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Moving from linear to circular household plastic packaging in Belgium : prospective life cycle assessment of mechanical and thermochemical recycling

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- 1 Moving from linear to circular household plastic packaging in Belgium: prospective life cycle
- 2 assessment of mechanical and thermochemical recycling
- 3 Didem Civancik-Uslu 19; Trang. T. Nhu 19; Bart Van Gorp²; Uros Kresovic³; Macarena Larrain^{4,5}; Pieter
- 4 Billen⁴; Kim Ragaert⁶; Steven De Meester⁷; Jo Dewulf¹; Sophie Huysveld^{1*}

- 6 ¹Sustainable Systems Engineering (STEN), Department of Green Chemistry and Technology, Faculty of
- 7 Bioscience Engineering, Ghent University, Coupure Links 653, 9000 Ghent, Belgium
- 8 ²ECO-oh! Recycling, Europark 1075, 3530 Houthalen, Belgium
- 9 ³WordMil bvba, Max Havelaarplein 21, 3111 Wezemaal, Belgium
- ⁴iPRACS, Faculty of Applied Engineering, University of Antwerp, Groenenborgerlaan 171, 2020
- 11 Antwerpen, Belgium
- 12 ⁵Department of Engineering Management, Faculty of Business and Economics, University of Antwerp,
- 13 Prinsstraat 13, 2000 Antwerpen, Belgium
- 14 ⁶Center for Polymer and Material Technologies (CPMT), Department of Materials, Textiles and Chemical
- 15 Engineering, Faculty of Engineering and Architecture, Ghent University, Technologiepark 130, 9052
- 16 Zwijnaarde, Belgium
- 17 Taboratory for Circular Process Engineering (LCPE), Department of Green Chemistry and Technology,
- 18 Faculty of Bioscience Engineering, Ghent University, Graaf Karel de Goedelaan 5, 8500 Kortrijk, Belgium
- 19 §These authors contributed equally to this work.
- ^{*}Corresponding author: Sophie.Huysveld@ugent.be

ABSTRACT

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Currently, Belgium is in a transition period after which more household plastic packaging waste will be collected separately in function of increased recycling. The challenge is to identify the most environmentally sound treatment option for the increased selectively collected plastic waste. In this study, mechanical recycling (MR) and thermochemical recycling (TCR) of four newly collected subfractions, being polypropylene (PP), polystyrene (PS), mixed polyolefins (MPO) rigids and polyethylene (PE) films, were investigated through prospective Life Cycle Assessment (LCA), in comparison to incineration with energy recovery. Results showed clear benefits of recycling over incineration with energy recovery. Generally, MR showed a better net environmental impact compared to TCR (for PP, PS, MPO rigids and PE films, respectively, e.g., a global warming impact of 100, -1580, 539 and 101 kg CO₂ eq. per ton by TCR, and -1183, -3096, -319 and -1162 kg CO₂ eq. per ton by MR, and 2339, 2494, 2108 and 2141 kg CO₂ eq. per ton by incineration). This could mainly be explained by the avoided burdens of virgin materials. Whereas TCR avoids the virgin supply of the feedstock for polymer production, MR avoids additionally polymerisation and granulation. MR products, i.e. regranulates or flakes, can be directly used in manufacturing, whereas TCR products require first processes like steam cracking, polymerisation and granulation before being used in manufacturing. As this study assumed a 1:1 substitution ratio between MR regranulates and their virgin alternatives, it presents the most favourable results for MR, which should be kept in mind and further investigated.

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- **KEYWORDS**: Mechanical recycling; Thermochemical recycling; Life Cycle Assessment, Plastic packaging;
- 41 Circular economy, post-consumer plastic waste

1 Introduction

In 2018, 29.1 million tons of post-consumer plastic waste were collected in Europe, of which only 32% was sent to mechanical recycling, 43% to energy recovery and 25% to landfill (PlasticsEurope, 2019). The circular economy concept aims to optimize resource use by providing circularity of products, components and materials by means of enhancing maintenance, reuse, remanufacture and recycle (Ellen MacArthur Foundation, 2012). To this end, increasing recycling rates of plastic waste is vital for the transition to a circular economy (European Commission, 2020). In this context, the European Commission (EC) has set a 50% recycling target for all the plastic packaging waste collected by 2025 and 55% by 2030 (European Commission, 2018a).

48% of post-consumer plastic waste was separately collected in Europe in 2018, while the remaining 52% was collected in residual waste fractions. The recycling rates were 62 and 6%, respectively, showing the impact of separate collection on recycling rates (PlasticsEurope, 2019). The collection of more plastic waste through an enhanced separate collection is vital to increase resource efficiency and contribute to circular economy targets (Tallentire and Steubing, 2020). Since 1994, Belgium has a kerbside collection system for some household's plastic packaging waste fraction with the so-called PMD system: it comprises Plastic bottles and flasks that are collected together with Metal packaging and Drink cartons. Belgium is currently in a transition phase during which an enhanced P+MD collection system is being introduced, also including other plastic packaging fractions like films, trays, tubes, etc., into a single bag. From 2021 onwards, the separately collected P+MD waste will be sorted in 14 fractions including 11 plastic fractions (containing a residual fraction), 2 metal fractions and drink cartons (Fostplus, 2019). The newly sorted plastic waste streams will include *polypropylene (PP) rigid, polystyrene (PS) rigid, mixed polyolefins (MPO) rigid, polyethylene (PE) films, and other films*. Evaluation of similar collection system expansions in neighbouring countries has shown that this leads to a significant reduction of plastic waste to be incinerated as a part of the residual household waste (Brouwer et al., 2019).

- There are two possible pathways of recycling for the individual fractions: mechanical and thermochemical.
- 67 Mechanical recycling (MR) is the recovery of plastics via mechanical means and leads to regranulates,
- 68 whereas in thermochemical recycling (TCR) plastics are converted into monomer building blocks (Ragaert
- 69 et al., 2017).
- 70 The challenge is to identify the environmentally most promising method for treatment of household
- 71 plastic waste within an economic context. Life cycle assessment (LCA) is a method that is used to compare
- 72 the environmental profile of different treatment options for plastic waste (Lazarevic et al., 2010).

Recycling is generally identified as a better solution than landfilling and incineration with energy recovery (Alston and Arnold, 2011; Gear et al., 2018; Hou et al., 2018; Perugini et al., 2005). One of the major benefits of recycling of plastics is the resource savings (Al-Salem et al., 2017). Incineration causes carbon emissions to the atmosphere, therefore contributes to global warming, whereas landfilling requires space (Khoo, 2019).

Household plastic waste is a complex stream to recycle, because its composition is usually hard to know and contaminated by organic and inorganic fractions (Ragaert et al., 2017). Its heterogeneous structure may affect the final product quality in mechanical recycling (Khoo, 2019; Ragaert et al., 2017; Rigamonti et al., 2020). Currently, plastic waste that cannot be recycled mechanically is sent to incineration with energy production in Europe. However, recent developments in chemical recycling have shown its potential to deal with heterogeneous streams like household plastic waste (PlasticsEurope, 2019).

A limited number of studies comparing environmental impacts of MR and TCR of plastic packaging waste was identified. By means of LCA, Khoo (2019) investigated 8 scenarios which are different combinations of TCR (i.e. gasification and pyrolysis), MR and waste-to-energy. The results showed that global warming impact of MR is lower than pyrolysis and slightly higher than gasification. Other studies (Chen et al., 2011; Cossu et al., 2017), however, showed higher global warming impacts than the study of Khoo (2019), but did not directly compare it to MR. The studies mentioned up to here considered plastic packaging waste as a mixed waste stream. To our knowledge only Meys et al. (2020) studied different polymer types as separate plastic waste streams, but this study was based on a theoretical chemical recycling model assuming ideal performance; it compared the environmental potential of chemical recycling technologies with real-case benchmark treatments, i.e. energy recovery in waste incineration, in cement kiln and MR. It was concluded that recycling into monomers and value-added products could reduce global warming impact compared to energy recovery in waste incineration, cement kiln and MR.

In this article we provide a prospective life cycle assessment of two possible recycling pathways, i.e. MR and TCR, for four newly collected and sorted plastic waste fractions in Belgium, which are *PP*, *PS*, *MPO rigids* and *PE films* separately. The results rely on the detailed design of MR and TCR processes of Belgian key actors for each specific waste fraction, considering realistic performances and taking into account impurities and waste object physical properties, and are presented in comparison to the current incineration as a benchmark scenario. Impacts from sorting the collected commingled bag in separate plastic fractions were included in case of recycling to be able to make a fair comparison between recycling and incineration as incineration does not require sorting. Sorting of the collected mixed waste has been

omitted in other LCA studies (Cossu et al., 2017; Meys et al., 2020). Landfilling was not selected for the assessment because it is not an option in Belgium (European Environment Agency, 2016).

2 Materials and methods

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106 107 2.1 Goal and scope definition 108 In this research, a prospective LCA on MR and TCR of four newly sorted plastic waste fractions, i.e. PP, PS, 109 MPO rigids and PE films, was performed following the ISO 14040/44 standards (ISO, 2006a, 2006b). 110 The functional unit was defined as 1 metric ton of a particular household plastic waste fraction (either PP, 111 PS, MPO rigids or PE films) to be treated through either MR or TCR. While doing this comparison, 112 incineration was considered as a benchmark scenario. The fraction other films was not investigated 113 because MR was considered not feasible by MR experts at this stage, based on the stream's very complex 114 composition (Ragaert et al., 2020). 115 The system boundary of the LCA starts when the household's mixed packaging waste, i.e. the P+MD bag, 116 enters the sorting facility. After being sorted, waste fractions continue to the recycling facility with 117 necessary treatment steps depending on the composition of each plastic waste fraction. Impacts from the 118 sorting process were included in case of recycling to be able to make a fair comparison between recycling 119 and incineration as incineration does not require sorting. The sorting process was modelled as a black box 120 (see for allocation, section **2.2.4**). 121 Depending on the modelled composition of each plastic waste fraction, prospective MR and TCR processes 122 were designed in a different way in collaboration with recycling experts from both academia and industrial 123 actors based on currently known technologies. The composition of each waste fraction after sorting was 124 modelled based on the studies of Roosen et al. (2020) and Kleinhans et al. (2020), see Table 1. More 125 detailed information on this waste's heterogeneity can be found in Roosen et al. (2020) and Kleinhans et

al. (2020). In the following sections, the MR and TCR scenarios are explained in detail.

Table 1. Composition of plastic waste fractions after sorting (%)

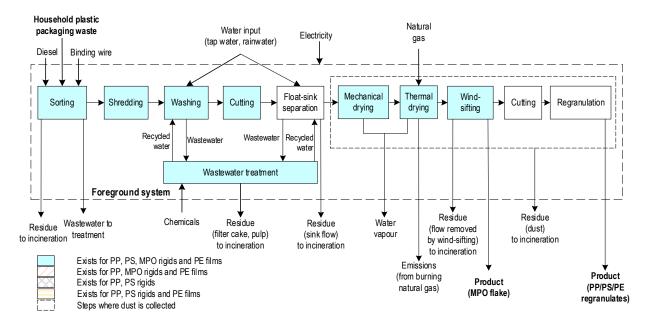
	PP rigid (%)	PS rigid (%)	MPO rigid (%)	PE films (%)
PP	90	-	25.9	-
PS	-	93.2	-	-
PE	3.5	0.2	48.2	78.8
PET	0.5	0.5	1.1	-
Dirt+moisture	5.3	4.2	6	9.1
Others (missorted plastics, EVOH, paper, etc.)	0.7	1.9	18.8	12.1

2.1.1 Mechanical recycling (MR)

An overview of the MR processes and the final products is presented in **Figure 1a**. After sorting, it starts with the pre-treatment steps, including shredding, washing and cutting into smaller pieces for removal of contaminants. Next, the waste flow is split through float-sink separation; the float fraction goes to mechanical and thermal drying, while the sink fraction goes to incineration. In case of the *PS rigid* fraction, this separation was not necessary because this fraction has a relatively low contaminant level and does not float in water, like polyolefins to separate it from other plastics. In the washing and float-sink separation, tap water and rainwater are used and treated afterwards at the MR facility by physical and chemical treatment methods. While the treated water is recycled, the filter cake and pulp residues from the wastewater treatment system go to incineration with energy recovery.

Natural gas is used as an energy source for thermal dryers. Next, wind sifting removes film particles from the fraction. For the *PP rigid* and *PS rigid* fractions, this is a residue that goes to incineration with energy recovery, while the main flow is further cut and regranulated with melt filtration. For the *PE films* fraction, it is actually the wind sifted sub-fraction that is the target fraction which then goes to regranulation. The cutting before regranulation is not necessary for a film fraction. The non-wind sifted residue from the *PE films* fraction goes to incineration with energy recovery. Cutting and regranulation were not included for the *MPO rigid* processing because MPO flakes are considered as a marketable recycled product. Finally, dust is collected at several steps following the mechanical drying and sent to incineration with energy recovery.

149 a. Mechanical recycling (MR)



b. Thermochemical recycling (TCR)

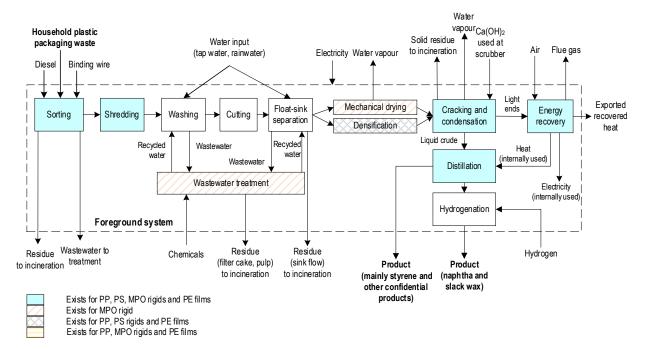


Figure 1. Flow diagram of (a) MR and (b) TCR scenario for four plastic fractions: *PP, PS, MPO rigids* and *PE films*, including inputs and outputs in the foreground system. The sorting plant (including different processes) was modelled as a black box. Regranulation includes melt filtration; however, the mass flow separated by melt filtration was considered negligible.

2.1.2 Thermochemical recycling (TCR)

An overview of the TCR process is presented in **Figure1b.** For all fractions, shredding is included with the aim of size reduction, but some other pre-treatment steps are only required depending on the composition of each waste stream. Due to the more contaminated and heterogeneous composition of the *MPO rigid* fraction, washing, cutting and float-sink separation are included to remove dirt, paper and missorted plastics such as PET, the latter to be removed because of its corrosive character (Butler et al., 2011). Tap water and rainwater are used in washing and float-sink separation and treated in a water treatment plant to be recycled. Residues (pulp and filter cake) from the water treatment plant are sent to incineration with energy recovery. The sink fraction from the float-sink separation also goes to incineration with energy recovery, while the float fraction continues to the densification, which is an extrusion process to reduce the water content before entering the cracking. In case of the *MPO rigid* fraction, a mechanical dryer is used instead of densification because it is sufficient to remove water.

After pre-treatment, cracking and condensation follow. Liquid products (with 6 carbon atoms or higher) from the cracker continue to distillation for further separation. After that, hydrogenation increases the product quality. Hydrogenation is not needed in case of PS *rigid* fraction because the product achieved after distillation is mainly styrene. The light ends (with lower than 6 carbon atoms) of the cracking are used for energy purposes, while the crude portion of it, which is HCl, is treated with Ca(OH)₂ in a scrubber before going to incineration with energy recovery as a part of the solid residue from cracking. Light ends go to energy recovery unit where electricity and heat are recovered. Recovered electricity and heat are consumed internally; the excess amount is exported except for *PS rigid* as there is no excess in this case. For the incineration of the residue from the cracker, energy recovery with metals recovery from bottom ash is considered. Final products from TCR are naphtha and slack wax for the *PP rigid*, *MPO rigid* and *PE films* fractions, while in case of the *PS rigid* fraction they are mainly styrene monomers.

2.2 Data Inventory

The studied system in each scenario can be divided into a foreground and a background system. The foreground system corresponds to all processes within the dashed frames in **Figure 1.** The background system consists of the processes which are outside of the foreground system (e.g. electricity production, tap water production, etc.). In this study, detailed data required for the foreground system were gathered from Belgian key actors and adapted for each waste fraction in collaboration with experts from both academia and industry during the period 2018-2020, whereas the data for the background system was retrieved from secondary sources like LCA databases.

2.2.1 Foreground system

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2.2.1.1 Mechanical recycling (MR)

Process flow diagrams and mass and energy data on MR of mixed household plastic waste were mainly established with the Belgian company ECO-oh!. Taking into account the composition of each waste fraction with its targeted plastic, missorted plastic, paper, dirt and moisture. All dirt and 95% of the paper in the waste stream were assumed to be removed during the washing step (based on Brouwer et al. (2018)) and end in filter cake and pulp. The remaining 5% of the paper goes to the sink flow through floatsink separation, except in case of the PS rigid fraction, where there is no float-sink separation; it was assumed to be lost as dust. 3% of dry plastic entering the MR facility is lost as dust (2.4%) and wastewater treatment residue (pulp, 0.6%). In case of the PS rigid fraction, dust was assumed to contain also paper besides plastic. The remaining plastic in the waste stream goes partly to the float flow and partly to the sink flow through float-sink separation. The float-sink mass balance could be defined based on the composition of each waste fraction after sorting (see Table 1), which was modelled based on the studies of Roosen et al. (2020) (waste composition before sorting) and Kleinhans et al. (2020) (sorting flows). The composition was specified in terms of material type (PE, PP, paper, Al, etc.) as well as original product (e.g. a bottle of a certain brand) and its components (e.g. bottle, cap, label etc.). For single-polymer plastic flakes (obtained after shredding and cutting), originating from a specific component (e.g. cap) of a specific representative waste item (e.g. Coca-Cola bottle), the float and sink flows were calculated based on the transfer coefficients mentioned in Table A.1 of Brouwer et al. (2018). For multi-material flakes coming from multilayers, the density of the multilayers was used to estimate the share between the float (density <10³ kg/m³) and sink (density >10³ kg/m³) flows. For the wind sifting mass balance, it was assumed that flexibles (films, labels and lids) are separated with 99% to the light fraction, while rigids (bottles, traps, caps) with the same efficiency to the heavy fraction. Finally, the remaining mass flow leading to the product was calculated (see the supplementary material, **Table S.1**). Next, the total energy consumption (incl. electricity and thermal energy of natural gas) was calculated based on Larrain et al. (under review). For a specific step (e.g. shredding), its energy consumption was obtained by multiplying its specific energy consumption (SEC) per dry ton processed. Several approaches were applied to estimate the SEC of each MR step. Firstly, the SEC of shredding, washing, cutting, float-

sink separation and mechanical drying was determined based on equipment specifications, i.e. power and

maximum throughput. Secondly, the SEC of thermal drying and regranulation was calculated based on the

physical characteristics and thermodynamic properties of each plastic fraction, taking into account energy

required for heating, melting and extruding. The latter was estimated based on defining the residual moisture content, expressed as the ratio between the water mass and the dry plastic mass of the flow coming into and going out the thermal dryers using the approach derived from Horodytska et al. (2018b). The efficiencies of thermal drying (48.9%) (Kemp, 2012) and extrusion (40%) (Chung, 2000) were also taken into account. Details of the foreground inventory data of MR can be found in the supplementary material (**Table S.2**).

2.2.1.2 Thermochemical recycling (TCR)

The mass and energy data required for the TCR foreground system were modelled on the basis of the chemical recycling company's pilot plant design specifications (scale: 50 kton for PS and scale: 120 kton for polyolefins (PO)). While the data for PS were directly used for the *PS rigid* fraction, experts relied on these data to model the processes for the *PP rigid*, *MPO rigid* and *PE films* fractions. Data for the pretreatment steps (i.e. washing, cutting and float-sink separation) were taken from the MR scenario in case of *MPO rigid* fraction. The water content of the waste fraction was reduced to 0.5% by densification for *PP rigid*, *PS rigid* and *PE films* and to 2.2% by mechanical drying in case of *MPO rigid*. The remaining water content was removed at the cracking and condensation stage. The modelling accounts for 90% yield of into liquid and gaseous products at the cracking. The remaining 10% was either lost as moisture or solid residue. Gaseous products form 11% of the products from the cracker in case of the *PP rigid*, *MPO rigid* and *PE films fractions*, whereas they were 5% in case of the *PS rigid* fraction. This gaseous fraction went to energy recovery unit and CO₂ release was considered. The rest of the products from the cracker continued to distillation and hydrogenation depending on the waste fraction.

Regarding electricity consumption, the same data were used as for MR, where applicable. For TCR-specific processes like densification, cracking and condensation and distillation, data on electricity consumption was provided by the recycling company and implemented for all the fractions. For heat, thermal oil was used for all waste fractions except the *PS rigid* fraction, where steam was used. The reason for that was the higher operational temperature of the distillation process in case of treatment of PO waste (350°C) compared to treatment of PS waste (100°C). Heat required to produce steam was assumed to be recovered from a nearby municipal waste incineration plant and being burden-free (see section 2.2.4). For the hydrogenation step, hydrogen was provided by a nearby cracking facility via a pipeline. In the foreground modelling of both MR and TCR, impacts of the infrastructure were excluded. Details of the foreground inventory data of TCR can be found in the supplementary material (Table S.3).

2.2.2 Background system

For the calculation of impacts from the background system (i.e. electricity, water, heat, chemicals, etc.) of MR and TCR, the ecoinvent v3.6 cut-off modelling library was used in SimaPro v.9 by excluding the impacts from infrastructure and long-term emissions. The most representative data were chosen from the database. A list of datasets can be found in the supplementary material in **Table S.4**. The impacts from incinerating residues (i.e. filter cake, pulp, dust, flow removed by wind shifting, sink flow and solid residue from the cracker) were quantified on the dry basis of residues.

Additionally, we noticed that, for some impact categories excluding global warming (e.g. resource consumption, terrestrial acidification), the environmental impact of virgin PS granulates (used to calculate the avoided burdens in case of MR) was unrealistically lower compared to virgin styrene (used to calculate the avoided burdens in case of TCR) based on the ecoinvent v3.6 database. This can probably be explained by the fact that the styrene dataset in ecoinvent v3.6 has been updated and is present in a disaggregated format, while the PS dataset is still in an aggregated format that will be updated only in the future by ecoinvent. As a sensitivity analysis, we therefore modelled the impact of virgin PS production starting from the styrene dataset and adding the average impact of polymerisation and granulation for PP, HDPE and LDPE (see section **3.3** and the supplementary material, section **C**).

2.2.3 Incineration (Benchmark scenario)

For the modelling of impacts from incineration of each waste fraction, calculations were done based on their defined compositions by considering the impacts of incineration of each component on a dry basis (e.g. PE, PS, paper, etc). With that purpose, the most representative data were used from the ecoinvent v3.6 database because no primary data were available. These data from ecoinvent can be considered representative for the praxis in Belgium. Details of the datasets used are provided in the supplementary material in **Table S.4**. In contrary to the MR and TCR scenarios, for which primary data were collected to model the foreground system, for modelling the incineration benchmark scenario we relied entirely upon secondary data. Impacts of infrastructure were excluded from the ecoinvent datasets to be consistent with the modelling of MR and TCR scenarios.

2.2.4 Allocation

In this study, a waste perspective LCA was applied with a "cut-off approach" (Baumann and Tillman, 2004), meaning that waste was considered burden-free. Following the same approach, heat from a municipal

waste incineration plant was also considered as burden-free (section **2.2.1.2**). For the allocation of the impacts of sorting process, mass allocation was done to the sorted fractions.

When crediting the system for the final products, avoided burdens were calculated based on the idea that recycled products (e.g. naphtha, regranulates, etc.) replace virgin materials at a 1:1 substitution ratio, meaning that the final products from MR and TCR have the same quality as their virgin counterparts (e.g. 1 ton of PP regranulates replace 1 ton of virgin PP). This study therefore presents the most favourable results for MR and TCR, which should be kept in mind when interpreting the results. On the one hand, assuming a 1:1 substitution ratio for the TCR products could be justified as post-treatment steps (distillation and hydrogenation) were included to increase the products' quality to a level similar to that of the virgin alternatives. On the other hand, MR products may have a lower technical quality than their virgin alternatives and also a limited market, eventually lowering the actual savings from avoiding virgin material production (Vadenbo et al., 2017). A sensitivity analysis on the substitution ratio for MR products is provided in section 3.4. The recycled products from MR and TCR and their corresponding virgin alternatives are listed in Table 2. The 1:1 substitution ratio was also applied when calculating the avoided burdens of electricity and heat production whenever incineration with energy recovery was considered.

Table 2. Overview of the recycled products and the substituted virgin alternatives in the mechanical recycling (MR) and thermochemical recycling (TCR) scenarios.

		PP rigid	PS rigid	MPO rigid	PE films
MR	Recycled product	PP regranulates	PS regranulates	MPO flakes ^a	LDPE regranulates
	Substituted virgin alternative	virgin PP granulates	virgin PS granulates	virgin HDPE granulates ^b	virgin LDPE granulates
TCR	Recycled product	naphtha and slack wax	mainly styrene ^c	naphtha and slack wax	naphtha and slack wax
	Substituted virgin alternatives	naphtha and slack wax	mainly styrene ^c	naphtha and slack wax	naphtha and slack wax

^a Flakes were considered instead of regranulates because experts have judged that potential applications for this fraction (street bench, pallet, etc.) allow direct extrusion from flakes without prior regranulation.

^b The substituted virgin alternative depends on the application of MPO flakes. In this study, we choose virgin HDPE granulates used for street bench production.

^cThere are minor amounts of other products in addition to styrene which cannot be disclosed because of confidentiality.

2.3 Impact assessment

Two impact assessment methods were chosen: (i) Cumulative Exergy Extraction from the Natural Environment (CEENE) v2013 (Alvarenga et al., 2013; Dewulf et al., 2007) for consumption of natural resources to establish a resource footprint (Berger et al., 2020) and (ii) ReCiPe 2016 (H) Midpoint v1.1 (Huijbregts et al., 2017) for impacts from emissions. CEENE results are presented as CEENE total which is the total consumption of natural resources, including fossil, nuclear, renewable (wind and hydro energy), water, mineral, metal and land resources, in Joules of exergy (Jex). In addition, global warming (kg CO₂ eq.) and terrestrial acidification (kg SO₂ eq.) impacts calculated with the ReCiPe method are presented as they are the most commonly studied impact categories among the LCA studies in plastic waste management (Lazarevic et al., 2010). ReCiPe 2016 was chosen as it can be considered as a state-of-the-art method for global warming and terrestrial acidification impacts, providing characterization factors representative on the global scale (Huijbregts et al., 2017). For global warming, ReCiPe 2016 relies on the fifth and latest assessment report of the IPCC (2013). Regarding terrestrial acidification, ReCiPe 2016 relies on Roy et al. (2014), providing spatially explicit characterization factors covering the global scale. The results of resource consumption and global warming impact are presented in sections 3.1 and 3.2, whereas terrestrial acidification results are presented in the supplementary material (section B).

3 Results

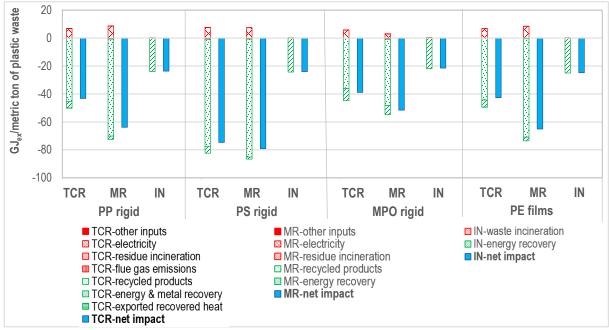
Figure 2 shows the potential environmental impacts (i.e., resource consumption and global warming) per ton plastic fraction treated through mechanical recycling (MR), thermochemical recycling (TCR) and incineration (IN), with in total twelve scenarios (i.e., three treatment options x four plastic fractions). For each scenario, the environmental impacts are presented in two ways, i.e. (i) a stacked bar composed of burdens (positive value; in red) and savings (negative value; in green), and (ii) a blue bar representing the net impact (burdens - savings).

3.1 Resource consumption

In **Figure 2a**, the resource consumption is expressed as the aggregated total CEENE (GJ_{ex}), which is the sum of eight impact categories quantified by the CEENE method (section **2.3**). The sorting plant was modelled as a black box; its net impact is negative (-0.9 GJ_{ex} /ton sorted waste), which is explained by the savings from energy recovery in the incineration of residues sorted from the collected P+MD waste.

For all of the four plastic fractions, electricity use is the main contributor to the environmental burdens of the two recycling processes in terms of resource consumption. Consequently, attention should be paid to the reduction of electricity use in the most energy-intensive steps of the recycling processes. It is regranulation in MR of the *PP rigid*, *PS rigid* and *PE films* fractions (which consumes 51, 44 and 43% of the total electricity consumption, respectively), shredding and cutting in MR of the *MPO rigid* fraction (which consumes 23 and 20% of the total electricity consumption, respectively). In case of TCR, it is cracking and condensation for all plastic fractions with 74, 71, 63 and 74% of the total electricity consumption for the *PP, PS, MPO rigids* and *PE films* fractions, respectively).

a. Resource consumption



b. Global warming

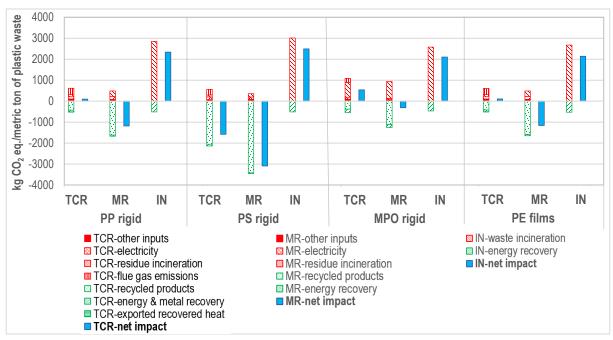


Figure 2. Potential environmental impacts: (a) resource consumption and (b) global warming impact of the three analysed treatment options: thermochemical recycling (TCR), mechanical recycling (MR) and incineration (IN) for the four plastic fractions: *PP, PS, MPO rigids* and *PE films*. Positive values on the y-as represent burdens, while negative values represent savings. Other inputs (TCR/MR): the burdens of sorting inputs, chemicals, water and heat, except electricity which is presented separately.

It is also noted that the burdens of electricity used in MR of the *MPO rigid* fraction is lower than in case of the other fractions. The main reason is that the second cutting process and the regranulation, the latter being the most energy-intensive step in MR, was not necessary in case of the *MPO rigid* fraction (**Figure 1**). The second reason is the lower (dry) flow in the processes following float-sink separation in case of the *MPO rigid* fraction and, therefore, a lower energy consumption of these steps. The lower (dry) flow can be explained by a higher share of contaminants (34.9%, incl. dirt, paper and mainly missorted plastics) compared to the other waste streams (3.6% for *PP rigid*, 3.2% for *PS rigid* and 22.5% for *PE films*), which are mainly removed through washing and float-sink separation.

The resource savings of the two recycling processes mainly come from the substituted virgin materials. This saving is quite higher for MR compared to TCR per ton waste (Figure2a). This will be further discussed in section 4.1. Additionally, the avoided burdens of electricity and heat due to the incineration of residues also contribute to the resource savings of TCR and MR.

For all of the four plastic fractions, the calculated net impacts for MR, TCR and IN are negative values, meaning that their products are environmentally more beneficial in terms of resource consumption than the production of their virgin alternatives. Comparing the net impacts amongst the three treatment options, the lowest (best) values are obtained for MR, while the highest (worst) are obtained for incineration, keeping in mind that this study presents full substitutability of virgin granulates by regranulates and flakes (section 2.2.4).

The benefit in terms of resource consumption between MR or TCR and incineration as benchmark was calculated by the absolute difference in the net impacts of two treatment options. TCR and MR achieve the highest benefit for the *PS rigid* fraction (50.7 and 55.1 GJ_{ex}/ton *PS rigid*, respectively) and the lowest benefit for the *MPO rigid* fraction (17.3 and 30.1 GJ_{ex}/ton *MPO rigid*, respectively). The reason for the former is that the two recycling processes gain the highest savings from the substitution of the recycled products: PS regranulates and recycled styrene (see further discussion in section **4.1**). The latter is explained by the high amount of missorted plastic that mainly ends in the sink flow for incineration in case of the *MPO rigid* fraction. This results in lower resource savings because of a lower yield of recycled products. As an example, MR delivers only 581 kg of dry *MPO flakes* compared to 848 kg of dry *PP regranulates*, while recycled naphtha and slack wax produced in TCR amounts only to 637 kg/ton *MPO rigid* compared to 814 kg/ton *PP rigid*. This reason also leads to the same conclusion for the other impact categories: global warming (section **3.2**) and terrestrial acidification (section **B**).

3.2 **Global warming**

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For all of the four plastic fractions, the global warming burdens of MR mainly originate from the incineration of residues and electricity use. The flue gas emissions from energy recovery together with these two sources are identified as the hotspots of the global warming burdens of TCR (Figure 2b). It highlights that there is room for improvement with TCR through reduction of electricity use (discussed in section 3.1) and of flue gas emission, while the savings from incinerating TCR residues (i.e. energy recovery) can offset its burden. The sorting inputs induce 74 kg CO₂ eq./ton sorted waste and contribute 12, 13, 7 and 12% to the burdens of MR for the PP, PS, MPO rigids and PE films fractions, respectively.

These values are 15, 21, 8 and 15% in case of TCR.

The analysis also shows that the global warming burdens of both recycling options for the MPO rigid fraction are higher than for the other plastic fractions due to a large contribution of the incineration of residues generated in MR and TCR pre-treatment. Due to a higher share of contaminants (34.9%) in the MPO rigid stream, TCR of this fraction requires pre-treatment and induces a higher amount of residues. Following the same reasoning, the burden from incineration of MR residues is considerably higher for the MPO rigid fraction than for the other fractions. Correspondingly, the savings by electricity and heat produced from incinerating TCR and MR residues are lower for the other fractions than for the MPO rigid fraction, which can be seen in all of the three considered impact categories, particularly in resource consumption (Figure 2a).

The calculated net impacts for MR are negative values for all four plastic fractions. This does not indicate that MR is a sink of greenhouse gas emissions but means that MR gains environmental savings (i.e., the negative part of the stacked bars) predominantly from avoiding virgin material production considerably higher than its burdens (i.e., the positive part of the stacked bars) in terms of global warming. The net impacts of TCR are positive (except for the PS rigid fraction), meaning that TCR products are environmentally less beneficial in terms of global warming than the production of their virgin alternatives in case of PO. For the PS rigid fraction, TCR products are environmentally more beneficial than the production of their virgin alternatives. However, TCR is still favourable over incineration.

Following the same reasons mentioned in section 3.1, TCR and MR show the highest benefit compared to incineration for the PS rigid fraction (4074 and 5590 kg CO₂ eq./ton PS rigid, respectively) and the lowest benefit for the MPO rigid fraction (1569 and 2427 kg CO₂ eq./ton MPO rigid, respectively) in terms of global warming impact. The high amount of missorted plastics in case of the MPO rigid stream results not only in lower savings in terms of global warming due to a lower amount of recycled products but also in a higher burden due to a higher amount of residues to be incinerated.

3.3 Sensitivity analysis for PS

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As explained in section **2.2.2**, due to the unrealistic impact results based on ecoinvent v3.6 data about virgin PS granulates for some impact categories (excl. global warming), the impact of virgin PS granulates was modelled by adding the average impact of polymerisation and granulation for PP, HDPE and LDPE to the impact of virgin styrene. Although the main findings presented in sections **3.1** and **3.2** remain valid, the adaptation results in a higher saving from the substituted virgin PS granulates for MR in terms of resource consumption and terrestrial acidification; therefore, MR gains a greater benefit compared to TCR and incineration. The benefit in terms of resource consumption increases 4.7 times and 1.3 times, respectively, but is lower (23 and 6%, respectively) in terms of global warming. More details can be found in the supplement material (section **C**).

3.4 Sensitivity analysis for the substitutability of virgin granulates by mechanical recyclates

Only the full substitutability (i.e. the 1:1 substitution ratio) of virgin granulates by mechanical recyclates

was investigated in sections 3.1 and 3.2; therefore, it presents the most favourable results for MR, which should be kept in mind when interpreting the results. However, technical quality degradation and a lower market uptake of mechanical recyclates may lead to a substitution ratio lower than 1:1 for MR (Ragaert et al., 2018; Vadenbo et al., 2017). Moreover, for specific applications such as food contact applications, there are currently no legislative approvals for the uptake of mechanically recycled content from the investigated waste fractions (De Tandt et al., 2021). Lazarevic et al. (2010) also indicated that the preference between MR and incineration becomes more uncertain as the substitution ratio at which the MR products substitute the virgin materials is reduced. For the ratio of 1:1 and between 1:1 and 1:0.5, MR was found to be favourable over incineration while it was harder to define a preference between MR and incineration when the ratio was less than or equal to 1:0.5. In our study, a sensitivity analysis of the substitution ratio for MR products was performed based on the identification of the "tipping point", i.e. the substitution ratio where MR obtains a net impact equal to TCR or incineration. In terms of resource consumption, MR obtains a worse net impact than TCR for substitution ratios for MR products less than 1:0.70, 1:0.95, 1:0.73, 1:0.68 for PP, PS, MPO rigids and PE films, respectively. Incineration is found to be environmentally beneficial compared to MR for substitution ratios for MR products lower than 1:0.42, 1:0.35, 1:0.37, 1:0.43 for PP, PS, MPO rigids and PE films, respectively. Regarding global warming impact, for the four plastic fractions, incineration remains the worst treatment option independently of the substitution ratio for MR products used. For substitution ratios for MR products higher than 1:0.21, 1:0.56, 1:0.23 and 1:0.21 in case of *PP*, *PS*, *MPO rigids* and *PE films*, respectively, MR maintains a better net impact than TCR. In terms of terrestrial acidification impact, TCR is the best treatment option independently of the substitution ratio for MR products used for the four plastic fractions. Except for *PS rigid*, this occurs only for substitution ratios for MR products lower than 1:0.67. MR obtains a worse net impact than incineration for substitution ratios for MR products less than 1:0.12, 1:0.05, 1:0.10 and 1:0.11 for *PP*, *PS*, *MPO rigids* and *PE films*, respectively. Identifying the "tipping point" as done here in this article is one step, but calculating the substitution ratio based on technical and/or market characteristics is another required step, which is challenging and needs further research. Based on a literature review by Rigamonti et al. (2020), they conclude that there is a lack of common procedure on how to calculate the substitution ratio. Moreover, the reported values for substitution ratios in literature are limited. For mechanically recycled plastics, Meys et al. (2020) reported that substitution ratios typically range from 0.7 for HDPE, LDPE and PP to 1 for PET. According to Rigamonti et al. (2020), the substitution ratio based on technical quality should be calculated in relation to a specific application and after identification of the substitutable (virgin) material. For mechanically recycled

plastics, only six values for application-specific substitution ratios were reported; they ranged from 0.69

for a recycled plastic mix (PET, PP, PVC and PS) substituting virgin PP in an injection moulding application

(Huysveld et al., 2019) until 9.23 for recycled PP substituting virgin PVC in an extrusion application

4 Discussion

(Rigamonti et al., 2020).

4.1 Closing the plastics loop by recycling

The results of this study show that, for four plastic fractions and three impact categories considered, the environmental savings (i.e. the negative part of the stacked bars) mainly come from the substitution of virgin materials by recycled products of TCR and MR. This benefit is higher in case of MR scenarios, which means that the production of virgin PP, PS and PE granulates causes considerably higher environmental burdens than the production of virgin styrene-related products, naphtha and slack wax. For example, this ratio is 1.4 in case of global warming impact of virgin PS production to virgin styrene production and 6.8 (as an average) in case of virgin PO production to virgin naphtha production. To better understand the reason behind this, impacts from the life cycle of plastics were studied in detail. As it can be seen from Figure 3, the life cycle of plastics starts with the crude oil extraction, followed by the oil refinery leading

to feedstock (e.g. naphtha) for steam cracking. For example, in case of PP, naphtha is used to produce propylene, from which PP granulates is produced through polymerisation and granulation. The PP enters a manufacturing process to be used in different potential product applications. At the end-of-life, it is collected, sorted and sent to incineration, MR or TCR, depending on the waste management system. Figure 3 shows that MR induces a shorter loop in the life cycle while plastics make a larger loop in case of TCR. Recycled styrene does not undergo the steam cracking process as in the case of recycled naphtha, resulting in a smaller loop.



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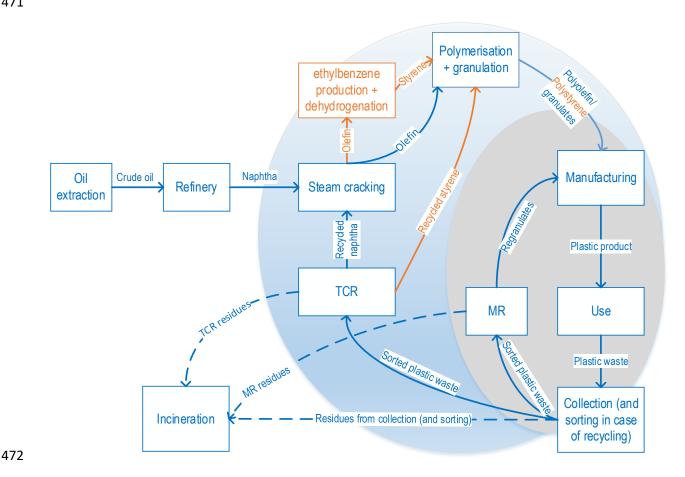


Figure 3. Visualization of linear and circular plastic waste management systems (Ecoinvent v3.6; PlasticsEurope for styrene production)

The environmental impacts of each life cycle stage of virgin plastic granulate production (i.e. PP, PS, LDPE and HDPE) were investigated using the ecoinvent datasets which were used in this study's LCA. It can be concluded that steam cracking, polymerisation and granulation are the main contributors to the environmental burdens in terms of global warming, as an example (Figure 4); MR avoids these two steps.

This explains why the environmental savings are higher in case of MR compared to TCR. Similarly, steam cracking is not necessary for recycled styrene; therefore, the savings of TCR are higher for the *PS rigid* fraction than for other fractions.

Incineration of plastics does not contribute to material circularity but causes release of high amounts of carbon to the atmosphere, while MR and TCR contribute to circularity of plastics in the society.



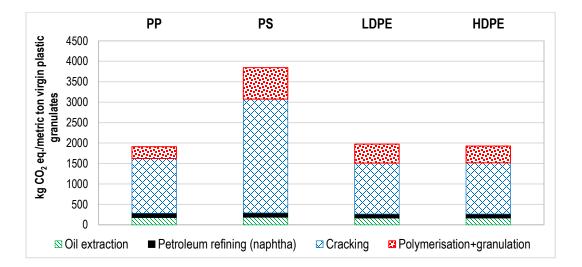


Figure 4. Relative contribution of different processes to the global warming impact of virgin plastic granulate production.

4.2 Comparison of our results with other case studies

In this study, a prospective LCA was performed based on the design of the processes in collaboration with academic and industrial experts. How the MR and TCR processes were designed and how the mass and energy balances were modelled were transparently reported in section 2.2.1 while not all LCA studies on the similar area provided such detailed information (Astrup et al., 2015). Detailed composition of each waste fraction and the foreground inventory data, which are important for the assessment but usually not provided (Antelava et al., 2019), are also mentioned in the supplementary material (section A). Additionally, this work, tackles some gaps of previous LCA studies on plastic waste management.

More specifically, the four individual plastic fractions after sorting (*PP, PS, MPO rigids* and *PE films*) were investigated while many LCA studies focused on mixed plastic waste instead (Cossu et al., 2017; Demetrious and Crossin, 2019; Gear et al., 2018; Hou et al., 2018; Khoo, 2019; Roy and Dutta, 2018). Consequently, the sorting of the collected mixed waste, which was usually omitted in other LCA studies

(Cossu et al., 2017; Meys et al., 2020), was included here and shows a contribution of 7-13% and 8-21% to the global warming burden of MR and TCR, respectively (section **3.2**). The sorting impact is even beneficial in terms of resource consumption (-0.9 GJ_{ex}/ton sorted waste).

Next to that, considering the difference between rigid and films (e.g. in the specific energy consumption (SEC), see section **2.2.1.1**) in modelling the MR processes adds value to this work. For the three impact categories analysed, the environmental performance of TCR and MR seems less dependent on whether the plastic fraction consists of rigids or films but especially dependent on the contamination degree of the household waste stream entering the recycling facility. The higher the contamination of the waste stream (e.g. 34.9% for *MPO rigid*), the worse the environmental performance for both recycling processes due to higher losses and thus a lower quantity of the final product. Resource consumption was quantified in addition to global warming and terrestrial acidification, the most commonly studied impact categories among the LCA studies in plastic waste management (Lazarevic et al., 2010).

Focusing on global warming, this work, on the one hand, shows the result in line with previous LCA studies reviewed in Lazarevic et al. (2010) and in Khoo (2019): both recycling options are favourable over incineration though MR is still more environmentally beneficial than TCR (i.e. pyrolysis). On the other hand, it highlights that this conclusion remains valid for the four individual polymer fractions, regardless of *rigid* (*PP, PS, MPO*) or *films* (*PE*) fractions, but under the full substitutability of virgin granulates by regranulates and flakes. Previous LCA studies showed no clear evidence that this result differed for individual polymer fractions (Lazarevic et al., 2010).

As presented in section **3.4**, the substitution ratio of MR products for virgin alternatives is an important factor in the comparison of the environmental performance of MR compared to TCR and incineration for the four household plastic fractions in Belgium. Regarding the *MPO rigid fraction*, recycled MPO flakes can be used for different applications (e.g. street benches, pallets, etc.); here we chose virgin HDPE granulates used for street bench production as the substituted alternative. Since a virgin street bench can be made of HDPE granulates or cast iron or tropical hardwood with a cast iron (Huysman et al., 2015), the choice in the applications of recycled materials and the virgin materials used for that application could influence the avoided burden of recycled products and thus the net impact of MR.

5 Conclusions

In this study, the environmental profile of MR and TCR of several household plastic waste fractions in Belgium was compared to incineration with energy recovery as a benchmark. In the transition phase to an enhanced household waste collection system, the newly sorted plastic waste fractions, i.e. *PP*, *PS*, *MPO rigids* and *PE films*, were studied as separate streams. For the modelling, detailed data were gathered from Belgian key actors and adapted for each waste fraction in collaboration with experts from both academia and industry during the period 2018-2020.

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The results showed that for all these fractions both MR and TCR perform better than incineration with energy recovery for the analysed environmental impacts (resource consumption, global warming, terrestrial acidification). MR is identified as an environmentally favourable option compared to TCR for the impact categories analysed when the products can substitute virgin materials in a 1:1 ratio. The major reason for the better results can be explained by the higher avoided burdens owing to a shorter loop with the production of MR regranulates or flakes, whereas TCR induces a larger loop with processes like cracking process and associated burdens. For example, global warming impacts of the production of 1 metric ton of virgin PS granulates are 1.4 times higher than for the production of 1 metric ton of virgin styrene, whereas the production of 1 metric ton of virgin PE and PP granulates shows 6.8 times higher (on average) impacts than the production of 1 metric ton of virgin naphtha. In other words, virgin plastic granulates have higher environmental impacts than chemical feedstocks per metric ton of material as the former are a few steps further in the production chain of plastics. However, in case of TCR, recycled styrene does not require the steam cracking process as in case of recycled naphtha, resulting in a smaller loop. On the other hand, in case of MR, further attention is to be paid to the substitution potential of regranulates and flakes as they may exhibit another level of technical quality and market uptake than the virgin materials. For example, mechanically recycled polymers, except PET, are currently not allowed as food contact materials (De Tandt et al., 2021); however, the EC is working on the rules to assure safe recycling of plastic materials other than PET into food contact materials (European Commission, 2020). The sensitivity analysis performed in this article showed the importance of a proper calculation of the substitution ratio depending on the envisaged application. Further research on the substitution ratio is recommended.

For both MR and TCR, electricity consumption is a major contributor to all impact categories analysed, whereas only for TCR, flue gas is an important cause for global warming and incineration of solid residue from the cracker is a cause for terrestrial acidification. Overall, incineration of residues from both recycling options contributes to global warming, while its impacts can be compensated through energy recovery. For all waste fractions studied, the highest benefits of both TCR and MR compared to the incineration benchmark are achieved for the *PS rigid* fraction and the lowest for the *MPO rigid* fraction. The former

can be explained by the avoidance of virgin production of (mainly) styrene in case of TCR, and of polystyrene in case of MR, both taking advantage of a relatively pure waste stream. The latter is explained by the high amount of residue sent to incineration as a result of relatively highly contaminated *MPO rigid* fraction.

Although the modelled electricity consumption of MR-related processes considered the physical difference between rigid and film fractions, the environmental performance of TCR and MR seems less dependent on whether the plastic fraction consists of rigids or films but especially dependent on the contamination degree of the household waste stream entering the recycling facility. The lower the contamination of the waste stream, the better the environmental performance for both recycling options.

Substantial levels of contamination are to a certain degree inherent to household plastic waste, where many polymer types are collected into a single commingled bag, including organic and paper contaminations (Ragaert et al., 2017). The purity of (sorted) household waste would benefit from simple systems that allow differentiation not only between the different polymer types, but also food-grade and non-food grade materials. Proposals for such systems have included the implementation of a harmonized labelling system or digital watermarks containing a variety of information, which have already been proposed by the EC (European Commission, 2020). Developing reverse logistics, extended producer responsibility and other innovative business models can also help to achieve plastic waste with less contamination (European Commission, 2018b). Extended pre-treatment can further reduce contamination in the waste before it goes to the recycling process. However, this is not preferred in all cases due to the extra (economic and environmental) cost it brings. In addition to the measures that can be taken during sorting and pre-treatment, reducing the complexity of the plastic packaging products at the design phase, which is known as Design for Recycling, can also contribute substantially to achieve higher separation and recycling efficiency (Roosen et al., 2020).

The EC stresses out the importance of the quality of separate collection and sorting for better plastic recycling performance (European Commission, 2020) and innovative solutions for chemical recycling (European Commission, 2018b) as the currently available waste management techniques (i.e. MR, incineration and landfilling) are not sufficient to deal with the increasing packaging waste problem (Vanapalli et al., 2021). In line with the EC's agenda, this study investigated the environmental profile of MR and TCR of four newly sorted plastic waste fractions that would otherwise go to incineration. This study also pointed out the important influence of contamination on the recycling efficiency. The results showed the potential of TCR for treating the household packaging waste in addition to existing MR,

especially when lower substitution ratios for MR products were considered in a sensitivity analysis. However, it should also be noted that when this study was performed TCR was less mature and still subject to upscaling and learning improvements.

Although this study was developed for a Belgian case, due to the similarities in the polymer types used in plastic packaging (Roosen et al., 2020), we think this study can put a light on other European and non-European countries' waste management policy. The separate kerbside collection bag for household plastic waste also exists in several other European countries such as the Netherlands, Germany and France. The sorting and mechanical recycling technologies used in Belgian installations are those that are representative for the recycling industry across OECD countries, such as cascades of near-infrared (NIR) and visual (VIS) spectrometry sensors, wind shifting, ballistic separators and extrusion (Ragaert et al., 2017). Finally, note that an analysis of the economic performance of this Belgian case study has also been investigated in other works (Larrain et al., 2020, under review).

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