



Plastic recycling in a circular economy; determining environmental performance through an LCA matrix model approach

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ABSTRACT

To ensure a circular economy for plastics, insights in the environmental impacts of recycling and optimal recycling choices for specific plastic polymers are crucial. This was obtained by determining the environmental performance of 10 selected recycling technologies with varying TRL levels, using the chemical properties of the top 25 produced polymers in Europe. The results were collected in a life cycle assessment (LCA) 'matrix' model. To simulate realistic plastic recycling challenges, case studies of PE/PP foils from municipal waste and ABS plastic with brominated flame retardants were developed, to be used as an addition to the LCA matrix model results. Potential emission reduction was assessed by combining LCA matrix outcomes with European polymer demand data. The LCA matrix model illustrates that potential environmental performance of recycling technologies varied strongly per polymer type and did not always follow the state-of-the-art recycling hierarchy. Commodity plastics performed well with tertiary recycling technologies, such as gasification and pyrolysis to monomers; secondary mechanical recycling was outperformed. A focus on primary recycling is environmentally beneficial for most engineering and high performance plastics. To enhance the performance of primary recycling technologies, a higher purity and improved sorting is required. As demonstrated in the case studies, low sorting efficiencies due to impurities reduces positive environmental impacts. Hence, optimal environmental performance of recycling is obtained where pre-treatment (sorting, cleaning) is adapted to the recycling technology. According to the model, recycling the 15 most demanded polymers in Europe reduces CO₂ emissions from plastics by 73% or 200 Mtonne CO₂ eq.

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1. Introduction

In 2018, 359 million tonnes of plastic was produced worldwide (PlasticsEurope, 2019). Still, only 9% of the plastics ever produced have been recycled (Geyer et al., 2017). The other fractions of waste plastic are either incinerated (12%), landfilled or lost to the environment (79%) (Geyer et al., 2017). Although plastic waste can negatively affect ecosystems in itself (Gall and Thompson, 2015), the continuous production of virgin fossil-based plastics has a significant impact on the environment as well. This is a result of fossil fuel depletion during material extraction, emissions associated with production from energy use, the transport of materials and (when applicable) incineration at end-of-life. Without value retention, the value from plastics in terms of material and embodied energy is lost. A solution to decrease environmental impact and to keep global temperature rise under 2 °C (UNFCCC, 2015) is to

develop a circular economy to replace the current, linear, economy. In a circular economy, products and materials are being re-used, refurbished or recycled according to the waste hierarchy, instead of being incinerated or disposed (Hansen et al., 2002). Consequently, fewer virgin material is required, which decreases raw material demand and raw material depletion, emissions and pressure on vulnerable ecosystems (Ellen MacArthur Foundation, 2013).

To make plastic a circular material, its current recycling rate of 9% needs to increase significantly. Plastic packaging from municipal waste is most recycled, whereas plastic recycling from other sectors (Construction, electronics, automotive) lags behind (Consultic, 2012). Additionally, to ensure re-use in products, the quality of recycled plastics has to increase (Hahladakis and Iacovidou, 2018). There is a general hierarchy available for plastic recycling, based upon the degree to which the polymer stays intact, which overlaps with the inner (material remains intact) and outer loops (material not intact) of the circular economy (Ellen MacArthur Foundation, 2017). This is captured in the categorization

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of primary (most intact), secondary, tertiary and quaternary recycling (least intact) (Singh et al., 2017). Hence, primary recycling is considered the most optimal (inner loop) and quaternary recycling (outer loop) the least (Singh et al., 2017). At this moment, plastic recycling occurs mainly through mechanical recycling from mixed waste streams, and is categorized as secondary recycling ('open-loop' recycling). In this system, the plastic is downcycled, meaning it is only partially re-used for the same purpose due to quality reduction (Paolo and Mantia, 2004; Ragaert et al., 2017; Sheppard et al., 2016). The quality and quantity recycling gaps is a result of plastic waste being collected in a mixed stream, consisting of different polymers and even materials (metals, cardboard, rubber and more). Furthermore, plastic products can contain a mix of materials and polymers, including multilayer material, copolymers, stickers, fillers and additives, which complicate the recycling process (Hopewell et al., 2009). These conditions vary strongly per plastic application and sector, and hence per waste stream. To summarize, presently recycling takes place for a limited selection of the total plastic waste streams, with only a few recycling technologies applied in a large scale, while the process itself is complex and suboptimal due to quality limitations. However, there are alternative, innovative recycling technologies that might fill these gaps, and surpass the limitations and boundaries associated with the plastics from different waste streams. This includes tertiary recycling options where the plastic waste is recycled to monomers or feedstocks with thermochemical methods (Vollmer et al., 2020). Other chemical recycling options are being developed as well, such as depolymerization, which breaks polymer bonds using chemicals, or dissolution with solvents that keeps polymers intact (Vollmer et al., 2020). Unfortunately, it is still unknown which existing or innovative recycling technology theoretically offers the most environmental benefits for each plastic application, and hence which technologies fit best in a circular economy.

Through Life Cycle Assessment (LCA), the environmental impact of products or technologies can be quantified. Existing LCA studies on plastic recycling are executed to analyse the environmental benefit of a recycling technology against the status quo. The recycled polymers are credited as 'avoided virgin polymer' and receive a negative value on the environmental impact balance, resulting in a 'positive' contribution (Gu et al., 2017; Wang et al., 2015). However, the scope and coverage of LCA studies are often limited and therefore unable to support a circular economy perspective. Often, only a single recycling technology is assessed, or only a specific waste stream (packaging or municipal plastic waste) (Chen et al., 2019; Dodbiba et al., 2006; Gu et al., 2017; Lazarevic et al., 2010). Furthermore, most LCA studies include existing boundaries associated with waste stream, hence becoming so specific that comparability is limited and the results cannot be used in another context or used for scaleup (Astrup et al., 2015). Thereof, LCA studies that focus on the current situation with its existing boundaries can potentially lead to suboptimal results, since their limitations can obscure the full potential of a recycling technology. In short, an overarching vision on optimal plastic recycling as a resource is lacking. Our goal with this study is to develop an overarching environmental assessment of the potentials of different plastic recycling technologies, applicable to all waste plastics, to support the development of a circular economy for plastics. By streamlining the LCAs functional units, system boundaries, conditions and input parameters for a wide selection of polymers and recycling technologies, an environmental impact matrix can be developed where polymer and recycling treatment are used as two dimensions. In such a matrix, variations between recycling of plastic polymer types become visible and the results comparable. The LCA matrix results will provide an overview on the environmental benefits of plastic recycling, where all main recycling options and plastic applications are assessed.

In this study the LCA matrix model is developed in order to assess the potential environmental performance of existing and innovative recycling technologies for 25 plastic polymers. To ensure comparability, the LCA matrix model includes polymer production versus a recycling treatment and processing into products. Use and collection phases are excluded. The recycling outputs are credited as 'avoided impact' (materials, feedstock or energy). The LCA matrix model will clarify on a polymer level which recycling technologies provide the most environmental benefit and whether this is in line with the existing recycling hierarchy. To ensure applicability to existing plastic recycling challenges, an extension of system boundaries is added to the model. These extensions are quantified in the 'case studies' and work as an additional layer to the LCA matrix model results. With the case studies, both the applicability and efficacy of the LCA matrix model are shown and the effect of boundary conditions from waste streams is quantified. Next, the matrix model is applied on European demand quantities to obtain the potential environmental impact reduction of recycling technologies. Together, these insights provide an environmental outlook on the mid and long term recycling strategies and the role of plastics recycling in a circular economy.

2. Methodology

2.1. Environmental data

Environmental data on materials, fuels and emissions was obtained from the ecoinvent 3.4 cut-off by classification database (Wernet et al., 2016). Environmental data used for polymers is discussed in section 2.2. The characterized unit data was used, focusing on the European data {RER} where possible, or else global datasets were used. This data was extracted using SimaPro 8 (Preconsultants, 2019), applying the midpoint impact categories from ReCiPe 2008 (h) (Goedkoop et al., 2013), and excluding long term emissions and including infrastructure processes. For aggregating the results of all midpoint impact categories, the total impact was calculated to a single score by applying shadow prices on the ReCiPe characterized results. The shadow price set used is developed by CE Delft and TNO and gives the most recent economic damage per impact category (De Bruyn et al., 2010; Ligthart and van Harmelen, 2019). The more recent ReCiPe 2016 impact assessment method (Huijbregts et al., 2017) does not have a full set of shadow prices, this is the reason why the previous version of ReCiPe was applied. All units are converted to Euro (€), so that midpoints can be combined and compared. The model outcomes were assessed through the total price and the midpoint character *climate change* (kg CO₂ eq) as this was chosen most representative proxy for the other impact categories.

2.2. Polymer selection

25 polymers were selected according to two criteria. Firstly, all polymers with European demand volumes of over 0.2 Mt/year were included (PlasticsEurope, 2019). Secondly, a wide variety of polymers had to be represented. This included commodity plastics, engineering plastics and high performance thermoplastics. Commodity plastics are strongly represented, as their production exceeds 0.2 Mt/year. Several engineering and high performance plastics with low production quantities were added (PSU, PEEK, PTFE). Additionally, another thermoset plastic besides PUR (bisphenol-A epoxy) and a bioplastics (Polylactic acid, PLA) were included to represent thermoset polymers and biopolymers. Appendix A includes the full list of polymers and database sources. The selected polymers represent at least 95% of 2018 European demand (PlasticsEurope, 2019). The final step included building a

dataset containing all information on the polymers chemical properties, molar mass, density, heat capacity, calorific value, type of production, melting point, glass temperature and data which was specifically required for recycling technologies, including monomer units, syngas yield and solvent to be recovered and solvents for dissolution purposes.

2.3. Recycling technologies

A large number of commercial and innovative recycling technologies are available for plastic recycling, including mechanical, chemical and thermochemical options. For this study, the technologies were classified according to the four categories of the recycling hierarchy (Singh et al., 2017; Vollmer et al., 2020). Biological recycling options were not included in this analysis, as this area of research is still young and has limited data available.

1. **Primary recycling** or closed-loop recycling. The material can be recycled to form products with the same properties as the previous product, hence the plastic polymer remains in the same 'loop'. Primary recycling takes place for pre- and post-consumer (mono-stream) plastics.
2. **Secondary recycling** or open-loop recycling. The material can be recycled but recycle is of lower quality than the original material, hence the plastic polymer is applied in other, usually lower value products and recycled in an 'open-loop'. Currently, most consumer plastic recycling follow this route.
3. **Tertiary recycling** includes plastic to feedstock and plastic to monomer recycling. The polymer is not kept intact, but valuable materials (feedstock, monomers) are being recovered.
4. **Quaternary recycling** or incineration of plastic material with energy recovery. The material is incinerated and fully destroyed. However, the high calorific value of plastic results in recovery of energy as heat and electricity.

There can be large variations within the same recycling category and technology in input parameters (such as energy use, product outcomes and emissions). Therefore, data for recycling technologies can deviate even though it is the same technology, for example at plant level. In this study, the recycling technologies that are described are a generalization of the wide variation possible within the technology itself. Furthermore, some technologies selected are innovative and are not yet applied on market scale. The level of development of the recycling technologies is defined on the Technology Readiness Level (TRL) scale (Mankins, 1995). Here, it is divided in three levels: low (1–4), medium (5–7), and high (8–9). The following recycling technologies are selected divided by recycling hierarchy category, where the definitions are based on (Vollmer et al., 2020):

1. Primary recycling

1.1. *Closed-loop mechanical recycling (TRL: high)*. This form of mechanical recycling considers the mechanical recycling for pure plastics. Plastics that are not mixed with other disturbing materials or polymers can be recycled through closed-loop.

1.2. *Dissolution of polymers with two solvents (TRL: low)*. This physical recycling technology resembles dissolution with supercritical solvent(s). The polymer is dissolved by a specific solvent and cleaned from potential contamination, using cosolvent to separate the polymer and retrieve the used solvent. The polymer is retrieved and regranulated. Solvents vary per polymer, and some dissolution technologies do not use cosolvents to retrieve the polymer. At this moment, recycling through dissolution is not realized for all polymers yet. In the model it is assumed that all polymers can be treated with dissolution.

2. Secondary recycling

2.1. *Open-loop mechanical recycling (TRL: high)*. When waste plastic polymers are collected in mixed waste streams, it contains a significant amount of contamination. This can be caused by other materials or polymers due to complex products, through contamination from the use phase, or through additives like colors. Hence, a (mixed) recycle with lower material properties (quality) than the virgin product is obtained after secondary recycling. This recycle cannot replace the original products.

3. Tertiary recycling

3.1. *Gasification to feedstock products (TRL: high)*. This chemical recycling technology resembles a high temperature gasification. For this technology the polymer is used as a refuse derived fuel and is transformed in a gasifier to syngas with a H_2/CO molar ratio of 2:1. The quantity of syngas and the accompanying CO_2 emissions depending on the polymer type.

3.2. *Pyrolysis to feedstock products (TRL: high)*. For this chemical recycling technology the polymer is used as a refuse derived fuel and is converted to pyrolysis oil, assumed equivalent to diesel. The energy content of the diesel depends on the polymer calorific value.

3.3. *Pyrolysis to wax products (TRL: high)*. For this chemical recycling technology, the polymer is used to obtain medium length hydrocarbon chains ($C_{10}-C_{14}$) to replace paraffin like waxes, which can potentially be used to replace lubricating oils as well.

3.4. *Thermochemical recycling to monomers through gasification (TRL: medium)*. The polymers are gasified at a medium to high temperature and a gas with a mix of chemicals are obtained, including BTX, monomers and short carbon molecules (C_2-C_5). The output can be used to make new polymers. The chemicals formed strongly depends on the polymer. An important requirement for monomer recovery is product separation.

3.5. *Thermochemical recycling to monomers through pyrolysis (TRL: medium)*. The polymer is pyrolyzed on a medium temperature and an oil with a mix of chemicals is obtained, which include monomers, BTX and shorter carbon molecules. These can potentially be re-used for making new polymers and the chemicals formed strongly depends on the polymer. An important requirement for monomer recovery is separation of the remaining oil.

3.6. *Depolymerization through glycolysis (TRL: medium)*. With this chemical recycling technology, ethylene glycol is added to specific polymers (PET, Nylon 6) in the presence of a catalyst, the polymer is depolymerized to its building blocks, which can be re-used in making new polymers.

3.7. *Hydrolysis with water for specific biopolymers (TRL: low)*. For specific biopolymers (in this case, PLA), the polymer can be dissolved in water when heated up and broken down to its monomer building blocks. These monomers can be re-used in making new polymers.

4. Quaternary recycling

Incineration for energy recovery (TRL: high). The polymer is incinerated but energy that is released is captured and can replace heat and electricity. The amount of heat and power depend on the calorific value (energy content) of the polymer and the configuration of the waste-to-energy plant. Table 1 gives an overview of all selected technologies, their recycling efficiencies and their main products. It is assumed that the material losses associated with recycling (efficiency) are recycled by incineration with energy recovery. In order to incorporate the loss in quality of the end products for secondary recycling, a literature assessment of what products can be substituted by the output of secondary mechanical recycling was executed (see appendix B). This is incorporated in a quality factor that is used to show decrease in output quality

Table 1

Overview of the recycling technologies assessed. The main substituted (avoided) products are mentioned and the recycling efficiencies, and quality factors that are taken into account. The quality factor and the efficiencies of 'polymer dependent' technologies, which vary per polymer input, are further elaborated in appendix B.

Technology	Substituted products	Recycling efficiencies	Quality factor
Incineration	None	0%	
Energy recovery	Electricity, Heat	21% electricity, 6% heat	
Gasification (Feedstock)	Syngas	Polymer dependent	
Pyrolysis (Feedstock)	Diesel	Polymer dependent	
Pyrolysis (Wax)	Paraffin, lubricating oil	50% paraffin, 10% lubricating oil	
Pyrolysis (Monomers)	Monomers, hydrocarbons	Polymer dependent	
Gasification (Monomers)	Monomers, hydrocarbons	Polymer dependent	
Mechanical open	Polymer (pellet)	80%	50%
Mechanical closed	Polymer (pellet)	74%	
Dissolution	Polymer (pellet)	90%	
Depolymerisation	Monomers	90%	
Hydrolysis	Monomers	95%	

(Table 1). An extensive description of all recycling technologies with all underlying assumptions for treatment and avoided products is described in Appendix B.

2.4. System boundaries

To assure comparability between polymers and recycling technologies, the system boundaries for the LCA matrix model include: polymer granulate production, recycling treatment impacts and avoided products. Hence, the model assumed pure polymers as waste input (Fig. 1). Outside the system boundaries are all parameters that vary greatly depending on product type, sector, and waste collection method. This includes polymer processing steps to products (e.g. injection moulding). Also the addition of additive materials, such as fibres, colours or flame retardants (Hahladakis et al., 2018) are excluded. Waste collection and pre-treatment (sorting, cleaning) are not included. Material inputs for treatment includes materials (solvents, soaps, chemicals), heat, electricity, water and waste products (emissions, waste treatment and wastewater). Through the substitution approach, recovered materials are credited as 'avoided products' (Ligthart and Ansems, 2002). These credits are affected by the type of output product

(e.g. polymer, monomer, energy), the assigned efficiencies of the recycling processes and material transfer coefficients. When feedstock is obtained from recycling, the technology obtains credits for the avoided, hence saved, feedstock.

2.5. Model development

The environmental data, polymer data and recycling parameters were combined within functions that were created for every recycling technology, where recycling data was used as input parameters. These functions were created in R Studio version 3.6.0 by using the following R libraries: dplyr, reshape2 and ggplot2 (R Core Team, 2019; Wickham, 2016, 2007; Wickham et al., 2019).

2.6. Case studies

In order to test applicability of the theoretical LCA matrix model to realistic, existing recycling challenges, several important steps in the plastic production and recycling chain have to be taken into account in the system boundaries. This was done by selecting two waste streams which were developed into *case studies*. For this, the system boundaries were expanded and an additional framework with conditions was created. The adapted system boundaries is visualized in Fig. 2, and the additional conditions for recycling technologies are summarized in Appendix C.

1. *PP and LDPE foil material* (formed through sheet extrusion) from packaging material, collected through municipal waste collection. Before primary, secondary or tertiary recycling, a sorting step is required with an energy use of 0.039 kWh per kg input (Perugini et al., 2005). For open-loop mechanical recycling and pyrolysis technologies, a high sorting effort is required with an efficiency 0.41 (0.59 to energy recovery). For gasification technologies, the sorting effort is assumed lower as biogenic materials can be co-gasified (Pinto et al., 2003), although this affects the outcome products (Fig. C1). The sorting efficiency is set on 0.5 (0.5 to energy recovery). Sorting is not included for energy recovery and incineration. The framework includes transport from collection to sorting facility of 100 kgkm.
2. *Injection molded ABS from electronic equipment, containing 2% flame retardants* (mix of Tetrabromobisphenol-A (TBBPA) and Antimony Trioxide (ATO)). The material is collected through old electronic equipment collection and therefore is part of a mixed waste stream. Added is a sorting step with 0.039 kWh

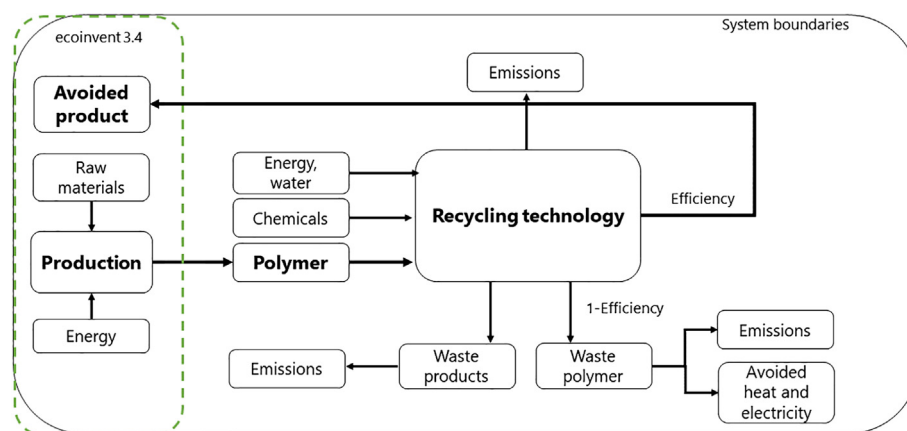


Fig. 1. The system boundaries for LCA the matrix model. The boundaries include production of the polymers raw materials and energy for treatment, waste streams and emissions from the recycling treatment and the avoided materials (output) for the output product. The green dashed line indicates data input from ecoinvent (or ILCD for specifically PAN fibers).

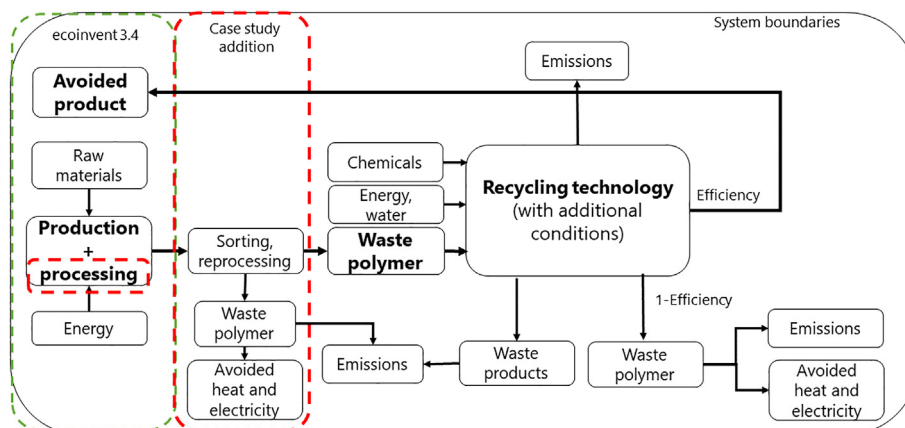


Fig. 2. The alternative system boundaries for using the LCA matrix model used within the defined case study frameworks. A) PE-PP foil mix. B) ABS plastic with flame retardants.

per kg input (Perugini et al., 2005) with an efficiency 0.5 and 0.5 to energy recovery (De Meester et al., 2019). The defined prior steps are not included for energy recovery and incineration. The framework includes transport from collection to sorting facility of 200 kgkm, due to fewer WEEE sorting facilities. Due to the presence of the flame retardants; dissolution is extended with additive removal and recovery.

2.7. Reduction potential

The 25 selected polymers in the model are not produced and disposed in similar quantities. This depends on the amount of polymer produced in a year and on the lifespan of the product. Depending on the application of the polymer and the sector in which it is applied, the lifespan of plastic may vary from a few months up to 35 years (Geyer et al., 2017). As the lifespan varies within the field of application, not disposal but demand data from plastics Europe was used instead (PlasticsEurope, 2019). With this information, potential environmental impact reduction was assessed using the developed LCA matrix model when these polymers are being recycled through their environmentally most optimal technology.

3. Results

3.1. Environmental impact per polymer and recycling technology

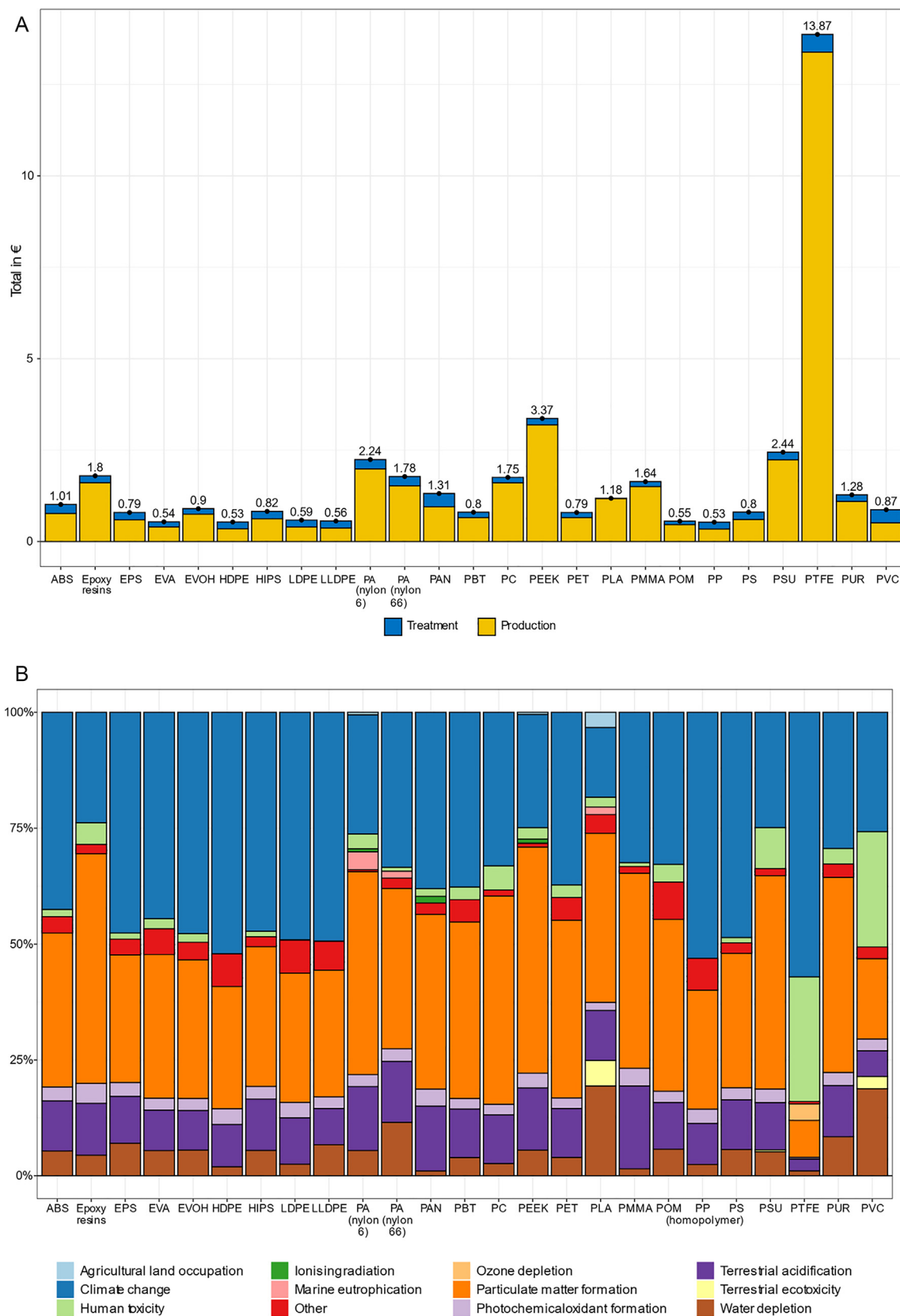
The single score environmental impact per polymer type varies when polymer production and incineration as waste treatment are visualized (Fig. 3a). The environmental impact is mainly caused by production of the polymer rather than by incineration. Hence, a circular recycling technology should focus on quality and quantity of avoided product production, to optimize environmental impact reduction. Due to high energy use, climate change and particulate matter formation are the largest contributors to the total environmental impact, combined between 50 and 75% for all polymers (Fig. 3b). Despite the high policy attention, climate change entails only 25–50% of the total environmental impact when regarding single scores. The contribution to the other impact categories is mainly caused by presence of additional molecules, including nitrogen (PAN, ABS, nylon), sulphur (PSU), fluorine (PTFE) and chlorine (PVC, Epoxy). These polymers contribute to human toxicity, eutrophication of the marine environment or ionizing radiation. Hence, PTFE has the highest environmental impact, mainly due to high human toxicity potential, while the polyolefins (PP, LDPE, HDPE, LLDPE) have the lowest. Although there are exceptions

(PLA), climate change is considered a suitable proxy for total environmental impact to be used in the model.

The variation of environmental impact through recycling methods is exemplified for PET in Fig. 4. Only for quaternary recycling technologies (incineration and energy recovery), the impact of the recycling treatment is slightly higher than the positive environmental impact of the avoided products (heat and electricity). Overall, the environmental impact for recycling treatment varies to some extent (for PET between €0.04–0.38 per kg, or 6% to 58% of the impact of polymer production) while the range for avoided materials (modelled as negative values) is larger (for PET between €0–0.58 or 0% to 89% of the impact of polymer production). Therefore, the quality and quantities of the avoided materials or feedstock affect environmental impact the most, hence the trade-off between the environmental impact of treatment and avoided products is positive. This means that for PET, increasing treatment intensity (e.g. energy) is environmentally beneficial when quality and quantity of output products increase. This is exemplified for dissolution and mechanical recycling (closed loop), with a high treatment impact but also a high value for avoided products. The open loop recycling has lower benefits due to the applied quality factor.

3.2. LCA matrix model

The developed polymer-technology LCA matrix (climate change) is visualized in Table 2. Incineration with energy recovery has the highest CO₂ emissions compared to the other recycling technologies. This is the only quaternary recycling technology in the model and most used technology at this moment. For the tertiary recycling options the CO₂ emissions vary widely between technologies and polymers. The tertiary recycling technologies of pyrolysis and gasification to waxes and feedstock have lower CO₂ emissions than incineration with energy recovery. This is a result of higher energy efficiency and value retention, however also a result of avoidance of the combustion emissions due to the system boundaries of the LCA. Gasification with monomer recovery has the lowest relative (color) and absolute (number) CO₂ footprint for the polyolefins (LDPE, HDPE, PP, LLDPE), with a CO₂ -eq of 0.9–1.1 kg per kg. For these polymers this tertiary recycling options has lower impact than the primary recycling options. This is due to the low environmental impact of production for polyolefins (Fig. 3a) resulting in low avoided impacts for primary recycling products. Also partial transformation to BTX chemicals during gasification to monomers contributes to the low CO₂ emissions, as these have a relatively high footprint. For another group of poly-



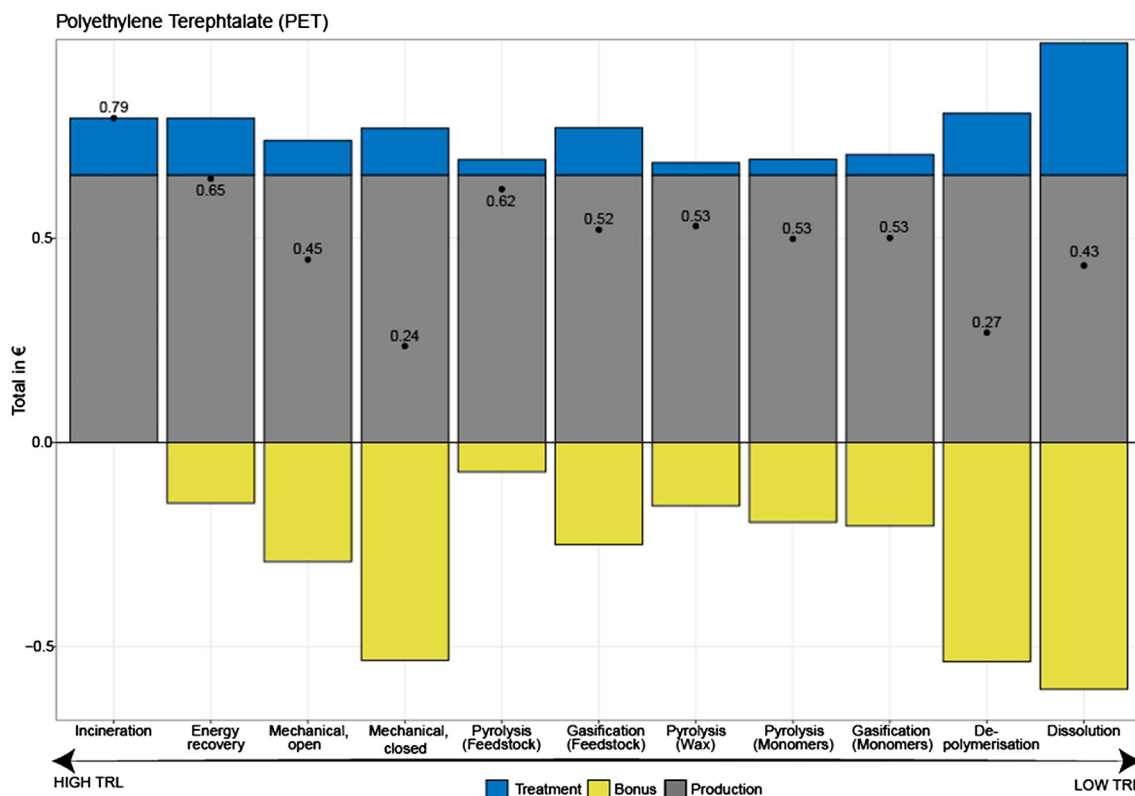


Fig. 4. The total environmental impact for recycling PET (per kg) with selected recycling technologies, sorted from high TRL level (left) to low TRL level (right). The analysis included polymer granulate production, recycling treatment and the recycling credits, which are visualized as negative values.

mers, particularly pyrolysis to monomers, result in a CO₂ reduction of more than 80% compared to incineration (1.5–2.9 kg CO₂ eq./kg polymer). This group includes polymers that are able to form a large amount of their monomer under the right pyrolysis circumstances (PMMA, PS, EPS, HIPS, Nylon 6). The depolymerization and hydrolysis, which are only applicable on a small selection of polymers (PLA, PET, Nylon 6), also result in a reduction of more than 80% compared to incineration, with 1.3–1.6 kg CO₂ eq. emissions/kg polymer. It should be noted that for tertiary recycling, a monomer product or chemical is obtained and ‘avoided’, and not a polymer like primary recycling. If these monomers or chemicals can be re-used in polymers, additional energy is required for polymerization. Although the polymer is broken down to a lower utility level, total yield and perhaps even quality can be higher than secondary or even primary recycling. According to the model, open-loop mechanical recycling (the only secondary recycling technology), does not seem to be the second best option for a number of polymers. For these polymers the quality decrease that comes with secondary recycling (assumed 50%) is significant enough for tertiary recycling to perform better. The secondary recycling does perform better than tertiary recycling for polymers that have high energy demand for production (and hence have a higher CO₂ footprint, about ~7 kg CO₂ eq. per kg after incineration). At last, the primary recycling technologies (dissolution and closed-loop mechanical recycling), show low environmental impacts. Primary recycling results in the lowest relative CO₂ emissions for a large group of polymers. For thermosets like PUR and Epoxy, mechanical recycling is not possible (and are incinerated after treatment); however, dissolution technologies can be a solution for these thermoset plastics when possible. The possibility for mechanical recycling to take place in a closed-loop or open-loop is crucial to determine the best recycling option for a large group of polymers, including ABS, PBT, PVC, PTFE and EVA. It determines whether

mechanical or an alternative, tertiary recycling technology is the best choice. If it is possible to recycle these polymers with closed-loop mechanical recycling, this technology can be considered the best recycling strategy. However, if a high quality output cannot be obtained due to potential boundary conditions, and open-loop mechanical recycling is required, a tertiary recycling method will result in lower CO₂ emissions

3.3. Case studies

In case study 1, LDPE and PP plastic films, the state of the art sorting efficiency (0.41) results in a low recovery rate compared to the outcomes of the LCA matrix model, which considers pure polymer recycling. Also, several recycling technologies are excluded since these excluded through the additional boundaries (stated in the SI) (Fig. 5A). This case study visualizes a similar technology ranking order for the polyolefins in the matrix model, however CO₂ reduction potential is lower due to the addition of sorting activities, losses, included product manufacturing and exclusion of technologies decided through the boundary framework. Furthermore, the technologies score in a more equal range due to the low sorting efficiency which leads to a high amount of material to energy recovery. Monomer recovery through gasification performs best, showing a total impact of 3.5 kg CO₂ eq. per kg, as the gasification technologies require a lower sorting effort (50%) compared to other technologies, as it is assumed biogenic material can be co-gasified (Pinto et al., 2003). Open-loop mechanical recycling has most CO₂ emissions after energy recovery, with 4.4 kg CO₂ eq. per kg.

For the ABS WEEE plastics, a high environmental impact is associated with dissolution treatment (Fig. 5B). However the credits for the avoided materials is also high; more than 4 times higher than monomer recovery through gasification. Dissolution results in low-

Table 2
The CO₂ emissions, relative to incineration (colour) and absolute (number), determined in the LCA matrix model for 25 most produced polymers versus available and innovative recycling technologies, per kg polymer. The technologies are ranked from top to bottom based on average polymer CO₂ emission and polymers categorized according to structural similarity.

Climate change (kg CO ₂ eq. / kg polymer)		polyolefins				Monomer forming polymers						Depolymerization/ hydrolysis polymers				Complex polymers										Thermoset polymers			
		PP	LDPE	HDPE	PS	EPS	HIPS	PMMA	PA (nylon 6)	PET	PLA	ABS	PVC	EVA	EVONH	PA (nylon 60)	PAN	PBT	PC	PEEK	POM	PSU	PTE	PUR					
High relative CO ₂ eq.	Incineration	5.2	5.3	5.1	5.2	6.8	7.0	9.4	10.1	5.4	3.2	7.8	4.0	4.4	7.7	10.6	8.9	5.5	10.3	14.5	3.4	10.7	138.9	6.7	7.6				
	Energy recovery	4.3	4.1	3.9	3.9	5.9	5.9	8.7	9.4	4.7	2.7	6.8	3.4	3.6	6.9	9.9	8.1	5.0	9.5	13.7	3.0	9.9	138.7	5.8	6.6				
	Pyrolysis (Energy)	1.9	1.9	1.7	1.8	3.4	3.3	3.4	7.2	7.5	3.4	3.2	4.4	2.0	2.3	5.5	8.1	5.6	3.4	8.0	11.6	2.5	7.9	137.4	4.7	5.2			
	Gasification (Energy)	1.5	1.7	1.4	1.5	3.6	3.5	3.6	7.0	7.2	3.3	2.3	4.5	2.0	1.7	5.1	7.7	5.9	3.1	7.8	11.7	1.8	8.2	137.6	4.4	5.1			
	Pyrolysis (Wax)	1.6	1.7	1.4	1.5	3.1	3.0	3.1	6.9	7.2	3.0	2.9	4.1	2.1	2.0	5.1	7.8	5.3	3.0	7.7	11.3	2.2	7.7	137.1	4.3	5.0			
Low relative CO ₂ eq.	Mechanical (Open loop)	2.0	2.0	1.9	1.9	3.0	2.9	3.0	5.0	5.2	2.5	2.2	3.5	1.9	1.9	3.9	5.6	4.3	2.6	5.4	7.7	1.7	5.6	83.0	-	-			
	Pyrolysis (Monomers)	1.3	1.4	1.2	1.3	1.5	1.4	1.7	2.7	2.9	2.9	2.5	4.2	2.2	2.0	5.3	7.6	5.1	2.8	7.4	10.9	2.0	7.4	70.6	4.3	4.7			
	Gasification (Monomers)	1.0	1.1	0.8	0.9	2.4	2.3	2.5	6.6	6.9	2.6	2.5	3.9	2.0	1.5	4.7	7.4	5.0	2.7	7.2	10.7	2.0	7.2	137.0	4.0	4.5			
	Mechanical (Closed loop)	1.4	1.3	1.3	1.3	1.8	1.8	2.4	2.6	1.5	1.0	2.0	1.2	1.2	2.1	2.8	2.3	1.6	2.6	3.6	1.1	2.8	33.7	-	-				
	Dissolution	1.6	1.6	1.6	1.6	1.8	1.8	1.9	2.0	2.2	2.3	-	1.8	1.5	1.5	1.9	2.2	2.0	1.7	2.3	2.5	1.4	2.3	15.0	1.8	2.0			
Hydrolysis	Depolymerization	-	-	-	-	-	-	-	1.6	1.3	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-				
	Hydrolysis	-	-	-	-	-	-	-	-	-	1.3	-	-	-	-	-	-	-	-	-	-	-	-	-	-				
	CO ₂ emission reduction	6%						100%						100%						100%									

est emissions, with 6.0 kg CO₂ eq. per kg, followed by monomer gasification and pyrolysis to waxes (6.5 kg CO₂ eq/kg input). End products of pyrolysis are assumed to be affected by additive presence; however technologies are being studied to remove flame retardants which may increase the potential of pyrolysis (Yang et al., 2013). This shows to be especially interesting for plastics that perform well with pyrolysis, such as HIPS or PMMA. The effect of flame retardants on gasification products is assumed to be minimal (B. Vreugdenhil, personal communication, October 2020). However it can result in a quality reduction of the output and decrease performance. Relatively, more CO₂ reduction can be obtained by recycling WEEE plastics instead of PE/PP foil, mainly due to the higher environmental impact of ABS production.

The results show that with the case studies, the order of best performing recycling technologies is identical to the LCA matrix model, the absolute environmental impact increases due to sorting effort and boundary conditions for recycling technologies. The overall performance of recycling decreased in all cases, due to adding the energy use of manufacturing of plastic products which is never avoided through recycling.

3.4. Reduction potential

When combining demand volumes of the various polymers in Europe with the LCA matrix model, some interesting patterns emerge on the absolute CO₂ reduction potential through recycling. In Fig. 6, the cumulative CO₂ emissions for the 25 analysed polymers are ranked from high to low contribution to the European footprint. In total, the top 15 polymers which contribute most to the CO₂ emissions in Europe are emitting about 90% or 254 Mtonne CO₂ together, representing a reduction potential of 73% or 200 Mtonne CO₂ when recycled with the environmentally optimal recycling technology. These polymers include polyolefins, which have the highest total CO₂ emissions when demand volumes are assessed, although per kg the CO₂ emissions are the lowest. Emissions are cut significantly when polyolefins are not incinerated (with energy recovery) but recycled through the lowest emission technology (gasification to monomers); 100 Mtonne CO₂, 37% of the emissions of all polymers combined. Polyolefins are used in all sectors, but mostly within packaging industry. This means that these polymers are both demanded in large volumes and end up in higher quantities in waste streams, due to the single use nature of packaging material (Geyer et al., 2017). Hence, emission reduction of treating these streams may be higher than estimated here. Following the polyolefins is PTFE, which is low in demand, but is the polymer with highest environmental impact, mainly due to the high production impact. PTFE shows an emission reduction of 90% when recycled properly through dissolution technology, because of the avoidance of virgin PTFE production. However, PTFE is used in electronics and as Teflon in household products (Omnexus, 2020). PUR and PVC are also in demand in large volumes and are most used in sectors where the lifespans are 20–30 years, such as building and construction. Especially for PUR, current recycling quantities are low (Zevenhoven, 2004). Dissolution shows a strong theoretical potential but as TRL is low, this recycling technology might not be available in the short term. PET has a higher material value compared to the polyolefins and recycling is already more common (particularly for beverage bottles (Yang et al., 2013)), especially in European countries (Eurostat, 2019). The potential reduction that can be obtained by focusing on PET recycling is therefore lower than visualized in the figure. In short, the CO₂ emissions when using the theoretical recycling potential of current plastics in Europe (excluding sorting and product manufacturing) can be reduced to 20% of current CO₂ emissions, indicating that the average circularity potential of plastics is around 80%, where circularity resembles the quantity of which the mate-

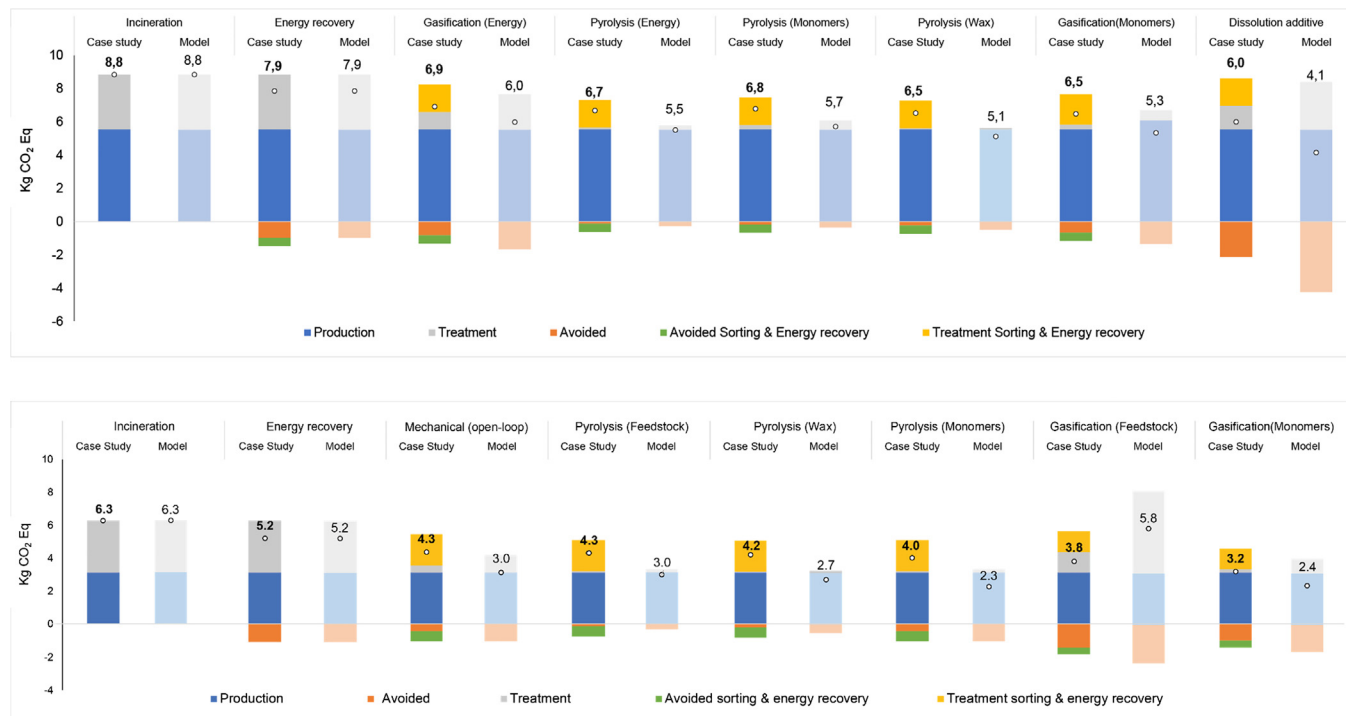


Fig. 5. Comparison of the CO₂ eq. emissions for treating 1 kg of plastic products. with the additional case study framework and the LCA matrix model. (A) the mixed LDPE-PP foil case study with additional sorting effort. per kg mix. (B) the WEEE case study where the ABS plastic contains flame retardants. per kg ABS.

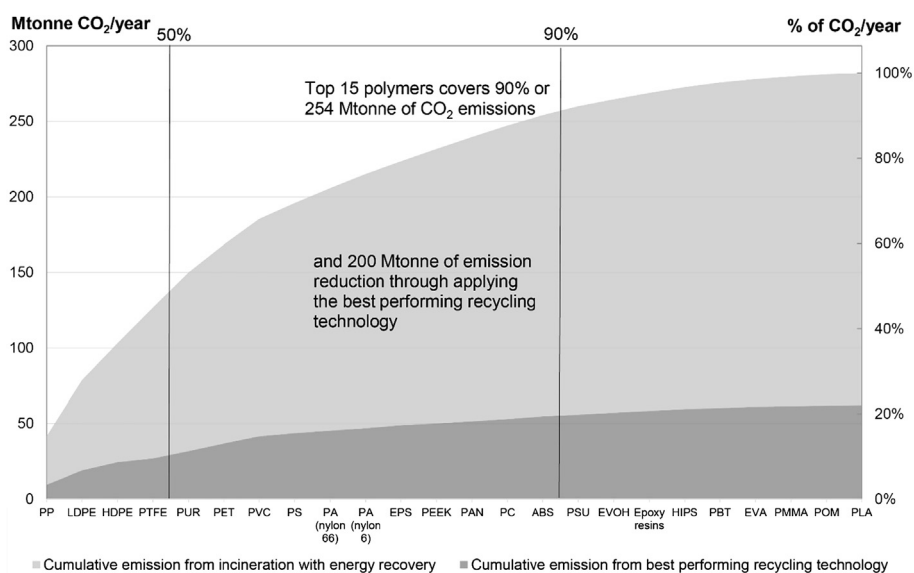


Fig. 6. Estimated CO₂ emissions resulting from recycling the (pure) polymer demand in Europe in 2018. comparing two technology types: incineration with energy recovery and the best performing technology according to the theoretical approach of the LCA matrix model.

rial can be reintroduced as new raw material. This is similar to five additional life cycles, assuming that the carbon emitted is not bound in the polymer material.

4. Discussion

4.1. LCA matrix model and waste pre-treatment

In the LCA matrix model, four main polymer categories were identified. The first are the complex plastics (engineering and high performance plastics) that have lowest environmental impact

through primary recycling (dissolution or closed-loop mechanical recycling). Due to the high environmental impacts of the production phase, recycled quantities and quality of the output increases the environmental performance of the primary recycling technology significantly. However, there are strict boundary conditions for treating the input waste stream with primary recycling technologies. For this the products from the waste streams have to be either pure or well sorted monostream equivalents. In the case studies, the sorting efficiency negatively affected the recycling treatment in various manners, including exclusion of specific recycling treatments (closed loop mechanical recycling) or decreasing

yields due to sorting efficiencies. In the ABS case study, the LCA matrix model determined dissolution as best recycling option. The legacy additives (hazardous flame retardant) disqualified mechanical recycling as recycling option (Wagner and Schlummer, 2020). For dissolution, the technology can treat polymers with additives when supplied to the technology without other impurities and contamination, hence purity through sorting has to be ensured. Improving sorting for ABS from WEEE waste might decrease yield, however, it ensures a primary (polymer) output, which is a valuable trade-off in this case study. The second group of polymers are the polyolefins, which is illustrated in the second case study. This resulted in a low environmental impact with tertiary recycling (gasification to monomers). The main difference with complex plastics described earlier is the lower environmental impacts associated with polymer production. Therefore, the quality-quantity trade-off shifts to a higher benefit of quantity. A lower intactness of the material (lower quality) is then beneficial because of the higher quantities recovered. Gasification with monomer recovery has the benefit that less sorting is required compared to technologies such as primary recycling, as impurities, such as biomass, do not disturb the gasification process (Pinto et al., 2003). Hence, the sorting efficiency was assumed higher, as sorting efforts can be reduced to increase recovery quantities. The sorting effort is mainly required to remove PVC, PS and PET plastic from the polyolefin packaging materials. Presence of oxygen (PET) can result in lower yields, or Cl (PVC) can result in reactor corrosion (Miranda et al., 1998). Thus, waste streams with high polyolefin content, which include packaging waste streams, PE/PP foils as shown in the case studies, or even waste from oceans and rivers (Schwarz et al., 2019), show good environmental results with gasification and pyrolysis. A third group identified in the matrix model are polymers that form monomers during pyrolysis also show a good environmental performance. Sorting and cleaning of these plastics (PS, EPS, HIPS, PMMA) is required as the influence of other polymers (e.g. containing O, N, Cl) or additives (e.g. metals) can reduce monomer concentrations and affect product outcomes (Hujuri et al., 2008). However, the purity degree required is expected to be lower compared to primary recycling technologies as additives are often collected in heavy fractions or tars (Miskolczi et al., 2004). As the fourth and last identified group, thermoset plastics (bisphenol-A epoxy resins and PUR) have lowest recycling potential in the LCA matrix model. If technically possible, dissolution or depolymerization recycling might be promising recycling technologies for thermoset polymers.

The approach of this study illustrated in the environmental most optimal recycling solution, where collection and pre-treatment are excluded in the LCA model to be added as boundary conditions as demonstrated in the case studies. With this approach, the pre-treatment does not limit the recycling, but instead can be optimized based on optimal technology. As illustrated by the case studies, these additional pre-treatment steps affect the environmental performance of technologies. As the best choice of technology was not altered by waste treatment, the LCA matrix model determines the optimal recycling technology and with that the most suitable collection and pre-treatment steps.

4.2. Recycling plastics in a circular economy

The role of plastics in the circular economy is a topic of high interest (Ellen MacArthur Foundation, 2017; European Commission, 2019). According to the LCA matrix model, increasing circularity of plastics through recycling technologies is possible to potentially four to five use cycles, resembling about 80% emission reduction and hence circularity performance. As demonstrated in the case studies this is unlikely to be reached by any recycling technology at the moment due to limitations in collection and

pre-treatments. Furthermore, when plastic products are to be recycled, it will always result into additional environmental impacts over its full life cycle, as the polymer may be kept intact and primary recycled, the product is not. To reach full circularity of plastic products, a push towards the high end of the waste hierarchy is required (re-use, refurbishment and long-lasting design of plastic materials) (Ellen MacArthur Foundation, 2017). In addition, for plastic recycling in a circular economy, recycling technologies have to be specified on properties of plastics in the waste stream, as this determines the environmentally most optimal and circular technology. According to the LCA matrix model, this includes either primary or tertiary recycling methods depending on the polymer. Overall, secondary recycling should be avoided and replaced with tertiary recycling for waste streams that are high in polyolefins and/or polystyrenes, and those collected in mixed waste streams. This result is not in line with the current, state-of-the-art (based on the degree of polymer intactness), recycling hierarchy which ranks secondary recycling above tertiary recycling options, which is still used in policymaking and literature as the status quo, even though it is not in line with this study's LCA results and supports earlier studies questioning the applicability of the waste hierarchy to plastics (Lazarevic et al., 2010). This existing recycling hierarchy shows its limitations and requires revision for a large group of (high demand) polymers.

4.3. Future recycling outlook

The proposed technologies in the matrix show the environmental potential of recycling for a wide range of polymers. However, some of these technologies have a low to medium TRL level indicating these are still in development. This has an effect on the recycling strategies to be determined for the near future. For the near future, suitable recycling technologies with the lowest environmental impact at a high TRL level are required. According to the LCA matrix, gasification has a low environmental impact for the polyolefins and pyrolysis has a low impact considering the polymers HIPS, EPS and PS, which are also produced in large volumes. For these polymers, a transition from the existing quaternary and even secondary recycling technologies towards these tertiary recycling options has a positive environmental impact and will contribute to CO₂ reduction. For the high performance and engineering plastics, an alternative strategy is required to improve environmental performance in the near future. Primary recycling (mechanical and dissolution) performs especially well for these polymers and should be pushed for. For products containing additives, such as WEEE plastics, illustrated in case study 1, primary mechanical recycling options are not possible and for the near future, tertiary recycling to monomers (pyrolysis and gasification) perform best. For the distant future, when dissolution is pushed to high TRL levels, this primary recycling option will result in improved recycling for complex plastics with additives. For the polyolefins and the monomer forming group, this shift is not required as gasification respectively pyrolysis to monomers already performs the best according to the matrix. Technologies that recover feedstock or wax from polymers can serve as an intermediate step in the transition towards optimal recycling of waste streams containing complex engineering polymers and thermosets or heavily contaminated or mixed waste streams. In combination with green electricity mixes in the future, it is likely that environmental impact of recycling is reduced further, as green electricity has lower impacts compared to fossil energy sources.

4.4. Limitations of an LCA approach

There are some methodological limitations to the LCA matrix model due to the LCA approach. An LCA is based on its system

boundaries, input data and assumptions. When these vary between studies, different outcomes and hence different conclusions are drawn. For example, Meys et al., 2020, using an LCA approach, concludes that quaternary recycling technologies outperform specific tertiary (chemical) recycling options. The system boundaries of this study and Meys et al., 2020 vary strongly. A main difference is that in this study, there is a focus on the potential of a technology, while Meys et al., 2020 uses a status quo approach, for example using a mix of plastics instead of pure polymers (Meys et al., 2020). Another important factor that has its limitations in LCA is quality. With open-loop mechanical recycling, significant quality reduction is observed, especially when applied to mixed waste streams (Jeannette M. Garcia, 2017). In LCA, the quality aspect is difficult to address. In this study, a quality factor is applied for secondary recycling to address quality reduction. However, the final results of the secondary recycling technology is very sensitive depending on the quality factor chosen, because substituted material plays an important role in the final LCA outcomes and hence is key on the performance of a technology. This supports the key role of quality of products when considering recycling of materials. Furthermore, with a quality factor the total mass balance of avoided materials is affected. If the polymers are used in products that are valued as 'lower grade' (e.g. plant pots, plastic lumber or roadside posts), an alternative credit method should be established instead of avoided products. In literature options are given through value-corrected substitution (Ligthart and Ansems, 2012), defining a substitution potential (Vadenbo et al., 2017) or using circularity indicators (Bracquen   et al., 2020). A related issue is the 'reward' of avoided impact for multiple uses (extended lifetime) of the materials. This is clearest when recycling to feedstock is applied, as in the subsequent life cycle, the feedstock is burned and stored carbon and other molecules are emitted to the atmosphere. The material is used in two life cycles but emissions during the second life cycle are not accounted for in the first life cycle. With a 'system expansion' approach, this can be put better into perspective (Shen et al., 2010).

4.5. Boundaries of the LCA matrix model

This study gives a framework for multiple recycling technologies, however the assumptions that are taken for the recycling technologies in the model come with boundaries and side notes. The model visualizes the theoretical, potential environmental impact of recycling. Local variables occur for all technologies and the model results are not applicable to specific recycling plants. The case studies that were developed propose the additional steps required to apply the model to existing recycling challenges. In order to structurally and coherently add the existing framework for these boundaries, such as performance, efficiencies and purity of waste streams, a full decision tool has to be developed together with the LCA matrix model results. Additionally, a waste perspective as executed in this study, can be adapted to a product perspective, where the different recycling technologies are compared through 1 kg of end product and not 1 kg of waste. Furthermore, for low to medium TRL technologies, lab or pilot level data used which can vary from large scale plants. Prospective LCA elements can help to improve these assumptions when technologies are applied to larger scales. These can be extended with data in future electricity mixes and the effect on environmental impacts. At last, the model is now limited to showing the dimensions of circularity and environmental impact, which is one of the prime motivations for recycling. In order for the technologies to be incorporated, life cycle costing can be incorporated in order to make viable decisions on recycling technologies.

5. Conclusions

The LCA matrix model demonstrates that the optimal recycling technology varies per polymer and that a multiple of technologies are to be applied for different plastic waste streams in a circular economy. Polyolefins have lowest environmental impact through gasification to monomers, with a CO₂ -eq of 0.9–1.1 kg per kg, more than secondary and even primary recycling alternatives, which challenges the existing recycling hierarchy. This is also the case for the plastics forming monomers, which can be recycled through pyrolysis to monomers, with a CO₂ -eq of 1.4–2.7 kg per kg. More high performance and engineering plastics still benefit most from primary recycling, such as dissolution or closed-loop mechanical recycling. The LCA matrix model assesses potential environmental benefit not limited by pre-treatment. Optimal environmental performance of recycling can only be obtained where pre-treatment is improved in line with the most suitable recycling method for a polymer. The case studies underline this, showing that of the quality of the polymers (mixed source, mixed materials) is important and affects the overall environmental performance of a technology; however not altering the performance ranking of the technologies. At last, a large CO₂ reduction potential is available by improving recycling. By executing recycling (and sorting) of the 15 most demanded polymers in Europe, life cycle CO₂ emissions from plastics can be reduced by 73% or 200 Mtonne CO₂ eq.

Declaration of Competing Interest

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

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2020.12.020>.

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