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

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**Abstract:** The purpose of this paper is to review the environmental profiles of petrochemical PET, (partially) bio-based PET, recycled PET, and recycled (partially) bio-based PET, and compare them with other bio-based materials, namely PLA (polylactic acid, a bio-based polyester) and man-made cellulose fibers (cellulose fiber produced from wood pulp, i.e. Viscose, Modal and Tencel). Life cycle assessment (LCA) studies on polymers, fibers and bottles made from these materials are reviewed. Only non-renewable energy use and greenhouse gas (GHG) emissions are considered. The scope is cradle to grave excluding the use phase. 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 studied, recycled (partially) bio-based PET has the lowest impacts, followed by recycled PET, (partially) bio-based PET, and petrochemical PET. PLA and man-made cellulose fibers produced in an integrated plant 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 open-loop recycling. © 2012 Society of Chemical Industry and John Wiley & Sons Ltd

**Keywords:** PET; bio-based; recycling; PLA; man-made cellulose fibers; LCA; NREU; GHG emissions; ILUC

## 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 third most important synthetic polymer next to PE and PP.<sup>1,2</sup> Plastics

and fibers 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 fiber, 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.<sup>1–3</sup>

In the past five to ten years, bio-based materials have attracted increased attention due to the public concerns about the depletion of 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.<sup>4</sup> With the large capacity of bio-based ethylene installed,<sup>4</sup> partially bio-based PET has been 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 consumption, thereby 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,<sup>5</sup> which is approximately over 40% of the total PET bottles consumed (estimated based on statistics from PlasticsEurope<sup>6</sup> and Kunststoffe<sup>3</sup>). Like virgin PET polymer, recycled PET is mostly used to make fibers and bottles. Worldwide over 70% of the recycled PET is converted into fibers, 10% goes back to bottles, and 20% is used for other applications (e.g. sheets and strapping tape).<sup>7</sup>

Detailed eco-profiles (life cycle inventories) of petrochemical PET have been published by PlasticsEurope.<sup>8,9</sup> In our previous studies<sup>10,11</sup> we reported the environmental impacts of recycled PET fibers and bottles made from post-consumer PET bottle waste using various allocation methods for open-loop recycling. The comparison among petrochemical PET, partially bio-based PET (from now onwards 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 fibers and bottles). PLA (polylactic acid, or polylactide) and man-made cellulose fibers 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 2011, approximately 0.225 Mt of PLA production capacity was installed worldwide.<sup>12,13</sup> It is expected that over 0.47 Mt will be installed by 2013.<sup>12</sup> Like PET, PLA can be used for both fiber- and bottle-making. Man-made cellulose fiber is regenerated cellulose made from wood pulp (less commonly, cotton linter is also a raw material to make cellulose fibers<sup>14,15</sup>). The worldwide production in 2007 was

approximately 3.5 Mt p.a.<sup>16</sup> representing the third most important commodity fiber following polyester and cotton.

In order to compare the polymers mentioned, in this paper we use publicly available LCA studies for polymers in primary form (polymer granulates; exception: man-made cellulose fibers). The environmental impacts considered are life cycle non-renewable energy use (NREU) and greenhouse gas (GHG) emissions.

In this paper, we focus on man-made polymers (made from both petroleum and biomass). Cotton, a natural polymer and one of the principal fibers in the market, is excluded. Cotton is neither energy intensive nor does it cause a large amount of GHG emissions.<sup>15</sup> Instead, the environmental issues of cotton are mainly related to ecotoxicity caused by herbicides and pesticides, eutrophication caused by fertilizer use and soil salinization caused by water consumption.<sup>15</sup> These are relatively less worrying issues for man-made polymers.<sup>15</sup> Furthermore, we limit ourselves to the most important types of synthetic fibers and exclude for example, nylon and acrylic.

## System definition

### Functional unit, system boundary, and product systems

The typical applications of PET polymer, PLA polymer and man-made cellulose fibers are shown in Table 1. In this review, the LCAs of three products are compared: polymer granulate, fiber, and bottle. Three functional units are accordingly defined:

- 1 kg of polymer granulates, amorphous grade
- 1 kg of staple fiber
- 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

**Table 1. Typical applications of PET, PLA and man-made cellulose fibers.**

Sector	PET <sup>2,3</sup>	PLA <sup>4</sup>	Man-made cellulose fibers
Packaging	32%	70%	
Injection moulding	1%	1%	
Agricultural films		1%	
Textiles and nonwovens	67%	28%	100%

stages: cradle-to-factory-gate and post-consumer waste management. We assume that the products (i.e. polymer, fiber 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. 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.<sup>6</sup> According to the current EU policy, landfilling will be soon phased out by legislation.<sup>17,18</sup> In many EU countries, such as the Netherlands, Germany, and Switzerland, landfilling of combustible waste is strictly prohibited.

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

#### Petrochemical PET

PET products (i.e. polymer granulates, fiber and bottle) that are made from petroleum feedstock, used once and disposed of in an MSWI facility with energy recovery. This is the reference system.

#### 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 produced by catalytic dehydration of bio-based ethanol. We assume that the bio-based ethanol is 60% maize-based and 40% sugarcane-based, approximately, representing the share of the two biggest producers of bio-based ethanol, the USA and Brazil, which use maize and sugarcane as the feedstocks.<sup>19</sup> The production of the two countries accounted for nearly 90% (the US 55% and Brazil 35%, resulting the ratio of 60:40) of the world bio-ethanol production in 2009.<sup>19</sup>

#### Recycled PET

Petrochemical PET bottles that are used once, recycled into PET polymer, fiber, or bottles, used again, and disposed of in an MSWI plant with energy recovery. Recycled PET granulates (polymer) and fibers are made from either 100% recycled pellets, or from a blend of virgin and recycled PET pellets. Table 2 shows the recycled content for polymers in primary form, fibers, and bottles; these products are

**Table 2. Recycled content (in weight %) delivered by the product systems *Recycled PET* and *Recycled bio-based PET* based on different allocation methods applied for open-loop recycling (see Section 2.2 for the description of the allocation methods).**

Allocation method	“Cut-off”	“Waste valuation”	“System expansion”
Recycled polymer	100%	100%	90%
Recycled fiber	100%	100%	90%
Recycled bottle	35% <sup>a</sup>	35% <sup>a</sup>	25% <sup>b</sup>

<sup>a</sup>A recycled bottle contains maximum 35% of recycled PET pellets; the remaining 65% is composed of virgin PET. See more detailed explanation in the text.

<sup>b</sup>Based on the “system expansion” method, per functional unit (1 kg of bottle) contains 0.28 kg of PET bottles from the first life (100% virgin) and 0.72 kg of PET bottle from the second life, which is made from 35% of recycled PET (0.25 kg) and 65% of virgin PET (0.47 kg). See detailed description in Section 2.2 and Figure 1b.

studied for both production from recycled petrochemical PET and from recycled bio-based PET. For polymer pellets and fiber, about 0.9 kg of recycled PET is delivered from the second life; about 0.1 kg of virgin PET is used to make up the PET loss during the recycling process. This ‘0.1 kg of loss’ is determined by the PET recycling efficiency (Fig. 1).<sup>11</sup> Moreover, according to the current commercial practice, the maximum share of recycled pellets used in a recycled PET bottle is 35%;<sup>11,20</sup> otherwise the level of discoloration becomes unacceptable.<sup>20,21</sup>

#### Recycled bio-based PET

Bio-based PET bottles that are used once, recycled into PET polymer, fiber, or bottles, used again, and disposed of in an MSWI plant with energy recovery.

#### PLA

PLA products contain 50% maize-derived PLA produced in the USA (i.e. by NatureWorks LLC) and 50% sugarcane-derived PLA produced in Thailand (i.e. by PURAC). NatureWorks and PURAC are the two companies which have large-scale PLA production today.<sup>4</sup> The actual production volumes of the two companies are unknown. Thus the LCA results which we present in this paper are based on the assumption of equal production shares of these two companies. It is assumed that the one-way PLA products are disposed of in an MSWI plant with energy recovery.

### Viscose

Viscose fiber is produced from wood pulp in an integrated plant and a non-integrated plant, used once and disposed of in an MSWI plant with energy recovery. The differences between an integrated and a non-integrated plant can be described as follows: in an integrated pulp/fiber plant, the process energy and material efficiencies are highly optimized. Most importantly, recovered bio-energy in the form of 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 plant (i.e. in the case of 'separate production'<sup>15</sup>), this is not possible because the pulp mill and the fiber plant are separately located. The process energy use of the pulp mill is partly covered by the recovered biomass energy (thick liquor) and partly by fossil fuels. Furthermore, the process energy of the fiber plant is entirely covered by local grid power and fossil fuels. A non-integrated production does not have by-products such as xylose, furfural, and acetic acid.

### Modal

Modal fiber is produced from wood pulp in an integrated plant, used once and disposed of in an average MSWI plant with energy recovery. Modal is a type of modified viscose fiber with higher degree of polymerization.<sup>15</sup>

### Tencel

Tencel fiber 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<sup>15</sup>) and the future Tencel (referred to as 'Tencel Austria 2012'<sup>15</sup>) are included. Unlike Viscose and Modal fibers, which are both based on viscose process, Tencel fiber is produced via the novel lyocel process which uses NMMO (N-methylmorpholine-N-oxide) to dissolve cellulose.<sup>15</sup>

For the two environmental impact indicators, NREU and GHG emissions, data are more readily available than for other impact indicators, therefore allowing to compare a large number of products. NREU is the cumulative non-renewable primary energy demand, including fossil fuels and nuclear energy.<sup>22</sup> Cumulative fossil fuel demand is a good proxy of the overall environmental profile of a product.<sup>23</sup> This is because the production of materials and many industrial processes are often energy intensive. However, for

processes which, for example, lead to the release of toxic emissions, energy provides an incomplete picture and other environmental impacts categories should be added.

GHG emissions are a proxy for the impact on climate change. In this paper, we compare GHG emissions calculated based on the global warming potential for a period of 100 years.<sup>24</sup> For bio-based materials (bio-based PET, PLA, and man-made cellulose fibers), the biogenic carbon embedded in the product is accounted for as negative GHG emissions from cradle to factory gate.<sup>25</sup> Consequently, in the grave stage, the biogenic CO<sub>2</sub> released from the combustion of the product is added to the cradle-to-grave GHG emissions. None of the LCA studies reviewed analyzed direct or indirect LUC (land-use change). Currently, there are still difficulties to include indirect LUC in an LCA because of limited data sources and large uncertainties related to the indirect effect of land use. Nevertheless, we tried to include indirect LUC for (partially) bio-based PET based on the current best available data.

### Allocation methods applied to open-loop recycling

In the case of open-loop recycling we are faced with one of the classical challenges of LCA, i.e. allocation.<sup>26–28</sup> In the context of open-loop recycling, the prime question is whether the recycled product should bear a part of the environmental impacts caused by the production of the virgin material (cradle)\* and if so, how the allocation should be implemented. This can be done with various methods.<sup>26</sup> Based on the literature review, three allocation methods were applied:<sup>10</sup>

- 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 (i.e. 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

\*The argument in favor of this approach is that recycling is only possible due to the production of the virgin material and hence should share part of the burden.

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, this approach was only applied for the LCA of recycled PET fibers.<sup>10</sup>

- The 'system expansion' method. This method applies the 'system expansion' principle (ISO14044:2006 Clause 4.3.4.2<sup>29</sup>), taking into account the entire life cycle and not allocating any environmental burden between the first and the second lives.<sup>10,11</sup> The method is illustrated in Fig. 1. Figure 1(a) shows the bottle-to-fiber recycling system: it is assumed that 1000 g of PET bottles produced from the first life, is recycled into 900 g of fiber in the second life (90% efficiency); to compensate the loss of recycling, 100 g of virgin PET fiber is added to the product system. Thus, the total output of the recycling system is 1000 g of bottle (from the first life) and 1000 g of fiber (900 g from the second life and 100 g added to compensate). In the reference system (still in Figure 1(a)), it is assumed that 1000 g of virgin polymer is produced, used, and disposed of. Since the reference system has exactly the same output as the recycling system, the two systems are comparable. Furthermore, because the production and the MSWI of virgin PET bottle (1000 g) are identical in both reference and recycling systems, the two unit processes can be excluded from the scheme, resulting in the FU of '1 kg of PET fiber' for both systems.

Figure 1(b) shows the bottle-to-bottle recycling system (FU = 1 kg bottle). Here, in the first life, 280 g of virgin PET are produced and converted to bottles. In the second life, these bottles (280 g) are recycled into 252 g of granulates (90% efficiency). These 252 g of recycled PET pellets are then mixed with 468 g of virgin PET. The blend is moulded into 720 g of recycled PET bottle (which contains 35% of recycled PET content). The total amount of PET bottle delivered from this product system is 1000 g (280 g from the first life and 720 g from the second life). The recycled content of the 1000 g of PET bottle is only 25% (=35% \* 720 g/1000 g; Table 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 3

shows the summary of the LCAs reviewed and the data and assumptions we used for our calculations.

## Results

### The ranges

Figures 2 and 3 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;<sup>9</sup> the upper range (marked with a dashed line) represents the results based on the previous ecoprofiles of PET.<sup>8</sup> The results of the two publications indicate that the environmental impacts caused by the production of petrochemical PET in western Europe have been substantially reduced over the last years, i.e. by 17% for NREU and by 38% for GHG emissions (Table 3). According to PlasticsEurope, the difference between the old and new eco-profiles originates from the improved PTA process.<sup>9</sup>

The resulting range of values for bio-based PET is comparatively small in both figures because bio-based EG accounts for only approximately one-third of the total mass of PET while PTA represents two-thirds. The lower values refer to sugarcane-based PET and the higher values refer to maize-based PET. For PTA, we used the new eco-profile<sup>9</sup> for the calculations, thereby assuming more efficient PTA production. The inventory data for the production of sugarcane-based ethanol were obtained from 40 plants in Brazil;<sup>30,43</sup> LCA data on maize-derived ethanol were based on the assumption that the bio-ethanol complies with the EU Directive 2009/28/EC,<sup>44</sup> according to which 35% of GHG emission savings should be by bio-ethanol compared to petrol. According to Chen and Patel,<sup>30</sup> currently only advanced maize-based ethanol production facilities meets the requirements of the Directive. For PLA, the lower values are sugarcane-based and the higher values are maize-based.

For Viscose fiber, the ranges are large in both Figs 2 and 3. The lower values are the LCA results of Viscose Austria<sup>15</sup> and the higher values are the LCA results of Viscose Asia.<sup>15</sup> Viscose Austria represents the production in the integrated pulp/fiber plant in Austria; Viscose Asia stands for a non-integrated fiber production in Asia. For Tencel fibers, the higher values refer to the product Tencel Austria,<sup>15</sup> which

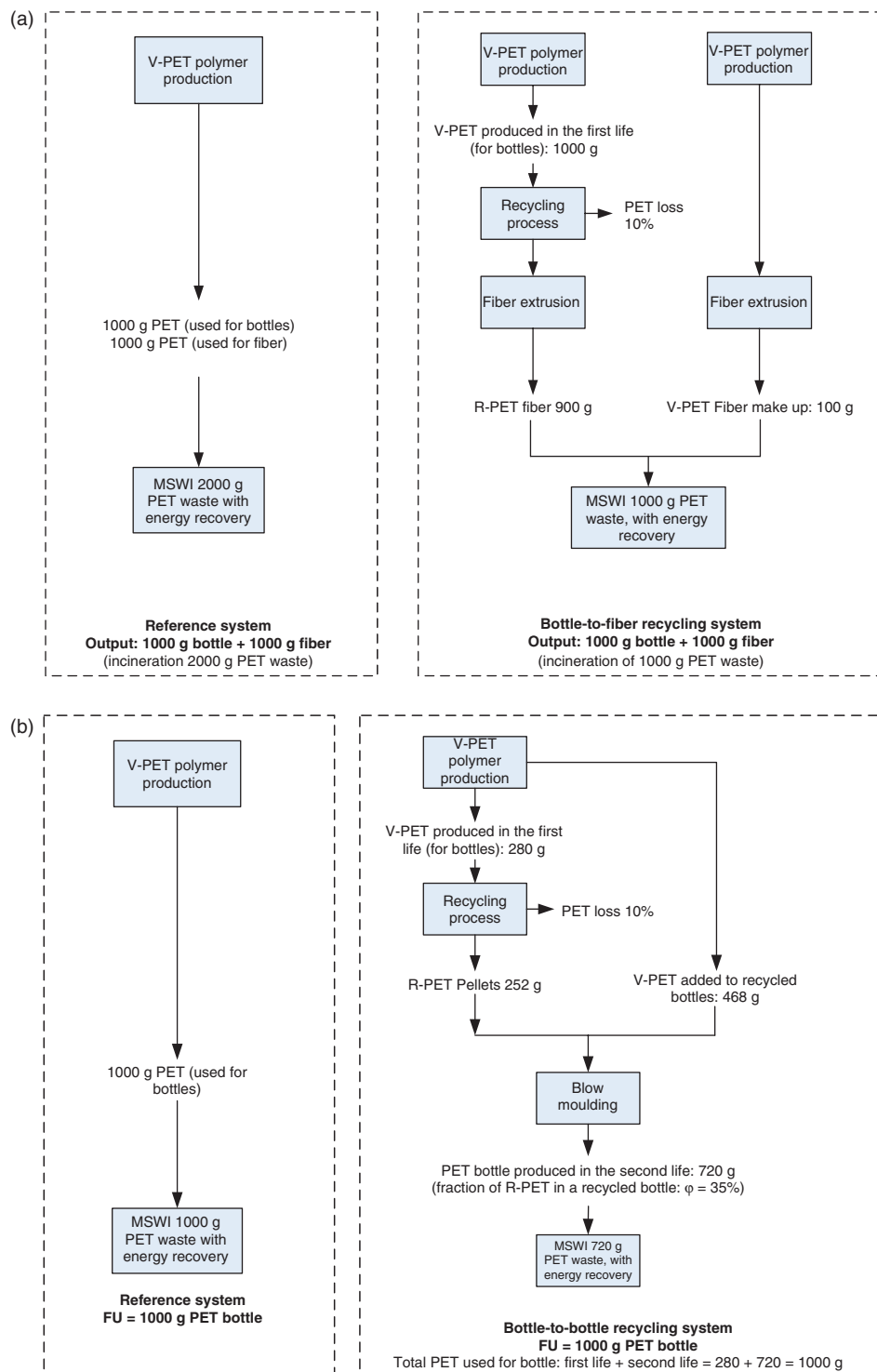


Figure 1. Applying the 'system expansion' method to PET bottle-to-fiber recycling (a) and PET bottle-to-bottle recycling (b). V-PET: virgin PET; R-PET: recycled PET. (a) PET bottle-to-fiber recycling: bottle waste recycled into fiber with 90% material efficiency; recycled fiber is made from 90% recycled PET pellets from the first life. Figure adapted from Shen *et al.*<sup>10</sup> (b) PET bottle-to-bottle recycling: bottle waste recycled into pellets with 90% material efficiency; recycled bottle contains 35% of recycled PET pellets from the first life. Figure adapted by Shen. *et al.*<sup>11</sup>

**Table 3. 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 <sup>9</sup> is used as the default value. The former eco-profiles of PET (in parentheses) <sup>8</sup> 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 <sub>2</sub> eq./kg	
B Bio-based PET, cradle-to-factory gate amorphous grade			Based on 40% sugarcane-derived ethanol from Brazil and 60% maize-derived ethanol from the US. <sup>30a</sup> 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 <sub>2</sub> eq./kg	
NREU (sugarcane-based)	51	MJ/kg	
GWP100a (sugarcane-based)	1.03	kg CO <sub>2</sub> 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). <sup>31,32</sup>
GWP 100a (Ingeo 2009)	1.30	kg CO <sub>2</sub> eq./kg	
NREU (PURAC PLLA)	31	MJ/kg	PURAC (sugarcane-based). <sup>33</sup>
GWP 100a (PURAC PLLA)	0.50	kg CO <sub>2</sub> eq./kg	
D Energy use of SSP (solid state polymerisation)	0.15	kWh/kg	Based on. <sup>8,34</sup> Process energy for polymer conversion was assumed to be identical for PLA, PET and recycled PET.
E Energy use of blow moulding process	2.1	kWh/kg	Based on. <sup>34,35</sup> Assumed that PLA requires 50% of the process energy compared to PET and recycled PET. <sup>b</sup>
F Energy use of the fiber extrusion process, converting amorphous PET to 1 kg fiber	0.64 5	kWh/kg MJ heat/kg	Based on. <sup>36</sup> Data was cross-checked with industry experts. Assumed that PLA requires 50% of the process energy compared to PET and recycled PET. <sup>b</sup>
G Cradle-to-factory gate, man-made cellulose fibers			Based on. <sup>15</sup> 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. <sup>15</sup>
GWP 100a, Viscose, integrated	-0.25	kg CO <sub>2</sub> eq./kg	
NREU, Viscose, non-integrated	61	MJ/kg	As "Viscose Asia" in. <sup>15</sup>
GWP 100a, Viscose, non-integrated	3.81	kg CO <sub>2</sub> eq./kg	
NREU, Modal	25	MJ/kg	As "Modal" in. <sup>15</sup>
GWP 100a, Modal	0.03	kg CO <sub>2</sub> eq./kg	
NREU, Tencel (current)	42	MJ/kg	As "Tencel Austria" in. <sup>15</sup>
GWP 100a, Tencel (current)	1.11	kg CO <sub>2</sub> eq./kg	
NREU, Tencel (future)	21	MJ/kg	As "Tencel Austria 2012" in. <sup>15</sup>
GWP 100a, Tencel (future)	0.05	kg CO <sub>2</sub> eq./kg	
H Cradle-to-factory gate recycled PET polymer (amorphous granulate)			Reproduced based on data from. <sup>11</sup>
NREU, cut-off	9.5	MJ/kg	Same values for both <i>Recycled PET</i> and <i>Recycled bio-based PET</i> .
GWP 100a, cut-off	1.01	kg CO <sub>2</sub> eq./kg	
I Cradle-to-factory gate recycled PET fiber			Calculated based on. <sup>11,36</sup> Same values for both <i>Recycled PET</i> and <i>Recycled bio-based PET</i> .
NREU, cut-off	22	MJ/kg	
GWP 100a, cut-off	1.70	kg CO <sub>2</sub> eq./kg	
NREU, cut-off	13-23	MJ/kg	Data ranges reported by <sup>10</sup> based on the production of two recycling companies.
GWP 100a, cut-off	0.96-1.88	kg CO <sub>2</sub> eq./kg	
NREU, waste valuation	40-49	MJ/kg	
GWP 100a, waste valuation	2.03-2.95	kg CO <sub>2</sub> 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 100a, cut-off, petrochemical virgin PET	3.08	kg CO <sub>2</sub> eq./kg	
NREU, cut-off, bio-based virgin PET	63	MJ/kg	Calculated from B, D, E and H.
GWP 100a, cut-off, bio-based virgin PET	2.55	kg CO <sub>2</sub> eq./kg	



**Table 3. Continued**

Parameters	Value	Unit	Source and notes
K Cradle to grave (without use phase) recycled PET polymer, fiber and bottle, applying system expansion			
NREU, recycled petrochemical polymer	15	MJ/kg	Calculated based on data from A, D, E and. <sup>10,11</sup>
GWP100a, recycled petrochemical polymer	1.11	kg CO <sub>2</sub> eq./kg	
NREU 100a, recycled bio-based polymer	14	MJ/kg	Calculated based on data from B, D, E and. <sup>10,11</sup>
GWP 100a, recycled bio-based polymer	1.03	kg CO <sub>2</sub> eq./kg	
NREU, recycled petrochemical fiber	23–33	MJ/kg	Data ranges reported by <sup>10</sup> based on the production of two recycling companies.
GWP 100a, recycled petrochemical fiber	1.33–2.21	kg CO <sub>2</sub> eq./kg	
NREU, recycled petrochem. fiber	28	MJ/kg	Calculated based on data from A, F and. <sup>10,11</sup>
GWP 100a, recycled petrochem. fiber	1.80	kg CO <sub>2</sub> eq./kg	
NREU, recycled bio-based fiber	27	MJ/kg	Calculated based on data from B, F and. <sup>10,11</sup>
GWP 100a, recycled bio-based fiber	1.72	kg CO <sub>2</sub> eq./kg	
NREU, recycled petrochem. bottle	66	MJ/kg	Calculated based on data from A, D, E and. <sup>10,11</sup>
GWP 100a, recycled petrochem. bottle	4.13	kg CO <sub>2</sub> eq./kg	
NREU, recycled bio-based. bottle	58	MJ/kg	Calculated based on data from B, D, E and. <sup>10,11</sup>
GWP 100a, recycled bio-based. bottle	3.47	kg CO <sub>2</sub> 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. <sup>37c</sup>
M Gross calorific values of polymers			
PET	23.1	MJ/kg	<sup>38</sup>
PLA	19	MJ/kg	<sup>39</sup>
Man-made cellulose fibers	~15	MJ/kg	<sup>15</sup>
N Electricity and heat production	Assumed to be the average EU electricity mix <sup>d</sup> and average European heat from industrial furnace (>100 kW) with low NOx. LCA data obtained from the Ecoinvent database Version 2.0. <sup>40</sup>		

<sup>a</sup>A recent publication from Tabone and colleagues<sup>41</sup> applied both LCA and the Green design principles to 12 polymers, among which there is (partially) bio-based PET. This study was heavily criticized for its scope of comparison, its LCA allocation method and its use of the single score approach.<sup>42</sup> 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.

<sup>b</sup>The life cycle inventory data of processing PLA are not publicly available. This assumption is an estimate based on the heat capacities ( $c_p$ ) and the drying and preforming temperatures of PLA and PET.

<sup>c</sup>This means that 1 GJ of waste yields 0.106 GJ<sub>e</sub> (electricity) and 0.223 GJ<sub>th</sub> (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<sub>e</sub> replaces 0.106/30% = 0.35 GJ<sub>p</sub> primary fossil fuels and 0.223 GJ<sub>th</sub> replaces 0.223/85% = 0.26 GJ<sub>p</sub> fossil fuels. The total primary fossil fuel that can be avoided is 0.35 GJ<sub>p</sub> + 0.26 GJ<sub>p</sub> = 0.61 GJ<sub>p</sub> – this is approximately 60% of the energy content of the waste.

<sup>d</sup>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. UCTE is the 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 the Nordic countries power association, including Denmark, Norway, Finland and Sweden. CENTREL stands for Central European power association, including Czech Republic, Hungary, Poland and Slovakia.

represents state-of-the-art Tencel production; the lower values represent the LCA results of Tencel Austria 2012,<sup>15</sup> which uses the energy recovered from municipal waste incineration for process heat and power. Modal fiber is exclusively produced in the integrated pulp/fiber plant in Austria. No ranges were calculated in earlier studies.

For the recycled systems, namely recycled PET and recycled bio-based PET, the new eco-profile of PET is used for the calculation (Table 2). The ranges reported here are large for polymers and fibers (not for bottles). They originate

primarily from the different allocation methods used for open-loop recycling.

The value ranges for the recycled bottles (for both NREU and GHG emissions) are small compared to the value ranges of recycled polymer granulates and fibers, because a recycled bottle contains only 35% recycled polymer and the remaining 65% is virgin polymer (petrochemical or bio-based). The overall impact of recycled PET bottle is therefore largely determined by the production of the virgin polymer. The impact reduction by recycling is lower than one may expect.

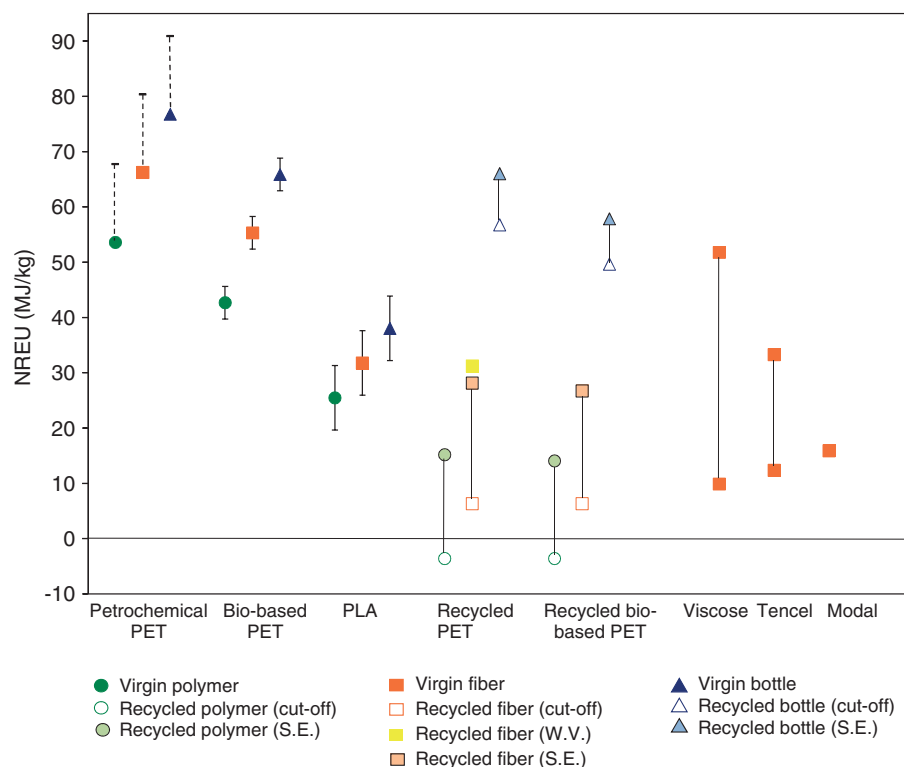


Figure 2: Comparison of cradle-to-grave NREU of partially and fully bio-based polymers in various forms (polymer granulates, fibers and bottles). Note: “grave” is MSWI with energy recovery, the use phase is excluded. The uncertainty ranges are explained in the text. S.E. stands for the “system expansion” method. W.V. stands for the “waste valuation” method. See the description of the methods applied for open-loop recycling in Section 2.2.

### The comparison

Across all types of functional units the rankings for polymer granulates, fibers, and bottles are identical in both Fig. 2 and Fig. 3: the impacts are the lowest for polymers, followed by fibers, and finally bottles.

In Fig. 2, the NREU of polymer and fiber 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, which can hence be considered as robust. When the ‘cut-off’ method is applied, fibers made from recycled PET and recycled bio-based PET both have a lower NREU than integrated produced Viscose fiber (the lower value for Viscose). When the system expansion or waste valuation method is chosen, the two recycled fibers have a higher NREU than the man-made cellulose fibers produced in the integrated plant (Modal and the lower value for Viscose) and also compare to future Tencel fiber (i.e. the lower of the two values for Tencel). If

produced in a non-integrated plant, man-made cellulose fiber is comparable with bio-based PET (comparing the higher value for Viscose and bio-based PET), or PLA (comparing the higher value for Tencel with PLA). The NREU of bottles is the highest for petrochemical PET and the lowest for PLA. Regardless of the choice of the allocation methods, the NREU of recycled bio-based PET bottle is lower than those of bio-based PET and recycled PET, but higher than the NREU of PLA. The NREU of recycled PET bottles is particularly strongly influenced by the choice of the allocation method. It is either lower than (based on the cut-off method) or comparable to (based on the system expansion method) the NREU of bio-based PET.

Like the results of NREU, in Fig. 3 the GHG emissions of polymer and fiber are the highest for petrochemical PET, followed by bio-based PET. The rankings of recycled PET, recycled bio-based PET, and PLA depend on the allocation method chosen. Viscose fiber produced in the integrated

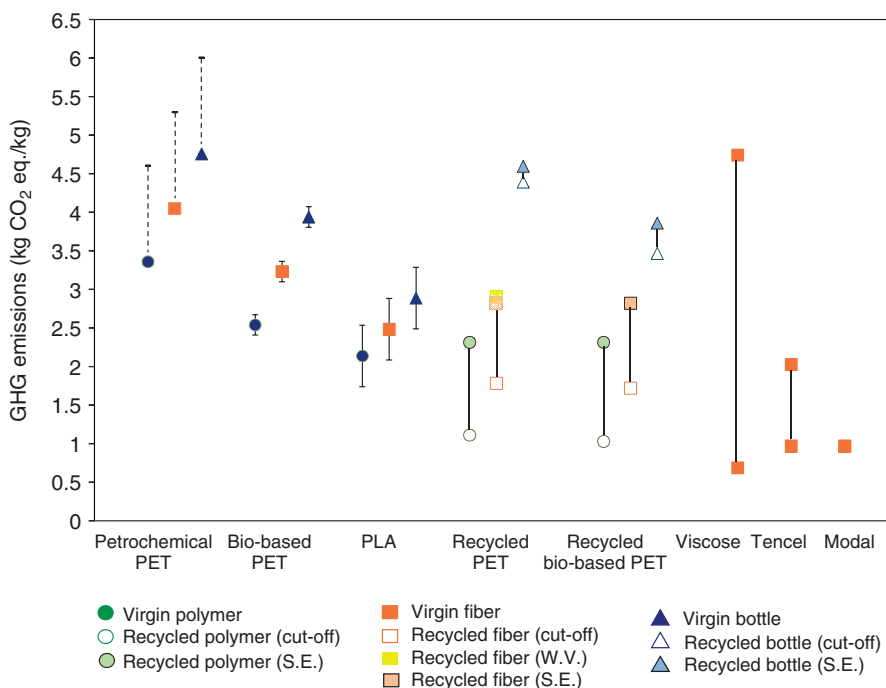


Figure 3: Comparison of cradle-to-grave GHG emissions of partially and fully bio-based polymers in various forms (polymer granulates, fibers and bottles. Note: “grave” is MSWI with energy recovery, the use phase is excluded. The uncertainty ranges are explained in the text. S.E. stands for the “system expansion” method. W.V. stands for the “waste valuation” method. See the description of the methods applied for open-loop recycling in Section 2.2.

plant shows the lowest GHG emissions among all fibers. Viscose fiber produced in a non-integrated plant has the highest GHG emissions of all fibers. The GHG emissions of 1 kg of bottles are highest for petrochemical bottles, followed by recycled PET, bio-based PET, and bio-based recycled PET. PLA bottle has the lowest GHG emissions. The choice of the system expansion method or the cut-off method does not have strong influence on the results for bottle.

In Figs 2 and 3, when recycled PET and recycled bio-based PET for polymers and fibers are compared, the ranking of the NREU and GHG emissions of the two systems are identical based on the cut-off method: here, the environmental impacts are caused by the recycling process and post-consumer MSWI; while the first life (virgin PET production) has no influence in this method. 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 Fig. 1 (the principle originates from Shen *et al.*<sup>10,11</sup>) the recycled system delivers 0.9 kg of recycled

PET (determined by the material efficiency of PET 90%<sup>11</sup>) 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 for the 0.1 kg of virgin PET (petrochemical vs bio-based).

### The allocation methods

In terms of NREU (Fig. 2), the cut-off approach leads to lower NREU because of the energy credits from MSWI (when applying the cut-off method the NREU of the virgin PET from the first life is excluded from the recycled product system). Both the waste valuation and system expansion methods lead to relatively high NREU compared to the cut-off method because less end-of-life waste is incinerated in the recycled product systems and therefore, fewer energy credits are obtained. In contrast, in terms of GHG emissions (Fig. 3), the system expansion method leads to lower GHG emissions due to less waste incinerated; the cut-off

method leads to higher GHG emissions because of the GHG emission from MSWI (even with the emission credits from energy recovery).

## Discussion

### Functionality

The results indicate that both recycling and bio-based alternatives are important ways of reducing the energy requirements and GHG emissions of material consumption. However, not all functionalities of the materials studied are strictly comparable, even though comparable products can be made from various materials.

#### Bottles: PLA vs PET

PLA has different material properties compared to PET. For example, the water gas-barrier property of PLA is weaker than that of PET,<sup>45,46</sup> and therefore, PLA bottles can only be used for non-carbonated drinks which have a limited shelf time. Furthermore, because of the different material properties, bottles made from PLA and PET are different in terms of thickness, resulting in different polymer mass required per functional unit. For example, a 500-ml water bottle made from PET weighs between 10 g and 20 g based on the technology level in the 2000s.<sup>47</sup> For a PLA bottle which offers the same function, NatureWorks suggested a bottle weight of 22 to 25 grams.<sup>46</sup>

In our default analysis, we assumed that bottles made from PLA and PET have the same weight (1 kg). Under this assumption, the PLA bottle has the lowest NREU and GHG emissions of all bottles studied (Figs 2 and 3). In Table 4, we present the sensitivity analysis for the NREU assuming different weights of the PLA bottle. If the weight of the PLA bottle is twice of the weight of a PET bottle, there would be little advantage of PLA compared to virgin PET (i.e. 1% reduction in NREU).

**Table 4. Reduction in cradle-to-grave NREU (excluding the use phase) of PLA bottle; the reference system: Petrochemical PET bottle.**

	NREU reduction of PLA
PLA has the same weight as PET (default)	50%
PLA is 10% heavier than PET	46%
PLA is 70% heavier than PET	16%
PLA is 100% heavier than PET	1%

#### PET: the recycled and the virgin

In general, recycled plastics offer a more limited range of applications compared to the virgin plastics. For instance, recycled PET fiber cannot be used to produce high performance textile products (e.g. with moisture management).<sup>10</sup> Another restriction of recycling is that fiber (both virgin and recycled) cannot be further recycled via mechanical recycling.<sup>10</sup> A more comprehensive discussion regarding the functional equivalence between recycled and virgin PET can be found in Shen *et al.*<sup>10</sup> For PLA recycling, there are still technological difficulties which need to be resolved; for example, polymer degradation during mechanical recycling. Although chemical recycling of PLA is technically feasible,<sup>48</sup> it has not been realized on an industrial scale so far.

#### Fibers: man-made cellulose vs polyester

Man-made cellulose fibers have different properties compared to polyester fibers (PET and PLA) in many aspects, for example density, dry tenacity, wet tenacity, water retention, dyeability,<sup>15</sup> which lead to different applications in the textile and the non-woven sector.

A last remark on functionality: 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, (partially) bio-based PET and petrochemical PET are truly functionally equivalent.

#### Indirect land-use change (ILUC)

The LCAs of man-made cellulose fibers used in this study reported that no direct LUCs (land-use changes) occurred during the wood production; the LCAs of bio-based PET and PLA do not discuss any LUCs. If either direct LUC or indirect LUC (ILUC) occurs in the biomass production, the GHG emissions of the bio-based materials will nearly certainly increase. In industrialized countries, direct LUC occurring after January 1, 1990 is unlikely. However, accounting for ILUC would increase the GHG emissions of bio-based products.

ILUC is not included in the PAS 2050 guidelines because the method and data required for the assessment are 'not fully developed'.<sup>25</sup> Nevertheless, in this study we made an attempt to estimate the possible effect of ILUC for bio-based PET, using the ILUC data published by CARB (Californian

**Table 5. GHG emissions of (partially) bio-based PET made from maize and sugarcane, taking into account indirect land use change. Own calculation based on the data obtained from.<sup>30,51</sup>**

1 kg of amorphous PET polymer	Cradle-to-factory gate (kg CO <sub>2</sub> eq.)	Cradle-to-grave (excluding the use phase, kg CO <sub>2</sub> eq.)
Bio-based, from maize, w/o ILUC <sup>a</sup>	1.36	2.67
Bio-based, from maize, with ILUC	1.65	2.96
Bio-based, from sugarcane, w/o ILUC	1.03	2.34
Bio-based, from sugarcane, with ILUC	1.47	2.78
Petrochemical PET	2.05	3.36

<sup>a</sup>w/o = without.

Air Resources Board). There are several models and ongoing projects to determine the ILUC of bio-fuels.<sup>49,50</sup> We use the CARB data because these are the first (and so far the only) government-adopted ILUC emission factors.

According to CARB, the GHG emissions caused by ILUC are 30 grams CO<sub>2</sub> eq./MJ maize-ethanol and 46 grams CO<sub>2</sub> eq./MJ sugarcane-ethanol.<sup>51</sup> Combining the CARB data and the bio-based PET production data from Chen and Patel,<sup>30</sup> (also Table 3), we re-calculated the GHG emissions of bio-based PET. The result is shown in Table 5. As expected, the GHG emissions of bio-based PET increases when ILUC is accounted for. From cradle to factory gate, the GHG emissions of bio-based PET increases by 21% (maize-based) to 42% (sugarcane-based); from cradle to grave, the GHG emissions increases by 11% (maize-based) to 19% (sugarcane-based). Based on the CARB ILUC emission factors, (partially) bio-based PET can still offer GHG emission savings compared to petrochemical PET. It should be reminded that this conclusion is made based on one set of ILUC data. More comprehensive research is required in order to draw a robust conclusion.

### Others aspects

In this paper, we aimed to review as many options for plastics and fiber materials as possible. However, some potentially important options had to be omitted. For instance, multiple recycling trips can potentially further reduce the environmental impact (according to Shen *et al.*,<sup>11</sup> by a maximum of six percentage points). Also the PET bottle-to-bottle recycling

technology is likely to be further improved. It can be calculated that from the use of 100% of recycled PET, instead of 35%, would further decrease the impacts of a recycled bottle (both NREU and GHG emissions) by 8 percentage points in the case of recycled bio-based PET and 12 percentage points in the case of recycled PET. Last but not least, novel bio-based polymers such as PLA and bio-based PET are still in their early stages of commercialization. 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<sup>52,53</sup> for PLA and furanic feedstocks for PET.<sup>54</sup> These new feedstocks and routes may further decrease the environmental footprint of bio-based materials, making them more competitive with recycled petrochemical polymers.

## Summary and recommendations

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

The NREU of polymers and fibers are highest for petrochemical PET, followed by bio-based PET, PLA, recycled PET and recycled bio-based PET. The GHG emissions of polymers and fibers are highest for petrochemical PET. Bio-based PET has higher GHG emissions than PLA, recycled PET, and recycled bio-based PET. The man-made cellulose fibers produced in integrated plants have the lowest GHG emissions of all fibers studied. The NREU and GHG emissions of bottles are highest for petrochemical PET. Bottles made from bio-based PET have higher impacts than bottles made from PLA. Bottles made from (partially) 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, they are considered robust conclusions.

However, the rankings for fibers and 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. For the recycled product systems, 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 NREU and GHG emissions. The wide range of results due to the choice of the allocation method does not cause problems when comparing recycled product systems with the virgin petrochemical product system. However, when the recycled are compared with the bio-based, 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<sup>10</sup> the three allocation methods used also in this paper were 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 policymakers should establish clear rules and procedures in order to support the decision-making for recycled products.

In this study, only NREU and GHG emissions were reviewed. Further methodological and empirical research on the impacts related to water use, LUC, ILUC, and biodiversity is urgently required.

For bio-based polymer producers, we recommend that efforts be made in process optimization, process integration, and the utilization of biomass for process energy (e.g. the biorefinery concept). The importance of these opportunities are demonstrated by the LCA results for man-made cellulose fibers. 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 reduction of the environmental impacts related to our material consumption will be achieved in the future.

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