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Environmental Impacts of End-of-Life Options of Biobased and Fossil-Based Polyethylene Terephthalate and High-Density Polyethylene Packaging

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Abstract: Plastic waste production increasingly causes environmental pollution. However, end-of-life (EoL) research often lacks detail and timeliness and fails to integrate the end-of-life option into a product's life cycle in a systemic perspective. This study addresses these knowledge gaps, by applying an improved anticipatory consequential life cycle assessment (LCA) approach. Reuse, mechanical and chemical recycling options were compared for (biobased and fossil-based) high-density polyethylene (HDPE) and polyethylene terephthalate (PET) plastic shampoo bottles in the European context using three types of impact categories: climate change, fossil resource scarcity and mineral resources scarcity. The completeness and detail of EoL were increased by modelling the polymer reprocessing within the collection system including all transport distances, while timeliness was improved by implementing the data applicable for the time of implementation of EoL options in the future. The results show that the reuse option has the largest benefits on climate change impact, and on fossil and mineral resource scarcity for both HDPE and PET, for both biobased and fossil plastics. Furthermore, all EoL options cause a net reduction in all climate change, fossil and mineral resource scarcity thanks to the avoided impact of virgin plastic. Finally, the improved LCA approach, utilized in this study, includes plastic production, use and EoL in one assessment, and thus can provide valuable information for adjusting policy and regulations for plastic manufacturers in their production of new virgin plastic polymer, as it requires alignment with its use and EoL options.

Keywords: plastic; anticipatory LCA; life cycle assessment; HDPE; PET; recycling; end-of-life

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Citation: Helmes, R.J.K.; Goglio, P.; Salomoni, S.; van Es, D.S.; Vural Gursel, I.; Aramyan, L. Environmental Impacts of End-of-Life Options of Biobased and Fossil-Based Polyethylene Terephthalate and High-Density Polyethylene Packaging. *Sustainability* **2022**, *14*, 11550. https://doi.org/10.3390/su141811550

Academic Editor: Gioele Capillo

Received: 15 July 2022
Accepted: 6 September 2022
Published: 15 September 2022

1. Introduction

The global economy continues to grow: global growth rate has been 2.6% in 2019 and it is projected to be 2.9% by 2022 despite global crises [1], which is accompanied by an increase in waste production. Global solid waste production is expected to reach 3.40 billion tonnes annually by 2050 [2]. Plastics are among the most important material because of their widespread use, and therefore their sustainability is an issue that needs to be addressed [3]. The world production of plastics (including thermoplastics, polyurethanes, thermosets, elastomers, adhesives, coatings and sealants and polypropylene-fibres) reached 368 million tonnes in 2019 of which 16% is produced in Europe [4]. It was estimated that till 2015 6.3 Gt of plastic waste has been produced and the total amount of plastic waste is expected to increase to 12 Gt by 2050 [5]. The largest market of plastics is packaging, the growth of which was accelerated by a global shift from reusable to single-use containers [5]. Due to regulatory efforts, plastic waste discard declined to 55% on average worldwide in 2015 [3].

The most common method for plastic recycling is mechanical recycling, while incineration is a very common plastic waste management [6]. Several other alternative recycling methods have been proposed, including depolymerization [6,7]. Plastic waste incineration contributes to global warming [6], while plastic waste resulted in up to 2% of the global greenhouse emissions in 2018 [7].

In response to growing concerns about climate change and the scarcity of non-renewable resources, the development of biobased plastics has been sought in alternative to fossil-based plastics [8–10]. Biobased plastics can be entirely or partly derived from biomass [11,12]. Biobased plastics are made from biorenewable sources, such as carbohydrate-rich crops such as corn or sugarcane. In 2016, the production capacity of biobased plastics amounted to 2.05 million tonnes [13]. Currently, biobased plastics represent under one percent of the 367 million tonnes of plastic produced annually. The global production level is expected to increase from around 2.41 to 9.20 million tonnes in the short term [14]. Packaging is the largest market segment for biobased plastics, accounting for 48% (1.15 million tonnes) of the total biobased plastics market in 2021. Biobased polyethylene terephthalate (PET), polyethylene (PE) and polylactic acid (PLA) are the main polymers used in the packaging market [14].

Several studies have analysed and estimated the environmental impacts of bioplastics production [15,16], and other studies have focused on the comparison between the impact of the biobased plastic production in relation to their petrochemical counterparts production, highlighting strengths and weaknesses [17–20]. Most literature on biopolymers does not take into consideration their end-of-life (EoL), which, according to several studies, significantly affects the overall life-cycle impacts of biopolymers [3,15]. It is very important to manage the biobased plastics waste disposal to optimise their potential environmental benefits [21,22]. This analysis should be done before biobased plastics are produced and implemented commercially, to enable a minimal impact of these products all along their life cycle [23]. Hence, anticipatory assessments are required.

Several anticipatory life cycle assessments for plastic bottles in Europe have been conducted previously [12], while plastic recycling technologies have been recently reviewed [7,24]. However, it was reported that only 27% of the EoL research studies were peer reviewed and often lacked completeness, detail and timeliness, often only broadly characterizing the EoL scenarios [3]. Completeness and detail are increased by modelling the polymer reprocessing within the collection system including all transport distances. Timeliness is improved by implementing the data applicable for the time of implementation of the end-of-life option in the future (anticipatory approach). The effects of the introduction of the individual end-of-life options are assessed (consequential approach). The effects of the introduction of the individual EoL options are assessed (consequential approach). To achieve the aim of the study, the following objectives are identified: (i) Assess reuse, mechanical and chemical recycling options for (biobased and fossil-based) polyethylene terephthalate (PET) and high-density polyethylene (HDPE) in the European context, with the shampoo bottle as an example; (ii) to identify environmental hotspots; (iii) to assess the environmental benefits of recycling; and iv) identify methodological issues related to anticipatory consequential LCA. A comparative perspective is taken to discuss results, but this study does not aim to claim a preference for a specific end-of-life option. Three environmental impacts are considered: climate change, fossil resource scarcity and mineral resources scarcity.

2. Materials and Methods

2.1. General Approach, Functional Unit, System Boundary

In this anticipatory consequential LCA, a use-to-grave system boundary is adopted including one recycling cycle. The functional unit is chosen so that it represents one of the common uses of biobased PET and PE within the packaging market [12]. The functional unit (FU) of the analysed system is defined as packaging 300 mL shampoo in a bottle with a circular end-of-life (EoL) option. The reference flow is 1 bottle with a weight of 40 g. Based on expert opinions it is assumed that differences in density between PET and PE are offset by applied differences in wall thickness due different strength and gas barrier properties of the materials, resulting in comparable weight for of bottles made from both materials.

The system boundaries of the shampoo bottle end-of-life (EoL) options include: (1) the collection and sorting and (2) recycling or refilling of the plastic after the use stage, (3) production of a shampoo bottle from recycled and virgin plastic, (4) transport to the user home and (5) the final waste processing after the second stage use including collection and incineration.

For each EoL option, some virgin plastic is necessary to replenish losses in the recycling process [12]. The virgin plastic compensates material losses in the entire recycling system and is required for quality requirements of the bottle. This approach is in agreement with several other LCA research works and reflects the consequential research question of introducing a new EoL treatment option in the current system [25,26]. Transport involved in each of these options are taken into account, but the shampoo contained in the bottle, as well as secondary and tertiary packaging, is not included. The consumer use phase and its impacts are also excluded. The geographic boundary covers Western Europe for all the recycling and reprocessing processes.

The study is carried out applying the LCA methodology, according to the ISO14040/14044 standards [27,28]. The model was developed in SimaPro 9.3 [29]. The impact categories considered are climate change, fossil resource scarcity and mineral resource scarcity. The impact assessment was done with the Life Cycle Impact Assessment method ReCiPe 2016 (Hierarchist version, Midpoint method version 1.04) [30].

2.2. LCA Scenario Development

Twelve scenarios covering three EoL options, two plastics and two feedstock sources were developed with assumptions consistent with the anticipatory consequential perspective of this LCA. Even though plastic waste treatment should follow 10R circular economy hierarchy favouring reuse over recycling over incineration [22], the three end-of-life (EoL) options are studied in separate scenarios. For the same reason, incineration is the default EoL option after the second life cycle for all scenarios, while plastic can be recycled a number of times in practice. The two different feedstock sources studied were 100% biobased or 100% fossil. The two different plastics were PET and HDPE. For each scenario, recycled plastic substitutes the corresponding amount of virgin plastic with the same feedstock: if the recycled bottle replaces a fossil plastic bottle, the virgin plastic included in the recycled bottle is also fossil based. This focuses our analysis on the introduction of the EoL option rather than on the replacement of fossil plastics with biobased plastics. Thus, the consequential scenarios analysed in this anticipatory LCA are:

- R-B: the biobased plastic shampoo bottle is reused, replacing an equivalent virgin biobased plastic shampoo bottle for one use (HDPE for R-B-H and PET for R-B-P);
- R-F: the fossil based plastic shampoo bottle is reused replacing an equivalent virgin fossil based plastic shampoo bottle for one use (HDPE for R-F-H and PET for R-F-P);
- M-B: the biobased plastic shampoo bottle is recycled through mechanical recycling replacing an equivalent virgin biobased plastic shampoo bottle for one use (HDPE for M-B-H and PET for M-B-P);
- M-F: the fossil based plastic shampoo bottle is mechanically recycled replacing an equivalent virgin fossil based plastic shampoo bottle for one use (HDPE for M-F-H and PET for M-F-P);
- C-B: the biobased plastic shampoo bottle is recycled through chemical recycling replacing an equivalent virgin biobased plastic shampoo bottle for one use (HDPE for C-B-H and PET for C-B-P);
- C-F: the plastic shampoo bottle, based on the current plastic market mix, is chemically recycled replacing an equivalent plastic shampoo bottle, based on the current market mix, for one use (HDPE for C-F-H and PET for C-F-P).

2.3. End-of-Life Assumptions and Data Sources

Data concerning collection and sorting of all plastics (fossil- and biobased) are based on current European conditions for fossil-based HDPE and PET (Table 1). Data necessary for the mechanical reprocessing and chemical reprocessing all plastics are based on literature research focusing on the fossil plastic recycling (Table 2). When specific HDPE data were not available, data on polyethylene in general have been collected and used instead. Plastic waste created during collection, sorting and recycling is assumed to be transported to the incineration plant and incinerated. After the second life cycle, bottles in each scenario are collected by the waste collection system and taken to the incineration plant. Transport assumptions regarding long distance transport follow Kouloumpis et al.'s (2020) approach, including transport distances for the virgin plastic supply chain [31] (Supplementary Materials). Bottle production with recycled plastic incorporates only 16% of recycled HDPE and for PET only 24%, the majority of the bottle consisting of virgin plastic [12]. Altogether, the most recent available data are used and extrapolated, where applicable, to the moment of implementation of the EoL option.

2.3.1. Reuse Scenarios

In the reuse scenario, the shampoo bottle user brings an empty bottle of shampoo back to the retailer where bottles are washed (optional) and refilled and returns to home with the same or another bottle. This scenario reflects some common reuse models identified by Muranko et al. (2021) [32]. The distance is assumed to be 5 km. The collection efficiency is assumed to be 100%, thus each bottle is brought back to retailer by the consumer (Table 2). Electricity, heat, water and detergent use are conservatively assumed to be the same as for washing a glass bottle [33] (Table 1). The reuse efficiency is assumed to be 99% with 1% of shampoo bottle disposed through incineration (Table 2). Due to this 1% loss in shampoo bottle, reuse is compensated by introducing new virgin plastics of the same type considered in the scenarios [12,31]. For instance, in the scenario R-B-P, the virgin plastic compensating the plastic loss is biobased PET. This reuse scenario applies to all types of plastic bottles in the same way.

2.3.2. Mechanical Recycling Scenarios

The mechanical recycling involves washing, cutting, shredding and extrusion to produce recycled PET and PE flakes [34]. After the use phase, the shampoo bottles are collected through the waste collection system and taken to the recycling facilities where the materials are sorted. The collection efficiency is 77% for PET and 72% for HDPE (Table 2) and the distance between the consumer house and the recycling facility is assumed to be 40 km (truck capacity considered is <10 tons). Once collection is completed, PET and HDPE are sorted with different efficiencies (Table 2). The fuel and energy requirements of sorting are shown in Table 1. Then, plastic waste is transported to the mechanical reprocessing plant with a distance assumed of 100 km (truck capacity considered >20 tons). A distance of 500 km covered by truck to transport the virgin plastic to the reprocessing plant is considered. Bottles are produced from the granulate with the blow moulding process from the ecoinvent database [35]. The bottle is transported to the shampoo production plant to be refilled, then to the retailer and to the consumer house.

2.3.3. HDPE Chemical Recycling

The chemical recycling of HDPE is carried out by pyrolysis which involves thermal degradation in the absence of oxygen [43]. The energy required for this process is estimated considering polyethylene as the only plastic used in the process. One of the products of pyrolysis is ethylene, which can be subsequently used in the polymerization to produce HDPE. Following the repolymerization, a blow moulding process is considered. The impact of the polymerization process was estimated by taking the environmental impact from HDPE production from the Ecoinvent database [35] and subtracting the impact of the fossil-based ethylene monomer from the same data source. The pyrolysis efficiency was set to 21%, derived from Donaj et al. (2012) [39]. The byproducts from the process (methane, ethane, propane, propylene, and butane) were assumed to be separated after the pyrolysis and applied as a substitute for their fossil equivalent chemicals in agreement with the consequential LCA approach [44–46].

Table 1. End-of-Life scenario efficiencies. Data are derived from literature sources [36–40]. PET: Polyethylene Terephthalate, HDPE: High-Density Polyethylene.

Scenario		Collection Efficiency		Sorting Efficiency		Recycling Efficiency	
		Value	**Source**	**Value**	**Source**	**Value**	**Source**
PET	Reuse	1	Assumption	-		0.99	Assumption
	Mechanical	0.77	[36]	0.31	[36]	0.86	[36]
	Glycolysis	0.77	[36]	0.31	[36]	0.87	[37,38]
HDPE	Reuse	1	Assumption	-		0.99	Assumption
	Mechanical	0.72	[36]	0.40	[36]	0.95	[36]
	Pyrolysis	0.72	[36]	0.40	[36]	0.21	[37,39,40]

Table 2. End-of-life process data. Data refer to 1 kg of processed/recycled plastics both for HDPE and PET.

Scenario		Value		Source
Sorting				
Diesel		0.084 MJ kg^{-1}		[34]
Energy		0.122 MJ kg^{-1}		[34]
Reuse				
Caustic soda		0.007 kg^{-1}		[33]
Heat		0.019 kWh kg^{-1}		[33]
Electricity		0.004 kWh kg^{-1}		[33]
Water		0.368 kg kg^{-1}		[33]
Mechanical reprocessing		**PET**	**HDPE**	
Sodium hydroxide		0.002 kg kg^{-1}	-	[34]
Methane		1.89 MJ kg^{-1}	0.52 MJ kg^{-1}	[34]
Electric energy		0.79 MJ kg^{-1}	1.75 MJ kg^{-1}	[34]
Water		2.24 kg kg^{-1}	1.56 kg kg^{-1}	[34]
Chemical reprocessing HDPE				
Heat		2.67 MJ kg^{-1}		Estimated value, see Section 2.3.3
Chemical reprocessing PET				
Heat		0.906 MJ kg^{-1}		Estimated value, see Section 2.3.4
Ethylene glycol		0.5 kg kg^{-1}		[41]
Repolymerization	Methane	1.63 MJ kg^{-1}		[38]
	Electricity	0.7 MJ kg^{-1}		[38]
	Steam	0.94 kg kg^{-1}		[38]
Incineration				
Natural gas		0.00986 MJ kg^{-1}		[42]
Electricity		0.252 MJ kg^{-1}		[42]

2.3.4. PET Chemical Recycling

PET is recycled through a glycolysis process in which the PET polymer is depolymerised using ethylene glycol, producing bis-2-hydroxyethyl terephthalate (BHET), which is then further used in the repolymerization for the production of recycled PET [47]. Due to lack of data regarding energy required for the glycolysis process, its impact was calculated considering the mass of the reagent (ethylene glycol), the specific heat capacity of ethylene glycol and PET and the temperature described in the patent [41] (Table 1). This energy requirement for glycolysis was adjusted to consider the increase in efficiency in the upscaling from lab-conditions. This type of depolymerization was chosen as it is eco-friendly due to the non-toxicity chemicals and products used [48], and it is also a very advantageous technique for its simplicity and flexibility [49]. After the glycolysis process, the repolymerization of PET (Table 1) and blow moulding process were accounted for using the ecoinvent database [35]. The repolymerization process efficiency is 87% (Table 2) [37,38].

2.4. Virgin Plastic Production

The same data sources were used for biobased and fossil-based plastics as much as possible. The impact of PTA production, polymerization to PET and blow moulding were taken from the EcoInvent database [35]. The impact of the polymerization to HDPE for the biobased HDPE production and the impact of the fossil-based PET and HDPE were taken from the same data source. Due to lack of data regarding conversion from HDPE to HDPE bottles, the conversion efficiency from PET to PET bottles was considered [37].

Bio-HDPE and bio-PET production is based on data taken from Liptow and Tillman (2012) [40] and Papong et al. (2014) [37]. The biobased PET bottle production utilises as input biobased MEG and fossil PTA. It is assumed that biobased monoethylenglycol (MEG) and fossil terephthalic acid (TPA) are shipped to the port of Rotterdam, then used in the polymerization and the blow moulding process in Europe. Bio-HDPE is produced in Brazil and then transported to Rotterdam and moulded into bottles. Transport data of both biobased plastics during production until reaching Rotterdam are taken from Liptow and Tillman (2012) [40]; Macedo et al. (2004) [50] and Tsiropoulos et al. (2015) [51].

The same assumptions regarding local transport, transport to the shampoo filling plant and to the retailer are applied to biobased and the fossil-based plastic: bottles of both plastics and feedstock types are transported to shampoo production plant. After filling, the bottles are transported to the retailer for a 500 km distance. Subsequently, the consumer transport from the retailer to the customer house is 5 km for each single trip (Supplementary Materials).

2.5. Contribution, Sensitivity and Uncertainty Analysis

A contribution, sensitivity and uncertainty analysis was carried out considering the three impact categories selected. A contribution analysis was carried out to identify the main processes that affect the overall impact of the EoL options in different scenarios, in agreement with Albers et al. (2019), Lefebvre et al. (2019) and Mendoza Beltran et al. (2018) [52–54].

The sensitivity analysis studied the effect of changes in key variables for both bioplastics: increasing the amount of recycled plastic in recycled bottles to 100% (derived from Nessi et al., (2020) [12]), a 25% lower yield of sugarcane production and a 10% reduction in blow moulding efficiency.

Finally, the uncertainty analysis assesses how key uncertainties in the life cycle inventory can affect the environmental impacts, in agreement with [27,53,54]. The simulation involved the estimated variables listed in this paper and the Supplementary Materials. A Monte Carlo simulation was performed with 10,000 runs for each scenario, with uniform distributions between a predefined minimum and maximum. The uncertainty range for energy input for the conversion of ethanol to ethylene, for pyrolysis and for glycolysis was assumed to be 33%. The spread in all efficiency values was identified from the literature, except for glycolysis and pyrolysis efficiency. In the absence of uncertainty information, the uncertainty range for these reprocessing options was assumed to be 10%. For the processes related to transport, a variation of 10% was considered.

3. Results

3.1. Scenario Performance and Contribution Analysis

The environmental impacts of all scenarios are shown in Figure 1. The contribution analysis is elaborated for climate change and only the trends are discussed for the other environmental impacts. The avoided impacts are shown with a negative sign and would decrease the environmental impact of the production and recycling system; these are due to the avoidance of virgin plastic use. Impacts with a positive sign would increase the environmental impact of the system; these are due to the reprocessing activities (washing for reuse, turning bottles into recycled granulate for recycling), collection and sorting and blow moulding and are referred to as end-of-life (EoL) impacts. Because the consequential LCA consists of the net effect of avoided impacts and recycling impacts, relative contributions are given in percentage of sum of the absolute values, indicating to what extent they affect the net outcome. Such percentage values shown below are marked as %abs.

3.1.1. Climate Change

The climate change impact of all scenarios is negative, implying that the introduction of any of the three EoL options results in a reduction of the impact for all scenarios. Introducing the different EoL options has very similar effects for fossil and biobased plastics (the pairwise difference ranging from -4.5×10^{-3} to -3.4×10^{-4} kg CO_2 eq). This is because the climate change impacts of both biobased plastics do not differ strongly from their fossil counterpart. Fossil HDPE has a 15% higher climate change impact than biobased HDPE, due to differences in ethylene production. The bio-ethylene monomer contributes 50% of the climate change impact of the plastic production, while fossil ethylene contributes 56%. Bio-based and fossil PET are similar in impact: the bioMEG monomer constitutes a smaller portion of weight of PET (32 wt.%) and contributes 10% to the climate change impact of the plastic production, while fossil MEG contributes 11%.

The reuse EoL option reduces the climate change impact most strongly for all four reuse scenarios (ranging from -0.16 to -0.12 kg CO_2 eq/bottle) because they imply less reprocessing impact (only 3%abs) and a high material efficiency across the entire chain from first use to second use (the use-to-use efficiency). This results in large avoided impacts: plastic production contributes 60–75%abs of absolute values across reuse options and blow moulding contributes 20–25%abs.

The mechanical recycling scenarios reduce climate change impact to a much lesser extent than their respective reuse scenarios (ranging from -0.019 to -0.0039 kg CO_2 eq/bottle). This is due to higher EoL impacts and lower use-to-use efficiencies. Plastic production and blow moulding contribute most (30–45%abs and 5–10%abs, respectively) to the avoided impacts. Regarding to the EoL impacts, the collection and sorting required for the reprocessing contribute more to the climate change impact (20 and 30%abs for HDPE and PET) than the reprocessing and blow moulding itself (15 and 10%abs for HDPE and PET). The larger effect of PET mechanical recycling compared to HDPE is explained by the larger impact of PET that is avoided by its substitution through recycling.

The chemical recycling scenarios reduce climate change to a lesser extent than their respective reuse scenarios as well (ranging from -0.010 to -0.0056 kg CO_2 eq/bottle). While chemical PET recycling causes more avoided impact than chemical recycling HDPE, it causes more reprocessing impact so that its net effect is limited. The reprocessing and blow moulding contribute strongly to the reprocessing impact for PET (30%abs) while the collection and sorting contribute modestly (15%). Thus, mechanical recycling reduces climate change impacts more strongly than chemical recycling for PET (2.2 and 2.4 times larger for fossil and biobased PET, respectively). In addition, the avoided impacts of the coproducts from the HDPE pyrolysis (methane, ethane, etc.) ensure that the chemical reprocessing of HDPE actually reduces the climate change impact. Collection and sorting contribute 30%abs, similar to mechanical HDPE recycling but in contrast with chemical PET recycling. The net effect is that chemical recycling HDPE reduces climate change impacts more strongly than mechanical HDPE recycling (3.1 and 3.3 times stronger effect for fossil and biobased HDPE, respectively).

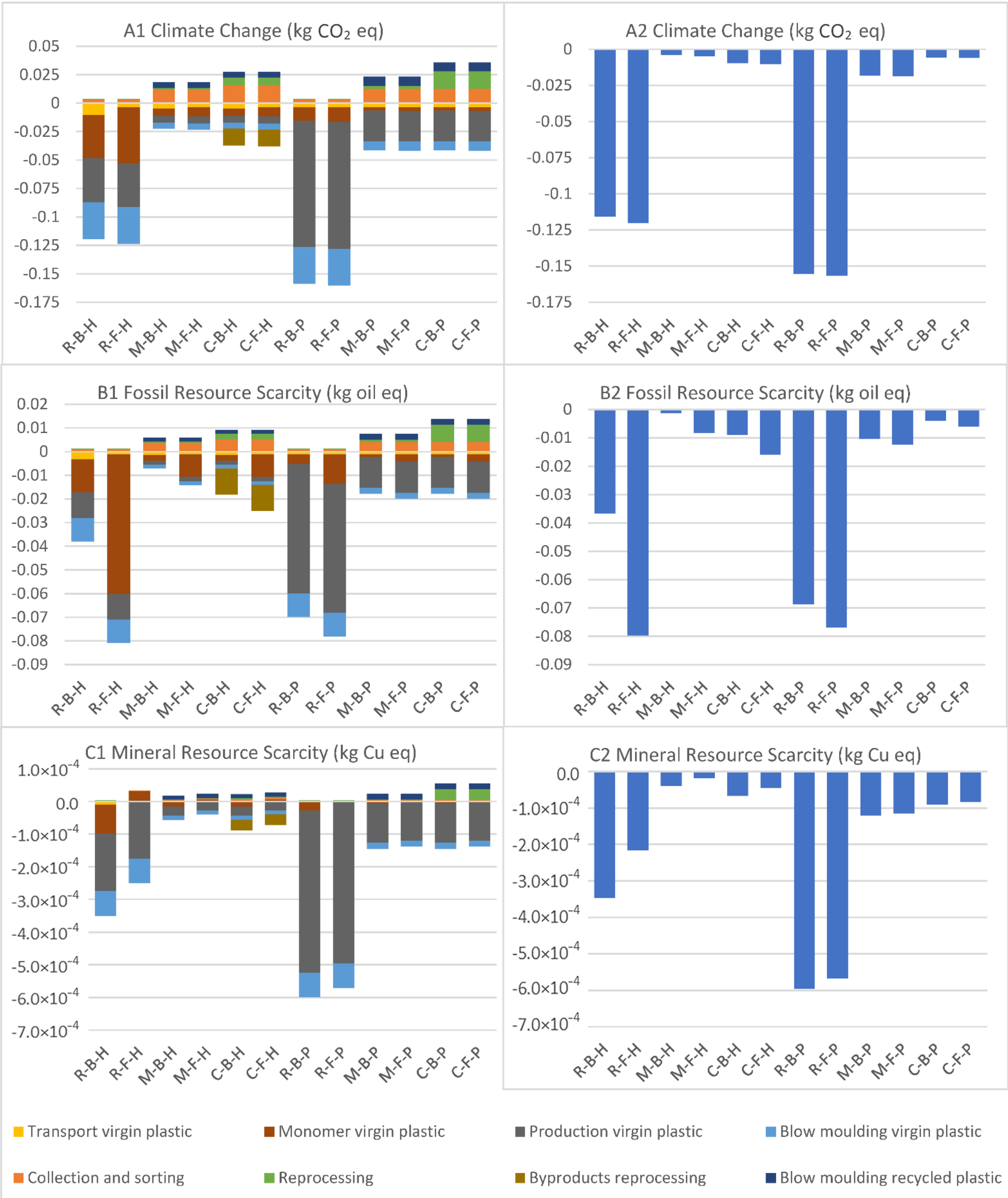

Figure 1. Environmental impacts of all 12 scenarios for A: Climate Change, B: Fossil Resource Scarcity, C: Mineral Resource Scarcity. On the left (**A1–C1**) absolute contributions are shown and on the right (**A2–C2**) the net effect is shown. Negative values indicate that the scenario causes a net reduction of the environmental impacts assessed. Scenario naming according to R = Reuse, M = mechanical recycling, C = chemical recycling, B = biobased virgin plastic, F = fossil virgin plastic, H = HDPE and P = PET. A figure excluding the reuse scenarios is included in the Supplementary Materials.

3.1.2. Fossil Resource Scarcity

Regarding fossil resource scarcity, trends are comparable to climate change, indicating a reduction of the impact for all scenarios. However, all scenarios with biobased plastics have lower avoided impacts and thus smaller effects on fossil resource scarcity than their fossil counterparts, thanks to their lower fossil resource dependence (ranging from 1.1 to 6.3 times lower). Reuse EoL options could reduce fossil resource scarcity most (ranging from −0.080 to −0.037 kg oil eq). For the same reason as for climate change, the effect of fossil resource scarcity is much lower for the mechanical recycling scenarios than for reuse. The effect is 10 and 28 times smaller for fossil and biobased HDPE and 7 and 6 times smaller for fossil and biobased PET, respectively (overall ranging from −0.012 to −0.0013 kg oil eq). The effect of the chemical recycling is about 5 times smaller for HDPE and about 14 times smaller for PET (ranging from −0.016 to −0.0040 kg CO_2 eq/bottle).

The trends mentioned for climate change are generally stronger for fossil resource scarcity. The trend that mechanical recycling has a larger effect than chemical recycling for PET is slightly more noticeable for fossil resource scarcity (2.1 and 2.6 times larger for fossil and biobased PET, respectively). The trend that chemical HDPE recycling has a larger effect than mechanical recycling is much stronger for biobased HDPE for fossil resource scarcity, because of the avoided fossil resource scarcity impact of the coproducts of pyrolysis (6.3 times larger, 1.9 for fossil HDPE).

3.1.3. Mineral Resource Scarcity

Regarding mineral resource scarcity, trends are comparable to climate change and fossil resource scarcity, indicating a reduction for all scenarios. However, all scenarios with biobased plastic have higher avoided impacts and thus they have larger effects on mineral resource scarcity then their fossil counterparts, due to their higher dependence on minerals and metals (ranging from 1.1 to 2.1 times higher). As for the other impacts, reuse EoL options could reduce mineral resource scarcity most (ranging from -6.0×10^{-4} to -2.2×10^{-4} kg Cu eq). Compared to reuse, the effect of mechanical recycling on mineral resource scarcity is about 10 times smaller for HDPE and 5 times for PET (overall ranging from -1.2×10^{-4} to -1.8×10^{-4} kg Cu eq). The effect of the chemical recycling is about 5 times smaller for HDPE and about 7 times smaller for PET (ranging from -9.0×10^{-5} to -4.6×10^{-5} kg Cu eq), because of the aforementioned trends. The relative difference between chemical and mechanical recycling is similar to climate change for mineral resource scarcity, where for PET the effect of mechanical recycling is 2.5 times larger for fossil PET and 1.7 larger for biobased PET. The relative difference between these recycling options is modest for mineral resource scarcity for both bio-based and fossil HDPE (chemical recycling has 1.4 times larger effect).

3.2. Sensitivity Analysis

The first sensitivity scenario assessed what would happen if the fraction of recycled plastic included in recycled bottles was increased from 16 and 24% for HDPE and PET, respectively, to 100%. This increase causes a strong growth in the use-to-use efficiency and shows what would happen if the collection or sorting efficiency were increased. For the reuse scenarios, the fraction of reused plastic in the reused bottle is already 100% and no change was induced. The effect of mechanical and chemical recycling on climate change can be 11 to 16 times larger for HDPE and 5.5 to 8.7 times larger for PET in this scenario (Table 3). The avoided impacts are increased, so the effect is strongest for the smallest net effects. Increasing the recycled fraction in mechanical and chemical recycling options has a smaller effect on fossil resource scarcity impacts (ranging from 4.3 to 7.2 times larger) because the avoided impacts are already large compared to avoided climate change impacts. The effect of the recycled fraction increase is more diverse for mineral resource scarcity impacts (ranging from 4.8 to 16 times larger), explained by the trends in the contribution analysis (Section 3.1.1).

Table 3. Relative changes to the environmental impacts for all 12 scenarios upon changes in the three sensitivity analyses. Large changes are depicted in factors, the smaller changes in percentages. Positive values indicate that the environmental benefit (net negative impact) of the scenario is increased. CC = Climate Change, FRS = Fossil Resource Scarcity, MRS = Mineral Resource Scarcity. Scenario naming according to R = Reuse, M = mechanical recycling, C = chemical recycling, B = biobased virgin plastic, F = fossil virgin plastic, H = HDPE and P = PET.

Impact	**R-B-H**	**R-F-H**	**M-B-H**	**M-F-H**	**C-B-H**	**C-F-H**	**R-B-P**	**R-F-P**	**M-B-P**	**M-F-P**	**C-B-P**	**C-F-P**
Sensitivity Analysis 1: 100% recycled plastic included in recycled bottle (expressed in factor changes)												
CC			16.5	14.9	11.5	11.2			5.6	5.5	8.7	8.4
FRS			6.7	7.2	6.6	6.8			4.3	4.3	4.3	4.3
MRS			16.2	7.8	8.1	7.3			5.0	4.8	6.2	5.5
Sensitivity Analysis 2: −25% sugarcane yield for biobased feedstocks (expressed in % changes)												
CC	0.5%		2.3%		1.0%		0.1%		0.2%		0.7%	
FRS	5.0%		7.2%		4.2%		0.8%		1.0%		1.3%	
MRS	2.4%		10.8%		1.6%		0.4%		0.6%		1.5%	
Sensitivity Analysis 3: −10% blow moulding efficiency for both virgin and recycled bottles (expressed in % changes)												
CC	−10%	−3%	−30%	3%	−30%	−14%	−10%	−2%	−15%	1%	−26%	26%
FRS	−10%	−3%	−11%	1%	−11%	−7%	−10%	−1%	−10%	0%	−10%	4%
MRS	−10%	−1%	−30%	1%	−17%	−7%	−10%	−1%	−13%	1%	−17%	12%

The second sensitivity scenario assessed what would happen if the yield of the sugarcane in the biobased plastic was reduced by 25%, resulting in 33% increase of the impacts in sugarcane cultivation The effect on climate change of the biobased EoL options can be 0.1% to 2.3% larger due to larger avoided impacts (Table 3). The effect on fossil resource scarcity can be 0.8% to 7.2% larger and on mineral resource scarcity the effect can be 0.4% to 10.8% larger. These small changes indicate the limited contribution of sugarcane cultivation in the scenarios. The variation in the resource scarcity impacts is larger because the net effect of recycling and avoided impacts is more variable for these impacts, and the trends are explained by the trends in the contribution analysis. The effects are smaller for PET than for HDPE, because the biobased monomer contributes a smaller mass fraction to the total polymer mass for PET (BioMEG vs. biobased ethylene). For fossil EoL options, no sugarcane is used, and no change was induced.

The third sensitivity scenario assessed what would happen if the blow moulding efficiency was reduced by 10%. This change in efficiency affects different contributions in each scenario. For all scenarios, the avoided impact of virgin plastic increases, while for both recycling options, the impact of reprocessing increases as well. The net effect can be large, as is the variation across scenarios, and is presented relative to variable baseline scenario results. For example, a 30% decrease is observed in the effect on climate change impact of mechanical bioHDPE recycling, because the avoided impacts decrease strongly while the impact of collection and sorting is not affected by blow moulding efficiencies. In contrast, a 26% increase is observed in the effect on climate change impact of chemical fossil PET recycling, because its large impact of reprocessing and blow moulding are both affected by blow moulding efficiency. Moreover, the effects are large because they are compared with the smallest baseline scenario impacts.

3.3. Uncertainty Analysis

The conducted uncertainty analysis demonstrates the compound effect of all uncertainty in the key variables in the foreground system from our study and does not represent a global uncertainty analysis. All results are presented in the Supplementary Materials. The uncertainty is variable across scenarios, with a coefficient of variation ranging from 2.9% to 38% for climate change. The smaller uncertainties occur in the reuse scenarios, because they contain fewer reprocessing data and transport data. The larger uncertainties occur in the chemical recycling scenarios, because of uncertainty in the reprocessing data. The 2.5 percentile from the Monte Carlo distributions could be seen as a global best-case scenario, considering optimistic assumptions. Reuse scenarios show less improvement potential with a 2.5 percentile 5 to 7% below the mean while chemical recycling options have a 2.5 percentile 38–73% below the mean. Mechanical recycling options are in between, with 26–58% potential improvement. All detailed trends in fossil and mineral resource scarcity follow trends described for these impacts in the sensitivity analysis (Section 3.2) and contribution analysis (Section 3.1).

4. Discussion

4.1. Comparison with Previous Research

This study confirms but also contrasts with key conclusions from previous studies. This is mainly because of the improved approach of a broadened focus on the total recycling system and the consequential LCA approach. The reuse option combines a limited reprocessing impact and a very high use-to-use efficiency (a high material efficiency across the entire chain from first use to second use). This causes large avoided impacts and a strong effect on the environmental impact. Mechanical and chemical recycling options have a limited use-to-use efficiency by the collection and sorting system and the practice of combining recycled plastic with virgin plastic during recycled bottle production.

This study confirms the previous work [7,55] that an optimised polymer production and inclusion fraction are important and could further reduce the environment impacts, for example through the first and third sensitivity analysis. Previous studies highlighted the need for more efficient recycling technologies from a circular economy perspective [7,25,56]. In addition, this study also illustrates that improved material recovery in the collection system is vital in mechanical and chemical recycling.

In contrast to previous findings [7,57], collection and sorting in the present study contribute significantly to all studied impacts with a large range of variability across scenarios. Nessi et al. (2020) indicated that distribution can be a relevant contributor to the three studied environmental impacts, despite different assumptions. End of life logistics and collection efficiencies should be optimised with equal emphasis as reprocessing technologies. Such optimisation will benefit the environmental impacts with the same trends as in the first sensitivity analysis.

It is likely that this specific reuse option will not be applied at large scale [7,22], as there are many packaging reuse models. It is expected that the use-to-use efficiency will determine the environmental effect of different reuse scenarios [32]. The current results support the suggestion by [31,58] that specific local conditions should be assessed as the logistics of the plastic management solutions have a relevant contribution. Because several studies highlight that with today's technology and current plastic user requirements recycled plastic cannot be utilised in the same manner as virgin plastic [24,57], our baseline scenarios considered the inclusion fraction of recycled plastic into recycled plastic bottles of present day. The first sensitivity analysis and the uncertainty analysis show that an increase of this fraction will strongly increase the benefits of the recycling options.

4.2. Study Limitations

This research study presents uncertainties which have been assessed in the uncertainty analysis for several parameters, as previously suggested [58]. In this study, several assumptions were made regarding the end-of-life (EoL) scenarios, production and transport. The weight of the bottle was kept the same across plastic types because higher density material can provide the same barrier properties and strength under a lower wall thickness. This approach is consistent with previous studies assessing plastic bottles [12]. For other packaging materials and for a detailed sensitivity and uncertainty analysis, a varying bottle wall thickness should be considered.

This study relies extensively on data, extrapolations and assumptions based on the literature. Specific data on emission, energy consumptions and performance were limited and protected by confidentiality. This is a common issue for new technologies [56,59]. As the chemical and physical properties are specific to each plastic type and chemical recycling is often carried with a mixed plastic waste stream [7], it is also hard to obtain specific data for a single type of plastic polymer. In the case of glycolysis, the specific energy consumptions data were not available and they were based on temperature requirements as described in the patent [41]. Further, plastic recycling technology is under development, thus technological parameters are subject to change with time [6,7]. All these aspects can affect the overall data quality, especially for the pyrolytic process. This issue is partially addressed with the uncertainty analysis, which shows that both mechanical and chemical recycling can have a much stronger effect on the environmental impact under more optimistic recycling efficiencies or inclusion fractions. This is also in line with previous works where assumptions were carried out and this results in a general lack of comparability among LCA studies, as discussed by Wang et al. (2021) [3]. Ideally, a structured and consistent method to account for missing data and for upscaling or extrapolating data should be applied across different studies.

The anticipatory consequential assessment carried out in this study used two substitution scenarios (i.e., biobased and fossil-based plastic); however, in contrast to other studies, this work used a 1:1 substitution assuming that the recycled plastics with a percentage of virgin plastic would achieve a satisfactory quality for a further use [24,57]. This was in agreement with previous research carried out for plastic bottles [12] and coherent with the objectives for this anticipatory consequential LCA focused on future technology [56,59]. The byproducts of pyrolysis also substituted their fossil counterparts on a one-to-one basis, for reasons of consistency. However, aspects of quality, byproduct separation and purification were not considered. Reducing or eliminating avoided impacts of pyrolysis byproducts limit the avoided impacts of chemically recycling HDPE. Future research could address variability in practice, such as in reuse scenarios. Furthermore, methodological uncertainty could be addressed by including numerous substitution scenarios for both recycled plastics as well as byproducts.

This study did not consider the simultaneous introduction of biobased plastics and a specific recycling option and did not explicitly compare the environmental impacts of biobased and fossil plastics, since the research focus was on evaluating the end-of-life (EoL) options. A larger number of substitution scenarios would be of interest here as well, although the trends in new scenarios would be explained with the trends in the current scenarios.

The current study assessed only three impact categories; however, a full assessment with more impact categories should be carried out to study possible environmental trade-offs, as largely discussed for other systems [60,61]. It is recommended to assess more environmental impact categories and to ensure data collection is suited to include impact categories.

Moreover, a major methodological issue is the lack of acceptable and widely used methods to account for temporary carbon storage. It cannot be determined what the climate change impact would be if carbon were stored away from the atmosphere for a limited time upon introduction of an EoL option. The carbon fixed in the plastics is only stored shortly and released upon incineration after the second life cycle. The storage of carbon in an individual shampoo bottle will not last longer than 20 years, but there is a collective effect on a scale of all bottles used over a period of time. A broader scenario study evaluating carbon pools in different materials at different places in society and including the latest methodology for dynamic greenhouse gas assessment could shed light on the combined effects of improved EoL options and increased use of biobased plastics. Assessment of carbon, stored in plastic packaging, would increase avoided impacts in correlation with the use-to-use efficiency of each end-of-life option, independent of the biobased or fossil origin of the stored carbon.

4.3. Recommendations

The production of new virgin plastic polymer should align with its use and the end-of-life requirements [7,12,55], which could be achieved through the regulation of plastic manufacturers [22]. Our approach and results include plastic production, use and end-of-life in one assessment, as suggested by [24], and can provide information for such alignment and regulation. It shows that small scale simpler solutions such as reuse could be very effective in the near future. It also highlights the impact of collection and sorting. This impact would be reduced by a smaller scale of the collection system, which contrasts with the scale requirements of chemical recycling for both HDPE and PET indicated by [55]. This scale issue has been largely discussed in previous research related to bioenergy systems [62–64] and within the 10R circular economy hierarchy [22].

In practice, the studied EoL options will be applied in combination with each other and with different EoL options, mixing large and small scales. Previous studies suggested a step towards closed-loop recycling should be made with new mechanical recycling of mixed systems, as recently proposed [7]. Garcia et al. (2017) [6] proposed to add compatibilisers to allow non miscible plastic compounds to become miscible, thus improving physico-chemical characteristics of recycled plastics. Another option would be to apply a smaller number of different plastic types that could be separated more easily. In this study, mechanical recycling seems the preferred option over chemical recycling for PET, thanks to the lower EoL impacts. Surprisingly, the reverse is the case for HDPE because of the avoided impact of the byproducts of chemical recycling (pyrolysis for HDPE). The difference in preference between mechanical and chemical recycling for the two plastic types might suggest that different recycling technologies might have a different order of preference for different plastics. The feasibility of increasing recycled plastic inclusion in recycled bottle production should be assessed, because of its strong beneficial effect. While previous studies suggest that quality requirements or virgin plastic quota might need to be reduced [7,24,55], issues of contamination, plastic quality and sorting accuracy for mixed plastics should be overcome.

Together with recycled plastic quality and purity and the use of recycled mixed plastics [24,57], the improvement of collection and sorting technologies are key in achieving the required purity for recycling process. Mechanical and chemical recycling should be made more suitable for smaller-scale facilities and mixed plastic streams, because this would reduce the collection and sorting impacts through reduced transport distances. As plastic waste streams are often contaminated, new recycling processes aimed at mixed plastic should be the focus of future research, as discussed previously [7]. However, there should be an alignment between the purity requirements of the plastic recycling process with the achievable sorting, collection and pre-treatment methods for recycling, which is key in achieving an environmental impact reduction [24,55].

5. Conclusions

This study has assessed end-of-life (EoL) scenarios for both biobased and fossil-based plastics, both PET and HDPE. This yields the following conclusions: (i) All EoL options cause a net reduction in all climate change, fossil and mineral resource scarcity thanks to the avoided impact of virgin plastic. Reuse options have the largest effect on the environmental impacts for both HDPE and PET while mechanical recycling has the smallest effect for HDPE and chemical recycling has the smallest effect for PET. Recycling biobased plastics has a smaller effect on fossil resource scarcity impact and a larger effect on mineral resource scarcity. (ii) Environmental hotspots are the avoided impacts of virgin plastic production (including the feedstock production and blow moulding) for all scenarios, and collection and sorting impacts for mechanical and chemical recycling options. (iii) The benefits of recycling occur in all scenarios for all environmental impacts but at very variable levels with variable uncertainties. Uncertainty and improvement potential are largest for mechanical and chemical recycling. (iv) The main methodological issues are the uncertain technological improvement for chemical recycling of both plastics and uncertainties regarding data gaps and extrapolations. In order to improve all EoL options in the future, optimization of the recyclability and quality of the recycled plastics as well as the collection, sorting and recycling efficiencies of the system deserve equal emphasis. Furthermore, the EoL options can be applied at different scales. At the optimal scale for each option, and in a combined fashion, the benefits of recycling will further increase.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su141811550/s1, Table S1 Transport distances and selected ecoinvent processes.; Table S2 Values and ranges for the uncertainty analysis for energy requirements during production and recycling. Table S3 Values and ranges for the uncertainty analysis for efficiencies and transport distances. Table S4 Results of the uncertainty analysis. Figure S1 Environmental impacts of 8 recycling scenarios excluding the reuse scenarios.

Author Contributions: Conceptualization, P.G. and S.S.; methodology, R.J.K.H. and P.G.; software, R.J.K.H., S.S. and P.G.; validation, D.S.v.E. and I.V.G.; formal analysis, R.J.K.H. and P.G.; data curation, R.J.K.H. and P.G.; writing—original draft preparation, R.J.K.H. and P.G.; writing—review and editing, L.A. supervision, L.A. and D.S.v.E. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Wageningen University Knowledge Base programme: Circular and climate neutral society (KB-34-010-001): Biobased materials and chemicals for relieving and replacing the fossil feedstock system (subtheme 3B), which is supported by funding from the Dutch Ministry of Agriculture, Nature and Food Quality.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data describing the analyzed system are presented in Tables S1 and S2 in the article in the Supplementary Materials. Restrictions apply to the availability of secondary process impact data. These data were obtained from the ecoinvent organisation through SimaPro 9.3 and are available through the ecoinvent organisation or through other LCA software under license charges. Names of process data items can be shared by the authors.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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