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Expanding the collection portfolio of plastic packaging: Impact on quantity and quality of sorted plastic waste fractions

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ABSTRACT

Expanding the curbside collection system for household plastic packaging could help in accomplishing the ambitious recycling targets set by the European Union. In this study, the effects of such expansion in terms of quality and quantity of sorted plastic waste fractions were investigated. By applying a validated sorting model, the flows of packages in Belgium from their use phase until the output of the sorting plant and the quantity and composition of sorted waste fractions were simulated. These data were subsequently used to calculate existing performance indicators, such as grade and recovery, as well as new indicators, such as the Simpson's Diversity Index (SDI), which is a measure for the compositional diversity of sorted waste fractions. The highest SDI was calculated for the fraction named 'other films', namely 82%, showing that this fraction comprised the most diverse polymer mixture. Results indicated that certain sorted fractions achieve high product grades of approximately 80% for the PE films. Material Flow Analysis showed that by expanding the collection portfolio the collection rate of plastic packaging increased from 33.6 to 64.4 m%, of which 77.3% is simulated to be separated in the targeted waste fractions. However, this will be insufficient to meet the recycling target of 50% by 2025 as in total only 49.8% of plastic packaging is sorted correctly. Hence, additional improvements in both recycling technologies and packaging design are needed to further increase plastic recycling rates.

1. Introduction

Efficient and sustainable end-of-life management of municipal solid waste is an enormous global challenge. Especially for plastics, end-of-life treatment and circularity are gaining more and more interests (Bening et al., 2021). Collection and recycling systems for post-consumer plastic packaging waste are indeed still less developed compared to some other packaging types such as paper, glass, and metal (Brouwer et al., 2019a). Over the years, various measures have been taken to improve recycling rates of plastics and to accomplish a transition towards a more circular

economy for plastics (De Tandt et al., 2021). In this context, the European Union (EU) has been introducing more and more stringent legislation to improve plastic waste management and to encourage environmental friendly end-of-use options such as re-use and recycling (Hahladakis and Iacovidou, 2019). A major waste management policy instrument which supports the implementation of the European waste hierarchy is the extended producer responsibility (EPR) (Milios et al., 2018). Today, most of the EU Member States have indeed introduced EPR schemes for packaging waste collection and recovery, which were implemented through different types of instruments, such as regulatory

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take-back schemes, recycling requirements, and deposit systems (Leal Filho et al., 2019). Yet, optimizing EPR schemes remains to date a major discussion point within the field of waste management (Kranzinger et al., 2017).

In 2018, the EU has stipulated more ambitious recycling targets as part of their circularity action plan (European Comission, 2018). These targets include recycling rates for packaging materials; in the case of plastic packaging the target to be met by 2025 is 50% while the 2030 target is 55% (European Comission, 2015). In order to meet these targets, significant advances are needed in design, collection, sorting and recycling (Hahladakis and Iacovidou, 2019). Collecting more plastics is obviously a key step in increasing recycling rates. Hence, various countries have changed their curbside collection system in the past decades. For example, in the Netherlands originally a collection system for various plastic packages existed (i.e., polyethylene (PE) and polypropylene (PP) bottles and trays, and plastic films), together with a deposit-refund system for large poly (ethylene terephthalate) (PET) bottles for water and soda drinks (Brouwer et al., 2018). In July 2021, this deposit-refund system was extended with small PET bottles. In 2015, the collection portfolio of the separate collection system was expanded with beverage cartons and metal packaging (Brouwer et al., 2019a).

Such combined co-collection systems for packaging materials are common in Europe and many variations exist. In Germany, for instance, a mandatory deposit-refund system for single-use beverage packaging was introduced in 2003. Additionally, Germany also applies a collection system for mixed residual waste and a separate collection system through the so-called Dual System, where packaging and non-packaging consisting of plastic, paper, metals and composite material are disposed for commingled collection (Picuno et al., 2021). The latter stream is sorted out in twelve fractions, which each must meet certain purity levels that are stipulated by the so-called Deutsche Kunststoff Recycling (DKR) specifications (e.g., DKR 310 plastic films, DKR 329 PE, DKR 324 PP, DKR 328–1 PET etc.).

In Belgium, certain commingled plastics, metal packaging and beverage cartons have been collected via the so-called PMD (plastic bottles and flasks, metallic packaging and drinking cartons) bag. In the period 2018–2020, the Belgian producer responsibility organization Fost Plus extended the collection system for plastics via the so-called P+MD bag (Watkins et al., 2017). In the previous limited collection system three fractions were included, namely (P) plastic bottles and flasks, (M) metal packaging including food and cosmetics sprays, aluminum plates, dishes and trays, and metal lids and caps, and (D) drinking cartons. In the newly expanded collection system, all post-consumer plastic packaging (P+), such as yoghurt pots, meat trays, shrink wraps, and plastic bags, are allowed in addition to the metals and beverage cartons that were already allowed for disposal via the previous system.

The expansion of the collection system obviously has a positive impact on the collected amounts of post-consumer plastic waste, yet, it also increases the complexity compared to the previously limited curbside collection system. The expansion of the collection system implies that next to the already existing sorted waste fractions or so-called 'bales', which are PET bottles (in various colors), PE rigids, beverage cartons and metals, now also a PP rigid fraction (which was in fact sometimes already sorted out voluntarily by the sorters in the existing PMD system), a PET tray fraction, a polystyrene (PS) rigid fraction, a PE film fraction, and a so-called other film fraction are intended to be sorted out by material recovery facilities (MRFs) (Civancik-Uslu et al., 2021).

PET bottles are in general already relatively well recycled because they are less prone to contamination than other plastic packaging types (Eriksen et al., 2019). However, there is still debate on how to properly manage and recycle the mixed and/or more contaminated waste streams (Hahladakis et al., 2018a). Picuno et al. (2021) stated that most of the material losses within the packaging value chain occur at the sorting stage as result of the complex waste feedstock, where a combination of different materials is common practice. Next to various polymers, also the addition of a wide range of contaminants, such as paper, aluminum, pigments/inks, etc., complicates sorting and recycling processes (Roosen et al., 2020).

It is stated that the more heterogeneous the feedstock is, the larger the challenge for MRFs to achieve high purity levels of sorted fractions and recycled plastics (Brouwer et al., 2019a). Thus, increasing the collected amounts might negatively affect the quality of the sorted fractions, which makes these fractions more challenging towards high-end recycling routes. This is in literature referred to as 'the quantity-quality trade-off assumption' (Brouwer et al., 2019a). However, real quantitative data to support this assumption as well as broadly supported and scientific-based arguments are still relatively scarce (Huysman et al., 2017).

More detailed data in terms of quantity and quality of sorted waste fractions are needed to further develop and optimize recycling techniques tailored on the specific amount and composition of a certain waste fraction, and, eventually, to accomplish the recycling targets set by the EU by 2025 and 2030. This ideally starts by applying holistic assessment methods, such as MFA combined with a set of performance indicators, that are able of evaluating the quantity and quality of sorted waste fractions. Research to improve EPR schemes, sorting techniques, and recycling methods are vastly increasing (e.g., Andreasi Bassi et al. (2020); Meys et al., (2020); Walker et al., (2020); Korley et al., (2021), and Woidasky et al., (2020)), yet, limited data on flow behavior and quantity/quality of sorted waste streams are available. Therefore, in this study, we aim to get better insights in the flow of plastic packaging through the collection and sorting chain, as well as in the composition and purity of sorted waste fractions by comparing two different collection systems for post-consumer plastic packaging waste, using the following approach:

- Model and compare the composition and quantities of the plastic sorted fractions generated within the limited 'PMD' system and the expanded 'P+MD' system in Belgium via a material flow analysis (MFA) approach;
- (2) Define new 'performance indicators' as an addition to the existing indicators (as already used before by e.g., Brouwer et al., (2019) and Kleinhans et al., (2021)) to assess the quality of sorted plastic waste fractions;
- (3) Apply these performance indicators to evaluate the expansion of the collection portfolio in terms of expected quantity and quality of the sorted plastic waste fractions.

2. Materials and methods

2.1. Material flow analysis

As can be seen in Fig. 1, this study builds upon two previous studies (Kleinhans et al., 2021; Roosen et al., 2020) to perform an MFA of the plastic bales separated via both the limited PMD and the expanded P+MD collection systems in Belgium.

In a first study of Roosen et al., (2020), 102 types of packaging products, consisting of 24 PET bottles, 12 PET trays, 10 PE bottles, 3 PP bottles, 14 PP trays, 9 PS trays, 11 monolayer films, and 19 multilayer films, were sampled at the outlet of a Belgian MRF (Roosen et al., 2020). Each of these products were characterized up to the level of separable packaging items (e.g., a bottle was disassembled into the bottle itself, the cap, and the label) in terms of polymer and elemental composition via various chemical analysis, comprising inductively coupled plasma optical emission spectrometry (ICP-OES), attenuated total reflection - Fourier transform infrared spectroscopy (ATR-FTIR), combustion ion chromatography (CIC), elemental analyzer, differential scanning calorimetry (DSC), and polarized optical microscopy (POM). A more detailed description of the sampling and analysis procedure, as well as the detailed polymer and elemental composition of the included

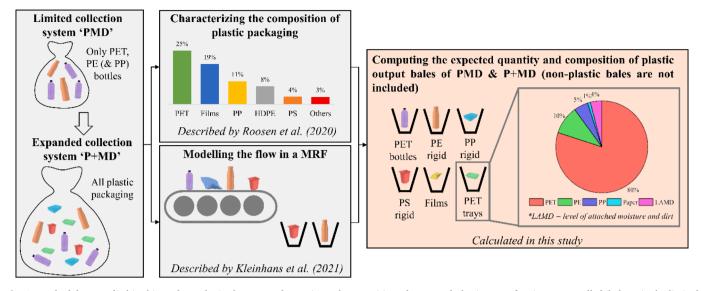


Fig. 1. Methodology applied in this study to obtain the expected quantity and composition of generated plastic waste fractions or so-called 'bales' via the limited PMD system and the expanded *P*+MD system.

packages (before sorting) can be found in Roosen et al., (2020).

As a next step, the behavior of these characterized packages through an MRF was modelled by applying the sorting model described by Kleinhans et al., (2021). The sorting model is based on existing sorting lines in which the transfer coefficients for every process (e.g., ballistic separation, near-infrared (NIR) separation, wind sifting, sieves & manual depollution) in the sorting stage were determined. The applied coefficients averaged per sorted waste fraction for PMD and P+MD facilities can be found in the study of Kleinhans et al., (2021) and are successfully validated via a case study of an operational post-consumer packaging waste sorting facility in Belgium. The waste flow was subdivided in material type (e.g., polymers, metals, carton) and packaging type (e.g., bottles, trays, and films) with its inherent product components (e.g., caps, labels, lids) in order to deliver a complete description of the composition of the sorted output fractions.

In order to perform the MFA, which is the new addition in this study, the separation efficiencies of the sorting model were combined with the detailed input compositions for both the PMD and the *P*+MD collection system. This study focuses on the sorted plastic fractions, being PET bottles (divided in different subcategories based on color as they are typically sorted in MRFs), PET trays, PE rigid, PP rigid, PS rigid, PE films, and other films, and thus excludes other fractions such as ferrous, nonferrous, and beverage cartons. The MFA follows the flow of the mentioned packages from collection to sorted fractions at an MRF in order to acquire data towards the potential of sorted fractions as input for recycling processes, including washing, float-sink, and effective processing. The data related to the plastic packaging that comes on the market are sourced from a report published by RDC Environment SA (2018). This report contains the total amount of plastic packaging that came on the Belgian market in 2015, together with an estimated capture rate which represents how much of the used plastic packaging is effectively captured by the waste collection system. The definition of capture rate used in this study is the ratio of the amount of packaging waste at the inlet of the sorting facility to the amount of packaging that is brought on the market (Tchobanoglous and Kreith, 2002). The capture rates determined by RDC Environment SA (2018) are based on manual sorting experiments performed by the Belgian EPR organization Fost Plus. In case of the PMD system, these manual sorting experiments are based on years of experience and data collection. In case of the P+MD system, data for determining the capture rates were acquired by a pilot project running in the period 2015-2016 in 6 Belgian municipalities. In this period, various sorting analyses were performed to investigate the influence of expanding the collection system in terms of capture rate. The applied capture rate values for the PMD and the P+MD system are included in Supplementary Material Table S1 and Table S2, respectively.

The amount of plastic packaging entering MRFs was calculated based on the indicated capture rates per packaging type. For instance, the report of RDC Environment SA (2018) indicated a capture rate of 81.3% for clear PET bottles via the *P*+MD system. Starting from the 41,983 ton clear PET bottles coming on the market, 34,132 ton (equaling to 81.3%) is assumed to be collected by the *P*+MD system. Based on the calculated collected amounts, the quantity of correctly sorted clear PET bottles was calculated. Kleinhans et al., (2021) reported a recovery rate of 87.0% for the clear PET bottles. Hence, it was simulated that 29,694 ton of the 34, 132 ton clear PET bottles (87.0%) end up in the clear PET bottle fraction.

2.2. Performance indicators

To evaluate the output and composition of sorted fractions, several indicators are typically considered. Following the suggestion of Testa (2015), *grade* and *recovery* were defined as the first two indicators to evaluate the performances of MRFs.

2.2.1. Sorting recovery and net recovery

The indicator *recovery* typically reflects the fraction of the targeted packages that are captured in the correct sorted fraction from the total amount of said packages entering an MRF. Hence, it can be considered as an indicator for the sorting efficiency of a certain packaging type.

By performing an MFA of packages from the moment they come on the market until they are sorted in bales, we can extend the recovery of the sorting process and define a new indicator which indicates how much of the plastic packages that come on the market also end up in the correct sorted fraction. Hence, this indicator comprises both the collection efficiency and the sorting efficiency. In this study, we thus apply two recovery-based indicators. The first indicator which only comprises the sorting efficiency will be referred to as **sorting recovery**, while the indicator comprising both the collection efficiency and the sorting efficiencies will be referred to as **net recovery**.

In terms of equations, the sorting recovery R_T of a target product T represents the mass of the target product T in the designated sorted fraction z divided by the mass of the target product T *that enters the facility*, as shown in Table 1. Likewise, the net recovery R_N of a target product T represents the mass of the target product T in the designated sorted fraction z divided by the mass of the target product T *that came on*

Table 1

Overview of the applied indicators and their corresponding definitions and equations, comprising sorting recovery R_T , net recovery R_N , product grade G_T , polymer grade G_P , SDI, total analyzed metal content C_M , total analyzed halogen content C_H, and C, H, N, O levels X_{CHNO}.

Performance indicator	Definition	Equation
Sorting recovery	Fraction of a desired product that is captured in the correct sorted fraction taken into account the sorting process	$R_T = \frac{f_z^T}{\mu^{TS}}$
Net recovery	Fraction of a desired product that is captured in the correct sorted fraction taken into account the collection and sorting process	$R_N = \frac{f_z^T}{\mu^{TO}}$
Product grade	Purity level of a desired product in a sorted fraction	$G_T = rac{f_z^T}{\sum_{m=1}^M f_z^m}$
Polymer grade	Purity level of a desired polymer in a sorted fraction	$G_P = \frac{f_z^p}{\sum_{m=1}^M f_\pi^m}$
SDI	Abundancy of different substances in a sorted fraction	$G_{T} = \frac{f_{z}^{T}}{\sum_{m=1}^{M} f_{z}^{m}}$ $G_{P} = \frac{f_{z}^{P}}{\sum_{m=1}^{M} f_{z}^{m}}$ $SCI = \left(1 - \frac{\sum_{i=1}^{S} (n_{i} - 1)n_{i}}{(N - 1)N}\right) \times 100\%$
Total analyzed metal content	Sum of the concentrations of Cd, Cu, Co, Zn, Fe, Mn, Pb, Li, Mg, Sr, Tl, Sb, Ti, Ca, Mo, V, As, Ni, Al, Be, Na, and Se present in a sorted fraction	$C_M = \frac{\sum_{i=1}^M f_z^i}{\sum_{m=1}^M f_z^m}$
Total analyzed halogen content	Sum of the concentrations of Cl, Br, and F present in a sorted fraction	$C_H = rac{\sum_{j=1}^M f_z^j}{\sum_{m=1}^M f_z^m}$
C, H, N, O levels	Mass fractions of C, H, N, S and O present in a sorted fraction	$X_{CHNO} = rac{f_z^{CHNO}}{\sum_{m=1}^M f_z^m} imes 100\%$

With R_T sorting recovery (%); R_N net recovery (%); G_T product grade (%); G_P polymer grade (%); f_z^T mass of target waste product T in bale z (kg); f_z^p mass of target polymer P in bale z (kg); f_z^m total mass of all waste products m in sorted fraction z (kg); μ^{TS} the input mass of the target product that enters the MRF (kg); μ^{TO} the mass of the targeted product that comes on the market (kg); S number of substances identified in a sorted fraction; ni proportional abundance of the nth substances (m%), and N total abundance of plastic entering the MRF (is always 100 m%), C_M total analyzed metal content (ppm); C_H total analyzed halogen content (ppm); X_{CHNSO} level of C, H, N, and O (m%); f_{π}^{i} mass of metal i in sorted fraction Z (mg); f_z^j mass of halogen j in sorted fraction Z (mg);); f_z^{CHNO} mass of C, H, N, and O in sorted fraction Z (kg).

the market in 2015.

2.2.2. Product grade and polymer grade

Grade is a measure of the purity of targeted packages in a certain sorted fraction and is relevant for the properties and applicability of the subsequently produced recycled plastics. Plastic sorted fractions, as defined by the Belgian EPR organizer Fost Plus, are named according to the targeted packaging types (e.g., PE rigids), which should then contain PE rigid packages (e.g., PE bottles and flakes). The original definition of grade relates to purity at such 'packaging type' level. This is also how different papers and legislations generally define the term grade.

However, plastic packages are composed of multiple components, materials and substances (e.g., a PET bottle regularly has a HDPE cap with closure ring, a PP label with prints, varnish and glue as well as contained product residues, attached dirt and moisture). By including compositional data in this study, we can go beyond product level and analyze the purity of a plastic sorted fraction in terms of polymer / material level. Hence, in this work a distinction is made between product grade and polymer grade. In the equations shown in Table 1, the product grade G_T of a sorted fraction z represents the mass of the target product T (without attached moisture and dirt) in sorted fraction z divided by the total mass of sorted fraction z. Likewise, similar to the

equation for product grade, the polymer grade G_P of a sorted fraction z represents the mass of the target polymerP (without attached moisture and dirt) in sorted fraction z divided by the total mass of sorted fraction z. The target polymer P in this definition corresponds to the target polymer contained in the target products. Hence, colored PET bottles ending up in the clear PET bottle fraction was considered cross-contamination and thus not categorized as target polymer for this sorted fraction. Likewise, clear PET bottles ending up in the colored PET bottle fraction were considered cross-contamination, and were consequently not categorized as target polymer. Fig. 2 shows a schematic flow diagram with indicated the symbols applied in the different equations in order to clarify the definitions of the different performance indicators.

2.2.3. Simpson's diversity index

We propose in this article a performance indicator called the Simpson's Diversity Index (SDI), previously unused in this field, as a measure for the diversity in terms of substances present in a sorted plastic waste fraction. The term substance used in this article comprises all polymers that are present, paper, and dirt and moisture. Additives, metals and halogens are, for instance, not included as substances in this indicator. The SDI is fundamentally different from the previously defined graderelated indicators, as it indicates how many different types of substances are present in a given sorted fraction, whereas product grade and polymer grade indicate the relative quantity of the targeted packages or polymers that are present in the corresponding sorted fraction. A higher diversity in polymer composition, i.e., a higher SDI, generally complicates the recyclability of plastics. Although certain sorted waste fractions can have the same purity grades, processing multiple polymers influences the morphology of the resulting blend (Ragaert et al., 2020; Vyncke et al., 2020) and it is stated that increasing the number of polymer components does indeed lead to complications towards mechanical recycling (Wang et al., 2011). For instance, a sorted fraction A, which would, for instance, consist of 90% PE, 5% PP and 5% PET, could be more challenging towards certain recycling options compared to a sorted fraction B, which would consist of 90% PE and 10% PP (albeit the polymer purities of both fractions are the same).

The SDI is already widely used to quantify the biodiversity of a habitat (Simpson, 1949), but will now for the first time be applied to indicate the diversity of sorted plastic waste fractions. To this purpose, the SDI is adapted to express the diversity of a sorted fraction based on the abundancy of different substances in that fraction (Gregorius and Gillet, 2008; Keylock, 2005). The SDI is calculated as the degree of concentration when the individual product components in the stream are classified into their respective categories, such as polymer types. The equation is shown in Table 1. In this equation, S stands for number of substances identified, ni for the proportional abundance of the nth substance, and N for the total abundance of plastic products entering an MRF (which is always 100%). For instance, when a sorted fraction would consist of 75 m% PET, 15 m% PE, 5 m% paper, and 5 m% dirt, the $\frac{(75-1)75+(15-1)15+(5-1)5+(5-1)5}{(100-1)100} \bigg)$ 1

SDI	would	be	calculated	as	(1 -	(75-1)75+(15-1)15-
						(100-1)

100%, which equals to an SDI of 41.4%. The value of the SDI always ranges between 0 and 100%; the closer the value is to 100%, the greater the polymer diversity in a given sorted fraction.

2.2.4. Performance indicators at elemental level

In order to further estimate the recyclability of outgoing flows, possible bottlenecks occur at a more elemental level. Plastic products can contain a variety of metal and halogen rich additives, such as pigments, stabilizers, plasticizers, processing aids, fillers, flame retardants etc. (Dimitrakakis et al., 2009). While during their use, these additives fulfill their function, the presence and degradation of these components, either during the course of their lives or while reprocessing, can be detrimental for the quality of the sorted waste fraction (Roosen et al., 2020). Halogens, such as chlorine, pose problems related to corrosion of equipment, among others issues. Metals can, for instance, interfere with

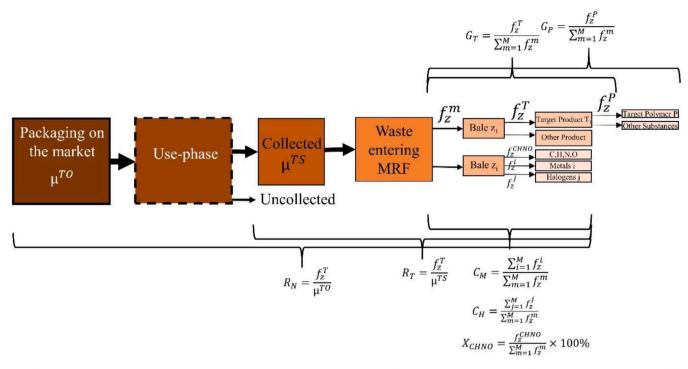


Fig. 2. Flow diagram of plastic packages through their life cycle with indicated the symbols and equations used to define the performance indicators applied in this study.

catalysts during pyrolysis. Also the C, H, N, and O content is key for thermochemical recycling, as this has an important effect on the yield of the process and the value of the obtained products (Toraman et al., 2014).

Therefore, the **total analyzed metal content, total analyzed halogen content**, and **carbon**, **hydrogen**, **nitrogen**, **and oxygen** (*C*, *H*, *N*, *O*) **levels** are identified as new performance indicators in this study. In the equations shown in Table 1, the total analyzed metal content of a sorted fraction z represents the sum of the mass of the analyzed <u>metals i</u> in sorted fraction z after washing divided by the total mass of stream z. Likewise, the total analyzed halogen content of a sorted fraction z represents the sum of the mass of the analyzed <u>halogens j</u> in sorted fraction z after washing divided by the total mass of sorted fraction z. The third indicator results in the C, H, N, and O fractions of a certain sorted fraction. These fractions are calculated by dividing the mass of the respective element (<u>i.e., C, H, N, or O)</u> in sorted fraction z by the total mass of sorted fraction z.

3. Results and discussion

3.1. Material flow analysis

A material flow analysis (MFA) was performed of plastic packaging collected via the previous limited collection system (i.e., the PMD system - 'bottles and bottle-like products'), which was the case until 2018–2020 in Belgium, and via the expanded collection system (i.e., the *P*+MD system – 'all post-consumer plastic packaging'), which is representative for the system in place today. The Sankey diagrams of both systems can be found in Fig. 3 and Fig. 4, respectively. Detailed data of the flows presented in the these figures can also be found in Supplementary Material Table S1 and Table S2. The MFAs start from the plastic packaging that is brought on the market in 2015, corresponding to 218.8 kton or 19.4 kg net. cap⁻¹ a⁻¹ (RDC environment SA, 2018). In 2015, 73.5 kton of the 218.8 kton plastic packaging was collected via the PMD collection system, which equals to an average capture rate of 33.6 m%. The collected plastics were typically sorted into four or five sorted plastic waste fractions (i.e., PET clear bottles, PET blue bottles, PET

green bottles, PE rigid, and optionally PP rigid). A sorted waste fraction typically consists of target and non-target packaging products. The latter are visualized by black lines in Fig. 3 and Fig. 4, respectively. Additionally, MRFs were also generating non-plastic sorted fractions such as ferrous and non-ferrous materials, beverage cartons, and a residue fraction, the calculation of which is beyond the scope of the current study. As can be seen in Fig. 3, 8.8 kton of the collected plastic packages will end up in the residue fraction and 0.4 kton in the non-plastic sorted fractions. The uncollected fraction, which equals to 145 kton or 66.4% in case of the PMD system, mainly goes to incineration via the mixed municipal solid waste stream in Belgium (Jacobsen et al., 2018).

3.2. Performance indicators

By expanding the collection portfolio, a large amount of materials is redirected from the mixed municipal solid waste stream to the separate collection system, as visualized in Fig. 4. Of the 218.8 kton that were put on the market in 2015, 140.8 kton is expected to be collected via the expanded packaging collection system, which corresponds to a capture rate of 64.3 m%. Next to the already existing sorted fractions in the previous collection system, the collected packaging waste is now also sorted in following additional sorted fractions: PET opaque, PET tray, PS rigid, PE film, other films, and mixed polyolefins (MPO).

3.2.1. Sorting recovery and net recovery

The calculated sorting recovery and net recovery of each sorted product has been determined and are given in Table 2. More detailed background data related to the calculations for both the PMD system and the P+MD system can be found in Supplementary Material Table S1 and Table S2, respectively. As the PMD system already incorporated sorted waste fractions such as PET clear bottles, PET blue bottles, PET green bottles, and PE rigid, these fractions are less affected in terms of recovery by the extension of the collection portfolio. Yet, it can be observed that the sorting recovery, and hence also the net recovery, is expected to decrease by around 5% for the PET bottle fractions. The reason for these lower recovery rates is twofold. Firstly, the P+MD waste that enters the sorting facility has a more complex composition as it consists of more

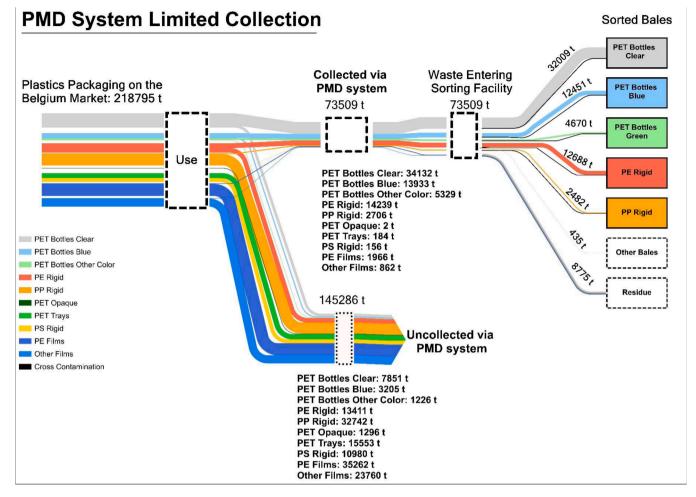


Fig. 3. MFA of plastic packaging as collected by the limited collection system (i.e., PMD system) from the moment that the packaging came on the market in 2015 until the end of the sorting process at MRFs, resulting in 5 sorted plastic waste fractions, 3 non-plastic waste fractions (i.e., ferro, non-ferro, and beverage cartons) which are here merged as 'other bales', and a residue stream. Cross-contamination by non-target packaging items in a certain sorted bale is visualized by black flow lines.

types of packages compared to PMD waste. Secondly, the output specifications for sorted P+MD fractions that were defined by the Belgian producer responsibility organization Fost Plus did not allow a significant quality reduction compared to the former sorted PMD fractions, leading to very stringent settings of the sorting equipment. For instance, the maximum allowed amount of impurities is 3% in the PET bottle clear fraction and 4% in both the PE rigid and the PP rigid fractions according to the tender specifications of Fost Plus (FostPlus, 2019).

The introduction of new categories such as PET trays, PP rigid, PS rigid and flexible packaging (separated in PE films and other films) has the biggest influence on the recovery indicators. Table 2 indicates that the expansion of the collection system will result in an estimated annual increase of the net recovery rate of 20.5%, which corresponds to 44.9 kton of plastic packaging in Belgium. With this additional recovered plastic material, the total net recovery at the end of an MRF is estimated to be around 49.7%. However, real recycling rates (i.e., amount of plastic that is actually reprocessed into products) will actually be lower as certain sorted fractions such as PET trays and other films are still very challenging to be effectively recycled (Ügdüler et al., 2020a, 2021) and since losses in the mechanical recycling process occur (Faraca and Astrup, 2019). This is an important result of this study, as it highlights the extra effort that is still necessary to be able to meet the European recycling targets of 50% by 2025 and 55% by 2030.

3.2.2. Product grade and polymer grade

A larger diversity in packaging products will be collected by expanding the collection portfolio, which will end up in more sorted fractions. Although the expansion of the collection system is needed in respect of the more ambitious European recycling targets, this expansion will come at a certain cost in terms of quality of the sorted waste fractions because of increased complexity of the feedstock that enters an MRF.

By applying the sorting model of Kleinhans et al., (2021), product grades of the sorted plastic fractions were calculated. Fig. 5a shows that the simulated product grades vary between 63.7% for the MPO fraction and more than 99% for the PS fraction. It can be observed that differences in product grades between sorted fractions generated via the PMD system and sorted fractions generated via the P+MD system are in most cases relatively limited. For instance, clear PET bottles have an estimated product grade of 99.0% when collected via the PMD system and an estimated product grade of 99.4% when collected via the P+MDsystem. For the PE rigid fraction, a more significant change can be observed with a decrease in product grade from 99.0% when collected via the PMD system to 96.3% when collected via the P+MD system. The limited influence of the expansion of the collection portfolio on the product grades can be explained by the fact that the Belgian producer responsibility organization Fost Plus did not allow a reduction in terms of product grades of the sorted *P*+MD fractions compared to the product grades of the former PMD fractions. In order to meet the quality

Sorted Bales

P+MD System Expanded Collection

PET Bottles Clear 298701 PET Bottles 11012 PET Bottles Other Colo 44431 Collected via Waste Entering P+MD system Sorting Facility 146151 Plastics Packaging on the PE Rigid Belgium Market: 218795 t 140811 t 140811 t 14829 t PP Rigid Use 871 t PET Opaque \$964 PET Bottles Clear: 34132 t PET Bottles Blue: 13933 t PET Trays PET Bottles Other Color: 5329 t 37551 PE Rigid: 15657 1 PP Rigid: 20342 t PET Bottles Clear PET Opaque: 934 t PS Rigid PET Trays: 8954 t PS Rigid: 6337 t PET Bottles Blue PET Bottles Other Color PE Films: 21182 t PE Rigid Other Films: 14011 t PE Films PP Rigid PET Opaque 77984 t PET Trays Other Film PS Rigid 1853 Uncollected via PE Films P+MD system Other Films MPO Ba Cross Contamination PET Bottles Clear: 7851 t PET Bottles Blue: 3205 t PET Bottles Other Color: 1226 t Other Bale PE Rigid: 11993 t PP Rigid: 15106 t PET Opaque: 364 t PET Trays: 6783 t PS Rigid: 4799 t Residue PE Films: 16046 t Other Films: 10611 t

Fig. 4. MFA of plastic packaging as collected by the expanded collection system (i.e., *P*+MD system) from the moment that the packaging came on the market in 2015 until the end of the sorting process at MRFs, resulting in 11 sorted plastic waste fractions, 3 non-plastic waste fractions (i.e., ferro, non-ferro, and beverage cartons) which are here merged as 'other bales', and a residue stream. Cross-contamination by non-target packaging items in a certain sorted bale is visualized by black flow lines.

Table 2

Overview of the simulated sorting recovery and net recovery of the sorted plastic waste fractions via both the limited (PMD) and the expanded (P+MD) collection system. The newly sorted fractions via the P+MD system are displayed in italic font. The total sorting and net recovery are calculated by dividing the total mass of packaging that is correctly sorted by the total amount of packaging entering MRFs and by the total amount of packaging that is brought on the market, respectively.

	Sorting 1	ecovery (%)	Net recovery (%)	
	PMD	P+MD	PMD	P+MD
PET bottles clear	92.9	87.0	75.5	70.7
PET bottles blue	88.5	83.3	72.0	67.7
PET bottles other color	87.4	83.3	71.1	67.7
PE rigid	89.0	91.0	45.8	51.5
PP rigid	90.0	72.8	6.9	41.8
PET opaque	_	84.3	-	60.6
PET trays	_	53.0	_	30.2
PS rigid	_	49.8	_	28.3
PE films	_	79.0	_	45.0
Other films	_	52.2	_	29.7
Total	86.8	77.3	29.2	49.7

requirements, MRFs have installed their sorting equipment in such a way that a sufficiently high purity level can be acquired. Yet, due to the increased complexity of the input stream, a higher reject rate of the sorting devices occur and, consequently, more plastic packages end up in the residue stream. It can indeed be seen in Fig. 3 and Fig. 4 that the share of plastic packages ending up in the residue stream increased from 11.9 to 18.4%.

The additionally generated sorted fractions from the P+MD system are more varied in terms of product grade. Certain sorted fractions have a high level of purity with product grades up to 99.9% in case of the PS trays, whereas others, such as the PE film and other film fractions, have significantly lower product grades of 82.9% and 85.7%, respectively. Again, this depends on the specific configuration of MRFs, which are designed to meet the imposed product grade levels.

As legislations often imply product grade levels, grades on polymer level are not yet well established in the recycling industry. In this study, we estimate the so-called polymer grade levels by first calculating the polymer composition and the level of attached moisture and dirt (LAMD) of the different bales after sorting, according to the definition of Brouwer et al., (2019a). The results can be seen in Fig. 6, showing the simulated polymer composition of the PET bottle clear, PE rigid, PP rigid, PET trays, PS rigid, MPO, PE film, and the other films sorted fractions. As the first three sorted fractions are separated via both the PMD system and P+MD system, their compositions are visualized in Fig. 6a. Fig. 6b shows the composition of the new sorted fractions from the P+MD system.

As can be seen in Fig. 6a, the simulated polymer compositions of

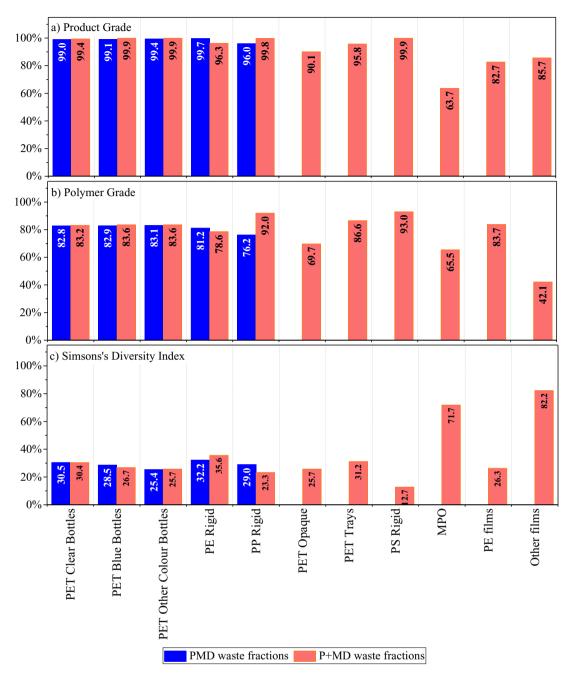


Fig. 5. (a) Simulated product grade and (b) polymer grade levels applied on sorted plastic waste fractions generated via the limited PMD system (indicated in blue) and the expanded *P*+MD system (indicated in red), as well as c) the Simpson's Diversity Index applied on the different sorted plastic waste fractions.

sorted plastic fractions that are included in both the PMD and the P+MD collection systems are in general only slightly influenced by the expansion of the collection portfolio. For the PET bottle clear fraction, no clear difference in composition between both collection systems can be observed. For the PE rigid sorted fraction, the simulated PE content decreases slightly from 81 to 79 m%, whereas the PET content in this sorted fraction increases by 2 m% as result of potential missorting of (multilayer) PET trays. The PP rigid sorted fraction is more subjected to change as the PP content substantially increased from 76 to 92 m%. This can be explained by the fact that PP was not officially part of the former PMD system. Moreover, by the expansion of the collection portfolio, the share of PP trays significantly increased within this stream and sorting lines became more tailored to separate out PP packages in the correct fraction.

PET trays often consist of a combination of PET and PE, together accounting on average for 92 m% of the PET tray fraction. The PS fraction is a relatively pure stream, consisting of 93 m% of PS. The MPO product is a combination of polyolefin-based (PP and PE) packages, which are not captured in their respective waste fractions, together accounting for 66 m%. Additionally, the MPO fraction also contains a relatively high amount of PET-based items of averagely 23.6 m%. This high level of PET contamination can be explained by the high level specification for the other plastic fractions and the applied sorting configuration as described by Kleinhans et al., (2021).

The PE film fraction consists on average of 84 m% of PE and contains 8 m% LAMD. The other film fraction is a much more heterogeneous mixture of polymers, with an average simulated PE content of 42 m%, PET content of 17 m%, and PP content of 15 m%.

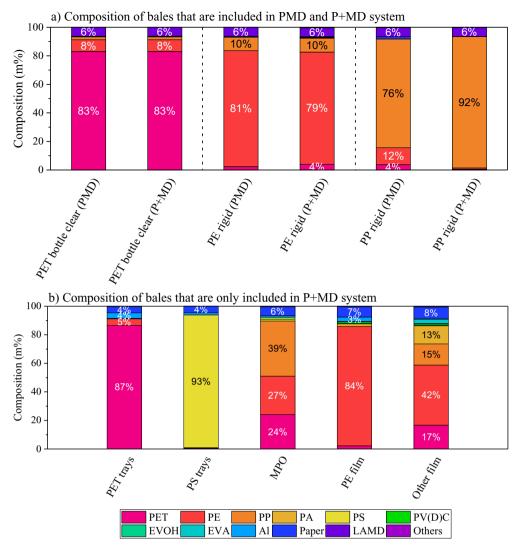


Fig. 6. Simulated polymer composition and paper and LAMD levels of different sorted plastic waste fractions, as occurring within both the limited (PMD) and the expanded (P+MD) collection system (a) for sorted fractions that are generated by both the PMD and P+MD system (i.e., PET bottles, PE rigid, and PP rigid), and (b) for sorted fractions that are only generated by the P+MD system (i.e., PET trays, PS trays, MPO, PE films, and other films).

The data presented in Fig. 6 place the composition of the sorted fractions and the reported recycling rates in perspective. In an amendment to the Packaging and Packaging Waste Directive (PPWD), the measuring point to calculate packaging recycling rates shifted to a later stage of the recycling process (Brouwer et al., 2019b). The new calculation method for recycling rates started to be applicable for 2020 data. Recycling rates are from that moment on calculated as the ratio between the outputs of recycling plants and the total generated amount of plastic packaging waste, whereas in the past they were calculated as ratio between the output of sorting plants and the total plastic packaging waste (Lombardi et al., 2021). The new approach is more correct as material losses occuring before the waste enters the recycling operation and during the recycing operation itself will not be included anymore in the waste amounts reported as recycled. As can be seen in Fig. 6, most sorted fractions comprise a significant amount of substances that are different from the main polymer of the packaging, varying between 7 m% for the PS rigids and 20 m% for the PE rigids. Separating such substances from the targeted polymer has an influence on the calculated recycling rates, especially if taken into account that separation processes are not 100% efficient and, thus, that besides the non-targeted substances, also a part of the target polymer might end up in the non-target fractions. It was indicated that the change of calulation methodology might drop recycling rates by 20% (Lombardi et al., 2021), which is in the same order of magnitude as the data reported in Fig. 6.

Similarly to the product grades, the differences between the simulated polymer grades of sorted fractions generated via both the PMD and P+MD systems are in general rather limited, typically between 1.0 and 3.0%, with exception of the PP rigids for which the polymer grade is simulated to increase from 76.2% to 92.0%. The limited influence of the expansion of the collection portfolio on the polymer grade is in agreement with the data presented by Brouwer et al., (2019a), that indicated that the average polymer grade after washing and sink/float separation was only slightly reduced from 91 \pm 6% to 90 \pm 7% by expanding the Dutch collection portfolio.

It can also be observed from Fig. 5 that a high product grade does not necessarily implicate a high level of polymer grade. For instance, PET trays have a product grade of 90.1%, yet due to the presence of inseparable multi-material trays and lids, the polymer grade is only 69.7%. This shows the relevance of the polymer grade as indicator to evaluate the polymer quality of sorted plastic waste fractions. In this study, the polymer grade is simulated at the end of the sorting process as this is an important feature for potential revenues from selling the sorted waste fractions to recyclers (Faraca et al., 2019). However, it should be noted that various steps that are applied after the sorting stage such as, among washing and separating processes other things. such as sink/float-separation, will still be able to improve the purity of waste fractions (Hahladakis and Iacovidou, 2019).

3.2.3. Simpson's diversity index

A new performance indicator was adopted from the domain of ecology, namely the SDI. Where the SDI is typically used to indicate the biodiversity of a certain habitat in ecology, this index is in this article applied to indicate the diversity of a certain sorted fraction in terms of comprising substances. The values of the SDI per sorted fraction can be found in Fig. 5c. For the PET bottle fractions and the PP rigid fraction, the estimated SDIs vary between 23.3 and 30.5%. The PE rigid fraction has a slightly higher SDI, namely 32.2% when generated via the PMD

system and 35.6% when generated via the P+MD system. When looking to the sorted fractions that are only generated within the P+MD system, varying results can be observed. For PET opaque, PET trays and PE films, the simulated SDIs are in the same order of magnitude, namely between 25 and 32%. The PS rigids have a significant lower SDI of 13.0%. This indicates that this sorted fraction is relatively pure and does not contain many different types of substances. This is confirmed by Fig 5a,b, showing that the sorting and polymer grades for PS rigid amount to 99.9 and 93.0%, respectively. As grade and recovery behave as communicating vessels, this high purity level also has an influence on the recovery of PS packages. Compared to the other sorted fractions, the PS

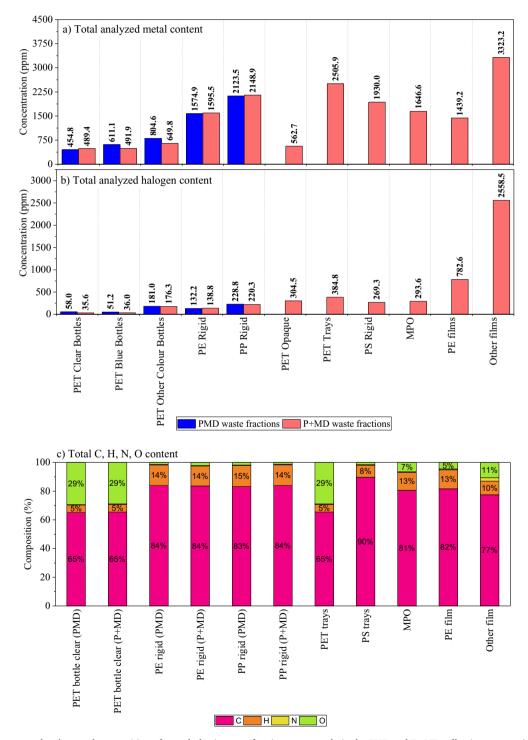


Fig. 7. Indicators to assess the elemental composition of sorted plastic waste fractions generated via the PMD and *P*+MD collection system: (a) the total analyzed metal content, (b) the total analyzed halogen content, and (c) total C, H, N, and O content.

fraction indeed has a significant lower sorting recovery (49.8%) and net recovery (28.3%), as can be seen in Table 2.

The MPO fraction, including PE and PP rigids, has a relative high simulated SDI of 71.7%. This can be explained by Fig. 6 which indicates that this product comprises various polymer types. Also the other film fraction has a high simulated SDI of 82.2% due to the fact that multilayer films typically have a complex polymer composition and can consist of various layers which are composed of several types of polymers (Kaiser et al., 2017; Walker et al., 2020).

The correlation between the variety in terms of polymer composition of a sorted fraction and the SDI, makes this performance indicator appropriate to signify the polymer diversity and thus, in most cases, the challenge of a certain waste fractions towards high-end purification and recycling methods.

3.2.4. Performance indicators at elemental level

Mechanical recycling remains to date the most industrially applied recycling technique for plastics (Larrain et al., 2021). The quality of mechanically recycled plastics strongly depends on the polymer composition of the input. Chemical recycling, on the other hand, is in a less mature stage and is affected by impurities that are situated at an elemental level, such as oxygen, chlorine, and metals. A high Cl content, for instance, can yield hydrochloric acid (HCl) which can be corrosive towards reactors and equipment (Al-Salem et al., 2017). Alkali and alkaline earth metals, such as Ca, K, Mg, and Na, are easily deposited on a catalyst's acid site, causing rapid deactivation of the acid catalyst (Ryu et al., 2020). The elemental composition of the feedstock also determines the yields of pyrolysis oil, char and gasses (Nanda and Berruti, 2020). As the performance indicators that were discussed in the previous sections do not take into account the elemental composition of sorted fractions, some additional indicators are defined to assess the suitability of these fractions as feedstock for (thermo)chemical recycling. In this study, we evaluate the elemental composition in terms of C, H, N, and O content, metal concentration, and halogen concentration. The results are visualized in Fig. 7.

Fig. 7a shows the total sum of the concentration of all the metals that are included in the performed analyses by Roosen et al., (2020) (i.e., Cd, Cu, Co, Zn, Fe, Mn, Pb, Li, Mg, Sr, Tl, Sb, Ti, Ca, Mo, V, As, Ni, Al, Be, Na, and Se). Clear differences can be observed within different sorted fractions, however, corresponding bales of the PMD collection system and the P+MD collection system have in general similar levels of total analyzed metal content.

Sorted fractions that are mainly composed of PET (i.e., PET clear bottles, PET blue bottles, PET other color bottles, and PET opaque) have in general a relatively low total analyzed metal content compared to the other generated bales. This is attributed to the fact that PET bottles have a relatively low total metal concentration of around 538 ppm before sorting (Roosen et al., 2020). As can be seen in Fig. 7a, the PET other color bottle fraction has a higher total simulated metal content (i.e., 649.8 ppm if generated via the P+MD system) compared to metal content of the PET clear bottles (i.e., 489.4 ppm if generated via the P+MD system). Another observation is that polyolefin-based fractions have a relatively high total analyzed metal content. Especially the PP rigid fraction shows high estimated metal concentrations of around 2100 ppm. Such differences across the metal content of different sorted fractions can be explained by considering the origin of metals in plastics. Metals can indeed originate from various sources and can be intentionally or unintentionally added to the packaging material. There are several reasons to add certain metals or metal-containing additives during the production of the packaging (Eriksen et al., 2018), e.g., as colorants, plasticizers, lubricants, and antioxidants (Hahladakis et al., 2018b). The influence of inks and printing on the metal content is noticeable in the plastic film products, which are typically heavily printed (van Putten, 2011). Another main source of metals in plastics are fillers such as CaCO₃ and Mg₃Si₄(OH)₂ which are used in concentrations up to 50 m% in order to increase the bulk of the plastics at a low cost

(Ügdüler et al., 2020b).

Besides certain metals, plastics can also contain certain halogens. The main source of halogens in packaging is chlorine-containing polymers such as PVC and PVDC (poly(vinylidene chloride)). This explains the substantial higher simulated chlorine content of the other film fraction (2559 ppm) compared to the PE film fraction (783 ppm), as shown in Fig 6b. The other sorted fractions have a lower halogen content, varying between 30 and 300 ppm. PET clear bottles have in general the lowest simulated halogen content, e.g., 35.6 ppm when collected via the PMD system and 58.0 ppm when collected via the P+MD system, which is in the same order of magnitude as the data reported by (Brouwer et al., 2020). PET other color bottles have averagely a higher Cl content compared to clear PET bottles, namely between 176 and 181 ppm. However, especially the PET opaque fraction has a significant higher estimated Cl content of 560 ppm. PET opaque bottles are often used as shampoo or detergent bottles, which might be one of the reasons why the halogen content is higher than for clear PET bottles which are mostly used for packaging of water, sodas or juices. As it is stated that the presence of halogens, such as chlorine, in a concentration of more than 10 ppm in pyrolysis oil limits its use as a fuel or chemical precursor, the halogen content of each sorted fraction might need to be monitored and certain pre-treatment steps are required prior to pyrolysis (Nanda and Berruti, 2020).

Fig. 7c visualizes the modelled C, H, N, O content of the different plastic waste fractions. As expected, a clear difference between PETbased products and polyolefin or PS-based products can be observed. For instance, the PET bottle fraction has an average simulated C content of 65 m% and an average O content of 29 m%, whereas the PE rigid fraction has a C content of 84 m% and O content of 2 m% and the PS rigid fraction a C content of almost 90 m% and O content of 2 m%. The hydrocarbon content plays an essential role in determining the calorific value of liquid oil (Wan Mahari et al., 2018). A higher carbon content will generally result in higher carbon efficiencies and, hence, higher gross margins (Ragaert et al., 2017). Also thermochemical recycling of a carbon-rich feedstock prefers a low oxygen content, since this might result in the production of undesired acids (e.g., benzoic acid and terephthalic acid) (Toraman et al., 2014). From this perspective, the PS, PE, and PP rigid bales and the MPO and PE films fractions are potentially interesting feedstock for thermochemical recycling.

4. Conclusions and perspectives

The extension of the collection system for plastic packaging resulted in larger amounts of collected plastic wastes. Also the simulated net recovery rates, comprising both the collection efficiency and the sorting efficiency, are significantly influenced for some sorted fractions. For instance, the net recovery rate of the PP rigids, which was in some cases already sorted in the former PMD system although not being officially part of it, is expected to increase from averagely 6.9% to 41.8%. For the PE rigid and the PET bottles, the influence on the recovery rate is less significant. The simulated net recovery rate for PET bottles decreased with around 5%. This is of course still a relevant fraction and opens again the discussion between, on one hand, a combination of a depositrefund system for PET bottles and a curbside collection system for other plastic packages as applied in, for instance, the Netherlands and, on the other hand, a curbside collection system comprising all plastic packaging as currently applied in Belgium. In addition, also the average net recovery rate of all plastic packaging significantly increased from 29.2% to 49.7%, as can be seen in Fig. 8.

Fig. 8 also shows that for the PET bottle fraction and the PE rigids the product grades are in the same order of magnitude when separated via the limited PMD system and via the expanded P+MD system. For instance, the product grade of the PET bottle clear fraction increased from 99.0% to 99.4%. For the PE rigids, a small decrease of the product grade can be observed, namely from 99.0% to 96.3%. The average product grades of the PET bottle fractions and the PE rigids, calculated

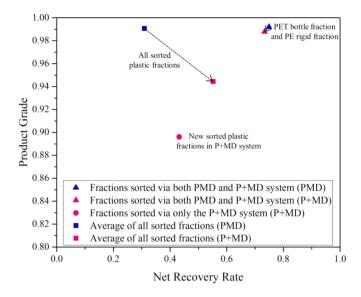


Fig. 8. Evolution of the sorted fractions separated via both the limited PMD and the expanded P+MD system (i.e., the PET bottle and PE rigid fractions), the fractions only separated via the P+MD system (i.e., PE rigid, PP rigid, PET opaque, PS rigid, MPO, PE films, and other films), and the average of all sorted fractions in terms of their product grade and net recovery rate.

by multiplying the product grade of each of these sorted fractions with the annual generated mass of the corresponding sorted fraction divided by the sum of the total amount of the plastic packages targeted for the respective sorted fractions, correspond to 99.2% in case of the limited collection system and 98.8% in case of the expanded collection system. However, the average product grade of the newly sorted fractions (i.e., PP rigid, PET opaque, PET trays, PS rigid, MPO, PE films, and other films) is only 89.6%. This indicates that these sorted fractions have a lower product grade compared to the sorted fractions that were already included in the previous PMD system.

Next to the more traditional performance indicators to evaluate the sorting process in terms of product purity, we have also defined new indicators such as the polymer grade and the Simpson's Diversity Index (SDI). The polymer grade of a sorted fraction is generally lower compared to its product grade due to the presence of other non-targeted substances such as other polymers, paper, dirt and moisture that are present. For PET bottles, for instance, these substances account for around 16 m% of the sorted fractions. Whereas the average product grade decreased with 4%, the average polymer grade was for the fractions generated via the PMD and the fraction generated via the P+MD system equal around 83%. Thus, in terms of polymer purity no significant decrease is expected by the expansion of the collection portfolio.

The SDI is an indicator to estimate the diversity of a sorted fraction. Whereas product grade and polymer grade give information on the share of the amount of target products or polymers in a certain sorted fraction, respectively, the SDI indicates how many different types of substances, such as polymers, paper, and dirt, are present in a sorted fraction. Two outliers can be observed, namely the MPO fraction with a simulated SDI of 71.7% and the other film fraction with a simulated SDI of 82.2%. The reasons behind these high values are different. The MPO fraction contains many different types of substances due to errors during the sorting process, whereas the other film fraction contains a broad range of films, including multilayer films with a diverse composition in terms of polymers.

Increasing collection rates for plastic packaging is a key step towards a more circular economy. Yet, collecting more plastic packaging comes at a certain cost in terms of complexity of the feedstock for the sorting process and induces cross-contamination within sorted fractions. Hence, a balance needs to be found between packaging design, collection rates, recovery rates, and the quality of the sorted fractions.

Even with an extensive expansion of the collection portfolio in Belgium, meeting the EU recycling targets will be challenging. Hence, further investments to improve pre-treatment, sorting and recycling technologies and easier recyclable packaging designs will be key to further increase plastic recycling rates.

CRediT authorship contribution statement

Martijn Roosen: Methodology, Investigation, Writing – original draft. Nicolas Mys: Methodology, Investigation, Writing – original draft. Kerstin Kleinhans: Software, Data curation, Validation. Irdanto Saputra Lase: Software, Data curation, Visualization. Sophie Huysveld: Validation, Writing – review & editing. Marieke Brouwer: Writing – review & editing. Eggo U. Thoden van Velzen: Writing – review & editing. Kevin M. Van Geem: Writing – review & editing, Funding acquisition. Jo Dewulf: Writing – review & editing, Funding acquisition. Kim Ragaert: Writing – review & editing, Funding acquisition. Ann Dumoulin: Writing – review & editing, Supervision. Steven de Meester: Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

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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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.106025.

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