LCAs reduces the ability to make comparisons [20, 22, 23]. In these studies, the outcomes of bio-based plastic LCAs were compared directly. By conducting reproductions, we could conduct LCIA for all ReCiPe impact categories. However, not only the scope of the results published was limited, but also the scope of the (underlying) LCIs. Some impact categories especially important for bio-based polymers, such as land and water depletion, were not included in a number of the studied LCIs. These scenarios then yielded very low environmental impacts in those categories. Transport was another potentially important factor that was not considered in all LCIs. Table S23 [1] contains an overview of the contribution of transport to the ReCiPe midpoint environmental impact categories discussed in section 4.3. Comparing these contributions for the different scenarios, transport emissions were not significantly different between scenarios that included transport in their LCI and scenarios that did not. This is because processes in the background database also include transport for many activities, and transport did not contribute significantly to environmental impact overall (<7%).

Furthermore, the limited data availability meant that we could only analyse two polymers in detail. As mentioned in section 4.2, multi-functionality and allocation differences were not consistent between scenarios, because they can be the result of different processes. In the three scenarios that used allocation (SCM-IN, SB-BE, and WH-BE), environmental impacts were relatively low. Especially when comparing SB-BE and WH-BE to their generic European market equivalents. However, the allocation factor of impact attributed to the biomass was 84% in the SB-BE scenario and 67% in the WH-BE scenario. These allocation factors do not fully explain the differences in environmental impacts. The analyses of the LCIs in table 4.2 revealed other differences such as agricultural chemical use and electricity use.

This study itself also had some limitations. We only considered polymer production, whereas the compounding of polymers with additives, their manufacturing, and end-of-life also significantly affect the environmental impact of bio-based polymers.

Table 4.5 and corresponding figure 4.7 compare our outcomes with those in previous literature, for the same biomass type and production location. In all cases, biogenic carbon was subtracted during production. Given that the goal of this study was to reduce differences between LCAs, some discrepancies with existing literature were to be expected. However, our findings were within the range of previous studies. Noteworthy is the Brazillian sugarcane scenarios, which showed a wide spread in results of previous studies. Our results showed significantly smaller variations compared to previous literature. This suggests that using a consistent methodology can reduce variations in LCA outcomes for bio-based plastics, but biomass type needs to be considered.

Table 4.5: Comparison of the CO_2-eq emission outcomes in this work with those in previously reported literature.

Polymer type and biomass type	Greenhouse gas emissions in this work, kg CO_2-eq	Greenhouse gas emissions in literature, kg CO_2-eq
PE: Sugarcane in Brazil	-2.46 – -1.57	-1.74 [29] -0.64 – 0.86 [32] -2.84 [34] -1.14 [60] -1.84 – 0.46 [61] -1.24 [62]
PE: Wheat	-1.43 – 4.48	-0.86 [45]
PE: Sugar beet in Belgium	-1.43	-0.44 [45]
PET: Sugarcane in Brazil MEG	-2.14 – 2.31	1.75 – 1.93 [32] 2.99 [47]
PET: Sugarcane molasses in India MEG	2.09	1.52 – 1.81 [32]
PET: Wheat MEG	1.99 – 3.16	2.70 [47] 5.44 [54]

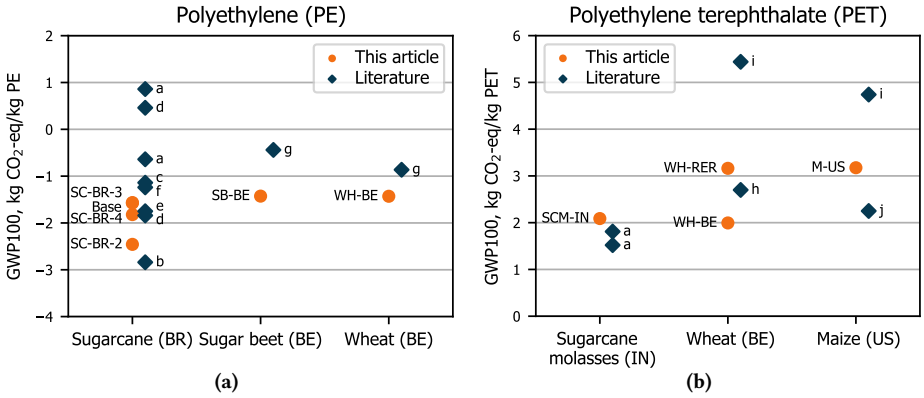

Figure 4.7: Comparison between the values from the methodologically consistent LCAs conducted in this research and those published in previous literature (a) PE, (b) PET. [a] [32], [b] [34], [c] [60], [d] [61], [e] [29], [f] [62], [g] [45], [h] [47], [i] [54], [j] [49].

We recommend that future LCA studies of specific bio-based plastics provide transparent and reproducible LCIs. Furthermore, values with and without biogenic carbon sequestration should be published and compared accordingly. In current comparisons, LCA outcomes are only compared outcomes for the same polymer type [22, 23, 63]. In order to make bio-based plastic LCA outcomes better comparable, they should be compared considering biomass type and production location. The further development of standardised methodologies for bio-based plastic LCAs will also contribute to bio-based plastic LCAs to become better comparable. This would guarantee that all impact categories are accounted for. Furthermore, the LCI data should be reported completely and transparently such that they can be replicated in future research.

4.5 Conclusions

Bio-based plastics hold the potential to be a sustainable alternative to petrochemical-based plastics. Scientific literature has been inconclusive about the environmental impact of bio-based plastics. Existing LCAs show large variations in environmental impact, attributed to methodological inconsistencies. We conducted a series of methodologically consistent LCAs of bio-HDPE and bio-PET based on LCIs from literature to reveal that methodological inconsistency is not the main cause of environmental impact variations. By only changing one production stage at a time, we tried to detangle the effect of different sourcing scenarios, data quality and chemical conversion processes. Different biomass sourcing had a significant effect on the environmental impact, so sourcing should be considered carefully when selecting bio-based polymers. Different processes could also cause variations in LCA outcomes, but this was difficult to verify due to the low quality of LCIs of many of the replicated studies.

Our results signal a need for a more nuanced approach to calculating and interpreting the environmental impact of bio-based plastics, taking into account the effect of biomass type. This is very different from the well-described source and production process of petrochemical-based polymers, which yield lower variations in outcomes even for different production locations. Comparing bio-based plastic options may ultimately require the replication of existing LCAs rather than a direct comparison of the LCA outcomes. It also remains important to further standardise LCA methodologies for bio-based plastics.

Future research is needed to elaborate on the effect of biomass type and production location on the environmental impact of bio-based polymers, as well as end-of-life. Conducting these studies for more bio-based polymers will contribute towards a better understanding of the sustainability of bio-based plastics. The contribution of additives to the environmental impact of bio-based plastics should also be assessed in order to move from polymers to plastic compounds.

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5

Sustainability of bio-based polyethylene: the influence of biomass sourcing and end-of-life

Supplementary information can be retrieved from: Ritzen, L., 2023. Supplementary information: Sustainable design with bio-based plastics in a circular economy. doi:10.4121/ba9bc787-9613-4bff-9209-00bc39ed9150 [1].

Abstract

Bio-based polymers may present a sustainable, circular way to reduce the environmental impact of plastics because they are produced from biomass that absorbs CO_2 during its growth. However, sourcing (type of biomass used and cultivation location), production, and end-of-life affect the environmental impact of bio-based plastics. We assessed the effect of sourcing and end-of-life options on the environmental impact of bio-based high-density polyethylene in 31 sourcing scenarios and five end-of-life options. Our study found that careful consideration of biomass sourcing (biomass type and production location) and end-of-life is needed to optimise the environmental impact of bio-based plastics. If these aspects are not considered, the environmental impact of bio-based high-density polyethylene may exceed that of its petrochemical-based counterpart. The direct availability of fermentable sugars indicated a lower environmental impact. The production location affected the resources needed for biomass cultivation, and the environmental impact of processing due to the energy mix. Recently published guidelines do not allow biogenic carbon to be accounted for during the production stage, but only upon the incineration of the plastic. Our results show that this way of attributing biogenic carbon results in an apparent disadvantage for bio-based plastics compared to petrochemical-based plastics. Furthermore, it disadvantaged mechanical recycling of bio-based plastics compared to incineration, a result out-of-line with circular economy principles.

5.1 Introduction

Plastics are so ubiquitous in modern society that the time we live in may well be looked back upon as the "plastic age" [2]. Over 5 billion metric tonnes of plastics have been produced since their commercial introduction in the 1950's [3]. However, plastics are associated with significant environmental problems, such as greenhouse gas emissions, plastic pollution [4] and fossil fuel use [5]. Bio-based plastics could present a sustainable, circular solution to reduce the environmental impact of plastics because they are based (at least in part) on biomass [6] that absorbs CO_2 during its growth. However, the production of bio-based polymers is fundamentally different from that of conventional, petrochemical-based polymers. Producing bio-based plastics involves the cultivation of biomass and often an extensive chemical conversion which may lead to higher environmental impacts than petrochemical-based plastics [7].

Published LCAs show high uncertainty and variations in outcomes. Walker and Rothman [7] compared 50 LCAs of bio-based plastics and found variations of over 1000% for the environmental impact of the same bio-based polymer. For instance, reported global warming potential (GWP100) values for the production of 1 kg polylactic acid ranged from 0.1 to 3.1 kg CO_2-eq. They noted three important reasons for these variations: methodological inconsistencies, feedstock source and processing. Bishop et al. [8] analysed the LCA methodologies of 44 LCAs for bio-based plastics. The variations in LCA outcomes were attributed to different system boundaries and different strategies for land-use change, biogenic carbon and allocation. They also highlighted the lack of reliable data for the chemical conversion processes involved in producing bio-based monomers as a source of variations.

The effect of feedstock sourcing for bio-based plastics has also been reported in literature by the comparison of different biomass types cultivated in different locations. PET with wood-based terephthalic acid (TA) produced in the USA was found to have a lower environmental impact compared to PET based on wheat or corn [9], or corn stover (a by-product of corn grain production) [10]. For the other building block of bio-based PET (monoethylene glycol), corn resulted in lower greenhouse gas emissions than switchgrass or wheat straw [10]. Wheat-based PET yielded lower environmental impacts than sugar beet-based PET from Germany [11], or sugarcane-based PET from Brazil [12]. However, Belboom and Léonard [13] reported a negligible 3% difference between wheat-based and sugar beet-based HDPE.

For ethanol-based polymers such as PE and PET, biofuel LCAs also provide an indication of the environmental impact of different feedstock sourcing scenarios. Muñoz et al. [14] compared six sourcing scenarios for bio-ethanol: maize grain or maize stover from the US, sugar beet or wheat from France and Sugarcane from two regions in Brazil. Sugar beet-based ethanol from France resulted in the lowest greenhouse gas emissions due to the high yield from sugar beets. Changing the feedstock for bio-ethanol from edible crops to agricultural by-products could lower greenhouse gas emissions, but were not beneficial for human and ecosystem health [15].

To the best of our knowledge, there are no publications about the effect of both biomass type and production location for bio-based polymers, as most of the aforementioned studies focus on limited scenarios and cannot be compared directly due to methodological inconsistencies. Furthermore, bio-based polymers do not necessarily solve plastic pollution issues since they are often not biodegradable in natural environments, so in order to be sustainable, recovery at end-of-life needs to be guaranteed [16]. While end-of-life options for bio-based plastics are not necessarily different to those of petrochemical-based plastics, the end-of-life emissions are different for some processes. Molecular decomposition of bio-based plastics can be considered a circular loop, as the CO_2 they emit has been previously derived from the atmosphere [17]. In order to assess whether bio-based plastics are a sustainable substitute for petrochemical-based plastics it is therefore important to also consider the end-of-life impacts.

In this article, we study the effect of biomass type, production location, and different end-of-life options on the environmental impact of bio-based high-density polyethylene (bio-HDPE). Polyethylene (PE) accounts for 30% of the entire plastics market [3]. Bio-based PE is currently produced from sugarcane in Brazil and makes up 14% of the bio-based plastics market [18]. Bio-based PE is a so-called “drop-in” bio-based polymer, which means it is chemically identical to petrochemical-based PE [19]. HDPE is a type of PE that has little branching in the polymer chain resulting in a high strength-to-density ratio. 31 scenarios for bio-HDPE production, covering five types of biomass and 12 locations, are analysed and compared to petrochemical-based HDPE (petro-HDPE). Additionally, we consider five end-of-life options for all aforementioned scenarios.

5.2 Methodology

5.2.1 Goal and scope definition

The environmental impact of bio-based HDPE (bio-HDPE) from different biomass resources in various locations was compared to that of petrochemical-based HDPE (petro-HDPE) in an LCA. Environmental impacts were calculated using the LCA software Activity Browser [20], with the Ecoinvent V3.8 background database [21]. ReCiPe Midpoint V1.13 impact categories were used for the lifecycle impact assessment (LCIA) [22]. Figure 5.1 displays the system diagrams for petro-HDPE and bio-HDPE. The environmental impact of production and end-of-life of 1 kg HDPE were compared. Since bio-HDPE is a drop-in bio-based polymer, we assume the same manufacturing methods and use scenarios; hence, they are outside the system boundary. Transport activities were not included in the system boundaries but they were studied in a separate analysis section 5.2.2.

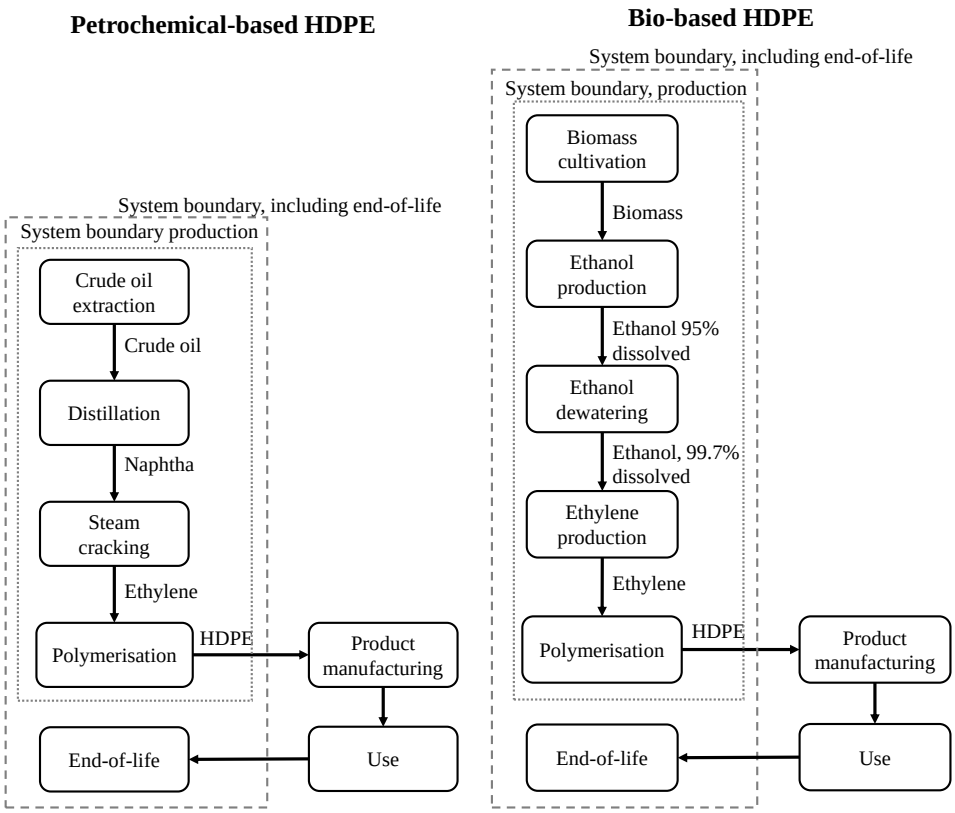

Figure 5.1: System diagram for the production of petro-HDPE and bio-HDPE.

5.2.2 Lifecycle inventory analysis

Petrochemical-based and bio-based high-density polyethylene production

The petro-HDPE production process from the background database was used unaltered, distinguishing between a European scenario (Petro-RER) and a scenario outside Europe (Petro-RoW). Bio-HDPE scenarios were established based on the availability of ethanol fermentation data, resulting in 5 types of biomass: sugarcane (SC), maize (M), sugar beet (SB), potatoes (P), and wood (WO). The amount of biomass needed to produce 1 kg of bio-HDPE varied: 18.6 kg - 23.4 kg for sugarcane, 6.7 kg for maize, 13.6 kg for sugar beet, 29.6 kg for potatoes, and 7.9 kg for wood.

Next, the cultivation for these biomass types were found in the background database, yielding a total of 22 sourcing scenarios spanning 11 locations. The biomass cultivation data were directly used from the background database as they accurately represent the emissions of growing the biomass in that location. Additional scenarios for biomass cultivation were found in the Global Feed LCA database [23]. Only locations for which scenarios from the background database were already found were included, yielding 9 additional scenarios. Table 5.1 provides an overview of the resulting 33 production scenarios and the abbreviations used throughout the article.

Table 5.1: Production scenario abbreviations and the corresponding biomass types and locations.

Scenario abbreviation	Resource	Location	Scenario abbreviation	Resource	Location
Petro-RoW	Crude oil	Outside of Europe	SB-CH	Sugar beet	Switzerland
Petro-RER	Crude oil	Europe	SB-DE	Sugar beet	Germany
SC-BR	Sugarcane	Brazil	SB-FR	Sugar beet	France
SC-CN*	Sugarcane	China	SB-SE*	Sugar beet	Sweden
SC-CO	Sugarcane	Colombia	P-CA	Potato	Canada
SC-IN	Sugarcane	India	P-CN	Potato	China
SC-US*	Sugarcane	United States of America	P-IN	Potato	India
M-BR	Maize	Brazil	P-US	Potato	United States of America
M-CA	Maize	Canada	P-CH	Potato	Switzerland
M-CN*	Maize	China	P-DE*	Potato	Germany
M-IN	Maize	India	P-FR*	Potato	France
M-US	Maize	United States of America	P-SE	Potato	Sweden
M-ZA	Maize	South Africa	WO-CA	Wood	Canada
M-CH	Maize	Switzerland	WO-CH	Wood	Switzerland
M-DE*	Maize	Germany	WO-DE	Wood	Germany
M-FR*	Maize	France	WO-SE	Wood	Sweden
SB-US	Sugar beet	United States of America			

* Scenarios from the Global Feed LCA database.

For the conversion of biomass into ethanol, localised processes were not always available and the energy mix and origin of other resources for ethanol conversion were adjusted to the country of biomass cultivation. Ethylene conversion was based on literature data [24], adjusting the energy mix and resources for the location of production. For the polymerisation step, we used the process for (petrochemical) HDPE production, replacing the ethylene with bio-based ethylene and adjusting for production location by changing the energy mix and the origin of other resources where possible. A complete lifecycle inventory can be found in the supplementary information (table S1-S3 [1]).

Biogenic carbon accounting

Bio-based polymers act as carbon storage until their biogenic carbon is reintroduced to the atmosphere, e.g., through incineration. According to methodological guidelines by the European Joint Research Commission (JRC) published in 2021, this biogenic carbon should not be subtracted from production emissions but only upon incineration of the polymer [25]. The rationale is that plastic products typically have a short lifetime, whereas global warming potential is often calculated in a period of 100 years or more, and therefore no meaningful carbon sequestration occurs. This is in contrast with some bio-based plastic LCAs published so far, where biogenic carbon was subtracted from production emissions [8].

In order to understand the effect of this approach to biogenic carbon accounting, we also considered an alternative approach, where biogenic carbon is accounted for during production. In this study, only the biogenic carbon that was converted into ethanol is taken into account. Biogenic carbon was accounted for during the biomass cultivation stage. The calculations used to determine the biogenic carbon in bio-HDPE can be found in the supplementary information [1]. From these calculations we determined the biogenic carbon stored in 1 kg of bio-HDPE to be 3.14 kg, which is in-line with existing literature [26].

End-of-life options

There are currently three realistic end-of-life options for polyethylene: landfilling, incineration and mechanical recycling [27]. In theory, there are also chemical recycling options for HDPE, but these currently do not exist at scale and there is no industry data for these processes [28, 29]. Therefore, five end-of-life scenarios were considered: mechanical recycling, incineration (with or without energy recovery), and landfilling (sanitary or unsanitary). We assumed that the end-of-life processing occurs in the same geographical region as production: in Europe if the plastic was produced in European countries and outside of Europe for non-European countries.

Mechanical recycling was modelled using the process "polyethylene production, high density, granulate, recycled". The replacement potential of recycled HDPE was implemented according to the following equation [30].

$$Impact_{withreplacement} = impact_{reprocessing} - A \cdot B \cdot impact_{virginfossilproduction} \tag{5.1}$$

A is the technical substitution ratio, i.e. the fraction of HDPE products that can be produced from recycled HDPE, and was set at 0.5 [31]. B is the avoided virgin production, which

is 0.94 kg virgin HDPE for 1 kg recycled HDPE [21]. Hence, mechanical recycling of 1 kg HDPE was assumed to avoid 0.475 kg virgin HDPE production. Recycled HDPE was assumed to replace virgin petro-HDPE since bio-based plastics only occupied 1% of the plastics market at the time of writing [18].

Incineration without energy recovery was based on the process "treatment of waste polyethylene, municipal incineration". Biogenic carbon was accounted for in the bio-based scenarios by removing it from CO_2 emissions upon incineration. To adapt the incineration process to include energy recovery, the energy recovered from the incineration of 1 kg of HDPE was subtracted from the impact of the incineration of HDPE. The lower heating value of HDPE is 42.2 MJ kg^{-1}, or 11.67 kWh kg^{-1} [32]. For electricity recovery efficiency, a value of 22% was chosen which is representative for European incineration facilities [33, 34]. With these assumptions, the incineration of 1 kg HDPE generated 2.57 kWh electricity, in-line with prior literature [13].

For landfilling, two scenarios were compared: a sanitary landfill and a non-sanitary landfill. A sanitary landfill is lined in order to isolate waste from its environment [5]. In an unsanitary landfill, leaking of waste into the environment (soil, water and air) is not prevented. The degradation of polyethylene in landfill conditions is 1% in 100 years [21], so we assumed no carbon emissions from landfilled HDPE.

Land-use change emissions

Direct land use change (LUC) is the direct repurposing of land for the cultivation of crops. For example, the change of forest land into agricultural land, or the repurposing of agricultural land for feed crops to agricultural land for crops for bio-based plastics. The ecoinvent and global feed LCA database both include direct land use change emission data. Indirect land-use change (iLUC) occurs when a land-use change inside the system boundary leads to a land use change outside of the system boundary. At the time of writing, there was no standard method to predict iLUC [25, 35]. Land-use change in one location due to biomass production for bio-based polymers may result in iLUC in other regions of the world and there are no methods to predict this. The location of this land largely affects the greenhouse gas emissions associated. Given the large scope of the analyses presented in this study and the many potential iLUC relations, including iLUC would result in a wide range of potential results that are ultimately not very reliable. iLUC is therefore not included in these analyses.

Transportation

Three transport scenarios were considered. In the first transport scenario, we assumed that the cultivated biomass would be transported 100 km by truck to the ethanol plant. In the second transport scenario, the ethanol was assumed to be produced locally and transported to the nearest port, by freight train if available and by truck otherwise. Next, it was shipped to a commercial ethylene production plant in Triunfo, Brazil, after which the ethylene was transported by freight train to a polymerisation facility in Sao Paulo, Brazil [36]. In the third and final scenario, the ethanol was shipped to Antwerp in Belgium, home to the largest petrochemical cluster in Europe [37]. An overview of the distances used in the travel scenarios can be found in the supplementary information (table S4-S5 [1]).

5.2.3 Sensitivity analysis

Data for the conversion of ethanol into ethylene is scarce and often varying [8], and reported efficiencies and material requirements may be too optimistic or may improve in the future. Therefore, we studied the sensitivity to the two largest contributors to the GWP100 of ethylene production: ethanol conversion efficiency and electricity needed, by a +10% or -10% increment. The contributions of all processes to ethylene production are reported in table S6 [1].

5.3 Results

5.3.1 Production of HDPE from different resources in different locations

16 ReCiPe Midpoint impact categories were calculated, but in this article, but we focus on four important ones for bio-based polymers: GWP100, agricultural land use, water depletion and fossil depletion. The other results can be found in the supplementary information (figure S1-S2 [1]). GWP100 is the most commonly assessed impact category in bio-based plastic LCAs [7], and it is the only impact category affected by biogenic carbon. Agricultural land use and water depletion are impact categories that are different for bio-based plastics compared to petrochemical-based plastics, since the cultivation of biomass for bio-based plastics requires both land and water. Finally, bio-based plastics hold the potential to reduce fossil fuel consumption. However, fossil fuels used in agricultural operations and the chemical conversion processes may exceed those needed to produce petrochemical-based plastics. Therefore, fossil fuel depletion was also included as an impact category to be discussed.

Global Warming Potential (GWP100)

Figure 5.2 displays the results for GWP100 of bio- and petro-HDPE, both without accounting for biogenic carbon uptake during production (in accordance with [25]) (figure 5.2a) and with biogenic carbon uptake (figure 5.2b). When excluding biogenic carbon uptake during production, six bio-HDPE scenarios resulted in a lower GWP100 than petro-HDPE from Europe: sugarcane in Brazil (SC-BR) and in Colombia (SC-CO) and sugar beets in Switzerland (SB-CH) and in France (SB-FR), and wood in Switzerland (WO-CH) and in Sweden (WO-SE). The GWP100 of bio-HDPE ranged from 1.1 kg CO_2-eq for sugarcane in Colombia to 12.7 kg CO_2-eq for potatoes in India. Figure 5.2b shows the GWP100 of bio-HDPE and petro-HDPE when accounting for biogenic carbon during production. If biogenic carbon was accounted for in the production stage, 14 additional bio-HDPE production scenarios yielded lower GWP100 than petro-HDPE from Europe: all scenarios produced from sugarcane, sugar beet and wood, as well as maize and sugar beet in Switzerland, Germany, France, and Sweden.

There was a clear distinction between biomass types, where maize and potatoes resulted in a relatively high GWP100 compared to sugarcane, sugar beet and wood. Maize- and potato-based bio-HDPE also yielded a broader variation in outcome between countries. These differences were primarily caused by the significant variation in the GWP100 of biomass cultivation (see figure 5.3). Furthermore, GWP100 due to ethanol production varied: for instance, GWP100 due to ethanol production from maize and potatoes were

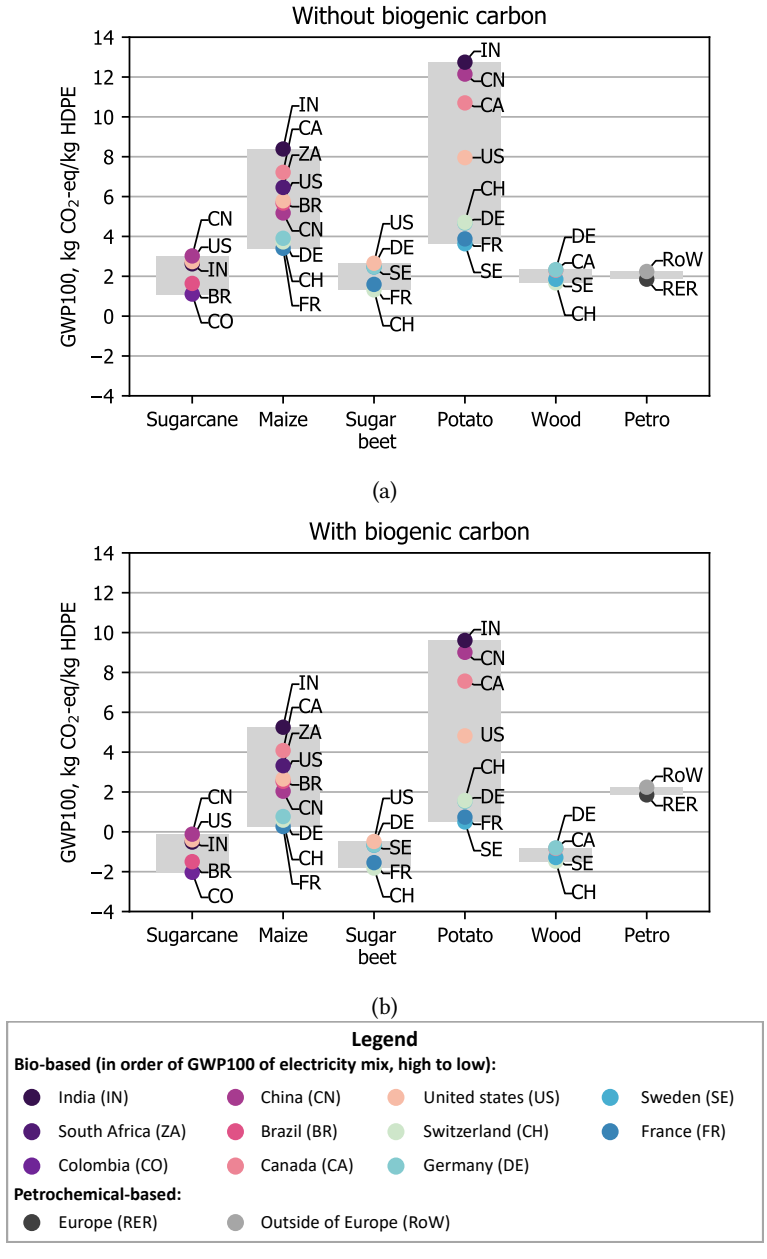

Figure 5.2: Comparison of the GWP100 of 1 kg petro-HDPE and bio-HDPE from various resources. (a) Not accounting for biogenic carbon uptake during production, and (b) accounting for biogenic carbon uptake during production. The order and colouring of the entries in the legend correspond with the GWP100 of 1 kWh of electricity at the location (from highest to lowest: IN, ZA, CO, CN, BR, CA, US, CH, DE, SE, FR).

8.5 to 37 times higher than those of ethanol production from sugarcane, attributed to the availability of fermentable sugars. Sugar-based biomass, such as sugarcane and sugar beet, contains high amounts of sugars directly available for fermentation into ethanol [14, 38]. Maize and potatoes are starch-based materials that require enzymatic hydrolisation into fermentable sugars, which increased the environmental impact of ethanol production [38].

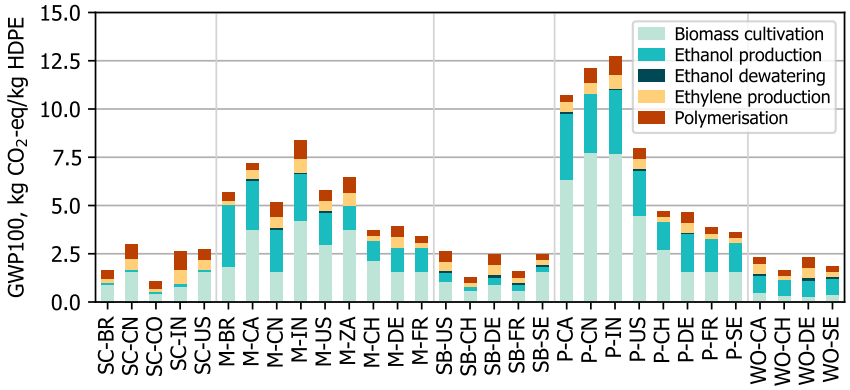

Figure 5.3: Process step contributions to the GWP100 of 1 kg bio-HDPE production, without accounting for biogenic carbon.

While wood is a cellulose-based biomass, which also needs additional conversion steps to yield fermentable sugars [38], it yielded a relatively low GWP100, as shown in figure 5.3. This could be attributed to the low maintenance of wood cultivation: wood was not irrigated or fertilised like the other biomass types. Furthermore, relatively low quantities of wood were needed to produce 1 kg of bio-HDPE: 7.9 kg. However, the GWP100 of ethanol production from wood were 6.6 – 10 times those of ethanol production from sugarcane, potentially due to the additional processing steps in converting the cellulose in wood into fermentable sugars.

Some locations consistently appeared at the higher or lower end of the GWP100 results for specific biomass types. For instance, India ranked as the location with the highest GWP100 for sugarcane, maize and potato. This was due to the environmental impact of the electricity mix of the production country, which had the highest GWP100 for 1 kWh out of all locations considered. However, this correlation did not always hold. For example, the GWP100 due to bio-HDPE production from sugar beet from Switzerland were lower than those of sugar beet in France, even though the Swiss electricity mix had a higher environmental impact compared to the French electricity mix. In this case, the difference could be attributed to emissions associated with the cultivation of these crops in these locations.

Table 5.2 compares the greenhouse gas emissions (GWP100) of this LCIA to greenhouse gas emissions reported in previous literature for each biomass type. In spite of the difficulty of directly comparing bio-based plastic LCA outcomes indicated in the introduction, we note that our outcomes are largely in-line with preexisting LCAs of bio-based HDPE. An

exception was the Braskem LCA which reported values twice as low as the ones in this study [36]. However, in the Braskem LCA, -1.10 kg CO_2-eq was attributed to land use change credits, whereas in our analysis LUC emissions were positive: 0.00097 kg CO_2-eq. Unfortunately, the lifecycle inventory for the Braskem LCA is not publicly available so the rationale for negative LUC emissions could not be derived.

Table 5.2: Comparison of the greenhouse gas emission outcomes of this study with greenhouse gas emissions from existing work.

Biomass type	This work	Other works
Sugarcane in Brazil (without biogenic carbon)	1.64 kg CO_2-eq	2.5 - 4.0 kg CO_2-eq [26] 0.3 kg CO_2-eq [39] 2 kg CO_2-eq [40] 1.3 - 3.6 kg CO_2-eq [41] 1.4 kg CO_2-eq [24] 1.9 kg CO_2-eq [42]
Sugarcane in Brazil (with biogenic carbon)	-1.50 kg CO_2-eq	-3.09 kg CO_2-eq [36]
Sugar beet (without biogenic carbon)	1.32 - 2.65 kg CO_2-eq	In Belgium: 2.7 kg CO_2-eq [13]

LUC emissions accounted for less than 5% of GWP100 outcomes (both with and without biogenic carbon) in all but two scenarios: P-IN (6.7%/8.9%, with/without biogenic carbon) and P-FR (8.9%/9.9%. with/without biogenic carbon). This was caused by a combination of LUC emissions due to potato cultivation in these locations, as well as the relatively large quantities of potatoes required to produce bio-HDPE (30 kg potatoes for 1 kg bio-HDPE). An overview of the results for LUC emissions can be found in the supplementary information (table S6 [1]).

Agricultural land occupation

In contrast with petrochemical-based plastics, bio-based plastics require land to cultivate biomass. Figure 5.4a shows the LCIA results for agricultural land occupation. Petro-HDPE resulted in relatively low agricultural land occupation (0.03 m^2-year) compared to bio-HDPE. Agricultural land-use for petro-HDPE was primarily attributed to wood cultivation for biofuels used in processing and the construction of pipelines and onshore wells. Agricultural land occupation of bio-HDPE was 31 to 1000 times as high, primarily due to biomass cultivation. The land needed depended on biomass type, location and the amount of biomass needed to produce the ethanol required to produce 1 kg of HDPE. Sugarcane and sugar beet required relatively little agricultural land, while maize, potato and wood required larger areas. Agricultural land occupation of the same biomass type also differed between locations. For instance, agricultural land needed to grow potatoes varied by a factor of 3.5 between India and China, potentially reflecting the local soil suitability and climate.

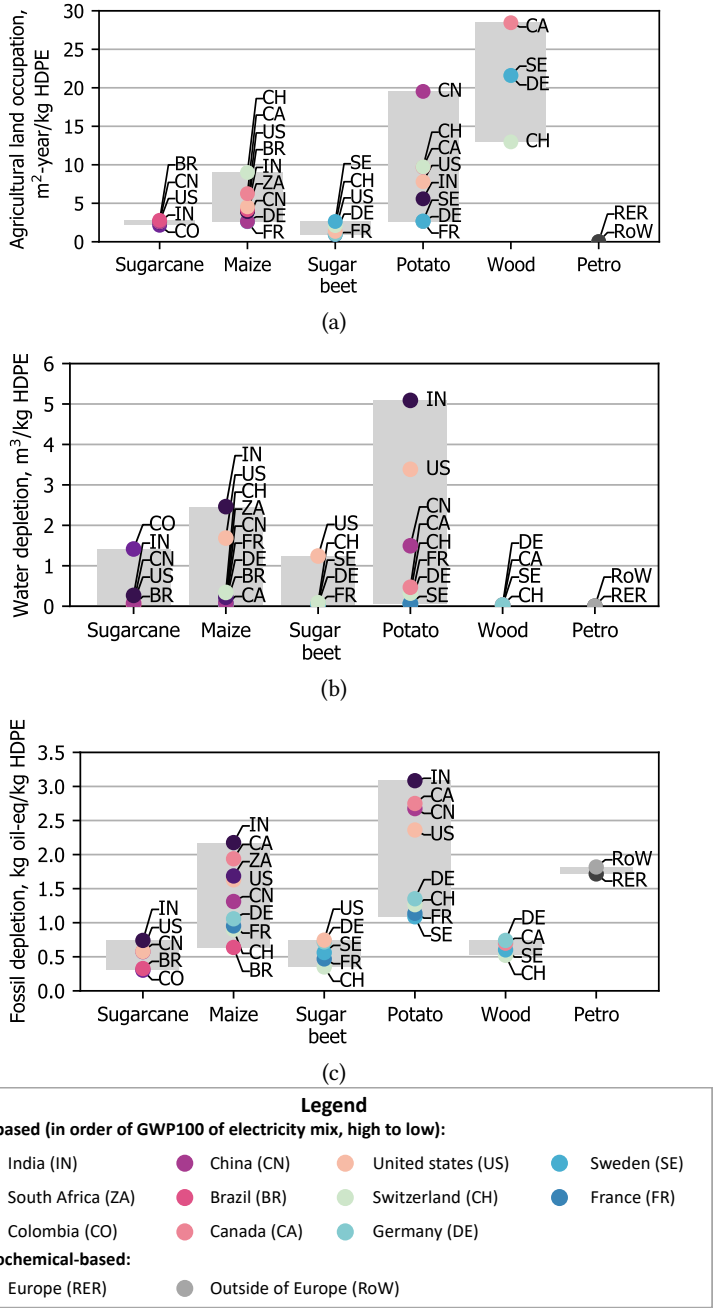

Figure 5.4: Comparison of the environmental impact of the production of 1 kg petro-HDPE and bio-HDPE from various resources in the following categories: (a) Agricultural land occupation, (b) water depletion, and (c) fossil depletion. The order and colouring of the entries in the legend correspond with the GWP100 of 1 kWh of electricity at the location (from highest to lowest: IN, ZA, CO, CN, BR, CA, US, CH, DE, SE, FR).

Water depletion

The biomass used for bio-HDPE needs water to grow, either supplied passively by rain or actively through irrigation. Figure 5.4b shows the LCIA results for water depletion of bio-HDPE and petro-HDPE. Petro-HDPE production resulted in less than 1 L water depletion. The amount of water needed for irrigation during biomass cultivation depended on the type of crop and the climate at the location where it was grown. In the scenarios for wood, biomass was not watered, so water was only consumed during ethanol production, resulting in a water depletion of less than 33 L water per kg bio-HDPE. For other biomass types, water depletion depended on location, up to over 1200 L, 1400 L, 2400 L, and 5000 L for sugar beet, sugar cane, maize, and potatoes, respectively.

Fossil depletion

Figure 5.4 shows the fossil fuel depletion of bio- and petro-HDPE. When producing bio-HDPE from maize or potatoes, the location determined whether the fossil depletion was larger or smaller than petro-HDPE. All sugarcane-, sugar beet- and wood-based bio-HDPE scenarios led to a lower fossil depletion than petrochemical HDPE. Similar to the GWP100 results, a larger spread in the outcomes for maize and potatoes compared to the other biomass types was observed, with the primary contributors being biomass production and conversion into ethanol. Fossil depletion outcomes correlated with the environmental impact of the local electricity mix, which was expected since an energy mix more reliant on fossil fuels also has a higher environmental impact. Any activity that consumes energy, e.g. treating the biomass with agricultural machinery, harvesting it, heating it to produce ethanol, also consumed more fossil fuels.

5.3.2 Effect of end-of-life options on global warming potential

Regarding the end-of-life options of bio- and petro-HDPE, we only present the results for GWP100. GWP100 is the only impact category that considers CO_2 emissions, allowing a comparison between biogenic carbon accounting approaches. The results for other impact categories can be found in the supplementary information (Figure S3 [1]).

The GWP100 results are shown with accounting for biogenic carbon either at end-of-life (following [25]) (figure 5.5a) or in the production stage (figure 5.5b). The results of both ways of accounting for biogenic carbon led to the same final result when considering incineration (with or without energy recovery). For HDPE based on sugarcane, sugar beet and wood the results were more favourable than for petro-HDPE. For maize and potatoes the result depended on the location of growth and production. When bio-HDPE is landfilled, it does not biodegrade significantly and the biogenic carbon is essentially stored. The landfilling processes resulted in 0.11 kg CO_2-eq (sanitary) – 0.17 kg CO_2-eq (unsanitary) emissions for 1 kg HDPE (bio-based or petrochemical-based), or a 0.9% - 15.0% increase in GWP100.

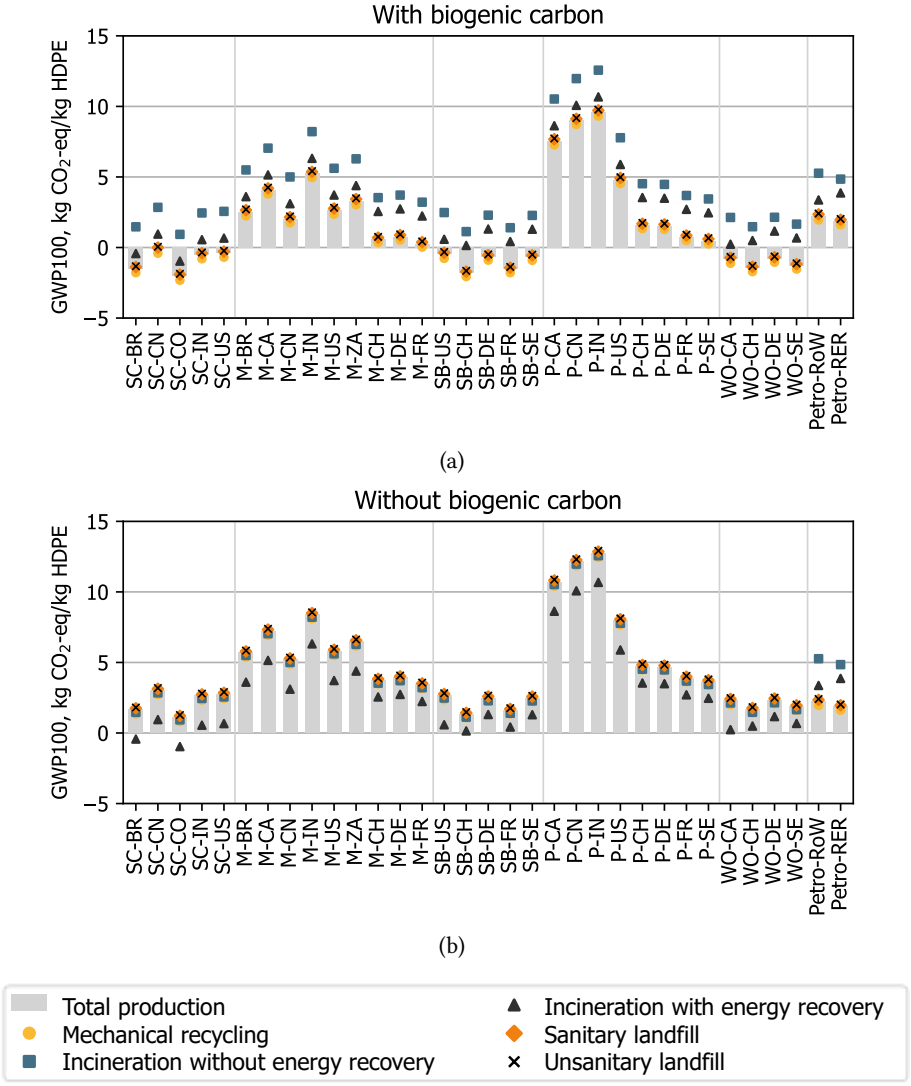

Figure 5.5: Comparison of five end-of-life scenarios for 1 kg bio-HDPE and petro-HDPE: mechanical recycling, incineration with energy recovery and incineration without energy recovery. Carbon accounting was considered in two ways: (a) biogenic carbon was accounted for upon incineration of the plastic, and (b) biogenic carbon was accounted for during production. The grey bar represents the total production GWP100, the markers represent the total GWP100 after the different end-of-life scenarios.

If energy is recovered from incineration, this energy replaces the local electricity mix, resulting in avoided emissions that reduced GWP100 outcomes. Hence, the avoided emissions were higher in locations with an energy mix more reliant on fossil fuels. If the GWP100 due to HDPE production were lower than that of the electricity it replaced after incineration, this resulted in net negative GWP100 outcomes. Net negative GWP100 outcomes only occurred for bio-HDPE from sugarcane in Brazil and in Colombia. However, the benefits of energy recovery from incineration may diminish in the coming decades. 10 out of the 11 countries considered in this article have committed to reducing the greenhouse gas emissions of the global energy sector to net zero by 2050 [43]. If the electricity mix becomes entirely based on renewables, the environmental impact of bio- and petrochemical-based polymers will decrease. If energy recovered from the incineration of bio-based plastics replaces a fully renewable electricity mix in the future, the benefits of incineration with energy recovery will diminish (at least in terms of greenhouse gas emissions).

The impact of the mechanical recycling process itself was always the same because it always concerned the mechanical recycling of HDPE with the same energy requirements, and avoiding the production of virgin petrochemical-based HDPE. Combined with production emissions, GWP100 after mechanical recycling led to a lower impact compared to virgin HDPE. Mechanical recycling was always the end-of-life option resulting in the lowest GWP100 for petro-HDPE. For bio-HDPE, the accounting method for biogenic carbon led to remarkable differences: mechanical recycling seemed to have the lowest impact when biogenic carbon is accounted for in production (figure 5.5b), whereas incineration with energy recovery appeared to result in the lowest impact in the case of accounting for biogenic at end-of-life (figure 5.5a).

5.3.3 The effect of transport

In the previously described scenarios for production and end-of-life, transport emissions were not included in the analysis. In this section, we assess the effect of including transport in three scenarios. The first transport scenario considers the transport of the biomass per freight lorry for 100 km. The effect on GWP100 ranged from 0.7% to 15.8%, depending on two factors: the GWP100 of polymer production and the weight of biomass transported. GWP100 of transport in scenario 1 were highest for potatoes, as 30 kg of potatoes are needed to produce 1 kg of bio-HDPE. The largest effect was observed in sugarcane-based HDPE, because the GWP100 due to production were relatively low for these scenarios.

In the other two transport scenarios, ethanol from biomass was transported to a factory in either Triunfo, Brazil (transport scenario 2) or Antwerp, Belgium (transport scenario 3). In the second transport scenario, the effect on GWP100 ranged from 3.5% to 33%. Scenarios with cultivation in China yielded the highest GWP100, since the distances were the largest. However, the largest difference of 33% was observed in the SC-CO scenario, because the GWP100 of production was low (0.36 kg CO_2-eq) compared to the emissions of transport (0.21 kg CO_2-eq). In the third transport scenario the ethanol was transported to a factory in Antwerp, Belgium. The effect on GWP100 ranged from 1% to 30%, with the largest effect on the WO-CA scenario, due to the relatively low environmental impact (2.30 kg CO_2-eq) and the relative large distance travelled (resulting in 0.69 kg CO_2-eq). The full results for each transport scenario can be found in the supplementary information (figure S4 [1]).

5.3.4 Sensitivity analysis

Table 5.3 displays the results of the sensitivity analysis. The highest sensitivity was found for the ethanol conversion efficiency. Reducing the conversion rate of ethanol into ethylene by 10% led to an increase in GWP100 of 3.6% - 9.2%, depending on the scenario. Changing the ethanol conversion efficiency affected the amount of ethanol needed and, by extension, the amount of biomass needed. Therefore, sensitivity to ethanol conversion efficiency depended on the part of the environmental impact caused by biomass and ethanol production combined. As such, the sensitivity to ethanol conversion efficiency of the sugarcane- and sugar-beet-based scenarios was relatively low compared to wood-, maize- and potato-based scenarios.

The sensitivity to the amount of electricity needed for the conversion of ethanol into ethylene was less significant. Increasing the amount of electricity by 10% resulted in GWP100 increases ranging from 0.01% - 2.46%, depending on the scenario. The sensitivity depended on the GWP100 of the local electricity mixture and on the fraction of the GWP100 of bio-HDPE production attributed to ethylene production. The scenario Sugarcane in India (SC-IN) had the highest sensitivity with 2.46%. Ethylene conversion caused 40% of the GWP100 in that scenario, and the GWP100 of electricity in India is also the highest of all locations studied (nearly three times as high as for electricity in Brazil). These two factors combined caused the relatively high sensitivity to the electricity needed for ethylene production in this case.

5.4 Discussion

Bio-based plastics hold the potential to yield lower greenhouse gas emissions and reduce fossil fuel dependency compared to petrochemical-based plastics. However, sourcing and end-of-life decisions significantly affect the environmental impact of bio-based plastics. Also, the applied LCA methodology affects how results are perceived. In this article, we developed 33 souring scenarios, 5 end-of-life scenarios and 3 transport scenarios for bio-based HDPE and compared these to petrochemical-based HDPE. Furthermore, we assessed the effect of different biogenic carbon accounting methods on the outcomes of the LCA.

Selecting a bio-based plastic for products should carefully consider three aspects: biomass type, production location, and end-of-life. Biomass type had the biggest effect on environmental impact outcomes. Sugar-based biomass such as sugarcane or sugar beet should be preferred over other biomass types, based on its lower environmental impacts across impact categories. This could be attributed to relatively high yields and the direct availability of fermentable sugars. Although the yield of cellulose-based biomass (wood) was high, associated land-use was also high as well as the energy required to convert cellulose into fermentable sugars. These outcomes were in agreement with previous work in biofuels [14, 44].

Location of biomass cultivation also affected the environmental impact. This could be attributed to three factors: energy mix, climate and local agricultural practice. The energy mix affected the impact of the chemical processes. The climate affected the need for agricultural operations such as irrigation and pesticide use. Local agricultural practice involved activities such as the method of cultivation (manual or machinal) and fuels used

Table 5.3: Outcomes of the sensitivity analysis of the GWP100 for ethanol conversion efficiency and ethylene production efficiency.

	Ethanol conversion efficiency		**Ethylene production energy**	
Scenario	**+10%**	**-10%**	**+10%**	**-10%**
SC-BR	+6.15 %	-6.15 %	+0.63 %	-0.63 %
SC-CN	+5.55 %	-5.55 %	+1.55 %	-1.55 %
SC-CO	+4.67 %	-4.67 %	+0.73 %	-0.73 %
SC-IN	+3.59 %	-3.59 %	+2.46 %	-2.46 %
SC-US	+6.13 %	-6.13 %	+0.85 %	-0.85 %
M-BR	+8.88 %	-8.88 %	+0.18 %	-0.18 %
M-CA	+8.83 %	-8.83 %	+0.02 %	-0.02 %
M-CN	+7.41 %	-7.41 %	+0.91 %	-0.91 %
M-IN	+7.99 %	-7.99 %	+0.77 %	-0.77 %
M-US	+8.17 %	-8.17 %	+0.40 %	-0.40 %
M-ZA	+7.72 %	-7.72 %	+0.79 %	-0.79 %
M-CH	+8.60 %	-8.60 %	+0.05 %	-0.05 %
M-DE	+7.20 %	-7.20 %	+0.67 %	-0.67 %
M-FR	+8.27 %	-8.27 %	+0.11 %	-0.11 %
SB-US	+6.01 %	-6.01 %	+0.88 %	-0.88 %
SB-CH	+6.05 %	-6.05 %	+0.14 %	-0.14 %
SB-DE	+5.59 %	-5.59 %	+1.05 %	-1.05 %
SB-FR	+6.31 %	-6.31 %	+0.23 %	-0.23 %
SB-SE	+7.83 %	-7.83 %	+0.08 %	-0.08 %
P-CA	+9.21 %	-9.21 %	+0.01 %	-0.01 %
P-CN	+8.89 %	-8.89 %	+0.39 %	-0.39 %
P-IN	+8.68 %	-8.68 %	+0.51 %	-0.51 %
P-US	+8.67 %	-8.67 %	+0.29 %	-0.29 %
P-CH	+8.90 %	-8.90 %	+0.04 %	-0.04 %
P-DE	+7.66 %	-7.66 %	+0.56 %	-0.56 %
P-FR	+8.48 %	-8.48 %	+0.10 %	-0.10 %
P-SE	+8.53 %	-8.53 %	+0.06 %	-0.06 %
WO-CA	+6.34 %	-6.34 %	+0.05 %	-0.05 %
WO-CH	+6.87 %	-6.87 %	+0.11 %	-0.11 %
WO-DE	+5.32 %	-5.32 %	+1.11 %	-1.11 %
WO-SE	+7.11 %	-7.11 %	+0.11 %	-0.11 %

in machinery [45]. The location with the lowest environmental impact therefore also depended on the biomass type. In most cases, the environmental impact correlated with the greenhouse gas emissions of the local electricity mix. Locations in Europe (Germany, Sweden, France, Switzerland) typically resulted in the lowest environmental impact.

Bio-HDPE has a relatively simple production process with few chemical conversion steps [46]. At the same time, molecular structure of PE ($(CH_2)_n$) means that most of the molecular weight consists of carbon atoms, and therefore the biogenic carbon storage of this polymer is relatively high. If more extensive chemical conversion is needed (such as for ethanol-based bio-based polypropylene [47], or bio-PET [12]), the effect of energy mix may become even more pronounced.

The results with respect to the different end-of-life scenarios reveal a notable preference for incineration with energy recovery over mechanical recycling of biobased plastics if biogenic carbon is only accounted for upon molecular decomposition following the JRC guidelines. This preference is not in line with circular economy principles, where materials should be kept at the highest possible value for as long as possible [48]. This preference emerges due to the omission of carbon stored in the polymer. In this approach, the CO_2 generated during incineration is biogenic and does not contribute to the GWP100. Notably, methane or other greenhouse gas emissions still contribute to the GWP100. The electricity generated by the incineration of a bio-based plastic does not have any CO_2 emissions, but replaces the local electricity mix leading to avoided emissions and a net-negative outcome.

Only accounting for biogenic carbon upon molecular decomposition means that biogenic carbon is not accounted for during the mechanical recycling process. Mechanical recycling requires energy and resources and has some avoided emissions because it replaces the production of virgin HDPE. However, with the emissions from the mechanical recycling process itself, the benefits in terms of GWP100 are relatively small. As a result, mechanical recycling resulted in a higher GWP100 compared to incineration in figure 5.5b. This approach does not acknowledge the potential value retention or the prolonged biogenic carbon storage of mechanical recycling. Although landfilling plastics has many drawbacks [49], prolonging the storage of biogenic carbon instead of re-releasing it into the atmosphere may be beneficial in terms of GWP100. If biogenic carbon is accounted during production, as in figure 5.5a, mechanical recycling resulted in a lower GWP100 compared to the incineration scenarios. This outcome better aligned with circular economy principles of retaining value.

Furthermore, in certain impact categories such as agricultural land occupation and water depletion, bio-HDPE consistently exhibits a higher environmental impact than petrochemical-based HDPE. This underscores the complexity of achieving circularity and sustainability in the plastics industry, emphasizing the importance not only of using renewable feedstocks but also of retaining the value of plastics at a high level for an extended duration [48].

It is therefore proposed that biogenic carbon be accounted for during production, despite the current non-supportive stance of the JRC guidelines. This approach offers two significant advantages: firstly, a more equitable comparison of the potential impact

reduction of bio-based plastics throughout the lifecycle against the impact of petrochemical-based plastics. When biogenic carbon uptake during production is considered, 20 bio-HDPE production scenarios result in lower GWP than petro-HDPE, compared to only 6 scenarios when biogenic carbon is accounted for upon incineration. Secondly, considering biogenic carbon during production demonstrates that value-retaining end-of-life options have a lower environmental impact compared to incineration with energy recovery. Mechanical recycling emerges as the end-of-life option with the lowest cradle-to-grave GWP100 in all scenarios, followed by landfilling (i.e., the sequestration of biogenic carbon). In contrast, incineration without energy recovery emerges as the least favorable end-of-life option.

This study still had several limitations, which also present opportunities for future research. The results presented in this article are likely accurate for bio-based low density polyethylene (LDPE) since it has the same production process until the polymerisation step which did not account for a large share of the environmental impact in this study. However, these results are not necessarily valid for other polymers. A similar study could be conducted with different polymers types in order to study if the same principles described here hold. However, data availability for chemical conversion processes involved in bio-based plastic production is notoriously poor [8]. A better understanding of the environmental impact of bio-based plastics under different sourcing conditions ultimately requires an improved access to bio-based polymer production data. Finally, indirect land-use change was not considered in the analyses, whereas its greenhouse gas emissions could exceed those of the entire bio-based polymer production [41]. The development of a clear method for accounting indirect land-use change is very important for increasing the accuracy of bio-based polymer LCAs and remains one of the most pressing subjects for future research in the field of bio-based polymer LCAs.

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6

Discussion

6.1 Introduction

Bio-based plastics can contribute to realising a circular economy for plastics due to their renewable feedstocks and the ability to capture and store CO_2 from the atmosphere. Nevertheless, their production involves many steps that have an environmental impact that can exceed that of petrochemical-based plastics. Furthermore, bio-based plastics still contribute to plastic pollution if not managed appropriately. Therefore, the circularity of bio-based plastics is not a given. Rather, it needs to be enabled through thoughtful material development and product design. This led to the main research objective: *To explore how material development and product design can enable bio-based plastics to be sustainable and circular.*

In order to meet this goal, four studies were conducted. In the first study in chapter 2, the challenges faced by practitioners when using bio-based plastics today were identified. In the other studies of the PhD research, two of these challenges that target the circularity and sustainability of bio-based plastics in particular were focused on: recovery at end-of-life and environmental impact. The recovery of bio-based plastics was covered in chapter 3, where the results of an elaborate literature review on the recovery of bio-based plastics are presented as well as the implications this has on product design. To clarify the environmental impact of bio-based plastics, two studies were conducted. In chapter 4, methodologically consistent reproductions of LCAs from literature were presented to understand where variations on outcomes came from. Finally, chapter 5 presented the results of a deeper investigation into the effect of sourcing on environmental impact of bio-based plastics.

In this chapter, the results presented in chapter 2-5 are synthesised. Section 6.2 starts with a brief summary of the four studies. In section 6.3, the results of all four studies are combined to discuss under which conditions bio-based plastics can be sustainable and circular, and what implications this has for product designers. Section section 6.4 reflects on lifecycle assessment (LCA) as a tool for sustainable product design with bio-based plastics. Section 6.5 contains the scientific and practical contributions of the work, and section 6.6 presents some recommendations for future work in this topic.

6.2 Recap of main findings

To achieve this research objective, four studies were conducted. The outcomes of the studies are summarised in figure 6.1. This section briefly details the rationale of these studies and their main outcomes.

The aim of the first study in chapter 2 was to identify the most pressing research gaps as identified by practitioners. The research question for this study was: *What challenges do actors throughout a durable product's value chain face when using bio-based plastics?* To answer this question, a workshop was organised involving stakeholders spanning a durable products value chain. While participants displayed a keen interest in bio-based plastics, they lacked access to dependable information on their usage. The circularity and sustainability of bio-based plastics were identified as strong drivers for their usage, but these concepts were also considered ill-defined. Participants indicated that there was confusion regarding the recovery of bio-based plastics at end-of-life. Furthermore, the environmental impact of bio-based plastics needed to be better qualified to justify their

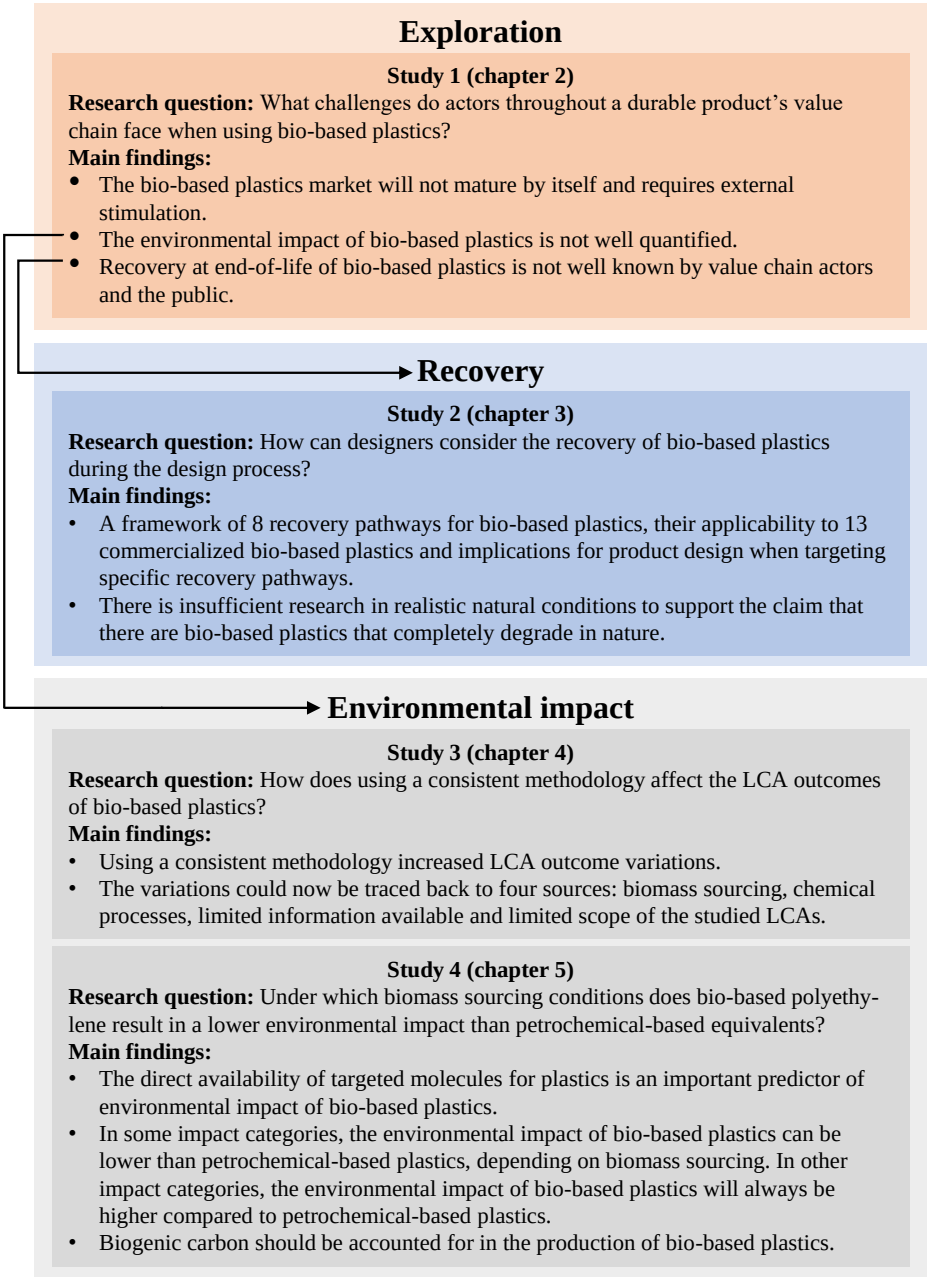

Figure 6.1: Overview of the four studies presented in this dissertation, their interrelations and their main outcomes.

usage. The lack of reliable information, combined with a lack of knowledge among value chain actors, lead to a risk of greenwashing when using bio-based plastics.

To better understand the recovery of bio-based plastics at end-of-life, the second study presented in chapter 3 was conducted. The research question was: *How can product designers consider the recovery of bio-based plastics during the design process?*. A framework was set up with 8 recovery pathways for bio-based plastics: mechanical recycling, dissolution, solvolysis, enzymatic depolymerisation, thermochemical recycling, anaerobic digestion, aerobic digestion, and incineration. The compatibility of these recovery pathways with 13 commercially available bio-based plastics was studied in a rigorous literature review. This led to the definition of implications for product design when targeting a specific recovery pathway based on the technical characteristics of that recovery pathway. The implications for product design are summarised in figure 6.2. Incineration and biodegradation could be considered circular for bio-based plastics due to their biogenic carbon, whereas they are linear for petrochemical-based plastics. However, the biodegradation of bio-based, biodegradable plastics was found to be highly dependent on the environmental conditions. Some bio-based plastics can degrade sufficiently in industrial conditions. However, at the time of writing there was insufficient research on biodegradation in realistic natural conditions to support the claim that there are bio-based, biodegradable plastics that fully degrade in nature within a reasonable timeframe.

The environmental impact of bio-based plastics has been a topic of debate since 2013 [2–4]. One main problem with bio-based plastics LCAs is methodological inconsistencies. The third study in chapter 4 aimed to answer the question: *How does using a consistent methodology affect the LCA outcomes of bio-based plastics?* LCAs based on LCI data in literature were conducted for polyethylene and polyethylene terephthalate in a total of 34 scenarios. The outcomes were also compared with petrochemical-based equivalents. A consistent methodology actually increased the variations in bio-based plastics LCAs. Four key factors that contribute to variations were identified. Two could be attributed to different processes in practice: biomass type and processing. The other two related to limited information and limited scoping of the studied LCAs

To better study the effect of sourcing, the fourth and final study of this PhD was conducted, corresponding to chapter 5. The research question of this study was: *Under which biomass sourcing conditions does bio-based polyethylene result in a lower environmental impact than petrochemical-based equivalents?* In this study, 31 sourcing scenarios for bio-based high density polyethylene were compared, as well as five end-of-life scenarios. These scenarios encompassed 5 biomass types and 11 locations. An important predictor of the environmental impact was the concentration and direct availability of targeted molecules. In the production of bio-based chemicals, there is typically one type of molecule in a plant that is targeted. In polyethylene (the subject of this study), this was glucose, which can be fermented into ethanol. Glucose is present in plants in many forms of varying size and complexity. The crops with high concentrations of relatively simple sugars, such as sugar beet or sugarcane, resulted in relatively low environmental impact. Crops where the sugar was stored in longer molecules such as starch or cellulose resulted in a higher environmental impact, due to a combination of lower concentration of the target molecules and more excessive processing required to obtain sugars. The production location affected

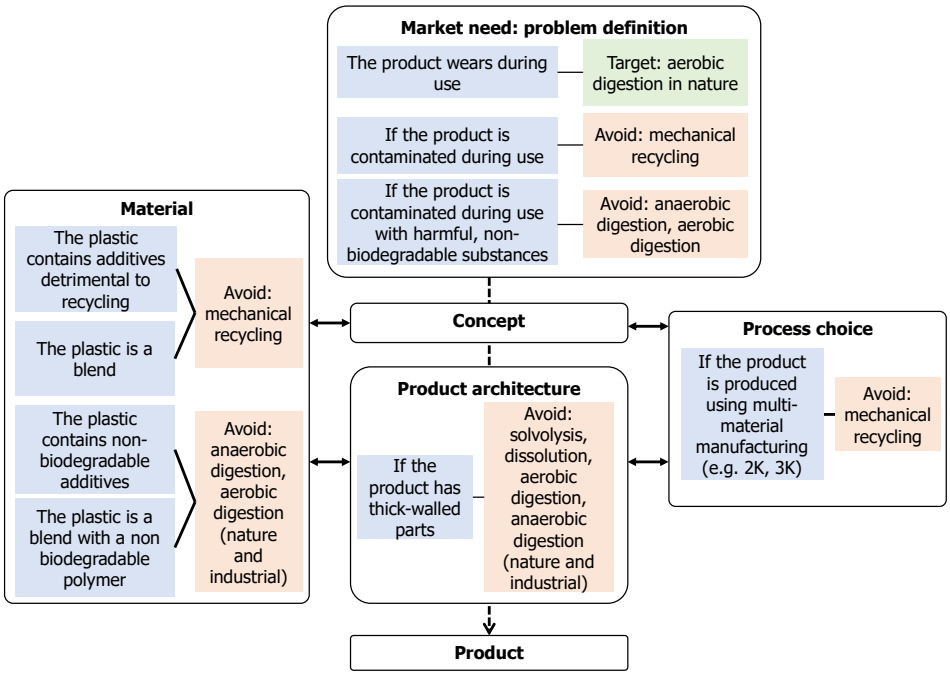

Figure 6.2: First iteration of the implementation of material-level recovery in a product design process using bio-based plastics. Figure adapted from Ashby et al. [1].

the resources needed for biomass cultivation, and the environmental impact of processing due to the energy mix.

Chapter 5 also assessed the effect of two biogenic carbon accounting strategies: accounting for carbon during the plastic production or upon molecular decomposition (in this case, incineration). In the study, accounting for biogenic carbon upon incineration led to results that were not in line with circular economy principles. Bio-based plastics could only 'benefit' from biogenic carbon if cradle-to-grave emissions were considered and the plastic was incinerated. This meant that in cradle-to-gate (i.e. only plastic production) CO_2-eq emissions, bio-based plastics were disadvantaged. Furthermore, incineration was the recovery pathway with the lowest CO_2-eq emissions, compared to e.g. mechanical recycling. This is counter-intuitive with circular economy principles, which prioritise keeping the plastic at its highest possible value. When biogenic carbon was accounted for during production, mechanical recycling resulted in the lowest CO_2-eq emissions. Therefore, biogenic carbon should be incorporated in the production stage of bio-based plastics LCAs.

6.3 Circular design with bio-based plastics

Bio-based plastics can address three important challenges for plastics: greenhouse gas emissions, fossil fuel consumption, and plastic pollution. The circularity of bio-based plastics depends on how they are produced and how they are applied. The sections below elaborate on how bio-based plastics can contribute to solving the environmental issues of plastics today, and the role of product designers.

6.3.1 Greenhouse gas emissions and environmental impact

Plastics were responsible for 4.5% of greenhouse gas emissions in 2015 [5]. However, in order to limit the mean global temperature increase below 1.5 °C, net-zero greenhouse gas emissions need to be reached in the second half of this century [6]. In order to reach climate change goals, greenhouse gas emissions from plastics must be mitigated. Two third of greenhouse gas emissions from plastics are released in the production phase, whereas one third is released upon their incineration [5]. Reducing the environmental impact of plastics should therefore address both the production of plastics and their incineration.

Bio-based plastics can yield reduced greenhouse gas emissions for plastics. However, the environmental impact due to the cultivation of biomass for bio-based plastics combined with the processing that is needed to produce monomers from this biomass that are then polymerised and compounded into plastics may exceed that of petrochemical-based plastic production. There are a wide variety of feedstock options for bio-based plastics, ranging from crops to by-products to algae [7–9]. Having many feedstock options was considered a major advantage of bio-based plastics in chapter 2, since they could have more stable and local supply chains. However, the environmental impact of transport was small (less than 10% of total impacts in most cases), hence minimising transport distance will not lead to significantly reduced greenhouse gas emissions. In chapter 4 and chapter 5, the type of biomass and cultivation location was found to have a major effect on the environmental impact of bio-based plastics. Some biomass types always yielded higher or lower greenhouse gas emissions than petrochemical-based plastic production, whereas for others, this depended on the cultivation location. The cultivation of specific crops has different environmental impact in different locations due to the local climate imposing different irrigation/fertilisation needs. Furthermore, agricultural practice also varies per location, for example machinal or manual harvesting or the use of specific pesticides. In the best-case scenario, production emissions from bio-based polyethylene could be up to 60% lower than those of petrochemical-based polyethylene, without accounting for biogenic carbon storage. If biogenic carbon would be accounted for, it could be up to 210% lower.

When 100% bio-based plastics are incinerated, the CO_2 produced is biogenic and their incineration can therefore be considered carbon-neutral. Nevertheless, incineration of bio-based plastics should be postponed whenever possible. A circular economy demands efficient resource use and recirculation at the highest possible value. Furthermore, incineration of bio-based plastics resulted in the highest environmental impact in chapter 5, compared to mechanical recycling and landfilling. Reducing virgin plastic plastic production and employing product lifetime extension strategies remain the most effective way to limit the environmental impact of bio-based plastics. Next, material-level recovery pathways that maintain the molecular structure of the plastic or produce monomers such

as mechanical or chemical recycling are also important because they avoid the production of virgin bio-based polymers or chemicals. Landfilling bio-based plastics is a more controversial topic. On the one hand, landfilling prevents reintroduction of biogenic CO_2 to the atmosphere. On the other hand, landfills are contested due to their effects on human health and the environment [10] and uncontrolled landfills can release methane into the atmosphere (which biodegradable plastics can contribute to). While landfilling bio-based plastics may be a more sustainable option in terms of CO_2-eq emissions, the other impacts on nature and human health mean that incineration should still be preferred.

The trade-off between the environmental impacts of landfilling and incineration already indicates that CO_2-eq emissions are not the only environmental impact that need to be considered for bio-based plastics. Bio-based plastics can make a positive contribution to this goal, but there are some environmental impact categories where bio-based plastics, by definition, have a higher environmental impact. Growing biomass for bio-based plastics will require land and water. Replacing all plastics with bio-based equivalents is expected to require significant amounts of biomass. Replacing all plastics produced in 2022 with bio-based equivalents would require 1% of all land on earth and 14% of the global annual freshwater use for crop lands [11–13]. However, the plastic economy is expected to have quadrupled by 2050. Extrapolating these numbers reveals that producing this quantity of bio-based plastics would require attributing 4% of all land on earth to bio-based plastic production. It would also require a 50% increase of freshwater use in agriculture. Agriculture will already need to grow substantially to feed an expected population of 9.7 billion [14]. This will put enormous stress on food production systems. Additionally, climate change will also affect food security and further stress agricultural systems [15]. Replacing all plastics with bio-based equivalents is not sustainable. Beside a switch to bio-based plastics with careful consideration of biomass sourcing, a circular economy for plastics will require a radical reduction in virgin plastic production.

Notably, this research primarily focused on first generation feedstocks, i.e. edible crops and wood (a second generation, non-edible, biomass type). If agricultural byproducts (as second generation feedstocks) would be assessed, the associated environmental impacts could be much lower. However, when this PhD research was conducted there was insufficient LCI data regarding the production of chemicals from agricultural byproducts to conduct comprehensive LCA studies. Furthermore, LCAs of agricultural byproducts come with multifunctionality. In the LCA of agricultural byproducts, the environmental impact is primarily attributed to the main product (i.e., the crop that is sold as food). There are several ways to distribute, or allocate, environmental impacts between main products and by-products. Common allocation methods in bio-based plastic LCAs are mass allocation, energy allocation, and economic allocation [2]. In mass allocation, the environmental impact is distributed according to the mass of the products. In energy allocation, the environmental impact is distributed according to the energy content of the products. In economic allocation, the most commonly reported allocation for bio-based plastics [2], the environmental impact is distributed according to the economic value of the products. For agricultural by-products, the economic value is currently low. However, when agricultural byproducts are used to produce high-value materials such as plastics, the economic value increases and so does the attributed environmental impact. The environmental impact of second generation bio-based plastics based on (for example) agricultural by-products could

still heavily depend on the biomass sourcing. However, in this case it may also depend on the economic value, comparative mass (crop compared to byproduct), or energy content of the type of crop.

Finally, it is important to note that the environmental impact of agriculture is highly variable. In chapter 5, the effect of biomass sourcing on the environmental impact of bio-based plastics was studied. Several biomass types and production locations were compared to show that biomass sourcing (both biomass type and production location) should be considered when producing bio-based plastics. In this study, only scenarios from different countries were compared. However, the food production system is highly heterogeneous: there are significant differences between individual farms growing the same crops in the same region [16]. Furthermore, fluctuations in climate may also cause the environmental impact of biomass production to vary per year or season [17]. The variations for biomass cultivation far exceed those of petrochemical production [18]. This implies that the environmental impact of bio-based plastics can vary per batch. In section 6.4 the implications of this variability on sustainable product design using LCA are elaborated.

6.3.2 Fossil fuel depletion

A common argument for a transition to bio-based plastics is that fossil resources are finite [19–21]. However, since the energy sector aims to rapidly transfer to renewable resources, there may be fossil reserves for petrochemicals for many years to come [22]. Plastic production consumes 8% of all fossil fuels produced today, a share that will grow to 20% if the plastic economy develops as predicted [23]. Moreover, the fossil fuel extraction has come with other types of environmental damage. Fossil fuel extraction is associated with biodiversity loss due to habitat destruction, visual and noise disturbance, and pollution [24]. Fossil fuel extraction also comes with the risk of oil spills, which have both short-term and long-term catastrophic effect on marine life [25]. It is therefore important to minimise fossil fuel use, also in the production of plastics.

In theory, bio-based plastics could be produced without any fossil fuels. However, as the results in chapter 5 demonstrated, the fossil depletion due to bio-based plastic production can exceed that of petrochemical-based plastic production. This was the result of the extensive use of fossil fuels in agriculture and energy production. Agricultural machinery still largely runs on fossil fuels [26], although they are expected to become electric in the future [27]. Furthermore, nitrogen and phosphorus fertilizers are currently produced exclusively using methane from natural gas [28]. Biogas production would need to grow by 360% in order to meet the demand for nitrogen fertilisers alone [28]. This makes decoupling agriculture from fossil fuels a major challenge, but not an insurmountable one. Agriculture with less or no synthetic fertilisers and pesticides could reduce demand [29], and greener methods for the production of fertilisers are also under development [30].

For product designers aiming to reduce the fossil fuel use of products by using bio-based plastics, the most important considerations are feedstock sourcing, product life extension and recovery. These are not different from those for reducing greenhouse gas emissions. In our analyses in chapter 5, outcomes for fossil depletion also correlated with CO_2-eq emissions because an energy mix more reliant on fossil fuels than renewables also has higher CO_2-eq emissions.

6.3.3 Plastic pollution

Plastic pollution is a ubiquitous problem. Plastic waste is found in oceans, rivers, soils, the atmosphere, animals and humans [31]. There are far reaching efforts to remove plastic waste from the environment and to prevent plastics from ending up there, but these are not expected to exceed the predicted growth in plastic waste [32]. It is a common misconception that bio-based plastics are inherently biodegradable [8]. However, many bio-based plastics are not biodegradable, and very few biodegrade in natural environments. Bio-based plastics do therefore not necessarily solve plastic pollution issues and it remains vital to fundamentally change both the material development and product design process, whether it is for petrochemical-based or bio-based plastics.

In order to reduce plastic pollution, recovery needs to be incorporated at the earliest stages of material development of and product design with both bio-based and petrochemical-based plastics [33, 34]. From a technical perspective, bio-based plastics do not necessarily behave differently from petrochemical-based plastics during recovery [35]. Much less is known about the recovery of bio-based plastics, because many bio-based polymers are relatively novel. Especially for dedicated bio-based plastics (for which no petrochemical-based counterpart exists), there is currently very little information available regarding recovery. Moreover, there is no infrastructure for recovery. This makes sustainable design that incorporates recovery with these materials very difficult.

Biodegradation of plastics in nature should be avoided for most products. Biodegradation in nature is complex and highly dependent on the natural environment; even different soil types can affect biodegradation behaviour [36], and incomplete biodegradation can result in microplastic formation. Furthermore, all value in the plastic is lost if it biodegrades in nature, whereas it could be captured or maintained through other recovery pathways. However, for some products it is inevitable that (micro)plastics end up in the environment. For instance, the wear of car tires is an important source of microplastics [37]. Microplastics from shoe soles have also been shown to have toxic effects on soils and plants [38]. Product design can play a vital role in reducing microplastic release from these products by using materials that biodegrade in nature.

6.4 A reflection on lifecycle assessment as a tool for sustainable product design with bio-based plastics

LCA is a powerful tool for sustainable product design. In product design, LCA can be used in numerous ways. For example, LCA can identify hotspots for environmental impact in products, which indicate an efficient way to reduce environmental impacts. Alternatively, LCA allows for a detailed analysis of a product and it can quantify the environmental impact of alternative designs [39, 40]. Product design often iterates on an existing product, which is ideal for a hotspot analysis or a detailed LCA. However, when a totally new product is being developed, detailed information is not available because the product does not exist yet. LCA is critical at this stage since decisions are made that have a major effect on a products environmental impact [41]. For some changes in the product, such as changing the plastic of a component to a bio-based alternative, detailed information about

this change may also not be available. In absence of exact information, the environmental impact of product design choices can be estimated using ex-ante LCA [42].

When product designers want to conduct an LCA to compare petrochemical-based and bio-based plastics for future products, they quickly run into issues. Participants in chapter 2 mentioned struggling with bio-based plastics LCAs, and chapter 4 further elaborated on these issues. Based on the experience that I have had with LCAs of bio-based plastics in the last years, I have found three shortcomings that make using LCA in sustainable product design with bio-based plastics challenging. Firstly, there is no generally agreed upon methodology for bio-based plastic LCAs. Secondly, there is very little high-quality lifecycle inventory (LCI) data available for the production of bio-based plastics. Thirdly, environmental impact agriculture is highly variable. Combined, these shortcomings may result in uncertainties that quickly become so large that they compromise the use of the LCA altogether [3]. Below, I elaborate on the three shortcomings, what they mean for circular product design practice, and how they could be overcome.

There are no widely accepted guidelines for how to conduct a bio-based plastic LCA. The poor comparability of bio-based plastic LCA outcomes have already been extensively discussed in this dissertation. This makes it difficult for product designers to compare alternative plastics directly, and requires a full LCA of each bio-based plastic alternative. Recently, the European Unions Joint Research Commission published guidelines for how to conduct bio-based plastic LCAs in order to make the outcomes more comparable [43]. These guidelines pose requirements for all step in an LCA, including system boundaries, data quality, allocation, and carbon sequestration accounting. However, the new guidelines have been met by considerable backlash from bio-based plastic producers [44]. They state that the guidelines result in an unfair treatment for bio-based plastics compared to petrochemical-based plastics, due to more stringent and specific data requirements. Additionally, the carbon sequestration in bio-based plastics may only be accounted for when the plastic returns to CO_2, for example upon incineration, instead of during the production of the plastic. The outcomes of chapter 5 indeed confirmed that this disadvantages bio-based plastics, and even circular strategies for bio-based plastics such as recycling.

If product designers conduct a full LCA of each bio-based plastic alternative, they are confronted with the second issue with bio-based plastic LCAs: low data availability. Poor data availability is a relatable issue for LCA practitioners [2, 39]. For many bio-based plastics produced today, there is no LCA data at all. The LCAs that are available for bio-based plastics are often not available publicly. In absence of high-quality LCA data, information is derived from secondary resources, at the expense of accuracy [45]. This data is often inconsistent, limited in scope, and comes with high uncertainties. In order to use LCA for product design with bio-based plastics, high-quality LCA data is required for all bio-based alternatives. While these outcomes are not yet comparable due to inconsistent LCA methodologies, these LCAs should preferably be transparently and publicly available.

The final shortcoming of LCA for sustainable design with bio-based plastics is how LCAs can deal with the inherent variability in the environmental impact of agriculture. The variability of the environmental impact of food and, by extension, biomass for plastics was already discussed in section 6.3. This inherent uncertainty poses a significant problem for sustainable product design: the plastic that is the most sustainable option today may

have a much higher environmental impact in the next batch. The variation due to inherent heterogeneity in agricultural systems could be incorporated in bio-based plastic LCAs as uncertainty. Including this uncertainty would require much more data regarding biomass cultivation. Furthermore, these uncertainties may render environmental impact comparisons inconclusive. In order to avoid this, producers could include environmental impact information for individual batches or seasons. Alternatively, they could use averages across multiple batches, which is less accurate but provides an indication for product designers looking to use the material for many years. The large variations also offer an opportunity for optimisation. By studying what conditions result in higher or lower environmental impacts, best practices for agriculture could be established.

While reliable data is still being developed and uncertainties remain high, LCA for bio-based plastics should mainly be used at a high level for decision making. LCA could be used to compare bio-based plastic options broadly, to incentivise the development of sustainable biomass sourcing scenarios and to highlight areas for improvement. Based on the observations from the research presented in this dissertation, a number of recommendations for a methodology for bio-based plastic LCA can be made.

- The scope of LCAs of bio-based plastic in products should be cradle-to-grave, preferably incorporating multiple use-cycles for recycled plastics. From a product-design perspective, it is important to clearly distinguish between the different phases of a lifecycle assessment of a bio-based plastic. Product designers should be able to compare bio-based plastics based on cradle-to-gate as well as cradle-to-grave environmental impact. In a circular economy, the recovery of a product or material needs to be considered during product design. In order to facilitate this, product designers need LCA data for the production of a bio-based plastic, as well as multiple recovery options.
- A standard set of environmental impact categories should be modelled, including at least: global warming potential, land-use, and water-use. This prevents differences in lifecycle impact assessment methods. It also prevents differences in LCA scoping. An example of this is that some of the studied LCAs in chapter 4 did not include water use in their analysis and corresponding data was missing from the LCIs.
  - Direct land-use change emissions need to be accounted for in bio-based plastic LCAs. In the analyses presented in chapter 5, direct land-use change did not contribute significant CO_2-eq emissions. Nevertheless, land use change is an important concern for bio-based plastics, similar to biofuels [46]. Furthermore, there are still developments in the modelling of land-use change emissions.
  - A standard for calculating indirect land-use emissions needs to be developed and incorporated in bio-based plastic LCAs. Indirect land-use change emissions may contribute significantly to the environmental impact of bio-based plastics [46–48]. Indirect land use is defined as '*changes in the use or management of land which is a consequence of direct land use change, but which occurs outside of the product system being addressed*' [49]. Models for calculating indirect land use emissions are under development, but are difficult to develop due to the complex nature of indirect land-use change [47].

- Biogenic carbon should be accounted for in the production stage for bio-based plastics. This incentivises value-retaining recovery pathways, which is in line with circular economy principles. Furthermore, it allows for a more fair comparison of bio-based plastics with petrochemical-based counterparts in cradle-to-gate assessments. This will also allow product designers to more rapidly compare bio-based plastics to petrochemical-based counterparts, without considering multiple recovery loops.

- There should be more stringent data requirements for bio-based plastic LCAs. The data used should be appropriate for LCA studies, and should be reported transparently. There should also be more strict requirements on the reporting of LCA data. Data reporting should be sufficiently transparent and detailed that the reported models are completely reproducable.

- LCAs of bio-based plastics need to integrate social sustainability. Bio-based plastics need to be both environmentally and socially sustainable. As discussed in section 6.3, bio-based plastics can have significant social effects, such as affecting food production systems. Furthermore, bio-based plastics are often produced in countries with weak legal conditions and poor working conditions [50]. Social lifecycle assessment (S-LCA) is a type of LCA that focuses on social impacts of products. Similar to LCA S-LCA can be used as a descision tool to compare options or to identify hotspots. However, compared to LCA, S-LCA is still considered relatively immature [51, 52]. There are already many S-LCA studies on biofuels, which could be extended to bio-based plastics [52].

6.5 Contributions

6.5.1 Contributions to science

The research presented in in this dissertation contributed to the development of the field of circular product design of products containing plastics. The contributions are listed below.

- An overview of drivers and barriers faced by actors throughout durable products' value chains (chapter 2). While there are already some resources for packaging design with bio-based plastics, there are no guidelines for the design of durable products. The overview of drivers and barriers for bio-based plastics was the first to target durable products. The identified barriers primarily related to a lack of knowledge and design tools. This presents further opportunities for material and design researchers to develop this knowledge.

- A clear and consistent terminology for the recovery of bio-based plastics in a circular economy, an overview of the suitability of specific bio-based plastic-recovery pathways, and recommendations for how recovery can be enabled through product design (chapter 3). Existing overviews focused on existing technologies. The review presented in this dissertation is the most elaborate to date, and includes information about existing as well as potential future recovery pathways for bio-based plastics. It is also the first to translate information about bio-based plastic recovery into concrete recommendations for product design.

- An exploration of the causes of environmental impact discrepancies in scientific bio-based plastic LCAs (chapter 4). Prior research on the environmental impact of bio-based plastics compared LCA outcomes to one another. The studies implied that a specific bio-based plastic should have a well-defined environmental impact. As a result, the differences between bio-based plastic LCA outcomes were mainly attributed to methodological inconsistencies and poor data availability. By studying the lifecycle inventories of bio-based plastic LCAs in depth, a new important cause for the discrepancies was identified: biomass sourcing. This shifts the interpretation of bio-based plastic LCAs from a defined impact for a specific plastic, to an impact that is also associated with a biomass sourcing scenario.

- An exploration of factors affecting the environmental impact of bio-based plastics in the most extensive comparison of biomass types, production locations and end-of-life options to date (chapter 5). The effect of biomass sourcing on the environmental impact of bio-based plastics was studied, but only with limited biomass sourcing scenarios. The research presented in chapter 5 encompasses a much wider variety of biomass sourcing scenarios: five biomass types, 11 production locations and five end-of-life options.

6.5.2 Contributions to practice

In addition to academic contributions, the work conducted in this PhD project also addressed practitioners. The circularity of bio-based plastics can be affected by many value chain actors, as well as policy makers. Below, the practical implications of the research are listed for specific fields of practice.

- Product design: a set of product design recommendations for design for recovery with bio-based plastics (chapter 3). Product design is vital for enabling the recovery of bio-based plastics, but the implications of this were never reported in scientific literature. The research presented in this dissertation presents the first set of product design recommendations for recovery of bio-based plastics (see figure 6.2).

- Material development: a set of recommendations for material composition based on the technical characteristics of existing as well as future recovery pathways (chapter 3). Existing research into the recovery of bio-based plastics considered the effect of material composition on the outcomes of recovery, but did not translate this back into recommendations for material development. In chapter 3, the effect of material composition on the ability to recovery is explicitly mentioned (see table 3.5).

- Waste management: an overview of potential recovery pathways for specific bio-based plastics. An insight into how recovery processes not yet in practical existence may affect the circularity of bio-based plastics and which recovery pathways may become important as the bio-based plastic market grows (chapter 3).

- Material development and product design: an insight into how biomass sourcing should be considered in the production and selection of bio-based plastics (chapter 5). Existing research has focused on comparing different bio-based plastics for products but has only considered one or a few biomass sourcing options. By studying a wider

variety of biomass sourcing scenarios, more generic conclusions about which factors related to biomass type and production location affect the environmental impact of bio-based plastics.

- An overview of potential future recovery pathways, and an improved insight into which factors affect the environmental impact of bio-based plastics. Recommendations that should improve the consistency of LCA outcomes and their comparability with petrochemical-based plastics (chapter 4, chapter 5, and section 6.4).

6.6 Recommendations for future work

This project was an exploration into the conditions under which bio-based plastics can be used in design for a circular economy. The research presented in this dissertation already mentioned a number of knowledge gaps that still need to be addressed.

- *Feasibility of various recovery pathways for bio-based plastics.*
  For many bio-based polymers, especially dedicated bio-based polymers, there is no research into material-level recovery pathways. Filling in these knowledge gaps is an obvious area for future research. Any cell labeled 'theoretically possible, but not researched yet' field in table 3.2 can be the topic of a new study. In particular, more research into biodegradation of (bio-based) biodegradable plastics in realistic natural conditions is needed. The soil composition affects biodegradation behaviour, but this effect has not been studied for most biodegradable polymers and in a wide variety of soil types [36, 53].

- *Environmental impact data for bio-based plastics: options for feedstocks and end-of-life.*
  LCA data for bio-based plastics remains very limited. Future research is needed to expand the LCA data for different bio-based plastics. This should not only include data for the production of various polymers, but also for manufacturing and end-of-life. When this data is generated and published, emphasis should be on transparent reporting.

- *Exploiting unique properties of bio-based plastics.*
  Bio-based plastics can have different properties compared to petrochemical-based plastics, which can enable them to be used in new ways or in new applications. At the time of writing, product design with bio-based plastics focuses mainly on marketing potential and perceived sustainability [54]. Exploiting the unique properties of dedicated bio-based plastics in product design presents a new way of interacting with these materials and can be used to identify new, value added applications.

- *How consumer behaviour affects the sustainability of bio-based and biodegradable plastics.*
  An important barrier encountered in chapter 2 was consumer education about bio-based and biodegradable plastics. Explaining what bio-based plastics are to consumers remains challenging [54, 55], and may result in harmful interactions with products containing bio-based plastics [56]. For example, in chapter 5, we assumed

that end-of-life of bio-based polyethylene would be identical to that of petrochemical-based polyethylene. However, the aforementioned consumer confusion about bio-based plastics may result in bio-based plastics ending up in the wrong recovery pathway. Future research should contribute to a better understanding of the consumer perception of bio-based plastics and how the concepts can best be conveyed in product design and marketing.

- *A tool for sustainable product design: an overview of bio-based plastic options and their properties.*
  In addition to consumers, value chain actors also reported a limited knowledge about bio-based plastics. This concerned an understanding of the concept of bio-based plastics, but they also identified that there is no consolidated resource about how to design durable products with bio-based plastics. In order to help product designers and other value chain actors with incorporating bio-based plastics, information could be consolidated into a method or tool. This could comprise of a tool for product design with bio-based plastics, including an overview of the bio-based plastic options, their properties, environmental impact (considering different biomass sourcing options), end-of-life options, and potential applications. Product designers should be included in the development of the tool in order to assure that it can meet their needs. Furthermore, the tool should be easily adaptable to future innovations, as the field of bio-based plastics is still rapidly developing.

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List of Publications

Journal publications

- *Ritzen, L.*, Sprecher, B., Bakker, C.A., & Balkenende, A.R., 2023. Bottlenecks in establishing the environmental impact of bio-based plastics: a case study of bio-based HDPE and bio-based PET. *Under Review.*
- Bos, P. *Ritzen, L.*, van Dam, S.S., Balkenende, A.R., Bakker, C.A., 2023. Bio-based plastics in product design: State of the art and challenges to overcome. *Under Review.*
- *Ritzen, L.*, Sprecher, B., Bakker, C.A., & Balkenende, A.R., 2023. Sustainability of bio-based polyethylene: the influence of biomass sourcing and end-of-life. *Under Review.*
- *Ritzen, L.*, Sprecher, B., Bakker, C.A., & Balkenende, A.R., 2023. Bio-Based Plastics in a Circular Economy: An Overview of Recovery Pathways and Implications for Product Design. Resources Conservation & Recycling, 199, p107268. doi: 10.1016/j.resconrec.2023.107268.
- *Ritzen, L.*, Montano, V., Garcia, S.J., Ritzen, L., Montano, V., & Garcia, S. J., 2021. 3D printing of a self-healing thermoplastic polyurethane through fdm: From polymer slab to mechanical assessment. Polymers, 13(2), 305. doi: 10.3390/polym13020305.
- Laroche, A., *Ritzen, L.*, Guillén, J.A.M., Vercillo, V., D'Acunzi, M., Sharifi Aghili, A., Hussong, J., Vollmer, D. & Bonaccurso, E., 2020. Durability of superamphiphobic polyester fabrics in simulated aerodynamic icing conditions. Coatings, 10(11), p.1058. doi: 10.3390/coatings10111058.

Conferences

- **Poster presentation** at *The European Bioplastics Conference (EBC2023)*, December 12 and 13, 2023, Berlin, Germany. Bos, P., *Ritzen, L.*, van Dam, S.S., Balkenende, A.R., Bakker, C.A., Design potential of bio-based plastics for durable products.
- **Oral presentation** at *11th International Conference on Industrial Ecology (ISIE2023)*, July 2-5, 2023, Leiden, the Netherlands. Title: Do bio-based plastics have a lower environmental impact than petrochemical-based plastics?
- **Conference paper and oral presentation** at *5th Conference on Product Lifetimes and the Environment (PLATE)*, May 29 - June 2, 2023, Espoo, Finland. *Ritzen, L.*, Bos, P., Brown, P., Balkenende, A.R., & Bakker, C.A., 2023. Drivers and barriers for bio-based plastics in durable products. PLATE 2023: the 5th Conference on Product Lifetimes and the Environment. Retrieved from: https://aaltodoc.aalto.fi/handle/123456789/122687
- **Oral presentation** at *8th International Conference on Self-Healing Materials*, June 20 - 26, 2022, Milan, Italy. Title: 3D Printing of a Self-Healing Thermoplastic Polyurethane through FDM.

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Biographical note

Linda Ritzen was on December 20th, 1993 in Roermond, the Netherlands. After finishing secondary school at B.C. Broekhin in Roermond, Linda pursued a degree in Aerospace Engineering at Delft Unversity of Technology. During her bachelor thesis, research into the opportunities that smart materials offer to improve the aerodynamic performance of commercial aircraft sparked her interest in materials. She continued her studies with a masters degree in aerospace engineering with a focus on materials. During this master, she spent 6 months on an internship in Airbus Central Research & Technology in Munich, working on aerodynamically efficient coatings. She conducted her masters thesis in the Novel Aerospace Materials (NovAM) research group, with the topic of 3D printing a self-healing thermoplastic polyurethane.

During her PhD in the Design for Sustainability (DfS) group in the faculty of Industrial Design Engineering, Linda expanded her knowledge of polymers to include recovery and environmental impact calculations through lifecycle assessment (LCA), as well as a product design perspective. During her PhD, she joined the University PhD Council to help disseminate the work of the council to the PhD candidates that it represents.