Bio-based and recycled polymers for cleaner production

An assessment of plastics and fibres

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Cover picture: Water drops condensed on a spider web. Spider web, made from one of the strongest polymers designed by nature, is 100% bio-based and biodegradable.
Picture taken in Portland, Oregon, USA
Photo credit: Jeff Williams-Gifford

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Bio-based and recycled polymers for cleaner production

An assessment of plastics and fibres

Duurzamere polymeren van biobased en gerecyclede grondstoffen

Een beoordeling van kunststoffen en vezels

(met een samenvatting in het Nederlands)

更清洁的生产：用生物质能和再生塑料生产聚合物

对塑料和纤维的环境评估

(附有中文总结论)

Proefschrift

ter verkrijging van de graad van doctor aan de Universiteit Utrecht op gezag
van de rector magnificus, prof. dr. J.C. Stoof, ingevolge het besluit van het
college voor promoties in het openbaar te verdedigen
op woensdag 16 maart 2011 des ochtends te 10.30 uur

door

Li Shen
沈莉

geboren op 12 mei 1977 te Nanjing, China

Promotor:	Prof. dr. E. Worrell
Co-promotor:	Dr. M.K. Patel

For my parents

献给我的父亲母亲

知之为知之，不知为不知，是知也。

Wisdom is recognizing what you know and what you don't. — Confucius

Contents

Chapter 1

Introduction

1.1 Historical outline

Plastics and fibres, like other polymer materials, had been used long before we started to understand their macromolecular structures. The original meaning of "plastic" is derived from Greek πλαστικος (plastikos), meaning *easy to process, shape and form.* Prior to the industrial revolution, animal horns and ivory were used to make many durable items. In the Middle ages, cow horns were heated and then shaped in a mould. By means of this type of processing, the thermoplastic properties of the material (animal horns) were made use of to prepare intarsia in wood [1]. Natural fibres, such as cotton, wool, silk and hemp, have been used as textile materials for thousands of years (see Figure 1.1 and Figure 1.2).

The industrial revolution in the 19^{th} century had a profound impact on our material consumption. In the 1870s, the invention of Celluloid, the first man-made thermoplastic, obsoleted ivory in most of its applications, e.g. jewelry and billiard balls. Two decades later, the viscose process was invented and viscose fibre was produced on large scale in the 1930s as an alternative to cotton and silk. Since then, viscose fibre has been one of the principle fibres. Before the use of petroleum was exploited, most of the technological innovations on plastics and fibres were focused on bio-based raw materials, among which, modified cellulose was, and still is, the most important (see Figure 1.2).

The petrochemical industry took off rapidly when large-scale oil extraction started in the early 20^{th} century. The transition from using bio-based feedstock to crude oil started. Many of today's important synthetic polymers were introduced between the 1930s and 1950s (see Figure 1.2). These "new" polymers, e.g. polyvinyl chloride (PVC), polystyrene (PS), polyethylene (PE), polypropylene (PP) and polyurethane (PUR) dramatically changed the history of plastics and our daily lives. Nylon (poly-amide, PA) and polyethylene terephthalate (PET) were the "new" choices for textile materials. In the same period, the technological development of bio-based materials halted. A well-known example is Henry Ford's research project which aimed at producing automobile parts out of soybeans. The project was interrupted by WWII and was never completed [3]. By the end of the 1960s, most of the early bio-based polymers had been replaced

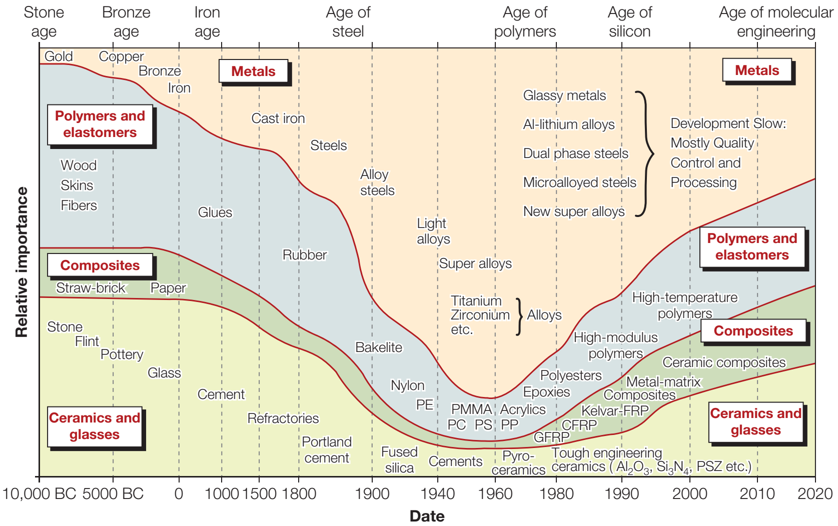
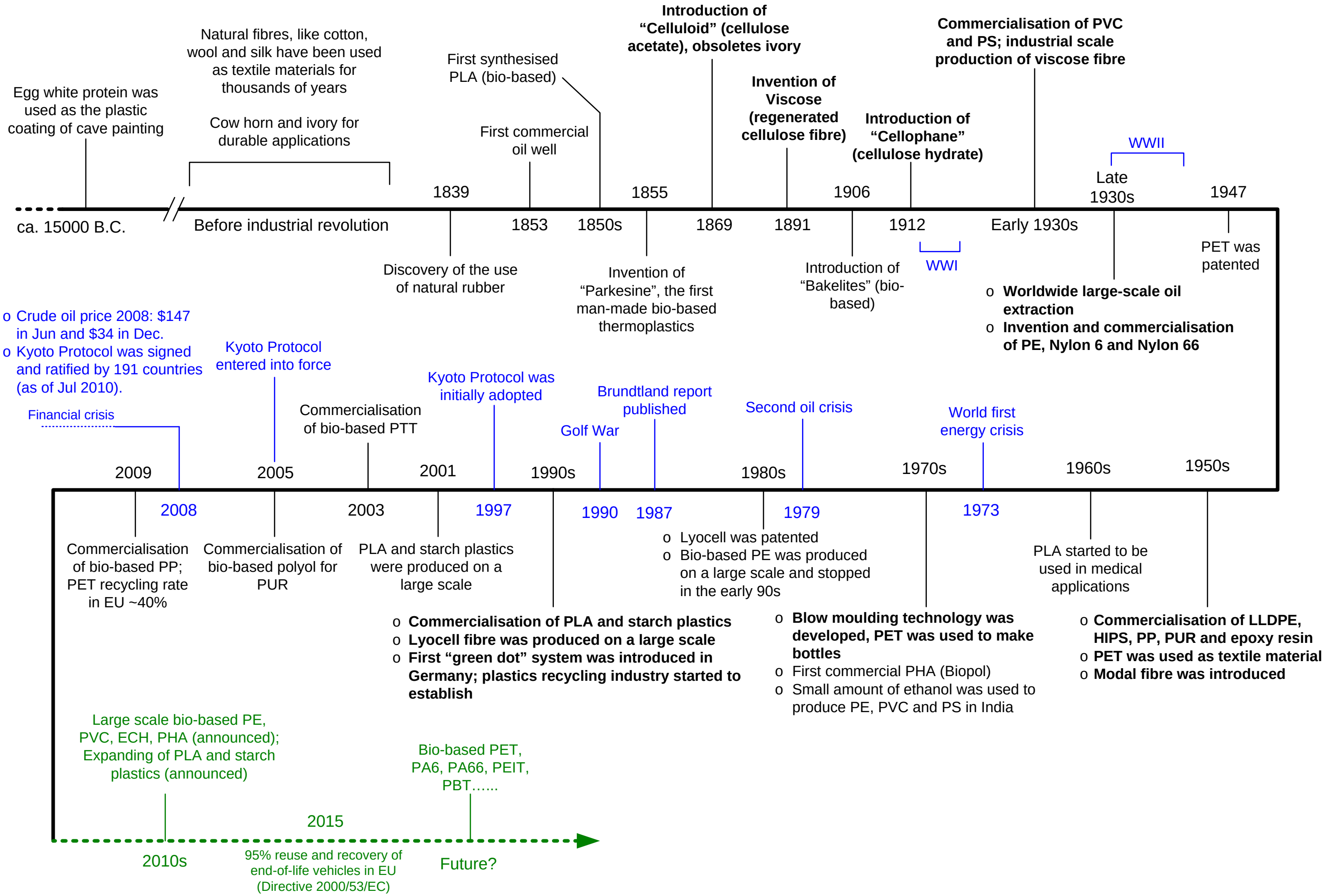


Figure 1.1: Relative importance of materials with time [2] (graph reproduced with permission from Elsevier).

by synthetic polymers. World plastics production increased from 1.5 million metric tonnes (Mt, or 10^6 metric tonnes) in 1950 to 30 Mt in 1970 [4] and world synthetic fibre production increased from 0.07 Mt to 4.8 Mt [5]. The transition from the use of carbohydrates to hydrocarbons as feedstocks for material production seemed completed [6]. The handful of bio-based survivors from this transition include wood pulp for paper-making, plant resin and vegetable oil used for coatings and man-made cellulose fibre (viscose).

The 1970s and 1980s were important for petrochemical plastics which experienced a fast growth: the global plastics production increased from 30 Mt in 1970, to 100 Mt in 1989 [4], although the plastics industry was still small compared to the paper industry with the global production of 230 Mt in 1989 [7]. PET, already in use as an important textile material, has increasingly also been used as a packaging plastic since the introduction of blow-moulding technology in the 1970s [8]. The first world energy crisis in 1973 caused public concern over the limited resource of oil. In 1987, the Brundtland report [9] proposed the concept of "sustainable development": *development that meets the needs of the present without compromising the ability of future generations to meet their own needs.* For the first time, sustainable development was formally introduced to the political agenda. In the 1970s and 1980s, despite the increase in the production of synthetic plastics, efforts were made to search for renewable alternatives [10]. For example, PHAs (polyhydroxyalkanoates) were introduced for the first time in the 1970s [11] and the cost of industrial enzymes dropped by 90% in this period [6]. The lyocell process was patented in the 1980s, representing an important technological innovation for man-made cellulose fibre. In Brazil in the 1980s, bio-based PE and PVC were produced (approx. 0.15 Mt per year, subsidised by the Brazilian government) [12].

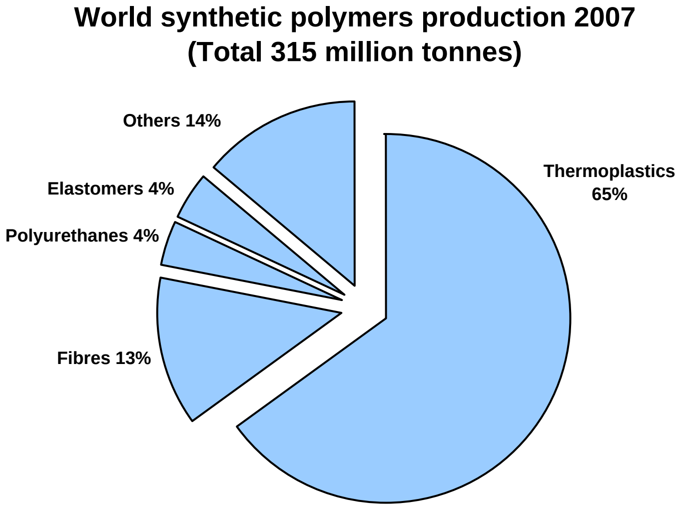


See the full names of the abbreviations in the text.

Figure 1.2: Historical outline of plastics and fibres.

The development of bio-based polymers continued in the 1990s, parallel to the growth of petroleum-based polymers. Both PLA (polylactic acid) and starch plastics were commercialised in this period. Meanwhile, world plastics production increased from 100 Mt in 1989, to 200 Mt in 2000 [13]. In the mid-1990s, the production of PET fibre exceeded that of cotton [14, 15]; since then, PET has been the most important fibre in the world to this day.

With the fast increase in the production and consumption of plastics and the increasing concerns over limited resources and plastics waste, recycling has been put on the political agenda. In the early 1990s, the first "Green Dot" system was introduced in Germany. A new industry, the plastics recycling industry, started to grow.



World synthetic polymers production 2007
(Total 315 million tonnes)

Thermoplastics: Standard plastics (PE, PP, PVC, PS, EPS, PET bottle grade) + Engineering Plastics (ABS, SAN, PA, PC, PBT, POM, PMMA, Blends, others inclu. high performance polymers)
Fibres: PA, Polyester, Acrylic and Other synthetic fibres
Others: Thermosets, Adhesives, Coatings, Sealants

Figure 1.3: World synthetic polymers production in 2007 [13].

At the beginning of the 21st century, the production of petroleum-based polymers continued to increase. Today, plastics and fibres are the two most important applications for synthetic polymer (see Figure 1.3). About 73% of the world synthetic polymers are used as thermoplastics, polyurethanes and elastomers, and 13% for fibres (see Figure 1.3). Meanwhile, bio-based polymers are also experiencing fast development. For instance, both PLA and starch plastics are now being produced on large scales. For more and more bio-based polymers, polymer precursors and monomers, industrial production has been demonstrated in pilot plants. Some of them are new polymers (e.g. PHA). Others are not entirely new, such as bio-based polyamides, bio-based polyols and bio-based epichlorohydrin (ECH), for which the bio-based routes were already investigated in the late 19th and the early 20th century. These bio-based polymers, polymer precursors and monomers were never produced on a large scale due to the transition from carbohydrates to hydrocarbons which started in the 1930s.

1.2 The sustainability challenges of plastics and fibres

1.2.1 Terms and definitions

Plastics and fibres, the subjects of this thesis, are both made from polymers. In this thesis, the term "man-made polymers" refer to both synthetic polymers and man-made bio-based polymers. The term "synthetic polymers" in this thesis strictly refers to petroleum-based polymers. "Plastics" is defined as the total of thermoplastics, thermosets and PUR (see Figure 1.4). The term "fibre" in this thesis is defined as man-made fibre, including both synthetic fibres, which are petroleum-based, and man-made fibres produced from renewable resources, such as cellulose fibres (e.g. viscose) and PLA fibre (see Figure 1.4).

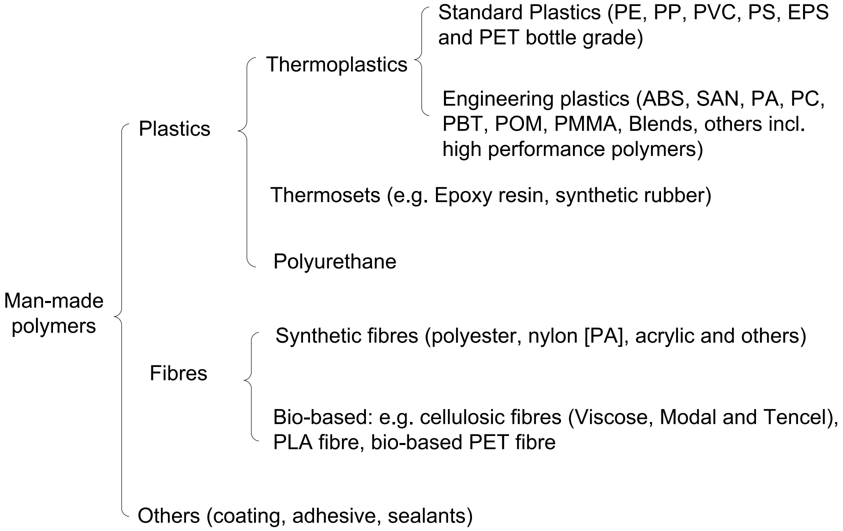
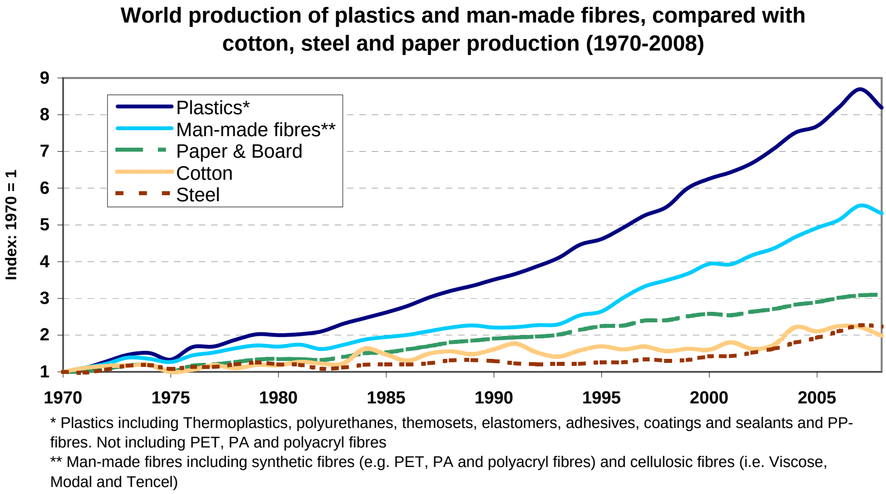


Figure 1.4: Chosen definition of the terms plastics and fibres in this thesis.

1.2.2 The sustainability challenges

The production of plastics and fibres has increased dramatically after WWII. Figure 1.5 shows that in the past 40 years plastics and fibres have outgrown their competing materials such as steel, paper and cotton. With population growth and the increase of wealth, the material consumption will continue to increase in the future. It is projected that the world polymer consumption will increase from 180 Mt in 2000 to 820 Mt in 2050, based on a "business-as-usual" scenario [16]. These demands need to be met sustainably. Petroleum, which took millions of years to form, is not a sustainable feedstock. Nearly all man-made plastics produced today are made from synthetic polymers. The polymer industry is the most important industry in the chemical sector; it accounts for approximately 60% of the sector's total primary energy consumption.[1] The chemical sector is one of the most energy intensive industrial sectors. Worldwide approximately 42 EJ of primary energy is used by the chemical sector (including feedstock energy),[2] representing about 9% of the total world primary energy consumption.[3] At the beginning of the 21st century, plastics and fibres have to face their first challenge: to meet the growing demand with non-renewable resources.


World production of plastics and man-made fibres, compared with cotton, steel and paper production (1970-2008)

* Plastics including Thermoplastics, polyurethanes, themosets, elastomers, adhesives, coatings and sealants and PP-fibres. Not including PET, PA and polyacryl fibres

** Man-made fibres including synthetic fibres (e.g. PET, PA and polyacryl fibres) and cellulosic fibres (i.e. Viscose, Modal and Tencel)

Figure 1.5: World production of plastics, man-made fibres, steel and paper (Index 1970 = 1). Based on statistics data from PlasticsEurope [13], CIRFS [5], IISI statistics [23] and FAO ForesStat [7].

The second challenge of plastics and fibres is the environmental impact of plastics waste management. Almost all synthetic polymers are non-degradable. In many countries, used plastics end up in landfills. The landfill space is limited. If not properly managed, landfill leachate can cause soil and water pollution. Moreover, the mismanagement of plastics waste can have severe impact on the environment. For example, the "Great Pacific Garbage Patch", the accumulation of non-degradable plastics waste in the North Pacific Ocean by gyre [24], has caused serious public concerns.

The third concern is related to climate change. In 2005, the chemical industry was the second largest GHG (greenhouse gas) emitter in the industry sector, accounting

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[1] Based on own estimate based on the chemical and polymer production volumes and energy statistics for 2006 [17, 18, 19] and cradle-to-factory gate primary energy consumption of major polymers produced [20, 21, 22].

[2] 1 EJ = 10^{18} Joule

[3] Estimated based on world energy balances for 2006 [17], assuming 35% power generation efficiency and 85% heat generation efficiency for conversion final energy to primary energy.

for about 4.1% of the global GHG emissions, following the cement industry (5%) and followed by the iron & steel industry (4.0%), aluminium/nonferrous industry (1.2%) and pulp & paper industry (1.1%) [25]. These environmental concerns bring the next challenge for plastics and fibres: to reduce the environmental impact of plastics waste and to combat climate change.

1.2.3 The solutions

There are two approaches to achieve the goal of sustainable development for material consumption [26]: dematerialisation (e.g. increase recycling) and transmaterialisation (e.g. shift to bio-based resources).

Dematerialisation requires efficient material production and consumption. Prevention and recycling are the current strategies proposed by the EU policy on the sustainable use of resources [27]. Both strategies require that the environmental impact is reduced by improving the material utilisation efficiency. Particularly, recycling re-introduces waste into the economic cycle in the form of valuable products. It not only provides a solution for plastics waste management, but also reduces the demand for non-renewable resources related to primary production [28, 29, 30, 31, 32].

Transmaterialisation seeks renewable alternatives to replace the current petroleum-based polymers. The concept of bio-based polymer is not entirely new. Before the industrial extraction of petroleum in the 1930s, most research activities were focused on bio-based chemicals/polymers (see Section 1.1). Today's sustainability challenge is calling for a new transition; a transition that can bring us from the hydrocarbon era to a new carbohydrate era. Bio-based material reduces our dependency on limited fossil fuels; it provides opportunities to produce compostable and/or degradable polymers, which avoids the use of non-degradable plastics [11, 29, 33, 34, 35]. At the same time, it is also possible to produce durable bio-based materials that are suitable for recycling.

1.3 Objectives, method and structure of this thesis

This thesis focuses on the two solutions to the sustainability challenges: bio-based alternatives and plastics recycling. The objective is to understand the environmental profiles of bio-based polymers and recycled polymers, in comparison with their petrochemical counterparts.

In the last few years, bio-based polymers have received much attention. Several studies have discussed the production technologies, future development and environmental impact of bio-based materials [11, 36, 37, 38, 39]. The past decades witnessed the commercialisation of many novel bio-based plastics, such as PLA, PHA, starch plastics, bio-based PTT and bio-based PE. The industry is responding to the call for the carbohydrate transition. However, so far, a thorough review on the current market volume and development status of this emerging industry does not exist. Chapter 2 answers the first research question: *what is the current status and what are the development potentials of the bio-based plastics industry?* In this chapter, the current market volume of the bioplastics industry is reviewed based on a company survey. We provide an overview (as complete as possible) of companies that have been active in the R&D, commercialisation and/or production of bio-based or biodegradable plastics in the past ten years. About 70 companies worldwide are interviewed. In the survey, information is collected based on three aspects: (i) the current (installed) capacity; (ii) the near future plans of capacity increase and (iii) technical substitution potential of bio-based products compared to their petrochemical counterparts. Chapter 2 aims at providing an overall picture where the bioplastics industry currently stands and how far the transition has proceeded from a hydrocarbon to a carbohydrate economy.

Chapter 3 and Chapter 4 answer the second research question: *What are the environmental impacts of bio-based polymers?* Chapter 3 provides a literature overview of environmental impact assessments of polysaccharide-based products. Polysaccharides (cellulose, starch and chitin) are the most abundant biomass feedstock [40]. Many environmental impact assessments were carried out for various bio-based products made from polysaccharides. In this review, not only the novel polysaccharide-based products are studied (e.g. starch polymers), but also conventional polysaccharide-based products that still have a very important position in the market (e.g. cotton). The review shows that little information was available on the environmental impact of modern man-made cellulose fibres. This leads to Chapter 4, where the environmental impact of three generations of man-made cellulose fibres (i.e. Viscose, Modal and Tencel) is analysed with the method of life cycle assessment (LCA). Chapter 4 focuses on the cradle-to-factory gate environmental impacts, including non-renewable energy use, renewable energy use and GHG emissions, land use, water use and the CML baseline environmental impact indicators (i.e. acidification, eutrophication, ozone layer depletion, photochemical oxidant formation, human toxicity, fresh water aquatic ecotoxicity and terrestrial ecotoxicity). The LCA results are compared with those for the conventional counterparts, i.e. cotton (bio-based) and polyester (petrochemical), at both mid-point level and end-point level (single-score analysis).

Chapters 5 and 6 focus on the environmental impact of recycled products and the LCA methodology for open-loop recycling. Parallel to the fast development of bio-based materials, the growth of the plastic recycling industry has been considerable in the past decade. For example, the amount of PET bottle waste collected increased from 0.4 Mt in 1998 to 1.3 Mt in 2008 in Europe [41]. Worldwide, approximately 4.5 Mt of PET bottles were collected and recycled into 3.6 Mt of flakes in 2007 [42]. Recycling of post-consumer PET bottles has become a well-established system with its own logistic chain including bottles collection, flake production and pellet production. Here, the first research question is: *what is the environmental impact of products made from recycled PET polymer?* We apply the method of LCA to answer this research question. During the analytical work, we found that there are very few case studies available in the public domain on open-loop recycling. The allocation issue in open-loop recycling has been widely discussed in literature. However, there is no consensus on the solution, because the simplified solutions are usually lacking justification; and the well justified solutions are usually demanding in terms of data and time. In the ISO standards, this issue has not been completely resolved yet. Neither are there clear policy guidelines on how the allocation should be done among several economic life cycles for the same physical material flow. This leads to the second research question on recycling: *How can LCA results be influenced by the choice of allocation methods applied to open-loop recycling?*

Chapter 5 provides a detailed LCA case study on PET bottle-to-fibre recycling by applying three methods for open-loop recycling. The LCI (life cycle inventory) data are based on real production data from three recycling companies covering mechanical recycling, semi-mechanical recycling and chemical recycling. The three allocation methods applied are the “cut-off”, “waste valuation” and “system expansion”. In this LCA, we compare the results across different methods and we discuss the implication of the choice of different methods.

Chapter 6 goes a step further on the topic of PET recycling by modelling a change-oriented LCA (or consequential LCA). In this case study, used PET bottles are recycled into both bottles and fibre, which are the two most important recycled PET products. In order to find the environmental optima of such a recycling system, four change-oriented effects are studied: the effect of multiple recycling trips, the effect of the shares of recycled PET used for bottle or fibre making, the effect of changing the market demand (changing the functional unit and thus, changing the reference system), and the effect of using bio-based PET polymer. The “system expansion” method, which implements life-cycle thinking, is applied to open-loop recycling.

Chapter 7 “synthesises” the findings from Chapters 3-5 by providing a comparison of the LCA results of polymer granulates, fibres and bottles made from bio-based PET, recycled PET, recycled bio-based PET and petrochemical PET, with the scope of cradle to grave without the use phase. A comparison among the bio-based, the recycled and the recycled bio-based has not been made so far. In Chapter 8, results are summarised and recommendations are put forward.

Chapter 2

Present and future development in plastics from biomass*

2.1 Introduction

Polymers abound in nature. Wood, leaves, fruit, seeds and animal furs all contain natural polymers. bio-based polymers have been used for food, furniture and clothing for thousands of years. Every year about 17×10^{10} metric tonnes biomass are produced by nature, of which only 3.5% are utilized by mankind [43]. Apart from wood used for conventional applications, for example energy, paper, furniture and construction, only a minor part of the total biomass is currently used for materials, for example clothing and chemicals.

The subject of this perspective is bio-based plastics. we define bio-based plastics as man-made or man-processed organic macromolecules derived from biological resources and used for plastic and fibre applications (without paper and board). Table 2.1 lists the main types of emerging bio-based plastics [44].

Bio-based plastics have a history of more than a century – much longer than petrochemical plastics. The first artificial thermoplastic – celluloid – was invented in the 1860s [45]. Since then, numerous inventions have been patented for new compounds and materials made from biological resources, such as ethylene produced by the dehydration of biobasedethanol in the 1940s [46]. However, many inventions made in the 1930s and 1940s remained in the laboratories and were never commercially exploited, due to the development of cheap, synthetic polymers from crude oil in the 1950s. The petrochemical industry has since taken off and plastics have become a daily necessity.

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*Published in *Biofuels, Bioproducts & Biorefining* (2010) **4**:1, 25-40. DOI: 10.1002/bbb. Co-authors: Ernst Worrell and Martin K. Patel (Utrecht University). Reproduced with permission from John Wiley and Sons.

In Western Europe in 2007, 43% of all plastics are used for packaging, 21% are used in building and construction, 8% for automobiles, 5% for electrical and electronic appliances, and the remaining 23% are used for various other applications [13]. The vast majority of the polymers used are polyolefins, i.e. polyethylene (PE) and polypropylene (PP) which together represent 54%, followed by polyvinyl chloride (PVC, 14%), which dominates in building and construction, and polyethylene terephthalate (PET, 8%) [13].

bio-based plastics have experienced a renaissance in the last few decades. Many new polymers from bio-based feedstocks were developed, for example polylactic acid (PLA) from sugars. One of the earlier drivers was to provide the market with biodegradable plastics in order to solve the problem of increasing amounts of waste and limited landfill capacity. Today, public concerns about the environment, climate change and limited fossil fuel resources have become more important drivers.

This perspective provides an overview of the market, material properties, applications, the technical substitution potential of emerging bio-based plastics between 2003 and 2007, and the projections of the future market until 2020. An extensive description of all polymers listed in Table 2.1 can be found in Shen et al. [44]. In this perspective, three bio-based plastics, namely PLA, bio-based PE and bio-based epoxy resins, are highlighted for their recent developments.

2.2 Market of bio-based plastics

2.2.1 Current market volumes

Before discussing the market volume of emerging bio-based plastics, it is worthwhile to understand the market size of traditional non-food bio-based polymers and materials. These include paper and board, man-made cellulosics (fibres and modified cellulose), non-food starch and alkyd resins. These bio-based polymers and materials have been produced and consumed in large quantities for long time. So far, the paper industry has been the largest non-food bio-based material producer in the world. The global paper and board production was approximately 365 million metric tonnes (Mt) in 2006 [47]. Together, non-food starch (without starch used for fuel ethanol[1]), man-made cellulose polymer[2] and alkyd resin account for approximately 20 Mt per year, of which nonfood starch takes the lion's share (75%, without starch used for fuel ethanol), followed by man-made cellulose polymers (20%, without paper) and alkyd resin (5%).[3]

Compared to the traditional non-food bio-based polymer and material market, the emerging bio-based plastics market is relatively small. The global capacity in 2007

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[1] In Europe starch used for fuel ethanol was approximately 1.9 Mt in 2007 [44].

[2] Including cellulose esters (e.g. cellulose acetate), cellulose ethers (e.g. carboxymethyl cellulose) and regenerated cellulose (e.g. viscose).

[3] The information on current capacity of the emerging bio-based plastics was collected from 30 companies who produced at least at pilot scale in 2007. These 30 companies are the current major players of starch plastics (8 companies), cellulose films (2), PLA (7), PTT (1), bio-based PA (1), PHA (6), bio-based PE (1), bio-based epichlorohydrin (1) and bio-based PUR (3). The capacity information for 2007 was collected by means of a questionnaire, via personal communication with companies and industry associations and through publicly available company announcements.

Table 2.1: Main emerging bio-based plastics for non-food applications [44].

No	**Bio-based plastics (group)**	**Type of polymer**	**Types/Structure/Production Method**
1	Starch plastics	Polysaccharides	Partially fermented starch; Thermoplastic starch (TPS); Chemically modified starch (e.g. starch acetate); Starch blends; Starch composites
2	Cellulose polymers	Polysaccharides	Organic cellulose esters; Regenerated cellulose
3	Polylactide (PLA)	Polyester	Bio-based monomer (latide) by fermentation, followed by polymerisation
4	Polytrimethylene terephthalate (PTT)	Polyester	Bio-based 1,3-propanediol (1,3-PDO) by fermentation plus petrochemical terephthalic acid (or DMT)
5	Polyamides		
	a. PA11	Polyamide	Bio-based monomer 11-aminoundecanoic acid from castor oil
	b. PA610		Monomer sebacic acid from castor oil
	c. PA6		Bio-based monomer caprolactam by fermentation of sugar
	d. PA66		Bio-based adipic acid by fermentation
	e. PA69		Bio-based monomer obtained from oleic acid via azelaic (di)acid
6	Polyhydroxyalkanoates (PHAs)	Polyester	Direct production of PHA by fermentation
7	Polyethylene (PE)	Polyolefin	Bio-based monomer (ethylene) obtained from ethanol which is produced by fermentation of sugar
8	Polyvinylchoride (PVC)	polyvinyl	Monomer vinyl chloride can be obtained from bio-based ehthylene (from ethanol)
9	Other thermoplastics[a]		
	a. Other polyesters (PBT, PBS, PBSL, PBSA, PBST, PBAT, PET, PEIT, PVAc, Polyacrylates, PTN, PTI, thermoplastic elastomers)	Polyester	Various carboxylic acids, various alcohols
	b. Other ethylene-based compounds (e.g. polystyrene and EPDM rubber)	Various	Ethylene by dehydration of bio-ethanol, reacted with other compounds
	c. Methanol-based compounds (e.g. phenolic resins, urea formaldehyde resins, melamine formaldehyde resins)	Various	Syngas by gasification of biomass, and synthesis of methanol, reacted with other compounds
	d. Propylene-based compounds (e.g. PP, polyacrylates, PUR, PA)	Various	Thermochemical propylene production via bionaphtha plus steamcracking or via biomethanol, followed by a methanol-to-olefins process and polymerisation
10	Polyurethanes (PUR)	Polyurethane	Bio-based polyol from vegetable oils, plus petrochemical isocyanate
11	Thermosets	Cross-linked polymers	
	a. Epoxy resins	Epoxy resin	Diglycidyl ether of bisphenol A derived from bisphenol A and epichlorohydrin (ECH); ECH produced by glycerine-to-ECH (GTE) process; glycerine is a by-product of bio-diesel production
	b. Epoxidised vegetable oils	Epoxide	Addition of oxygen to alkenes
	c. Thermosets based on 1,2-PDO and 1,3-PDO	Unsaturated polyester	Polycondensation of unsaturated and saturated dicarboxylic acids with diols
	Alkyd resins	Alkyd resin	Condensation polymerisation of polyols, organic acids and fatty acids or triglyceride oils

[a] Abbreviations: DMT = Dimethyl terephthalate; PBT=polybutylene terephthalate; PBS=polybutylene succinate; PBSL=polybutylene succinate-co-lactate; PBAT=polybutylene adipate-co-butylene terephthalate; PET=polyethylene terephthalate; PEIT=polyethylene-co-isosorbite terephthalate; PVAc=polyvinyl acetate; PTN=polytrimethylene naphthalate; PTI=polytrimethylene isophthalate; EPDM=ethylene propylene diene M-class rubber; PP=polypropylene.

was estimated at 0.36 Mt, equivalent to only 0.1% of the world's paper and board production and 0.2% of the global petrochemical plastics production. However, the emerging bio-based plastics market is growing rapidly. Between 2003 and 2007, the average annual growth rate was close to 40% worldwide (Figure 2.1) and nearly 50% in Europe.[4]

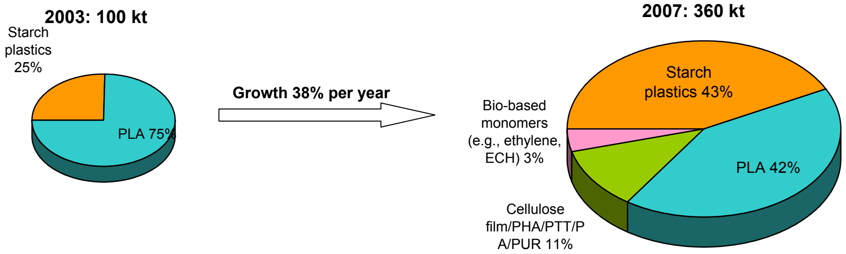

Figure 2.1: Global production capacities of emerging bio-based plastics in 2003 [11] and 2007.

The developments in the past five years in emerging bio-based plastics are spectacular from a technological point of view. Starch plastics and PLA have been the frontrunners in the renaissance of bio-based plastics. They were the only bio-based plastics produced on a large scale in 2003 [11] (Figure 2.1). Today, starch plastics and PLA are still the most important bio-based plastics in the market, but other types of bio-based plastics will soon be produced on a large scale, such as bio-based polyethylene (PE) and bio-based epoxy resin (made from bio-based ethylene and epichlorohydrin, respectively).

Recent technological breakthroughs have allowed substantial improvement in the material properties of novel bio-based plastics, such as heat-resistance (e.g. for PLA) and tensile strength (e.g. for PHA). This makes bio-based plastics eligible for a wider variety of applications. Moreover, many old processes have been revisited, with ethanol dehydration probably the most prominent one. Many first-of-its-kind plants are being (or will be) constructed. Hence, we are at the very beginning of the learning curve for bio-based plastics. Most of the plant capacities are still small (e.g. the capacity of Tianan's PHA plant is only 2 kt (metric kilotonnes) [48]), but others are very sizable (e.g. Braskem's bio-based PE plant will be 200 kt [49]). With a growing demand for bio-based plastics, it can be expected that turn-key plants with large capacities will be commercially available within a few years, thus allowing accelerated growth.

2.2.2 Technical substitution potential

Table 2.2 provides an overview of the typical physical, mechanical and thermal properties of emerging bio-based plastics. Note that there are no differences in terms of material properties between bio-based plastics and their petrochemical counterparts which have identical chemical structures (e.g. PE).

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[4] The capacity information for 2003 was obtained from Crank et al. [11] and updated by personal communication with companies.

Table 2.2: Typical physical and thermal properties, renewable contents, biodegradability and commercialisation stages of emerging bio-based plastics[j].

	Density (g/cm^3)	**Tensile strength (MPa)**	**Young's modulus (GPa)**	**Flexural modulus (GPa)**	**T_m(°C)**	**T_g(°C)**	**Renewable content (wt%)**	**Biode-gradable**	**Development phase, end of 2008**
Starch plastics [44]	1.20-1.35	20-40	1.0-1.7	-[h]	˜60	-	40-100%	Yes&No[a]	Large scale
PLA [50, 51][b]	1.24	53-100	-	3.83	140, 220	55 to 70	100%	Yes	Large scale
PHA [52][c]	1.17-1.25	18-40	1.2-3.0	-	50-190	-13 to 4	100%	Yes	Pilot scale
Cellulose film	1.45 [53]	70-125 [54]	-	-	-	-	100%	Yes	Medium scale
Bio-based PTT [55][d]	1.35	66	-	2.7	228	45 to 55	35%	No	Small scale
Bio-based PET [56][d]	1.31	52	2.7	-	220-225	75	˜30%	No	Research
Bio-based LDPE [53][d]	0.92	10-18	0.15-0.20	-	105-130	-30	100%	No	Pilot
Bio-based PP [57][d,e]	0.91	33-35	-	1.45-1.55	160-170	-13 to 0[f]; -18 to -15[g]	100%	No	Pilot
PA11[58, 59]	1.05	57	-	1.17	180-189	45	100%	No	Medium scale
PA610 [60]	1.08	55	-	2.00	225	-	˜60%	No	Small scale
Bio-based PA66 [58][d]	1.14	83	-	2.83	269	65 to 85	100%	No	Research
Bio-based PA6 [58][d]	1.14	81	-	2.70	228	65 to 75	100%	No	Research
Bio-based PUR	-	-	-	-	-	-	8-90% [44]	No	Small scale
Bio-based LER[d, j]	-	-	-	-	-	-	30%	No	Small scale

[a] The biodegradability of starch plastics depends on the copolymer blended with thermoplastic starch (TPS). Starch plastics are only biodegradable if the copolymer is also biodegradable.

[b] The thermal property of PLA is for amorphous, semi-crystalline and crystalline PLA.

[c] PHAs here include P(3HB), P(3HB-co-HV), P(3HB-co-HHx) and P(3HB-co-HA).

[d] These bio-based plastics are chemically identical with their petrochemical counterparts.Therefore, the physical and thermal properties are also identical.

[e] Refers to PP homopolymer

[f] For isotactic PP

[g] For atactic PP

[h] Not applicable or not available

[i] See the full names of the abbreviations in Table 2.1 .

[j] LER = Liquid Epoxy Resin.

Table 2.3: Technical substitution potential of bio-based plastics (plastic applications including thermoplastics and thermosets, excluding fibres).

% substitution[a]	LDPE	HDPE	PP	PVC	PS[b]	PET	PUR	PA	ABS[c]	PC	PBT	PMMA	Other Polyacrylates	Epoxy resin	Synth. rubber	Other
Starch plastics	8	8	8		8		8					4				
PLA		10	10		10	20		10				5				
PHA	20	20	10	10	20	10	10		10			5				
Cellulose films			10	10	10	15										
Bio-based PE	72	62														
Bio-based PP			57													
Bio-based PVC[d]				80												
Bio-based PET[d]						35										
Bio-based PTT[d]			5			20		30		20	100	5				
Bio-based PUR[d]							80									
Bio-based PA[d]								30								
Bio-based polyacrylates[d]													100			
Bio-based epoxy resin[d]														75		
Bio-based ABS[d]									90							
Bio-based PB[d]															80	
Sum percentages	**100**	**100**	**100**	**100**	**48**	**100**	**98**	**70**	**100**	**20**	**100**	**19**	**100**	**75**	**80**	**0**

Unit: 1000t	LDPE	HDPE	PP	PVC	PS[b]	PET	PUR	PA	ABS[c]	PC	PBT	PMMA	Other Polyacrylates	Epoxy resin	Synth. rubber	Other	Total	%subst.
2007 Global consumption[e]	37100	30700	44900	35280	16105	15498	12285	2730	7455	3150	954	1400	660	1150	10889	6930	227186	100
Technically replaceable volumes	37100	30700	44900	35280	7730	15498	12039	1911	7455	630	954	266	660	863	8711	0	204698	90

Note: See abbreviations in Table 2.1.

[a] The values on substitution potential (in the upper table) were established based on interviews with industrial experts (see text).

[b] PS (all types) and EPS.

[c] ABS/SAN, including also other styrene copolymers.

[d] Partially bio-based polymer.

[e] For PE, PP, PVC, PS, PUR, ABS, PA, PC and PBT, data are for 2007 based on the projection of Kunststoffe [61]. The PET data is also projected for 2007 but based on the data for 2006 from PlasticsEurope [62] and annual growth projection according to Kunststoffe [61]. For PMMA, the consumption data is for 2006 [61]; no projection for 2007 is available. For other polyacrylates, data are for 2003 [63]. For epoxy resin and synthetic rubber, consumption data are for 2000 [64, 65].

In order to understand to what extent bio-based plastics could replace petrochemical plastics from a technical point of view, we estimated the maximum technical substitution potential. This was done by interviewing industrial experts based on the material properties and applications of bio-based plastics. The outcome of the interview is shown in Table 2.3 (for plastics applications) and Table 2.4 (for fibres) in *% substitution*. Complete substitution (100%) is achieved when petrochemical plastics are replaced by chemically identical bio-based plastics (e.g. PE). In all other cases, the substitution percentages are lower than 100% because petrochemical plastics can only be replaced partially by fully bio-based or partially bio-based plastics. Depending on the type of plastic, between 20% and 100% of the current volume could be in principle replaced by bio-based plastics. The substitution potentials of 8 out of the 16 plastics are 100% (Table 2.3).

Table 2.4: Technical substitution potential of bio-based man-made fibres (both staple fibres and filament).

%Substitution[a]	PET	PA	Acrylic	Other synthetic	Cellulosic
PLA	10	0	5	0	5
PTT	20	20	5	0	5
PHA	5	0	5	0	5
Bio-based PET	65	0	0	0	0
Bio-based PA6, PA66	0	80	0	0	0
Sum Percentages	**100**	**100**	**15**	**0**	**15**

(Unit: 1000 t)	PET	PA	Acrylic	Other synthetic	Cellulosic	Total	%subst.
2007 World fibre consumption [66]	30804	3836	2407	575	3081	40703	100
Technically replaceable volumes	30804	3836	361	0	462	35463	87

Note: See abbreviations in Table 2.1.

[a] The values on substitution potential (in the upper table) were established based on interviews with industrial experts (see text).

By multiplying the substitution percentage by the consumption volume of the respective petrochemical polymer in 2007, volume estimates are obtained for each bio-based plastic. Thus, the overall maximum technical substitution potential for plastic applications[5] is estimated at 205 Mt, which corresponds to 90% of the total global plastic consumption in 2007. For fibres (Table 2.4), the substitution potential is estimated at 35 Mt, or 87% of the world's fibre consumption in 2007. Therefore, **the total maximum technical substitution potential of bio-based plastics and fibres** replacing their petrochemical counterparts is estimated at 240 **Mt, or 90% of the total consumption of plastics and fibres in 2007**. This substitution potential is purely based on the technical properties of the bio-based plastics. It does not account for resource availability and economic viability. As a note of caution, the substitution potential of 90% should be regarded as indicative because it has not yet

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[5] Important plastic applications are injection moulding, blow moulding, extrusion and foaming; in contrast, the use of polymers for coatings and fibres is usually excluded.

been proven that the large-scale production of all bio-based plastics (including fibres) shown in Table 2.3 and Table 2.4 is technically feasible.

2.2.3 Market projections of bio-based plastics

The market projections of bio-based plastics for 2020 were carried out as follows:

1. The companies' expected production capacity was collected by a questionnaire, via personal communication with companies and with the industry associations *European Bioplastics* and *Japan Bioplastics Association*, and through publicly available company announcements. About 70 companies worldwide were investigated in our market projection, including both current producers and the future producers of bio-based plastics. The current activities range from lab scale to large, commercial scale.

   The questionnaire was sent to all the member companies of the *European Bioplastics* association. Seven out of about 50 companies replied. About 25 companies' data were collected via personal communication or communication with industry associations. Company capacity data which could not be obtained by the questionnaire or personal communication was collected from company announcements in the public domain. The companies were requested to provide projections of their planned capacity expansion. The companies' views were collected in the first half of 2008 and once more in March 2009 in order to account for the economic crisis.

   Our questions primarily focused on the period 2007–2020. The projections for 2009–2013 are based on concrete plans, which are currently being implemented; in contrast, the statements made for the year 2020 have more of a visionary character. The projection prepared on this basis is referred to as 'projection based on company announcements'.

   The result of Step 1 was compared to the technical market potentials of bio-based plastics and fibres.

2. In the survey, we also asked companies to provide their expectations of the growth rate of the bio-based plastics sector as a whole, for the next 10–20 years. We then used the average growth rate to derive the projection for 2020, which we refer to as the 'projection based on industry expectations'.

3. Three scenarios (BAU, HIGH and LOW) were constructed in the third step not only taking into account the companies' announcements and their expectations but also considering technical barriers, the estimated market size for bulk applications, cost competitiveness and the raw material availability for the production of bio-based plastics until 2020. These scenarios are referred to as 'PRO-BIP 2009 Scenarios'.[6]

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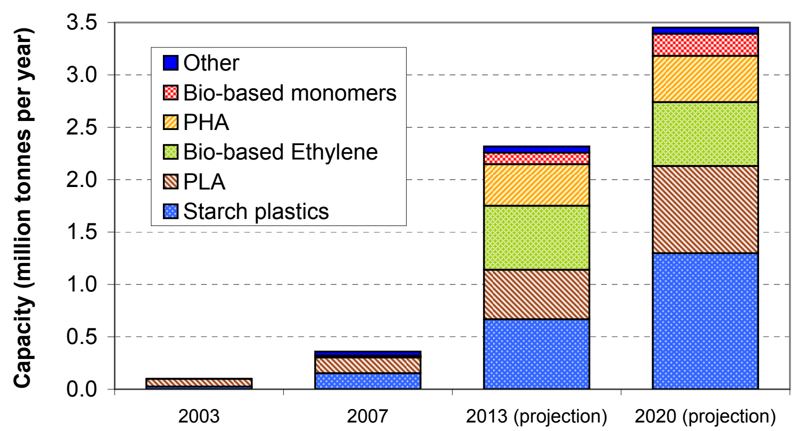
[6] PRO-BIP is the short name of the project 'Product overview and market projection of emerging bio-based plastics'. See Shen et al. [44]

4. Finally, the outcome from Steps 1, 2 and 3 was compared to the projections prepared in the earlier study by Crank et al. [11].

Projection based on company announcements (Step 1)

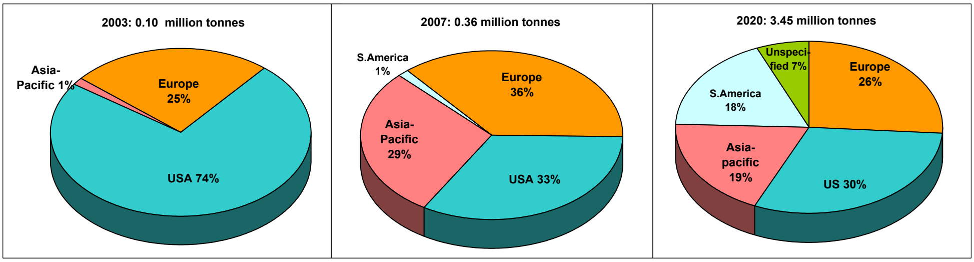
According to company announcements, the worldwide capacity of bio-based plastics is expected to increase from 0.36 Mt in 2007 to 2.32 Mt in 2013 and to 3.45 Mt in 2020 (Figure 2.2). The announced capacities are (ordered by size): starch plastics (1.30 Mt), PLA (0.83 Mt), bio-based ethylene (0.61Mt), PHA (0.44 Mt), bio-based epichlorohydrin (ECH, 0.21 Mt) and other bio-based plastics such as bio-based PTT, PA 11, PA 610 and bio-based PUR (total approximately 0.06 Mt). Based on these announcements, the capacity breakdown can be presented by regions over time. As shown in Figuire 2.3, the leading position of the USA and Europe in the years 2003 and 2007 changes to a more balanced regional distribution by the year 2020.



Note: Category "other" includes cellulose films, PTT from bio-based 1,3-PDO,bio-based polyamide and PUR from bio-based polyols; category "Bio-based monomers" includes primarily bio-based epichlorohydrin.

Figure 2.2: Worldwide capacities of bio-based plastics until 2020 based on company announcements (the most recent data used for making this graph were received in March 2009; the reported values refer to the capacities at the end of each year).

During the preparation of this study, the world economy experienced a dramatic downturn, with very serious decreases in demand (in 2008/2009). As a consequence, the oil prices dropped from \$130/barrel in July 2008 to \$40/barrel in December 2008, followed by an oil price level of \$40–60/barrel in the first half of 2009. Most interviewed companies are still optimistic about their long-term plans (we re-contacted the major players in March 2009). Some companies have, however, delayed their expansion plans. For instance, Dow announced that its bio-PE project will be delayed to 2012 [67] and Telles postponed the start-up of its 50 kt PHA plant from end 2008 to the second



Note: "Unspecified region" in the right figure means that there are no concrete plans about the location of these capacities.

Figure 2.3: Breakdown of worldwide capacity of bio-based plastics by region, projection of 2020 is based on company announcements (the most recent data used for making this graph were received in March 2009; the reported values refer to the capacities at the end of each year).

quarter of 2009 [68]. However, it is not clear whether these delays are the consequence of the global economic crisis or whether there are other reasons. As described earlier, the maximum technical substitution potential of bio-based plastics is 240 Mt. The worldwide capacity of bio-based plastics in 2007, i.e. 360 kt, was only 0.15% of this potential market; and even the projected capacity in 2020 will only meet approximately 1.5% of the technical potential market (Figure 2.4).

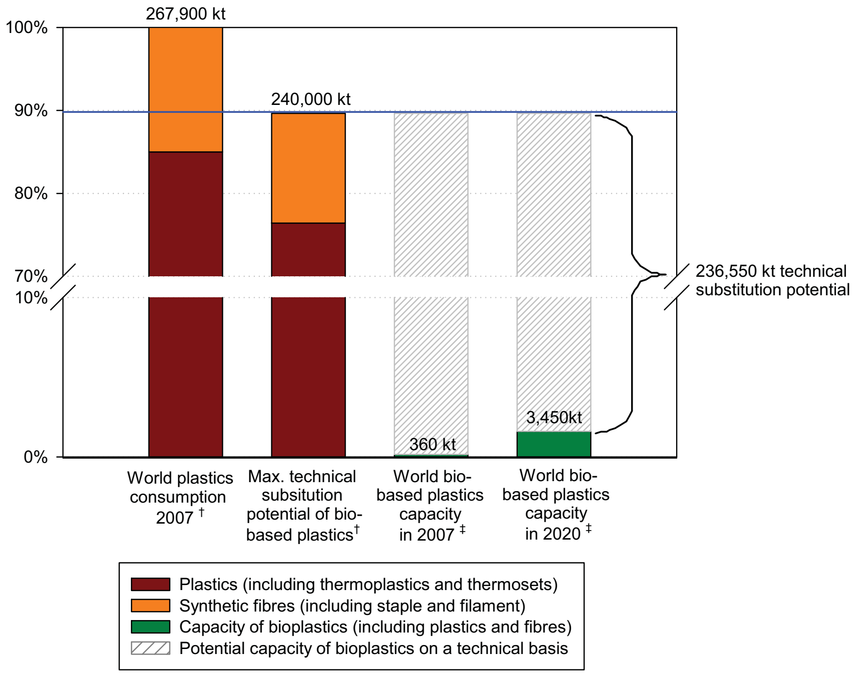
Projection based on bio-based plastics industry expectations (Step 2)

Our company survey also included the question about the development of the bio-based plastics sector as a whole (Step 2). According to this survey, the bio-based plastics industry expects that production will grow on average by 19% per year during 2007 and 2020. The resulting projection in 2020 (3.44 Mt) is very close to the projection for 2020 from Step 1, which is based on company announcements (3.45 Mt).

PRO-BIP 2009 scenarios: BAU, HIGH and LOW (Step 3)

The projections based on the company announcements are not necessarily consistent across the various types of bio-based plastics. For example, starch plastics producers are rather optimistic about their future development, in comparison with PLA producers. For this reason, independent projections have been prepared. Three scenarios were built for 2020 (Step 3): business-as-usual (BAU), HIGH and LOW. These scenarios were designed considering four major influencing factors, namely technical barriers, bulk applications, cost competitiveness and raw material availability. We identify starch plastics, PLA, bio-based PE and bio-based epoxy resin as the four most important plastics for the future bio-based plastic sector.

The BAU scenario assumes a steady growth of the four key plastics and a modest growth for cellulose films, PHA and bio-based PUR. The remaining plastics are expected to contribute little to the overall growth. The BAU projection results in a



†See data in Table 2.3 and Table 2.4. The overall maximum technical substitution potential for plastics and fibres is 89% (the blue line in the graph).
‡See data in Figure 2.3. The projection in 2020 is based on company announcements. The most recent data used for making this graph were received in March 2009; the reported values refer to the capacities at the end of each year.

Figure 2.4: Comparing the projections for 2020, based on company announcements with the market potential, based on the maximum technical substitution.

global production capacity of approximately 3 Mt for 2020, which is somewhat more modest compared to the companies' targets (approx. 3.45 Mt).

The HIGH scenario shows an optimistic future and a fast growing sector. The four key plastics will grow strongly and a steady growth rate is assumed for cellulose films, PHA and bio-based PUR. The major technical barriers will be overcome for bio-based succinic acid, bio-based PA6, PA66 and bio-based PP. The HIGH scenario projects that the global production will reach 4.40 Mt by 2020, approximately 30% higher than the projections aimed at by the companies (approx. 3.45 Mt).

The LOW scenario describes a relatively pessimistic future. The four key plastics will have slow growth rates and the growth from the remaining plastics will be insignificant. The LOW scenario projects that only 1.47 Mt capacity will be installed by 2020. This is approximately 60% lower than the projections aimed at by the companies (approx. 3.45 Mt).

Comparison with Crank et al.[11] (Step 4)

Table 2.5 shows the comparison with an earlier projection made by Crank et al. (i.e. PRO-BIP 2005). For 2020, the projections based on company announcements (3450 kt), based on the industry expectation (3440 kt) and based on the PRO-BIP 2009 BAU scenario (3000 kt), lie between the former projection with and without policy and measures (PM). Thus the 2020 projections from this study coincide well with the earlier projections published in 2005 by Crank et al.

Table 2.5: Worldwide production capacity of bio-based plastics until 2020, comparison of old and new projections.

			PRO-BIP 2009			For Comparisons	
All values in kt	Announced by companies	Expected by industry	BAU	HIGH	LOW	Crank *et al.* w/o PM [11]	Crank *et al.* with PM [11]
2003	100	100	100	100	100	71	71
2007	360	360	360	360	360		
2010						1 275	2 200
2013	2 320						
2020	**3 450**	**3 440**	**3 000**	**4 400**	**1 470**	**2 500**	**4 175**

2.3 Production, properties and applications of three selected bio-based plastics: PLA, bio-based PE and partially bio-based epoxy resin

Emerging bio-based plastics are still at an early stage of commercialisation. Only a few have entered large-scale production, while most are still in the pilot or R&D stage (Table 2.2). In the past, for starch plastics and PLA, efforts have been made to overcome key technical challenges. In the past decade, the first large-scale plants have been set up and a niche market has been established. For starch plastics, the technique of native starch blend-extruded with other compounds is nowadays well understood and applied by multiple players. For PLA, important remaining challenges include downstream processing of lactic acid, alternative raw materials, plastic processing and material property improvements. For bio-based PE, PA 11 and cellulose films, the technologies are relatively mature and therefore relatively little technical challenges will be encountered. For PHA, companies' expectations are high and the first large-scale plant (50 kt p.a.) is currently being constructed. However, the time and effort required to overcome the technical challenges, the market price and the material properties will strongly determine the market uptake of PHA. Being the first large scale plant of its type, the uncertainties are still relatively high. Shen et al. [44] elaborated in detail on the production, properties and application of the bio-based plastics listed in Tables 2.1 and 2.2.

In this perspective, we select three bio-based plastics as examples and report the latest technology and market developments. The three types of plastics are PLA (polyester), bio-based PE (polyolefin) and (partially) bio-based epoxy resin (thermoset). PLA is one of the most important bio-based plastics today (Figure 2.1); bio-based PE is likely to be produced on a large scale (> 500 kt) in the near future and will be operated by big chemical companies; and bio-based epoxy resin is an emerging opportunity for renewably sourced thermosets.

2.3.1 PLA

PLA is an aliphatic polyester, produced via the polymerisation of lactic acid which is a sugar fermentation product. With the start of the NatureWork's production plant in 2002, PLA became the first bio-based plastic produced on a large-scale (name plate capacity 150 kt p.a. in 2009). In 2007, the world's largest lactic acid producer PURAC started to produce lactide for technical applications on a large scale (capacity 75 kt p.a. lactide in 2008, plant located in Thailand). Recently, a new company, Pyramid, announced its plan to produce 60 kt p.a. PLA in 2012 in Germany.

Production and material properties of PLA

Lactic acid is produced by fermenting glucose, which can be obtained from various sugar sources. NatureWorks' PLA is produced from maize and PURAC's lactides are produced from cane sugar, potato starch and tapioca starch. In the future, it is expected that cellulosic biomass will be used to produce PLA. NatureWorks expects to use cellulosic feedstocks to produce PLA in the timeframe 2013–2018 [69].

Specific production of either of the isomers of lactic acid – i.e. L (+) or D (-) lactic acid – can be achieved by using an appropriate lactobacillus [70]. Polymerisation of L-lactide results in PLLA and polymerisation of D-lactide results in PDLA. The majority of current commercial PLA is poly (meso-lactide), which is a mix of L-lactide ($> 95\%$) and D-lactide ($< 5\%$).

Poly (meso-lactide) can be used in a wide range of applications, such as film- and tray- packaging, bottle packaging and textiles. This type of PLA exhibits no stereochemical structure. It is highly amorphous, does not rotate polarized light and is optically inactive. It has a relatively low glass-transition temperature (T_g = 55-60 °C), low vicat-softening point and low heat-deflection temperature. End products made from this PLA are not suitable for applications requiring high temperatures (similar to PET). The recently announced heat-resistant PLA is based on stereocomplex technology. Stereocomplex formation between PLLA and PDLA occurs when L-lactide unit sequences and D-lactide unit sequences coexist in one system [71]. PURAC describes the synthesis as a transesterification process in the presence of a catalyst; the starting materials are obtained from separately polymerized L-lactide and D-lactide [72]. Stereocomplexation of PLA sometimes is also called stereocomplex (sc) crystallization or racemic crystallization. Melt blending PLLA and PDLA with a D/L ratio of 1:1

produces sc-PLA crystals with a melting temperature (T_m) of 210-240 °C, which is about 30–60 °C higher than the T_m of homocrystalline PLLA. The crystal growth rate of sc-PLA was reported to be comparable with that of PA6 and PE [72].

Current and future applications of PLA

The current applications of PLA cover a wide range, for example packaging (cups, bottles, films and container), textiles (shirts, furniture textiles), non-wovens (diapers), agricultural mulch films (usually blended with TPS) and cutlery. Stereocomplex PLA is potentially suitable for meltspun fibres and biaxially stretched films. An example is the heat-resistant PLA fibres used for automobile textiles, developed by Mazda and Teijin [73]. Also, there is a potential of foamed PLA used as insulation material, for example PLA foam developed by Synbra, Sulzer and PURAC; foamed PLA is a bio-based alternative for expanded polystyrene (EPS) foam [74]. Furthermore, PLA blends and composite products experience increasing demand. For example, NEC and UNITIKA announced a mobile phone housing made from PLA reinforced with kenaf fibre [75]. Mazda, PURAC and Nishikawa Rubber of Japan collaborated on the development of heat-resistant automotive interior parts based on a combination of starch with stereocomplex PLA (Reichert R and van der Pol M, PURAC, private communication).

Table 2.6 shows the results of interviews on current and future market shares of PLA. Presently NatureWorks' PLA is primarily used in the packaging and the textile sector. For the future, NatureWorks sees the market potential not only in textile and packaging, but also in transportation and E&E (electrical appliances and electronics). PURAC sees the future PLA market mainly in textiles, buildings and transportation, while packaging and E&E have a relatively lower importance.

Table 2.6: Main applications for PLA – share of total production by market sector (interview results).

Sector	% of total production		
	2007	2020	
	NatureWorks	NatureWorks	PURAC
Packaging	70	20	20
Building			20
Agriculture	1		
Transportation		20	20
Electrical appliances and electronics (E&E)	1	10	10
Textile (fibres and fabrics)	28	50	40
Total	**100**	**100**	**100**

2.3.2 Bio-based PE

Bio-based polyethylene (PE) is produced from bio-based ethylene. In nature, many plants produce ethylene when their fruit are ripening. Industrial bio-based ethylene is produced from ethanol through a chemical dehydration process. The emergence of bio-based PE on the market is not a new phenomenon. In the 1970s, India used a small amount of its ethanol to produce ethylene and subsequently, PE, PVC (polyvinyl chloride) and styrene [12]. In the 1980s, subsidized by the Brazilian Government, small producers in Brazil produced in total 150 kt of ethylene per year; this ethylene was then converted into PE and PVC [76]. In the early 1990s, the subsidies for bio-based ethylene were stopped in Brazil, due to low oil prices. Consequently, production ceased. Today, given the public concern about global warming and limited fossil fuels and as a result of the increased oil price prior to the economic crisis in 2009, bio-based PE has become attractive again. In 2007, three large chemical companies, namely Braskem, Dow-Crystalsev and Solvay, announced the production of bio-based ethylene on a large scale in Brazil; the planned annual capacities are 200 kt of PE in 2010 by Braskem, 350 kt of PE in 2012 by Dow-Crystalsev and 60 kt of ethylene in 2010 by Solvay [67, 77, 78, 79].

Production of bio-based ethylene

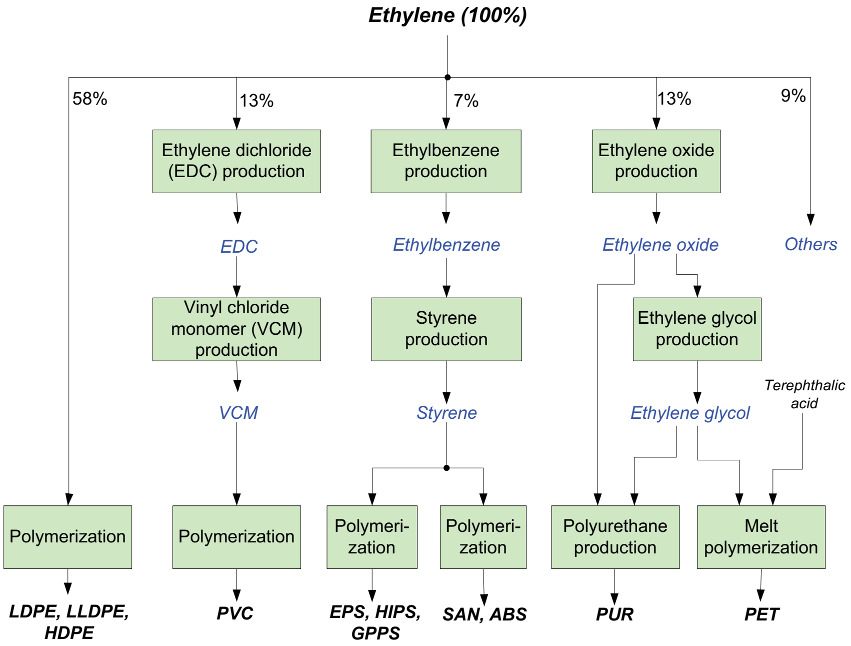
At present, bio-based PE is exclusively produced from sugarcane-based ethanol. In a sugar mill, the harvested sugarcane is cleaned, sliced and shredded, resulting in sugarcane juice as the main product and bagasse as the by-product. In Brazil, the bagasse is typically combusted to generate heat and power to fuel the sugar mill. The power surplus from bagasse is usually sold to the grid. The sugarcane juice is then fermented to ethanol under anaerobic conditions. Ethanol is distilled to yield hydrous ethanol (95.5 vol.-%) [80]. Ethylene is produced by dehydrating ethanol at temperatures varying from 300 °C to 600 °C in the presence of heterogeneous catalysts [81].

Bio-based ethylene as a building block

Ethylene is an important platform chemical in the chemical industry. PE is by far the most important product made from ethylene (Figure 2.5). In addition, ethylene is an important intermediate to produce PVC, PET, PS and polyols for polyurethanes (PUR). Figure 2.5 shows the intermediate chemicals and final plastics which can be derived from ethylene. In 2007, the global consumption of all ethylene-derived polymers was approximately 185 million tonnes including plastics and fibres [61]. Today, bio-based PE and PVC have been scheduled for production on a large scale. In the future, more bio-based plastics may be expected from bio-based ethylene.

Application of bio-based PE

bio-based PE can, just as petrochemical PE, be used for a large variety of applications (Table 2.7). For bio-based PE, Braskem will offer grades for food packaging, cosmet-



Note: percentages showed in the graph are on weight basis, according to global ethylene consumption in 2004 [82].

Figure 2.5: Ethylene as a platform chemical.

ics and personal care, automotive parts and toys [77]. Dow-Crystalsev will produce bio-based PE mainly for food packaging industry and for agricultural and industrial purposes (Gregorio M; Dow Chemical, private communication).

2.3.3 Partially bio-based epoxy resin

Approximately 75% of all epoxy resins are liquid epoxy resins (LER), which are derived from diglycidyl ether of bisphenol A; the remaining 25% are composed of various epoxy resins [64]. In this perspective, we limit ourselves to LER. The key materials to produce LER are epichlorohydrin (ECH) and bisphenol A. Today, bisphenol A is exclusively manufactured from petrochemical resources, whereas ECH can be produced from bio-based glycerol.

The conventional, petrochemical process to produce ECH is the chlorohydrination of allyl chloride, which in turn is produced by propylene chlorination. Until recently, ECH was also used to produce glycerol. However, in the past five years, this production process has become superfluous because of the large availability of bio-based glycerol as a by-product of biodiesel production. Thus the reverse process, which produces ECH from glycerol, has now become economically attractive.

Table 2.7: Market segments in Western Europe (figures in % for 2005) for HDPE, LDPE and LLDPE [61].

Market Segment	**HDPE**	**LDPE**	**LLDPE**
Films	18%	74%	82%
Blow moulding small parts	19%	1%	5%
Blow moulding large parts	12%		
Pipes and extruded products	19%	4%	4%
Extruded coating		11%	1%
Caps and closures	4%		
Petro tanks	3%		
Injection moulded parts	14%[a]	4%	5%
Cable		4%	5%
Textiles	3%		
Other	8%	2%	1%
Total: 12.7 Mt	**5.2 (Mt)**	**4.3 (Mt)**	**3.1 (Mt)**

[a] Excluding injection moulded HDPE caps, closures and petro tanks.

In the 1920s and 1930s, several publications and patents described the synthesis of glycerol-derived ECH or dichloropropanol by the reaction of glycerol with HCl (aqueous or gaseous), in the presence of catalysts (e.g. acetic acid) [83, 84, 85, 86, 87]. However, these approaches were never used to produce ECH and subsequently, epoxy resin.

In April 2007, Solvay started the production of ECH from bio-based glycerol in Tavaux, France (10 kt per year) based on its Epicerol™process patent [88]. Meanwhile, Solvay announced the building of a bio-based ECH plant in Thailand with an annual capacity of 100 kt and which will start production in 2009 [89]. In 2007, Dow Epoxy announced the set up of a glycerol-to-ECH plant (150 kt per year) in Shanghai, using Dow's proprietary technology. The plants are scheduled to start up in 2009/2010 [90].

Production of bio-based ECH from glycerol

In biodiesel production, glycerol is obtained as a by-product of the transesterification of crude vegetable oil. For example, in palm oil biodiesel production, approximately 10 kg of glycerol is obtained for every 100 kg biodiesel [91]. According to Solvay's patent [92], glycerol reacts with HCl at a temperature of 80–120 °C. The reaction is carried out with catalysts such as hexanedioic acid. The output of the reaction is a pseudoazeotropic mixture, containing 1,3-dichloropropanol, HCl and water. Through either steam stripping or distillation, HCl and water are separated from 1,3-dichloropropanol, which is then further purified and dehydrochlorinated into ECH (Figure 2.6). Solvay claims that this process leads to fewer by-products and lower water consumption.

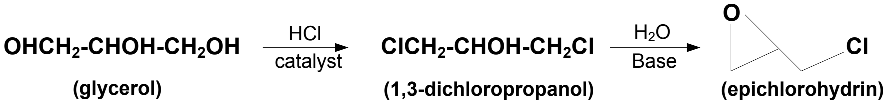


Figure 2.6: Conversion of glycerol into epichlorohydrin according to the Solvay Epicerol™ process.

Partially bio-based epoxy resin in the future

For every 100 kg LER, approximately 30 kg ECH and 80 kg bisphenol A are required based on the reaction stoichiometry. Since bisphenol A is petrochemically derived, the share of the bio-based component in LER is approximately 30 wt-%. The global consumption of epoxy resins was estimated at 1.15 Mt for the year 2000 [64], of which LER accounted for 860 kt (76%). If all these LER were obtained from bio-based ECH, 260 kt per year of bio-based ECH would be required (860 kt $*$ 30 kg ECH/100 kg LER). It is likely that a substantial part of demand for ECH in 2010 can be covered by the bio-based ECH capacities announced by Dow (150 kt) and Solvay (110 kt).

Applications of epoxy resin

Epoxy resins are primarily used for protective coatings and in electrical and structural applications. bio-based LER is chemically identical with petrochemical LER and therefore there are no differences regarding applications.

2.4 Conclusions

The historical use of natural products for plastics production demonstrates that bio-based products are neither fictional nor new. Instead, for many decades, bio-based products have been an industrial reality on a million-tonne scale (e.g. paper and board). Today, the combined volume of non-food and non-plastics applications of starch and man-made cellulose fibres is 55 times larger than the total of all emerging bio-based plastics (approx. 20 Mt versus approx. 0.36 Mt in 2007). This demonstrates that the production of bio-based products on very large scale is not unprecedented and that related challenges, for example concerning logistics, can be mastered. If emerging bio-based plastics succeed in following this example, it is possible that they can substitute their petrochemical counterparts in large quantities.

Between 2003 and 2007, the annual growth rate of the emerging bio-based plastics was nearly 40%, resulting in a global capacity of the emerging bio-based plastics of 0.36 million tonnes in 2007. The global production of bio-based plastics is likely to grow strongly in the next decade and to reach 2.3 Mt in 2013 and 3.5 Mt in 2020. The maximum technical substitution potential of bio-based plastics (including man-made fibres) replacing their petrochemical counterparts is estimated at 240 million tonnes, or 90% of the total plastics and fibres based on the 2007 market demand. Based exclusively on the technical feasibility (and hence disregarding economic and other aspects), the growth potential for bio-based plastics is hence enormous.

Of all the emerging bio-based plastics, we highlighted three products – PLA, bio-based PE and partially bio-based epoxy resins. PLA is a new polymer. In the past decade, it has experienced technological breakthroughs that will allow a wider range of applications. bio-based PE, which was applied on a large scale in India and Brazil before the 1990s is now being revisited. The economic feasibility of bio-based ethylene opens up enormous opportunities for renewably sourced ethylene-derived chemicals and plastics in the future. The increasing availability of bio-based glycerol from biodiesel production makes it possible to produce bio-based epichlorohydrin. Based on the announced capacity expansions, it is likely that by 2010 a large part of liquid epoxy resins in the market will be partially renewably sourced.

To conclude, several factors clearly speak for bio-based plastics. These are the limited and uncertain supply of fossil fuels (i.e. oil and gas), economic viability, environmental considerations (e.g. savings of non-renewable energy and greenhouse gas abatement), innovation offering new opportunities (technical, employment, etc.) and rejuvenation in all steps from chemical research to the final product and waste management. Challenges that need to be successfully addressed in the next years and decades are the low performance of some bio-based plastics (e.g. thermoplastic starch), their relatively high cost for production and processing and the need to minimize agricultural land use and forests, in order to avoid competition with food production and adverse effects on biodiversity and other environmental impacts.

Chapter 3

Life cycle assessment of polysaccharide materials: A review*

3.1 Introduction

Polysaccharides are among the most important renewable resources for mankind. They have been widely used for a long time for food (starch), clothing (cotton, flax, and jute), communication (paper), packaging (paper and board), and construction (wood). Next to these traditional usages, other non-food products have been developed to partly replace conventional products which are either based on non-renewable resources or based on traditional polysaccharide materials. For instance, starch polymers are used for packaging films and loose fills; and natural fibre reinforced polymer composites substitute glass fibre-reinforced polymer composites in automobile components. Table 3.1 lists the production volumes of some bulk polysaccharide products.

Table 3.1: Polysaccharide products, global production, large scale producers and volumes (kt = 1000 metric tonnes)

Polysaccharide materials	Global production	Production EU	Production US
Man-made cellulose fibre	2700-3300 kt (2005) [93, 94]	416 kt (2005) [94]	46 kt (2005) [94]
Starch polymers [95]	40 kt (2006)	30 kt (2006)	10 kt
Natural fibre composites [96]	n/a	51 kt (2003)	n/a
Wood plastic composites [96, 97]	720 kt (2003)	65 kt (2003)	655 kt (2003)[a]

[a] Data for North America region, estimated.

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Bio-based materials are generally considered to be more sustainable than conventional petrochemical materials because they are made from renewable instead of non-renewable raw materials [98]. The purpose of this paper is to obtain insight into the environmental impacts of polysaccharide products in comparison to their counterparts, which are either petrochemical products or conventional polysaccharides. The most widely accepted method to assess environmental impact is the method of life cycle assessment (LCA). In this paper, we review readily available LCA studies or environmental assessment studies for polysaccharide-based textile products, natural fibre composites, and thermoplastic starch. The most traditional usages of polysaccharides, such as food, wood, pulp, paper prints and paper packaging products, are not included in this review.

3.2 Life cycle assessment

Life cycle assessment has been standardised by the International Standardisation Organisation (ISO) in the ISO 14040 series. There are four ISO standards which address the various areas of LCA, namely [99]:

- ISO 14040: 1997 - Principles and framework
- ISO 14041: 1998 - Goal and Scope definition and inventory analysis
- ISO 14042: 2003 - Life Cycle Impact assessment
- ISO 14043: 2003 - Interpretation

The two most commonly used systems chosen in LCA studies are cradle-to-factory gate and cradle-to-grave. A cradle-to-factory gate LCA study includes all steps from the extraction of raw materials and fuels, followed by all conversion steps up and until the product is delivered at the factory gate. Cradle-to-factory gate analyses are often published by material producers. The system cradle-to-grave covers all steps of the system cradle-to-factory gate and in addition, also the usage and the disposal phase. Cradle-to-grave analyses have the advantage of covering all phases of the life cycle. Since waste management differs by country and not all waste treatment options can be taken into account, cradle-to-grave analyses for a given product can lead to very different results depending on the type of waste management. If comparisons across the various waste management options are not available, cradle-to-factory gate analyses can provide first insight into the environmental impacts.

The results in LCA studies can be presented in different forms. In most studies the so-called mid-point level results are presented. Here, the life cycle inventory data, which represents the various types of emissions and the raw material requirements, are converted into environmental impact categories, such as the contribution to global warming or to acidification. End-point level results are calculated by aggregating different impact categories with weighting factors, leading to an overall environmental score for a product. Analyses leading to end-point level results are also referred to as single score analyses. They involve subjective judgement when determining the weighting factors for the various impact categories. In a comparative LCA study, sometimes mid-point results are sufficient to draw conclusions, e.g. when one product is clearly better than the other for all impact categories (or when it is comparable with the other option for all categories). If, on the other hand, the environmental impacts are larger for some impact categories and lower for others, judgements need to be made about the relative importance of each impact category. For this purpose, it is a rather common to apply single score methods (examples are Eco-Indicator 99 [100], EPS 2000 [101], IMPACT2002 [102, 103] and EDIP [104, 105]).

We focus on primary non-renewable energy use (NREU) and greenhouse gas (GHG) emissions. For these indicators, data are more readily available from environmental assessments than for other impact indicators and therefore offer a wider basis for comparison across alternative options. Non-renewable energy use (NREU) represents the total of fossil energy and nuclear energy, of which fossil energy usually dominates. Fossil energy requirements from cradle to factory gate, also referred to as cumulative fossil energy demand, has been proven to be a good indicator for environmental performance of a given product or service [106]. This is because in many cases materials and processes are strongly energy-related. However, for impacts with less energy-dominated activities, for example, when involving the use of toxic compounds, energy provides an incomplete picture and environmental impacts should be extended by other, more specific indicators.

3.3 Textiles

In general, the object of study in an LCA should be as close as possible to an end product (such as a shirt or a pair of trousers). However, many data in LCA studies are available for fibres, which are the starting material for fabrics (e.g. by weaving or knitting). Table 3.2 shows an overview of NREU for the production of fibres and fabrics, and the energy recovery from waste incineration. As Table 3.2 shows, the NREU for fibre production differs very substantially across the different types of fibres; in contrast, the energy use of fabric processing is less dependant on the type of the raw material and instead, primarily depends on the type of processing, which determines the final functions and qualities.

In this section, we discuss the results of comparative LCA studies for fibres and end products (but not for fabrics) made from cotton, polyester, and man-made cellulose fibres. For cotton and polyester, the available LCA studies allow us to compare results for both fibres and end products; while for man-made cellulose fibres, the comparison is only possible at the level of fibres. In the following sections, we first provide results for the system “cradle-to-factory gate”; we then proceed with the system “cradle-to-grave” including use phase and incineration with energy recovery.

3.3.1 Cradle-to-factory gate, fibres and end products

Per kg fibre

For fibre production, we found that the polysaccharide-based fibres have lower NREU (non-renewable energy use) than petrochemical-based fibres. As shown in Table 3.3, man-made cellulose fibres have the lowest NREU requirements among all the fibres

Table 3.2: Non-renewable energy use (NREU) for the production of different fibres and fabrics and the energy recovery by waste incineration [107]

Process	Wool	Cotton	Viscose	Polyester	Acrylic
Feedstock in raw mat.	n/a [a]	n/a	0[b]	46 MJ/kg fibre	60 MJ/kg fibre
Production of raw mat.	8 MJ/kg (Rough estimate for raw wool fleece, excluding energy for raw wool scouring)	49 MJ/kg (Baled raw cotton)	2 MJ/kg (Pulp of fibres, external energy)	50 MJ/kg	52 MJ/kg
Production and spinning of fibre	Not relevant	Not relevant	33.3 MJ/kg (Polymerisation and spinning of fibres)	13.6 MJ/kg (Polymerisation and spinning of fibres)	46.3 MJ/kg (Polymerisation and spinning of fibres)
Subtotal for fibre production	8 MJ/kg	49 MJ/kg	35 MJ/kg	109 MJ/kg	158 MJ/kg
Spinning of staple fibre yarn	Rough estimate based on cotton/polyester data 15-45 MJ/kg				
Warp-size and weaving	Rough estimate based on cotton/polyester data 10-30 MJ/kg				
Knitting	Rough estimate based on cotton/polyester data 5-20 MJ/kg				
Dyeing/washing/drying	Rough estimate based on cotton/polyester data 3.5-13 MJ/kg				
Finishing	Rough estimate based on cotton/polyester data 4-8 MJ/kg				
Subtotal for fabric processing	40-116 MJ/kg (estimates based on cotton/polyester data)				
Energy recovery from combustion	20.5 MJ/kg	16.3 MJ/kg	16.3 MJ/kg	22 MJ/kg	Not available

[a] Unknown according to [107].

[b] Feedstock of viscose fibre is renewable energy (biomass): 36 MJ/kg fibre.

reviewed. Viscose fibres require 10-30% less NREU than cotton fibres and 50-80% less NREU than petrochemical-based fibres.

Table 3.3: Summary of cradle-to-factory gate NREU for the production of different types of fibres (MJ/kg)

Cotton fibre	Man-made cellulose fibre	PET fibre	Source
49			[108]
59		97	[109]
49	35	109	[107]
	44		[110]
	39		[111]
		95[a]	[112]

[a] According to [112], the polymer (raw material) production for PET amorphous requires energy 81 MJ/kg. According to [107], spinning of polyester fibre requires 14 MJ/kg. So the energy use for the PET amorphous fibre production is estimated at 81 + 14 = 95 MJ/kg.

Per piece and per kg end product

Table 3.4 presents the NREU for two end products, namely a sofa cover and a hotel bed sheet. The results are presented firstly per piece and secondly for 1 kg of the respective material. It can be seen that the NREU of 1 kg end product made from cotton and PET is practically identical (160 MJ/kg and 159 MJ/kg, respectively [108]). However,

Table 3.4: Cradle-to-factory gate NREU for cotton and PET textile end products

Functional unit	Cotton fabric		PET fabric		Cotton/PET fabric	
	MJ/end product	MJ/kg	MJ/end product	MJ/kg	MJ/end product	MJ/kg
One three-seat sofa cover [108]	796	160	568	159		
One hotel single-bed sheet [109]	72	94			34	110

because of the higher density of cotton, which causes higher material demand for the cotton products, the cotton products have a higher NREU per piece of end product than the polyester products (796 MJ/ piece cotton cover versus 568 MJ/piece PET cover in [108]).

From Table 3.4 it can also be seen that much more energy is required per kg cotton used for sofa covers (160 MJ/kg [108]) than per kg cotton used for bed sheets (94 MJ/kg [109]). This implies that the production of the dyed cotton sofa cover requires more energy than the bleached-only cotton bed sheets. This again demonstrates that the energy requirement of fabric manufacturing strongly depends on the type and function of the final textile products and is relatively independent from the type of fibres (see also Table 3.2).

3.3.2 Use phase

The environmental impact from the use phase of textiles is dominated by the maintenance of the textile products, particularly cleaning and drying [107]. Dahllöf report 54% higher energy consumption for the cotton sofa cover than for the PET sofa cover during the use phase [108]; for the hotel bed sheet, Kalliala and Nousianinen report 20% higher laundering energy use for the cotton sheet than for the 50/50 cotton/PET sheet [109]. We can conclude from these data that cotton products require more energy for cleaning and drying than polyester products. One reason is that the heavier mass weight of cotton fabrics increases the washing load [108]. Another reason is the high water absorption capacity of cotton; as a consequence, more water is needed for washing and therefore, more energy is needed to heat water. Also, the amount of energy required to dry the textiles increases in proportion to the amount of water evaporated, under the condition that an electric dryer is used rather than simple drying in air [107]. No LCA data is available for the maintenance of cellulose products.

3.3.3 Incineration with energy recovery

Incineration with energy recovery is seen as a common end of life management for textile products [108]. Contrary to the use phase, it is rather simple to capture the advantages of energy recovery in waste incineration. To this end, we use the indicator “net NREU”. It is defined as the NREU of production (energy spent) minus the energy recovered from waste incineration (energy gained; due to the complexity involved, we exclude the use phase). In Table 3.5, this indicator is compared across different fibres at an energy recovery rate of 50% and a theoretical recovery rate of 100%. It is found that polysaccharide fibres have less net NREU requirements than petrochemical fibres, even the calorific values of natural fibres is much lower than that of petrochemical fibres (Table 3.5). Viscose fibres have lower net NREU values than cotton for both 50% and 100% recovery rates (Table 3.5). However, a thorough literature review did not yield energy consumption data for the viscose fabric production. Assuming that the energy use for textile production from fibres does not differ substantially across the various types of polymers, it can be concluded that the viscose fibre is the preferred textile material for the system cradle-to-grave excluding the use phase (due to lack of data) but including the waste management stage (incineration with energy recovery). This conclusion is based on the assumption that the end products can be compared on a mass basis (identical functionality of 1 kg fibre material).

In contrast, Dahllöf [108] compared a 3.56 kg PET sofa cover with a 4.99 kg cotton sofa cover. The comparison includes production, the use phase and incineration with 100% heat recovery. Although cotton has a lower calorific value compared to PET (calorific value: 16 MJ/kg cotton and 22 MJ/kg PET), the higher weight of the cotton sofa cover results in a similar energy recovery as the PET sofa cover (energy recovered: 81 MJ per cotton cover and 78 MJ per PET sofa cover). Thus, Dahllöf concludes that from cradle to grave the cotton sofa cover is a less favourable choice compared to the PET sofa cover [108]. This finding differs from the conclusion drawn based on Table 3.5 (see preceding paragraph) and hence shows the importance of the amount of material required per end product.

Table 3.5: Energy recovery and net energy requirement for different fibres (MJ/kg)

Type of fibre	NREU, cradle-to-factory gate (A)	Calorific value (LHV) (B)	Net NREU at 100% recovery (A-B)	Net NREU at 50% recovery (A-50%B)
Cotton	50-60 [107, 109]	16.3 [108]	34-44	42-52
Viscose	35-44 [107, 110, 111]	16.3 [107]	19-28	27-36
Lyocell	39 [111]	15 [113]	24	32
PET	95-109 [107, 109, 112]	22.6 [114]	73-87	84-98
Polyacrylic	158 [107]	n/a	n/a	n/a
Nylon 66	154 [107, 115]	30.1 [114]	124	139
Nylon 6	134 [107, 116]	30.1 [114]	104	119

3.3.4 Discussion

We have so far only discussed energy use but there are environmental impacts which are not related to energy consumption. For example, cotton production causes several impacts during cultivation, namely, water consumption for irrigation and processing, fertiliser use and herbicides/insecticides use [107, 108]. Based on the studies reviewed, we identify two general problems which arise when assessing the environmental impacts related to textile fibres.

The first problem is the data quality, namely old data and geographically differing agriculture practices. In some literature sources, data for cotton growing are 20-40 years old [109]. The use of dated information is not a problem if the agriculture practice of cotton cultivation has not changed much in the last decades, which might be the case in some parts of the world. Moreover, location-dependent agriculture practices cause inaccurate estimates of water consumption, fertiliser use and herbicides/insecticides use. According to [107], huge differences of these agriculture practices not only exist between regions, but also within the same region/country. For example, the amount of insecticides used in Spain is in general 300% higher than in Brazil. Within Spain, the amount of insecticides used for cotton growing can differ by a factor of seven from one farm to another, causing difficulties in obtaining a generic dataset.

The second problem is a lack of toxicity assessment for the production of polysaccharides. In particular, a quantitative analysis of the toxic effects of cotton cultivation, viscose production, and *N*-Methylmorpholine-*N*-oxide (NMMO) solvent in Lyocell production (which is another type of man-made cellulose fibre) would be of interest. However, the lack of harmonised and consistent methodologies for the toxicological assessment in LCAs and the incompleteness of the databases with regard to toxicological information pose serious problems (compare [117] and [118]).

3.4 Natural fibre composites

In this section we discuss natural fibre composites made of plant fibre-reinforced thermoplastic polymers. The commonly used fibres are flax, hemp and abaca. Natural fibre composites have been used in automotive components as substitutes for glass fibre composites. For both environmental and economic reasons, natural fibres are becoming more and more attractive as reinforcing agent for composite materials. Compared to glass fibre, natural fibres require less energy to manufacture (Table 3.6). In addition, natural fibre composites reduce the weight of vehicles and therefore improve the fuel economy. According to FNR [119], currently German car manufacturers used on average 3.5 kg natural fibre per passenger car, especially in medium and upper classes. Assuming that the average natural fibre used in a European passenger car is 1.5-3.5 kg, this amounts to 22-51 kt per year natural fibre demand in automotive industry in EU 15 (in 2004 passenger car production in EU15 was 14.7 million units according to VDA [120]).

Table 3.6: Non-renewable energy requirements for production of different fibres

Non-renewable energy requirements (MJ/kg fibre)							
Glass fibre[a]		Flax fibre[a]		China reed fibre[b]		Hemp fibre[c]	
Raw materials	1.7	Seed production	0.05	Cultivation	2.5	Cultivation	0.5
Mixture	1.0	Fertilisers	1.0	Transport plant	0.4	Fertiliser&seeds	1.3
Transport	1.6	Transport	0.9	Fibre extraction	0.08	Transport	0.2
Melting	21.5	Cultivation	2.0	Fibre grinding	0.4	Fibre production	1.8
Spinning	5.9	Fibre separation	2.7	Transport fibre	0.3	Mat. Production[d]	2.9
Mat. Production	23.0	Mat. Production	2.9	Mat. Production[d]	2.9		
Total	54.7	Total	9.6	Total	6.5	Total	6.8

[a] Original source [121], reproduced in [98].

[b] Original source [122], back up calculation by [123].

[c] Source [124].

[d] Material production is estimated from flax fibre, using reference [121].

In the next section, we will review several studies of natural fibre composites materials used in automotive components (three studies) and for transportation pallets (one study). In these studies, NREU and GHG emissions for production phase, use phase and waste management are discussed (Section 3.4.1). In addition, an LCA study, which links environmental impacts (single scores) and composites material properties [125], is reviewed in Section 3.4.2.

3.4.1 Automotive and transportation components

Four LCA studies on natural fibre composites are reviewed in the application area of automotive and transportation components. In these studies, the natural fibres chosen are flax, hemp and china reed; the matrix materials are mainly PP (polypropylene) and EP (epoxy resin). The applications are interior panels of a car and a transportation pallet. We separately discuss the results for the production phase, the use phase and waste management.

Production phase (Cradle-to-factory gate)

Table 3.7 and Table 3.8 show the energy and GHG savings per component (panel or pallet) and per kg composites. It can be seen that all the studies show benefits of natural fibre composites in terms of NREU saving; moreover, a higher fibre fraction results in less NREU. The cradle-to-factory gate NREU are mainly determined by the matrix material, since the production of 1 kg natural fibre requires approximately 7-10 MJ/kg (Table 3.6) while the NREU of the natural fibre composites amounts to 60-90 MJ/kg (Table 3.8). In most cases natural fibre composites show advantages for GHG emissions over glass fibre composites (Table 3.7 and Table 3.8). As an exception, according to study [31], one kg hemp/EP has a higher GHG emission than ABS copolymer (see Table 3.8) due to the strong impact from the production of the epoxy resin. However, per piece of panel, the hemp/EP panel leads to less GHG emissions than the ABS panel (see Table 3.7).

Use phase

The use phase of natural fibre composites, especially as a component in transportation systems (panel and pallet), entails a more important environmental benefit than the production phase [122, 124, 125]. At least 95%[1]of the production energy can be saved during the use phase according to [122] and [124] (see Table 3.9); for long distance transportation, the energy savings can be as high as 300%[2] of the energy required to produce a new pallet.

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[1] From Table 3.9, Hemp/EP composites used for side panel: 71 MJ / 73 MJ = 97%.

[2] From Table 3.9, China-reed composites used for transportation pallets: 2300 MJ / 717 MJ = 320%.

Table 3.7: NREU and GHG emission savings of natural fibre composites

Functional unit (1 piece of)	Natural fibre composites (A)	Conventional material (B)	Saving (1-A/B)	Source
Non-renewable energy, NREU (MJ per piece)				
Underfloor panel[a]	132 (flax/PP)	155 (GF/PP)	16%	[121]
Interior side panel[a]	73 (hemp/PP)	132 (ABS)	45%	[98, 124]
Transportation pallets[a,b]	717 (CR/PP)	1345 (GF/PP)	47	[122]
GWP (kg CO_2 eq. per piece)				
Interior side panel[a]	4.6 (hemp/PP)	5.4 (ABS)	13%	[98, 124]
Front subframe[c]	4.0 (hem/EP)	21 (GF/PP)	81%	[126]
Transportation pallets[a]	40 (CR/PP)	75 (GF/PP)	47%	[122]

Note: GF = glass fibre, EP = epoxy resin, CR = china reed

[a] Cradle to factory gate.

[b] 717 MJ = PP production (562 MJ) + china reed fibre production (19 MJ) + china reed fibre transportation (4 MJ) + pallet production (132 MJ); 1345 MJ = PP production (883 MJ) + glass fibre production (303 MJ) + pallet production (168 MJ) [122].

[c] Cradle to grave; end of life recycle.

Table 3.8: Cradle-to-factory gate NREU and GHG emissions of 1 kg composite materials

Material	Fibre content (wt%)	NREU (MJ/kg)	GHG emissions (kg CO_2 eq./kg)	Source
China reed/PP	53	61[a]	3.4[b]	[122]
Hemp/EP	47[c]	89	5.7	[124]
Glass fibre/PP	42	89[d]	5.0[b]	[122]
Glass fibre/EP	n/a	126[e]	5.9[e]	[127]
ABS copolymer	0	117	4.8	[124]
Carbon fibre/EP	26	176	5.8	[127]

[a] 61 MJ/kg is calculated from 717MJ/functional unit. 717 MJ = PP production (562 MJ) + china reed fibre production (19 MJ) + china reed fibre transportation (4 MJ) + pallet production (132 MJ) [122].

[b] GHG emissions for system boundary of production plus incineration with energy recovery.

[c] 47 wt% of hemp fibre content is own calculated. According to [124], hemp fibre has a volume fraction of 66 vol.%. Since the entire volume is same for both ABS and hemp/EP panels, the mass of hemp fibre can be calculated based on the density (ρ) data. In this calculation, $\rho_{\mathrm{ABS}} = 1.05$ g/cm^3 [128], $\rho_{\mathrm{EP}} = 1.20$ g/cm^3 [125].

[d] 89 MJ/kg is calculated from 1345 MJ/functional unit. 1345 MJ = PP production (883 MJ) + glass fibre production (303 MJ) + pallet production (168 MJ) [122].

[e] Assume the weight is the same as the carbon fibre/PE blade, which is 300 kg.

End of life

In the disposal phase, incineration with energy recovery reduces the net NREU of natural fibre composites (compared to cradle-to-factory gate), whereas, for glass fibre, it leads to an increase by 1.7 MJ/kg glass fibre due to the extra energy required in waste incineration [122]. Due to the low calorific value of natural fibres, the energy

Table 3.9: Energy saving by natural fibre composites during the use phase

Natural fibre composites	Substituted product	Energy requirement for production (MJ/functional unit)	Energy saving during use (MJ/functional unit)	Source
China reed/PP pallet	Glass fibre/PP pallet	717	660-2300[a]	[122]
Hemp/EP side panel	ABS side panel	73	71-118[b]	[124]

[a] Transportation distance 5000 – 200000 km.

[b] Low range for a light car and high range for a heavy car.

credit from waste incineration is small for natural fibres (the calorific value of flax fibre is 16 MJ/kg [114], while the calorific value of ABS and PP is around 40-45 MJ/kg [114]). However, the overall energy saving of natural fibre composites is dominated by the fuel saving during the use phase (see section "Use Phase"). Hence, from cradle-to-grave, natural fibre composites allow to save considerably more NREU than glass fibre composites and petrochemical polymers.

Besides incineration, recycling is an alternative disposal option. The recycling rate is a critical factor for the extent to which environmental impacts can be reduced [122]. A further disposal option is landfilling. However, due to changed legislation, landfilling is not an option any more in many countries especially in EU.

3.4.2 A stiff beam made from flax fibre composites

Bos [125] studied the environmental impacts of beams and ties which were made from natural fibre composite materials and were designed based on stiffness-limited criteria. Stiffness is one of the most important mechanical requirements for engineering materials. Typical examples of stiffness-determined products are a beam and a tie (other important functions are, for example, shaft, plate and column). In Bos' study there are six hypothetical unidirectional composite materials which have the same level of stiffness[3] and are made from three types of matrices, EP (epoxy), UP (unsaturated polyester) and PP, reinforced with either glass fibre or flax fibre with different fibre weight fractions (0.2, 0.4, 0.6 and 0.8). The system boundary is cradle-to-factory gate. The environmental impact is expressed using the single score method EcoIndicator 95 (see Figure 3.1).

It can be seen from Figure 3.1 that the higher the fibre weight fraction, the lower the single score, except for the glass fibre/PP composite, for which the fibre content does not have significant influence on the environmental impact of the composites. Furthermore, for both flax and glass fibres, the PP composites are significantly better than the EP and the UP composites from an environmental point of view. Bos also studied the relationship between beam weight and fibre fraction since weight reduction is considered to be a major advantage of natural fibre composites (especially if applied for moved parts). Figure 3.2 shows that the beam weight of the flax fibre composites is lower than the beam weight of the glass fibre composites at all fibre contents

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[3] The functional unit is defined as "deflection beam, width 100 mm and length 1 m, with variable thickness, designed to give a maximum deflection of 10 mm at a load of 1000 kN" [125].

(see Figure 3.2); and particularly, the beam weight reduction at high fibre contents is significant.

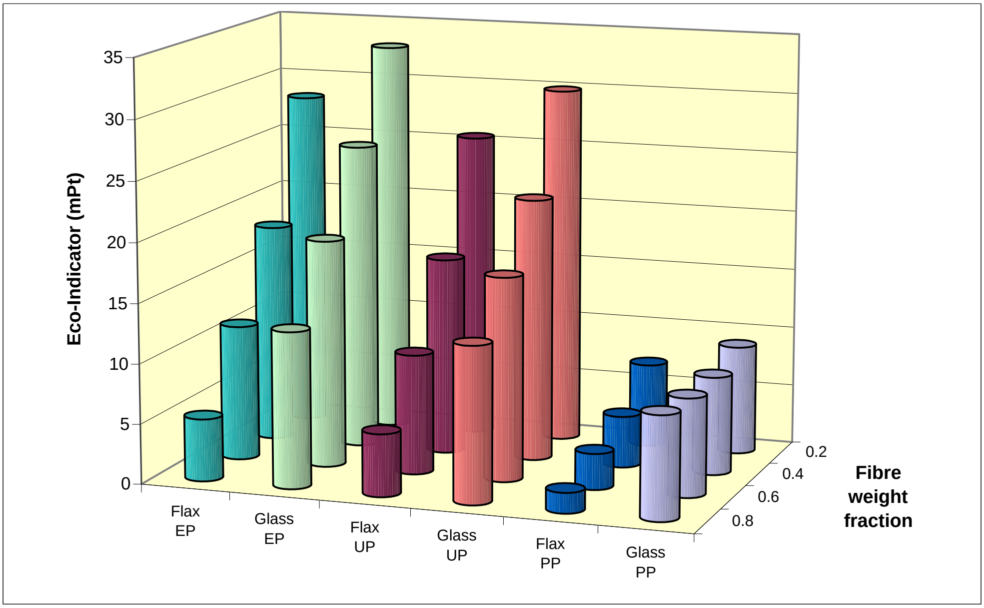

Figure 3.1: Cradle-to-factory gate environmental impacts of a stiff beam made from the six hypothetical composites, data rearranged from [125].

Besides the LCA studies of a stiff composite, Bos also presented an LCA study for a strong tie for tensile loading[4] made from the above six hypothetical materials. It was found that because of the low strength of flax fibres in comparison with glass fibres, the element becomes much thicker and consequently a relatively larger amount of matrix resin is required than the glass fibre composites. Since the matrix material has strong influence on the final environmental impact, the environmental impact of a strong tie made from the flax composites is clearly higher than the glass fibre composites. Therefore, it is concluded that flax fibre reinforced composites are a better choice from an environmental point if stiffness is required in combination with limited strength.

3.5 Starch polymers

Thermoplastic starch (TPS) is produced from natural starch, destructurised in the presence of specific amount of plasticizers and under certain extrusion conditions. The type of starch polymer varies from 100% pure starch to different kinds of blends with different shares of petrochemical copolymers. In this review, we mainly present the

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[4] When bearing loads, a strong tie requires strength and it may show elastic property and hence bend (imagine a plastic film), while a stiff tie does not change its shape (imagine a bookshelf).

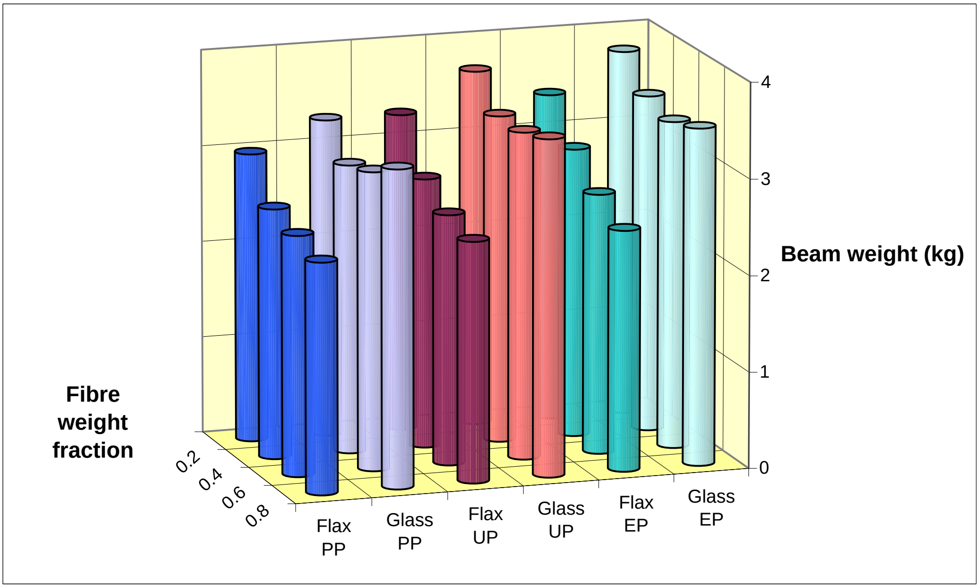

Figure 3.2: The mass of a stiff beam made from six hypothetical composite materials, as a function of fibre weight fraction, data rearranged from [125].

results of the review by Patel et al. [98] and a comparative LCA study by James and Grant [129]. Patel et al. [98] presented a review of six LCA studies for end products from starch polymers, namely, starch pellets, loose fills, films and bags. James and Grant [129] conducted a comparative LCA of biodegradable grocery bags made from starch polymers/blends, paper, cotton and petrochemical plastics.

All LCA studies that were available to us report that, per kg, starch polymers require less NREU than petrochemical polymers (see Table 3.10). According to Patel et al. [98], the energy saving of starch polymers ranges from 23 to 52 MJ/kg (±15% depending on LDPE or LLDPE is chosen as reference [130, 131]. However, due to the low density of starch polymers, usually larger amounts of them (in mass terms) are needed to produce a final product that fulfils a same function (e.g. as loose fills, packaging films and bags) than their petrochemical counterparts (see Table 3.11). For the cases studied, TPS is nevertheless at least comparable to petrochemical products and in some cases, the NREU savings are even substantial (for example, the TPS film requires half of the energy to produce the PE film, see Table 3.11).

As shown in Table 3.12, starch polymers have lower calorific values than petrochemical LDPE; therefore, less energy can be recovered from TPS by incineration. This could mean that for the system cradle-to-grave, the inclusion of waste incineration with energy recovery could invert the energy saving benefit for starch polymers. However, as Table 3.12 shows, when comparing TPS to petrochemical LDPE, the net NREU (cradle-to-factory gate NREU minus energy recovered from incineration) of starch polymers is still by far smaller than that of the pure petrochemical products (note that the net NREU as defined here excludes the use phase, see also Section 3.3.3; due to lack of detailed data the values refer to plastic pellets and not to end products).

In terms of GHG emissions, 1 kg TPS leads to lower GHG emissions than petrochemical plastics. Patel et al. [98] reported that the GHG emission saving of TPS is 1.2-3.7 kg CO_2 eq./kg, ±15% (depending on whether LDPE or LLDPE is the reference [130, 131]); James and Grant [129] reported that the GHG emission saving of TPS is about 1.1-2.1 kg CO_2 eq./kg depending on whether HDPE or LDPE is chosen as the reference (Table 3.10). Per functional unit of an end product, the GHG emission savings of TPS is partially compensated by the higher weight [98, 129] of TPS products compared to petrochemical plastics. For the functionally fully comparable cases (e.g. comparison of single use petrochemical grocery bags with single use bio-based bags), the bio-based polymers are better than the petrochemical based polymers. But the single-use bio-based grocery bags cannot compete with multi-use petrochemical bags.

It should be considered in this context that the high share of landfilling as assumed in most of the cases leads to an advantage for the GHG profile of petrochemical polymers because the fossil carbon embodied in the polymers is not released, while for bio-based polymers biodegradation may lead to methane emissions with high global warming potential (unless the landfill is operated with methane capture); in contrast, full or predominant incineration (practically absent from Table 3.11) would result in a GHG advantage for bio-based polymers because the carbon embodied in the polymers has been extracted earlier from the atmosphere and therefore does not represent a net addition.

So far the comparisons refer to *virgin* petrochemical polymers as conventional counterparts. One of the sources reviewed compared loose fill packaging material made of starch polymers with loose fill made of *recycled* petrochemical polymers. In this case, starch polymers can hardly compete with petrochemical polymers from an environmental point of view [133]. This finding may well be representative also for other products.

Table 3.10: Summary of energy and GHG emissions for per kg plastic pellets; product listed are all commercial products manufactured by state-of-the-art technologies.

Type of plastic	Functional unit (FU, kg)	Cradle-to-gate NREU[a] (MJ/FU)	Type of waste treatment assumed for calculation of emissions	GHG emissions (kg CO_2 eq./FU)	References
TPS	1	25.4	Incineration	1.14	[29]
	1		80%incin.+20%compost.	1.20	[132]
	1		100% composting	1.14	[132]
TPS+12.7%PVOH	1	18.9	Non-(cradle-to-factory gate)[b]	1.1[b]	[133]
TPS+15%PVOH	1	24.9	Incineration	1.73	[29]
TPS+52.5%PCL	1	48.3	Incineration	3.36	[29]
TPS+60%PCL	1	52.3	Incineration	3.60	[29]
TPS foam grade	1	32.4-53.5	Composting	0.89	[134]
			Waste water treatment plant	1.43	[134]
			Composting	1.21	[135]
TPS+50%PBS/A	1	n/a	70.5%landfill.+10%compost.+0.5%litter +19% reuse	0.80	[129]
TPS+50% PBAT	1	n/a	70.5%landfill.+10%compost.+0.5%litter +19% reuse	0.92	[129]
TPS film grade (50%TPS+PCL)	1	n/a	70.5%landfill.+10%compost.+0.5%litter +19% reuse	1.18	[129]
HDPE	1	80	Incineration	4.84[c]	[98, 136]
			99.5% landfill.+ 0.5% litter	2.92	[129]
LDPE	1	81	Incineration	5.04[c]	[98, 130]
		92	80% incin.+ 20% landfill.	5.20[c]	[133]
			97.5%landfill.+2%recycle +0.5%litter	2.65	[129]
EPS	1	84	Incineration	5.88[c]	[98, 137]
		88	Non-(cradle-to-factory gate)[b]	2.80	[134]
PET	1	77	Incineration	4.93[c]	[98, 112]
PCL	1	77-83	Incineration	3.1-5.7[c]	[135, 138]
PVOH	1	58-102	Incineration	2.7-4.3[c]	[134, 138]

Abbreviation: TPS = thermoplastic starch; PVOH = polyvinyl alcohol; PCL = polycaprolactone; PBAT = polybutylene adipate terephthalate; EPS = expandable polystyrene; HDPE = high density polyethylene; LDPE = low density polyethylene; PET = polyethylene terephthalate

[a] Total of process energy and feedstock energy. Non-renewable energy only, i.e. total fossil and nuclear energy. In the "cradle-to-factory gate" concept the downstream system boundary coicides with the output of the polymer or the end product. Hence, no credits are ascribed to valuable by-products from waste management (steam, electricity, secondary materials).

[b] No credit for carbon uptake by plants.

[c] Only CO_2 embodied carbon: 3.14 kg CO_2/kg PE, 2.34 kg CO_2/kg Nylon6, 2.29 kg CO_2/t PET, 3.38 kg CO_2/t PS, 2.32 kg CO_2/t PCL, 2.00 kg CO_2/t PVOH.

Table 3.11: Summary of energy and GHG emissions for per functional unit plastic products; products listed are all commercial products manufactured by state-of-the-art technologies

Type of plastic	Functional unit	Cradle-to-gate NREU (MJ/FU)	Type of waste treatment assumed for calculation of emissions	GHG emissions (kg CO_2 eq./FU)	Reference
Loose fills					
Starch loose fills	1 m^3 (10 kg)	492	Waste water treatment plant	21	[134]
Starch loose fills	1 m^3 (12 kg)	277	30% incin.+70% landfilling	33.5	[133]
EPS loose fill	1 m^3 (4.5 kg)	680	Incineration	56	[134]
EPS loose fill	1 m^3 (4 kg)	453	30%incin.+70% landfilling	22.5	[133]
EPS loose fill (by recycling of PS waste)	1 m^3 (4 kg)	361	30%incin.+70% landfilling	18.6	[133]
Films					
TPS film	100 m^2	649	80% incin.+20%landfilling	25.3	[132]
PE film	100 m^2	1340	80% incin.+20%landfilling	66.7	[132]
Grocery bags[b]					
50% starch + PBS/A (single use)	3.12 kg	n/a	70.5% landfill.+10% compost.+0.5%litter + 19%reuse	2.5	[129]
50% starch + PBAT (single use)	3.12 kg	n/a	70.5% landfill.+10% compost.+0.5%litter + 19%reuse	2.88	[129]
50% starch + PCL (single use)	4.21 kg	n/a	70.5% landfill.+10% compost.+0.5%litter + 19%reuse	4.96	[129]
HDPE (single use)	3.12 kg	n/a	78% landfill.+2% recycle +0.5%compos.+ 19%reuse	6.13	[129]
PP (multiple use)	0.48 kg	n/a	99% landfilling + 0.5% litter	1.95	[129]
LDPE (multiple use)	1.04 kg	n/a	97.5% landfill.+2% recycle + 0.5% litter	2.76	[129]

[a] Total of process energy and feedstock energy. Non-renewable energy only, i.e. total fossil and nuclear energy. In the ”cradle-to-factory gate” concept the downstream system boundary coicides with the output of the polymer or the end product. Hence, no credits are ascribed to valuable by-products from waste management (steam, electricity, secondary materials).

[b] The functional unit is defined as the grocery bags needed for “a household carrying approximately 70 grocery items home from a supermarket each week for 52 weeks; the functional unit is determined by the weight, the capacity (volume) and the lifetime of the bag. The volume of TPS bags and HDPE singlet bag are same (6-8 items); the volume of the PP bag is 1.2 times the volume of the HDPE singlet bag; and the volume of LDPE bag is three times the volume of the HDPE singlet bag. All the TPS bags and the HDPE singlet bag are for single use; the PP bag is multi use and has a life time of 2 years; and the LDPE bag is multi-use and has a life time of 1 year [129].

Table 3.12: Energy recovery by incineration and net energy input of starch polymers

Type of Plastic	Share of petrochemical compounds % (wt)	Cradle-to-factory gate NREU[a] MJ/kg product (A)	Calorific value[b] MJ/kg product (B)	Net NREU input with 100% energy recovery= $A-B$	Net NREU input with 50% energy recovery= $A-0.5B$
TPS	0	25.4	13.6	11.8	18.6
TPS/PVOH	15	24.9	15.0	9.9	17.4
TPS/PCL	53	48.3	18.6	29.7	39.0
TPS/PCL	60	52.3	19.2	33.1	42.7
LDPE	100	80.6	43.3	37.7	59.0

[a] Non-enewable energy (total fossil fuel and nuclear), including feedstock energy; these values are listed in Table 3.10; they originate from different sources.

[b] Calorific values of TPS and LDPE originate from [114].

3.6 Conclusions

In this paper we reviewed LCA studies for polysaccharide products including textile products, natural fibre composites and starch polymers. In the review we chose non-renewable energy use (NREU) and greenhouse gas (GHG) emissions as important indicators for environmental profiles of products. The conclusions of this review are:

1. From cradle to factory gate, the comparisons per kg material show that polysaccharides can offer important potentials for NREU savings and GHG emission reduction. In the application area of textiles, man-made cellulose fibres can save about 10-30% NREU relative to cotton and up to 50-80% NREU relative to PET if the comparison is made on a kg basis. As engineering materials, natural fibre composites can save about 25-30% NREU and reduce 3-40% GHG emissions compared to glass fibre composites on a kg basis. Also, the higher the fibre content in natural fibre composites, the lower NREU and GHG emissions are. For packaging materials, on a kg basis, TPS can save about 25-75% NREU and reduce 20-70% GHG emissions compared to virgin petrochemical polymers (±15% depending on whether HDPE, LDPE or LLDPE is the reference).

   Making use of results for individual products we estimated to which extent polysaccharidebased products have already offered savings of NREU and GHG emissions in the EU-25 today. As shown in Table 3.13 (first and second column from the right) a total of around 30 PJ (26-34 PJ) non-renewable energy have been saved and approximately 0.1-1.2 million tonnes CO_2 emissions have been avoided. Man-made cellulose fibres, due to their large production volume, account for about 80% of the total energy savings and almost 70% of the total GHG emission reduction.

2. Because polysaccharide materials have lower density than petrochemical polymers, the material (in mass terms) required to fulfil the same end use is usually higher than that of petrochemical products. Consequently, cotton offers neither NREU savings nor GHG emission savings compared to PET textile products; no data is available for the production of man-made cellulose textiles (end products). In contrast, natural fibre composites, from cradle to factory gate, do offer savings

Table 3.13: Cradle-to-factory gate NREU and GHG emission savings by the novel polysaccharide-based materials in the EU 25.

Polysaccharide products	Energy saving (MJ/kg)	GHG emission saving (kg CO_2 eq./kg)	Production volume in EU (kt)[a,b]	Total energy savings in EU (PJ)	Total GHG emission savings in EU (kt CO_2 eq.)
Man-made cellulose fibres[c]	55 – 65[d]	0.4 – 2.2[e]	416	23 – 27	170 – 195
Total natural fibre composites in automotive applications[f]	28 – 65	−0.9 to 2.5[g]	85	2.4 – 5.5	−76 to 213
Starch polymers[h]	23 – 52[i]	1.1 – 3.7[j]	30	0.7 – 1.6	33 – 111
			Total savings	26 – 34	122 – 1240

[a] Production volume in year 2003, except for man-made cellulose fibres, which is for the year 2005.
[b] See Table 3.1.
[c] Viscose fibre vs. polyester fibre.
[d] See Table 3.3.
[e] Own calculation based on [107, 109]
[f] China reed or hemp composite vs. fibreglass composite or ABS.
[g] Own calculation based on [122, 124].
[h] Own calculation based on Table 3.10.
[i] TPS vs. LDPE or LLDPE.
[j] TPS vs. LDPE, LLDPE or HDPE.

of NREU and GHG emissions when the comparison is made with conventional composites at the level of the end product. Also in the case of packaging products, TPS offers NREU and GHG emissions savings over petrochemical polymers. There are two exceptions for this conclusion. First, single-use TPS products cannot compete with multi-use petrochemical products. Second, (virgin) TPS can hardly compete with recycled petrochemical polymers.

3. For the use phase, the NREU and the GHG emissions of polysaccharide products strongly depend on the application. For textiles, due to the higher density, cotton requires more energy for washing than polyester and hence, the polysaccharide product is disadvantageous during the use phase; no data is available for the use phase of man-made cellulose textiles. Natural fibre polymer composites can enable weight reduction in vehicles and therefore strongly improve the fuel economy during the use phase. For packaging, there is no significant difference during the use phase between starch polymers and petrochemical polymers, because the weight difference is minimal while the volume is the limiting factor for transportation (e.g. capacity of a lorry).

4. For waste treatment at the end of life, incineration with energy recovery is a common option for both polysaccharide and petrochemical products. Although the calorific values of polysaccharides are much lower compared to many petrochemical polymers (e.g. compared to PE or PA while the difference is marginal for PET), polysaccharide materials show a lower net NREU than petrochemical polymers if the comparison is on a kg basis. Landfilling without CH_4 capture,

which is another common waste treatment in some regions, can cause higher GHG emissions for starch polymer products than for petrochemical products due to the methane emissions from bio-degradation.

To summarise, it is not possible to generalise with absolute certainty that polysaccharide-based products are better than their petrochemical based counterparts from an environmental point of view, but important advantages do exist. The overall conclusion can be drawn that from cradle to grave, in terms of non-renewable energy requirements and GHG emissions, the polysaccharide products are better than their conventional counterparts, which are mostly petrochemical-based materials. Cotton is the exception because its cultivation requires a relatively high amount of water and chemicals. Up-to-date information on the production and use of man-made cellulose textiles is rarely available, calling for further studies on the environmental impact assessment covering the overall life stages of production, use and waste management.

Chapter 4

Environmental impact assessment of man-made cellulose fibres*

4.1 Introduction

The production of textile materials has undergone dramatic changes in the last century. Prior to the industrial revolution in the 19th century, natural materials, e.g. cotton, animal furs and silk, had been used for thousands of years. In the first decades of the 20th century, cotton accounted for more than 70% of all textile raw materials in the world [139]. It was not until the 1930s that man-made cellulose fibres became one of the principle fibres. After World War II, the production of man-made cellulosics kept increasing, until in the 1960s synthetic fibres "swept" the textile market (Figure 4.1). In the meantime, water and air pollution caused by toxic compounds darkened the image of the man-made cellulosics [14]. After decades of fierce competition, man-made cellulose fibres have maintained their characteristic position in the market and are now primarily used for high-value applications.

Man-made cellulosics are synthetic polymers made from natural resources. Wood pulp and cotton linters are the common raw materials. Viscose staple fibres are by far the most important man-made cellulose fibres. In 2002, the world-wide man-made cellulose fibre production was 2800 kilo tonnes, of which staple fibres (including viscose, modal and lyocell) accounted for 62%, viscose filaments (including modal) 13%, acetate tow 21%, and acetate & cupro filaments 4% [142, 143]. The viscose process has set the standard for quality, variety and price which other man-made cellulose fibres had to compete with [14]. In this study we analyze the man-made cellulose fibres produced by Lenzing AG. The company produces 1/5th of the world's man-made cellulose

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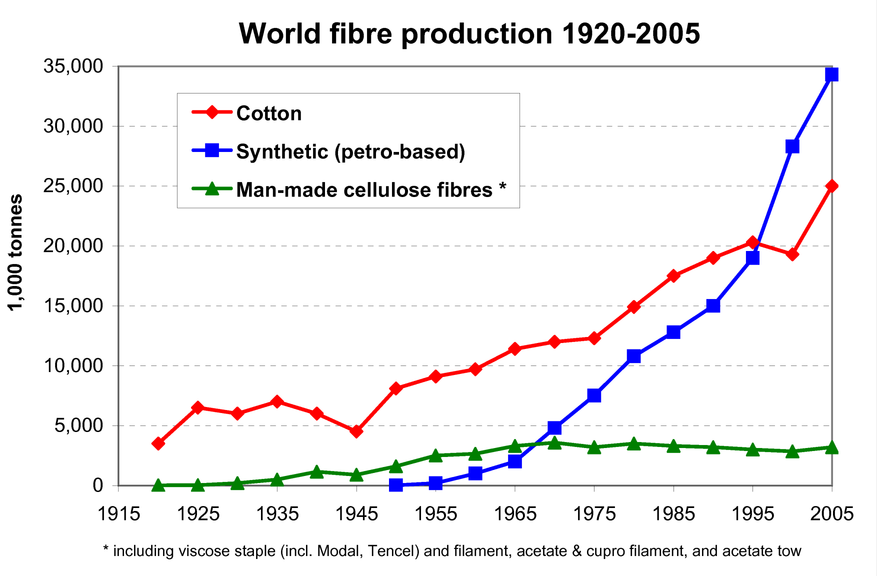

Figure 4.1: World fibre production 1920 – 2005 [14, 15, 66, 139, 140, 141]

fibres by the application of three generations of technologies: the conventional viscose process (Lenzing Viscose®), the modified viscose process – for high wet modulus fibres (Modal®), and the Lyocell process (Tencel®),[1] which is a solvent based process that was commercialized in the early 1990s. All three generations of fibres are produced nowadays simultaneously in large quantity.

The purpose of this study is to assess the environmental impact of man-made cellulose staple fibres using the tool of life cycle assessment (LCA).[2] In the past, several studies have compared the energy consumption for the production of natural, man-made and synthetic fibres [111, 107]. However, the data used in those studies are dated and the studies are limited to energy use while not covering any environmental impacts. Moreover, all publicly available studies address viscose only in general terms and do not distinguish between viscose, modal and lyocell. In the past decades, efforts have been made to optimize and integrate the production process for viscose and modal fibres, reducing pollution and improving material and energy efficiency. The lyocell process is a novel production process; it does not use toxic compounds as reagents (e.g. CS_2); and it substantially reduces total chemical use (e.g. NaOH). The

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[1] Lenzing Viscose®, Modal® and Tencel® are registered trade names by Lenzing AG. In this paper, Viscose, Modal and Tencel (capitalized spelling) refer to the trade names. The production processes and generic fibre names are spelt with lower cases, i.e. viscose, modal and lyocell.

[2] Staple fibre refers to short fibres that can be spun into yarn [144]. The term originated from natural fibres, e.g. cotton and wool. For man-made fibres, the primary spinning results in continuous filament fibre, which can be cut short to produce staple fibre.

environmental impacts of cellulose fibres produced via modern production and process technologies have not been assessed so far.

A further reason for preparing this study is that the production volume of man-made cellulose fibres is by far larger than all the other man-made biobased polymers together (except for paper and board) [145]. Bio-based materials have attracted much attention in the last few years due to concerns of the environment and the limited fossil fuels resources. Comparative environmental assessments between man-made cellulose fibres, conventional cultivated natural fibres (e.g. cotton) and fossil fuel-based synthetic fibres do not exist so far.

Against this background, the research questions addressed by this paper are:

1. What are the cradle-to-factory gate environmental impacts of the three types of man-made cellulose staple fibres, i.e. Viscose, Modal and Tencel? Compared to cotton, PET (polyethylene terephthalate) and PP (polypropylene) fibres, what are the advantages and disadvantages of man-made cellulose fibres from an environmental point of view?

2. Which steps in the process chain contribute most to the overall environmental burden of man-made cellulose staple fibres?

4.2 Goal, functional unit and system boundary

We applied the method of LCA to assess the environmental impacts of man-made cellulose fibres. LCA has been standardized by the ISO 14040 series [146, 147].

The goal of this LCA is to assess the environmental impacts of three types of man-made cellulose fibres produced by Lenzing, namely, Viscose, Modal and Tencel. The functional unit is defined as one metric tonne of staple fibres. The system boundary of this LCA is cradle to factory gate. This includes all steps from the extraction of raw materials and fuels, followed by all conversion steps until the product (staple fibre) is delivered at the factory gate. When delivered at the factory gate, Viscose and Modal fibres have a moisture content of 11%; Tencel fibre has moisture content of 13%. Table 4.1 introduces the five product systems analyzed in this article:

1. Viscose (Asia) is produced in Asia, using market pulp produced from eucalyptus wood planted in the southern hemisphere. The fibre plant is representative for the state-of-the-art separate viscose fibre plant based on wood pulp in the world.

2. Viscose (Austria) is produced in Austria. The most important difference between Viscose (Asia) and Viscose (Austria) is the integration of pulp and fibre plants. The Austrian viscose fibre plant is integrated with a pulp mill, while pulp and fibre production are separate in the case of Viscose (Asia); in this sense, Viscose (Austria) represents the best available technology of the current global viscose fibre production.

3. Modal is also produced at the integrated site in Austria. Both Viscose (Austria) and Modal use wood from Europe. In the integrated pulp/fibre plant, process

energy is supplied by internal biomass (e.g. bark and thick liquor from the pulp production), external biomass, municipal solid waste incineration (MSWI, with external waste from municipalities), and a very small amount of fossil fuels.

4. Tencel is produced based on the Lyocell process in Heiligenkreuz in Austria. Both imported pulp and pulp from the Lenzing mill are used. The process energy for the current Austrian Tencel production is supplied by natural gas (70%) and external biomass (30%).
5. In addition, we include a future Tencel fibre - Tencel (2012) - in the analysis. It is physically the same Tencel fibre as is produced today, but it will be produced using a different energy supply. According to Lenzing AG, in 2012 the process energy of Tencel fibre will be entirely based on energy recovered from municipal solid waste incineration (MSWI).

Furthermore, we compare the LCA results of conventional cotton, PET and PP. The inventory data of conventional cotton is based on the weighted average of US cotton and Chinese cotton. The cotton production in these two countries represented about 43% of world cotton production in the season 2004/2005 [148]. For PET and PP fibres, we assume production in Western Europe. Table 4.1 gives an overview of the product systems analyzed and compared in this article.

Table 4.1: Product systems included in this study: types and geographic scope of man-made cellulose fibres in comparison with cotton, PET and PP

Man-made cellulose fibres

Names used in this article	Trade name (fibre type[a])	Wood	Pulp	Fibre plant	Process energy
Viscose (Asia)	Lenzing Viscose (viscose)	Eucalyptus	Market pulp	Asia	Local electricity, coal, gas, oil
Viscose (Austria) Modal	Lenzing Viscose (viscose) Modal (modal)	Beech	Integrated pulp and fibre production in Austria		Biomass, recovered energy from MSWI
Tencel	Tencel (lyocell)	Eucalyptus and Beech	Mixed Lenzing pulp & Market pulp	Austria	70% gas and 30% biomass
Tencel (2012)	Tencel (lyocell)				100% recovered energy from MSWI

Other commodity fibres

Names used in this article	Type	Geographic scope	Data source
Cotton (US&CN)	Natural fibre	US & CN	Literature data (see Section 4.3.2)
PET (W.Europe)	Polyester	Western Europe	
PP (W.Europe)	Polyolefin	Western Europe	

[a] Fibre designation according to ISO/TC 38.

Staple fibres are not end products, but are important semi-finished products which are ready to be converted to many textile end products. The properties of the staple fibres studied are not identical (see Table 4.2). The end applications of these fibres may not be exactly the same. Ideally, the comparison should hence be made for relevant end products, which is beyond the scope of this paper. It is recommended to take into account the specific properties of fibres when using this LCA study for decision making.

Table 4.2: Selected mechanical, thermal and water rentention properties of staple fibres

Fibre name	Trade name	Density (g/cm^3)	Tenacity[a] (wet) (cN/tex)	Tenacity[a] (dry) (cN/tex)	Water retention (%)	Melting point (°C)
Cotton[b]		1.5 – 1.54[144]	26 – 40[149]	24 – 36[149]	38 – 45[143]	~400[144]
Viscose	Lenzing Viscose	1.52 – 1.54[144]	10 – 13[149]	24 – 36[149]	90 – 100[143]	n/a
Modal	Modal	1.52 – 1.54[144]	19 – 21[149]	34 – 36[149]	60 – 65[143]	n/a
Lyocell	Tencel	1.50[144]	34 – 36[149]	40 – 42[149]	60 – 70[143]	n/a[c]
PET[144]	Dacron	1.36 – 1.41	30 – 55	28 – 55	3 – 5	250 – 260
PP[149]	Herculon	0.9 – 0.92	25 – 60	25 – 60	0	160 – 175

[a] Tenacity is a fineness indicator measured in tex. 1 tex = 1 g per 1000 m.

[b] Natural fibres such as cotton usually have some impurities in the staple fibre product (e.g. dust and ginning residues), whereas man-made fibres, both biobased and petroleum based, are produced from chemical processes and usually have very high purity. In general, man-made fibres do not need purification before further textile processing.

[c] n/a = data not available or not applicable.

4.3 Life cycle inventory

4.3.1 From trees to fibres

Wood is the raw material used by Lenzing for producing man-made cellulose fibres. Viscose (Austria), Modal and a part of Tencel are produced from Lenzing pulp, which is made from European beech plantation. Half of the beech wood is from Austria and most of the other half is from other European countries. The wood is transported by rail or truck to the integrated pulp and fibre plant located in the village Lenzing, Austria. Viscose (Asia) and Tencel are produced from imported market pulp, which originates from eucalyptus plantations in the southern hemisphere. The market pulp is transported by ship to the fibre production sites in Asia and Europe. The beech and eucalyptus plantation used for the pulp production have existed for more than 20 years (Personal communication with Lenzing AG and the market pulp supplier). Based on the definition chosen by the IPCC and PAS 2050, the GHG emissions from land use change are considered negligible.[3]

Data for European beech production were obtained from the Ecoinvent database [152]. According to Ecoinvent, the average yield of European beech wood is 3.4 oven-dried-tonne/ha-yr (including bark). The European beech is neither fertilized nor irrigated, and it is machine-harvested.

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[3] Both IPCC [150] and PAS [151] define that only the direct land use change occurring "on or after 1 January 1990" should be included.

Data for the production of eucalyptus wood are not available in the Ecoinvent database (version 1.3). Instead, literature data were collected (see Appendix 4.A) and they were cross-checked with the pulp producer. The eucalyptus plantation is not irrigated. Small amounts of nitrogen and phosphate fertilizers are applied. Since the amount of fertilizer use is relatively small, only the direct N_2O emissions are taken into account. Harvesting is mainly done (80%) by hand (Personal communication with the market pulp supplier). The harvested wood is transported from the forest to the pulp mill by rail and road.

The pulp used to produce man-made cellulose fibres is so-called dissolving grade pulp. The difference between dissolving grade pulp and paper grade pulp can be described as follows: in paper grade pulp, lignin and resins are removed from wood and the pulp contains both cellulose and hemicellulose; in contrast, dissolving pulp process removes not only lignin and resins, but also large amounts of hemicellulose, resulting in a very high content (90-94%) of alpha cellulose (Personal communication with Schmidtbauer J. of Lenzing AG). For dissolving pulp production, the acid sulphite or the Kraft process is used.

Man-made cellulose fibres are produced by the regeneration of alpha cellulose. Two types of technologies for cellulose regeneration, i.e. the viscose process and the lyocell process, are applied to produce three types of man-made cellulose fibres, namely Viscose, Modal and Tencel. The two technologies are illustrated in Figure 4.2.

In the viscose process, pulp is first alkalized in caustic soda, then depolymerised and reacted with carbon disulfide (CS_2) to form cellulose xanthate, which is dissolved in caustic soda. After filtration, degassing and ageing, the viscose solution is ready to be spun from a precipitation bath containing sulphuric acid, sodium sulphate and zinc sulphate. Here, cellulose is regenerated in filament form. Classic spinnable xanthate solution contains 7–10% cellulose, 5–7% sodium hydroxide (caustic soda, NaOH), 25–35% CS_2 [14]. The solution is spun into regular Viscose fibres in an acid salt bath (80g/l H_2SO_4, 150–300g/l sodium sulphate ($NaSO_4$), 10–20g/l $ZnSO_4$ at 45–55°C [14]. In Modal's production, the xanthate solution contains 6–8% cellulose, 6.5–8.5% NaOH and 30 – 40% CS_2; small amounts of modifier may also be added (Personal communication with Schmidtbauer J. of Lenzing AG) [14]. Modal fibres are spun into filament in a slightly acidic bath of low temperature and with a strong coagulating effect [14]. The viscose process requires a large amount of caustic soda (0.5 – 0.8 kg NaOH per kg fibre) [153] and leads to sodium sulphate (Na_2SO_4) as by-product. Nowadays up to 70% of the CS_2 is directly recycled and reused. Most of the remaining 30% is converted into sulphuric acid which is also recycled to the process (Personal communication with Schmidtbauer J. of Lenzing AG).

The viscose process has been applied on industrial scale since the 1930s and nowadays the process is used for the production of both Viscose and Modal fibres. Modal fibres are manufactured by a modified viscose process with a higher degree of polymerization and modified precipitating baths [14]. This leads to fibres with improved properties such as higher wet strength and better washability.

The lyocell process represents a complete technology innovation. NMMO (N-methyl-morpholine-N-oxide) is used to dissolve pulp and regenerate cellulose. The process has a nearly closed solvent cycle (see Figure 4.2). This not only avoids the use

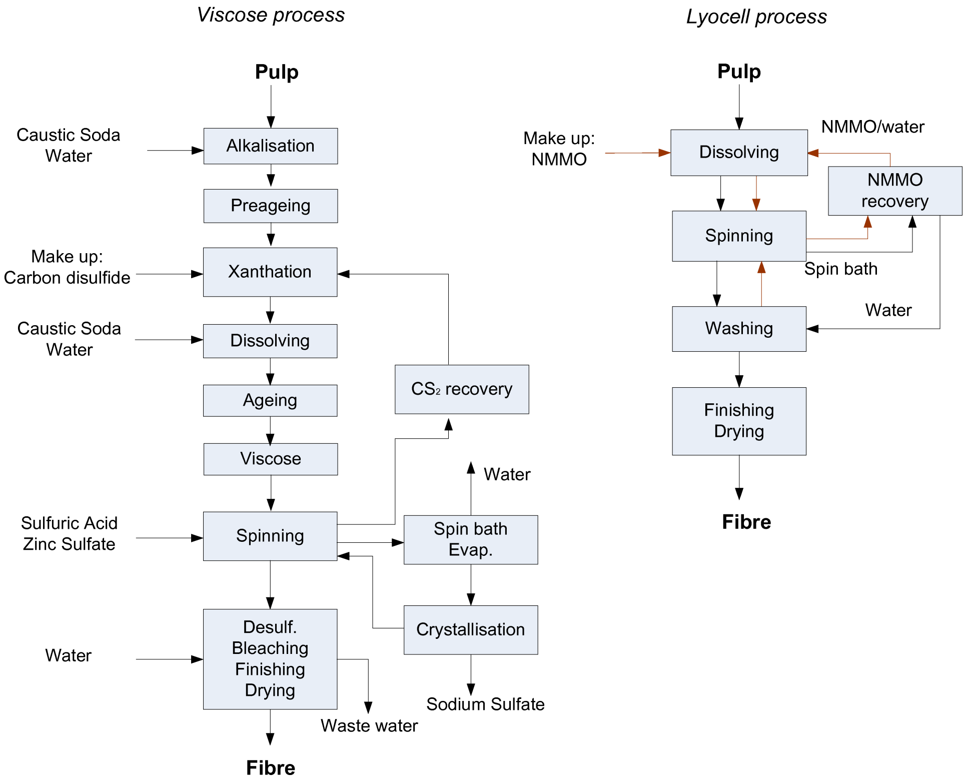

Figure 4.2: The viscose process and the lyocell process

of the highly toxic solvent CS_2, but also reduces the number of the process steps and total chemical use.

Figure 4.3 shows the two different production systems: integrated (for Viscose (Austria) and Modal) and separate production (for Viscose (Asia) and partially Tencel). In the case of integrated production, process energy use has been highly optimized and only a small amount of fossil fuels is required. Bark, thick liquor and soda extraction liquor from the pulping process are used fuel the pulp and fibre production. The remaining heat requirements (about 40% of the total heat requirements) are covered by externally purchased bark and a municipal solid waste incineration plant which is located next to the integrated plant. The integrated production is self-sufficient in terms of electricity use.

In this article, the pulp produced from the integrated plant is called Lenzing pulp. Lenzing pulp yields several wood-derived by-products, i.e. xylose, acetic acid, furfural and thick liquor;[4] it is not traded in the market.

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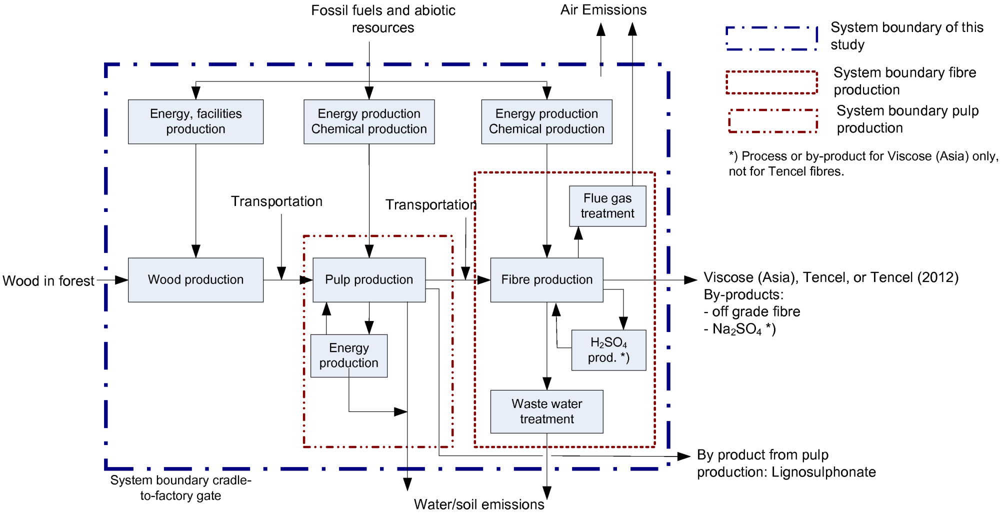
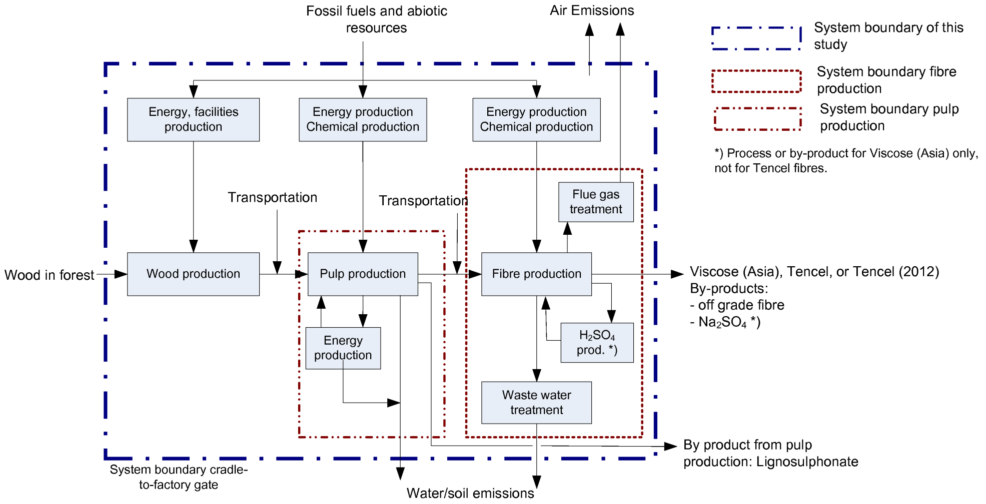
[4] The major part of the thick liquor is used to fuel the integrated pulp and fibre production process. A small amount of thick liquor is sold as a by-product.



(a) integrated pulp-fibre plant



(b) separate pulp and fibre production

Figure 4.3: System description of man-made cellulose fibre production

More commonly, pulp and fibre are separately produced. Viscose (Asia) and part of the Tencel fibres are produced from market pulp. For average market pulp, the pulping process yields lignosulphonate as the by-product (see Figure 4.3b). A substantial amount of the process heat and power is provided by combustion of thick liquor.

Additional process energy is provided by combustion of fossil fuels. Small amounts of electricity are purchased from public grid.

In a fibre plant of the separate production system, fossil fuels are the main energy source. For example, in the fibre plant of Viscose (Asia), over 99% of the process heat and power originate from fossil fuels, which are mainly coal and oil; nearly half of the electricity is supplied from public grid.

4.3.2 Data and assumptions

The data can be categorized into two levels: site-specific data and region-specific data. Site-specific data are collected from the production sites, i.e. energy and material balances of the pulp and fibre production, and the distance and the means of transportation. Site-specific data were provided by Lenzing AG for the preparation of the LCI (life cycle inventory) of market pulp, Lenzing pulp and the cellulose fibres. Unless otherwise reported, the technological level assumed represents the mid 2000s. These site-specific data are confidential and therefore will not be reported in this article. Region-specific data refer to the country or regional level. For example, electricity from the public grid is used for the production of NaOH. The data on the regional electricity mix and efficiencies were obtained from the Ecoinvent database (version 1.3), IEA statistics and scientific reports. A summary of data used can be found in Table 4.3.

The LCI data on cotton were obtained from unpublished work by Dinkel and Stettler (for US Cotton) and the Ecoinvent database (version 2.0, for Chinese cotton) [21]. The allocation between cotton fibre and cotton seed is conducted based on mass and economic values, resulting in factors of 85% and 15%, respectively.

The inventory data for the production of amorphous PET and of PP resin were obtained from PlasticsEurope [112, 163], representing average Western European production in the 2000s. The life cycle inventory of PET and PP consists of both polymer production and fibre spinning. The energy requirements of the spinning process are based on data from [164]. These data are relatively dated, but they have been validated by several industrial experts.

4.3.3 Allocation principles

Allocation is needed where one process has multiple products. In this study, three types of allocation problems are distinguished: allocation of by-products from pulp and fibre production, allocation of energy from waste incineration, and other multi-output situations.

Allocation of by-products from pulp and fibre production

According to ISO guidelines, allocation should be avoided if possible [147]. This can be done by applying system expansion, i.e. by assuming that the product would be produced otherwise by standard technology. In the integrated Viscose production, acetic acid is one of the by-products from the pulp production. The standard method to produce acetic acid is from petrochemical ethylene. By applying the system expansion

Table 4.3: Summary of data used in this study

	Data source	Notes
Pulp production (Lenzing pulp, market pulp)	Lenzing AG	Site specific
Fibre production (viscose, modal and tencel)	Lenzing AG	Site specific
Public grid power, country specific	Ecoinvent database (Version 1.3) [154] and IEA energy statistics [17, 155]	European electricity mix (used for NaOH and other chemical production): 65% from the UCTE grid, 13% from the NORDEL grid, 9% from the CNETREL grid, 12% from the UK grid and 1% from the Irish grid.
Public grid heat, country specific	Ecoinvent database (Version 1.3) [156]	Grid heat from industrial gas boiler
Production of chemicals (e.g. caustic soda)	Ecoinvent database (Version 1.3) [157]	Region specific (Europe, Asia)
Production of fuels	Ecoinvent database (Version 1.3) [156, 158, 159]	Region specific (Europe)
Transportaion	Ecoinvent database (Version 1.3) [160]	Including road, rail, barge and transoceanic transportation
Municipal solid waste incineration	Ecoinvent database (Version 1.3) [161]	Average Switzerland
Energy recovery from MSWI (for post-consumer waste incineration)	Literature data and personal communication with Dr. D.O. Reimann of CEWEP and [162]	Average Western Europe
Cotton, conventionally cultivated	US cotton: unpublished work by Dinkel and Stettler; see also the appendix in [145]. CN cotton: Ecoinvent (Version 2.0) [21]	Cradle-to-factory gate cotton fibre production including tillage, planting, fertiliser and pesticide use, harvesting, transportation, ginning and baling
PET, PP polymer production	PlasticsEurope publication [112, 163]	Average Western Europe
Energy requirements of PET and PP fibre spinning (from resin)	0.64 kWh electricity and 5 MJ heat (from fossil fuel) based on Brown et al. [164]	This energy data have been cross-checked by several polyester fibre industrial experts

Abbreviations: UCTE stands for Union for the Co-ordination of Transmission of Electricity; countries included in UCTE are Austria, Bosnia and Herzegovina, Belgium, Switzerland, Germany, Spain, France, Greece, Croatia, Italy, Luxemburg, Macedonia, Netherlands, Portugal, Slovenia and Serbia and Montenegro. NORDEL stands for Nordic countries power association, including Denmark, Norway, Finland and Sweden. CENTREL stands for Central European power association, including Czech Republic, Hungary, Poland and Slovakia.

method, credits are assigned to the integrated pulp and fibre production, representing the avoided petrochemical production of acetic acid. This method is applied for acetic acid and sodium sulphate (Na_2SO_4) for the integrated production process.

It is not possible to apply the system expansion method for the wood-derived by-products (e.g. xylose), for which the Lenzing production process represents the standard production technology. Instead, we apply economic allocation, i.e. the assigned environmental impacts are proportional to the economic values of the by-products. Apart from the wood-derived chemicals, economic allocation is applied for off-grade fibres (fibres with lower quality and sold at a discount).

To summarize, the default allocation method applied for by-products is system expansion; if not applicable, economic allocation is used. In the sensitivity analysis (see Section 4.6.1) we will additionally discuss alternative approaches, including system expansion in combination with calorific-value allocation and economic allocation without system expansion.

Allocation of heat from waste incineration plant

In the integrated Viscose and Modal production, as well as in the future Tencel production, part of the energy is supplied in the form of heat from a municipal solid waste incineration (MSWI) plant, operated with external waste. The MSWI plant consumes a small amount of fossil fuels and it delivers two services: waste disposal service and energy production. We now discuss three methods, namely two allocation approaches and one method based on system expansion.

For this allocation problem, physical allocation based on energy, exergy or mass is not possible because the two services are not comparable in terms of energy, exergy or mass content. Therefore, allocation based on economic values is applied. According to Lenzing AG, the average cost (not including collection and transportation) for the disposal of one tonne of solid waste in a MSWI plant in Austria is 200 euro (value for 2006). Assuming that the price of recovered heat is the price paid for heat from fossil fuels, which is the case in Switzerland according to Doka [161], the income from selling the recovered heat is about 17.5% of the total income of a MSWI plant; 82.5% can hence be assigned to the waste disposal service of the MSWI plant. We use this allocation method as the baseline method (default method) in our analysis (see Table 4.4).

The income composition of a MSWI plant, however, differs across regions and countries. In Switzerland, the revenue from energy recovered from waste incineration is only 5 – 10% of total revenue of a MSWI plant [161]. Therefore, Ecoinvent assigns no environmental burden to energy recovery from waste incineration, while the waste disposal service bears the entire environmental burden. In this study, we chose the Ecoinvent approach as our “free heat” case (see Table 4.4).

One could also argue that using heat from waste incineration is a coincidental situation. If there was no on-site MSWI plant, the fibre production would most likely obtain the required heat from a natural gas-fired boiler. We use system expansion approach as the third method, which is shown in Table 4.4 as the “natural gas” case.

Table 4.4: Allocation methods for heat from MSWI

Name	Method	Description
Baseline	Econimc allocation	17.5% of environmental burden of MSWI plant is assigned to recovered heat
“Free heat”	Economic allocation	0% of environmental burden of MSWI plant is assigned to recovered heat
“Natural gas”	System expansion	Heat is supplied by a natural gas-fired boiler

Other multi-output processes

The allocation methods described in Table 4.4 are also used for other types of combustible solid wastes, which are generated during the pulp/fibre production process. They are then disposed of by a MSWI plant or a HWI (hazardous waste incineration) plant with energy recovery.

Caustic soda (NaOH), which is one of the most important input materials for the pulp, Viscose and Modal production, is produced as one of the co-products (the other one is hydrogen) of chlorine production through the electrolysis of an aqueous solution of sodium chloride. We use data from the study of PlasticsEurope [165] in which the allocation to chlorine, caustic soda and hydrogen is carried out on a mass basis. Using this approach, about 52% of the impacts of the electrolysis process are assigned to NaOH (46% are assigned to Cl_2 and 1.3% are assigned to H_2).[5]

4.4 Environmental impact assessment methods

4.4.1 Energy, land use, water use and the CML baseline method

In this paper, cumulative energy demand (CED) represents cradle-to-factory gate primary energy [167]. Primary energy is "energy found in its original or natural form" [168]. CED is the sum of non-renewable energy use (NREU) and renewable energy use (REU). NREU is the total of fossil fuel (oil/gas/coal) and nuclear energy (uranium); REU consists of biomass, solar, hydro and wind. Cumulative fossil energy demand is a good proxy for overall environmental impacts [106].

The indicator of land use in this article refers to the land use for biomass production, i.e. agricultural and forest land use. Land use for infrastructure (e.g. land use of a fibre plant or a cotton spinning factory) and land use for transportation are excluded. We report forest and agricultural land use as separate inventory results, because a suitable impact assessment method of land use does not exist so far.

Water use is expressed as the sum of original natural fresh water consumption. Sea water is excluded in this study. Three categories of water use during the production are taken into account, namely, process water, cooling water and irrigation water. So far no aggregation methods have been developed to assess the impacts of water consumption. The three types of water consumption may have different environmental impacts. Process water usually requires higher quality; energy and materials input are required for the production [166]. For cooling water, the energy use of pumping is included; the water loss due to the evaporation is not taken into account. Irrigation water may have strong direct impact on local hydrological systems (e.g. depletion of local available surface or ground water). Using surface water and ground water for irrigation has different impacts such as soil salination, depending on the local soil condition [169]. In this article, we report the three types of water consumption as separate inventory results.

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[5] Allocation based on economic values of caustic soda, chlorine and hydrogen is not suitable because all three co-products develop in different markets; the price ratios vary extremely [166].

In this study, the following impact categories are selected from the CML baseline method [170]: global warming potential (GWP100a) [171] abiotic depletion, ozone layer depletion, human toxicity, fresh water aquatic ecotoxicity, terrestrial ecotoxicity, photochemical oxidant formation, acidification and eutrophication.

4.4.2 Normalization

Normalization is an optional step in an LCA according to ISO 14040 and 14044. This step allows determining the relative contribution of the product system to the impact categories at a national, regional or global level. To this end, the results per impact category are divided by the respective values for a given area (e.g. Europe or world). The normalized results do not imply weighting among impact categories, they merely give an indication to which extent the product system studied contributes to the total environmental loads of a region in a year. In this article, the CML global normalization factors for 2000 [172] were used.

4.4.3 Single-score analysis

Single-score analysis is an optional step according to the ISO guideline. A single-score analysis leads to an overall conclusion of an LCA by means of introducing subjective weighting factors for different environmental categories. In this study, three single score methods are introduced. Single scores I and II are based on the same weighting factors (always equalling 1) among the different environmental categories. Single score III is based on different weighting factors.

Single score I assumes that all the environmental categories are of the same importance. The score is directly derived from the mid-point results. The environmental categories are the selected CML baseline indicators plus land use and water use (see Section 4.4.1). The energy indicators (NREU, REU and CED) are excluded from this single score in order to avoid the double counting of abiotic depletion. As a reference, the score of cotton is set to 100. No normalization step is applied. The calculation according to this method is shown in Equation 4.1.

Similar to Single Score I, Single score II assumes that all the environmental categories are of the same importance. The mid-point results are, however, divided by the normalization factors before the total scores are added up (see Equation 4.1). Again, the score of cotton is set to 100 for comparison. Single score II includes the environmental categories of the CML baseline method. Water use and land use are excluded because they are not impact indicators.

Single score III uses the NOGEPA[6] weighting factors (see Appendix 4.B). It should be noted that this set of weighting factors represents the opinion of the Dutch oil and gas industry. Climate change is hence the most important environmental issue for the energy industry. It is, however, not necessarily the case for the fibre industry to have the same opinion as the energy industry. We nevertheless use this set of weighting factors because the opinions from the fibre industry are not available.

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[6] NOGEPA stands for Netherlands Oil and Gas Exploration and Production Association.

$$SingleScore_i = \frac{\sum_j \frac{E_{i,j}}{N_j} \times W_j}{\sum_j \frac{E_{cotton,j}}{N_j} \times W_j} \qquad (4.1)$$

In which:

i - the fibre type;

j - the environmental theme (e.g. abiotic depletion, eutrophication, etc.);

E - the mid-point result of the environmental impacts (in kg substances equivalent/tonne fibre);

N - the normalisation factors (in kg substances equivalent); for Single Score I, $N = 1$; for Single Score II and III, N are the CML normalisation factors for world in the year 2000 [172]; and

W - the weighting factor, $W = 1$ for each environmental category for Single Score I and II. The weighting factors used in Singe Score III are obtained from NOGEPA study (See Appendix 4.B).

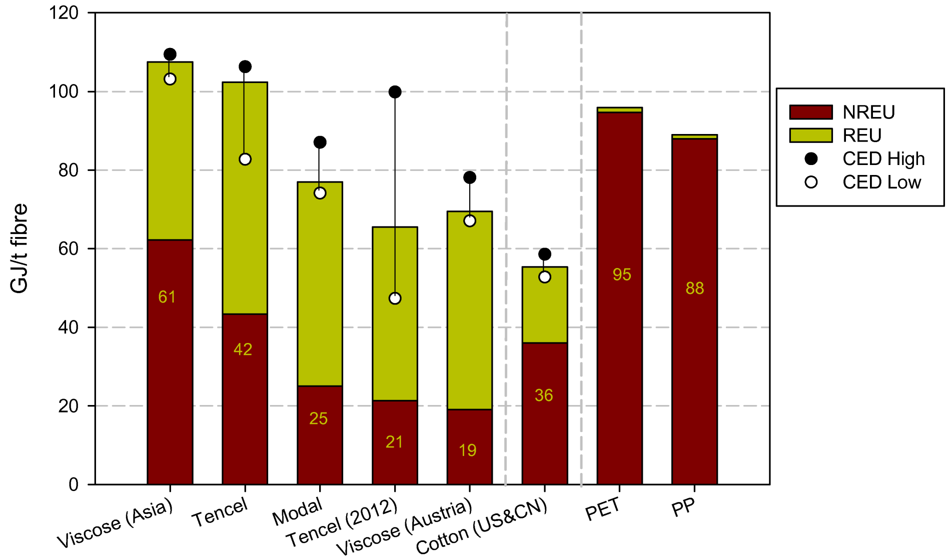
4.5 LCA results

4.5.1 Energy, water use and land use

The result of primary energy requirements is shown in Figure 4.4. The NREU of all cellulose fibres are lower than those of the synthetic fibres (PET and PP). Viscose (Austria) has the lowest NREU and PET has the highest. Cotton is not an energy-intensive product; it has slightly lower NREU than Tencel. Viscose (Asia) requires 70% more NREU than cotton. Modal, Tencel (2012) and Viscose (Austria) require 30%, 40% and 50% less NREU than cotton, respectively.

The particularly large value for Viscose (Asia) is related to the relatively inefficient coal-based heat and power production (Figure 4.5). Next to fossil fuel use in fibre production, the production of chemicals (including caustic soda) is the second most important factor. For Viscose (Austria) and Modal, process energy from fossil fuels does not play such an important role. Instead, caustic soda accounts for more than half of the NREU (Figure 4.5). Other important processes are the production of other chemicals such as sulphur, CS_2 and NaOCl (sodium hypochlorite). Compared to the viscose process, the lyocell process requires substantially less chemicals in fibre production (see Figure 4.2 and Figure 4.5). With more than 70% of the total NREU, natural gas contributes most to the NREU of the current Tencel fibre. In 2012, the NREU of Tencel will decrease by half (from 42 GJ/t to 21 GJ/t), if process energy is entirely supplied by energy recovered from external MSWI (see Figure 4.5).

Cotton has the lowest CED among all fibres studied. This refers, however, to the calculation based on the default allocation method; a different allocation method (see



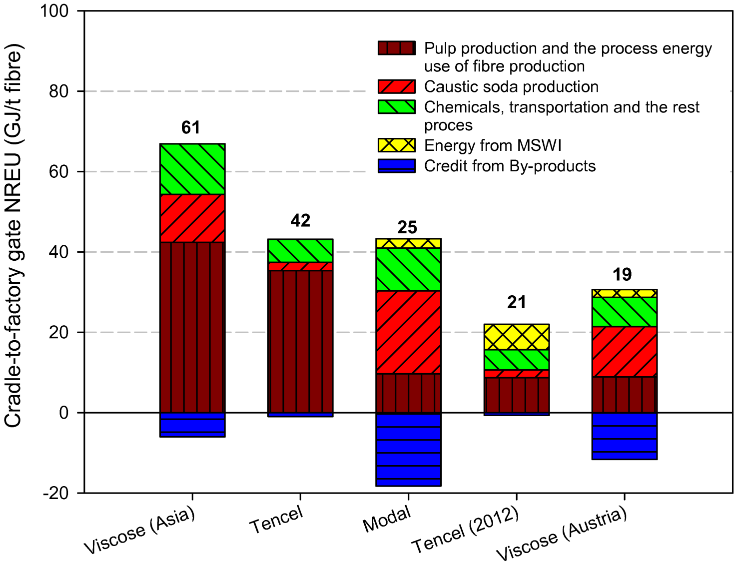
- The uncertainty ranges of Viscose (Asia) and Tencel originate from the mix sources of market pulp.
- The uncertainty ranges of Viscose (Austria) and Modal: the lower ranges show the results based on the “free heat” case and the higher ranges show the results based on the “natural gas” case (see Table 4.4).
- The relatively large uncertainty range of Tencel (2012) is a result from the combination of the mix market pulp sources and allocation methods applied for the energy obtained from MSWI.

Figure 4.4: Cradle-to-factory gate primary energy requirements (NREU, REU and CED) of one tonne of staple fibre (default allocation method for by-products)

Table 4.4), as well as a different mix of market versus Lenzing pulp may change the ranking (see the uncertainty ranges in Figure 4.4). Man-made cellulose fibres require relatively large REU compared to cotton, PET and PP. This is caused not only by the renewable feedstock which is embedded in product, but also by the large amount of biomass energy used in the production.

Figure 4.6 shows the results of land use. The cellulose fibres which use the European wood require more land than the eucalyptus wood-based fibres, because of the low forestry biomass yields in Europe compared to the high yields in warmer regions. Tencel requires more land than Tencel (2012). The difference is caused by the saved land due to the use of process heat from MSWI instead of biomass. Applying simple addition of the various types of land, we find that cotton required most land among all fibres studied.

In Figure 4.6 , land use for wood plantations in Europe and in the southern hemisphere, and for agriculture are reported in the same chart, even though the environmental impacts of land use is different depending on types of land, local climate and local ecosystem. In this study we limit ourselves to the inventory analysis (quantity). We



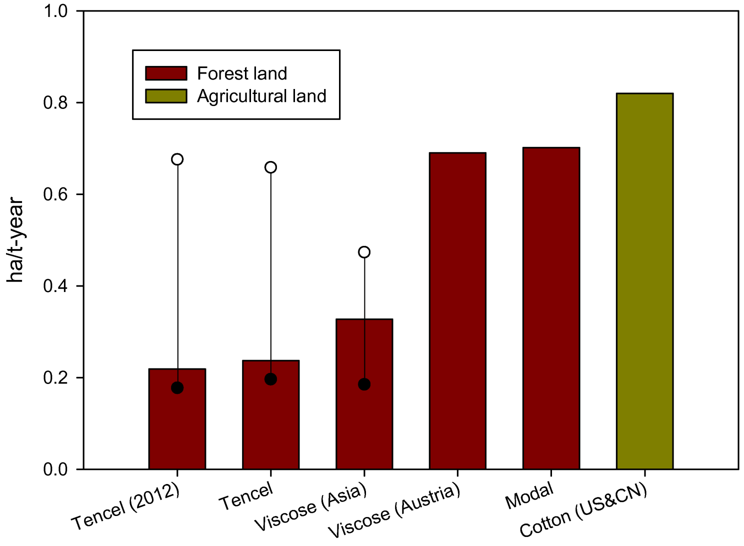
Note: the number reported above each column represents the total NREU and was calculated by deducting the negative bar section from the positive bar section

Figure 4.5: Breakdown of cradle-to-factory gate NREU of Lenzing man-made cellulose fibres (default allocation methods for by-products)

do not assess the overall environmental/ecological impacts of land use because suitable aggregation methods are still missing.

Table 4.5 shows the water use of the fibres studied. The water use of cellulose fibres is dominated by cooling water, accounting for about 90 – 95% of the total water consumption. The remaining 5 – 10% is process water which is the sum of softened water, deionized water, decarbonizes water and tap water. No irrigation is needed for the plantation in Europe and neither for the eucalyptus plantation. Allocation between fibres and by-products has been conducted based on economic values. If we exclude cooling water, cotton's water consumption is over 100 – 500 times higher than the water use of man-made cellulose fibres. If we include cooling water, the water use of cotton still about 10 – 20 times higher than the water use for man-made cellulose fibres. Almost all (>99%) water used by cotton is for irrigation. For the average Chinese and US cotton, about 70% of the irrigation water originates from ground water and 30% from surface water (see Figure 4.6).

It should be noted that the environmental impacts of various forms of water use are rather different. For example, cooling water does not cause local fresh water resource depletion, but irrigation water may do. Moreover, irrigation with ground water and with surface water (and also irrigation efficiency) may have different impacts depending on the local hydrological conditions. Moreover, irrigation may cause environmental impacts such as soil salination and water shortage downstream the river [169]. The



Note: The uncertainty range shows the land use assuming different sources of pulp (various sources of market pulp and/or Lenzing pulp).

Figure 4.6: Land use for biomass production for one tonne of staple fibre (economic allocation for by-products)

Table 4.5: Water use of one tonne of staple fibre, based on natural water origin (m^3 per tonne of fibre, default allocation method for by-products)

Type	Fibre	Process water	Cooling water	Irrigation water
Petrochemical fibres	PP (W.Europe)	<2	74	-
	PET (W.Europe)	<5	125	-
Man-made cellulose fibres	Viscose (Asia)	11	308	-
	Tencel (2010)	20	243	-
	Tencel	20	243	-
	Viscose (Austria)	42	403	-
	Modal	43	429	-
Cotton	Cotton (US&CN)	<5	37	5690 (4300-6860)[a]

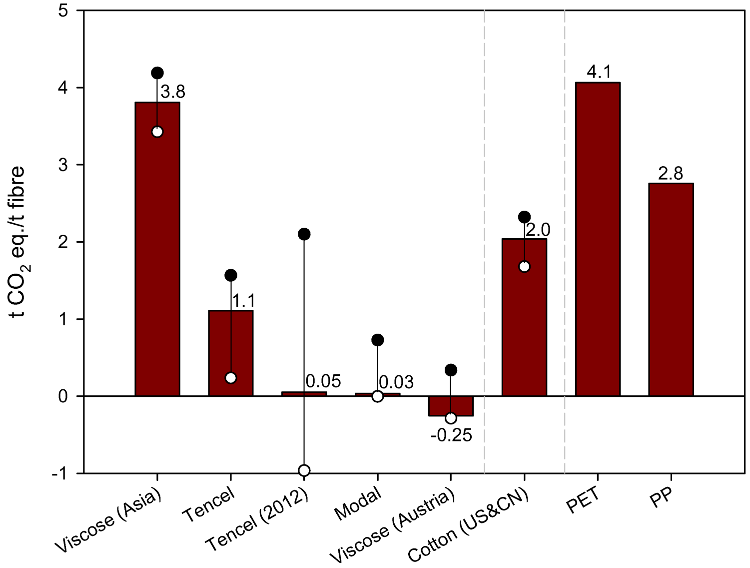
[a] The lower range represents the average US cotton, the higher range represents the average Chinese cotton.

data shown in Table 4.5 should hence be interpreted as inventory data instead of being considered environmental impacts.

4.5.2 CML baseline method

GWP100a

Figure 4.7 shows the results of cradle-to-factory gate GWP100a. All man-made cellulose fibres have lower GWP than PET. Tencel, Tencel (2012), Modal and Viscose (Austria) have the lowest GWP among all fibres studied. Modal and Tencel (2012) have nearly zero GWP. Viscose (Austria) has a negative GWP, which means that it sequestrates more carbon into the product than it emits. This refers, however, to the default calculations for which the entire heat demand is covered by a municipal waste incineration plant; a different allocation for the source of the heat used, as well as a different mix of market versus Lenzing pulp may change the ranking (see the uncertainty ranges in Figure 4.7).



- The uncertainty ranges of Viscose (Asia) and Tencel originate from the mix sources of market pulp.
- The uncertainty ranges of Viscose (Austria) and Modal: the lower ranges show the results based on the “free heat” case and the higher ranges show the results based on the “natural gas” case (see Table 4.4).
- The relatively large uncertainty range of Tencel (2012) is a result from the combination of the mix market pulp sources and allocation methods applied for the energy obtained from MSWI.

Figure 4.7: Cradle-to-factory gate GWP 100a of one tonne of staple fibre

Figure 4.8 shows the process contribution. For Viscose (Asia) the market pulp and the process heat and power used in fibre production account for more than three quarters of its total fossil carbon emissions; the production of caustic soda and other chemicals are also important processes. For Viscose (Austria) and Modal, the most important fossil carbon emissions are from the production of caustic soda; it accounts

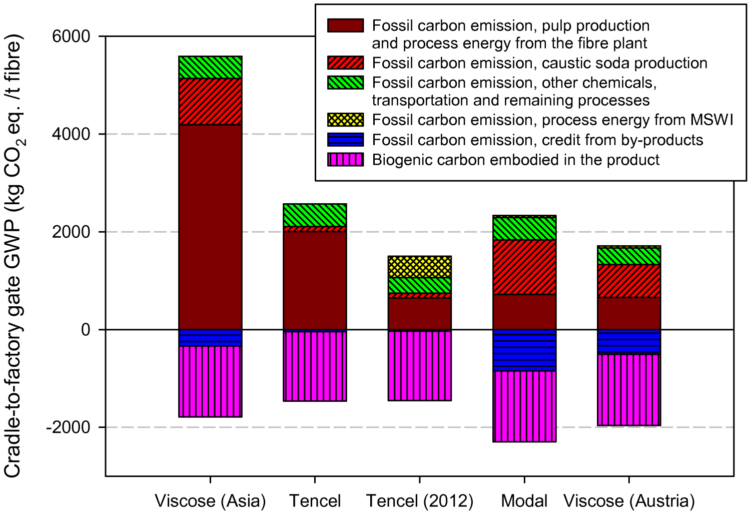


Figure 4.8: Process contribution of cradle-to-factory gate GWP of one tonne of man-made cellulose fibres

for more than half (for both fibres) of the total fossil carbon emissions. The low GWP of Modal is caused, to a large extent, by the avoided fossil carbon emissions from the by-products (especially Na_2SO_4 and acetic acid); the avoided carbon emissions are about 60% of total fossil carbon emissions of Modal. For Tencel, more than half of the fossil carbon emissions originate from natural gas combustion for process heat. The GWP of Tencel (2012) is reduced more than 90% compared to Tencel today by replacing the energy source.

Other CML impact categories

Table 4.6 shows the LCA results for other CML 2 baseline impact categories.

Abiotic depletion

PET and PP have the highest abiotic depletion impact due to the fossil fuel use. Viscose (Asia) also has a relatively high impact on abiotic depletion. Coal, market pulp and caustic soda account for nearly 60% of the abiotic depletion impact. More generally, for the fibres produced with viscose process, caustic soda, CS_2 and sulphur production are the most important factors next to process energy use. For fibres based on the lyocell process (i.e. Tencel), the process energy and market pulp are the most important factors, while the material consumption plays a less important role.

Ozone layer depletion

In this study, processes which require oil as input have a relatively high ozone layer depletion impact because of the Halon emissions from crude oil production. Halon is used

Table 4.6: Cradle-to-factory gate environmental impact assessment of one tonne of staple fibre, CML 2 baseline method

	Cotton	PET	PP	Viscose (Asia)	Viscose (Austria)	Modal	Tencel	Tencel (2010)
Abiotic depletion (kg Sb eq./t)	17	45	42	40	14	18	20	7
Ozone layer depletion ($\times 10^{-4}$ kg CFC11 eq./t)	2.0	0.7	0.7	2.8	0.3	0.4	1.1	0.7
Human toxicity (kg 1,4-DB eq./t)	1700	4393	369	1490	630	765	470	660
Fresh water aquatic ecotoxicity (kg 1,4-DB eq./t)	17310	58	53	160	74	93	85	75
Terrestrial ecotoxicity (kg 1,4-DB eq./t)	1568	12	12	16	11	16	5.0	5.0
Photochemical oxidant formation (kg C_2H_4 eq./t)	0.7	1.0	0.6	1.8	0.5	0.5	0.6	0.4
Acidification (kg SO_2 eq./t)	41	21	11	45	14	15	17	13
Eutrophication (kg PO_4^{3-} eq./t)	22	1.2	1.0	2.3	1.2	1.3	1.8	1.9

in fire extinguishing systems, especially in the Middle East, Russia and Africa [158].[7] Viscose (Asia) has the highest impact of ozone layer depletion of all fibres studied. Approximately 95% of the impact of Viscose (Asia) is related to the oil consumption for transportation, process fuels and the production of grid electricity.

Human toxicity, fresh water aquatic ecotoxicity and terrestrial ecotoxicity

For the toxicity impacts of cotton (i.e. human toxicity, freshwater aquatic ecotoxicity and terrestrial ecotoxicity), we only consider the impacts from US cotton. The reason is that Chinese cotton uses different pesticides and fertilizers and many of them cannot be assessed with the CML methods, which would cause underestimation. We therefore decide to use the toxicity impacts of US cotton as a proxy for the toxicity impacts of cotton. However, this approach most probably still underestimates the toxicity impacts of Chinese cotton, because US cotton farming has to comply with stricter legal requirements on fertilizer and pesticide use than many other conventional cotton cultivations in the rest of the world.

PET fibre has the highest human toxicity impact. More than 90% of the impact is caused by air emission of PAH (polycyclic aromatic hydrocarbon) in amorphous PET production [112]. For the human toxicity of viscose fibres, the most important processes are the production of caustic soda, market pulp and external electricity use. These three factors account for more than 70% of the total human toxicity impact of Viscose (Asia). These factors cause little or no impact for the Tencel fibres. Tencel (2012) has a slightly higher human toxicity than Tencel because of the emissions from the waste incineration plant.

Cotton has the highest fresh water ecotoxicity and terrestrial ecotoxicity mainly

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[7] As laid down in the Montreal Protocol the production of Halon has been banned in developed nations since 1994 [173]. However, due to existing stocks, Halon 1301 is likely to continue to be emitted for about 25 years [174]. This implicates that the current status of the consumption of Halon still makes ozone layer depletion an important environmental issue in the next decade.

due to pesticides use. More than 80% of the freshwater aquatic ecotoxicity and more than 90% of the terrestrial ecotoxicity are caused by soil emission of one insecticide, i.e. aldicarb. According to a survey of USDA (US Department of Agriculture) on average about 0.67 lb of aldicarb per acre (approx. 0.75 kg/ha) was applied to about 19% of the cotton fields in the US in 2005 [175] (thus, averagely 0.14 kg aldicarb per hectare (=0.75 × 19%) has been applied).

For cellulose fibres, pulp and caustic soda production are the most important factors for fresh water ecotoxicity and terrestrial ecotoxicity. For all cellulose fibres studied, the credits related to by-products, especially Na_2SO_4 and acetic acid, significantly contribute to lower human toxicity impacts and fresh water aquatic ecotoxicity. Terrestrial ecotoxicity is not particularly influenced by the credits of the by-products.

Photochemical oxidant formation

For man-made cellulose fibres the most important factor for photochemical oxidant formation are the SO_2 emissions. These mainly originate from two sources: the emissions from the production of SO_2 (SO_2 is used as a process input for the pulp production) and the SO_2 emissions from energy production. Viscose (Asia) has the highest photochemical oxidant formation due to high SO_2 emissions from the energy production in the fibre plant – the SO_2 emissions from Viscose (Asia) are about 10 times higher than those from Viscose (Austria).

Acidification

Acidification is mainly caused by SO_2 emissions. For Viscose (Austria) and Modal, the SO_2 emissions from the fibre plant are 10 times less than those of Viscose (Asia). For Tencel fibres, there are no SO_2 emissions from the energy production in the fibre plant. The acidification impacts of Viscose (Austria), Modal, Tencel and Tencel (2012) mainly originate from the production of SO_2 in the pulp mill.

Eutrophication

Cotton has the highest eutrophication impact due to its fertilizer use. For cellulose fibres, pulp production and caustic soda production contribute most to eutrophication. These two processes contribute about 50% of the total impact for Viscose (Asia). About 80% of the eutrophication of Viscose (Asia) and Modal is caused by pulp production, caustic soda production and NOx emissions. For Tencel (2012), energy recovered from MSWI also makes an important contribution.

4.5.3 Normalized results

Figure 4.9 shows the normalized results. The normalized environmental impacts of ozone layer depletion and photochemical oxidant formation are nearly invisible. Furthermore, all man-made cellulose fibres studied cause comparatively insignificant impacts regarding human toxicity, fresh water aquatic ecotoxicity and eutrophication. None of the impact categories of Viscose (Austria) and Tencel represent a significant contribution. On the other hand, the normalized results show that for Viscose (Asia), global warming, abiotic depletion and acidification are relatively important environmental issues. For Modal and Tencel, relatively important environmental issues are abiotic depletion and acidification.

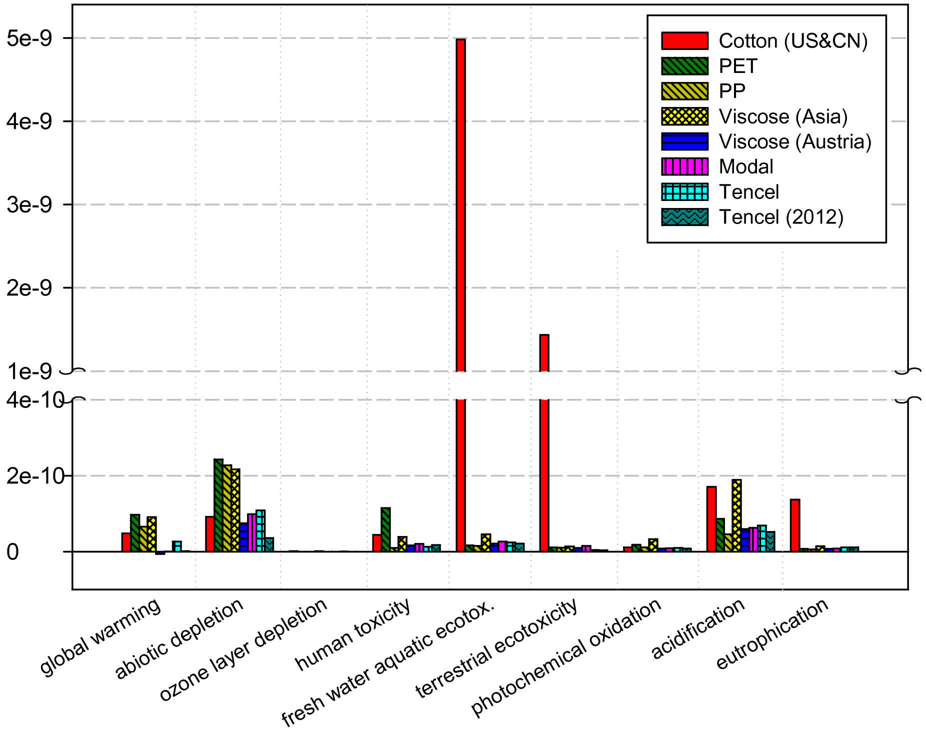

Figure 4.9: Comparing cradle-to-factory gate environmental impacts, one tonne of staple fibre, CML 2 baseline 2000 method, Normalization to World 2000

4.5.4 Single scores

Figure 4.10 shows the results according to Single score I. Viscose (Asia) scores slightly higher than, but is similar to cotton. The most important environmental impacts of Viscose (Asia) are GWP, ozone layer depletion, abiotic depletion and photochemical oxidation. However, the normalized result (see Figure 4.10) shows that ozone layer depletion and photochemical oxidation are less important environmental issues for fibres in a global context. Therefore, it is advisable to use an improved single-score method which uses the results of the normalization.

Single score II comes to a different conclusion than Single score I, in terms of the ranking between cotton and Viscose (Asia) (Figure 4.11). Here, cotton has by far the highest score due to its very high normalized ecotoxicity impacts. The fresh water

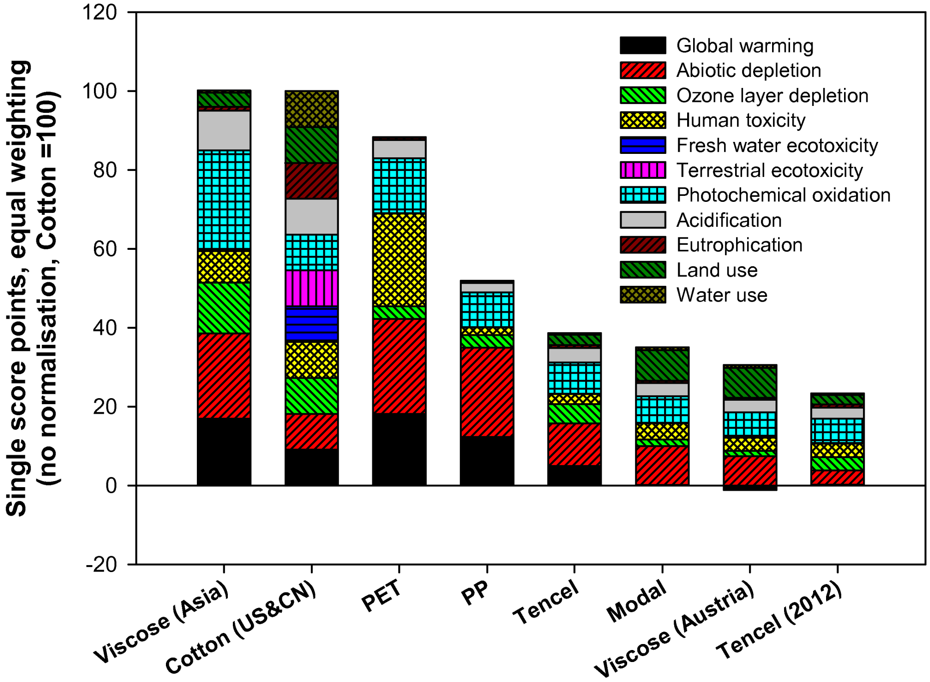

Figure 4.10: Single Score I, cradle-to-factory gate, no normalization, equally weighted eleven environmental impact categories (cotton =100)

ecotoxicity and terrestrial ecotoxicity of cotton account for 70% and 20% of the total impacts of cotton, respectively. Viscose (Asia), which has only 9% of the score of cotton, is ranked as the second least favourable choice by Single score II. The other four man-made cellulose fibres, namely Tencel, Modal, Viscose (Austria) and Tencel (2012), have very low scores compared to cotton (<5% of the cotton's score).

Single score III is constructed based on Single score II, using the weighting factors established by NOGEPA (see Section 4.4.2). As presented in Figure 4.12. Single score III shows a similar pattern as Single score II – cotton has the highest score and Tencel (2012) has the lowest score. Cotton is neither an energy-intensive nor GHG-intensive product. NOGEPA has rated global warming as the most important environmental category. Still, the high fresh water ecotoxicity and terrestrial ecotoxicity results in a high overall impact for cotton. Unlike Single score II, for Single score III Viscose (Asia) is placed after cotton and PET, although it is still comparable with PET in spite of the high impacts of abiotic depletion and GWP100a. Like the result from Single scores I and II, the other four man-made cellulose fibres, namely Tencel, Modal, Viscose (Austria) and Tencel (2012), have the lowest scores.

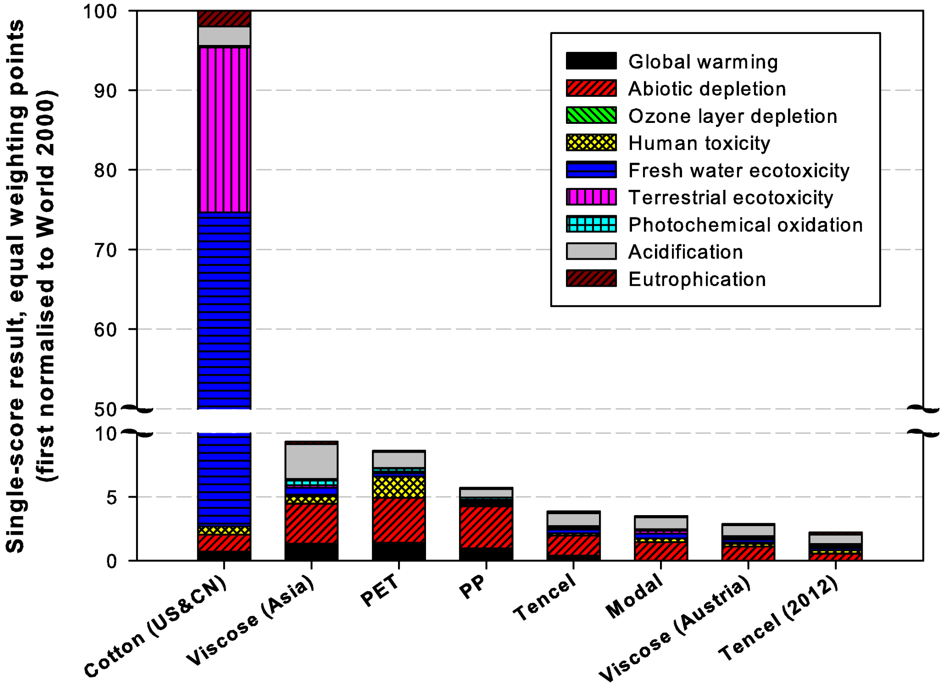

Figure 4.11: Single Score II, cradle-to-factory gate, first normalized to World 2000, equal weighting (cotton =100)

4.6 Discussion

4.6.1 Sensitivity analysis allocation methods

In Section 4.3.3 we explained the allocation method applied for the by-products from the integrated pulp and fibre production, namely system expansion in combination with allocation based on economic values. In this section, we compare the results based on two alternative allocation methods.

Alternative method 1: system expansion + allocation based on calorific values

As an alternative, we can also choose the approach of system expansion in combination with allocation based on calorific values, because by-products such as thick liquor, xylose and furfural (for which the system expansion method cannot be applied) can be otherwise considered as fuel.

The LCA results based on this approach can be found in Appendix 4.C. This approach leads to slightly higher allocation factors for by-products compared to the default approach. For Viscose (Asia), Tencel and Tencel (2012), the change in the

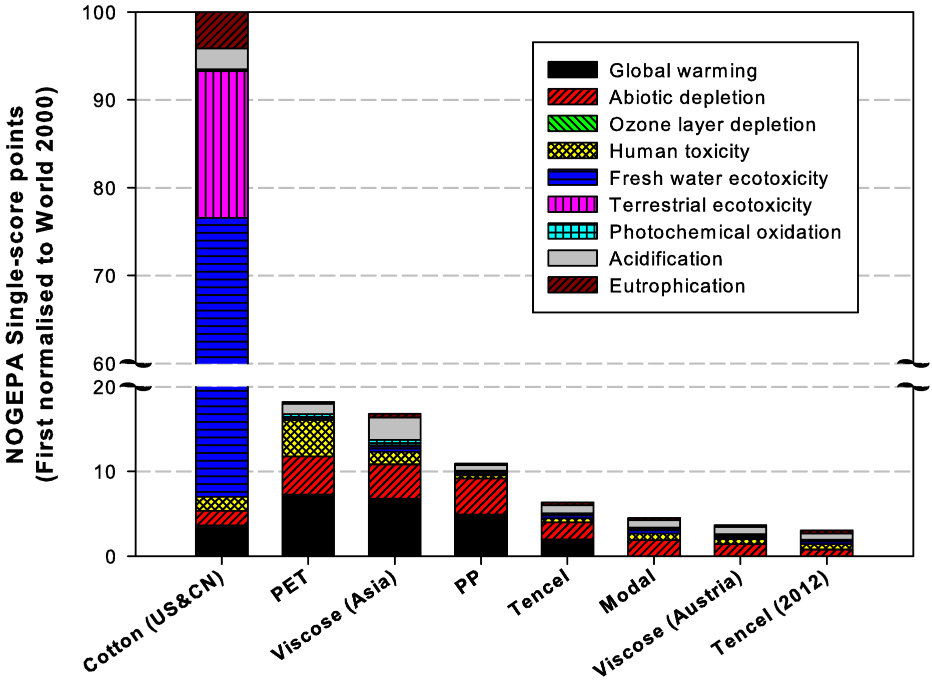

Figure 4.12: Single Score III, cradle-to-factory gate LCA result, first normalized to World 2000, NOGEPA weighting factors (Cotton =100)

environmental impacts is insignificant (<1%). The impacts of Viscose (Austria) and Modal decrease by about 8-9%. Figure 4.13 illustrates the analysis of the NREU of Viscose (Austria). The credits given for acetic acid and Na_2SO_4 are identical for both methods. The small difference is caused by the increase of the allocation factors for xylose, furfural and thick liquor. In comparison with the default method, the ranking relative to cotton, PET and PP does not change by this alternative allocation method.

Alternative method 2: allocation based on economic values (no system expansion applied)

In the second alternative approach, no system expansion is applied, i.e. no credits are given for acetic acid and Na_2SO_4. The total environmental burden is allocated based on economic values. This approach substantially increases the environmental impacts of the main product (fibres). The price of acetic acid and Na_2SO_4 are much lower than the fibre prices. Consequently, the environmental burden assigned to acetic acid and Na_2SO_4 is much smaller than the avoided environmental burden from the production of acetic acid and Na_2SO_4 (see Figure 4.13).

For Tencel and Tencel (2012), the changes of the LCA results are negligible (< ±3%,

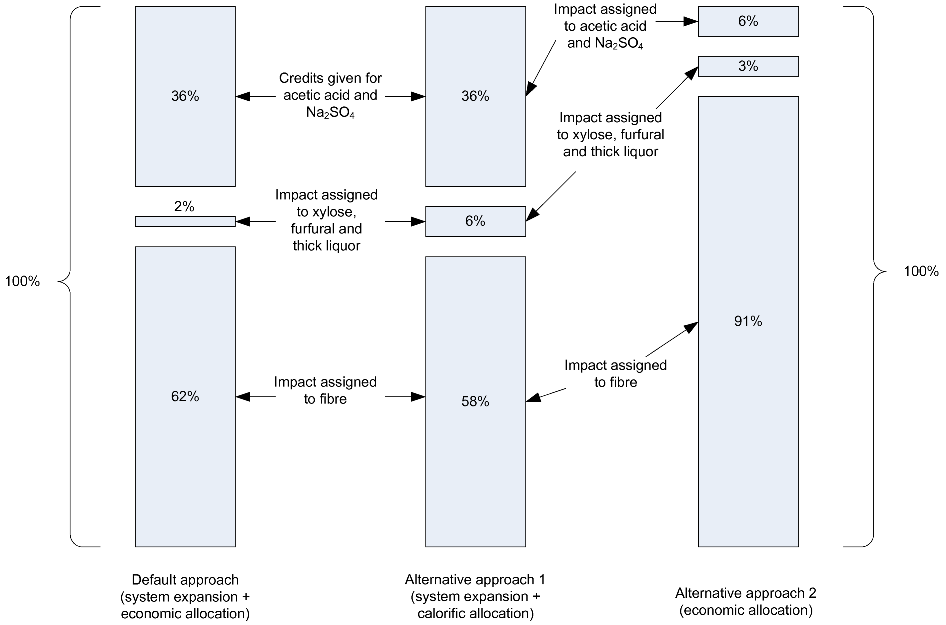

Figure 4.13: Comparison of the allocation of NREU of Viscose (Austria) based on the default method and two alternative methods

see Appendix 4.C). For Viscose (Asia), the changes are also insignificant (0.1-9%). For Tencel, Tencel (2012) and Viscose (Asia), the influence of this alternative method is minor. The ranking relative to cotton, PET and PP does not change.

For Viscose (Austria) and Modal, the LCA results change significantly. The LCA results increase by approximately 15-60% for NREU, abiotic depletion, human toxicity, fresh water aquatic ecotoxicity, photochemical oxidant formation and acidification. Ozone layer depletion is the most sensitive indicator to this allocation method – the impact increases by 140-170%. Appendix 4.C shows the complete result of this sensitivity analysis.

Although this allocation method increases the environmental impacts of Viscose (Austria) and Modal, the ranking relative to cotton, PET and PP does not change substantially. Viscose (Austria) and Modal still have lower scores for most of the impact categories than cotton, PET and PP. The exception is the NREU of Modal which becomes slightly higher than cotton (39 GJ/t vs. 36 GJ/t) and the photochemical oxidant formation of both Viscose (Austria) and Modal becomes comparable with that of cotton, PET and PP.

4.6.2 CML method, land use and water use

CML 2 baseline 2000 method

The CML baseline method has 10 environmental impact categories. In this study, we reported nine of them. The one that we excluded is marine aquatic ecotoxicity because of the data uncertainties in the characterization factors (Heijungs et al., 2004). Several studies have discussed these uncertainties of ecotoxicity from the CML method [118, 176, 177]. When using the LCA results of this study, readers are recommended to treat the toxicity impacts with caution.

Environmental impacts of land use and water use

The results of land use in this study only refer to the land use for biomass production. The impacts of land use are region-dependent. The yield of biomass may be influenced by many factors, such as climate, soil fertility, land use change (if relevant) and fragility or stability of the local ecosystem. It is beyond the scope of this study to look into these factors. Amongst all currently available impact assessment methods, only Eco-indicator 99 provides land use as one of the mid-point indicators. This method is not suitable for our analysis. In Eco-indicator 99 all forms of land use are assumed to occur within Europe (the environmental impact is also assumed to occur in Europe)[8] [100]. In our analysis, only the forest land use is located in Europe; while the eucalyptus is grown a country in the southern hemisphere; furthermore, cotton is cultivated in different climate zones in China and the US. So far, there are no generally accepted methods for aggregating different forms of land use.

We reported the water use (by natural origin) for each type of fibre. Water use causes different types of environmental impacts depending on the regional hydrological system, the local climate and ecological system. To our knowledge, there is no mature method for the aggregated assessment of different types of water use.

4.6.3 Scope extension: Cradle to factory gate plus post-consumer waste incineration

The system boundary of this LCA is "cradle to factory gate". After leaving the factory gate, fibres are used to produce various end-products. The end-products are then consumed and disposed of. In the waste stage, the used products may be incinerated, recycled or landfilled. Direct landfilling is prohibited in several European countries and will be phased out in the EU [178]. In this section, we extend the cradle-to-factory gate analysis by adding the waste management stage to the system boundary. We assume that by the end of life, the fibre product will be incinerated in an average MSWI facility in the EU, with or without energy recovery.

We introduce a new indicator for this analysis: Net NREU, which is defined as the total gross NREU for production minus the energy recovered from waste incineration,

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[8] According to the methodology description of Eco-indicator 99 (section 2.3.3, page 13) [100], the environmental impact (damage) of land use is expressed in Potential Disappeared Fraction (PDF) of species; the species numbers are determined by observations (counting), not by models.

i.e. the system boundary includes the cradle-to-factory gate stage and the waste management stage; the production and consumption of the end-products are excluded. In the case of an MSWI with energy recovery, the cogeneration of electricity and heat is assumed. The recovered energy replaces grid electricity and conventionally raised heat. Incineration with a recovery rate of 60% (in primary energy terms) has been estimated to represent the average level in Europe (Personal communication with Dr. D. O. Reimann of CEWEP) [162]. If no energy is recovered from the end-of-life MSWI, the Net NREU is equal to the cradle-to-factory gate NREU.

Table 4.7 shows the comparison of man-made cellulose fibres with other polymers in terms of Net NREU. In the case of MSWI with energy recovery, all studied man-made cellulose fibres have a lower Net NREU than PP and PET. This is remarkable because man-made cellulose fibres have a lower calorific value than petrochemical polymers and hence receive a lower credit from energy recovery in the waste management stage.

Similarly, Net GWP can be defined as the sum of the GWP of production and post-consumer waste incineration, minus the credit from energy recovery (processing of the staple fibre and the use phase are excluded). The analysis of Net GWP leads to a similar conclusion as for Net NREU. The analysis can be found in Appendix 4.D.

Table 4.7: Comparison of Net NREU (GJ/t fibre) of man-made cellulose fibres, cotton, PET and PP (including cradle-to-factory gate plus waste incineration with energy recovery, excluding fabric production and use phase)

	Cradle-to-factory gate NREU	Avoided NREU (MSWI with energy recovery)	Net NREU (MSWI with energy recovery)	Net NREU (MSWI without energy recovery)
Cotton (US&CN)	36	−10	26	36
PET	95	−14	81	95
Viscose (Asia)	61	−9	52	61
Tencel	42	−9	33	42
Modal	25	−9	16	25
Tencel (2012)	21	−9	12	21
Viscose (Austria)	19	−9	10	19

4.7 Conclusions

We conducted a comparative LCA to assess the environmental impacts of three types of man-made cellulose fibre (Viscose, Modal and Tencel). The LCA results are compared with the conventional textile fibres (cotton) and synthetic fibres (PET and PP). The environmental indicators we assessed include primary energy demand (NREU, REU and CED), land use, water use and the CML baseline impact categories. The functional unit is one tonne of staple fibre. The analysis was carried out for the system of cradle-to-factory gate. Three single score methods were introduced and applied. The main findings of this study are:

1. Based on all the mid-point results and three single scores, we conclude that all man-made cellulose fibres, except for Viscose (Asia), have better environmental profiles than PET, PP and cotton; Tencel (2012) has the lowest impact of all. Viscose (Asia) has a lower impact than cotton; it is comparable to PET, but less preferable than PP and other man-made cellulose fibres.
2. The environmental benefits of Viscose (Austria) and Modal are largely attributed to low fossil energy requirements in the pulp and fibre production. This is a result of process integration, the use of renewable energy and credits from by-products. Furthermore, Viscose (Austria) and Modal have much lower process emissions (e.g. SO_2 and NOx) compared to Viscose (Asia), leading to low human toxicity, photochemical oxidant formation, acidification and eutrophication. The environmental benefits of Tencel (2012) are the result of low energy consumption, low chemical use, low CO_2 emissions, low SO_2 emissions and low water consumptions, leading to low impacts on abiotic depletion, terrestrial ecotoxicity, photochemical oxidant formation and acidification.
3. Viscose (Asia) is less favourable than the other studied man-made cellulose fibres. The higher impacts of Viscose (Asia) are primarily attributed to process energy, the use of market pulp and local sourcing of chemicals while emissions from the viscose process are a minor contributor to the overall impact.
4. Cotton is not an energy-intensive product; it has the lowest CED of all fibres studied. However, cotton is ranked as the least favourable choice by Single scores II and III. The major environmental issues of cotton include land use, water use, fresh water aquatic ecotoxicity, terrestrial ecotoxicity and eutrophication. The use of pesticides in cotton cultivation causes most of the ecotoxicity impacts. Furthermore, the use of fertilizer is the main cause of the eutrophication impact.
5. Two alternative allocation methods are applied. Both lead to less favourable results for man-made cellulose fibres compared to the default method (see Section 4.6.1), although the ranking of all fibres studied does not change.
6. Based on the system of cradle-to-factory gate plus waste incineration with and without energy recovery, all man-made cellulose fibres studied are better than PET and PP in terms of Net NREU.

State-of-the-art LCA methodologies have been applied in this study. However, the quality of toxicity calculations in LCA tools is currently still doubtful and research is underway to improve the methodologies and to make the databases more complete. Furthermore, only environmental indicators that are generally considered in LCA studies have been taken into account. For example, an environmental indicator which has not been considered is the impact on biodiversity. Land use and water use have been exclusively reported as inventory results, i.e. different types of land (and water) have not been aggregated due to the lack of suitable methods. The risk of explosion has neither been taken into account.

In conclusion, modern man-made cellulose fibres have a clear potential in reducing the environmental impacts over cotton and petrochemical synthetic fibres.

4.A Appendix: Key data on the eucalyptus wood production in southern hemisphere

No.	Name of parameter	Value	Data Source	Note
1	Density	650 kg/m^3	Ecoinvent [152]	Dry mass, dry volume density for average hardwood.
2	Carbon content	49.1%	Ecoinvent [152]	Assumed the same as average European hardwood.
3	Bark content in wood	12%vol.	Ecoinvent [152]	
4	CO_2 sequestered in 1 m^3 wood	1319 kg CO_2	Calc. from 1-3	1 m^3 wood without debarking
5	Calorific value of eucalyptus wood	19.8 GJ/odt	Personal communication with the market pulp supplier	
6	Yield of eucalyptus wood	12 odt/ha/year	Personal communication with the market pulp supplier	
7	Fertiliser use		Personal communication with the market pulp supplier	
	N fertiliser	25 kg/ha/yr		
	P fertiliser	17 kg/ha/yr		
8	Machinery use	1.2 kg diesel/m^3 (dry matter)	Literature data [179] and personal communication with the market pulp supplier	
9	N_2O emissions from applying fertilisers	0.01 kg N_2O-N/kg N fertiliser	IPCC 2006 guidelines [150]	Direct emission from N fertiliser use

odt = oven dried tonne

4.B Appendix: Weighting factors based on the CML environmental themes from the Dutch Oil and Gas Exploitation and Production Association (NOGEPA)

Environmental theme[a,b]	Weight
Climate Change	32
Ozone layer depletion	5
Abiotic depletion	8
Human toxicity	16
Fresh water aquatic ecotoxicity	6
Terrestrial ecotoxicity	5
Photochemical oxidant formation	8
Acidification	6
Eutrophication	13
Total[c]	99

Note: [a]Abiotic depletion is not weighted by NOGEPA, we add it in because we consider it to be an important environmental theme in this study. [b]Marine aquatic ecotoxicity is not included in this study and thus not listed in this table. It is, however, weighted as 8 by the NOGEPA. [c]Data source: [180]. According to Huppes, et al. [180], totals do not add up to 100 due to rounding.

4.C Appendix: Comparison of LCA results based on different allocation methods for by-products (see Section 4.6.1).

	NREU (GJ/t)	REU (GJ/t)	CED (GJ/t)	GHG total (kg CO_2 eq./t)	Abiotic depletion (kg Sb eq./t)	Ozone layer depletion (kg CFC-11 eq./t)	Human toxicity (kg 1,4-DB eq./t)	Fresh water aquatic ecotoxicity (kg 1,4-DB eq./t)	Terrestrial ecotoxicity (kg 1,4-DB eq./t)	Photochemical oxidant formation (kg C_2H_4 eq./t)	Acidification (kg SO_2)	Eutrophication (kg PO_4^{3-} eq./t)
Default method: system expansion + economic allocation												
Viscose (Austria)	19	50	70	-250	14	3.0×10^{-5}	630	74	11	0.45	14	1.2
Modal	25	52	77	33	18	3.7×10^{-5}	765	93	16	0.48	15	1.3
Viscose (Aisa)	61	45	106	3810	40	2.8×10^{-4}	1490	160	16	1.81	45	2.3
Tencel	42	59	101	1110	20	1.1×10^{-4}	470	85	5	0.6	17	1.8
Tencel (2010)	21	44	65	53	7	7.2×10^{-5}	663	75	5	0.4	13	1.9
Alternative method 1: System expansion + Calorific allocation (Section 4.6.1)												
Viscose (Austria)	18	47	64	-345	13	2.8×10^{-5}	580	68	10	0.4	13	1.1
Modal	23	47	70	-90	17	3.4×10^{-5}	700	85	15	0.4	14	1.2
Viscose (Asia)	61	45	106	3800	40	2.8×10^{-4}	1490	159	16	1.8	45	2.3
Tencel	42	59	101	1090	20	1.1×10^{-4}	470	83	5	0.6	16	1.8
Tencel (2010)	21	44	65	43	7	7.2×10^{-5}	658	74	5	0.4	12	1.9
Alernative method 2: Economic allocation, without applying system expansion (Section 4.6.1)												
Viscose (Austria)	28	48	75	94	17	7.2×10^{-5}	750	100	12	0.6	17	1.2
Modal	39	49	88	650	23	1.0×10^{-4}	990	140	18	0.8	19	1.4
Viscose (Asia)	64	44	108	3910	41	3.0×10^{-4}	1550	170	16	1.8	46	2.3
Tencel	43	59	102	1115	20	1.1×10^{-4}	475	84	5	0.6	17	1.8
Tencel (2010)	22	44	66	58	7	7.4×10^{-5}	664	75	5	0.5	13	1.9
For comparison												
Cotton (US&CN)	36	19	55	2040	17	2.0×10^{-4}	1700	17310	1568	0.7	41	22
PET (W.Europe)	95	1	96	4063	45	7.0×10^{-5}	4393	58	12	1.0	21	1.2
PP (W.Europe)	88	1	89	2760	42	7.0×10^{-5}	370	53	12	0.6	11	1.0

4.D Appendix: Analysis of Net GWP (see Section 4.6.3)

Net GWP is defined as the total GWP for production and for post-consumer waste incineration. In the case of MSWI with energy recovery (recovery rate = 60%), the electricity and heat recovered from the calorific value of the waste avoids the use of fossil fuels combustion. Here, we assume that the avoided fossil fuel is natural gas. The emission factor of natural gas is 56 kg CO_2 eq/GJ [181]. Therefore, 1 GJ of primary fuel saved also avoids 56 kg CO_2 eq. of GHG emissions. The Net GWP is computed as cradle-to-factory gate GHG emissions plus the biogenic emissions from waste incineration minus the saved CO_2 emissions from energy recovery (34 kg CO_2 eq./GJ at a 60% recovery rate). In the case of MSWI without energy recovery, there are no avoided CO_2 emissions. The amount of GHGs emitted by incineration is equal to the CO_2 embedded in product.

The results of Net GWP are presented in Table-Appendix IV. In the case of MSWI with energy recovery, Tencel (2012), Modal and Viscose (Austria) have Net GWP of around or less than 1 tonne CO_2 eq./t fibre, which is substantially lower than Cotton, PET, PP, Viscose (Asia) and Tencel. Viscose (Asia) is similar to PET and PP but is less favorable than cotton. In the case of MSWI without energy recovery, PET and PP have the highest Net GWP among all fibres studied because of their high carbon content. All Lenzing man-made cellulose fibres except for Viscose (Asia) have lower Net GWP than cotton. Viscose (Asia) has a slightly lower Net GWP than PP but significantly higher than cotton.

Table 4.8: Comparison of Net GWP 100a (t CO_2 equivalent/t fibre) of man-made cellulose fibres, cotton, PET and PP (including cradle-to-factory gate plus waste incineration with energy recovery, excluding fabric production and use phase)

	Cradle to factory gate GWP	Embedded carbon in product (as CO_2 eq.)	Avoided GWP (MSWI with energy recovery)	Net GWP (MSWI with energy recovery)	Net GWP (MSWI without energy recovery)
Cotton (US&CN)	2.04	1.65	-0.55	3.13	3.70
PET	4.06	2.29	-0.80	5.56	6.35
PP	2.76	3.14	-1.64	4.25	5.90
Viscose (Asia)	3.81	1.45	-0.52	4.74	5.26
Tencel	1.11	1.42	-0.52	2.03	2.53
Modal	0.03	1.45	-0.52	0.97	1.48
Tencel (2012)	0.05	1.42	-0.50	0.97	1.47
Viscose (Austria)	-0.25	1.45	-0.5	0.69	1.20

Chapter 5

Open-loop recycling: An LCA case study of PET bottle-to-fibre recycling*

5.1 Introduction

Polyethylene terephthalate (PET) bottles have experienced rapid growth since the 1970s when the technique of blow moulding was introduced [8]. Today, bottle grade PET is one of the most important packaging plastics. In 2007, the worldwide consumption of bottle grade PET was 15 million metric tonnes (10^6 metric tonnes or Mt) [13], representing 8% of the total demand of standard plastics.[1] Meanwhile, recycling of post-consumer PET bottles has become a well-established system with its own logistic chain including bottles collection, flake production and pellet production. In 2007, approximately 4.5 Mt of PET bottles were collected and recycled into 3.6 Mt of flakes worldwide [42]. Most of the recycled PET flakes were converted into fibres (Figure 5.1). Recycled PET fibre accounted for approximately 8% of the world PET fibre production in 2007 [42, 66].

In Europe, the amount of collected post-consumer PET bottle waste has increased from 0.2 Mt in 1998 to 1.26 Mt in 2008 [183], representing an annual growth rate of approximately 19% (see Figure 5.2). About 40% of all used PET bottles in Europe were collected for recycling in 2009 [4]. It is expected that PET bottle waste collection in Europe will continue to increase by 10% p.a. in the near future [8] (see Figure 5.2).

The primary purpose of this study is to understand the environmental impacts of

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[1] According to PlasticsEurope's definition, "Standard Plastics" refer to standard thermoplastics, including PE (polyethylene), PP (polypropylene), PVC (polyvinylchloride), PS (polystyrene), EPS (expanded polystyrene) and PET (bottle grade).

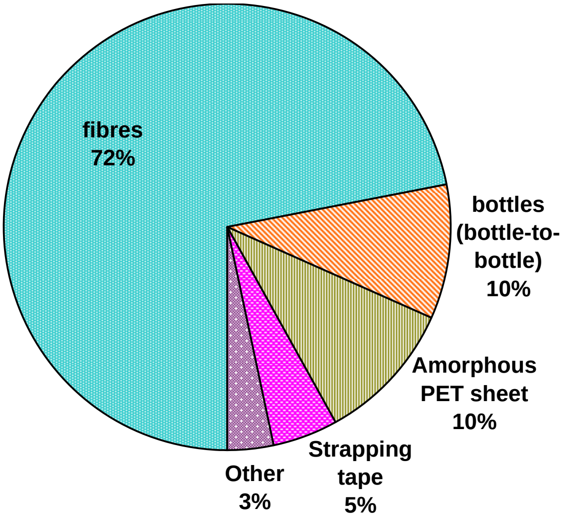

Figure 5.1: Application of recycled PET flakes, worldwide 2007, based on data from [182].

recycled PET fibre compared to virgin PET. Several studies reported the environmental impacts of PET recycling [184, 185, 186]. In these studies, PET recycling was seen as a post-consumer waste management option and was compared with other options such as landfilling and incineration. The goal of this study is not to analyse different waste management options, but to understand the environmental impact of making recycled PET fibres.

The second purpose of this study is to apply different allocation methods for this open-loop-recycling case. In LCA, there has been so far no standardised procedure for open-loop recycling. Several studies have discussed this methodological problem [189, 190, 191, 192]. A common practice is to follow the "cut-off" principle which distinguishes the first life (virgin product) and the second life (recycled product) as separate systems; the post-consumer waste from the first life does not bear any environmental burden when it is used as the feedstock in the second life. The cut-off rule has been widely applied for recycled or recovered products. For example in the Ecoinvent database, heat recovered from waste incineration is considered free of environmental impact [193]. Another example is the EU Directive 2009/28/EC, in which crude glycerol is treated as waste and is considered to be free of greenhouse gas emissions [194]. The cut-off method is considered simple and easy to apply, because no data of the first life is needed.

In this study, we started the analysis with the "cut-off" approach. Two alternative methods were introduced in order to further develop the methodology for open-loop recycling. The first alternative method is the "waste valuation" method, which follows the principle of economic allocation. The second alternative method is the "system expansion" method, in which the entire system (cradle to grave) is analysed.

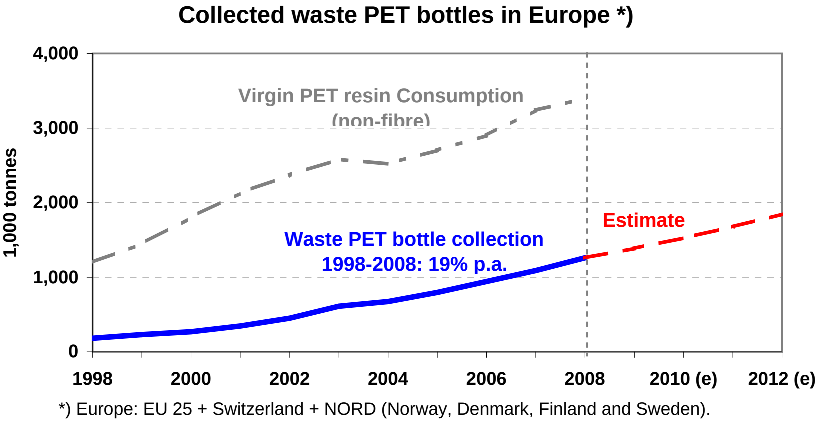

Figure 5.2: Collected post-consumer waste PET bottles in Europe over the last 10 years based on data from [187] and [41] and the projection for the near future based on 10% p.a. growth rate [8]. Virgin PET consumption data were obtained [4, 188].

Four PET recycling cases are investigated in this study, including mechanical recycling, semi-mechanical recycling, back-to-oligomer recycling and back-to-monomer recycling. For each of the first three types of recycling technologies, the respective process data for the year 2008 were provided by three companies. Due to confidentiality issues, no plant data were available for back-to-monomer recycling. Therefore, the analysis was performed based on publicly available information. Virgin PET fibre produced in Western Europe was taken as the reference system. In addition, the LCA result was compared with commodity fibres, i.e. cotton, viscose and polypropylene (PP) as well as novel bio-based fibres, i.e. man-made cellulose fibres (Viscose and Tencel) and polylactic acid (PLA) fibres.

5.2 Methodology

LCA has been standardised by the ISO 14040 series, namely:

- ISO 14040: 2006 - Principles and framework [146]; and
- ISO 14044: 2006 – Requirements and guidelines [147].

5.2.1 Goal, functional unit and system boundary

Goal and functional unit

The goal of this LCA is to assess the environmental impacts of recycled PET fibre compared with virgin PET fibre. The functional unit is defined as “one metric tonne of fibre”. Fibres are important intermediate products for the textile and nonwoven industry. There are two types of PET fibre, staple fibre and POY (partially oriented yarn, which is generally called filament fibre). It should be noted that staple fibre and POY are different products in terms of material properties and consequently, they are used for different end-use applications (see Table 5.1). The goal of this LCA is not to compare staple fibre with POY, but to understand the environmental impacts of recycled PET fibres compared with the two main types of virgin PET fibres.

The chosen functional unit implies the assumption that recycled PET fibre and virgin PET fibre are functionally equivalent. One may argue that recycled fibre might not reach the same quality as virgin fibre. However, it depends on the recycling technology and the scope of such a comparison. For chemical recycling back to monomers, the quality of the recycled polymer is identical with virgin polymer. PET fibre produced by chemical recycling back to oligomers has very similar properties as virgin fibres except for dyeability, which is generally inferior to that of virgin fibre (Private communication with Far Eastern New Century Co., Ltd.). For mechanical and semi-mechanical recycling, the quality of recycled fibre strongly depends on the purity of the waste stream. According to one of the recycling companies investigated in this study, recycled staple fibre can reach the same quality as virgin staple fibre if a clean bottle source is used, bottles are properly sorted and the impurities are carefully removed. In addition, because polyester has such a wide range of applications, it is always possible to find suitable applications for recycled fibres, where virgin fibres are also used.

System boundary

The scope of this LCA is cradle to factory gate. For a virgin product, this includes all steps from the extraction and transportation of raw materials and fuels, followed by all conversion steps until the product – i.e. fibre – is delivered at the factory gate. The production of the end product (e.g. a shirt), the use phase and the post-consumer waste management are excluded. A cradle-to-grave analysis, including the waste disposal phase but excluding the use phase, is discussed in Section 5.5.2.

For open-loop recycling, it is typically a problem to define the "cradle" stage of the recycled product. As default case, we choose the conventional "cut-off" approach to define the system boundary.[2] Figure 5.3 illustrates the concept of the "cut-off" approach: the first life and second life are cut into two independent product systems. Based on the cut-off principle, the used bottles from the first life are considered to be waste; waste does not bear any environmental burden from the first life. We follow this rule and define the "cradle" of the second life as the collection and transportation of used PET bottles.

Next to the "cut-off" approach we introduce, apply and discuss two alternative methods, in Section 5.5, namely the "waste valuation" method and the "system expansion" method. The "waste valuation" method has the same scope as the "cut-off" approach, i.e. cradle to factory gate. The "system expansion" method covers the entire system from cradle to grave.

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[2] It is considered "conventional" because this method has been applied for many recycled products, such as secondary steel, aluminium and glass [193, 146].

Table 5.1: Product systems in this study, comparing type of fibre, property and application.

	Recycling case 1	Recycling case 2	Recycling case 3	Recycling case 4	Reference
Technology	Mechanical	Semi-Mechanical	Chemical, back-to-BHET recycling	Chemical, back-to-DMT recycling	single-use virgin PET
Current technology level	Large scale production	Large scale production	Small scale production	Small scale or pilot scale production	Large scale production
Inventory data	Wellman-International Ltd. (Wellman)	Long John Group (LJG)	Far Eastern New Century Co. (FENC)	Literature data[a]	Literature data[b]
Geographic scope	Western Europe	Taiwan	Taiwan	Western Europe	Western Europe
Type of fibre studied	Staple	Filament (POY)	Filament (POY)	Filament (POY)	Staple and filament (POY)
Property	High to medium denier Staple No microfibre	High to medium denier Staple and filament Limited microfibre	Medium to low denier Mainly filament Microfibre	Medium to low denier Mainly filament Microfibre	Full denier range Staple and filament Microfibre
Application	Non-woven Technical end use	Footwear Technical textile Bags	Apparel Soft hand feel Moisture management Limited colours available	Performance apparel Soft hand feel Moisture management All colours available	Non-woven Apparel Performance apparel All colours available

Abbreviations: BHET: bis-hydroxylethylene terephthalate; DMT: dimethyl terephthalate; POY: partially oriented yarn.

[a] See data sources in Section 5.2.2 and 5.3.3.

[b] See data sources in Section 5.2.2.

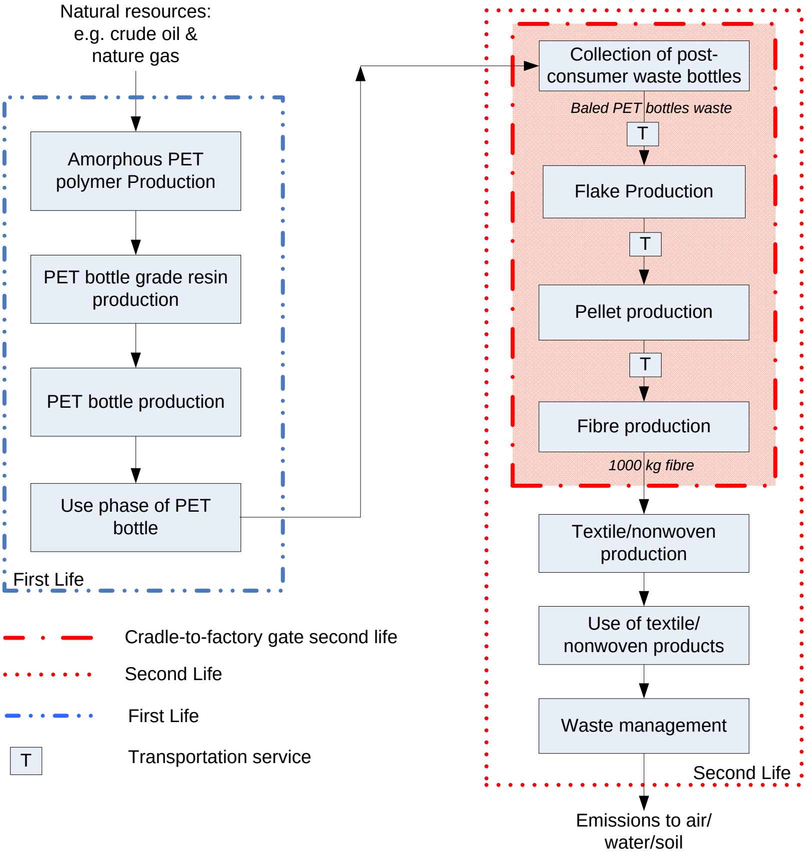

Figure 5.3: Cradle-to-factory gate system boundary of recycling PET fibres from waste PET bottles, splitting the first life and the second life based on the “cut-off” approach.

5.2.2 General data and assumptions

The geographic boundary covers Western Europe and Taiwan depending on the product system (Table 5.1). All three companies recycle PET bottles on a large scale. Wellman International Ltd. (in short “Wellman”) recycles about 10% of the collected bottles in Europe every year. Both Long John Group (in short “LJG”) and Far Eastern New Century Co., Ltd. (in short “FENC”) are among the largest recycling companies in Taiwan. Thus, the result of this analysis is expected to be representative for mechanical recycling of PET in Europe and in Taiwan. The virgin PET fibre produced in Western Europe is chosen as the reference system. The LCI data of virgin PET polymer production is based on average technology in Western Europe [195]. The transportation of raw materials, intermediate products and fuel is included in the system boundaries. A detailed description of the recycling process can be found in Section 5.3.

For all three recycling companies, the inventory data was provided for the year of 2008. For both virgin polymer production and the inventory data from the Ecoinvent database, the production represents the technologies in the 2000s [193, 195].

The data for heat and power generation, chemical production, transportation, waste management and virgin polymer production were obtained from various sources including LCA databases, scientific publications, governmental statistics and personal communication. Table 5.2 provides a summary of the general data and assumptions. For chemical recycling back to dimethyl terephthalate (DMT), our attempt to obtain data was not successful due to confidentiality issues. The analysis was carried out based on the information available from the public domain. The detailed assumptions and data sources used for this case are described in Section 5.3.4.

Since the chosen allocation methods may strongly influence the outcome of LCA studies we summarize here which methods are applied and which system they refer to:

1. As mentioned in Section 5.2.1, the "cut-off" approach is applied as the default method for open-loop recycling. An alternative allocation based on economic values ("waste valuation" method) and an approach which follows the "system expansion" principle will be discussed in Section 5.5.

2. By-products from the flake production, mainly consisting of coloured bottles and polyethylene (PE) and accounting for about 6-11% of the total mass of the input, are allocated based on economic values. The average selling prices of both by-products and main products (flakes) were provided by the companies for the year 2008, resulting in the economic value of the by-products of typically 5-6% of the total value of the products.

3. The system expansion method is applied for the process waste and for post-consumer solid waste which is assumed to be disposed of in a municipal solid waste incineration (MSWI) facility with energy recovery. Credits were assigned to the recovered electricity and/or heat since the production of the grid electricity and/or heat can be avoided. In Western Europe, the energy recovery rate in primary energy terms is approximately 60% in primary energy term (private communication with Dr. Reimann of CEWEP) [155, 162]. In Taiwan, the energy recovery rate of an average waste-to-energy facility is approximately 43% in primary energy term [203, 204].

5.2.3 Environmental impact assessment

In Life Cycle Impact Assessment (LCIA), the life cycle inventory data, which represent all emissions released by the product system to the environment and all raw material requirements, are converted into environmental impact categories. The results are generally referred to as LCA mid-point results. In this study, the environmental indicators are: NREU (non-renewable energy use), GWP (global warming potential) (IPCC, 2007) and the indicators from the CML 2 baseline 2001[3] impact assessment method

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[3] CML: Centrum voor Milieuwetenschappen Leiden (Institute of Environmental Sciences), Leiden University, the Netherlands.

Table 5.2: Data sources of this study

Data	Sources	Note
PET bottle-to-fibre recycling	Collected from three recycled PET fibre producers (Wellman, LJG and FENC).	Site-specific, for year 2008
Grid electricity	Ecoinvent V2.0 [196]; OECD and Non-OECD country energy balances 2005/2006 [17, 155].	Country-specific. European electricity mix: 65% from the UCTE grid, 13% from the NORDEL grid, 9% from the CENTREL grid, 12% from the UK grid and 1% from the Irish grid. Taiwan electricity fuel mix: 58% coal, 20% nuclear, 12% natural gas, 8% oil and 3% renewables.
Production and combustion of natural gas, LPG, fuel oil and diesel	Ecoinvent V2.0 [197, 198]; EIA statistics [199]; US EPA report [200].	Country-specific energy profiles, except for LPG for which global data is used based on Ecoinvent database.
Production of chemicals	Ecoinvent V2.0 [21].	Western Europe mid-2000 technology level
Transportation distances and means for raw materials, chemicals and intermediate products	Collected from three recycled PET fibre producers (Wellman, LJG and FENC).	
Road and water transportation	Ecoinvent V2.0 [201].	32 t lorry for road transportation. Water transportation refers to transoceanic shipping.
Rail transportation	Ecoinvent V2.0 [196, 201]	Only occurs in France, modified by French grid electricity data in Ecocinvent.
Waste management - sanitary landfilling	Ecoinvent V2.0 [202]	Switzerland mid-2000 technology level.
Waste management - incineration with energy recovery	Ecoinvent V2.0 [202]; CEWEP report [162]; EPA reports [203, 204].	Country-specific.
Virgin polymer production	Plastics Europe Eco-Profiles [112, 205].	Western Europe polymer production.
Energy use for staple and filament fibre spinning process (for melt-spinning virgin PET fibre)	Assumption based on [164]: 0.64 kWh electricity and 5 MJ heat (from fossil fuel) per kg fibre.	This data was cross-checked by polyester industry experts.

Abbreviation: UCTE stands for Union for the Co-ordination of Transmission of Electricity; countries included in UCTE are Austria, Bosnia and Herzegovina, Belgium, Switzerland, Germany, Spain, France, Greece, Croatia, Italy, Luxemburg, Macedonia, Netherlands, Portugal, Slovenia and Serbia and Montenegro. NORDEL stands for Nordic countries power association, including Denmark, Norway, Finland and Sweden. CENTREL stands for Central European power association, including Czech Republic, Hungary, Poland and Slovakia. LPG stands for Liquefied Petroleum Gas.

[206, 207], namely abiotic depletion, acidification, eutrophication, human toxicity, fresh water aquatic ecotoxicity, terrestrial ecotoxicity and photochemical oxidant formation. For chemical recycling via the DMT route, due to the limited data availability, only NREU and GWP were assessed.

In addition, normalisation was performed using CML normalisation factors for World 2000 (see Table 5.3). This step determines the relative contribution of the product system to the impact categories at a global level. The normalised results do not imply weighting of the impact categories, they merely give an indication to which extent the product system contributes to the total environmental loads of a region for a given year.

Table 5.3: CML normalisation factors, global impact per year, World 2000 [172].

Environmental impact categories	Normalisation factors
Global warming (kg CO_2 eq./yr)	4.18×10^{13}
Abiotic depletion (kg Sb eq./yr)	1.83×10^{11}
Ozone layer depletion (kg CFC-11 eq./yr)	2.30×10^{8}
Human toxicity (kg 1,4 DB eq./yr)	3.82×10^{13}
Fresh water ecotoxicity (kg 1,4 DB eq./yr)	3.48×10^{12}
Terrestrial ecotoxicity (kg 1,4 DB eq./yr)	1.09×10^{11}
Photochemical oxidation (kg C_2H_4 eq./yr)	5.44×10^{10}
Acidification (kg SO_2 eq./yr)	2.39×10^{11}
Eutrophication (kg PO_4^{3-} eq./yr)	1.58×10^{11}

5.3 Recycling PET bottles into fibre

5.3.1 Collection of used PET bottles

In Western Europe, used PET bottles are collected either under the GreenDot scheme (`www.gruener-punkt.de`), or under other schemes, such as a mandatory deposit system [208]. In Taiwan, used PET bottles are collected either together with other household waste before they are sorted out manually (`www.epa.gov.tw`), or via the deposit-refund system [209]. In all cases, used PET bottles are collected on a local scale, e.g. they are from consumers and brought to a waste separation centre where bottles are sorted out, baled and compacted. The energy consumption related to sorting, baling and compacting is very small compared to the energy requirements of the recycling process [184, 185]. In this study we assume that the energy requirements associated with sorting, baling and compacting are negligible.

The major environmental burdens from the collection step are related to the fuel consumption and air emissions from the transportation of baled bottles (i.e. from waste separation centres to flake production facilities). In the case of Wellman's recycling operation in Western Europe, the baled bottles are transported by truck over a distance of about 300-400 km. In the case of the two recycling companies in Taiwan, the baled bottles are transported by truck for about 100-350 km.

5.3.2 Production of recycled PET flakes

Figure 5.4 shows the flowsheet of the production of recycled PET flakes. After baled bottles are opened, loose bottles are sorted by colour and material type. Transparent (uncoloured) bottles have a higher economic value than blue and green ones. The unwanted colour fractions and unwanted materials (e.g. paper and metal) are either sold as by-products, or disposed of in local municipal solid waste (MSW) management

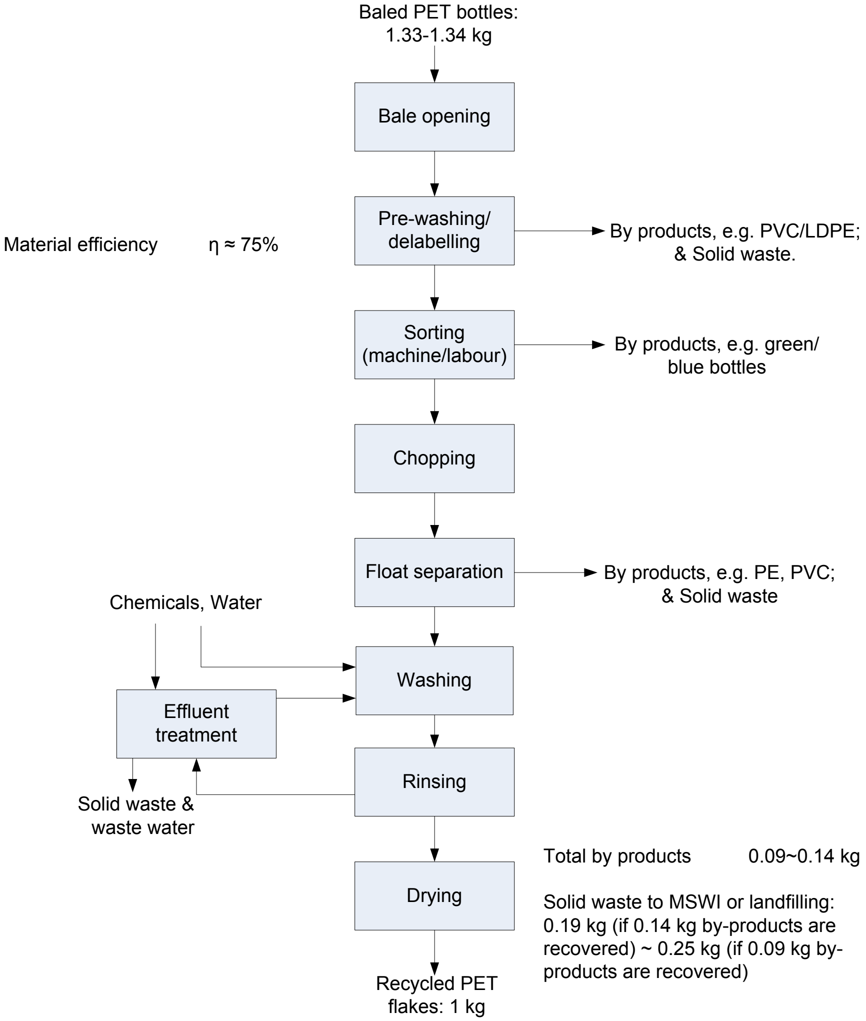

Figure 5.4: Producing recycled PET flakes from baled PET bottles.

facilities or landfilled, depending on the available local infrastructure. MSW can be incinerated with or without energy recovery. Next, the bottles are sorted. The typical plant in Europe uses automated sorting (through colour recognition technology), while the Asian producers use manual sorting. Some producers wash the bottles with hot water to remove the labels before the sorting process. The plastics labels are either sold as by-products (mainly consisting of LDPE and/or PVC), or sent to local MSW management. The bottles are then chopped into flakes, followed by a float separation step to separate PET from other plastics (e.g. HDPE caps) based on density differences. PE obtained from this step is sold as a by-product. The PET flakes are then washed in a cleaning solution, rinsed and dried. In some production lines, a second chopping step (also called "fine crushing") is required to ensure that the PET flakes meet the quality requirements. Finally, the dried PET flakes are ready to be transported to a pellet plant or a fibre plant.

5.3.3 Mechanical and semi-mechanical recycling

Mechanical recycling is the physical conversion of flakes into fibre or other products by melt-extrusion. Currently, there are two ways to produce recycled fibre from mechanical recycling:

1) directly extrude flakes into fibre; or more commonly,
2) first convert flakes into pellets or chips (pelletizing) and then melt-extrude pellets or chips into fibre.

Flake to fibre (mechanical, Wellman International Ltd.)

Wellman produces recycled PET staple fibre directly from melt extrusion of recycled PET flakes (see the left graph of Figure 5.5). After flakes are off-loaded, they are dried in a column dryer before they are melt-extruded. The extruded polymer is filtered before it passes through the spinneret where filament spinning takes place. After the filaments pass a denier setter, they enter the finishing process where the spun filaments are drawn, dried, cut into staple fibre and baled. Approximately 1% of the flakes end up as solid waste which is disposed of in a landfill.

Flake to pellet, then to fibre (semi-mechanical, Long John Group)

In many other mechanical recycling plants, flakes are first extruded into pellets and then converted into fibre and other products. LJG (Long John Group) produces recycled PET fibre through the flake-pellet-fibre route (see the right graph of Figure 5.5. PET flakes are dried prior to the melt extrusion step. The extruded polymer is further purified through a filtration step. After a cooling process, the polymer is pelletised and dried. The PET pellets are then transported to the fibre spinning plant where they are melt-spun into filament fibre (POY). In LJG's process, a small amount of ethylene glycol (EG) is added to meet the final quality requirements. We therefore classify LJG's process as a semi-mechanical recycling process. The solid waste from the recycling process is disposed of in a MSWI with electricity recovery (recovery rate = 43%, see Section 5.2.2).

5.3.4 Chemical recycling

In chemical recycling, PET polymer is broken down into monomers or oligomers via various depolymerisation technologies. Chemical recycling is more expensive than mechanical recycling. It usually requires a large scale in order to become economically feasible [210]. The important advantage of chemical recycling is that the quality of

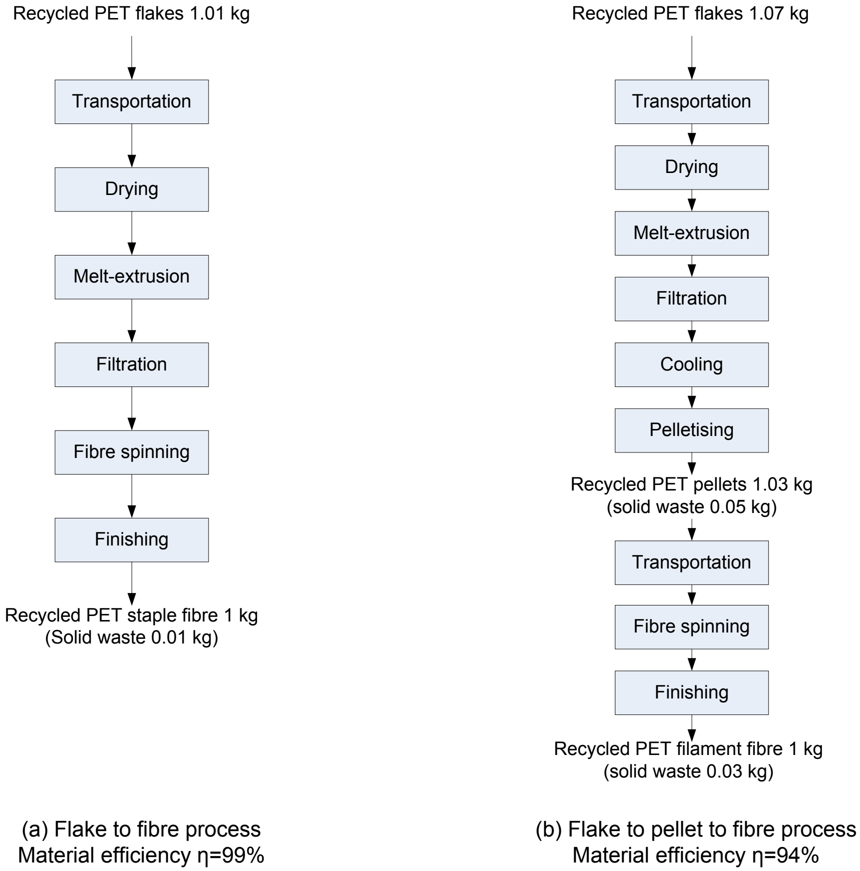

Figure 5.5: Producing recycled PET fibre from PET flakes via mechanical (left) and semi-mechanical (right) recycling.

virgin PET can be achieved. Current commercially available chemical recycling technologies include glycolysis, methanolysis and alkaline hydrolysis [210]. In our study, recycled PET produced via the glycolysis route was analysed based on data received from Far Eastern New Century (FENC). The methanolysis route was analysed based on publicly available data.

Glycolysis to BHET (chemical recycling, back to oligomer, Far Eastern New Century Co., Ltd. (FENC))

Figure 5.6 shows the back-to-oligomer recycling by FENC. The glycolysis of PET yields the oligomer bis-hydroxyl ethylene terephthalate (BHET). The process is usually conducted in a temperature range between 180 and 250 °C with excess EG and in the presence of catalysts [211]. After the glycolysis process, the oligomer passes through a fine filtration step before it is repolymerised into PET. The recycled polymer is then spun into fibre. The process solid waste is disposed of in a MSWI facility with electricity recovery (recovery rate = 43%, see Section 5.2.2).

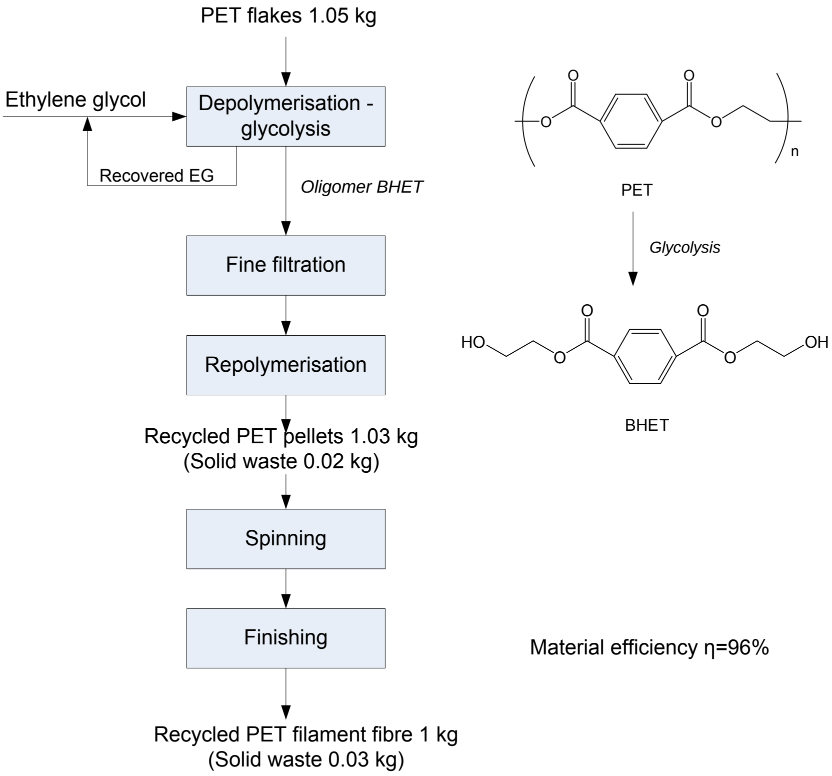

Figure 5.6: Chemical recycling PET via the glycolysis process.

Methanolysis to DMT (chemical recycling, back to monomer)

In methanolysis, PET is depolymerised with methanol to DMT and EG in the presence of catalysts under a pressure of 2-4 MPa and a temperature of 180-280 °C [211]. The reaction mix is cooled and DMT is recovered from the mix via precipitation, centrifugation and crystallization [211]. Figure 5.7 shows the flowsheet for chemical recycling of PET via the methanolysis route. The recycled polymer is then converted into fibre via spinning and finishing processes.

A recent patent by Teijin [212] illustrates that PET is depolymerised with EG and sodium carbonate to yield BHET; the BHET is then further broken down into DMT with methanol [213]. This process is considered more economically attractive than the direct methanolysis of PET into DMT [214].

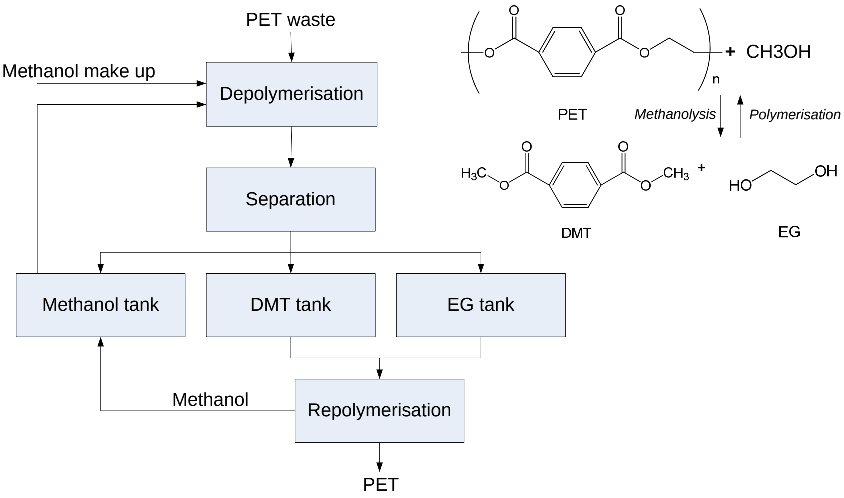

Figure 5.7: Chemical recycling PET via the methanolysis process [211].

The methanolysis route is commercially operated but no process data could be obtained. In this study, we use publicly available data to estimate the NREU and GWP (the available data did not allow to also include the environmental impact categories according to the CML method).

Our estimate is primarily based on the LCA published by Patagonia [215] for recycled DMT. According to Patagonia's LCA results, the cradle-to-factory gate non-renewable energy requirements (NREU) and GHG emissions of 1 tonne of recycled DMT are 11.96 GJ and 0.98 t CO_2 eq., respectively. The "cradle" of Patagonia's LCA follows the "cut-off" rule since the cradle was defined as collection of PET waste.

Based on the stoichiometric equation, depolymerising 1000 kg of PET requires 333 kg methanol and yields 1010 kg of DMT (or 76% by weight) and 323 kg EG (or 24% by weight). Patagonia's LCA results were allocated based on the weight of the products. Using this information we estimate that the NREU and GWP values for the total process yielding 1 tonne of DMT and 0.32 tonnes of EG are 15.78 GJ/t DMT and 1.29 t CO_2 eq./t DMT, respectively (here, the energy use and the emissions related to EG production have been assigned to DMT).

The material efficiencies and the monomer recovery rates are not published by Patagonia. We assume three cases, namely, a low case, a high case and an average case. In the low case, the PET loss is assumed zero, which is the theoretical optimum (stoichiometric conversion). In the high case, we assume 10% PET loss based on Marathe et al. [216] who reported that the yield of methanolysis does not exceed 90%. The loss of 10% refers to rather clean and sorted PET waste, while the losses may be substantially larger for other products, e.g. finished textiles (due to the use of textile auxiliaries, dyes etc.). As average case, we assume that the loss is somewhere in-between, i.e. 5%. Furthermore, the net methanol input (the "make-up" in Figure 5.7 is assumed to be zero for the low case, 10% for the high case and 5% for the average.

Since Patagonia's study only reports the production of DMT, it is not known whether and how much EG is recovered and reused in the repolymerisation step. In the low case, the recovery and reuse of EG is assumed to be 100% (stoichiometric conversion and complete recovery). In the high case, we assume that EG is not recovered at all and that the EG required for repolymerisation, which is estimated at 323 kg EG/t PET based on the stoichiometric equation, is purchased. In industrial practice the unrecovered EG may be incinerated together with other compounds, with or without energy recovery. In the low case, we assume that there is no energy recovery. In the average case, 50% of EG is assumed to be recovered and the rest 50% is purchased externally. Finally, in the high case, no energy credits are assigned to the lost amounts of EG. The environmental impact of the purchased EG is obtained from the Ecoinvent database for "Ethylene glycol, at plant" [21]: the cradle-to-factory gate NREU and GWP100a of EG are 52 GJ/t and 0.82 t CO_2 eq./t.

The repolymerisation step is technically identical with the polymerisation process leading to virgin PET. According to an earlier publication of AMPE [217], the NREU for synthesizing 1 tonne of PET via the PTA/EG route is 10.16 GJ and the GWP 100a is 0.61 t CO_2 eq. We assume that the repolymerisation of recycled PET via the DMT/EG route has the same energy requirements.

Next, the recycled amorphous PET polymer is sent to the fibre production plant. It is assumed that the energy requirement of fibre spinning is the same as for virgin fibre production (see Table 5.2). Typically, monomer recycling is combined with filament production because the value of the high purity of the recycled compounds is fully exploited; this will be taken into account in the interpretation of this study. Table 5.4 shows the summary of the data and assumptions for chemical recycling via the methanolysis route.

Table 5.4: Data and assumptions for chemical recycling through methanolysis.

	Low case	High case	Average
NREU of depolymerisation (GJ/t DMT)[a]		15.78	
GWP100a of depolymerisation (t CO_2 eq./t DMT)[a]		1.29	
Overall PET loss (by weight)[b]	0	10%	5%
Overall MeOH make-up (by weight)[c]	0	10%	5%
EG recovery (by weight)[c]	0	100%	50%
External EG required (by weight)[c]	100%	0%	50%
Repolymerisation[d]			
Fuel (GJ/t)		1.63	
Electricity (GJ/t)		0.70	
Steam (t/t)		0.94	

[a] Data source: [215]. The allocation is based on mass.

[b] Assumed based on Marathe et al. [216].

[c] Own estimate or assumption.

[d] Data source: [217].

5.4 LCA results based on the “cut-off” approach

Table 5.5 shows the cradle-to-factory gate LCA results for one tonne of recycled PET fibre based on the “cut-off” approach. Recycled fibres offer 45-85% of NREU savings compared to the virgin fibre. Note that due to the cut-off approach, the embedded energy (calorific value) of the recycled PET is set to zero, whereas for virgin PET fibre, the embedded energy accounts for about 40% of its total NREU.

As Table 5.5 shows, recycled PET fibres offer significant GWP savings compared to virgin PET fibres. The GWP of recycled PET fibres is 76% (mechanical recycling), 54% (semi-mechanical recycling), 36% (back-to-oligomer recycling) and 24% (back-to-monomer recycling) lower than that of virgin PET.

Table 5.5: LCA result for 1 tonne of recycled PET fibre, based on the “cut-off” approach, cradle to factory gate for second life.

Recycling route	Mechanical	Semi-mechanical	Chemical BHET	Chemical DMT	V-PET (W.Europe)
Company	Wellman	LJG	FENC	n/a	n/a
Fibre type	Staple	POY	POY	POY	Staple or POY
NREU (GJ)	13	23	39	51 (40-62)[a]	95
GWP 100a (t CO_2 eq.)	0.96	1.88	2.59	3.08 (2.71-3.44)[a]	4.06
Abiotic depletion (kg Sb eq.)	6	11	18	n/a	45
Acidification (kg SO_2 eq.)	3	9	14		21
Eutrophication (kg PO_4^{3-} eq.)	0.8	0.7	2.3		1.2
Human toxicity (kg 1,4-DB eq.)	362	415	745		4393
Fresh water aqua. ecotox. (kg 1,4-DB eq.)	296	250	303		58
Terrestrial ecotoxicity (kg 1,4-DB eq.)	7	7	17		12
Photochemical oxidant formation (kg C_2H_4 eq.)	0.2	0.3	0.6		1.0

[a] For chemical recycling via the DMT route only NREU and GWP were assessed. The range in bracket represents the low case and the high case (see Table 5.4).

Compared to virgin fibres, mechanically and semi-mechanically recycled fibres offer lower impacts for all seven CML environmental categories except for freshwater aquatic ecotoxicity. Back-to-oligomer recycling offers a lower impact in six out of nine categories. The exceptions are eutrophication, freshwater ecotoxicity and terrestrial ecotoxicity. For all three recycling companies investigated, the impact of freshwater aquatic ecotoxicity originates from the incineration of solid waste from flake production. More than 90% of the freshwater ecotoxicity impact can be traced back to the water emission of a small amount of vanadium. Vanadium oxides are commonly used in municipal waste incineration plants as catalysts to treat NO_x emissions [202]. About 50% of the eutrophication impact of FENC’s fibre originates from the production of chemicals (e.g. EG) used for the chemical recycling process. Atmospheric emissions of vanadium (from fuel oil combustion) are responsible for more than 60% of its terrestrial ecotoxicity impact.

Mechanical recycling (Wellman) causes the lowest impact in eight out of nine environmental categories, compared to the other three product systems shown in Table 5.5.

Process energy use is responsible for the major part of the environmental impacts, represented by NREU, GWP, abiotic depletion, acidification, terrestrial ecotoxicity and photochemical oxidant formation. The process waste management in flake production, including both emissions from waste water treatment (e.g. COD) and from solid waste management (e.g. MSWI), is the most important factor for eutrophication, human toxicity and freshwater aquatic ecotoxicity. The production of chemicals and the transportation of raw materials, intermediate products and solid waste treatment contributes very little (<10%) to the overall environmental impact.

The process energy use of the semi-mechanical recycling (LJG) is the most important factor for eight out of nine environmental indicators. The exception is freshwater aquatic ecotoxicity. The process energy use for fibre production from flakes is the most important contributor (40-70%) to NREU, GWP, abiotic depletion and photochemical oxidant formation. The process energy use of flake production is the most important contributor (40-80%) to acidification, eutrophication, human toxicity and terrestrial ecotoxicity. Fresh aquatic ecotoxicity is mainly caused by the solid waste which is sent to MSWI. Compared to flake production and fibre production, pellet production causes smaller environmental impacts. Transportation and the production of chemicals (e.g. small amount of EG) have minor impact (<5%).

For back-to-oligomer recycling by FENC, the glycolysis process contributes most to the overall environmental profile. The chemicals and energy use (electricity and fuels) are responsible for the major part of NREU, GWP, abiotic depletion, acidification, eutrophication, human toxicity, terrestrial ecotoxicity and photochemical oxidant formation. Like Wellman and LJG, FENC's freshwater aquatic ecotoxicity originates from solid waste management.

For back-to-monomer recycling, only NREU and GWP were analysed because of the lack of information (see Section 5.2.1). The depolymerisation process contributes most to the overall impact, with shares of 30-40% of the total NREU and 45-50% of the total GWP.

Figure 5.8 shows the LCA results normalised to World 2000. Compared to virgin production, recycled PET fibres cause substantially lower environmental impacts. Particularly, the impact reduction of abiotic depletion, acidification and human toxicity is substantial. Furthermore, for all PET fibres studied (both recycled and virgin), eutrophication, terrestrial ecotoxicity and photochemical oxidant formation are negligible in a global context. Recycled fibres cause a relatively high environmental impact on freshwater aquatic ecotoxicity compared to virgin PET because following the cut-off approach, all impacts from post-consumer waste management including fresh water ecotoxicity are exclusively assigned to the recycled product. Thus, the allocation method and the chosen system boundary have strong influence on the results of this open-loop recycling case.

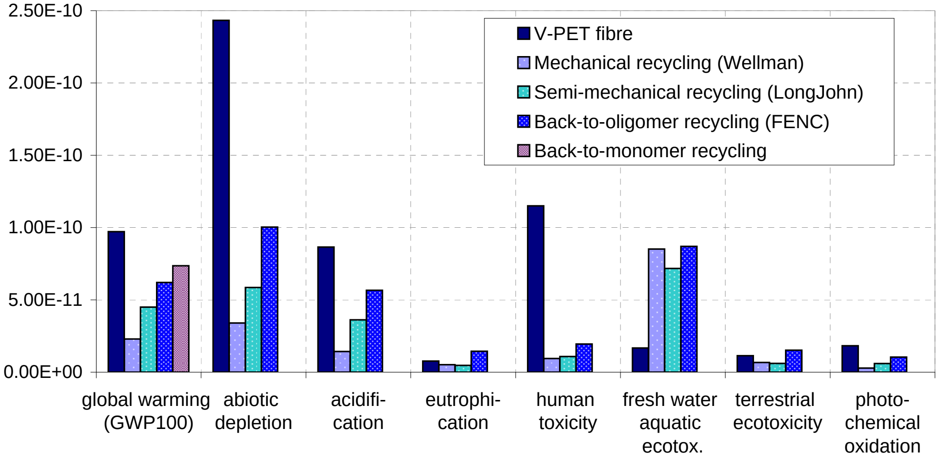

Figure 5.8: Normalised results for 1 tonne of PET fibre, "cut-off" approach, cradle to factory gate for second life, normalised to World 2000

5.5 Alternative approaches for open-loop recycling

5.5.1 "Waste valuation" method

Until now, we have only discussed the LCA results based on the "cut-off" approach. The environmental burden of the first life was not considered in the system boundary (see Figure 5.3). However, one can argue that this method is oversimplified, because in reality bottle waste is traded and it does have a commercial value. In other words, waste is a valuable resource. Thus the environmental impact of the production of virgin polymer should be shared between the first life and the second life (see the illustration in Figure 5.9).

In this study, allocation based on mass is not a feasible choice, because bottles and fibres are different products. We therefore apply the second approach, i.e. economic allocation. In this article we name this alternative method "waste valuation" method. This represents a variant of the "cut-off" approach which makes use of economic values (prices):

$$E_{WV} = E_{cut-off} + AF \times E_{\text{vPET resin}} \tag{5.1}$$

where E_{WV} stands for the environmental impact of recycled PET fibre; $E_{cut-off}$ is the environmental impact of recycled PET fibre based on the "cut-off" approach; $E_{\text{vPET resin}}$ is the environmental impact of virgin PET bottle grade resin; and AF is the allocation factor. $AF \times E_{\text{vPET resin}}$ is the environmental burden which is shifted from the first to the second life.

The determination of the allocation factor is the key step for the "waste valuation" method. We define the allocation factor (AF) as the ratio of the market value of baled

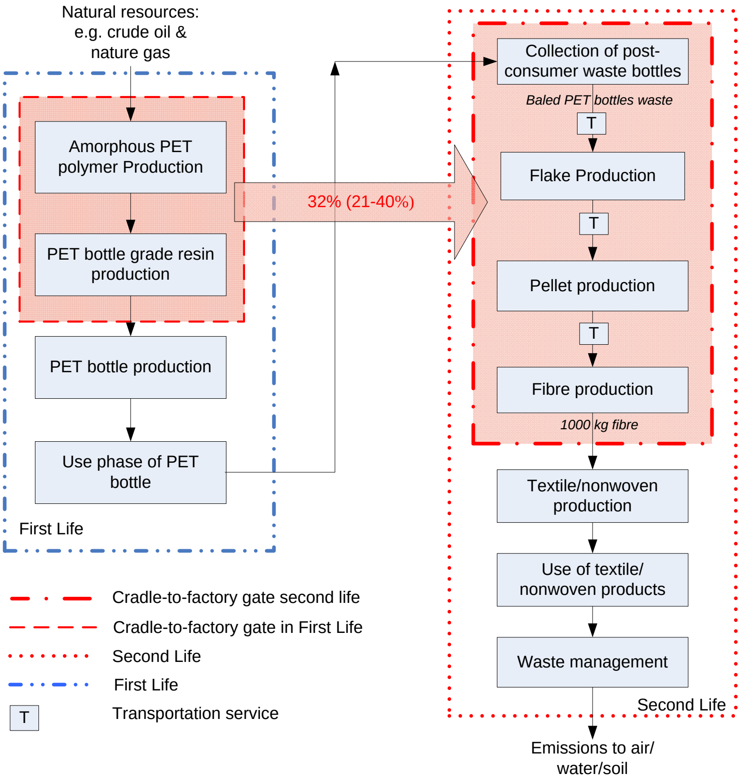

Figure 5.9: System boundary based on the "waste valuation" method.

bottle waste to the market value of virgin PET bottle grade resin:

$$AF = \frac{\text{Price of Baled bottle waste}}{\text{Price of virgin PET bottle grade resin}} \tag{5.2}$$

The price of baled bottle waste was collected from three companies as average value for the year 2008. The price of virgin PET bottle grade resin was obtained from the monthly prices of North America plastics resins published by Plastics Online Technology [218]. Due to the regional differences and the strong fluctuation of crude oil prices in 2008, the AFs differ by companies, although not substantially. In general, the AF is in the range of 21-40%; the average AF is approximately 32%. For the "waste valuation" method, we therefore assumed that 32% (21-40%) of the environmental burden of virgin PET bottle grade resin is shifted to the recycled PET fibres. The LCA results are shown in Table 5.6.

Compared to virgin fibre, mechanically and semi-mechanically recycled PET fibres still offer environmental benefits in all categories except for freshwater aquatic ecotoxicity. Back-to-oligomer recycling offers an impact reduction in six out of nine categories. By analogy with the "cut-off" approach, recycled fibre produced from chemical recy-

Table 5.6: LCA result for 1 tonne of recycled PET fibre, based on the "waste valuation" approach, cradle to factory gate for second life.

Recycling route	Mechanical	Semi-mechanical	Chemical BHET	V-PET (W.Europe)
Company	Wellman	LJG	FENC	
Fibre type	Staple	POY	POY	Staple/POY
NREU (GJ)	40	49	66	95
GWP 100a (t CO_2 eq.)	2.03	2.95	3.66	4.06
Abiotic depletion (kg Sb eq.)	19	23	31	45
Acidification (kg SO_2 eq.)	8	14	19	21
Eutrophication (kg PO_4^{3-} eq.)	1.1	1.0	2.6	1.2
Human toxicity (kg 1,4-DB eq.)	1640	1700	2030	4390
Fresh water aqua. ecotox. (kg 1,4-DB eq.)	300	250	305	58
Terrestrial ecotoxicity (kg 1,4-DB eq.)	8	7	17	12
Photochemical oxidant formation (kg C_2H_4 eq.)	0.4	0.6	0.8	1.0

cling back to BHET has a relatively high impact on eutrophication, freshwater aquatic ecotoxicity and terrestrial ecotoxicity compared to virgin fibre. Due to lack of data, it is not possible to analyse the back-to-monomer recycling based on the "waste valuation" method.

Figure 5.10 shows that the shifted environmental impact has a strong influence on NREU and GWP. The shifted NREU accounts for 40-65% of the total NREU and the shifted GWP is 30-50% of the total GWP. Figure 5.11 shows the increase of the environmental impact (which is equal to the shifted burden from the virgin bottle grade) for the CML indicators. The shifted burden has the strongest influence on human toxicity which increases by 170-350%, because the production of PET has relatively high impact on human toxicity.[4] For abiotic depletion, acidification, eutrophication, photochemical oxidant formation, together with NREU and GWP, the increase ranges from 30% to 200%. For freshwater aquatic ecotoxicity and terrestrial ecotoxicity, the impact from the shifted environmental burden is negligible (≤ 5%).

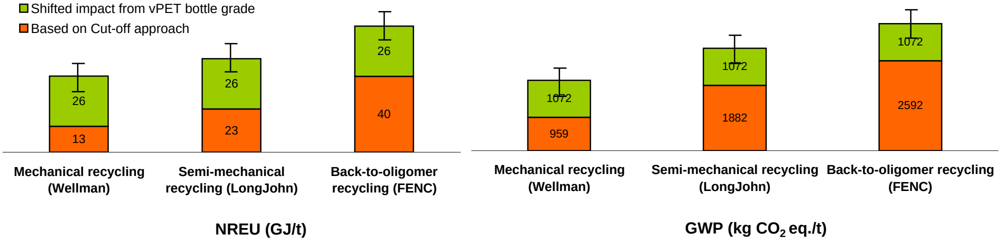

Figure 5.10: Breakdown of NREU and GWP for 1 tonne recycled staple fibre, based on the "waste valuation" method, cradle to factory gate for second life.

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[4] The relatively high human toxicity of virgin PET fibre can be also seen in Table 5.5 or Figure 5.8. More than 90% of the impact is caused by the air emission of PAH (polycyclic aromatic hydrocarbon) in virgin PET resin production [112].

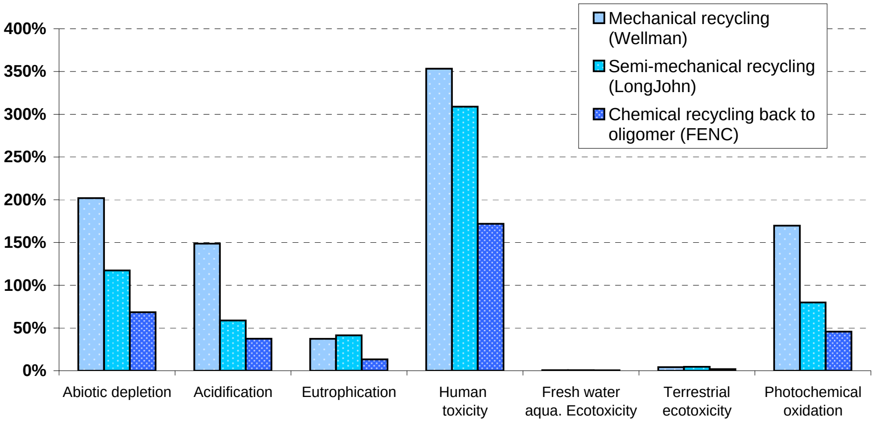

Figure 5.11: Change of environmental impact from the “cut-off” method to the “waste valuation” method, cradle to factory gate for second life, for one tonne of recycled staple fibre, CML 2001 baseline method.

5.5.2 “System expansion” method (cradle to grave)

Open-loop recycling faces two methodological problems. The first problem is how to allocate the environmental impact of the production of the original product throughout several life cycles. In this study, we have so far discussed two methods: the “cut-off” method and the “waste valuation” method. However, both methods are not entirely satisfactory. The “cut-off” method cannot be justified if waste is considered to be a valuable resource. The result from the “waste valuation” method depends on market prices, which are determined by supply and demand, the crude oil price and other economic aspects; they can therefore fluctuate considerably over time.

The second methodological problem is how to allocate the environmental burden of the ultimate “grave” of the product throughout several life cycles. So far this has not been included because the primary system boundary of this study is “cradle to factory gate”. If we extend the product system to the “grave” stage, according to the “cut-off” principle, the environmental impact of end-of-life waste management (e.g. incineration) would be entirely allocated to the last recycled product, the recycled PET fibre. The first life (the virgin PET bottle) does not bear any environmental burden originating from the ultimate waste management.

In short, these two problems are both caused by allocation. The allocation problem for open-loop recycling has not been resolved in current ISO standards. In this study, we propose a method which follows the principle of system expansion.

In a complete cradle-to-grave bottle-to-fibre recycling system, two products are delivered in two lives, i.e. bottles and fibres. If a reference system is to be established for comparison, this system must deliver identical products: bottles and fibres. This concept is illustrated in Figure 5.12. In the reference system, 1000 kg of virgin PET bottle grade, 100 kg PE (for caps and labels)[5] and 1000 kg of virgin fibre are produced and incinerated, i.e. the life cycles are complete.[6] In the bottle-to-fibre (B2F) product system, it is assumed that 1000 kg of virgin PET bottle grade is produced and recycled into approximately 800 kg of PET fibre.[7] The 100 kg of PE is separated and either sold as a by-product or disposed of in a MSWI plant with energy recovery. The 800 kg of recycled PET fibre is used and incinerated; and the life cycles are complete. In such a product system, 1000 kg of bottle grade and 800 kg of fibre are the output function of the product system. In order to make the functional unit comparable with the reference system in terms of mass, an additional 200 kg of fibre is required, which is assumed to be produced from crude oil (see Figure 5.12).

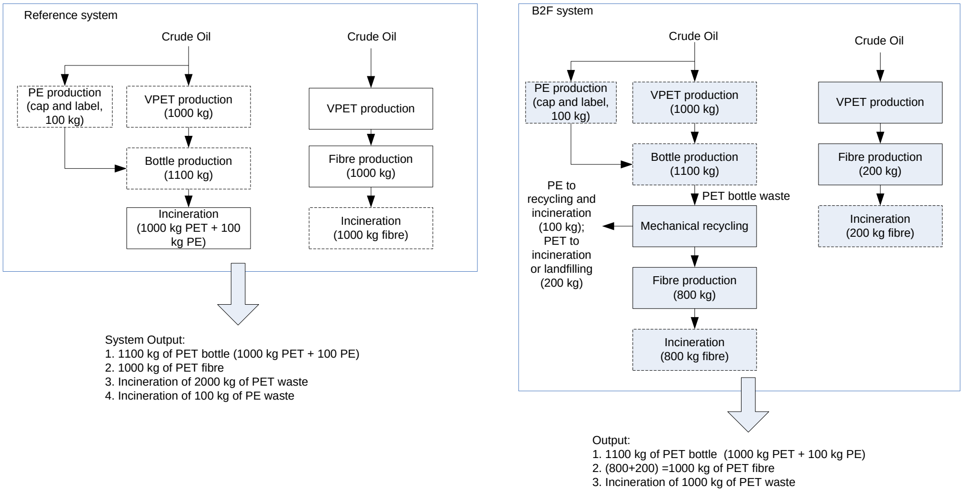

Figure 5.12: "System expansion" method applied for open-loop recycling, functional unit 1 t of PET bottle and 1 t of PET fibre, cradle to grave without the use phase (the mass balances shown in the graph are indicative)

To summarize, both the reference system and the B2F system have the same output in terms of mass, i.e. 1000 kg of PET bottle grade and 1000 kg of PET fibre. The difference is that in the reference system, 2000 kg of PET waste and 100 kg PE waste are incinerated, whereas in the B2F system, only 1000 kg of PET waste is incinerated (PE leaves the system either as a by-product or it is incinerated). In this way, it is possible to study the environmental impact of recycling versus single-use without cutting off life cycles. We name this method the "system expansion" method. An important pre-assumption of this method is that the quality of virgin PET fibre is

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[5] According to Detzel et al. [185] depends on the size of bottles, the weight of caps and labels is approximately 10% (ranging from 7 to 13%) of the weight of PET in a bottle.

[6] The use phase is excluded, because it is the same for both product systems.

[7] This recycling efficiency (80%) is indicative; it does not coincide exactly with the values received from the companies. In general, the recycling efficiency of the PET material flow is about 80-90%, according to the inventory data provided by the companies.

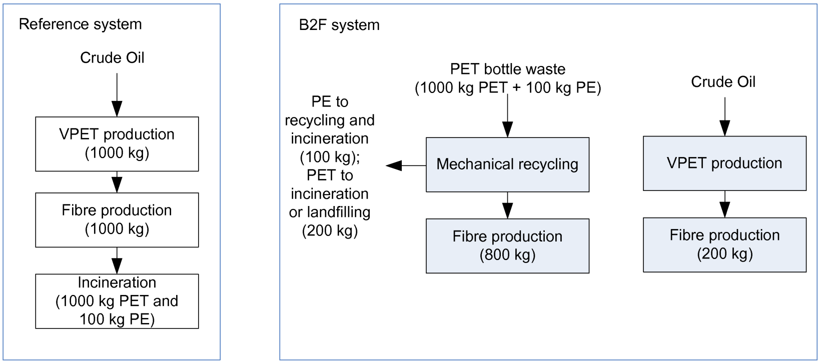


Figure 5.13: "System expansion" method applied for the open-loop recycling, pruned from Figure 5.12, functional unit 1 tonne of PET fibre, cradle to grave without the use phase (the mass balances shown in the graph are indicative)

assumed to be identical to the quality of recycled fibre. In other words, the 1000 kg (800 kg recycled + 200 kg virgin) of fibre from the B2F system is assumed to be fully comparable to the 1000 kg of virgin fibre from the reference system (see also Section 5.2.1 and Table 5.1).

Taking a close look at the two product systems in Figure 5.12, we find that several unit processes are the same in the reference system and in the B2F system, for example, the production of virgin polymers (PET, PE), bottle production and the incineration of PET waste (see dashed boxes in Figure 5.12). Since our primary focus is on the differences between the two product systems, removing the identical unit processes does not change the result of the comparison. Therefore, the dashed boxes in Figure 5.12 can be trimmed out. The pruned product systems are shown in Figure 5.13. In these two product systems, the production of virgin bottle is not presented. In other words, only fibres are studied. We could, in theory, rename the functional unit back to "one tonne of fibre" with the system boundary of cradle to grave (excluding the use phase).

The LCA results based on the "system expansion" method are shown in Table 5.7. Recycled fibres produced by mechanical and semi-mechanical recycling (Wellman and LJG) offer low environmental impacts for all nine indicators, compared to the single-use virgin fibre. Back-to-oligomer recycling (FENC) has a low environmental impact in all categories except for eutrophication and terrestrial ecotoxicity.

From cradle to grave, the NREU of recycled fibre is 70% (Wellman), 60% (LJG) and 40% (FENC) lower than that of virgin fibre; the GWP of recycled fibre is 76% (Wellman), 60% (LJG) and 50% (FENC) lower compared to virgin fibre. Figure 5.14 shows the breakdown of NREU and GWP based on different life cycle phases. In the cradle-to-grave B2F recycling systems, post-consumer waste incineration is avoided. The major part of the impact on energy and GHG emissions is related to the recycling processes which are referred to as "RPET fibre" in Figure 5.14. For each recycling system, the impact from the virgin PET fibre is different (see "VPET fibre" in the figure for the three recycling companies), depending on the recycling efficiency, it ranges from 80% to 90%.

Table 5.7: LCA result for 1 tonne of recycled PET fibre, based on the "system expansion" approach, cradle to grave, excluding the use phase.

Recycling route	Mechanical	Semi-mechanical	Chemical BHET	V-PET (W.Europe)
Company	Wellman	LJG	FENC	
Fibre type	Staple	POY	POY	Staple/POY
NREU (GJ)	23	33	48	79
GWP 100a (t CO_2 eq.)	1.33	2.21	2.82	5.54
Abiotic depletion (kg Sb eq.)	11	16	22	38
Acidification (kg SO_2 eq.)	5	10	15	19
Eutrophication (kg PO_4^{3-} eq.)	0.9	0.8	2.1	1.5
Human toxicity (kg 1,4-DB eq.)	845	1020	1310	6150
Fresh water aqua. ecotox. (kg 1,4-DB eq.)	270	220	265	2540
Terrestrial ecotoxicity (kg 1,4-DB eq.)	8	8	16	10
Photochemical oxidant formation (kg C_2H_4 eq.)	0.3	0.4	0.6	0.9

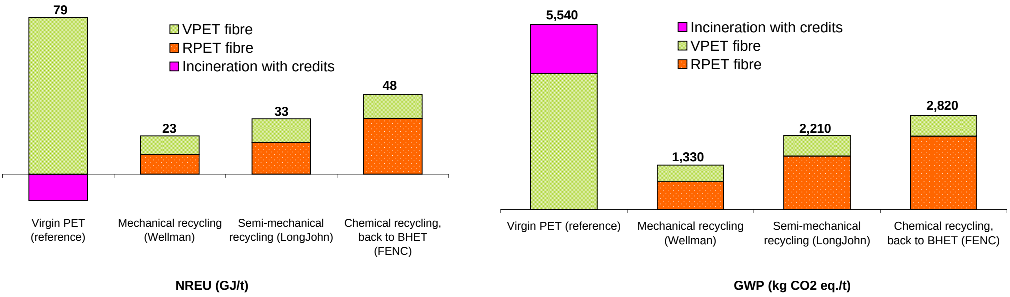


Figure 5.14: Breakdown of NREU and GWP100a for 1 t of PET fibre for the three product systems, cradle to grave without use phase (the life cycles are shown in Figure 5.13; B2F = bottle-to-fibre)

5.6 Discussion

5.6.1 Comparison with other studies

As mentioned in the introduction, most PET recycling studies have focused on waste management rather than the production of recycled products [185, 186, 219]. The results of these LCA studies are not directly comparable with our results because the goal and the functional units are different. Moreover, a transparent dataset on PET recycling is hardly available in public domain. A few studies reported inventory data of flake production. The comparison of flake production shows that the inventory data and the results reported by this study fit well with those reported by Arena et al. [184] and Detzel et al. [185] (see Table 5.8).

Table 5.8: Comparison of flake production with other studies

Output: 1 t of recycled PET flakes	This study	Arena et al. [184]	Detzel et al. [185]
Yield of PET flakes (or material efficiency, wt%)	75%	76%	80%
By-products (wt%)	7-10%	7%	5%
NREU (GJ/t flake)	2.5-6.0	2.7	n/a
GWP100a (kg CO_2 eq./t flake)	310-720	635	n/a

5.6.2 Use of LCA results from the three methods

In this study, three methods were applied for the B2F open-loop recycling case. The three methods take different perspectives. The "cut-off" approach follows the natural business-to-business boundary and is the most commonly used LCA method for recycled products. It is easy to apply and no data is required from outside of the investigated product system. The disadvantage is that the method oversimplifies the environmental impact of the "cradle" and the "grave" stages.

The "waste valuation" method uses economic values to elaborate the "cradle stage" by shifting part of the environmental impact from the virgin polymer to the second life cycle. It is also a method which is easy to apply. However, the allocation factor strongly depends on the market prices that are determined by demand and supply and the macroeconomic development. It is possible to further elaborate the "waste valuation" method by introducing more comprehensive economic indicators (e.g. long-term price elasticity) [189, 192]. These methods are usually more complicated and require data from economic models.

The third approach we applied, the "system expansion" method, takes the real "cradle" and "grave", merges two life cycles into one product system and compares systems with and without recycling. The most important advantage of this method is that it avoids allocation. This method applies life-cycle thinking to the whole system. It is our preferred method for open-loop recycling. The disadvantage of this method is that it is not easy to apply; it results in large systems and the data requirements from extended product systems can be demanding [190].

The use of these LCA results depends on the perspective of a decision maker. From a manufacturer's point of view, it is important to reduce the environmental impact of the production process and the suppliers. The system boundary of cradle-to-factory gate (the "cut-off" and "waste valuation" methods) fits well to the business boundary. Both methods are easy to apply and to communicate. From a life-cycle-thinking perspective, the benefit of recycling is the improvement of the material utilisation efficiency by avoiding further resource extraction and waste management. The overall impact can only be assessed when the entire system and the effect of the system are considered. Therefore, the "system expansion" method represents a life-cycle-thinking perspective.

5.6.3 Comparison with other commodity fibres and renewable alternatives

So far, we only compared PET fibres. It is also interesting to understand the position of recycled PET fibre among other commodity fibres, such as cotton, viscose and PP, as well as novel bio-based fibres, such as Tencel and PLA. Figure 5.15 and Figure 5.16 show the comparisons of NREU and GWP among these fibres. The LCA results of Lenzing Viscose and Tencel fibres were obtained from Shen and Patel [220]. The cotton data is a weighted average of Chinese and US cotton [221, 222]. The eco-profiles of PP resin and PLA resin are obtained from Plastics Europe [163] and NatureWorks LLC [223, 224], respectively. The energy consumption of melt spinning PP and PLA is assumed to be the same as that of PET (see Table 5.2).

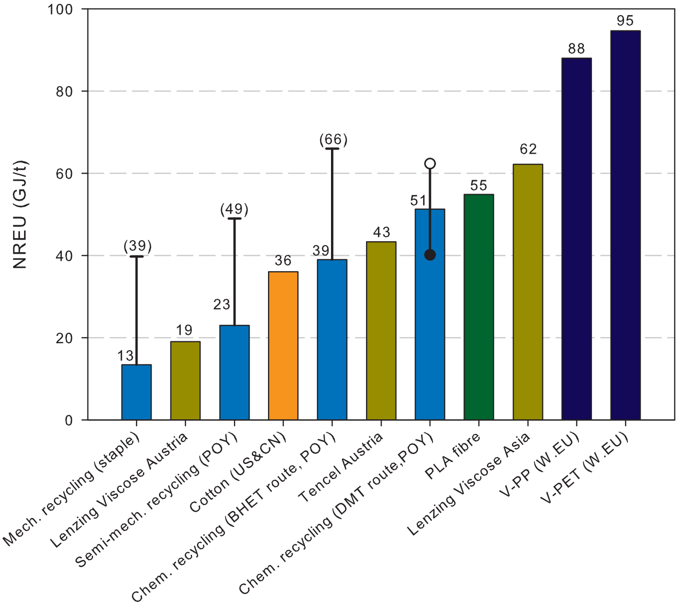
Figure 5.15(a) shows the results for the system boundary cradle to factory gate. For recycled fibres, the default method is the "cut-off" method (for the second life), with the error bar showing the results based on the "waste valuation" method. For recycled fibre produced from the DMT route, only the "cut-off" method was applied; the error bar shows the results based on the high and low cases assumptions (see Section 5.3.3). Figure 5.15(b) shows the cradle-to-grave comparison without the use phase. For recycled PET fibres, the "system expansion" method is applied (except for chemical recycled fibre produced via the DMT route). For other fibres, it is assumed that all the fibre products are used and disposed of in Western Europe in an average MSWI plant with energy recovery (recovery rate = 60%, see Section 5.2.2).

Based on the "cut-off" approach, staple fibre produced from mechanical recycling (Wellman) has the lowest cradle-to-factory gate NREU among all fibre studied; recycled PET fibre produced from semi-mechanical recycling (LJG) has slightly higher NREU than Lenzing Viscose Austria; the NREU of chemically recycled fibre produced by FENC is slightly higher compared to cotton; and chemical recycled fibre via the DMT route has a higher NREU value than Tencel Austria. All recycled PET fibres have lower NREU values than virgin PET and virgin PP, based on the three methods.

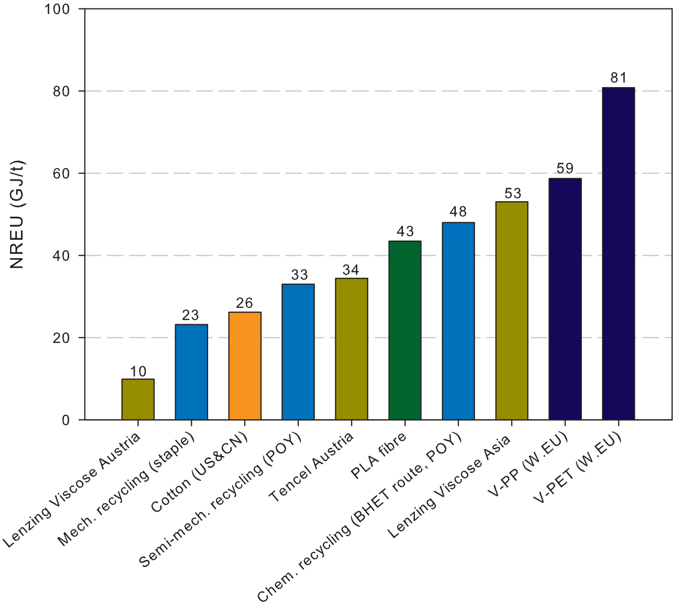
Figure 5.16(a) presents the comparison of cradle-to-factory gate GWP100a based on the "cut-off" method with the error bar showing the results based on the "waste valuation" method. For chemically recycled fibres based on the DMT route, only the "cut-off" approach was applied. Figure 5.16(b) shows the cradle-to-grave comparison without the use phase. For recycled PET fibres, the "system expansion" method is applied. For other fibres in the case study, it is assumed that the fibre products are single-use and the post-consumer waste is incinerated with energy recovery.

Based on the "cut-off" approach (Figure 5.16a), recycled fibre produced via mechanical recycling (Wellman) has a lower GWP value than all the other fibres listed except for Lenzing Viscose Austria; recycled fibre produced from semi-mechanical recycling (LJG) is has a slightly lower GWP than PLA and cotton; recycled fibres produced via chemical recycling (BHET route and DMT route) are comparable with virgin PP. Based on all three methods applied, all studied recycled PET fibres have lower GWP than Lenzing Viscose Asia and virgin PET.

When we compare the energy use and GWP of various fibre products (as in Figure 5.15 and Figure 5.16), it should also be taken into account that fibres are inter-

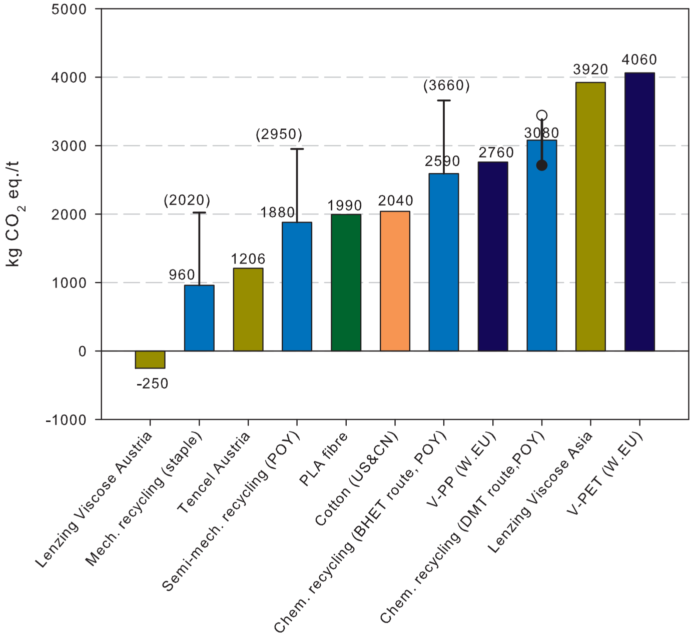


(a) Cradle-to-factory gate NREUof 1 t fibre based on the "cut-off" approach. The error bar shows the results corrected by the "waste valuation" method. For "Chem. recycling (DMT route, POY)", only the "cut-off" approach was applied.

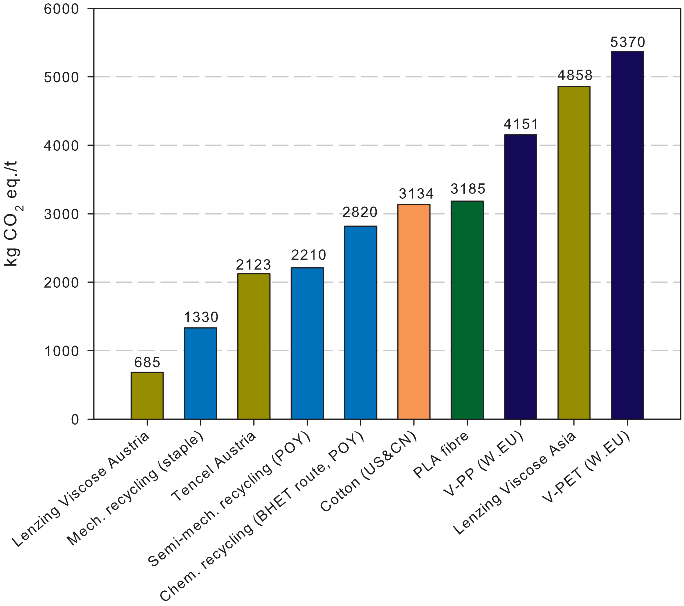


(b) Cradle-to-grave NREU of 1 tonne staple fibre excluding the use phase. For recycled fibres, the "system expansion" method is applied. For other fibres, it is assumed that the post-consumer waste is incinerated with 60% energy recovery rate.

Figure 5.15: Comparison of NREU among recycled PET, virgin PET, virgin polypropylene, cotton, viscose, Tencel and PLA. Data sources (except for recycled PET): [112, 163, 164, 220, 222, 223, 224].



(a) Cradle-to-factory gate GWP of 1 tonne staple fibre based on the "cut-off" approach. The error bar shows the results corrected by the "waste valuation" method. The carbon sequestration of bio-based fibres has been taken into account. For "Chem. recycling (DMT route, POY)", only the "cut-off" approach was applied.



(b) Cradle-to-grave GWP of 1 tonne staple fibre excluding the use phase. For recycled fibres, the "system expansion" method is applied. For other fibres, it is assumed that the post-consumer waste is incinerated with 60% energy recovery rate. The carbon sequestration of bio-based fibres has been taken into account.

Figure 5.16: Comparison of GWP100a among recycled PET, virgin PET, virgin polypropylene, cotton, viscose, Tencel and PLA. Data sources (except for recycled PET): [112, 163, 164, 220, 222, 223, 224].

mediate products. The fibres studied are designed to delivery different functionalities and to fulfill various end-use purposes, and they cannot always replace each other. Table 5.9 shows that the mechanical, thermal and water retention properties of fibres compared are very different. Therefore, if fibre A has a higher environmental impact than fibre B, it does not immediately imply that fibre A should be replaced by fibre B.

Furthermore, in both Figure 5.15(b) and Figure 5.16(b) the use phase is excluded. Here, the use phase includes the fabric and the end product (e.g. shirt) manufacturing stages and the use of the end-product. Depending on the type of fibre, the environmental impacts in the use phase can be substantially different. For example, different types of fibres have different energy requirements, chemical use and generate different types of waste in dyeing, finishing, washing and drying processes; the type of fibre/fabric also determines the life time of product.

Table 5.9: Selected mechanical, thermal and water retention properties of fibres

Fibre name	Fibre type	Company or trade name	Density (g/cm^3)	Tenacity[a] (wet) (cN/tex)	Tenacity[a] (dry) (cN/tex)	Water retention (%)	Melting point (°C)
R-PET[c]	Staple	Wellman	1.36-1.40	30-48	28-48	0-2	245-260
R-PET[d]	Filament	FENC	1.36-1.41	35-45	35-45	3-5	240-250
V-PET[e]	Staple	Dacron®	1.36-1.41	30-55	28-55	3-5	250-260
V-PET[e]	Filament	Serene®	1.36-1.41	40-60	38-60	3-5	250-260
Cotton	Staple		1.5-1.54[e]	26-40[f]	24-36	38-45[g]	n/a[b]
Viscose	Staple	Lenzing Viscose®	1.52-1.54[e]	10-13[f]	24-26	90-100[g]	n/a[b]
PP[e]	Staple	Herculon®	0.9-0.92	25-60	25-60	0	160-175
PLA[h]	Staple	Ingeo™	1.25	n/a[b]	32-36	n/a[b]	170

[a] Tenacity is expressed in relative to the fineness (1 tex = 1 gram per 1000 metres). Figures for tenacity are based on both fiber fineness (tex) and cross-sectional area of the sample.

[b] n/a = data not available or not applicable.

[c] Private communication with Wellman-International Ltd. (2009).

[d] Private communication with Far Eastern New Century Co.(2009).

[e] Schultze-Gebhardt & Herlinger [144].

[f] Abu-Rous & Schuster [149].

[g] Lenzing AG [143].

[h] NatureWorks LLC [225].

5.7 Summary, conclusions and future research

In this study, the environmental impacts of bottle-to-fibre (B2F) recycling were assessed. We investigated four recycling technologies, namely mechanical recycling, semi-mechanical recycling, back-to-oligomer recycling and back-to-monomer recycling. The LCA results were compared with the eco-profile of virgin PET fibre. Three methods were applied for this open-loop recycling case, namely, the "cut-off", "waste valuation" and "system expansion" methods. The "cut-off" and the "waste valuation" methods follow the system boundary of cradle to factory gate. The cradle to grave system is analysed based on the “system expansion” method. The use phase is excluded in this LCA.

Based on all three methods, recycled PET fibre offers 40-85% non-renewable energy savings and 25-75% GWP savings compared to virgin PET, depending on the technology, the chosen allocation method and/or system boundaries. Based on all three methods, bottle-to-fibre recycling reduces impacts for most of the environmental categories studied. In addition, in terms of NREU and GWP100a, recycled PET fibres are comparable to cotton, modern viscose (i.e. Viscose Austria), Tencel and PLA, and they are better than PP, traditional viscose (i.e. Viscose Asia) and virgin PET. Both mechanical and semi-mechanical recycling have lower impacts than chemical recycling via the BHET route. However, it must be acknowledged that fibres produced from chemical recycling can be applied more widely than fibres produced from (semi-) mechanically recycled fibres. This also applies to chemical recycling via methanolysis, which has the highest impacts on NREU and GWP100a among the four recycling technologies investigated, but yields the highest product quality.

The three methods applied in this study take different perspectives. The “cut-off” method is easy to apply and straightforward to communicate. It focuses only on the recycled product and no data is required outside of the investigated product system. However, it simplifies the open-loop allocation issues especially for the “cradle” and the “grave” stages. The “waste valuation” method can be seen as an elaborated “cut-off” method. It uses economic values to allocate the environmental impacts of the production of virgin polymer (which is used for both life cycles). This method follows the suggested procedures by ISO/TR 14049 for recycling. However, the price fluctuation may lead to significant uncertainties for this method. The “system expansion” takes the perspectives of life-cycle thinking. The “system expansion” method is our preferred method to deal with open-loop recycling, although this method is not easy to apply because it requires detailed data outside of the life cycle of the investigated product.

Among the three methods we applied, the “cut-off” approach reflects current environmental policy (e.g. emission trading), where companies or sectors are addressed as individual actors and their actual energy use and emissions are fully taken into account. This is not the case for the “waste valuation” method because it shifts part of the impacts from primary to secondary production. Compared to the “cut-off” approach the “waste valuation” method is less favourable for the recycling industry. However, it can encourage the product design for recyclability because producing recyclable product results in a credit by shifting part of the impacts to the recycled products. The “system expansion” method reflects the overall efficiency of material utilization without distinguishing different players. In a policy context where responsibilities are assigned to individual companies or sectors, it is difficult to apply the “system expansion” method.

We conclude that PET B2F recycling offers important environmental benefits over single-use virgin PET fibre. PET fibre is a product that cannot be further recycled via mechanical recycling. Chemical recycling is technically possible, but the economic viability of large scale operation is still to be proven. Another important way of recycling PET bottles is bottle-to-bottle recycling (see Figure 5.1). This is an example for closed-loop recycling system. In theory, PET can be recycled multiple times before it is finally converted into fibre. The environmental impact of such recycling systems, the effect of the number of cycles and the influence from different allocation methods for open-loop and/or closed-loop recycling should be further investigated.

Chapter 6

Life cycle energy and GHG emissions of PET recycling: change-oriented effects*

6.1 Background and goal

The demand of bottle grade PET (polyethylene terephthalate) has experienced two-digit growth rates in the past two decades [8], making PET the most important packaging plastic next to polyolefins [13]. Meanwhile, PET recycling has become a well-established business. Worldwide in 2007, approximately 10% of used PET bottles were collected and recycled into 3.6 million tonnes of PET flakes [42]. Approximately 72% of these PET flakes were converted into fibres, about 10% were converted into recycled bottles and 18% into other products (e.g. sheets and strapping tapes) [42]. It is expected that in the future more and more used bottles will be recycled back into bottles [8].

Many studies reported on the life cycle assessment (LCA) of PET recycling. Most of these studies were conducted in order to support the decision-making on waste management, e.g. recycling was compared with landfilling or incineration, or to optimise the recycling process [184, 186, 219, 226, 227]. A few studies dealt with the impact of the recycled products where cases of open-loop and/or closed-loop recycling were analysed [185, 228, 229]. Despite the different goals of these studies, all of them concluded that recycling of PET reduces the environmental impact.

The goal of the present study is to gain further insights into PET recycling system, in which used bottles are recycled into both bottles and fibre, the two most important products of virgin and recycled PET. We study the effects related to changes of the following four parameters in this open-loop recycling system: i) the number or recycling

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*Submitted to *International journal of LCA*. Co-authors: Evert Nieuwlaar, Ernst Worrell and Martin K. Patel (Utrecht University).

trips, ii) the shares of recycled PET pellets used for B2B (bottle-to-bottle) and B2F (bottle-to-fibre) recycling, iii) the change of the market demand of bottle and fibre and iv) PET polymer made from bio-based feedstock. The motivations of studying these effects are stated as the following:

- In the previous LCA studies on PET recycling, the effect of multiple recycling trips has not been discussed. The industry has grown fast and is likely to continue in the future [8]. It is expected that the quantity of recycled PET will increase and the recycled polymer can be further recycled. We therefore formulate as our first research question:*what is the effect of multiple recycling trips on the overall environmental impact of PET recycling?*
- The second parameter is related to the market demand of recycled PET pellets. Currently, about seven times more recycled PET pellets are used for recycled fibre than for recycled bottles [42]. If more recycled PET pellets are available for B2B recycling, less of them would be available for fibres. The optimisation of B2B and B2F recycling should be studied to understand how the environmental impact can be minimized. The second research question of this study is: *how does the overall environmental impact change when the share of recycled PET pellets used for B2F and for B2B recycling changes?*
- Worldwide in 2005 approximately 65% of the PET polymer was used to produce fibre and 30% was used to produce bottles [8].[1] In contrast, in Europe only about 35% of PET went into the fibre sector [8]. It is interesting to investigate whether the share of the market demand of bottle and fibre influences the overall environmental impact of the recycling system. This leads to the third research question: *how does the overall environmental impact change when the market demands of PET bottle and fibre change?*
- The fourth parameter is related to bio-based feedstock. Bio-based plastics have attracted much attention in the past decades due to the concerns of limited fossil resources and climate change. Several studies have shown that bio-based materials have lower environmental impacts than their petrochemical counterparts [11, 37, 98, 230, 231]. Bio-based PET and petrochemical PET are chemically identical. A comparative LCA of recycled PET, bio-based virgin PET and bio-based recycled PET has not been conducted so far. Our fourth research question is raised: *how does recycled PET compare to bio-based virgin PET and bio-based recycled PET?*

In LCA, the methodology of open-loop recycling has been extensively discussed but only a few case studies are available in the public domain. In our previous study [228], three allocation methods were applied to open-loop recycling. The three methods are the "cut-off" method, "waste valuation" method and the "system expansion" method. All three methods have different perspectives and thus have different system boundaries. It was concluded that the "system expansion" method is the preferred choice because it implements the life-cycle thinking. In the present study, all four research questions are related to the effect of increased recycling in different forms. There-

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[1] The remaining 5% is used for other applications such as films and injection moulded products [8].

fore, we follow the principle of consequential LCA and apply the “system expansion” method. The details of the model will be presented in Section 6.2. The environmental impacts analysed are non-renewable energy use (NREU) and global warming (i.e. impact on climate change). Section 6.3 shows the results. In Section 6.4, the sensitivities of the key assumptions are analysed. Section 6.5 concludes this study.

6.2 Methodology

6.2.1 Functional unit and system boundary

Considering that, in 2005, 65% of all virgin PET was converted into fibres and 30% into bottles [8], we define the functional unit as 350 kg of PET bottles and 650 kg of PET fibre. Compared to the market shares we increased the amount of PET bottles in the functional unit (from 30% to 35% of the total) in order to arrive at a total output of 1000 kg.

In this study, the functionality of the virgin and recycled products is considered to be identical. In order to meet the regulations of food safety, the recycled PET bottle-grade resin is produced via super-clean recycling processes [8, 229] and a recycled bottle should contain at least 65% of virgin PET, even in the case of repeated recycling trips (thus, a recycled bottle contains a maximum of 35% of recycled PET). This ratio is the practical maximum value because the discoloration effect is acceptable for commercial use [232, 229].

Virgin PET fibre has a wider application spectrum than recycled PET fibre. Shen et al. [228] distinguished the following three main differences: i) PET fibres produced via mechanical recycling are mainly staple fibre, while virgin PET is converted into both staple and filament fibres; these two products serve different applications. ii) Recycled PET fibre cannot be used to produce microfibre. iii) The dyeability of recycled fibre is limited. In short, recycled PET fibre is mainly used in so-called technical applications, while virgin PET fibre can be used in technical applications, apparel and also high performance applications. This should be taken into account when using the results of this LCA.

The system boundary of this LCA is cradle to grave. Since the research questions are related to a system change, we applied consequential LCA modelling (or change-oriented LCA, prospective LCA) [206, 233, 234]. Consequential LCA is used when comparing two (or more than two) systems to support decision-making. In the present study, we distinguish two product systems: the reference system and the recycling system. The reference system is a single-use (or one-way) PET system. The use phases of PET bottles and fibre are identical for both reference and recycling systems and hence cancel out. Furthermore, the amount of PET that is extruded into fibre and blow-moulded into bottles is identical for both systems; for this reason the impacts related to fibre extrusion and bottle blow-moulding cancel out and are consequently not included in this analysis (since this is a change-oriented analysis).

In this study, the “grave” refers to the ultimate end-of-life of a product. This means that the material is not recycled any further and is disposed of. Recycling is an intermediate step between different life stages (see Figure 6.1). This is different from many other LCA studies where recycling is also considered as the "grave". In this study, the "grave" is assumed to be municipal solid waste incineration (MSWI) with energy recovery. The recovery rate is 60% in primary energy terms (see Table 6.2 in Section 6.2.3).

We assume both virgin and recycled PET products to be produced and disposed of in Western Europe. The inventory data are obtained based on the average technology level of mid- or late-2000s (see Section 6.2.3).

6.2.2 Life cycle inventory modelling

Figure 6.1 shows the flow diagram of the reference system and the baseline recycling system, which will be modified in subsequent change-oriented modelling. In the reference system, bottles and fibres are produced from virgin PET (V-PET) polymer and incinerated with energy recovery. In the baseline recycling system, bottles are produced from V-PET polymer. The recycling process started with bottle collection. It is assumed that the used bottles are recycled with a collection rate of 100%[2] and a material efficiency (η) of 95% based on Shen et al. [228] and van der Velden [229]. Based on today's global material flows for recycled PET it is assumed in the baseline recycling system that 88% of the recycled PET (R-PET) pellets is used for B2F recycling and 12% go to the B2B recycling.[3] Since PET fibre cannot be further recycled,[4] the used fibre is disposed of in a MSWI plant with energy recovery. The R-PET pellets going to the B2B recycling are mixed with V-PET polymer in order to produce recycled bottles. In the current commercial practice, the maximum fraction of R-PET used in a recycled PET bottle (ϕ) is 35% [232, 229]. In the baseline recycling system, we assume that the recycled bottles are not further recycled, although in theory it is possible to carry on the recycling.[5] Here, used bottles from the second life are sent to a MSWI plant with energy recovery and the life cycle is completed. Figure 6.1 shows the baseline recycling system and the reference system.

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[2] According to [42], only 10% of the used PET was collected worldwide in 2007. If we would include a low collection rate in the inventory modelling, the changing effect of the recycling system would be hardly visible, i.e. the majority of the impact would originate from the 90% virgin PET which is not recycled. Since the goal of the LCA is to understand the impact of PET recycling, 100% collection rate is assumed here to be able to focus on the PET recycling system.

[3] According to Thiele [42], in 2007 worldwide 72% of the PET flakes were used to produce fibre and 10% were used to produce bottles. In this study we assume all available R-PET are used for B2F and B2B recycling since fibre and bottles are the most important applications of PET. Thus the share of R-PET used for B2F is $72/(72 + 12) \times 100\% = 88\%$; the share of R-PET goes to B2B recycling is $1 - 88\% = 12\%$.

[4] It is difficult to obtain pure stream of PET textile waste because in many cases PET fibres are blended with other textile fibres. Also, it is difficult to remove various additives, e.g. dyes and finishing chemicals, and to sort out a clean stream of PET. Furthermore, PET fibre has a relatively high crystallinity which further restricts mechanical recycling, although chemical recycling is possible; and it has been done [215].

[5] This is not precisely in line with our earlier assumption of 100% collection rate (see footnote 2). However, the baseline case should be seen as the starting point of the analysis. Scenario 1 shows the analysis of multiple recycling trips, with 100% collection rate for each loop.

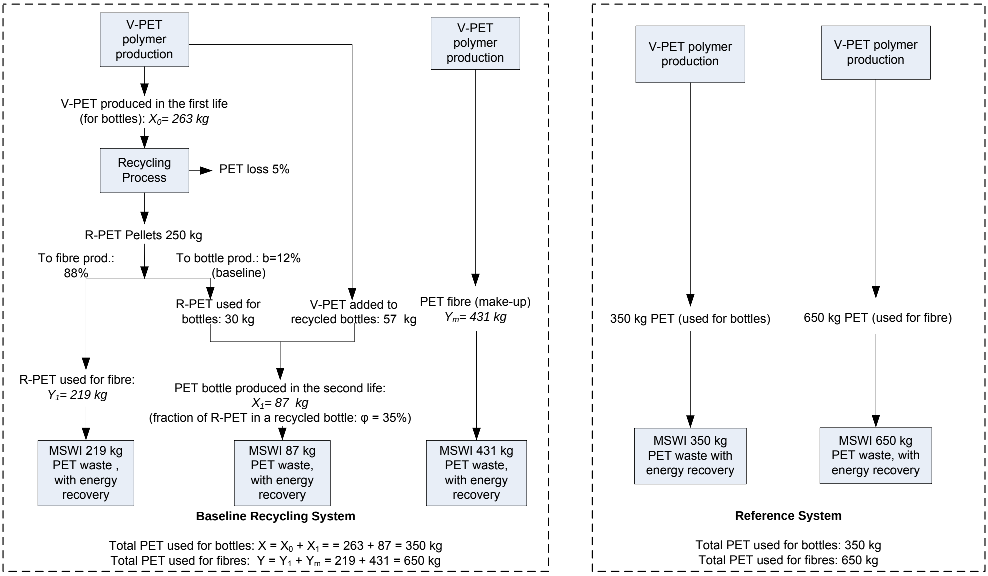

Figure 6.1: Product systems: the baseline recycling system and the reference system (see the modelling in Equations 6.1, 6.2 and 6.3)

Since the total amount of bottle delivered by the first life and the second life has to be 350 kg, it can be calculated that 263 kg of bottles are delivered in the first life and that 30 kg of recycled PET is combined with 57 kg of virgin PET to produce 87 kg of bottles in the second life (see Figure 6.1). The amount of recycled fibre delivered in the second life is 219 kg, which is less than the amount of fibre required for one functional unit. Here, we apply system expansion and make up 431 kg (= 650 – 219) of virgin fibre for this product system. Thus, the amounts of PET bottle and fibre output in this recycling system are the same as those in the reference system. In the reference system, the total amount of incinerated post-consumer PET waste (bottle + fibre) is 1000 kg; while in the recycling system, the total amount post-consumer PET waste is 263 kg less, which is the amount of PET recycled.

Based on the baseline model, four scenarios are constructed in order to answer the four research questions. A summary of the four scenarios is shown in Table 6.1.

In Scenario 1, the effect of multiple recycling loops is analysed. PET bottles are assumed to be recycled multiple times while all other parameters remain unchanged. The mass balance of bottles and fibres can be calculated with the following equations:

$$X_r = \sum_{i=0}^{n} X_i = X_0 \times \sum_{i=0}^{n} (\frac{\eta b}{\phi})^i \tag{6.1}$$

$$Y_r = \sum_{i=0}^{n} Y_{i+1} = \sum_{0}^{n} \eta(1-b) \times X_i = \eta(1-b) X_0 \times \sum_{i=0}^{n} (\frac{\eta b}{\phi})^i \tag{6.2}$$

Table 6.1: Four scenarios in this study

	Baseline	Scenario 1	Scenario 2	Scenario 3	Scenario 4
Changing Parameters	Baseline (current situation)	Multiple recycling loops	Change of the share of R-PET pellets used for B2B recycling	Change of PET fibre and bottle demands	Bio-based PET, and recycled bio-based PET
n (number of recycling trips)	1	>1	>1	>1	1
b (share of R-PET pellets used for B2B recycling)	12%	12%	0-100%	0-100%	12%
Functional unit: bottles/fibre (kg)	350/650	350/650	350/650	650/350	350/650
PET polymer	Petrochem.	Petrochem.	Petrochem.	Petrochem.	Bio-based ethylene glycol & petrochem. PTA

$$X = X_r + X_m \quad Y = Y_r + Y_m \tag{6.3}$$

Where:
X: total amount of bottle per functional unit.
Y: total amount of PET fibre per functional unit.
X_0: amount of V-PET bottle produced in the first life.
X_r: total amount of bottle delivered from the recycling system, including the first-life bottles and all recycled bottles, excluding V-PET used for bottle make-up.
Y_r: total amount of fibre delivered from the recycling system, including all recycled fibre, excluding V-PET for fibre make-up.
X_m: V-PET added to make up the bottle requirement of one functional unit. If $X_r = X$, then $X_m = 0$ (see Figure 6.1).
Y_m: V-PET added to make up the fibre requirement of one functional unit. If $Y_r = Y$, then $Y_m = 0$ (see Figure 6.1 and Figure 6.2).
b: the share of R-PET pellets used for B2B recycling; in the baseline case b=12% (see Figure 6.1).
η: material efficiency of PET bottle-to-pellet recycling; $\eta = 95\%$ (see Table 6.2).
phi: fraction of R-PET used in a recycled bottle; V-PET required is $(1 - \phi)$; ϕ=35% (see Table 6.2).
n: number of recycling trips.

From Equation (6.1) it can be seen that $\sum X_i$ is an infinite geometric series. Since the value of the common ratio $(b \times \eta/\phi)$ is less than 1,[6] it is possible to calculate the finite sum provided that i is large enough:

$$\sum_{i=1}^{\infty} X_i = X_0 \times \sum_{i=0}^{\infty} (\frac{\eta b}{\phi})^i = \frac{X_0}{1 - (\eta b/\phi)} \tag{6.4}$$

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[6] Take the values of b, η and ϕ from Figure 6.1: $\eta b/\phi = 12\% \times 95\%/35\% \approx 0.33 < 1$.

In Scenario 2, the effect of using different shares of the available R-PET pellets for making bottles is analysed. In the baseline case, 12% of the R-PET obtained from the previous life is used to produce bottles (i.e. b = 12%); and the remaining 88% is used to produce fibre. In this scenario, b is changed from 0% (i.e. all R-PET pellets are used for B2F recycling) to 100% (i.e. all R-PET pellets are used for B2B recycling). Furthermore, the change of the environmental impact related to the change of b is analysed for multiple recycling trips (i.e. $n > 1$).

In Scenario 3, the functional unit is changed into 650 kg of bottles and 350 kg fibre (i.e. the quantities are inversed) while other parameters remain unchanged. Figure 6.2 shows the material flow of this scenario. In contrast to the baseline case, V-PET is needed for bottle make-up instead of fibre. Additionally, we also investigate the effect of changing parameter b (i.e. $b = 0 - 100\%$), as well as the effect of multiple recycling trips (i.e. $n > 1$) with the new functional unit.

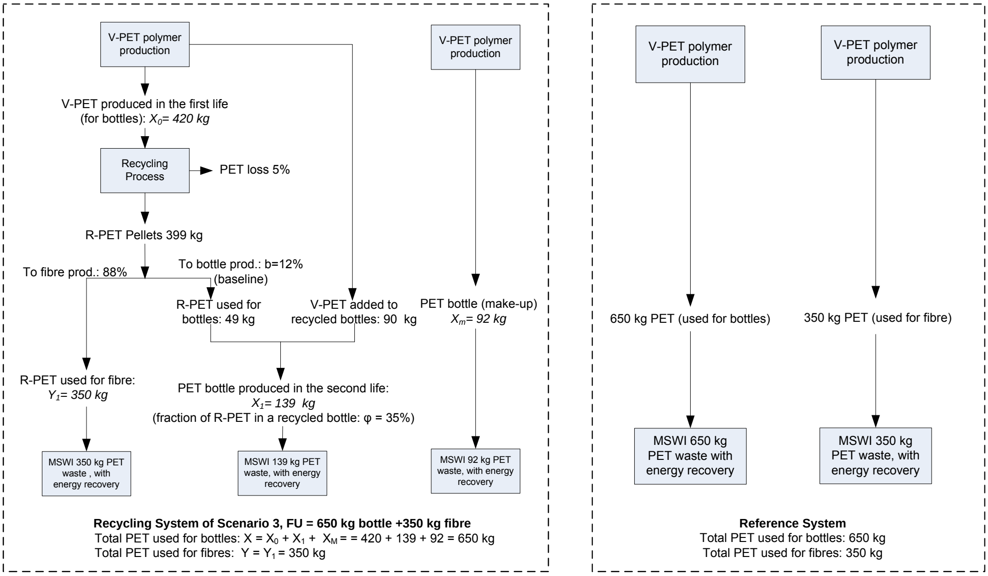

Figure 6.2: Product systems of Scenario 3, functional unit: 650 kg bottle + 350 kg fibre

In Scenario 4, the effect of renewably sourced PET is studied. We change the data for the cradle-to-factory gate polymer production while all other parameters remain the same as in the baseline case. In this scenario, bio-based PET is produced from bio-based EG (ethylene glycol) and petrochemical PTA (purified terephthalic acid); strictly speaking we are hence studying partially bio-based PET. Bio-based EG is produced from bio-based ethylene which is the dehydration product of bio-based ethanol. Currently, the two most important bio-based feedstocks of ethanol production are sugarcane and maize.[7] We assume that the bio-based ethylene in this study is produced

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[7] The US and Brazil are the two biggest fuel ethanol producing countries in the world. In 2009,

from 50% maize-based ethanol and 50% sugarcane-based ethanol. The detailed data used to calculate the impact of bio-based PET can be found in Section 6.2.3. We assume petrochemical PTA because there is no commercially available bio-based equivalent.

6.2.3 Input data

The cradle-to-factory gate impact of virgin PET was acquired from the latest eco-profiles published by PlasticsEurope; the data represents the average technology in Western Europe in the late 2000s [236] (see Section 6.3.5 for the discussion on the former PlasticsEurope's eco-profile). The inventory data of PET bottle-to-pellet recycling were based on literature data, most of which have been cross-checked with industrial experts. Other background data, such as road transportation and grid power generation, were obtained from the Ecoinvent database (Version 2.0). A summary of data and assumptions is shown in Table 6.2.

Table 6.2: Summary of data used in this study

Parameters	**Value**	**Unit**	**Source**
Virgin PET amourphous grade			Based on PlasticsEurope [236], the NREU and GHG emissions (100 years) of PET bottle grade are 68.6 MJ/kg and 2.15 kg CO_2 eq./kg. Based on [112, 205], the NREU and Global warming (100 years) of the SSP step (solid state polymerisation) are 1.96 MJ/kg and 0.10 kg CO_2 eq./kg. The NREU and Global warming impact of PET amorphous are calculated: 68.60-1.96 = 66.64 MJ/t and 2.15-0.10 = 2.05 kg CO_2 eq./kg.
NREU	66.64	MJ/kg	
Global warming (100 years)	2.05	kg CO_2 eq./kg	
Transportation distance, bottle waste collection (d_1)	400	km	Assumed; to be checked in the sensitivity analysis.
Energy use for bottle sorting, compacting and baling	negligible	-	Assumed based on [184, 185, 228].
PET bottle-to-flake production			
Baled PET bottle waste	1316	kg/t flake	[184]
Electricity	278	kWh/t flake	[184]
Heat (from natural gas)	2500	MJ/t flake	[184]
NaOH (30%)	10	kg/t flake	[184]
Sulphuric acid (30%)	20	kg/t flake	[184, 228]
By-products (e.g. PE)	88	kg/t flake	[184]
Allocation factor of by-products	5%	-	Economic allocation [184, 228]
Solid waste[a]	222	kg/t flake	[184]
Transportation distance, flake to pellet production (d_2)	400	km	Assumed; to be checked in the sensitivity analysis.
Pellet production			
Flake input	1031	kg/t pellet	[228]

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the sum of the fuel ethanol production in the two countries accounted for nearly 90% of the world fuel ethanol production [235]. Fuel ethanol is produced from maize in the US and from sugarcane in Brazil.

Table 6.2 – continued from previous page

Parameters	**Value**	**Unit**	**Source**
Heat (from natural gas)	252	MJ/t pellet	[237]
Pellet extrusion	447	kWh/t pellet	[238]
Material efficiency of PET (PET bottle to pellet, PET flow, η)	95%		Assumed based on [228, 229]; to be checked in the sensitivity analysis.
Fraction of R-PET pellet in a recycled bottle (ϕ)	35%		Assumed based on [229]; to be checked in the sensitivity analysis.
MSWI plant with energy recovery			
Gross calorific value of PET	23	MJ/kg	Ecoinvent database V2.0 [202].
Energy recovery from MSWI in Western Europe	60%[b]	in primary energy terms	[162] and Personal communication with Dr. Reimann; to be checked in the sensitivity analysis.
Bio-based PET			
- Bio-based EG			[239]: no land use change is assumed for maize and sugarcane production in the US and Brazil. Values reported in this table are based on 50% maize and 50% sugarcane as the feedstock.
NREU	17	MJ/kg EG	
Global warming (100 years)	−0.55[c]	kg CO$_2$ eq./kg EG	
- Petrochemical PTA			PlasticsEurope [236].
NREU	53	MJ/kg PTA	
Global warming (100 years)	1.33	kg CO$_2$ eq./kg PTA	
- Polymerisation			Patel et al. [240]
Natural gas	2.29	GJ/t PET	
Electricity	101	kWh/t PET	
Steam	240	kg/t PET	
PTA	867	kg/t PET	
EG	334	kg/t PET	

Data obtained from the Ecoinvent database V2.0	**Process names in the Ecoinvent database**
Transportation by road	"Transportation, >32t lorry, EURO3/RER"
Heat from natural gas	"Heat, natural gas, at industrial furnace low-NO$_x$ >100 kW/RER"
EU grid electricity mix[d]	"Electricity, low voltage, production [grid name], at grid/[grid name]"
NaOH	"Sodium hydroxide, 50% in water, Production mix, at plant/RER"
Sulphuric acid	"sulphuric acid, liquid, at plant/RER"
Nitrogen	"Nitrogen, liquid, at plant/RER"
MSWI of PET	"Disposal, polyethylene, 0.4% water, to municipal incineration/CH"

[a] Solid waste is sent to a MSWI plant with energy recovery.

[b] The efficiencies of electricity and heat are 10.6% and 22.3% in an average MSWI plant in Europe according to [162]. This means that 1 GJ waste yields 0.106 GJ$_e$ (electricity) and 0.223 GJ$_{th}$ (heat). These amounts of electricity and heat would be otherwise produced conventionally with an electricity efficiency of 30% and a heat efficiency of 85% (assumed). Thus, 0.106 GJ$_e$ electricity replaces 0.106/30% = 0.35 GJ$_p$ primary fossil fuels and 0.223 GJ$_{th}$ heat replaces 0.223/85% = 0.26 GJ$_p$ primary fossil fuels. The total primary fossil fuel that can be avoided is 0.35 GJ$_p$ + 0.26 GJ$_p$ = 0.61 GJ$_p$ – this is approximately 60% of the energy content of the waste.

[c] Bio-based carbon has been considered as negative CO$_2$ emissions. See Section 6.2.4 for the method of GWP.

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Table 6.2 – continued from previous page

Parameters	Value	Unit	Source

[d] European electricity mix: 65% from the UCTE grid, 13% from the NORDEL grid, 9% from the CENTREL grid, 12% from the UK grid and 1% from the Irish grid. UCET is Union for the Co-ordination of Transmission of Electricity; countries included in UCTE are Austria, Bosnia and Herzegovina, Belgium, Switzerland, Germany, Spain, France, Greece, Croatia, Italy, Luxemburg, Macedonia, Netherlands, Portugal, Slovenia and Serbia and Montenegro. NORDEL is Nordic countries power association, including Denmark, Norway, Finland and Sweden. CENTREL stands for Central European power association, including Czech Republic, Hungary, Poland and Slovakia.

6.2.4 Environmental impact categories: NREU and Climate Change

NREU (non-renewable energy use) is the sum of cumulative fossil energy and nuclear energy [167]. Cumulative fossil energy is a good proxy of the environmental performance of a product [106]. The impact on global warming is calculated based on the characterisation factor of global warming potential based on IPCC guidelines with the timeframe of 100 years [171]. For bio-based PET, the biogenic carbon embedded in the polymer is taken into account as negative GHG emissions for the system cradle to factory gate [151]. Consequently, in the “grave” stage, the biogenic CO_2 released from the combustion of PET are added to the cradle-to-grave GHG emissions.

6.3 Results

6.3.1 The baseline case

Figure 6.3 shows that both NREU and global warming of the baseline recycling system are approximately 20% lower compared to the reference system. The impact reductions are mainly attributed to the decrease in V-PET fibre requirement, which is 650 kg in the reference system and 431 kg in the recycling system (see Figure 6.1). The decrease of V-PET for bottles is not as significant as for fibre. Only 30 kg (or 9%) of V-PET bottle are saved by the recycling system. As a result, the NREU and global warming impact are both reduced by only approximately 9%. Moreover, the recycling process, which converts 263 kg of used bottles (i.e. first life V-PET bottles) into 250 kg of R-PET pellets, has a minor contribution to the overall impact of the recycling system. The NREU of the 250 kg of R-PET pellets is about 2.4 GJ (including recycling process, excluding the feedstock energy), which is only $1/4^{\text{th}}$ of the NREU required to produce the same amount of V-PET pellets (excluding feedstock energy). Both systems receive energy credits and GHG-emission penalties from the combustion of post-consumer (PC) waste in an MSWI with energy recovery (ER). The baseline case saves a total amount of PC waste of 263 kg (i.e. the same amount as the first life V-PET bottles), which results in a lower impact on global warming; these savings are slightly compensated by the lower energy credits.

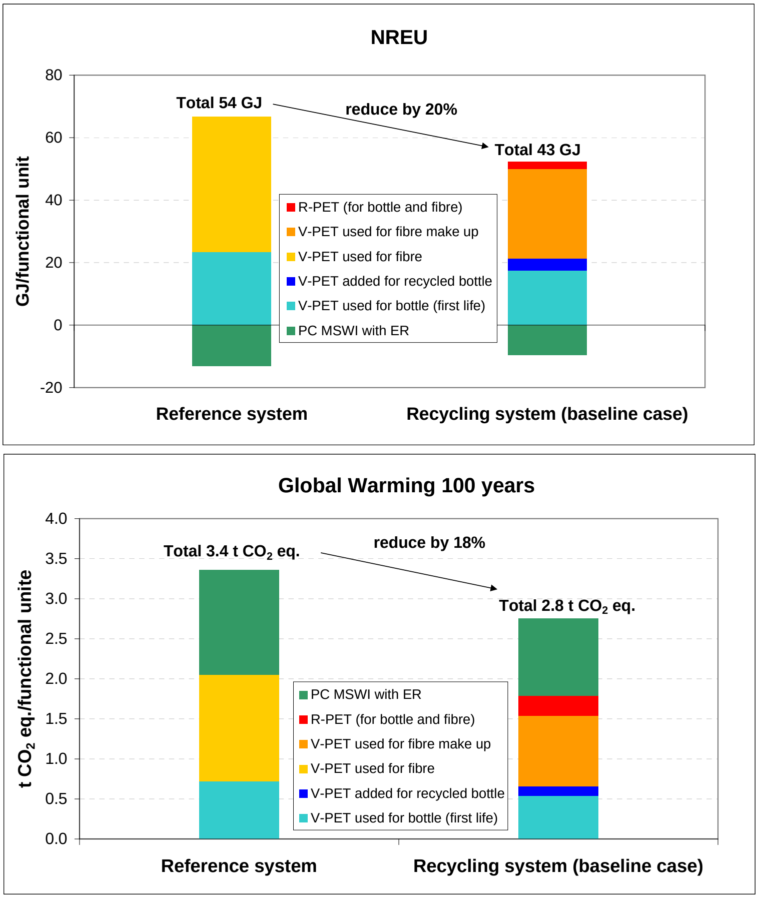

Figure 6.3: Cradle-to-grave NREU (above) and global warming (below) of the baseline recycling system and the reference system

6.3.2 Scenario 1: multiple recycling trips – effect of n

Figure 6.4 demonstrates that if the number of recycling trips (n) increases, both NREU and global warming decrease dramatically until n is equal to four. After four recycling trips, both impacts remain constant. This pattern reflects the property of the geometric series when the common ratio is less than 1 (see Equation 6.1). If the recycling would be carried on for infinite times, the NREU and global warming would decrease to a marginally lower value, i.e. to 39.5 GJ/functional unit and 2.58 t CO_2 eq./functional unit, representing the maximal NREU and GHG-emission savings of 26% and 23% compared to the reference system. Four and more cycles hence allows to increase the NREU and GHG-emission savings by a maximum of six percentage points compared to the baseline recycling system.

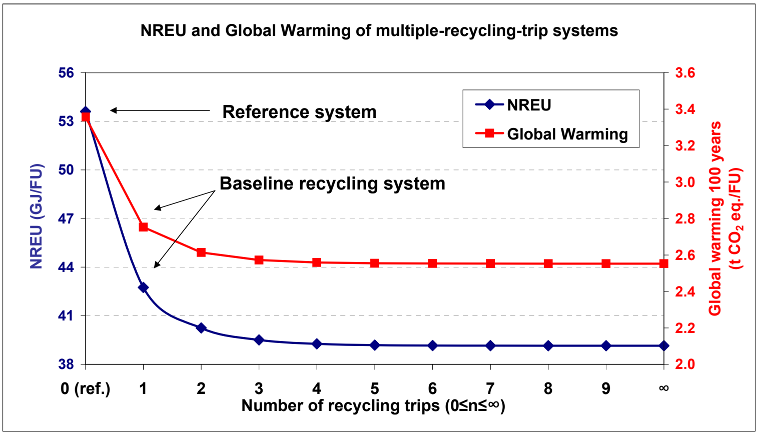

Figure 6.4: Cradle-to-grave NREU and global warming of multiple-recycling-trip systems in Scenario 1

6.3.3 Scenario 2: Change the share of R-PET pellets used for B2B recycling - effect of b

In the baseline recycling system, 12% of the bottle waste is used for B2B recycling (b = 12%). If more R-PET pellets are used for B2F recycling (i.e. decreasing b), the overall impacts decrease (see Figure 6.5). When all R-PET pellets are used for B2F recycling (i.e. b = 0%), NREU and global warming decrease by approximately 27% and 24% compared to the reference case. When b is decreased to 0%, the environmental impacts of the recycling system do not change with the number of recycling trips (n) (see Figure 6.5), because the first-life virgin bottles are all converted into recycled fibre and fibre cannot be further recycled. If more R-PET pellets are used for B2B recycling (i.e. increasing b), the overall impacts increase (see Figure 6.5). When all R-PET pellets are used for B2B recycling (i.e. b = 100%) and after four recycling trips (i.e. $n > 4$), the NREU and global warming decrease only about 10% compared to the reference case.

These dependencies can be understood by breaking down the mass balance of the system. Table 6.3 shows such a breakdown for one recycling trip ($n = 1$). When more R-PET pellets are used for B2B recycling (i.e. with increasing value of b), the amount of V-PET used for first-life bottles decreases, lowering the impact of V-PET used for first-life bottles. However, this also leads to less recycled PET fibre. Consequently, the system requires more V-PET for fibre make-up in order to fulfil the demand. The increase of V-PET used for fibre increases the total V-PET requirement. As shown in Table 6.3, the total V-PET requirement increases from 67% of the system's total material requirements when b = 0%, to 91% when b = 100%. In addition, less V-PET used for first-life bottles also leads to less recycled PET in the system, resulting in more PC waste at the end-of-life stage.

Based on the analysis of this Scenario, we conclude that the environmental benefit of B2F recycling is greater than B2B recycling, when the demand of fibre is higher than the demand of bottle. This is caused by a larger total amount of R-PET produced in the B2F system than in the B2B system. In other words, if the market does not require many bottles, increasing the amount of B2B recycling does not bring additional environmental benefits. In Scenario 3, we will further discuss the change of the environmental impact based on a different market demand.

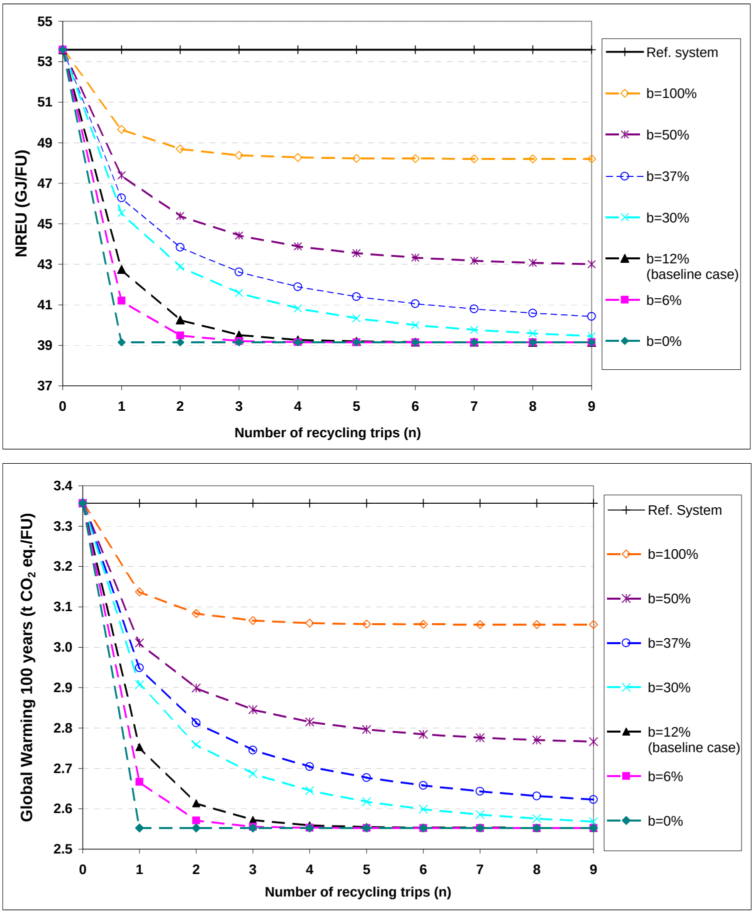

Figure 6.5: Scenario 2: Effects of changing the share of recycled PET pellets used for B2B recycling (b) (above: NREU, below: global warming)

6.3.4 Scenario 3: Change the demand of PET fibre and bottle (functional unit = 650 kg bottle and 350 kg fibre)

As explained earlier, the functional unit is defined based on the current market demand of PET (see Section 6.2.1). However, the PET market varies considerably in different

Table 6.3: Mass balance for different values of b in Scenario 2 (functional unit: 350 kg of bottles + 650 kg of fibre; $n = 1$; b is the share of recycled PET pellets used for B2B recycling)

(kg per functional unit)		Share of bottle waste going to B2B recycling (b)							Reference
		0%	6%	12% (baseline)	30%	37%	50%	100%	
A	V-PET (first-life bottle)	350	301	263	193	175	148	94	350
B	V-PET (added to r-bottle)	0	32	57	102	114	131	166	0
C	V-PET fibre (make up)	318	382	431	522	545	579	650	650
D	R-PET (used for r-bottle)	0	17	30	55	61	71	90	0
E	R-PET (used for r-fibre)	333	268	219	128	105	71	0	0
F	Total PC MSWI with ER	650	700	737	807	825	852	906	1000
V-PET total (A+B+C)		67%	71%	75%	82%	83%	86%	91%	100%
R-PET total (D+E)		33%	29%	25%	18%	17%	14%	9%	0%

regions. For example, in Asia over 80% of the PET polymer was used to produce fibre in 2005 [8]. In this scenario, we intend to understand how the change of PET demand can influence the environmental impact of the recycling system. We now change the reference system, assuming the inversed case where the demand of fibre is less than PET bottle, i.e. the functional unit is defined as 650 kg of PET bottle and 350 kg of PET fibre. The results of NREU and global warming are shown in Figure 6.6.

In this scenario, the recycling system reduces NREU and global warming by approximately 30% compared to the reference system. The impact reductions of the recycling system in this scenario are larger than those of the baseline recycling case and also larger compared to all scenarios discussed above. The reason is that the quantities of R-PET are larger than those of the systems discussed above, i.e. 400 kg (or 40%) as compared to a maximum of 333 kg (or 33%) in the earlier cases (see Table 6.3 when $b = 0\%$). More R-PET leads to less V-PET required in the system, which substantially reduces the impact (see Figure 6.6). In the recycling system, no V-PET is required for fibre-making, i.e. the 350 kg of fibre is 100% produced from R-PET. Figure 6.6 also shows that the savings from the V-PET bottles are not significant. Only 7% of the V-PET bottle is saved by recycling. And $1/7^{\mathrm{th}}$ of the total V-PET are used for the bottle make-up purpose.

It should be noted that this scenario implies that virgin fibre production is not needed anymore. In reality, while recycled PET fibre can partly replace virgin PET fibre, 100% substitution is rather unlikely. The functional equivalence of V-PET and R-PET bottles and fibres has been discussed in Section 6.2.1.

Furthermore, it should be emphasised that the reference system in this Scenario is not comparable with the reference system in the baseline recycling case because the functional units differ. Both reference systems have the same impacts (compare Figure 6.4 with Figure 6.3) only because the fibre extrusion and bottle blow-moulding steps are cancelled out in the consequential LCA modelling. The comparison between the baseline recycling case and this scenario should be strictly about the impact reduction.[8]

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[8] The NREU and GHG emissions of converting amorphous PET into 1 t of fibre are about 13 GJ and

In Scenario 2 we found that the overall environmental impact of the recycling system is sensitive to the share of R-PET pellet used for B2B recycling (b) (see Figure 6.5). In Scenario 3, the overall environmental impact is also sensitive to the value of b, but in a different way. Figure 6.7 shows that with one recycling trip (i.e. $n = 1$), both NREU and global warming reach the lowest point when b is 17%. The breakdown of mass balance (see Table 6.4) indicates that when b is 17%, no make-up V-PET is required.[9] The amount of R-PET produced in the recycling system is maximised (58%). As a result, the impact of the recycling system is minimised. When b is less than 17%, V-PET is needed to make up the demand of bottles; when b is more than 17%, V-PET is needed to make up the demand of fibre. Either way of make-up increases the V-PET requirements and reduces the R-PET produced in the system, which in turn increases the overall impact. If more recycling trips are assumed (i.e. $n > 1$), similar patterns can be observed. In Figure 6.7 the minimal impact of the recycling system are observed when b is 26%, 31% and 33% (with $n = 2$, 3 and 4, respectively). These points represent the recycling systems where no make-up fibre or bottles are needed.

Based on this scenario, we conclude that the impact reduction of the recycling system is sensitive to the choice of the reference system. When the reference system contains more bottles (650 kg) than fibres (350 kg), the maximal savings of the recycling system can be achieved when no V-PET is required for make-up purpose, i.e. when the demand of bottles and fibre can be fulfilled by only recycling the used bottles. All these optima are characterised by a maximum of R-PET in the system.

The environmental benefit of recycling originates from producing R-PET to replace V-PET in the system. From this point, we carry the analysis a step further and calculated the energy/GHG emission savings[10] per tonne of R-PET produced in the system. The results show a linear relationship between NREU (or GHG emissions) savings and the quantities of R-PET in a recycling system. Thus, impact savings per tonne of R-PET is a constant value, regardless of the scenarios taken, i.e. recycling every tonne of R-PET results in a NREU saving of 43.5 GJ and a GHG-emission saving of 2.4 t CO_2 eq./t. This value is determined by the credit received from saving V-PET, the penalty received from the recycling process (from bottle collection to pellet production), the credit and penalty received from post-consumer MSWI, and the material efficiency of the recycling process (η). A detailed reasoning on this linear relationship can be found in the Appendix of this paper.

0.7 t CO_2 eq. (calculated based on [164]). The NREU and GHG emissions of converting amorphous PET into 1 t of PET bottles (SSP + bottle moulding) are 23 GJ and 1.4 t CO_2 eq. (calculated based on [112, 205]). Therefore, making bottles is more energy- and GHG-intensive than making fibre. However, the goal of this study is not to compare PET bottles with fibre, but to understand the impact of the recycling system. The fact that making bottles requires more energy than making fibre is not relevant if the recycling systems are compared with the reference system.

[9] In Equations (6.1), (6.2) and (6.3), let $X = X_r = 650$ kg and $Y = Y_r = 350$ kg (thus, $X_m = Y_m = 0$, i.e. no make-up V-PET is needed). Since η is 95% and ϕ is 35%, it can be calculated that $b \approx 17\%$ when n is 1, $b \approx 26\%$ when $n = 2$, $b \approx 31\%$ when $n = 3$ and $b \approx 33\%$ when $n = 4$.

[10] NREU (or GHG-emission) saving is defined as the NREU (or the GHG emissions) of the reference system minus the NREU (or the GHG emissions) of the recycling system.

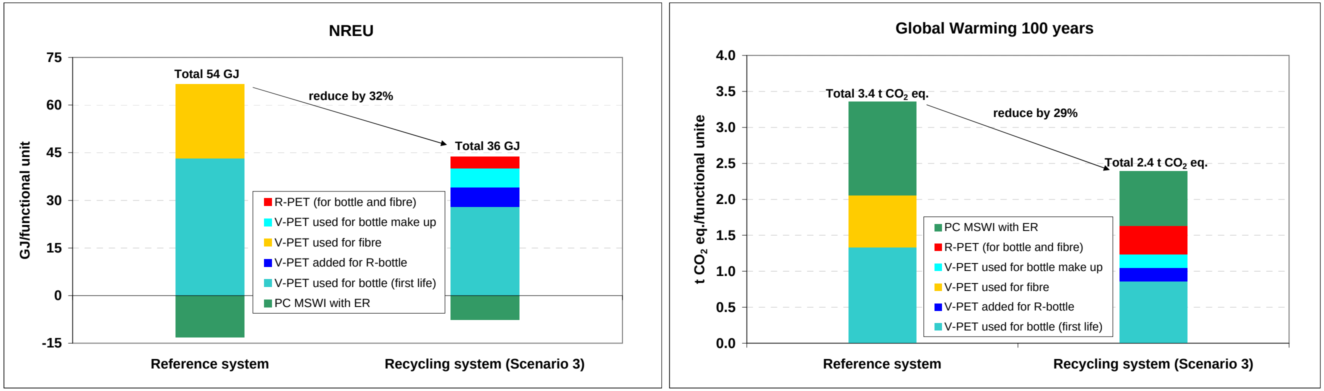


Figure 6.6: Cradle-to-grave NREU (left) and global warming (right), functional unit: 650 kg bottle and 350 kg fibre in Scenario 3, with default $b = 12\%$ (b is the share of recycled PET pellets used for B2B recycling)

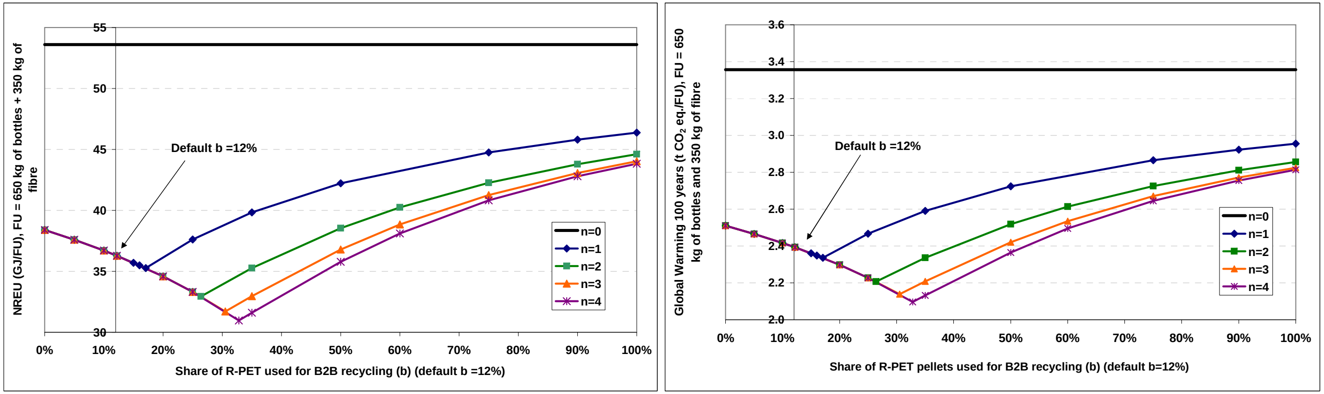


Figure 6.7: Effect of changing the share of recycled PET pellets used for B2B recycling (b) in Scenario 3 (left: NREU, right: global warming)

Table 6.4: Mass balance for different values of b in Scenario 3 (functional unit: 650 kg bottle + 350 kg fibre; n = 1, b is the share of recycled PET pellets used for B2B recycling)

(kg per functional unit)		Share of bottle waste going to B2B recycling (b)							Reference
		0%	5%	10%	12% (default)	17% (minimal)	50%	100%	
A	V-PET (first-life bottle)	368	388	409	420	444	276	175	650
B	V-PET (added to r-bottle)	0	34	72	90	134	243	309	0
C	V-PET fibre (make up)	0	0	0	0	0	219	350	350
D	V-PET bottle (make up)	282	210	130	92	0	0	0	0
E	R-PET (used for r-bottle)	0	18	39	49	72	131	166	0
F	R-PET (used for r-fibre)	350	350	350	350	350	131	0	0
G	Total PC MSWI with ER	632	612	591	580	556	724	825	1000
V-PET total (A+B+C+D)		65%	63%	61%	60%	58%	74%	83%	100%
R-PET total (E+F)		35%	37%	39%	40%	42%	26%	17%	0%

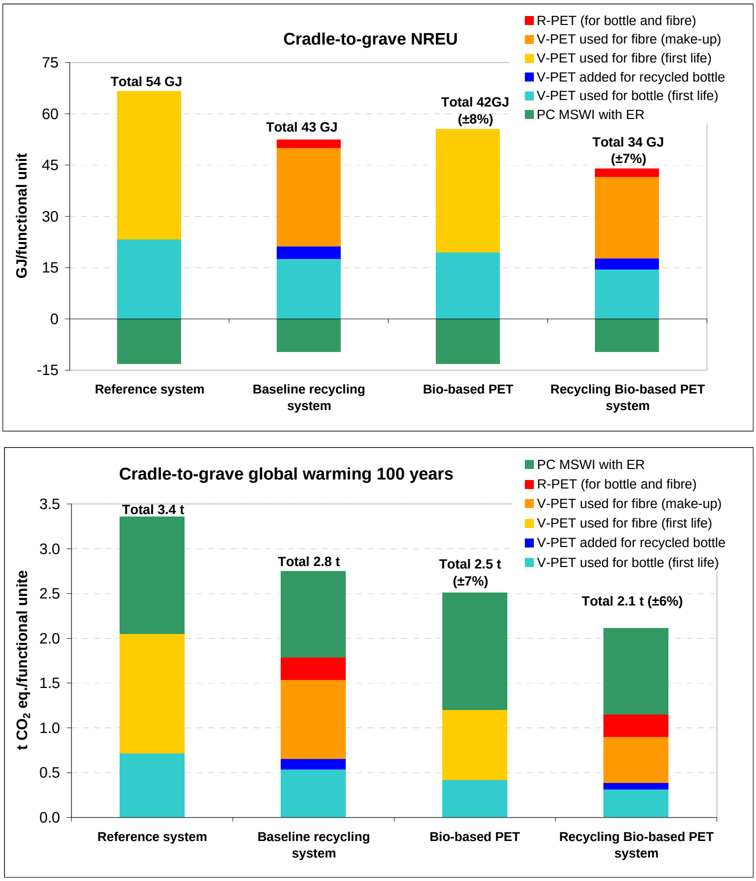
6.3.5 Scenario 4: Renewably sourced PET

Figure 6.8 shows the comparison of four product systems including: the reference system, the baseline recycling system, bio-based PET (single-used system) and recycled bio-based PET for the functional unit of 650 kg fibres and 350 kg bottles. The following results can be observed:

- The system “Recycled bio-based PET” has the lowest impact among all four product systems; it offers at least 35% of the impact reductions (for both NREU and GHG emissions) compared to the reference system and at least 20% of impact reductions compared to the baseline recycling system.
- The product system of (virgin) bio-based PET, i.e. without recycling, saves NREU and GHG emissions by 21% and 25%, respectively, compared to the reference system where petrochemical PET is used (also without recycling).
- The (virgin) bio-based PET system is comparable to the recycled, petrochemical PET system (i.e. the baseline recycling system).

The environmental benefits of bio-based PET and recycled bio-based PET system originate from the low impact of the production of bio-based PET. The cradle-to-factory gate NREU of bio-based PET is 55 GJ/t, which is 17% lower compared to petrochemical PET. The cradle-to-factory gate global warming of bio-based PET is 1.2 t CO_2 eq./t, which is 40% lower compared to petrochemical PET. In the default case, we assume 50% sugarcane-based ethanol and 50% maize-based ethanol for bio-based PET (see Table 6.2). If only sugarcane-based ethanol or only maize-based ethanol is used, the overall impact changes by less than ±8%. Thus the uncertainty of the bio-based feedstock is small. The overall conclusions on bio-based PET and recycled bio-based PET do not change.

From the results of Scenario 1, 2 and 3, we understand that the impact of the recycling system can be further reduced by increasing the number of recycling trips,



Note: 1) For global warming, carbon sequestration of bio-based PET has been included in the cradle-to-factory gate analysis. 2) Percentages in brackets show the uncertainties originate from different bio-based feedstocks; the lower range stands for 100% sugarcane-based ethanol and the higher range stands for 100% maize-based ethanol.

Figure 6.8: Cradle-to-grave NREU (above) and Global warming (below) of bio-based PET and recycled bio-based PET, in comparison with reference system and the baseline case; functional unit: 350 kg of PET bottle + 650 kg of PET fibre

maximizing the quantity of R-PET in the system, and/or by reversing the functional unit into 650 kg of bottle and 350 kg of fibre. These conclusions are also valid for the system “recycled bio-based PET”. It can be calculated that the impact of the system “recycled bio-based PET” can be further reduced by approximately 50% if the functional unit is 650 kg of bottles and 350 kg of fibre, n is 4 and b is 33% (the optimization point when n is 4, see Section 6.3.4).

It should be noted that the eco-profile of petrochemical PET has been substantially improved over the last five years in Western Europe. The most recently published eco-profile from PlasticsEurope shows that the cradle-to-factory gate NREU and GHG emissions of amorphous PET have been reduced by 17% and 38%, respectively, compared to the previous eco-profile of PET [112, 236]. The low impact of virgin PET polymer leads to the low impact of PET recycling system. As a result, recycled PET becomes competitive with the virgin bio-based PET.

6.4 Sensitivity analysis

Figure 6.9 shows the sensitivity of three assumed parameters ϕ, η and d (the transportation distances, see Table 6.2, $d = d_1 + d_2 = 400 + 400 = 800$ km) and energy recovery (ER) rate of MSWI to the environmental impact of the baseline recycling case. Since the results for global warming are very similar those for NREU, we only present the latter in this section.

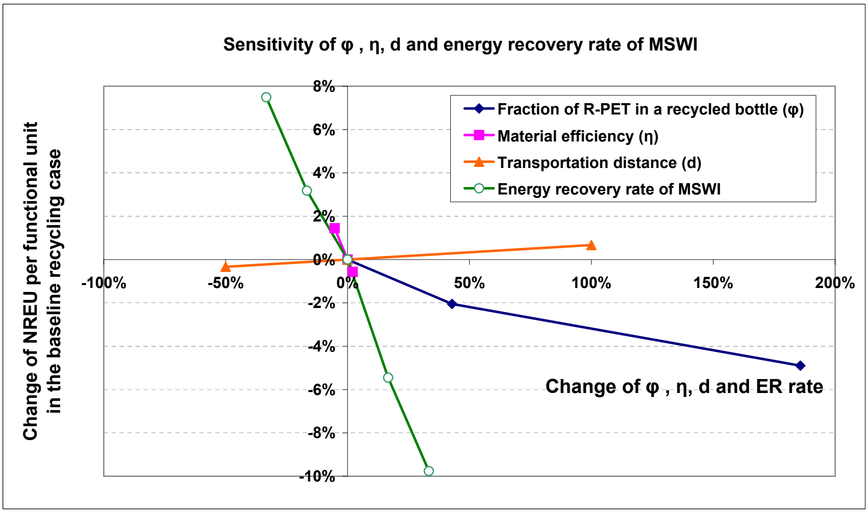


Figure 6.9: Sensitivity of the fraction of R-PET in a recycled bottle (ϕ), material efficiency of bottle-to-pellet recycling (η), transportation distance (d) and energy recovery rate of a MSWI plant in the baseline case

In the LCA, it is assumed that a recycled bottle contains 35% of R-PET and 65% of V-PET. If we increase the share of R-PET in the recycled bottle, less V-PET is required. Consequently, the overall impact of the recycling system decreases. When ϕ is increased from 35% to 100%, NREU decreases by less than 5%. Therefore, the sensitivity analysis shows that the impact reduction is not substantial.

In the LCA, it is assumed that 95% of the used PET can be converted into recycled PET. This assumption was made based on the current industrial practice. Since the material efficiency is a relatively stable parameter in the PET recycling industry, in the sensitivity analysis we slightly change the value of η, i.e. between 90% and 97%. A higher material efficiency leads to a lower environmental impact of the recycling system. Figure 9 shows that NREU is not sensitive to the change of η. When η is varied between 90-97%, the change of NREU is less than ±2%.

The third parameter for which the sensitivity is analysed is the transportation distances (d) in PET recycling. In the LCA, a total amount of 800 km of transportation distance is assumed, including 400 km for bottle waste collection and 400 km for flake transportation. When d is changed between 400 km and 1600 km, NREU changes less than ±1%. Therefore, the overall impact is not sensitive to the transportation distance.

In the default analysis, we assumed that the average energy recovery rate of a MSWI plant is 60% (in primary term, see Table 6.3). In the sensitivity analysis, we altered the energy recovery rate from 40% to 80%, representing the changes of ±33%. The result shows that under a very high energy recovery rate (80%), the overall NREU decreases by 10%; under a very low energy recovery rate (40%), NREU increases by 8%. Thus the impact is not sensitive to the energy recovery rate.

6.5 Conclusions

In this study, the environmental impacts (i.e. NREU and global warming) of PET B2B and B2F recycling were assessed. The functional unit is linked to the market demand of PET bottle and fibre. We used the “system expansion” approach for open-loop recycling. Consequential LCA modelling was applied to gain insights of the change-oriented effects. The analysis was started with the baseline recycling case which is established on the current situation of PET B2B and B2F recycling. Based on the baseline recycling case, four scenarios were built to analyse the change-oriented effects. The main findings of this study are summarised as the following:

1. When comparing the baseline recycling system with the reference system, the overall environmental impacts (including both NREU and global warming) are reduced by approximately 20% by the baseline recycling system; the savings mainly originate from the decrease of V-PET fibre requirements. Multiple-recycling trips can further reduce the environmental impact of the recycling system by maximally 26% compared to the reference system. The additional savings become negligible when n is more than three.

2. Based on the reference system where more fibre (650 kg) is needed than bottles (350 kg), B2F recycling can achieve greater impact reductions (in both NREU and global warming) than B2B recycling; if all R-PET pellets are used to make fibre, the impact of the recycling system reduces to the lowest point, characterised by a maximum of R-PET pellets produced by the system. Increasing the amount of B2B recycling does not bring additional environmental benefits, when the market does not require many bottles.

3. Based on Scenario 3, we conclude that the impact reductions of the recycling system are sensitive to the choice of the reference system. If more bottles (650 kg) are needed than fibre (350 kg) in one functional unit, both NREU and global warming are reduced by approximately 30% for one recycling trip compared to

the reference system. A further finding is that the impacts are sensitive to the share of R-PET pellets used for B2B and B2F recycling. When no extra V-PET is needed for the make-up purpose, the quantities of recycled PET in the system are maximised.

4. Based on the first three scenarios, we conclude that all the optima are characterised by a maximum of R-PET in the system.

5. In Scenario 4, we analysed the role of bio-based PET. Compared to the reference system, the single-used, bio-based PET system reduces NREU and global warming by 21% and 25%, respectively. The recycling bio-based PET system has the best environmental profile among the four systems studied; it offers 35% NREU savings and 37% GHG-emission savings compared to the reference system. Considering the outcome of Scenario 3 we can conclude that the savings would be even larger if bio-based PET were recycled for the purpose of a functional unit of 650 kg bottles + 350 kg fibres.

We consider that the uncertainty of the study is small. Most data are collected from peer-reviewed publications or industrial data. Furthermore, the sensitivity analysis shows that results are not sensitive to key assumptions.

In this study, we analysed the effect of certain changes to the PET recycling system. It should be noted that for all four scenarios, we assumed ceteris paribus conditions. For example, the change of the share of R-PET pellets used for B2B and B2F recycling does not affect the market demand of virgin bottle and fibre, and vice versa. However in reality, the PET material flow is not a static system, but a dynamic one. Further research is recommended to understand the dynamics of PET production and consumption. In that case, dynamic LCA modelling should be applied. It is recommended that other ways of using PET which may lead to a lower environmental impact (e.g. re-use of PET bottles) should be investigated and compared to the recycling approach.

6.A Appendix: The linear relationship between NREU saving and the mass of R-PET in the system

Define $NREU_{saving} : E_{ref} - E_{recycl}$
Where:
E_{ref} : cradle-to-grave NREU of the reference system (GJ/FU)
E_{recycl} : cradle-to-grave NREU of a recycling system (GJ/FU)

E_{ref} consists of the NREU of two life stages: the production of V-PET and MSWI with energy recovery.

$$E_{ref} = FU \times (NREU_{\text{V-PET}} + NRUE_{MSWI}) \tag{6.5}$$

Where:
FU: functional unit, which is 1000 kg PET polymer (regardless of the shares of bottles and fibre)
$NREU_{\text{V-PET}}$: cradle-to-factory gate NREU of one tonne of V-PET, which is approximately 67 GJ/t (see Table 6.2)
$NREU_{MSWI}$: NREU of incinerating one tonne of PET waste in a MSWI plant with energy recovery, recovery rate = 60% (see Table 6.2)

E_{recycl} is determined by three elements: the amount of V-PET (and the related impact), the amount of R-PET (and the related impact) and the amount of PC waste in the system (and the related impact).

$$E_{recycl} = M_{\text{V-PET}} \times NREU_{\text{V-PET}} + M_{\text{R-PET}} \times NREU_{\text{R-PET}} + M_{MSWI} \times NREU_{MSWI} \tag{6.6}$$

Where:
$M_{\text{V-PET}}$: quantity of V-PET in the recycling system (tonne)
$M_{\text{R-PET}}$: quantity of R-PET in the recycling system (tonne)
M_{MSWI} : quantity of total amount of post-consumer PET ended up in a MSWI with energy recovery in the recycling system (tonne)
$NREU_{\text{R-PET}}$: NREU (GJ) of the recycling process per tonne delivered PET (the recycling process includes bottle waste collection, sorting, flake production and pellet production; not including the feedstock energy of PET)

$\because$ $FU = M_{\text{V-PET}} + M_{\text{R-PET}}$, and $M_{MSWI} = FU - (M_{\text{R-PET}}/\eta)$
($\eta = 95\%$, is material efficiency of the recycling process)

Equation (6.2) can be rewritten into:

$$\begin{aligned} E_{recycl} &= (FU - M_{\text{R-PET}}) \times NREU_{\text{V-PET}} + \\ &\quad M_{\text{R-PET}} \times NREU_{\text{R-PET}} + (FU - M_{\text{R-PET}}/\eta) \times NREU_{MSWI} \qquad (6.7) \\ &= FU \times (NREU_{\text{V-PET}} + NREU_{MSWI}) + \\ &\quad M_{\text{R-PET}} \times (NREU_{\text{R-PET}} - NREU_{\text{V-PET}} - NREU_{MSWI}/\eta) \qquad (6.8) \end{aligned}$$

$NREU_{saving}$ = Equation (6.5) − Equation (6.8):

$$E_{ref} - E_{recycl} = M_{\text{R-PET}} \times (NREU_{\text{V-PET}} - NREU_{\text{R-PET}} + NREU_{MSWI}/\eta)$$

$$\begin{aligned} \therefore \quad (E_{ref} - E_{recycl})/M_{\text{R-PET}} &= NRUE_{\text{V-PET}} - NREU_{\text{R-PET}} + NREU_{MSWI}/\eta \\ &= 43.5GJ \end{aligned} \tag{6.9}$$

($NREU_{\text{V-PET}}$=66.6 GJ/t, $NREU_{\text{R-PET}}$=9.5 GJ/t, $NREU_{MSWI}/\eta$=−13 GJ/t, η=95%)

Replace NREU with global warming in Equation (6.9), the GHG-emission saving per tonne R-PET can be calculated at 2.4 t CO_2 eq./t.

Chapter 7

Comparing life cycle energy and GHG emissions of bio-based PET, recycled PET, PLA and man-made cellulosics*

7.1 Introduction

Worldwide PET (polyethylene terephthalate) polymer production in 2007 was 46 Mt (1 Mt = 10^6 metric tonnes), which was approximately 15% of the total synthetic polymer production, making PET the second most important synthetic polymer next to polyolefins [13, 66]. Plastics and fibres are the two most important applications of PET. Approximately 31 Mt (or 67%) of the total amount of PET produced in 2007 was used to make polyester fibre, 12.5 Mt (or 27%) went to plastic bottles, 2 Mt (or 5%) was converted into films and sheets and 0.4 Mt (1%) was used for injection moulded products [13, 66, 8].

In the past decades, bio-based materials have attracted much attention due to the public concerns over limited fossil fuels and climate change. Between 2003 and 2007, the global capacity of bio-based plastics increased from 0.1 Mt to 0.36 Mt [241]. With the large scale production of bio-based ethylene announced in the near future [241], partially bio-based PET can be produced from bio-based ethylene glycol (EG) and petrochemical purified terephthalic acid (PTA). Moreover, PET recycling has experienced steady and fast growth in the past decade along with the growth in PET bottle

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*Submitted to *Environmental Science & Technology.* Co-authors: Ernst Worrell and Martin K. Patel (Utrecht University) consumption and therefore, increasing pressure on bottle waste management. In Europe, the collected PET bottle waste increased from 0.2 Mt in 1998 to 1.3 Mt in 2008 [183], which is approximately over 40% of the total PET bottles consumed.[1] Like virgin PET polymer, recycled PET is mostly used to make fibres and bottles.[2]

The eco-profiles of petrochemical PET have been reported extensively by "PlasticsEurope" [112, 236]. In our previous studies [228, 242], we reported the environmental impacts of recycled PET fibres and bottles made from used PET bottles using various allocation methods for open-loop recycling. The comparison among petrochemical PET, partially bio-based PET (from now onwards "partially bio-based PET" is referred to as "bio-based PET"), recycled PET and recycled bio-based PET has not been made so far.

Moreover, it is of interest to compare the environmental profiles of bio-based and/or recycled PET with other bio-based polymers which can be used for similar purposes (i.e. for fibres and bottles). PLA (polylactic acid, or polylactide) and man-made cellulose fibres are two types of bio-based polymers that are currently produced on large scales. PLA is a polyester made via the fermentation of sugar (sugar is obtained from sugar crops such as sugarcane or starch crops such as maize). In 2007, approximately 0.15 Mt of PLA production capacity was installed worldwide [44]. It is expected that over 0.47 Mt will be installed by 2013 [44]. Like PET, PLA can be used for both fibre- and bottle-making. Man-made cellulose fibre is regenerated cellulose made from wood pulp.[3] The worldwide production in 2007 was approximately 3.5 Mt p.a. [5], representing the third most important fibre following polyester and cotton.

The purpose of this article is to provide an overview of the environmental impacts of petrochemical PET, bio-based and/or recycled PET, and to compare the PET systems with other bio-based product systems, namely PLA and man-made cellulose fibres. We intend to include all publicly available LCA studies to prepare this review. The environmental impacts considered are life cycle non-renewable energy use (NREU) and greenhouse gas (GHG) emissions.

In this review, we focus on man-made polymers (made from both petroleum and biomass). Cotton, a natural polymer and one of the principle fibres in the market, is excluded from this review. Cotton is neither energy intensive nor GHG-emission intensive [243]. Instead, the environmental issues of cotton are mainly related to ecotoxicities caused by herbicides and pesticides, eutrophication caused by fertiliser use and soil salination caused by water consumption [243]. These are relatively less worrying issues for man-made polymers. Furthermore, we limit outselves to the most important types of synthetic fibres and exclude for example, nylon and acrylic due to their comparatively low production volumes.[4]

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[1] According to PlasticsEurope [4], PET consumption (excluding fibre) in 2008 was approximately 3.4 Mt in Western Europe. Approximately 90% was used for bottles [8]. Thus the total amount of PET bottles consumed in 2008 in Western Europe can be estimated at approximately 3.1 Mt.

[2] Over 70% of the recycled PET in 2007 was converted into fibres, 10% went back to bottles, and 20% was used for other applications (e.g. sheets and strapping tape) [182].

[3] In a rare case, cotton linter is used to make cellulose fibres [14, 243].

[4] According to the statistics from [66], worldwide polyester produced in 2008 was five times as much as the total of nylon and acrylic fibres.

7.2 System definition

7.2.1 Functional unit, system boundary and product systems

The typical applications of PET polymer, PLA polymer and man-made cellulose fibres are shown in Table 7.1. In this review, the LCAs of three products are compared: polymer granulate, fibre and bottle. Three functional units are accordingly defined: i) *1 kg of polymer, amorphous grade*; ii) *1 kg of staple fibre*; and iii) *1 kg of bottles.* The system boundary is defined as *cradle to grave excluding the use phase.* Thus the life cycle can be divided into two stages: cradle-to-factory gate and post-consumer waste management. We assume that the products (i.e. polymer, fibre and bottle) are consumed in Western Europe and are disposed of using a municipal solid waste incineration (MSWI) plant with energy recovery, which is a common waste treatment method in Western Europe.[5]

Table 7.1: Typical applications of PET, PLA and man-made cellulose fibres.

Sector	PET [8, 66]	PLA [241]	Man-made cellulose fibres
Packaging	32 %	70%	
Injection moulding	1%	1%	
Agriculture		1%	
Textiles and nonwovens	67%	28%	100%

Based on the functional unit and system boundary, we describe the product systems as follows:

1. *Petrochemical PET*: PET products that are made from petroleum feedstock, used once and disposed of in a MSWI facility with energy recovery. This is the reference system.

2. *Bio-based PET*: PET products that are made from bio-based EG and petrochemical PTA. Bio-based EG is obtained from bio-based ethylene which is the dehydration product of bio-based ethanol. We assume that the bio-based ethanol is 50% sugarcane-based and 50% maize-based, approximately representing the share of the two biggest producers of bio-based ethanol, the US and Brazil,[6] which use maize and sugarcane as the feedstocks [235].

3. *Recycled PET*: Petrochemical PET bottles that are used once, recycled into PET polymer, fibre or bottles, used again, and disposed of in a MSWI plant with energy

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[5] Landfilling is also a common waste management option. Almost 50% of the plastics waste were still landfilled in the EU in 2008, approximately 21% was recycled (including both mechanical recycling and feedstock recycling) and 30% was incinerated with energy recovery [4]. However. according to the currently EU policy, landfilling will be soon phased out [178, 27]. In many EU countries, such as the Netherlands, Germany and Switzerland, landfilling of combustable waste is strictly prohibited.

[6] The production of the two countries accounts for nearly 90% (the US 55% and Brazil 35%) of the world bio-ethanol production in 2009 [235].

recovery. Recycled granulates (polymer) and fibres are made from 100% recycled pellets,[7] while recycled bottles consist of 35% recycled pellets and 65% virgin pellets.[8]

4. *Recycled bio-based PET*: Bio-based PET bottles that are used once, recycled into PET polymer, fibre or bottles, used again, and disposed of in a MSWI plant with energy recovery. Recycled granulates (polymer) and fibres are made from 100% recycled pellets (see footnote 7). Recycled bottles contain 35% recycled PET and 65% virgin PET (see footnote 8).
5. *PLA*: PLA products contain 50% maize-derived PLA produced in the US (i.e. by NatureWorks LLC) and 50% sugarcane-derived PLA produced in Thailand (i.e. by PURAC), used once, and disposed of in a MSWI plant with energy recovery.
6. *Viscose*: Viscose fibre that is produced from wood pulp in an integrated plant and a non-integrated plant, used once and disposed of in a MSWI plant with energy recovery.[9]
7. *Modal*: Modal fibre that is produced from wood pulp in an integrated plant, used once and disposed of in an average MSWI plant with energy recovery.
8. *Tencel*: Tencel fibre that is produced from wood pulp, used once and disposed of in an average MSWI plant with energy recovery. Both the state-of-the-art Tencel (referred to as “Tencel Austria” in [243]) and the future Tencel (referred to as “Tencel Austria 2012” in [243]) are included.

In this comparison we focus on two environmental impact indicators, namely NREU and GHG emissions. NREU is the cumulative non-renewable primary energy demand, including fossil fuels and nuclear energy [167]. Cumulative fossil fuel demand is a good proxy of the overall environmental profile of a product [106]. GHG emission indicates the impact on climate change. In this article, we compare GHG emissions calculated based on the global warming potential for a period of 100 years [171]. For bio-based

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[7] The exception is the recycled products assessed by the “system expansion” method, where the output (i.e. polymer or fibre) consists 90% recycled and 10% virgin polymer which is for the “make-up” purpose. The “90%/10%” ratio is determined by the PET material flow efficiency of the recycling process [242]: the more efficient the recycling process is, the more recycled polymer is delivered by the whole system. The scheme of applying the “system expansion” method can be found in Figures 12 and 13 in [228].

[8] In the current commercial practice, the maximum fraction of recycled pellets used in a recycled PET bottle is 35% [242, 229]. This ratio is the practical maximum value because the discoloration effect at this composition is acceptable for commercial use [232, 229].

[9] The differences between an integrated and a non-integrated plant can be described as the following: in an integrated pulp/fibre plant, the process energy and material efficiencies are highly optimised. As a result, recovered bio-energy such as thick liquor is used to fuel the process and more by-products are recovered (e.g. xylose, furfural and acetic acid). In a non-integrated production (or a “separate production”, as described in [243]), the pulp mill and the fibre plant are separately located. The process energy use of the pulp mill is supplied partially by the recovered biomass energy (thick liquor) and partially by fossil fuels. The process energy of the fibre plant is entirely supplied by local grid power and fossil fuels. A non-integrated production does not have by-products such as xylose, furfural and acetic acid. More detailed description can be found in [243].

materials (bio-based PET, PLA and man-made cellulose fibres), the biogenic carbon embedded in the product is taken into account as negative GHG emissions from cradle to factory gate [151]. Consequently, in the "grave" stage, the biogenic CO_2 released from the combustion of the product is added to the cradle-to-grave GHG emissions. Moreover, the LCA study of man-made cellulose fibres [243] reported that no direct land use change (LUC) occurred during the wood production after to 1 January 1990.[10] No direct LUCs were taken into account in the studies reviewed for bio-based PET and PLA [224, 239, 244, 245, 246]. None of the LCA studies reviewed took into account indirect LUC.

7.2.2 LCAs reviewed in this paper

The LCA data were obtained from peer-reviewed journal papers, scientific reports, life cycle inventory databases and personal communications with industry experts. Table 7.2 shows the summary of the LCAs reviewed and the data and assumptions we used for our calculations.

Based on the literature review, three allocation methods were applied for the two recycled PET systems, i.e. *Recycled PET* and *Recycled bio-based PET*. The three methods which were described in detail in [228] are:

- The "cut-off" method - the product system of the recycled product starts from waste collection (here chosen as the "cradle"). The production and use of the virgin product is outside the system boundary of the recycled product. Following this principle, the environmental burden of the final disposal of the material (the ultimate "grave") is entirely assigned to the recycled product.
- The "waste valuation" method - the environmental burden of virgin polymer production is shared between the two lives (i.e. virgin and recycled products). The environmental burden of the "grave" stage is also shared between the two lives. The economic values of virgin and recycled products are used to determine the allocation factors. In this review, the only study used this method is the LCA of recycled PET fibres [228].
- The "system expansion" method - this method applies the "system expansion" principle (see ISO14044:2006 Clause 4.3.4.2 [147]), taking into account the entire life cycle and not allocating any environmental burden between the first and the second lives (see more in [228, 242]).

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[10] According to PAS 2050 [151], only the GHG emissions arising from direct land use change occurring after 1 January 1990 should be taken into account.

Table 7.2: Summary of data used in this study.

	Parameters	**Value**	**Unit**	**Source and notes**
A	Petrochemical PET (amorphous), cradle-to-factory gate			PlasticsEurope's new eco-profile of PET [236] is used as the default value. The former eco-profiles of PET (in parentheses) [112] are used for comparison. Production technology in Western Europe in the late 2000s.
	NREU	67 (81)	MJ/kg	
	GWP100a	2.05 (3.30)	kg CO_2 eq./kg	
B	Bio-based PET, cradle-to-factory gate amorphous grade			Based on 50% sugarcane-derived ethanol from Brazil and 50% maize-derived ethanol from the US [239].[a] Production technology in the mid-2000s in the US and Brazil. LUCs are not taken into account for the sugarcane and maize production.
	NREU (maize-based)	59	MJ/kg	
	GWP100a (maize-based)	1.36	kg CO_2 eq./kg	
	NREU (sugarcane-based)	51	MJ/kg	
	GWP 100a (sugarcane-based)	1.03	kg CO_2 eq./kg	
C	PLA, cradle-to-factory gate amorphous grade			Site specific technology in the late 2000s. LUCs are not taken into account for the maize and sugarcane production.
	NREU (ingeo 2009)	42	MJ/kg	NatureWorks LLC (maize-based, from the US) [224, 245].
	GWP (ingeo 2009)	1.30	kg CO_2 eq./kg	
	NREU (PURAC PLLA)	31	MJ/kg	PURAC (sugarcane-based, from Thailand) [246].
	GWP (PURAC PLLA)	0.50	kg CO_2 eq./kg	
D	Energy use of SSP (solid state polymerisation)	0.15	kWh/kg	Based on [112, 205]. Assume the process energy of polymer conversion is the same for PLA, PET and recycled PET.
E	Energy use of blow moulding process	2.1	kWh/kg	Based on [205, 247]. Assumption of the same energy use for both PLA and PET polymers.
F	Energy use of the fibre extrusion process, converting amorphous PET to 1 kg fibre	0.64 5	kWh/kg MJ heat/kg	Based on [164]. Data was cross-checked with industry experts. Assume the process energy of polymer conversion is the same for PLA, PET and recycled PET.
G	Cradle-to-factory gate, man-made cellulose fibres			Based on [243]. Average technology level of the mid-2000s. No land use changes occurred in the wood production.
	NREU, Viscose, integrated	19	MJ/kg	As "Viscose Austria" in [243].
	GWP, Viscose, integrated	−0.25	kg CO_2 eq./kg	
	NREU, Viscose, non-integrated	61	MJ/kg	As "Viscose Asia" in [243].
	GWP, Viscose, non-integrated	3.81	kg CO_2 eq./kg	
	NREU, Modal	25	MJ/kg	As "Modal" in [243].
	GWP, Modal	0.03	kg CO_2 eq./kg	
	NREU, Tencel (current)	42	MJ/kg	As "Tencel Austria" in [243].
	GWP, Tencel (current)	1.11	kg CO_2 eq./kg	
	NREU, Tencel (future)	21	MJ/kg	As "Tencel Austria 2012" in [243].
	GWP, Tencel (future)	0.05	kg CO_2 eq./kg	

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Table 7.2 – continued from previous page

	Parameters	Value	Unit	Source and notes
H	Cradle-to-factory gate recycled PET polymer (amorphous granulate)			
	NREU, cut-off	9.5	MJ/kg	Reproduced based on data from [242]. Same values for both *Recycled PET* and *Recycled bio-based PET*.
	GWP, cut-off	1.01	kg CO_2 eq./kg	
I	Cradle-to-factory gate recycled PET fibre			
	NREU, cut-off	22	MJ/kg	Calculated based on [242, 164]. Same values for both *Recycled PET* and *Recycled bio-based PET*.
	GWP, cut-off	1.70	kg CO_2 eq./kg	
	NREU, cut-off	13-23	MJ/kg	Data ranges reported by [228] based on the production of two recycling companies.
	GWP, cut-off	0.96-1.88	kg CO_2 eq./kg	
	NREU, waste valuation	40-49	MJ/kg	
	GWP, waste valuation	2.03-2.95	kg CO_2 eq./kg	
J	Cradle-to-factory gate recycled PET bottle (with 35% recycled content)			
	NREU, cut-off, petrochem. virgin PET	70	MJ/kg	Calculated from A, D, E and H.
	GWP, cut-off, petrochem. virgin PET	3.08	kg CO_2 eq./kg	
	NREU, cut-off, bio-based virgin PET	62	MJ/kg	Calculated from B, D, E and H.
	GWP, cut-off, bio-based virgin PET	2.53	kg CO_2 eq./kg	
K	Cradle to grave (without use phase) recycled PET polymer, fibre and bottle, applying system expansion			
	NREU, recycled petrochemical polymer	15	MJ/kg	Calculated based on data from A, D, E and [228, 242].
	GWP, recycled petrochemical polymer	1.11	kg CO_2 eq./kg	
	NREU, recycled bio-based polymer	14	MJ/kg	Calculated based on data from B, D, E and [228, 242].
	GWP, recycled bio-based polymer	1.03	kg CO_2 eq./kg	
	NREU, recycled petrochem. fibre	23-33	MJ/kg	Data ranges reported by [228] based on the production of two recycling companies.
	GWP, recycled petrochem. fibre	1.33-2.21	kg CO_2 eq./kg	
	NREU, recycled petrochem. fibre	28	MJ/kg	Calculated based on data from A, F and [228, 242].
	GWP, recycled petrochem. fibre	1.80	kg CO_2 eq./kg	
	NREU, recycled bio-based fibre	27	MJ/kg	Calculated based on data from B, F and [228, 242].
	GWP, recycled bio-based fibre	1.72	kg CO_2 eq./kg	
	NREU, recycled petrochem. bottle	66	MJ/kg	Calculated based on data from A, D, E and [228, 242].
	GWP, recycled petrochem. bottle	4.13	kg CO_2 eq./kg	
	NREU, recycled bio-based. bottle	57	MJ/kg	Calculated based on data from B, D, E and [228, 242].
	GWP, recycled bio-based. bottle	3.49	kg CO_2 eq./kg	
L	MSWI with energy recovery			
	Energy recovery rate (in primary energy terms)	~60%	-	Calculated based on the efficiencies of electricity and heat of 10.6% and 22.3% in an average MSWI plant in Europe according to Reimann [162].[b]
M	Gross calorific values of polymers			
	PET	23.1	MJ/kg	[202]
	PLA	19	MJ/kg	[114]

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Table 7.2 – continued from previous page

	Parameters	Value	Unit	Source and notes
	Man-made cellulose fibres	˜15	MJ/kg	[243]
N	Electricity and heat production	Assumed to be the average EU electricity mix[c] and average European heat from industrial furnance (>100 kW) with low NOx. LCA data obtained from the Ecoinvent database Version 2.0 [196].		

[a] A recent publication from Tabone and colleagues [248] applied both LCA and the Green design principles to 12 polymers, among which there is (partially) bio-based PET. This study was heavily criticised for its scope of comparison, its LCA allocation method and its use of the single score approach [249]. We carefully reviewed this article. We appreciate the concept of comparing the Green Design Principles with LCA. However, we consider the LCA results for bio-based PET unreliable and therefore they are not included in this review.

[b] This means that 1 GJ of waste yields 0.106 GJ_e (electricity) and 0.223 GJ_{th} (thermal). These amounts of electricity and heat would be otherwise produced conventionally with a cradle-to-factory gate electricity efficiency of 30% and a heat efficiency of 85% (approximately). Thus, 0.106 GJ_e replaces 0.106/30% = 0.35 GJ_p primary fossil fuels and 0.223 GJ_{th} replaces 0.223/85% = 0.26 GJ_p fossil fuels. The total primary fossil fuel that can be avoided is 0.35 GJ_p + 0.26 GJ_p = 0.61GJ_p – this is approximately 60% of the energy content of the waste.

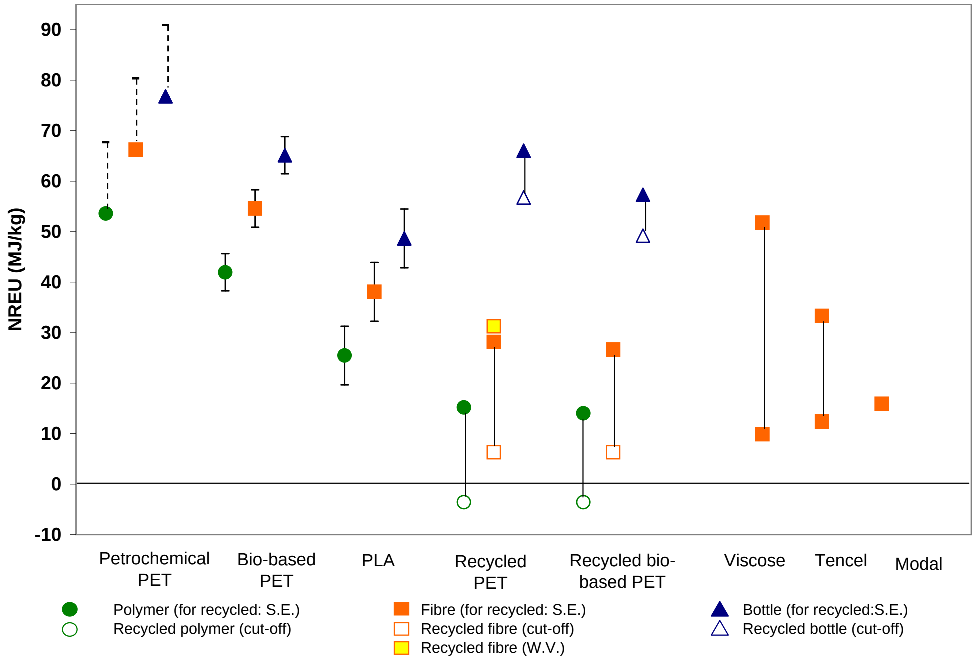
[c] European electricity mix: 65% from the UCTE grid, 13% from the NORDEL grid, 9% from the CENTREL grid, 12% from the UK grid and 1% from the Irish grid. UCET is Union for the Co-ordination of Transmission of Electricity; countries included in UCTE are Austria, Bosnia and Herzegovina, Belgium, Switzerland, Germany, Spain, France, Greece, Croatia, Italy, Luxemburg, Macedonia, the Netherlands, Portugal, Slovenia and Serbia and Montenegro. NORDEL is Nordic countries power association, including Denmark, Norway, Finland and Sweden. CENTREL stands for Central European power association, including Czech Republic, Hungary, Poland and Slovakia.

7.3 Results

7.3.1 The ranges

Figure 7.1 and Figure 7.2 show the results of cradle-to-grave (without the use phase) NREU and GHG emissions for the eight product systems. The results of *Petrochemical PET* are based on the new eco-profile of PET; the dashed upper range represents the results based on the previous eco-profiles of PET. The eco-profile of petrochemical PET has been substantially improved over the last years in Western Europe. Based on the new eco-profile, the cradle-to-factory gate NREU and GHG emissions of amorphous PET have been reduced by 17% and 38% (see also in Table 7.2), respectively, compared to the previous eco-profile of PET [112]. The difference between the the old and new eco-profiles originates from the improved PTA process [236].

The ranges of *Bio-based PET* are comparatively small in both figures. Bio-based PET has approximately one-third bio-based content (EG, on mass basis) and two-third petrochemical content (PTA). The lower values refer to sugarcane-based PET and the higher values refer to maize-based PET [239]. For PTA, we used the new eco-profile [236] in the calculation and the reported ranges do not include the previous eco-profile of PTA. The inventory data on the production of sugarcane-based ethanol were obtained from 40 plants in Brazil [239, 244]; LCA data on maize-based ethanol were based on the assumption that the bio-ethanol complies with the requirements of



The uncertainty ranges are explained in the text. S.E. stands for the "system expansion" method. W.V. stands for the " waste valuation" method. See the description of the methods applied for open-loop recycling in Section 7.2.2.

Figure 7.1: Comparison of cradle-to-grave NREU, "grave" is MSWI with energy recovery, the use phase is excluded.

the EU policy.[11] For *PLA*, the lower values are sugarcane-based (from PURAC) and the higher values are maize-based (from NatureWorks).

For *Viscose* fibre, the ranges are large in both Figures 7.1 and 7.2. The lower values are the LCA results of "Viscose Austria" in [243] and the higher values are the LCA results of "Viscose Asia" in [243]. "Viscose Austria" is produced in the integrated pulp/fibre plant in Austria; "Viscose Asia" is produced in a non-integrated fibre production in Asia. For *Tencel* fibres, the higher values refer to the product "Tencel Austria" in [243], which represents state-of-the-art Tencel production; the lower values represent the LCA results of "Tencel Austria 2012" in [243], which uses the energy recovered from municipal waste incineration for process heat and power supply. *Modal* fibre is exclusively produced in the integrated pulp/fibre plant in Austria. No ranges were calculated in earlier studies.

For the recycled systems, namely *Recycled PET* and *Recycled bio-based PET*, wherever virgin PET is involved, the new eco-profile of PET is used for the calculation (see

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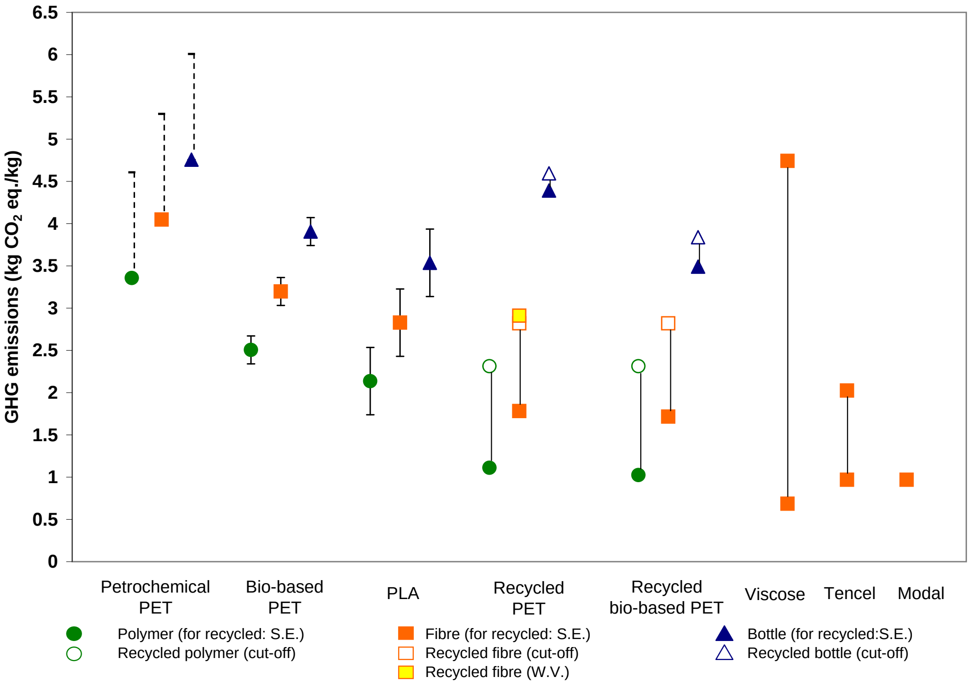
[11] According to EU Directive 2009/28/EC [194], 35% of GHG emission savings should be achieved for bio-ethanol comopared to petrol. According to [239], currently for the maize-based ethanol production, only the best-available-technology (BAT) meets the requirements of the Directive.



The uncertainty ranges are explained in the text. S.E. stands for the "system expansion" method. W.V. stands for the " waste valuation" method. See the description of the methods applied for open-loop recycling in Section 7.2.2.

Figure 7.2: Comparison of cradle-to-grave GHG emissions, "grave" is MSWI with energy recovery, the use phase is excluded.

also Table 7.2). The ranges reported here are large for polymers and fibres (not for bottles, see the next paragraph). The large ranges originate primarily from the different allocation methods used for open-loop recycling (see Section 7.2.2).

The value ranges of the recycled bottles (for both NREU and GHG emissions) are small compared to the value ranges of recycled polymer granulates and fibres. Because a recycled bottle contains only 35% recycled polymer and the remaining 65% is virgin polymer (petrochemical or bio-based) (see Section 7.2.1). The overall impact of recycled PET bottle is greatly influenced by the production of the virgin polymer. As a consequence, the (expected) impact reduction from recycling is strongly undermined.

7.3.2 The comparison

Across all types of functional units the rankings for polymer granulates, fibres and bottles are identical in both Figures 7.1 and 7.2: the impacts are the lowest for polymers, followed by fibres and finally bottles.

In Figure 7.1, the NREUs of polymer and fibre are the highest for *Petrochemical*

PET, followed by *Bio-based PET*, *PLA*, *Recycled PET* and *Recycled bio-based PET*. The ranking is not affected by the applied allocation method; and can hence be considered as robust. When the "cut-off" method is applied, fibres made from *Recycled PET* and *Recycled bio-based PET* both have lower NREU than integrated produced Viscose fibre (the lower value for *Viscose*). When the "system expansion" or "waste valuation" method is chosen, the two recycled fibres have higher NREU than the two man-made cellulose fibres produced in the integrated production (*Modal* and the lower value for *Viscose*) and the future Tencel fibre (the lower value for *Tencel*). If produced from a non-integrated plant, man-made cellulose fibre is comparable with *Bio-based PET* (comparing the higher value for *Viscose* with *Bio-based PET*), or *Recycled PET* (comparing the higher value for Tencel with the highest value for *Recycled PET*). The NREU of <u>bottle</u> is highest for *Petrochemical PET*, followed by *Bio-based PET* and *PLA*. The NREUs of the two recycled bottles are influence by the choice of the allocation method. If the "cut-off" method is applied, *Recycled bio-based PET* bottles are comparable with *PLA* bottles; and *Recycled PET* bottles have a similar NREU as *Bio-based* PET bottles.

Like the results of NREU, in Figure 7.2 the GHG emissions of <u>polymer</u> and <u>fibre</u> are the highest for *Petrochemical PET*, followed by *Bio-based PET*, *PLA*, *Recycled PET* and *Recycled bio-based PET*. *Viscose* fibre produced via integrated production have the lowest GHG emissions among all fibres. *Viscose* fibre produced via a non-integrated production has the highest GHG emissions of all. The GHG emissions of 1 kg of <u>bottles</u> are the highest for *Petrochemical bottles*, followed by *Recycled PET* and *Bio-based PET*. The GHG emissions of *Recycled bio-based PET* and *PLA* bottles are the lowest of all bottles studied. When the "system expansion" method is applied, *PLA* and *Recycled bio-based PET* are comparable. When the "cut-off" method is applied, *PLA* bottles are more favourable than bottles made from *Recycled bio-based PET*.

Compare *Reccled PET* and *Recycled bio-based PET* for <u>polymers</u> and <u>fibres</u>, the NREU and GHG emissions of the two systems are identical based on the "cut-off" method: the environmental impacts were contributed by the recycling process and post-consumer MSWI; the first life (virgin PET production) has no influence on the second life. If the "system expansion" method is applied, the differences between *Recycled bio-based PET* and *Recycled PET* are subtle. Based on the "system expansion" method described in [228, 242], the recycled system delivers 0.9 kg of recycled PET (determined by the material efficiency of the PET flow is 90% [242], see also footnote 7) and the remaining 0.1 kg of PET needs to be "made up" from virgin PET production (which is either petrochemical or bio-based PET). The minor differences between the two recycled systems are caused by the differences of the 0.1 kg of virgin PET (petrochemical vs bio-based).

7.3.3 The allocation methods

In terms of NREU (see Figure 7.1), the "cut-off" approach leads to lower NREU because of the energy credits from MSWI. Both the "waste valuation" and "system expansion" methods lead to relatively high NREU because less end-of-life waste is incinerated in the recycled product systems and therefore, less energy credits are obtained. In contrast, in terms of GHG emissions (see Figure 7.2), the "system expansion" method leads to lower GHG emissions due to less waste incineration; the "cut-off" method leads to higher GHG emissions because of the net GHG emission penalties from MSWI (even with the emission credits). The "waste valuation" method leads to slightly higher results for GHG emissions than the "cut-off" method.

7.4 Discussion

The results indicate that both recycling and bio-based alternatives are important ways to reduce the environmental footprint of material consumption. However, not all functionalities of the materials studied are strictly comparable, although same products can be made from various materials.

Firstly, PLA has different material properties compared to PET. For example, the gas-barrier property (e.g. for CO_2) of PLA is much weaker than that of PET, which determines that PLA bottles can only be used for non-carbonate drinks and have limited shelf time.

Secondly, recycled products have a limited range of applications compared to the virgin ones. For instance, recycled PET fibre cannot be used to produce high performance textile products (e.g. with moisture management) [228]. Another restriction of recycling is that fibre (both virgin and recycled) cannot be further recycled via mechanical recycling [228]. A more comprehensive discussion regarding the functional equivalence between recycled and virgin PET can be found in [228]. For PLA recycling, there are still technological difficulties which need to be resolved, e.g. polymer degradation during mechanical recycling. Although chemical recycling of PLA is technically feasible [250], it has not been realised on an industrial scale so far.

Thirdly, man-made cellulose fibres have different properties compared to polyester fibres (PET and PLA) in many aspects, e.g. density, dry tenacity, wet tenacity, water retention, dyeability, etc. [243], which lead to different applications in the textile/nonwoven sector.

Finally, although petrochemical PET has higher environmental impacts compared to recycled PET and PLA, it does offer the widest range of applications that neither recycled PET nor PLA can completely cover using the state-of-the-art technology. In this sense, bio-based PET and petrochemical PET are truly functionally equivalent.

In addition, the reviewed LCAs of man-made cellulose fibres reported that no direct LUCs (land use changes) occurred during the wood production and the LCAs of bio-based PET and PLA do not report any LUCs. If either direct or indicrect LUC occurs in the biomass production, the GHG emissions of the bio-based materials will increase.[12]

In this article, we intend to review as many as possible man-made alternatives for plastics and fibre materials, although it is not possible to cover all possibilities. For instance, multiple recycling trips can potentially further reduce the environmental

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[12] In industrialised countries, direct LUC occurring after 1 January 1990 is unlikely. However, the impact of indirect LUC will increase the GHG emissions of bio-based products. Indirect LUC is not included in the PAS 2050 guidelines because the method and data required for the assessment are not fully developed [151].

impact by maximally six percentages [242]. Also, the PET bottle-to-bottle recycling technology is still under development. It can be calculated that if 100% of recycled PET pellet is allowed in a recycled bottle instead of 35%, the impacts (both NREU and GHG emissions) of recycled PET bottle will further decrease by 8% and 12% for *Recycled Bio-based PET* and *Recycled PET*, respectively. Last but not least, novel bio-based polymers such as PLA and bio-based PET are still in their early stages of commercialisation. Efforts have been, and will continue being made to improve the material properties of PLA and recycled PET. In the meantime, other bio-based feedstocks and routes are being investigated, such as cellulosic feedstocks [69, 251] for PLA and furanics feedstocks for PET [252]. These new feedstocks and routes may further decrease the environmental footprint of bio-based materials, making them more competitive with recycled petrochemical polymers.

7.5 Conclusions and recommendations

In this paper, we reviewed the *cradle-to-grave (without the use phase)* NREU and GHG emissions of 1 kg of polymer, fibre and bottles for four PET product systems (i.e. petrochemical PET, bio-based PET, recycled PET and recycled bio-based PET) and two groups of bio-based materials (i.e. PLA and man-made cellulose fibres). Based on the findings, the following conclusions are drawn.

The NREU and GHG emissions of polymers and fibres are the highest for *Petrochemical PET*, followed by *Bio-based PET*, *PLA*, *Recycled PET* and *Recycled bio-based PET*. Man-made cellulose fibres produced via the integrated production has the lowest GHG emissions of all fibres studied. The NREU and GHG emissions of bottles are the highest for *Petrochemical PET*. Bottles made from *Bio-based PET* have higher impacts than bottles made from *PLA*. Bottles made from *Recycled PET* have higher impacts than bottles made from *Recycled bio-based PET*. These results are not affected by the choice of the allocation method. Hence, these conclusions are considered robust.

However, the rankings for bottles made from *PLA*, *Bio-based PET*, *Recycled PET* and *Recycled bio-based PET* are strongly depending on the choice of the allocation method applied to open-loop recycling. Compared to the“system expansion” method, the “cut-off” approach leads to relatively low NREU but high GHG emissions. The “waste valuation” leads to relatively high results of both NREU and GHG emissions. The wide range of results due to the choice of the allocation method do not cause problems when comparing recycled product systems with the virgin petrochemical product system. However, when the recycled products are compared with other renewable alternatives, the ranking becomes less straightforward.

So far in the LCA community, there is no uniform standards on how the environmental impact should be assessed for open-loop recycling. In our previous study [228], the three allocation methods are applied and evaluated. In our opinion, the “system expansion” method is the preferred choice because it implements life-cycle thinking. With the increasing amount of recycled products in our daily life, we recommend that the LCA community and policy makers should establish clear rules and procedures in order to support the decision making for recycled products.

For bio-based polymer producers, we recommend that efforts should be made on the process optimisation and the utilisation of biomass feedstock for process energy (e.g. the biorefinery concept). The LCA results of man-made cellulose fibres show that process integration substantially reduces environmental impact. It is also very important to continue improving the material properties of the novel bio-based polymers (e.g. PLA). If the bio-based polymers are able to cover a wide range of applications and if recycling is implemented as part of their waste management, a significant impact reduction on our material consumption will be achieved in the future.

Chapter 8

Summary and Conclusions

Today, almost all man-made plastics and fibres are produced from synthetic polymers. Synthetic polymers, made from petroleum which took millions of years to form, are facing three sustainability challenges: (i) the limited fossil fuel resources, (ii) the environmental impacts caused by non-degradable plastics waste, and (iii) greenhouse gas emissions caused by combusting fossil fuels. To tackle these sustainability challenges, two strategies have been proposed. First, use bio-based polymers to replace the traditional petrochemical polymers. Second, increase the overall material utilisation efficiency by means of recycling. Plastics and fibres are the two most important applications of synthetic polymers. The objective of this thesis is to assess the environmental impacts of plastics and fibres made from bio-based and recycled materials and to compare them with the conventional counterparts.

The first research question of this thesis is: "*what is the current status and what are the development potentials of the bio-based plastics industry?*" In the past decades we witnessed the commercialisation of several novel bio-based plastics, such as PLA (polylactic acid), PHAs (polyhydroxyalkanoates), starch plastics, bio-based PTT (polytrimethylene terephthalate) and bio-based PE (polyethylene). However, there is so far no thorough review of the current market volume and development status of this emerging industry. Chapter 2 provides an overview of the current global market of bio-based plastics, their material properties, technical substitution potential and future market (for 2020). About 70 companies worldwide were included in our survey.

The historical use of natural feedstocks for plastics production demonstrates that bio-based products are neither fictional nor new. Instead, bio-based products have been an industrial reality on a million-tonne scale (e.g. paper and board) for many decades. Today, the combined volume of non-food and non-plastics applications of starch and man-made cellulose fibres is 55 times larger than the total of all emerging bio-based plastics (approx. 20 Mt versus approx. 0.36 Mt in 2007).

Between 2003 and 2007, the annual growth rate of the emerging bio-based plastics was nearly 40%, resulting in a global capacity of 0.36 Mt in 2007. The global production of bio-based plastics is likely to continue growing strongly and reach 2.3 Mt in 2013 and 3.5 Mt in 2020. Starch plastics, PLA, bio-based PE, PHAs and bio-based epoxy resin are expected to be the major types of bio-based plastics in the future. The maximum technical substitution potential of bio-based plastics (including man-made fibres) replacing their petrochemical counterparts is estimated at 240 Mt, or 90% of the total amount of plastics and fibres based on the 2007 market demand. Based exclusively on the technical feasibility (and disregarding economic and other aspects), the growth potential of bio-based plastics is hence enormous.

Chapter 2 concludes that several factors clearly speak for bio-based plastics. These are the limited and uncertain supply of fossil fuels (e.g. oil and gas), economic viability of biomass feedstock, environmental considerations (e.g. savings of non-renewable energy and greenhouse gas abatement), innovation offering new opportunities (technical, employment, etc.) and rejuvenation in all steps from chemical research to the final product and waste management. Challenges that need to be successfully addressed in the next years and decades are the low performance of some bio-based plastics (e.g. thermoplastic starch), their relatively high cost for production and processing and the need to minimise agricultural land use and forest land use, in order to avoid competition with food production and to minimise adverse effects on biodiversity and other environmental impacts.

Following the first research question, we posed our second research question: "*What are the environmental impacts of bio-based polymers?*" This research question is answered by Chapter 3 and Chapter 4. The purpose of Chapter 3 is to obtain insight into the environmental impacts of polysaccharide products in comparison to their counterparts, which are either petrochemical products or conventional polysaccharides. Polysaccharides are among the most important renewable resources for mankind. They have been widely used for a long time for food (e.g. starch), clothing (e.g. cotton, flax, and jute), communication (e.g. paper), packaging (e.g. paper and board), and construction (wood). Next to these traditional usages, other non-food products have been developed to partly replace conventional products which are either based on non-renewable resources or based on traditional polysaccharide materials. In Chapter 3 we review published LCA studies of polysaccharide-based textile products (e.g. viscose), natural fibre composites, and thermoplastic starch. In the review we choose non-renewable energy use (NREU) and greenhouse gas (GHG) emissions as important indicators for environmental profiles of products.

Chapter 3 concludes that polysaccharides can offer important potentials for NREU savings and GHG emission reduction from cradle to factory gate for one kg material as functional unit. Man-made cellulose fibres can save about 10-30% NREU relative to cotton and up to 50-80% NREU relative to PET. For engineering materials, natural fibre composites can save about 25-30% NREU and reduce up to 40% GHG emissions compared to glass fibre composites. Also, the higher the fibre content in natural fibre composites, the lower NREU and GHG emissions are. For packaging materials, thermal plastic starch (TPS) can save about 25-75% NREU and reduce 20-70% GHG emissions compared to virgin petrochemical polymers (±15% depending on whether HDPE, LDPE or LLDPE is the reference).

Taking into account also the use phase and the end-of-life waste management phase, it is not possible to conclude that polysaccharide-based products are always better than their petrochemical counterparts, but important advantages do exist. The overall conclusion can be drawn that from cradle to factory gate, in terms of non-renewable energy requirements and GHG emissions, the polysaccharide products are better than their conventional counterparts, which are mostly petrochemical products. Up-to-date information on man-made cellulose textiles was not available, which leads to Chapter 4 of this thesis.

In Chapter 4, the environmental impact of man-made cellulose fibres is assessed. Life cycle assessment was conducted for three types of fibres (i.e. Viscose, Modal and Tencel) produced by Lenzing AG. Man-made cellulose fibres are one of the three principle fibres in the world (the other two are cotton and polyester). In this LCA, the functional unit is one tonne of staple fibre. Five man-made cellulose fibre products, namely, Viscose (Asia), Viscose (Austria), Modal, Tencel and Tencel (2012), were investigated. The system boundary is cradle to factory gate. We compared the LCA results with other commodity fibres, namely cotton, PET and PP. Cumulative energy demand (CED, which is the total of non-renewable energy use and renewable energy use), water use, land use and the CML baseline impact categories were assessed. Sensitivity analyses were carried out to understand the influence of different allocation methods. In addition, three single score methods (Single Score I, II and III) were introduced and applied in order to draw an overall conclusion.

Based on the LCA results, we conclude that all man-made cellulose fibres, except for Viscose (Asia), have better environmental profiles than PET, PP and cotton; Tencel (2012) has the lowest impact of all. Viscose (Asia) has a lower impact than cotton; it is comparable to PET, but less preferable than PP and other man-made cellulose fibres. The environmental benefits of Viscose (Austria) and Modal are largely attributed to low fossil energy requirements in the pulp and fibre production. This is a result of process integration, the use of renewable energy and credits from by-products. Furthermore, Viscose (Austria) and Modal have much lower process emissions (e.g. SO_2 and NOx) compared to Viscose (Asia), leading to low human toxicity, photochemical oxidant formation, acidification and eutrophication. The environmental benefits of Tencel (2012) are the result of low energy consumption, low chemical use, low CO_2 emission, low SO_2 emission and low water consumption, leading to low impacts regarding abiotic depletion, terrestrial ecotoxicity, photochemical oxidant formation and acidification. Cotton is not an energy-intensive product; it has the lowest CED of all fibres studied. However, cotton is ranked as the least favourable choice by Single score II and III. The major environmental issues of cotton include land use, water use, fresh water aquatic ecotoxicity, terrestrial ecotoxicity and eutrophication. The use of pesticides in cotton cultivation causes major ecotoxicity impacts. Furthermore, the use of fertilizer results in eutrophication. In the sensitivity analysis, two alternative allocation methods are applied, i.e. allocations based on calorific values and economic values. Both lead to less favourable results for man-made cellulose fibres compared to the default method, although the ranking of all fibres studied does not change. In addition, based on the system of cradle to factory gate plus waste incineration with and without energy recovery, all man-made cellulose fibres studied are better than PET and PP.

State-of-the-art LCA impact assessment methodologies have been applied in this study. However, the quality of toxicity calculations in LCA tools is currently still doubtful and research is underway to improve the methodologies and to make the databases more complete. Furthermore, only environmental indicators that were generally considered in LCA studies have been taken into account. For example, an environmental indicator which has not been considered is the impact on biodiversity. Land use and water use have been exclusively reported as inventory results, i.e. different types of land (and water) have not been aggregated due to the lack of suitable methods. The risk of explosion has neither been taken into account. Disregarding these uncertainties we conclude that modern man-made cellulose fibres have a clear potential to reduce the environmental impacts compared to cotton and synthetic fibres.

In Chapters 5 and 6 we shift our focus from bio-based materials to recycling. With these two chapters, we aim to answer the third research question: "*What is the environmental impact of products made from recycled PET polymer?*" Parallel to the fast development of bio-based materials, the growth in plastic recycling industry has been considerable in the past decade. Recycling of post-consumer PET bottles has become a well-established system with its own logistic chain including bottles collection, flake production and pellet production. The topic is also of interest because in LCA there has been so far no standardised procedure for open-loop recycling. There are hardly any case studies available for open-loop recycling, although the methodological discussion has been extensively published.

The first purpose of Chapter 5 is to understand the environmental impacts of recycled PET fibre compared to virgin PET fibre. The second purpose is to apply different allocation methods for this open-loop-recycling case. In this LCA, we investigated four recycling technologies, namely mechanical recycling, semi-mechanical recycling, back-to-oligomer recycling and back-to-monomer recycling. The LCA results were compared with the eco-profile of virgin PET fibre. Three methods were applied for this open-loop recycling case, namely, the "cut-off", "waste valuation" and "system expansion" methods. The "cut-off" and the "waste valuation" methods assess the system of cradle to factory gate, with bottle waste as the "cradle". The "system expansion" method is applied to assess the system of cradle to grave. The use phase is excluded in this LCA. Nine environmental impact indicators were analysed, i.e. NREU, global warming potential (GWP), abiotic depletion, acidification, eutrophication, human toxicity, fresh water aquatic ecotoxicity, terrestrial ecotoxicity and photochemical oxidant formation. The LCA results are compared with virgin PET fibre and other commodity fibre products, i.e. cotton, viscose, PP and PLA.

The LCA results show that recycled PET fibres offer important environmental benefits over virgin PET fibre. Depending on the allocation methods applied for open loop recycling, NREU savings of 40-85% and GWP savings of 25-75% can be achieved. Mechanical recycling has a better overall environmental profile than chemical recycling, but chemically recycled fibres have the advantage that they can be applied in a wider range of applications than mechanically recycled fibres.

Among the three methods we applied, the “cut-off” approach reflects current environmental policy (e.g. emission trading), where companies or sectors are addressed as individual actors and their actual energy use and emissions are fully taken into account. This is not the case for the “waste valuation” method because it shifts part of the environmental burden from primary to secondary production. Compared to the “cut-off” approach, the “waste valuation” method is less favourable for the recycling industry. However, it can encourage product design for recyclability because producing a recyclable product results in a credit by shifting part of the impacts to the recycled products. The “system expansion” method reflects the overall efficiency of material utilization without distinguishing different players. In a policy context where responsibilities are assigned to individual companies or sectors, it is difficult to apply the “system expansion” method. However, in a LCA context, this method implements life-cycle thinking and it is therefore our preferred method.

Chapter 6 goes a step further on the topic of PET recycling by modelling a change-oriented LCA. In this case study, used PET bottles are recycled into both bottles and fibre, which are the two most important recycled PET products. The purpose of this LCA is to find the environmental optima of such a recycling system and to compare it with the reference system where the virgin PET is produced, used and disposed of (i.e. no recycling). The functional unit is 650 kg of fibres and 350 kg of bottles (total 1000 kg of PET products). The system boundary is cradle to grave without the use phase. We applied the “system expansion” method for open-loop recycling. Starting from the analysis of the baseline recycling system (where virgin PET bottles are produced, used and recycled into both bottles (12%) and fibre (88%), used again and disposed of), four change-oriented effects are analysed: the effect of multiple recycling trips, the effect of the shares of recycled PET used for bottle- or fibre- making, the effect of changing market demand (changing the functional unit and thus, changing the reference system), and the effect of using bio-based PET.

The baseline recycling system reduces both NREU and GHG emissions by 20% compared to the reference system. Multiple recycling trips can maximally reduce the impacts by 26% but the additional savings are negligible after three recycling trips. In the baseline case, more fibres are needed than bottles in one functional unit (650 kg fibre and 350 kg bottles). This leads to the LCA result that bottle-to-fibre recycling offers more impact reduction than bottle-to-bottle recycling. When all recycled PET pellets are used to make fibre, the savings are approx. 25%. In contrast, if the functional unit is reversed into 350 kg of fibre and 650 kg of bottles, i.e. if there is more market demand for bottles than for fibre, 30% of the impact reduction can be achieved. Both NREU and GHG emissions can be further reduced when the quantity of the recycled PET pellets is maximised. The bio-based PET recycling system offers at least 36% impact reduction, representing the lowest impact among all systems studied. We conclude that the system’s environmental impact can be minimised by maximising the amount of recycled PET polymer in the system and by using bio-based polymers.

Chapter 7 “synthesises” the findings from Chapters 2-6. In this chapter, the environmental impact of bio-based materials, recycled materials and their petrochemical counterparts are reviewed and compared. The PET systems include petrochemical PET, bio-based PET, recycled PET and recycled bio-based PET. Other bio-based product systems include PLA and man-made cellulose fibres. The single use of petrochemical PET is set as the reference system. The LCAs on three products made from these materials are reviewed, namely polymer granulates, fibres and bottles. The system boundary is cradle to grave without the use phase. The environmental impacts considered are NREU and GHG emissions.

The results show that both recycled and bio-based materials offer important environmental benefits over single-use petrochemical PET. Among the four PET product systems, recycled bio-based PET has the lowest impacts, followed by recycled PET, bio-based PET and petrochemical PET. Man-made cellulose fibres produced in integrated plants and PLA have lower impacts than both petrochemical PET and bio-based PET.

The impacts of recycled products are strongly influenced by the choice of the allocation method applied to the open-loop recycling system. Compared to the "system expansion" method, the "cut-off" approach leads to relatively low NREU but high GHG emissions. The "waste valuation" method leads to relatively high results of both NREU and GHG emissions. In spite of the wide range of results as consequence of the choice of the allocation method, it is safe to conclude that recycled product systems cause clearly lower environmental impact than the virgin petrochemical product systems. However, when the recycled products are compared with bio-based alternatives, the ranking becomes less straightforward. The lack of uniform standards for open-loop recycling in LCA leads to uncertainty regarding the environmental performance of recycled products. Policy makers and the LCA community should therefore establish a clear procedure in order to support decision making for recycled products.

Bio-based products have attracted much attention in recent years. The technical substitution potential of bio-based polymers replacing petrochemical polymers is enormous (Chapter 2). The bioplastics industry grew strongly in the past decade and will continue growing in the future (Chapter 2). Bio-based polymers reduce our dependency on limited fossil fuels and offer environmental benefits (Chapters 3 and 4). However, bio-based materials face their own sustainability challenges, e.g. the potential GHG emissions arising from land use change and the potential competition with food production. For land use change, especially indirect land use change, there is so far no generally accepted methodology and database, calling for future research in this area. The development of bio-based material is still in an early stage. We recommend that decisions are made with caution to avoid competition with food production.

Like bio-based products, recycled products reduce our dependency on limited fossil fuels and reduce the amount of municipal waste. Recycled products offer at least as much savings of NREU and GHG emissions as bio-based products (Chapters 5, 6 and 7). Unlike bio-based polymers, recycled polymers do not cause problems related to land use change and food competition. The outcome of the environmental assessments presented in Chapters 5, 6 and 7 strongly indicate that more attention should be paid to recycling. In this thesis we only studied PET; in practice, more plastics are recycled on a large scale (e.g. PE, PVC and EPS). The full potential of plastic recycling has not been explored. Major research efforts are under-way for bio-based products while the activities in the area of recycling are very limited.

Overall, we conclude that bio-based and recycled materials offer important opportunities for cleaner production. The environmental impact assessments presented included the standard categories of impact assessment (e.g. the CML method applied in Chapters 4 and 5), or only NREU and GHG emissions (Chapters 3, 6 and 7). Future research on the impact of water use, land use and biodiversity is urgently required. Furthermore, for both bio-based and recycled polymers, continuous efforts should be made to improve material properties. If a wide range of applications is covered, a significant impact reduction will be achieved in the future. However, improvement of material properties may lead to a higher production cost, which is a potential barrier to the implementation of bio-based and recycled polymers. In this thesis, we assessed the sustainability only from the environmental point of view. Future research on the economics and the social aspects of bio-based and recycled polymers is required.

Samenvatting

Vrijwel alle kunstmatige plastics en vezels zijn tegenwoordig gebaseerd op synthetische polymeren. Deze synthetische polymeren, op basis van petroleum dat miljoenen jaren nodig heeft gehad om te vormen, staan voor drie uitdagingen op het gebied van duurzaamheid: (i) de beperkte beschikbaarheid van fossiele brandstoffen, (ii) de impact van afval uit niet-afbreekbare kunststoffen op het milieu, en (iii) de emissie van broeikasgassen veroorzaakt door de verbranding van fossiele brandstoffen. Om deze uitdagingen het hoofd te bieden zijn twee strategieën voorgesteld. Ten eerste het vervangen van traditionele petrochemische polymeren door biobased polymeren. Ten tweede het verbeteren van de algehele efficiëntie van materiaalgebruik door middel van recycling. Kunststoffen en vezels zijn de twee belangrijkste toepassingen van synthetische polymeren. De doelstelling van dit proefschrift is om te bepalen wat de impact is die kunststoffen en kunstmatige vezels uit biobased en gerecyclede materialen hebben op het milieu en ze hierin te vergelijken met hun conventionele tegenhangers.

De eerste onderzoeksvraag van dit proefschrift luidt: *Wat is de huidige stand van zaken en wat zijn de potentiële ontwikkelingen in de biobased kunststofindustrie?"* In de afgelopen decennia zijn we getuige geweest van de commercialisering van verscheidene nieuwe biobased kunststoffen, zoals PLA (polylactide), PHA (polyhydroxyalkanoaten), zetmeelplastic, biobased PTT (polytrimethyleentereftalaat) en biobased PE (polyetheen). De huidige marktomvang en de status van ontwikkeling van deze opkomende industrie zijn echter nog niet grondig onderzocht. Hoofdstuk 2 geeft een overzicht van de huidige wereldmarkt van biobased kunststoffen, hun materiaaleigenschappen, hun potentieel om andere materialen technisch te vervangen, en toekomstige marktontwikkelingen (tot 2020). Zo'n 70 bedrijven wereldwijd zijn opgenomen in ons marktonderzoek.

De lange historie die het gebruik van natuurlijke grondstoffen heeft in de productie van kunststoffen toont aan dat biobased producten niet fictief en ook niet nieuw zijn. Integendeel, biobased producten vormen al vele decennia lang een industriële realiteit op een schaal van miljoenen tonnen (o.a. papier en karton). Tegenwoordig is het gecombineerde volume van non-food/non-plastics toepassingen van zetmeel en kunstmatige cellulosevezels 55 keer groter dan het totale volume van alle opkomende biobased kunststoffen (ca. 20 mln. ton tegenover ca. 0.36 mln. ton in 2007).

In de periode van 2003 tot 2007 was de jaarlijkse groei van opkomende biobased kunststoffen bijna 40%. Dit resulteerde in een wereldwijde capaciteit van 0.36 mln. ton in 2007. De wereldwijde productie van biobased kunststoffen zal waarschijnlijk sterk blijven groeien, 2.3 mln. ton bereiken in 2013, en 3.5 mln. ton in 2020. De verwachting is dat zetmeelplastic, PLA, biobased PE, PHAs en biobased epoxy de belangrijkste biobased kunststoffen in de toekomst zullen zijn. Het potentieel dat biobased kunststoffen (waaronder kunstmatige vezels) hebben om hun petrochemische tegenhangers technisch te vervangen wordt geschat op 240 mln. ton, of 90% van de totale hoeveelheid plastics en vezels gebaseerd op de marktvraag van 2007. Uitsluitend op basis op de technische haalbaarheid (economische en andere aspecten niet beschouwd) kan dus gesteld worden dat het groeipotentieel van biobased kunststoffen enorm is.

Hoofdstuk 2 concludeert dat biobased kunststoffen in meerdere aspecten gunstiger zijn dan hun petrochemische tegenhangers. Dit zijn de beperkte en onzekere levering van fossiele brandstoffen (o.a. olie en gas), economische haalbaarheid van biomassagrondstof, milieukundige afwegingen (bijv. besparing van niet-hernieuwbare energie en broeikasgasvermindering), innovaties die nieuwe mogelijkheden bieden (op gebied van techniek, werkgelegenheid, etc.) en vernieuwing in alle stappen van chemisch onderzoek tot aan het eindproduct en afvalverwerking. Uitdagingen die met succes moeten worden aangegaan in de komende jaren en komende decennia zijn de ondermaatse prestaties geleverd door sommige biobased kunststoffen (o.a. thermoplastisch zetmeel), relatief hoge productie- en verwerkingskosten en de noodzaak om het grondgebruik van land- en bosbouw te minimaliseren om concurrentie met voedselproductie te voorkomen en om negatieve effecten op biodiversiteit en andere impacts op het milieu te minimaliseren.

Volgend op de eerste onderzoeksvraag poneren we onze tweede onderzoeksvraag: *"Wat zijn de impacts van biobased polymeren op het milieu?"* Deze onderzoeksvraag wordt beantwoord in hoofdstuk 3 en hoofdstuk 4. Hoofdstuk 3 beschrijft het inzicht dat verworven is in de impacts die producten uit polysachariden hebben op het milieu in vergelijking met hun tegenhangers, petrochemische producten dan wel conventionele polysachariden. Polysachariden behoren tot de belangrijkste hernieuwbare grondstoffen voor de mensheid. Ze worden al lange tijd op grote schaal gebruikt voor voedsel (zetmeel), kleding (katoen, vlas, jute), communicatie (papier), verpakking (papier, karton) en in de bouw (hout). Naast deze traditionele toepassingen zijn andere non-food producten ontwikkeld om conventionele producten gedeeltelijk te vervangen, zoals producten gebaseerd op niet-hernieuwbare grondstoffen en traditionele producten gebaseerd op polysachariden. In hoofdstuk 3 beschouwen we gepubliceerde resultaten van levenscyclusanalyses (life cycle assessment, LCA) van textielproducten (o.a. viscose) gebaseerd op polysachariden, composieten met natuurlijke vezels en thermoplastisch zetmeel (thermoplastic starch, TPS). In de beschouwing hebben we gekozen voor niet-hernieuwbaar energieverbruik (non-renewable energy use, NREU) en de emissie van broeikasgassen als belangrijke indicatoren voor de milieuprofielen van producten.

Hoofdstuk 3 concludeert dat polysachariden belangrijke mogelijkheden bieden voor de vermindering van NREU en emissie van broeikasgassen, waarbij een "cradle-to-factory gate" analyse is uitgevoerd met 1 kg materiaal als functionele eenheid. Kunstmatige cellulosevezels kunnen ca. 10-30% NREU verminderen ten opzichte van katoen en tot 50-80% ten opzichte van PET. Wanneer we technische materialen beschouwen, kunnen composieten met natuurlijke vezels 25-30% NREU en tot 40% broeikasgassen verminderen ten opzichte van glasvezelcomposieten. Ook geldt: hoe hoger het vezelgehalte in composieten met natuurlijke vezels, des te lager de NREU en emissie van broeikasgassen. Wanneer we verpakkingsmaterialen beschouwen, kan TPS 25-75% NREU en 20-70% emissie van broeikasgassen verminderen ten opzichte van petrochemische polymeren (±15% afhankelijk van de gebruikte referentie: HDPE, LDPE of LLDPE).

De gebruiksfase en de hierop volgende afvalverwerking in acht nemend kan niet geconcludeerd worden dat producten gebaseerd op polysachariden altijd beter zijn dan hun petrochemische tegenhangers, maar belangrijke voordelen zijn aanwezig. Als algehele conclusie kan gesteld worden dat "from-cradle to-factory gate", in termen van NREU en emissie van broeikasgassen, producten uit polysachariden beter zijn dan hun conventionele tegenhangers, welke vooral bestaan uit petrochemische producten. Up-to-date informatie over kunstmatige cellulosetextiel was niet beschikbaar, wat heeft geleid tot het schrijven van hoofdstuk 4 van dit proefschrift.

In hoofdstuk 4 is de impact van kunstmatige cellulosevezels op het milieu onderzocht. Een LCA is uitgevoerd voor drie vezelsoorten, te weten Viscose, Modal en Tencel, die worden geproduceerd door Lenzing AG. Kunstmatige cellulosevezels behoren tot de drie belangrijkste vezelsoorten ter wereld (samen met polyester en katoen). In het uitgevoerde LCA-onderzoek wordt als functionele eenheid 1 ton stapelvezel gebruikt. Vijf kunstmatige cellulosevezelproducten zijn beschouwd, te weten Viscose (Azië), Viscose (Oostenrijk), Modal, Tencel en Tencel (2012). Het toegepaste systeemkader is "cradle to factory gate". We vergelijken de resultaten van de LCA met andere commodity-vezels, te weten katoen, PET en PP. De cumulatieve energiebehoefte (de optelsom van NREU en hernieuwbaar energieverbruik), het waterverbruik, het grondgebruik en categorieën uit de CML- baselinemethode zijn beschouwd. Gevoeligheidsanalyses zijn uitgevoerd om de impact van verschillende allocatiemethoden te nader te bepalen. Daarnaast zijn drie single-scoremethoden (Single Score I, II en III) geïntroduceerd en toegepast om een algehele conclusie te kunnen vormen.

Op basis van de resultaten van de LCA concluderen we dat, met uitzondering van Viscose (Azië), alle kunstmatige cellulosevezels betere milieuprofielen hebben dan PET, PP en katoen; van alle beschouwde vezels heeft Tencel (2012) de laagste impact. Viscose (Azië) heeft een lagere impact dan katoen en is vergelijkbaar met PET, maar minder gunstig dan PP en andere kunstmatige cellulosevezels. De milieutechnische voordelen die Viscose (Oostenrijk) en Modal hebben zijn vooral toe te wijzen aan de geringe behoefte aan fossiele brandstoffen in de pulp- en vezelproductie. Dit is het resultaat van procesintegratie, het gebruik van duurzame energie en kredieten verkregen uit bijproducten. Verder hebben Viscose (Oostenrijk) en Modal veel lagere procesemissies (o.a. SO_2 en NOx) vergeleken met Viscose (Azië), dit leidt tot lagere toxiciteit voor de mens, verminderde fotochemische oxidantvorming, verminderde verzuring en verminderde eutrofiëring. De milieutechnische voordelen van Tencel (2012) zijn het resultaat van laag energieverbruik, beperkt gebruik van chemische middelen, lage CO_2-emissie, lage SO_2-emissie en laag waterverbruik, dit leidt tot lage impacts op het milieu wat betreft abiotische uitputting, terrestrische ecotoxiciteit. fotochemische oxidantvorming en verzuring. Katoen is geen energie-intensief product; het heeft de laagste cumulatieve energiebehoefte van alle beschouwde vezels. Toch is katoen beoordeeld als de minst gunstige keuze door Single Scores II en III. De belangrijkste milieutechnische problemen waar katoen mee te maken heeft zijn grondgebruik, waterverbruik, zoetwater-ecotoxiciteit, terrestrische ecotoxiciteit en eutrofiëring. Het gebruik van pesticiden bij de verbouwing van katoen heeft het grootste aandeel in de ecotoxiciteiten. Daarnaast resulteert het gebruik van kunstmest tot eutrofiëring. In de gevoeligheidsanalyses zijn twee alternatieve allocatiemethoden toegepast, gebaseerd op de calorische en economische waardes. Hoewel beide leiden tot minder gunstige resultaten voor kunstmatige cellulosevezels vergeleken met met de standaardmethode, verandert de rangorde van alle beschouwde vezels hierdoor niet. Wanneer het systeemkader bestaat uit "cradle to factory gate" plus afvalverbranding met en zonder terugwinning van energie zijn bovendien alle beschouwde kunstmatige cellulosevezels beter dan PET en PP.

In dit onderzoek zijn state-of-the-art LCA impact assessment-methoden toegepast. De kwaliteit van bepaling van toxiciteit met behulp van LCA-tools is op het moment echter nog in twijfel te trekken, en er lopen verschillende onderzoeken om de methoden te verbeteren en databanken te vervolledigen. Daarnaast zijn alleen milieukundige indicatoren beschouwd die gangbaar waren in LCA-onderzoek. Een voorbeeld van een indicator die niet is beschouwd is de impact op biodiversiteit. Resultaten voor grondgebruik en waterverbruik zijn alleen gerapporteerd als inventarisatie, wat wil zeggen dat verschillende soorten grond (en water) niet bij elkaar zijn opgeteld bij gebrek aan geschikte methoden. Ook is geen rekening gehouden met bijvoorbeeld het risico op explosies. Deze onzekerheden buiten beschouwing gelaten kunnen we concluderen dat moderne kunstmatige cellulosevezels een duidelijk potentieel hebben om impacts op het milieu te verminderen ten opzichte van katoen en synthetische vezels.

In hoofdstukken 5 en 6 verleggen we onze aandacht van biobased materialen naar recycling. De doelstelling van deze hoofdstukken is het beantwoorden van de derde onderzoeksvraag: *"Wat is de impact van producten uit gerecyclede PET-polymeer op het milieu?"* Parallel aan de snelle ontwikkeling van biobased materialen heeft de kunststofrecyclingindustrie een aanzienlijke groei doorgemaakt in het afgelopen decennium. De recycling van gebruikte PET-flessen is inmiddels een gevestigd systeem met een eigen logistieke keten, inclusief inzameling van flessen, productie van PET-vlokken en productie van plastic pellets. Het onderwerp is ook van belang omdat in de LCA-methode tot dusver geen standaard bestaat voor open-loop recycling. Naar open-loop recycling bestaan nauwelijks case studies, hoewel discussie van de methode uitgebreid gepubliceerd is.

De eerste doelstelling van hoofdstuk 5 is om de impacts te begrijpen die gerecyclede PET-vezel heeft op het milieu in vergelijking met virgin (nieuw geproduceerde) PET-vezel. De tweede doelstelling is het toepassen van meerdere allocatiemethoden op dit voorbeeld van open-loop recycling. In deze LCA hebben we vier technieken voor recycling onderzocht, te weten mechanische recycling, semi-mechanische recycling, afbreken tot oligomeren en afbreken tot monomeren. De LCA-resultaten zijn vergeleken met het ecologische profiel van virgin PET-vezel. Drie methoden zijn toegepast op deze open-loop recycling, te weten “cut-off” (afkapping), “waste valuation” (toekenning van waarde aan afval) en “system expansion” (systeemuitbreiding). De eerste twee methoden, “cut-off” en “waste valuation”, beschouwen het “cradle to factory gate” systeem met flesafval als “cradle”. De derde methode, “system expansion” wordt toegepast om het “cradle to grave” systeem te beoordelen. De gebruiksfase is niet opgenomen in deze LCA. Negen indicatoren voor impact op het milieu zijn geanalyseerd, te weten NREU, klimaatverandering, abiotische uitputting, verzuring, eutrofiëring, toxiciteit voor de mens, zoetwater-ecotoxiciteit, terrestrische ecotoxiciteit en fotochemische oxidantvorming. De LCA-resultaten worden vergeleken met virgin PET-vezel en andere commodity-vezelproducten, te weten katoen, viscose, PP en PLA.

De LCA-resultaten laten zien dat gerecyclede PET-vezels belangrijke milieutechnische voordelen bieden ten opzichte van virgin PET-vezel. Afhankelijk van de allocatiemethoden die zijn toegepast voor open-loop recycling kan een NREU-vermindering van 40-85% en een GWP-vermindering van 25-75% worden bereikt. Het algehele milieuprofiel van mechanische recycling is beter dan dat van chemische recycling, hoewel vezels die zijn geproduceerd via chemische recycling een uitgebreider toepassingsgebied hebben.

Van de drie toegepaste methoden is de “cut-off”-benadering een weerspiegeling van huidig milieubeleid (o.a. emissiehandel), waarbij bedrijven of sectoren worden beschouwd als individuele actoren en volledig rekening wordt gehouden met hun daadwerkelijke energieverbruik en emissies. Dit is niet het geval bij de “waste valuation”-methode, die een deel van de milieubelasting verschuift van primaire naar secundaire productie. In vergelijking met de “cut-off”-benadering is de “waste valuation”-methode minder gunstig voor de recyclingindustrie. Toch kan dit stimuleren dat in productontwerp rekening wordt gehouden met recycling, omdat het produceren van een recyclebaar product resulteert in een krediet doordat een deel van de impact wordt verschoven naar het gerecyclede product. De “system expansion”-methode weerspiegelt de algehele efficiëntie in materiaalgebruik zonder onderscheid te maken tussen verschillende spelers. In een beleidscontext, waarin verantwoordelijkheden worden toegewezen aan individuele bedrijven of sectoren, is de “system expansion”-methode moeilijk toepasbaar. Echter, in een LCA-context geeft deze methode gestalte aan de life-cycle filosofie en krijgt daarom als methode onze voorkeur.

Hoofdstuk 6 neemt een stap verder in het onderwerp PET-recycling door een LCA te modelleren die is gericht op verandering (change-oriented). In deze case-study worden gebruikte PET-flessen gerecycled tot flessen en tot vezel, de twee belangrijkste gerecyclede PET-producten. Het doel van deze LCA is om de optimale impact te vinden die een dergelijk recyclesysteem kan hebben op het milieu en om deze te vergelijken met het referentiesysteem, waarin de virgin PET wordt geproduceerd, gebruikt en weggedaan (dat wil zeggen, geen recycling). De functionele eenheid is 650 kg vezels en 350 kg flessen (in totaal 1000 kg PET-producten). Het systeemkader is “cradle to grave” zonder de gebruiksfase. We passen de “system expansion” methode toe voor open-loop recycling. Beginnend met de analyse van het baseline-recyclesysteem (waarin virgin PET-flessen worden geproduceerd, gebruikt en gerecycled tot flessen (12%) en vezel (88%), hergebruikt en weggedaan) worden vier change-oriented effecten geanalyseerd:

Het effect van meerdere recycleronden, het effect van welk gedeelte gerecycled PET wordt gebruikt om vezel dan wel flessen te maken, het effect van veranderingen in de marktvraag (verandering van de functionele eenheid en daarmee ook het referentiesysteem), en het effect van het gebruik van biobased PET.

Het baseline-recyclesysteem vermindert zowel NREU als broeikasgasemissies met 20% in vergelijking met het referentiesysteem. Het toepassen van meerdere recycleronden kan de impacts verminderen met maximaal 26%, verdere besparing is verwaarloosbaar na drie recycleronden. In het baseline-recyclesysteem zijn meer vezels dan flessen benodigd per functionele eenheid (650 kg vezel en 350 kg flessen). Dit leidt tot het LCA-resultaat dat recycling van flessen tot vezel meer impactvermindering biedt dan recycling van flessen tot flessen. Wanneer alle gerecyclede PET-pellets worden gebruikt om vezels te maken, is de besparing ca. 25%. Wanneer echter de functionele eenheid wordt omgedraaid naar 350 kg vezel en 650 kg flessen, dat wil zeggen, als er een grotere marktvraag is naar flessen dan naar vezel, dan kan 30% van de impactvermindering worden behaald. NREU en broeikasgasemissies kunnen beide verder worden verminderd door de hoeveelheid gerecyclede PET-pellets te maximaliseren. Het biobased PET-recyclesysteem biedt tenminste 36% impactvermindering, waarmee het de laagste impact vertegenwoordigt van alle systemen die in beschouwing zijn genomen. We concluderen dat de impact die het systeem heeft op het milieu kan worden geminimaliseerd door de hoeveelheid gerecyclede PET-polymeer te maximaliseren in het systeem en door het gebruik van biobased polymeren.

Hoofdstuk 7 combineert de bevindingen van hoofdstukken 2-6. Het geeft een overzicht en een vergelijking van de impact die biobased materialen, gerecyclede materialen en hun petrochemische tegenhangers hebben op het milieu. De beschouwde PET-systemen zijn petrochemische PET, biobased PET, gerecyclede PET en gerecyclede biobased PET. Andere beschouwde biobased productsystemen zijn PLA en kunstmatige cellulosevezels. Het eenmalig gebruik van petrochemische PET is gebruikt als referentiesysteem. Een evaluatie wordt gegeven van de LCA's van drie producten gebaseerd op deze materialen, te weten polymeergranulaten, vezels en flessen. Het systeemkader is "cradle to grave" zonder de gebruiksfase. De beschouwde impacts op het milieu zijn NREU en broeikasgasemissies.

De resultaten laten zien dat gerecyclede en biobased materialen beide milieutechnische voordelen bieden ten opzichte van eenmalig gebruikte petrochemische PET. Van de vier PET-productsystemen heeft gerecyclede biobased PET de minste impact, gevolgd door gerecycled PET, biobased PET en tenslotte petrochemische PET. Kunstmatige cellulosevezels die zijn geproduceerd in geïntegreerde fabrieken en PLA hebben minder impact dan zowel petrochemische als biobased PET.

De impacts van gerecyclede producten worden sterk beïnvloed door de keuze van de allocatiemethode die is toegepast op het open-loop recyclesysteem. Vergeleken met de "system expansion"-methode leidt de "cut-off"-methode tot een relatief lage NREU maar tot hoge broeikasgasemissies. De "waste valuation"-methode leidt tot relatief hoge resultaten voor zowel NREU als broeikasgasemissies. Ondanks de uiteenlopende resultaten als gevolg van de keuze van de allocatiemethode kunnen we veilig concluderen dat de gerecyclede productsystemen een duidelijk lagere impact op het milieu met zich meebrengen dan de virgin petrochemische systemen. Echter, wanneer de gerecyclede producten worden vergeleken met biobased alternatieven is de rangorde minder eenduidig. Het gebrek aan uniforme standaarden voor open-loop recycling in LCA-onderzoek leidt tot onzekerheden wat betreft de milieutechnische prestaties van gerecyclede producten. Beleidsmakers en de LCA-gemeenschap zouden daarom een duidelijke procedure moeten opstellen teneinde beleidsvorming op het gebied van gerecyclede producten te ondersteunen.

De afgelopen jaren hebben biobased producten veel aandacht getrokken. De mogelijkheden om petrochemische producten technisch te vervangen door biobased producten zijn enorm (hoofdstuk 2). De bioplastics-industrie is sterk gegroeid in de afgelopen decennia en zal blijven groeien in de toekomst (hoofdstuk 2). Biobased polymeren verminderen onze afhankelijkheid van beperkte fossiele brandstoffen en bieden milieutechnische voordelen (hoofdstukken 3 en 4). Biobased materialen hebben echter hun eigen uitdagingen op het gebied van duurzaamheid, zoals de potentiële broeikasgasemissies die voortkomen uit verandering in grondgebruik en de mogelijke concurrentie met voedselproductie. Voor verandering in grondgebruik, met name indirect grondgebruik, bestaan tot dusver geen geaccepteerde methode en databank, iets wat dringend vraagt om aanvullend onderzoek op dit gebied. De ontwikkeling van biobased materialen verkeert nog in een vroeg stadium. We adviseren dat beslissingen zorgvuldig worden gemaakt om zo concurrentie met voedselproductie te voorkomen.

Net als biobased producten verminderen gerecyclede producten onze afhankelijkheid van beperkte fossiele brandstoffen en verminderen ze de hoeveelheid afval. Gerecyclede producten bieden minstens zoveel vermindering van NREU en broeikasgasemissies als biobased producten (hoofdstukken 5, 6 en 7). In tegenstelling tot biobased polymeren veroorzaken gerecyclede polymeren geen problemen met betrekking tot verandering in grondgebruik en concurrentie met voedselproductie. De uitkomst van de milieukundige beschouwing die wordt gepresenteerd in hoofdstukken 5, 6 en 7 duidt erop dat meer aandacht aan recycling moet worden besteed. In dit proefschrift hebben we alleen PET beschouwd; in de praktijk worden meer kunststoffen op grote schaal gerecycled (o.a. PE, PVC en EPS). Het volledige potentieel van kunststofrecycling is nog niet verkend. Grote onderzoeksinspanningen worden verricht op het gebied van biobased producten terwijl de activiteit op het gebied van recycling zeer beperkt is.

Over het geheel genomen concluderen we dat biobased en gerecyclede materialen belangrijke mogelijkheden bieden voor duurzamere productie. De gepresenteerde beoordelingen van impacts op het milieu betreffen de standaardcategorieën voor impactbeoordeling (zoals de CML-methode toegepast in hoofdstukken 4 en 5) of alleen NREU en broeikasgasemissies (hoofdstukken 3, 6 en 7). Toekomstig onderzoek naar de impact van waterverbruik, grondgebruik en op biodiversiteit is dringend nodig. Daarnaast zou continu inspanning moeten worden verricht om de materiaaleigenschappen van zowel biobased als gerecyclede polymeren te verbeteren. Als een breed scala aan toepassingen kan worden gedekt zal op den duur een significante impactvermindering worden bereikt. De verbetering van materiaaleigenschappen kan echter leiden tot hogere productiekosten, een potentiële barrière voor de implementatie van biobased en gerecyclede polymeren. In dit proefschrift hebben we de duurzaamheid alleen beschouwd vanuit milieuoogpunt. Toekomstig onderzoek is nodig naar de economische en sociale aspecten van biobased en gerecyclede polymeren.

总结论

当前，几乎所有的塑料和人造纤维制品都是由人工合成的高分子聚合物(synthetic polymers)制造而成。人工合成的高分子聚合物是从石油(化石燃料)中提取生产得到，而石油的形成需要上百万年的地质条件。总体说来，高分子聚合物面临着三个主要的可持续发展的挑战：一、有限的石油资源；二、无法降解的塑料垃圾；三、化石燃料燃烧所形成的温室气体。为了应对这些可持续发展的挑战，我们提出两点策略。首先，使用生物质(bio-based)的材料[1]来代替传统的、由石油生产出来的高分子材料；其次，回收循环利用现有的高分子材料，提高材料的整体利用效率。塑料和纤维是高分子材料运用的两个最主要方面，本论文旨在对从生物质原料生产出来的塑料和纤维，以及对塑料和纤维产品的循环再利用的塑料进行环境影响评估，并且将此评估的结果与传统的、同等的石油制品进行比较。

本论文的第一个研究问题是：**“生物塑料产业的现状以及发展的潜力如何？”**在过去的几十年里，我们目睹了众多生物塑料新材料的产业化过程，诸如聚乳酸(PLA)，聚羟基脂肪酸酯(PHAs)，淀粉塑料，基于生物质的对苯二甲酸丙二醇(PTT)以及基于生物质的聚乙烯(PE)。然而，对于目前生物质塑料的市场现状以及此产业的发展定位和前景，产业界和学术界尚未提出完整的回顾性评价和预测。在第二章里，我们对目前全球生物质塑料的市场发展，新材料的特性，技术意义上的替代潜力，以及未来的市场发展(到2020年)进行了调查，分析与回顾。全球约70家公司参与了此调查。

从历史上看，很多产品都是由自然原料生产出来，这说明生物质产品既非虚构亦非全新概念。其实，几十年以来，生物质产品已经实现了百万吨规模的工业化(譬如造纸业)。现今，用于非粮食和非塑料的淀粉和人造纤维[2]的产量是全部新兴生物质塑料的产量的55倍(对比：前者全球2007年产量约两千万吨，后者约36万吨)。

从2003-2007的五年间，新兴生物质塑料的年增长率接近40%，2007年的全球产能达到36万吨。未来全球生物质塑料行业将持续强劲的发展势头。据预计，2013年全球产能将达到230万吨，2020年将达到350万吨。未来最重要的几个生物质塑料产品的种类为：淀粉塑料，聚乳酸(PLA)，基于生物质的聚乙烯(PE)，聚羟基脂肪酸酯(PHA)以及基于生物质的环氧树脂(epoxy resin)。单从技术意义上来看，生物质塑料(包括人造纤维)替代同等的石油产品的潜力最大可达到两亿四千万吨，或全球2007年全部塑料和纤维制品需求量的90%。所以仅从技术意义上的替代来看，生物质塑料的发展潜力将是巨大的。

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[1] 译注：生物质材料从可再生的资源里生产出来，例如：以玉米为原料，利用发酵技术生产的聚乳酸塑料。

[2] 译注：如用作粘合剂的工业淀粉。

在第二章中，我们得出这样的结论：生物质塑料的发展得益于诸多有利因素，这些因素包括有限的化石燃料资源(石油和天然气)，生物质原料的经济可行性，对环境的考虑(如节约不可再生能源和减少温室气体的排放)，创新所创造的机遇(科技、就业等)，以及从基础化学研究，到最终产品生产和废弃物处理的所有步骤的复兴。然而，在未来几年内，生物质塑料亦应面对诸多挑战。例如，某些生物质塑料较低的性能(如热塑淀粉材料)，生物质材料较高的生产和工艺成本，以及对农业和林业用地需求的最小量化，从而避免与粮食生产产生竞争，同时也减少对生物多样性以及其它环境指标的不利影响。

在回答了第一个研究问题的基础上，论文的第二个研究问题是“**生物质高分子材料对环境的影响如何？**”在第三章和第四章里，我们对这个问题进行研究。第三章旨在对多糖聚合物(polysaccharide)产品的环境影响进行深度的了解，并且与同等的传统产品进行比较，这些传统产品既可为石油制品，也可为传统的多糖聚合物产品。多糖聚合物是为人类所使用的最重要的可再生资源之一，长期广泛地被用于食品生产(如淀粉)，衣着(如棉花、亚麻)，通讯(如纸张)，包装(如纸、纸板)和建筑(如木制品)。除了这些传统的用途之外，它也被用在非食品的新产品领域，用于替代或部分替代传统的产品(传统产品基于非可再生原料或传统多糖聚合物的产品)。在第三章里，我们回顾了已发表的诸多对多糖聚合物产品的生命周期评估(Life Cycle Assessment, 或LCA)的研究成果，包括纺织材料(如粘胶)，天然纤维的复合材料(composites)，和热塑淀粉材料，并且将它们与传统的同等产品(如棉花和石油制品)进行比较。在这个回顾性分析里，我们选择不可再生能源使用(non-renewable energy use，或NREU)和温室气体(greenhouse gas，或GHG)排放作为所分析产品的两个重要的环境指标。

第三章的结论为，从“摇篮”到“工厂大门”，以每千克为功能单位(functional unit)，多糖聚合物对于减少不可再生能源的使用以及降低温室气体的排放具有重要的潜力。相对于棉纤维和涤纶，人造纤维素纤维可分别减少10-30%和50-80%的不可再生能源使用。就工程材料而言，天然纤维复合材料比玻璃纤维复合材料要减少25-30%对不可再生能源的需求和40%的温室气体排放。与此同时，纤维含量越高，自然纤维复合材料对不可再生能源的需求及温室气体的排放就越低。而就包装材料来说，每千克热塑淀粉材料相对于石油高分子材料可以减少25-75%对不可再生能源的需求，以及20-70%的温室气体排放(±15%取决于选择高密聚乙烯，低密聚乙烯或者线性低密聚乙烯为参考)。

如果同时考虑产品的使用过程以及使用后的废弃物的处理阶段，我们无法得出多糖聚合物产品总是优于同等的石油制品的结论，但是重要的优势依然存在。总体说来，从“摇篮”到“工厂大门”，对不可再生能源的需求及温室气体的排放而言，多糖聚合物优于同等的传统产品(大多数传统产品为石油制品)。同时，对于人造纤维素纤维的纺织材料，目前依然缺乏最新的数据和分析，于是本论文第四章由此而生。

在第四章里，我们对人造纤维素纤维对环境的影响进行了评估。生命周期评估(LCA)应用于三类纤维产品，分别为粘胶，木代尔(Modal)和天丝(Tencel)，三者均为兰精纤维有限公司(Lenzing AG)的产品。人造纤维素纤维是世界上三种最主要的纤维产品之一(其他两者分别为棉花和涤纶)。此LCA的功能单位设定为一吨短纤(staple fibre)。我们对五种人造纤维素纤维产品进行了分析，它们分别是粘胶(亚洲)，粘胶(奥地利)，木代尔，天丝，天丝(2012)。系统边界设定为从“摇篮”到“工厂大门”。我们将分析的结果与重要的商品短纤(即棉花、涤纶和丙纶)进行比较。此分析的环境指标包括：累积能源需求(cumulative energy demand，或CED，定义为全部不可再生能源与

可再生资源的总和)，水资源需求，土地使用，以及CML[3]基准环境指标。同时，我们对不同的分配方法(allocation method)进行了敏感性分析(sensitivity analysis)。最后，我们引进三种“单一评分”方法(单一评分I，II和III)，从而得出总体结论。

生命周期分析的结果显示，除粘胶(亚洲)之外，其他人造纤维素纤维对环境的影响均优于涤纶、丙纶和棉花的环境影响。天丝(2012)对环境的影响最低。粘胶(亚洲)的环境表现优于棉花，与涤纶环境表现相当，但劣于丙纶及其它人造纤维素纤维。粘胶(奥地利)和木代尔的优势主要体现在木浆和纤维制造中极少的化石燃料的使用，这是由于木浆和纤维制造的工艺整合程度很高，并且大量地使用可再生能源以及副产品的优势。再者，在粘胶(奥地利)和木代尔的生产过程中，二氧化硫和氮氧化合物的排放比粘胶(亚洲)要低很多，这直接导致了较低的人类毒性(human toxicity)，光化学氧化剂的形成(photochemical oxidant formation),[4] 酸雨的形成(acidification)和富营养化(eutrophication)。天丝(2012)的主要的环境优势可归功于少量不可再生能源和化学品的使用，较低的二氧化碳和二氧化硫的排放以及对水资源较低的使用需求。这些优势减少了对非生物资源枯竭(abiotic depletion)，土壤生态毒性(terrestrial ecotoxicity)，光化学氧化剂的形成以及酸雨的形成。棉花的生产无需大量能源，棉花的累积能源需求在所研究的短纤中是最低的。可是，所有三个单一评分方法均指出棉花为最不利的选择。棉花的环境问题主要集中在耕地的使用，水资源的需求(大量灌溉导致土壤盐碱化及地下水的减少)，淡水水域生态毒性(fresh water aquatic ecotoxicity)，土壤生态毒性和富营养化。在棉花的耕作过程中，杀虫剂和除草剂的使用导致其较高的生态毒性，而化肥的施用增加了(土壤和水域的)富营养化。在敏感性分析中(sensitivity analysis)，我们分析了两种替代的分配方法，即基于热值的分配和基于经济价值的分配。与缺省的分配方法相比，两种替代的分配方法对人造纤维素纤维均导致较高环境影响，然而，所有短纤的排名依然不变。此外，如果系统边界条件变更为“摇篮”到“工厂大门”，加上使用后的废物焚烧(兼顾能源回收和无能源回收)，此课题中所有的人造纤维素短纤的环境表现均优于涤纶和丙纶。

在此研究中，我们应用了目前最先进的LCA环境影响评估方法论。然而，当前的LCA毒性影响计算工具仍然具有不确定性，很多科研项目正着力更进此方法论并且完善所需的数据库。此外，此研究中我们只考虑了LCA中广为使用的环境指标，并非所有的指标都考虑在内，诸如对生物多样性的影响就没有在研究中体现。而对土地使用和水资源的需求，我们只汇报了直接考证的结果，即：不同的土地资源和不同的水资源需求所产生的影响并没有被累加起来，这是由于当前学术界仍然缺乏合适的对应的方法论。同样地，我们也没有考虑其他风险诸如化学产品的爆炸性等等。若抛开诸如此类的不确定性问题，我们可以下如此的结论：现代化的人造纤维素纤维相对于棉花和化纤来说，对于减少对环境的影响具有明显的优势。

在第五章和第六章里，我们的研究兴趣从生物质材料转移到循环利用。此两章节旨在回答第三个研究问题：**“循环使用的PET(聚对苯二甲酸乙二酯)产品对环境的影响如何？”** 在过去的几十年里，在生物质材料快速发展的同时，塑料的回收再利用产业也得到了迅速的发展。回收再利用废旧的PET塑料瓶已经建立起一条龙式的物流产业链，包括收集废瓶，生产干净的塑料碎片(flake)和热塑整合生产塑料粒子。此外，在LCA中，对于处理开环回收(open-loop recycling)的理论已被深入广泛讨论过，可是

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[3] 译注：CML是莱顿大学环境科学中心的简称。

[4] 译注：即光化学污染烟雾。

到目前为止，对开环回收的案例并没有可操作的标准，也鲜有文献记载实际案例。这是我们对这个课题的另一个兴趣所在。

在第五章里，首先我们想了解利用回收的PET废瓶生产出来的涤纶纤维(以下简称“回收涤纶”，涤纶即为PET化学纤维)的对环境影响，并且与原生涤纶(virgin PET fibre)对环境的影响做比较。其次，我们想对这个典型的开环回收的案例应用不同的分配方法(allocation methods)。这个LCA分析了四种回收再利用的工艺，包括物理回收，半物理回收，由低聚物(又称“寡聚物”，oligomer)回收以及由单体(monomer)回收。对于开环回收，我们应用了三种不同的分配方法，即：“截断法”(cut-off)，“废物价值法”(waste valuation)和“系统延展法”(system expansion)。根据“截断法”和“废物价值法”，系统范围定义为从“摇篮”到“工厂大门”，以废瓶为“摇篮”。根据“系统延展法”，系统范围定义为从“摇篮”到“坟墓”，以对自然资源的采伐为“摇篮”，产品的使用过程被排除到系统之外。在此LCA中，我们分析了九个环境指标，即：不可再生能源的使用，全球变暖效应，非生物资源的枯竭，酸雨的形成，富营养化，人类毒性，淡水水域生态毒性，土壤生态毒性和光化学污染。LCA分析的结果与其他主要的短纤产品的环境影响做了比较，包括棉花，粘胶，丙纶和聚乳酸纤维(基于生物质的聚酯纤维)。

意料之中，LCA的结果显示回收涤纶相对于原生涤纶有重要的环境优势。对于不同的开环回收的分配方法，回收涤纶可以节约40-85%的不可再生能源，减少25-75%的全球变暖效应。物理回收对环境的影响小于化学回收，但是，化学回收所生产出来的涤纶比物理回收生产出来的涤纶具有更广泛的应用范围，材料性能也更接近于原生涤纶。

对于这三种分配方法，“截断法”体现了当前环境政策(如碳排放交易)的制定方式：以单个的企业或产业作为政策针对的对象，考虑企业或产业的实际能耗和排放。然而“废物价值法”采取了截然不同的观点，因为初级生产所产生的环境负担，一部分被转移到次级生产(回收产品)。相对于“截断法”而言，“废物价值法”并不有利于回收产业的发展。然而，此法可促进产品可回收性的设计，因为生产出可回收的初级产品，则部分环境影响可以转移到回收产品中去，大大减少了初级产品(如新PET)对环境的影响。“系统延展法”体现了系统中材料使用的整体效率，并不着力于区分初级产品或回收产品。从具体政策制定的角度来看，“系统延展法”难以体现单个的企业和产业的责任。然而，从LCA的理论角度来看，这个方法清楚地贯彻了生命循环的思路，从而也是我们所倾向的方法。

在第六章里，我们建立了“面向变化”(change-oriented)的LCA模型，对PET的回收再利用进行了更深入的讨论。在这个案例里，用过的PET瓶子被回收并同时转化成两个最主要的产品：PET瓶和PET化纤(涤纶)。此文的主旨在于寻找此系统的环境最优化点并且与参考系统的环境表现相比较。参考系统被定义为原生PET塑料从生产(从石油中)到使用再到丢弃(无回收再利用)的过程。功能单位定义为650千克纤维和350千克瓶子(共1000千克PET产品)(译注：此份额是由目前市场份额决定)。分析系统边界条件是从“摇篮”到“坟墓”，产品的使用阶段被排除在外。我们将“系统延展法”应用到这个开环回收的案例中。整个模型始于一个基础回收系统(a baseline recycling system)，在这个系统里，新的PET瓶子被生产出来，使用过后被回收再利用，12%的回收塑料粒子用于生产PET瓶子(瓶-瓶回收)，88%用于生产回收涤纶纤维(瓶-纤回收)，这些二次生产的瓶子和化纤，使用过后，不再进行回收再利用，而是被送入城市垃圾焚烧厂处理掉。基于这个基础回收系统，我们分析了四个面向变化的效应：一、多次反复回收利用的效应(译注：多次意为大于二次回收)；二、回收塑料粒子被用于生产瓶子和

纤维的份额对整体环境影响的效应；三、改变目前市场需求的份额的效应(从而改变功能单位，即同时需改变参考系统)；四、使用从生物质原料里生产出的PET的效应。

相对于参考系统，整个基础回收系统可以减少20%的不可再生能源和温室气体排放。多次循环回收可以最多减少26%的环境影响，但经过三次循环回收后，回收的额外的优势不再显著地增加。在基础回收系统里，每功能单位对纤维的需求量多于瓶子(650千克纤维和350千克瓶子)，因此LCA的结果显示，在这样的一个系统里，瓶-纤回收比瓶-瓶回收对环境更有利。当所有回收的PET都用于制造涤纶，整个回收系统对环境的影响可以减少25%。与之相反，如果我们调换功能单位，即350千克纤维和650千克瓶子，市场对PET瓶子的需求大于对涤纶的需求，则回收系统可减少30%的环境影响。如果我们在此模型中将回收PET粒子的量进行最大化，则整个系统对不可再生能源的需求和温室气体的排放可达到最小化。基于生物质的PET的回收系统可以减少36%的环境影响(相对于基础回收系统而言)，在研究的所有的系统中，回收利用基于生物质的PET具有最低的环境影响。因此，本章的结论为，当回收材料的份量在整个体系中最大化，或使用生物质材料，则整个体系对环境影响可最小化。

在第七章里，我们“综合”了2-6章的分析结果。我们对生物质材料和/或回收材料对环境的影响和与之相对应的石化材料对环境的影响做了回顾和比较。PET体系包括了石化PET(传统的PET)，生物质的PET，回收的PET，以及回收的生物质的PET。其他生物质的材料包括了聚乳酸和人造纤维素纤维，一次使用的石化PET产品系统设为参照体系。我们对三个产品种类的LCA进行了回顾, 即塑料粒子(中间产品)，纤维和瓶子。分析的范围包括从“摇篮”到“坟墓”，不包括产品的使用阶段。考虑的环境指标为不可再生能源的使用和温室气体的排放。

结果显示，回收的材料和生物质的材料，相对于传统的一次使用的石化材料来说，均有重要的环境优势。在所分析的四个PET产品系统中，回收的生物质PET产品具有最低的环境影响，其次为回收的PET，生物质PET，最后为传统的石化PET产品。由整合工艺生产出来的人造纤维素纤维和聚乳酸纤维的环境影响均小于石化PET纤维以及生物质PET纤维。

回收产品的环境影响很大程度上取决于用于开环回收的分配方法。相对于“系统延展法”而言，“截断法”的结果往往具有较低不可再生能源的需求和较高的温室气体排放；“废物价值法”的结果往往是同时具有较高的不可再生能源需求和温室气体排放。尽管不同分配方法产生结果范围广泛，但回收产品系统总体上优于非回收的石化产品系统。然而，回收产品系统与生物质的产品系统相比较时，结论并非如此直截了当。对于LCA中的开环回收问题，目前(学术界和产业界)仍然没有统一的标准，这导致了开环回收产品的环境影响有很大的不确定性。对此，我们建议政府以及LCA的社群应建立清晰明了的开环回收的LCA操作标准，以便对各类回收产品的决策决定提供技术支持。

近年来，生物质产品引起人们广泛的注意。从技术角度来看，生物质高分子聚合物替代传统石化高分子聚合物的潜力非常巨大(第二章)。过去几十年里，塑料产业的发展非常强劲，预计在未来几十年内，它将继续保持强劲的发展势头(第二章)。生物质聚合物能够减少我们对有限的化石燃料的依赖并且有利于环境保护(第三章与第四章)。然而，生物质材料也面临着自身的可持续发展的挑战，如：潜在的由于土地使用转换(land use change)而产生的温室气体，潜在的与粮食生产的竞争(有限的土地资源)。对于土地使用转换而言，特别是间接的土地使用转换(indirect land use change)，目前为止尚未建立被一致接受的方法论以及完整的数据库，未来的科研应着重于此。

生物质材料的发展尚处于初级阶段，我们建议任何决策都必须谨慎而定，以避免与粮食生产产生对土地资源的竞争。

与生物质产品一样，回收产品也能减少我们对有限的化石燃料的依赖，且减少垃圾的产生。回收产品至少能够减少与生物质产品相当的不可再生能源消耗和温室气体排放(第五、六、七章)。与生物质产品不同的是，回收产品不会产生土地使用转换的影响和与粮食生产对土地的竞争关系。第五、六、七章的环境影响分析结果强烈建议，我们应该将更多的注意力放在回收再利用资源上。本论文只研究了PET，实际上更多的塑料都被大规模地回收再利用(如聚乙烯，聚氯乙烯(PVC)和膨胀聚苯乙烯(EPS))，塑料回收的潜力尚为被完全发掘。现今主要的科研努力都集中在生物质产品上，而对于回收产品的研究却非常有限。

总之，生物质和回收产品为清洁生产提供重要的机遇。我们所分析的环境影响包括了一系列标准的环境影响指标(如第四章和第五章所使用的CML方法)，或仅限于能源需求与温室气体的排放(第三、六、七章)。未来的科研应急切地关注生物质和回收产品对水资源，生物多样性和土地使用的影响。另外，它们的材料特性也应该被进一步地提高。当材料能被广泛地应用于各个领域，其对环境的影响才可达到显著的降低。然而，材料特性的提高将会导致较高的生产成本，这也将成为生物质和回收产品被广泛运用的阻碍之一。此论文只从环境角度讨论了这两种产品的可持续发展，未来的研究方向亦须包括它们在经济以及社会影响方面的可持续性。

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Nomenclature

1,3-PDO	1,3-propanediol
1,4-DB	1,4-dichlorobenzene
Cl_2	Chloride
CO_2	Carbon dioxide
CS_2	Carbon disulfide
H_2	Hydrogen
N_2O	Nitrous oxide
$NaSO_4$	Sodium sulphate
NO_x	mono-nitrogen oxides
SO_2	Sulfer dioxide
ABS	Acrylonitrile butadiene styrene
AF	Allocation factor
AMPE	Association of Plastics Manufacturers in Europe
B2B	Bottle-to-bottle (recycling)
B2F	Bottle-to-fibre (recycling)
BAT	Best Available Technology
BAU	Business as usual
BHET	Bis-hydroxylethylene terephthalate
CED	Cumulative energy demand
CEWEP	Confederation of European Waste-to-Energy Plants

CIRFS	Comité International de la Rayonne et des Fibres Synthétiques (The International Rayon and Synthetic Fibres Committee)
CML	Centrum voor Milieuwetenschappen Leiden (Institute of Environmental Sciences), Leiden University, the Netherlands
COD	Chemical oxygen demand
DMT	Dimethyl terephthalate
ECH	Epichlorohydrin
EDC	Ethylene dichloride
EG	Ethylene glycol
EJ	Exa Joule; 1 EJ = 10^{18} Joule
EP	Epoxy resin
EPDM	Ethylene propylene diene M-class rubber
EPS	Expanded polystyrene
FAO	Food and Agriculture Organisation
FENC	Far Eastern New Century Co.
GHG	Greenhouse gas
GJ	Giga Joule; 1 GJ = 10^9 Joule
GPPS	General purpose polystyrene
GTE	Glycerine-to-epichlorohydrin (process)
GWP	Global warming potential
ha	hectare
HDPE	High density polyethylene
HIPS	High impact polystyrene
IEA	International Energy Agency
IISI	International Iron and Steel Institute
IPCC	Intergovernmental Panel on Climate Change
ISO	International Standardisation Organisation
JCFA	Japan Chemical Fibers Association

kg	kilogram
kt	kilo tonnes (=1000 metric tonnes)
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life Cycle Impact Assessment
LDPE	Low density polyethylene
LER	Liquid epoxy resin
LJG	Long John Group
LLDPE	Linear low density polyethylene
LUC	Land use change
MeOH	Methanol
MJ	Mega Joule; 1 MJ = 10^6 Joule
MSW	Municipal solid waste
MSWI	municipal solid waste incineration
Mt	million metric tonnes, or 10^6 metric tonnes
NaOCl	Sodium hypochlorite
NaOH	Sodium hydroxide, caustic soda
NMMO	*N*-Methylmorpholine-*N*-oxide
NOGEPA	Netherlands Oil and Gas Exploration and Production Association
NREU	Non-renewable energy use
p.a.	per annum
PA	Polyamide
PAH	Polycyclic aromatic hydrocarbon
PAS	Publicly Available Specification
PBAT	Polybutylene adipate-co-butylene terephthalate
PBS	Polybutylene succinate
PBSL	Polybutylene succinate-co-lactate

PBT	Polybutylene terephthalate
PC	Polycarbonate
PCL	Polycaprolactone
PDLA	Poly-D(-)lactide
PE	Polyethylene
PEIT	Polyethylene-co-isosorbite terephthalate
PET	Polyethylene terephthalate
PHA	Polyhydroxyalkanoates
PLA	Polylactic acid, or Polylactide
PLLA	Poly-L(+)lactide
PM	Policy and measures
PMMA	Polymethyl methacrylate
POM	Polyoxymethylene
POY	Partially oriented yarn
PP	Polypropylene
PRO-BIP	PRoduct Overview and market projection of emerging BIo-based Plastics
PS	Polystyrene
PTA	Purified terephthalate acid
PTI	Polytrimethylene isophthalate
PTN	Polytrimethylene naphthalate
PTT	Polytrimethylene terephthalate
PUR	Polyurethane
PVAc	Polyvinyl acetate
PVC	Polyvinyl chloride
PVOH	Polyvinyl alcohol
R-PET	Recycled polyethylene terephthalate
R&D	Research and Development

REU	Renewable energy use
SAN	Styrene acrylonitrile
SSP	Solid state polymerisation
T_g	Glass-transition temperature
T_m	Melting temperature
TPS	Thermoplstic starch
UCTE	Union for the Co-ordination of Transmission of Electricity
UP	Unsaturated polyester
USDA	US Department of Agriculture
V-PET	Virgin polyethylene terephthalate
VCM	Vinyl chloride monomer
w/o	without
WWII	World War II

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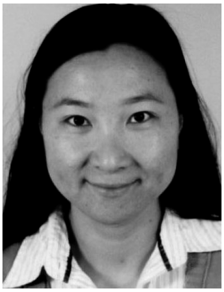
Curriculum Vitae



I was born on 12 May 1977 in Nanjing, China. From 1995 to 1999, I studied energy and power engineering, track refrigeration and cryogenics, in the University of Shanghai for Science and Technology (China). There I received my Bachelor degree in Engineering *cum laude* in 1999. After working in Shanghai for three years (first at a quality/environmental management consulting firm, later at a machinery manufacturing company), I decided to go back to university. In 2003, I came to the Netherlands to pursue my Master education. I followed the programme "Sustainable Development", track energy and resources, at Utrecht University and received my Master degree in Environmental Science in 2006. During my studies, I worked as a student research assistant for the BREW project (an EU project, see `www.chem.uu.nl/brew`) in the group of Science, Technology and Society in Utrecht University. Since 2006, I have worked as a junior researcher in the same group, under the supervision of Dr. Martin Patel. My PhD project was initially funded by an EU project – EPNOE (European Polysaccharide Network of Excellence, see `www.epnoe.org`). Later, the research was co-funded by EPNOE, industrial associations and companies. My research area includes impact assessment of bio-based and recycled materials, as well as LCA methodology development. In the past five years, I have been working closely with partners in industry. Several studies of mine have been used by industries to establish references or to contribute to decision making. In 2009 I received the *Paul Schlack Honoury Prize* for the research on man-made cellulose fibres.

Publication list

Journal publications

Shen L, Worrell E and Patel MK (Forthcoming) Comparing life cycle energy and GHG emissions of bio-based PET, recycled PET, PLA and man-made cellulosics. Submitted to *Environmental Science & Technology*

Shen L, Nieuwlaar E, Worrell E and Patel MK (Forthcoming) Life cycle energy and GHG emissions of PET recycling: change-oriented effects. Submitted to *International Journal of LCA*

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