Technical and market substitutability of recycled materials

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Technical and market substitutability of recycled materials: Calculating the environmental benefits of mechanical and chemical recycling of plastic packaging waste

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ABSTRACT

Most plastics are today mechanically recycled (MR), whereas chemical recycling (CR) is an emerging technology. Substitutability of virgin material is vital for their environmental performance assessed through life cycle assessment (LCA). MR faces the reduction in the material's technical quality but also the potential market because legal safety requirements currently eliminate applications such as food packaging. This study presents a data-driven method for quantifying the overall substitutability (OS), composed of technical (TS) and market substitutability (MS). First, this is illustrated for six non-food contact material (non-FCM) applications and three hypothetical future FCM applications from mechanical recyclates, using mechanical property and market data. Then, OS results are used in a comparative LCA of MR and thermochemical recycling (TCR) of several plastic waste fractions in Belgium. For mechanical recyclates, TS results for the studied non-FCM and FCM applications were comparable, but OS results varied between 0.35 and 0.79 for non-FCM applications and between 0.78 and 1 for FCM applications, reflecting the lower MS results for the current situation. Out of nine application scenarios, MR obtained a worse resource consumption and terrestrial acidification impact than CR in six scenarios. MR maintained the lowest global warming impact for all scenarios. This study contributes to an improved understanding of the environmental benefits of MR and TCR. Inclusion of other criteria (e.g. processability, colour, odour) in the quantification of the overall substitutability for MR products should be further investigated, as well as the environmental performance of TCR at industrial scale.

1. Introduction

1.1. *High-quality recycling of plastic packaging waste*

Proper management of plastic waste is a key priority to avoid plastic pollution of the environment (Kurniawan et al., 2021) and for the transition to a circular economy (Blomsma and Brennan, 2017). The packaging sector was responsible for 61 % of the collected post-consumer plastic waste in the European Union, Switzerland and Norway in 2018 (PlasticsEurope, 2019). For instance in the European Union, plastic packaging recycling targets were set at 50 % by 2025 and 55 % by 2030 (European Parliament and Council, 2018).

Recycling of plastics can be done through mechanical and chemical pathways. Mechanical recycling (MR) typically consists of sorting, grinding, washing, and usually compounding of the material, leading finally to flakes (without compounding) or regranulates (after compounding) (Ragaert et al., 2017). Chemical recycling (CR) is a process where the chemical structure of the polymer is changed, and is defined

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by the European Chemical Industry Council as”the conversion into chemical building blocks including monomers that can be used again as a raw material in chemical processes” (Cefic, 2022), such as the production of new polymers. Thermochemical (e.g. conventional pyrolysis, gasification) and catalytic conversion processes (e.g. catalytic pyrolysis, hydrocracking) can be distinguished (Ragaert et al., 2017). For post-consumer plastic packaging, being a mixture of food and non-food packaging, MR is the main recycling method today (PlasticsEurope, 2019). However, its heterogeneous composition including inorganic and organic contaminants, as well as polymer degradation during its lifetime and during reprocessing, pose large challenges to MR. Mechanical post-consumer recyclates may therefore not meet the technical quality of virgin alternatives leading to only a partial substitution of the virgin or primary material instead of a full substitution depending on the application (Golkaram et al., 2022; Ragaert et al., 2017; Roosen et al., 2020).

In addition to varying technical material quality requirements, there are also differences in legal material requirements amongst the different plastic applications. Whereas food packaging has the strictest legal material requirements, other applications such as building and construction have minimal legal requirements (Eriksen et al., 2019). Barring some very specific small-scale exceptions, closed-loop MR into food contact materials (FCM) is currently not possible for plastics other than PET due to food safety regulations and the challenge of separating food from non-food packaging (De Tandt et al., 2021). MR of food packaging waste, therefore, leads to open-loop recycling into different fields of application than the original one. If these applications demand less material quality, the label ‘downcycling’ or cascading into lower-valued applications may be used (Ragaert et al., 2017). In contrast, CR is an emerging technology suitable for closed-loop recycling of food packaging (Ragaert et al., 2017), as it is not subject to European Food Safety Authority (EFSA) approvals. The European Commission emphasized, in its recent’Circular Economy Action Plan for a cleaner and more competitive Europe’, the need for high-quality recycling toward qualitative secondary materials. Also in this plan, it was included that legislation for the safe mechanical recycling of plastics other than PET into FCM needs to be established in the future (European Commission, 2020).

1.2. *Environmental performance of recycling strategies and substitutability of secondary materials*

To compare the environmental impact of different treatment options for plastic waste, Life Cycle Assessment (LCA) is a commonly used method (Lazarevic et al., 2010). When secondary resources are recovered, the so-called multi-functionality issue needs to be addressed to obtain a fair result for the different products of the multi-functional system. The environmental impacts of the primary material production, the recovery processes and the final waste management should be shared amongst the products (“multiple functions”) of consecutive life cycles (Ekvall and Tillman, 1997; Rehberger and Hiete, 2020). To handle multi-functionality issues, the international standard ISO 14,044 prescribes an order of procedures to be followed: (1a) subdivision, (1b) system expansion, (2) allocation based on an underlying physical relationship, and (3) allocation based on another relationship (e.g. economic) (ISO, 2006). The substitution or avoided burden approach is regarded as a special or simplified type of system expansion, where, in the context of secondary resource recovery, the environmental impact of the primary resource that can be displaced by the secondary resource in an equivalent way is subtracted from the impact of the multi-functional system (Rigamonti et al., 2020). The calculation of these avoided impacts, also called savings or credits, is a key aspect in LCAs of recycling strategies because they can dominate the overall LCA results (Vadenbo et al., 2017). Often a 1:1 substitution ratio, meaning that 1 kg recycled material can substitute 1 kg of virgin material, cannot be justified and may be a reason for overestimation of the benefits from recycled material use (Rigamonti et al., 2020). For example, Civancik-Uslu et al. (2021) covered the prospective LCA of MR and thermochemical recycling (TCR) of four plastic fractions and assumed a 1:1 substitution ratio between both the MR and TCR products and their virgin alternatives. The results showed that MR performed better than TCR for all plastic fractions mainly because of the higher avoided impact of virgin materials in case of MR. Knowing the heterogeneous composition of household plastic waste (Roosen et al., 2020) and the challenges of MR described in section 1.1, the results of Civancik-Uslu et al. (2021) most probably overestimate the environmental benefits from MR. When comparing the environmental performance of MR and CR of post-consumer plastic waste, two aspects should be taken into account in the substitution modelling: (1) the reduction in the technical quality of the plastic after MR and (2) the potential fields of application for the recycled material, which is directly related to its potential market size.

Vadenbo et al. (2017) introduced a framework to account for product displacement or substitution related to resource recovery in LCA. In this framework, two main ways to calculate the substitution of virgin materials were distinguished. The first one calculates the displacement rate obtained through market-based modelling (Zink et al., 2016). Because of issues with data availability and varying market prices causing unstable results, this approach is not commonly applied by LCA practitioners. The second, more commonly applied, approach uses the product of the (technical) substitutability and the market response as a proxy for the displacement rate (Vadenbo et al., 2017).

Technical substitutability starts from two (or more) materials that are fully or partially substitutable alternatives for the functionality of a particular application. Rigamonti et al. (2020) gave an overview of the technical substitutability factors available in literature and concluded that there was no harmonisation in how they were calculated. They proposed therefore an equation based on the ratio of the main technical property values of the substitutable materials. A technical property was considered as the main one when it is necessary to fulfil the key function in relation to a specific application. The selection of the main technical property, however, can be difficult knowing that multiple material and geometric characteristics are essential to deliver the full product functionality (Ragaert et al., 2017). Demets et al. (2021) developed a method to calculate the technical substitutability of recycled plastics composed of two quality subfactors, one accounting for processability and one for mechanical properties. Golkaram et al. (2022) developed a quality model for recycled plastics that accounts for a very broad range of properties, such as mechanical and processability properties but also odour and colour.

Because functional equivalence does not necessarily lead to market displacement, in the framework of Vadenbo et al. (2017) the technical substitutability is multiplied with the market response. Vadenbo et al. (2017) defined the market response as the “expected change in consumption/activity level(s) of the affected compensatory product system (s)”. Little guidance, however, was offered on how to calculate the market response, as all the presented case studies include a market response of 100 %. In Horodytska et al. (2020), the substitution rate, equivalent to the technical substitutability, is multiplied with the potential market share of the intended applications, to compare closed-loop and open-loop recycling of post-industrial printed films. While open-loop recycling could only target 24 % of the European low-density polyethylene (LDPE) market (including sectors such as building and construction, automotive, etc.), closed-loop recycling including a de-inking step provided materials suitable for all sectors and thus allowed a market share of 100 % (Horodytska et al., 2020).

1.3. *Focus and objective of this study*

In this article a data-driven method for the calculation of the substitutability of recycled materials from plastic waste is presented. To address the reduction in the quality of the plastic after MR and to differentiate the potential market size between MR and CR products due to legislative safety constraints, the (overall) substitutability of the recycled product is modelled by the calculation of a technical substitutability (TS) factor and a market substitutability (MS) factor, respectively. The method is illustrated by applying it to the LCA case study from Civancik-Uslu et al. (2021), allowing a more fair comparison of the environmental benefits from MR and TCR. For the TS of mechanical recyclates, the equation from Demets et al. (2021) for recycling quality, based on several mechanical properties, is used and simplified for the case study. The calculation of TS is illustrated for in total nine applications from mechanical recyclates, including both currently possible non-FCM applications and hypothetically possible future FCM applications. The TS of chemical feedstock from TCR is considered equal to 1, because post-treatment steps to increase the feedstock's quality to a level similar to that of the virgin alternatives are included within the scope of the LCA. The MS of mechanical recyclates is calculated for two sets of scenarios in order to distinguish between currently possible non-FCM applications and hypothetically possible future FCM applications. The MS of TCR products is set equal to 1, because there are no legislative constraints that prevent them from targeting the same market as their virgin alternatives.

2. Material and methods

This section is divided in two main sections, i.e., a first section describing the LCA case study, based on Civancik-Uslu et al. (2021), and a second section explaining the method for the calculation of the substitutability of recycled materials.

2.1. LCA case study

2.1.1. Introduction of the case study

In Belgium, the former so-called "PMD" system collected since 1994 **P**lastic bottles and flasks together with **M**etal packaging and **D**rink cartons at the kerbside. From 2021 onwards, the separately collected "P + MD" waste includes also other plastic packaging like films, trays, tubes, etc. and is sorted at the sorting plant in 14 fractions including 11 plastic fractions, two metal fractions and a fraction of drink cartons (Fostplus, 2019). This article focuses on the recycling of four newly sorted plastic fractions, i.e., polypropylene (PP) rigid, polystyrene (PS) rigid, mixed polyolefins (MPO) rigid, and polyethylene (PE) films. The composition of these fractions is presented in Table 1.

2.1.2. Goal and scope definition

The goal of the LCA is to assess the environmental performance of MR and TCR of four household plastic waste fractions in Belgium. The functional unit is defined as 1 metric ton of a specific plastic waste fraction (including dirt and moisture) that is mechanically or thermochemically recycled. While doing this comparison, incineration with electricity and heat recovery (IN) is used as a reference scenario. Fig. 1 presents the process flow diagrams of the MR and TCR scenarios for the four plastic fractions.

A detailed description of the included processes can be found in Civancik-Uslu et al. (2021). The system boundary includes the sorting of the collected bags of household mixed packaging waste, thus excluding the steps of primary material production, manufacturing of the initial products and waste collection because these steps are similar amongst the MR and TCR scenarios. The sorted waste fractions are recycled, including all necessary pre-treatment and post-treatment steps depending on their composition. Sorting is not included in the incineration scenario because it is not required before incineration of plastics as part of the residual waste. The system boundary ends with the production of the recycled products that are listed in Table 1. To compare the environmental benefits from MR and TCR, credits are calculated for these recycled products following the methodology explained in section 2.2. In addition to the recycled (material) products presented in Table 1, in both MR and TCR scenarios, also minor amounts of recovered energy from incineration of residues were taken into account.

Table 1
Composition of the sorted plastic fractions (wt.%), modelled based on the studies of Roosen et al. (2020) and Kleinhans et al. (2021). Recycled products, as well as the recycling yields, in the mechanical recycling (MR) and thermochemical recycling (TCR) scenarios for each of the sorted plastic fractions.

Plastic waste fraction		PP rigids	PS rigids	MPO rigids	PE films
Composition of the sorted fraction					
PP		90	–	25.9	–
PS		–	93.2	–	–
PE		3.5	0.2	48.2	78.8
PET		0.5	0.5	1.1	–
Dirt and moisture		5.3	4.2	6.0	9.1
Others (missorted plastics, EVOH[c], paper, etc.)		0.7	1.9	18.8	12.1
Recycled products and yields					
MR	Recycled product	PP regranulates	PS regranulates	MPO flakes[a]	LDPE regranulates
	Yield (%)	85.2	89.5	58.4	81.2
TCR	Recycled product	naphtha and slack wax	mainly styrene[b]	naphtha and slack wax	naphtha and slack wax
	Yield (%)	81.5	81.7	53.0	79.8

[a] The focus is on MPO flakes because possible applications for this fraction are feasible through direct extrusion from flakes.

[b] Because of confidentiality reasons, small quantities of other products cannot be revealed.

[c] Ethylene vinyl alcohol.

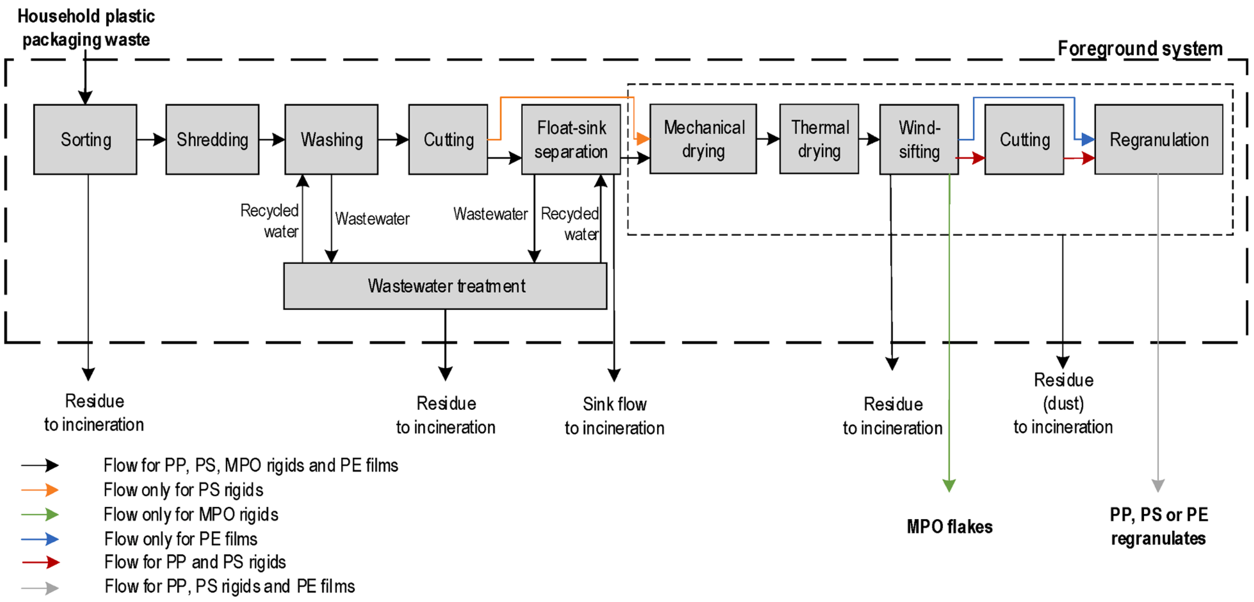
2.1.3. Inventory analysis

As mentioned in section 1.4, this study applies the methodology explained in more detail in section 2.2 to the LCA case study from Civancik-Uslu et al. (2021). Depending on the modelled composition of each plastic waste fraction, prospective MR and TCR processes were designed for each fraction based on currently known technologies in collaboration with experts from both academia and industry. While MR scenarios were based on industrial scale data, TCR scenarios were based on a pilot plant's design specifications (50 kton for PS and 120 kton for polyolefins). For 1 metric ton of sorted plastic waste (including dirt and moisture), the recycling yields are summarized for all MR and TCR scenarios in Table 1. These yields take into account all mass losses happening during pre-treatments, recycling and post-treatments.

The systems studied can be separated into foreground and background subsystems. The foreground subsystem consists of all processes shown in Fig. 1. The background subsystem includes the other processes that are linked through mass or energy exchanges with foreground processes. The production of electricity is an example of a background process. While foreground data were primary data collected during 2018–2020 from Belgian companies and modified for each waste fraction together with both industrial and academic experts, secondary background data were extracted from the ecoinvent v3.6 database (2019). For modelling the impacts of the incineration with energy recovery scenario (IN), ecoinvent datasets were used because no primary data were available. Based on the waste fractions' composition, the impacts of incinerating each component (e.g. PE, PS, paper, etc) were calculated through their respective ecoinvent datasets and finally added. The avoided impacts of electricity and heat production were taken into account, based on ecoinvent data. The life cycle data inventory for all MR, TCR and IN scenarios can be found in the supplementary material (section A).

2.1.4. Impact assessment

Two impact assessment methods were applied to the case study in this article: (i) Cumulative Exergy Extraction from the Natural Environment (CEENE) v2013 for consumption of natural resources (Alvarenga et al., 2013; Dewulf et al., 2007) and (ii) ReCiPe 2016 (H) Midpoint v1.1 (Huijbregts et al., 2017) for impacts from emissions. The CEENE



b. Thermochemical recycling (TCR)

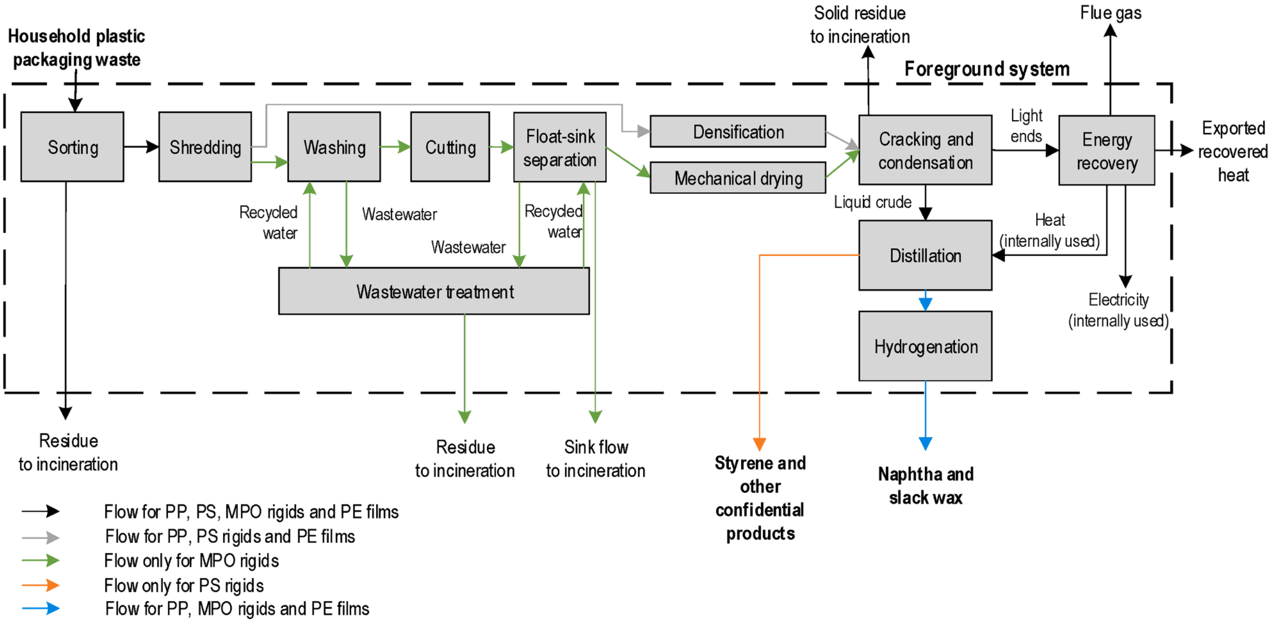


Fig. 1. Process flow diagram of MR (a) and TCR (b) scenarios for PP, PS, MPO rigids and PE films. The dashed lines include the foreground systems. The sorting system was modelled as a black box. Redrafted after Civancik-Uslu et al. (2021b).

method has been recommended as the most appropriate method to quantify a resource footprint based on thermodynamics (Berger et al., 2020; Liao et al., 2012). CEENE results are presented as CEENE total which is the total consumption of natural resources, which include fossil, nuclear, renewable (wind and hydro energy), water, mineral, metal and land resources, in joule exergy (J_{ex}). Additionally, global warming (kg CO_2 eq.) and terrestrial acidification (kg SO_2 eq.) impacts calculated with the ReCiPe method were selected for analysis in this article. These impacts are among the most commonly studied impact categories in LCA studies on plastic waste management (Gear et al., 2018; Hou et al., 2018; Khoo, 2019; Lazarevic et al., 2010), and were also covered in Civancik-Uslu et al. (2021) to which this study compares. The results of resource consumption and global warming impact are presented in section 3, whereas terrestrial acidification results are presented in the supplementary material, section E.

To compare the environmental performance of MR and TCR, the net environmental impact per metric ton of waste is calculated according to **Eq. (1)**.

$$\textit{Net environmental impact} = B - \left(S_{vm,1:1} \times TS \times MS\right) - S_{e,1:1} \quad (1)$$

With the net environmental impact [impact unit/metric ton waste]; *B*, the burdens of sorting, pre-treatment, recycling and post-treatment in case of MR and TCR, or the burdens of incineration in case of IN [impact unit/metric ton waste]; $S_{vm,1:1}$, the savings from avoiding virgin material based on a 1:1 substitution in case of MR and TCR [impact unit/metric ton waste]; *TS*, the technical substitutability of the recycled material [0,1]; *MS*, the market substitutability of the recycled material [0,1]; and $S_{e,1:1}$, the savings by energy recovery through incineration based on a 1:1 substitution for heat and electricity [impact unit/metric ton waste]. *TS*

× *MS* results in *OS*, the overall substitutability [0,1]. The next section explains the calculations of TS and MS.

2.2. *Technical and market substitutability of recycled materials*

2.2.1. *Technical substitutability (TS)*

Starting from the framework of Vadenbo et al. (2017), the technical substitutability (TS) is defined as the degree of technical functional equivalence between different products for a particular application. In this article, the technical substitutability for TCR products (TS_{TCR}) was set equal to 1, because the quality of the produced naphtha, slack wax and styrene-related products was considered to be similar as their virgin alternatives because post-treatment steps (distillation and (in some cases also) hydrogenation, see Fig. 1b) were included. During MR, thermal–mechanical degradation of polymers takes places and contaminations are not completely removed. For MR products, TS_{MR} can be interpreted as the degree of dilution with virgin material that is required to achieve technical functional equivalence with the virgin reference material for a specific application. The calculation of TS_{MR} thus depends on the envisaged application; in other words, it is not possible to define a generic TS_{MR} valid for all possible applications. It should, however, be possible, when for a large sample of applications data about technical properties are available, to calculate an average TS_{MR} for a group of applications that are all technically feasible for a particular mechanically recycled material. In this article, at least two (in some cases three) technically feasible applications for each mechanically recycled material from the four plastic fractions were defined by recycling experts (Table 2). Besides the technical properties of the recycled material, the selection of feasible applications also depends on the legislation EC No 282/2008 regarding the use of recyclates in FCM applications (European Commission, 2008). As the current FCM legislation makes it extremely challenging to use non-PET mechanical recyclates for FCM application, a first group of applications only consists of non-FCM applications. Additionally, a second group of FCM applications was defined in order to address hypothetical (optimistic) future scenarios where also non-PET recyclates that are technically suitable for FCM application are able to receive the corresponding legislative status via EFSA approval. As the additional efforts to make these hypothetical scenarios possible could not yet be estimated in the current study, they should be kept in mind when interpreting the results. In total, nine applications (six non-FCM and three FCM) were defined and are presented in Table 2. More information on the selection process for the applications can be found in the supplementary material (section B).

Table 2
Overview of the defined applications for the mechanically recycled materials in this article for both the current and the potential future legislative status regarding FCM application.

Plastic fraction	Recycled material	Applications		
		Current non-FCM applications		Potential future FCM applications
PP rigids	Regranulates	Storage box through injection moulding (**PP_c,box**)	Large decorative flower pot through injection moulding (**PP_c, flowerpot**)	Caps for beverage bottles (**PP_f, capbottle**)
PS rigids	Regranulates	Non-food tray through thermoforming (**PS_c,tray**)		Food (yoghurt) pots through thermoforming (**PS_f,yoghurtpot**)
MPO rigids	Flakes	Street bench through extrusion (**MPO_c, bench**)	Pallet through injection moulding (**MPO_c, pallet**)	Food packaging was considered not technically feasible for this recycled material
PE films	Regranulates	Garbage bag or agricultural film through film blowing (**PE_c,garbagebag**)		Seal film for food packaging (**PE_f, sealfilm**)

After defining the applications, the technical substitutability for MR products (TS_{MR}) was calculated according to Eq. (2), which is a simplification of the scoring function to calculate the mechanical recycling quality (RQ^{mech}) factor from Demets et al. (2021). The simplification in Eq. (2) implies that ‘the higher the mechanical property value, the better the quality of the recycled material for the intended application’. This simplification is not always adequate, depending on the property. Properties like impact strength, tensile strength and strain at break only have a lower threshold value, so the higher the value, the better. However, other properties like elastic modulus are constricted by an optimal value range, defined by both a lower and an upper threshold limit. In our case study, all rigid applications (PP, PS and MPO) benefit from higher elastic modulus values, while the PE film application must remain sufficiently flexible (corresponding to an upper threshold for elastic modulus). However, for the PE film application, elastic modulus values for the recycled and virgin material are statistically identical (Table 3), so there is no need to further complicate the calculation. Therefore, the simplified Eq. (2) does hold for all the selected applications in this study.

$$TS_{MR} = \sum_{i=1}^{n} \left(w_i \cdot \frac{Pi_{recycled}}{Pi_{virgin}} \right) \tag{2}$$

with $Pi_{recycled}$ the value of the mechanical property i of the mechanical recyclate, Pi_{virgin} the value of the mechanical property i of the virgin reference material for a specific application and w_i: application-specific weighting factors for the mechanical property i [0,1]. For **Eq. (2)** it holds that $\sum_{i=1}^{n} w_i = 1$ and $\frac{Pi_{recycled}}{Pi_{virgin}} \leq 1$; when $Pi_{recycled} > Pi_{virgin}$ the ratio $\frac{Pi_{recycled}}{Pi_{virgin}}$ is set equal to 1. **Eq. (2)** enables to account for n relevant mechanical properties of the mechanical recyclate compared to those properties of its virgin alternative that is defined for a specific application. The application-specific weighting factors reflect the relative importance of the selected mechanical properties for a specific application. Both the selection of relevant mechanical properties and the definition of the application-specific weighting factors were done by recycling experts (supplementary material, section B). An overview of the mechanical properties and their values for the mechanically recycled material and its virgin alternative in each of the nine applications is presented in Table 3. Information on how the data for the mechanical properties of the recycled and virgin materials were obtained can be found in the supplementary material (section C). The application-specific weighting factors are shown in Table 4. With **Eq. (2)**, three TS_{MR} values were calculated for each application: an average TS_{MR} based on the average values of the mechanical properties, and an optimistic and pessimistic TS_{MR} based on the standard deviation (stdv) data of the mechanical properties. For the optimistic TS_{MR}, the average values + stdv for the recycled material was used, while the average value - stdv was used for the virgin material. For the pessimistic TS_{MR}, the average value - stdv for the recycled material was used, while the average value + stdv was used for the virgin material.

2.2.2. *Market substitutability (MS)*

MS is defined as the potential share of the total market size of the reference virgin material that can be targeted by the recycled material, taking into account legislative constraints (e.g. regarding chemical composition and/or migration behaviour of the applied material). The market substitutability for TCR products (MS_{TCR}) was set equal to 1, because there are no legislative constraints that prevent them from being used in the same market as their virgin fossil-based alternatives. For MR products, MS_{MR} was defined for two sets of scenarios, i.e. currently possible scenarios and hypothetical future optimistic scenarios (Table 5). The current situation relies on Eriksen et al. (2019) for data on

Table 3
Overview of the mechanical property data for the mechanically recycled materials and their virgin alternatives for each application.

Plastic fraction	Recycled material or virgin reference material	Applications	Impact strength [kJ/m^2]	Elastic modulus [MPa]	Tensile strength [MPa]	Strain at break [%]
PP rigids	Recycled	**PP_c,box, PP_c,flowerpot and PP_f,capbottle**	6 ± 0	1201 ± 35	26.6 ± 0.2	31.3 ± 4.3
	Virgin	**PP_c,box**	3 ± 1	1450 ± 44	26.2 ± 4.0	8.4 ± 2.0
		PP_c,flowerpot	7	1650	29	n/a
		PP_f,capbottle	3 ± 0	1455 ± 22	34.9 ± 0.4	26.0 ± 6.8
PS rigids	Recycled	**PS_c,tray and PS_f,yoghurtpot**	7 ± 1	2392 ± 54	27.4 ± 0.4	42.2 ± 7.4
	Virgin	**PS_c,tray**	12	1800	24	35
		PS_f,yoghurtpot	n/a	1900	28	70
MPO rigids	Recycled	**MPO_c,bench and MPO_c,pallet**	3 ± 0	1096 ± 81	14.0 ± 0.5	n/a
	Virgin	**MPO_c,bench**	3 ± 0	864 ± 48	23.8 ± 0.4	268.9 ± 53.9
		MPO_c,pallet	10 ± 1	1141 ± 41	23.0 ± 0.5	n/a
PE films	Recycled	**PE_c,garbagebag and PE_f,sealfilm**	n/a	106 ± 15	17.0 ± 1.6	644.5 ± 118.6
	Virgin	**PE_c,garbagebag and PE_f,sealfilm**	n/a	105 ± 15	17.7 ± 1.6	639.5 ± 34.5

Table 4
Weighting factors based on expert judgement for each application.

Plastic fraction	Application	Weighting factors			
		Impact strength	Elastic modulus	Tensile strength	Strain at break
PP rigids	**PP_c,box**	0.5	0.2	0.3	0
	PP_c, flowerpot	0.4	0.4	0.2	0
	PP_f, capbottle	0.1	0.4	0.3	0.2
PS rigids	**PS_c,tray**	0	0.5	0.3	0.2
	PS_f, yoghurtpot	0	0.5	0.3	0.2
MPO rigids	**MPO_c, bench**	0.1	0.7	0.2	0
	MPO_c, pallet	0.3	0.5	0.2	0
PE films	**PE_c, garbagebag**	0	0.1	0.5	0.4
	PE_f,sealfilm	0	0.2	0.4	0.4

European virgin plastic-specific market shares of the most important application sectors (see supplementary material, section D). Based on the strictness of legal requirements regarding chemical composition and/or migration behaviour, Eriksen et al. (2019) divided eight key application groups into three categories from high over medium to minimal legal requirements: (i) food packaging (European Commission, 2008), (ii) toys (European Commission, 2009), pharmaceutical applications (European Commission, 2007), and electrical and electronic equipment (European Commission, 2011), and (iii) building and construction, non-food packaging, automotive and others. In the scenarios for the current situation, food packaging, toys and pharmaceutical applications were excluded as potential application sectors for the calculation of MS_{MR}. Electrical and electronic equipment was not excluded in the current scenario as the requirements (e.g. maximum allowable metal concentrations) were never exceeded in the analysis of household plastic packaging waste in Belgium by Roosen et al. (2020). Considering the excluded market sectors for the current situation, it can be noted that the market share of food packaging represents the main share. In the hypothetical future scenarios, the MS_{MR} was set equal to 1, reflecting full market substitution potential. The hypothetical future scenarios should be considered as optimistic scenarios; they were included in this article as they can bring useful insights about the role of MR towards higher closed-loop recycling rates of products such as food packaging. While the MS_{MR} for the current situation will be multiplied with TS_{MR} for the defined currently possible non-FCM applications, the TS_{MR} for the potential future FCM applications will be multiplied with MS_{MR} equal to 1, resulting then in OS_{MR} values.

It is important to highlight that in our approach presented in this article only the calculation of TS_{MR} is specific to a particular application, such as a flower pot or a street bench (see **section 2.2.1**), while MS_{MR} is not defined specifically for that (single) application. As an example, this means that the current MS_{MR} for PP rigids of 0.81 (Table 5) should be interpreted as 81 % of the total market size of virgin PP that can potentially be displaced by the mechanically recycled PP material, taking into account legislative constraints regarding chemical properties of the recycled material. As said before, the set of analysed applications should ideally be expanded in future research in order to enable the calculation of an average TS_{MR} for a group of applications that are all technically feasible for a particular mechanically recycled material. Finally, also note that the market substitutability does not account for the discrepancy between the (currently still lower) available amount of both mechanically and chemically recycled materials and the (higher) demand for recycled materials when they would displace all the virgin material demand.

Table 5
Overview of the European virgin polymer market shares of food packaging, toys and pharmaceutical applications for PP, PS, mixed plastic and film, based on data from Eriksen et al. (2019), and the thereupon based calculated market substitutability factors for MR products (MS_{MR}) in the current and hypothetical future scenarios for each plastic waste fraction.

European virgin polymer markets (Eriksen et al., 2019)	Food packaging (%)	Toys (%)	Pharmaceutical applications (%)	Other applications[a] (%)	Plastic waste fraction in this study	MS_{MR} current	MS_{MR} hypothetical future
PP	18	0	1	81	**PP rigids**	0.81	1
PS	15	0	1	84	**PS rigids**	0.84	1
Mix[b]	34	1	1	64	**MPO rigids**	0.64	n/a[c]
Film	54	0	1	45	**PE films**	0.45	1

[a] ‘Other applications’ include electrical and electronic equipment, building and construction, non-food packaging, automotive and others (Eriksen et al., 2019).
[b] Based on polymer markets that were weighted in relation to European polymer production (Eriksen et al., 2019).
[c] For this fraction, food packaging production was considered not technically feasible by recycling experts.

3. Results

3.1. Technical substitutability (TS_{MR}) and overall substitutability (OS_{MR}) in case of MR

Table 6 presents the TS_{MR} for the nine defined applications of the four MR products. Overall, the TS_{MR} range from 0.63 for the use of MPO flakes for pallet production (**MPO_c,pallet**) to 1.00 for the use of LDPE regranulates for garbage bag or agricultural film production (**PE_c, garbagebag**) or for seal film (for food packaging) production (**PE_f, sealfilm**). For the applications analysed in this article, it can be seen that the TS_{MR} for the potential future FCM applications are not noticeably lower than the TS_{MR} for the current non-FCM applications, showing the potential of using mechanical recyclates for FCM applications from a pure technical point of view.

In Table 6, the OS_{MR}, calculated as TS_{MR} (Table 6) × MS_{MR} (Table 5), are shown. The OS_{MR} range from 0.35 for the current use of LDPE regranulates (**PE_c,garbagebag**) to 1.00 for the potential future use of LDPE regranulates (**PE_f,sealfilm**). These results clearly reflect the influence of the MS_{MR} factor. In case of PE films, the European market share of food packaging is the highest amongst all polymer types (Table 5), therefore, lowering the OS_{MR} to a great extent in case of the current scenario where only non-FCM applications are possible.

3.2. Effect on environmental impact results

Fig. 2 presents the resource consumption and global warming impacts per ton plastic fraction for seventeen scenarios (9 × MR, 4 × TCR and 4 × IN). Figure S.1 in the supplementary material (section E) presents the results for terrestrial acidification. In case of MR, two or three scenarios are presented for each plastic fraction depending on the defined application by recycling experts (section **2.2.1**). The environmental impacts for each scenario are shown in two manners. First, a stacked bar including burdens in red and savings in green. Second, a black dash on the bars shows the net impact. Additionally, the reduction in MR savings by moving from the approach where a full substitution for MR products (equivalent to $OS_{MR} = 1$) was assumed (Civancik-Uslu et al., 2021) to the approach in this article, where (the average) TS_{MR} (Table 6) and MS_{MR} (Table 5) are considered, is visually represented by a black upwardly directed arrow.

While, by assuming a full substitution for both MR and TCR products, Civancik-Uslu et al. (2021) concluded that for the net environmental impact in terms of resource consumption, global warming and terrestrial acidification MR was always favourable over TCR, this study shows that for several scenarios the net impact of MR becomes worse than TCR. The next sections describe the results more in detail.

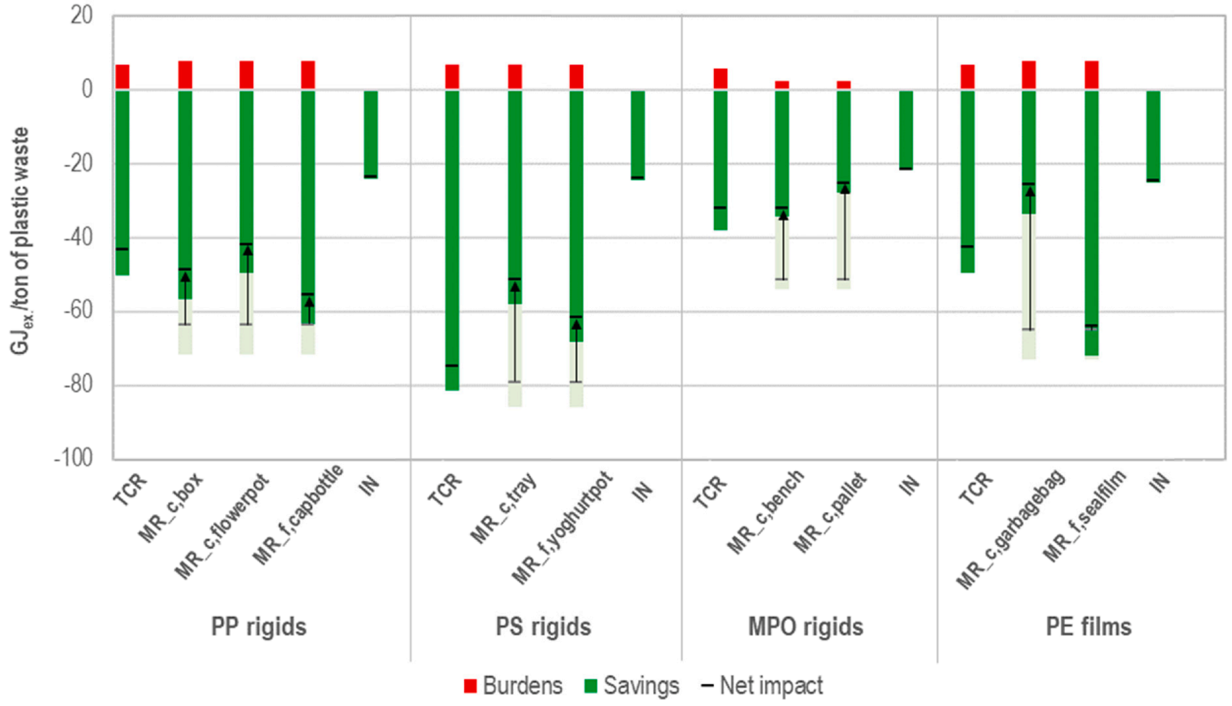
3.2.1. Resource consumption

Compared to IN, both MR and TCR obtain better results for resource consumption for all defined applications. Whether the net impacts differ substantially, varies amongst the scenarios. In case of small differences, they should be carefully interpreted considering the data and model uncertainties involved in LCA. Compared to IN, MR obtains a 4–158 % better impact, and TCR a 50–211 % better impact. Out of nine scenarios, MR obtains a 1–40 % worse net resource consumption impact than TCR in six scenarios (Fig. 2**a**). The major differences (>10 %) between MR and TCR scenarios are described here. The net impact of MR is worse than TCR, about 40 %, for PE films when LDPE regranulates are used for, currently possible, non-FCM film production (**PE_c,garbagebag**), and about 31 %, for PS rigids when PS regranulates are used for the, currently possible, non-FCM application (**PS_c,tray**). In case of the future FCM application from PS regranulates (**PS_f,yoghurtpot**), the net impact of MR is 17 % worse than TCR. Focusing on MPO rigids, the net impact of MR is 21 % worse than TCR when MPO flakes are used for currently possible pallet production (**MPO_c,pallet**). MR maintains the best environmental performance in terms of resource consumption in case of only one currently possible non-FCM application (**PP_c,box**), about 13 % better, and two potential future FCM applications: about 29

Table 6
Overview of the technical (TS_{MR}) and overall substitutability (OS_{MR}) for MR products for the defined applications of the plastic fractions. stdv: standard deviation.

Plastic fraction	Application		Technical substitutability for MR products (TS_{MR})		
			Average value	Pessimistic value based on stdv	Optimistic value based on stdv
PP rigids	**Current**	**PP_c,box**	0.97	0.92	0.98
		PP_c,flowerpot	0.84	0.81	0.87
	Future	**PP_f,capbottle**	0.88	0.87	0.90
PS rigids	**Current**	**PS_c,tray**	0.80	0.80	0.80
	Future	**PS_f,yoghurtpot**	0.79	0.79	0.80
MPO rigids	**Current**	**MPO_c,bench**	0.92	0.91	0.92
		MPO_c,pallet	0.70	0.63	0.74
PE films	**Current**	**PE_c,garbagebag**	0.98	0.79	1.00
	Future	**PE_f,sealfilm**	0.98	0.78	1.00
Plastic fraction	**Application**		**Overall substitutability for MR products (OS_{MR})**		
			Average value	**Pessimistic value based on stdv**	**Optimistic value based on stdv**
PP rigids	**Current**	**PP_c,box**	0.78	0.74	0.79
		PP_c,flowerpot	0.68	0.66	0.71
	Future	**PP_f,capbottle**	0.88	0.87	0.90
PS rigids	**Current**	**PS_c,tray**	0.67	0.67	0.67
	Future	**PS_f,yoghurtpot**	0.79	0.79	0.80
MPO rigids	**Current**	**MPO_c,bench**	0.59	0.58	0.59
		MPO_c,pallet	0.45	0.40	0.47
PE films	**Current**	**PE_c,garbagebag**	0.44	0.35	0.45
	Future	**PE_f,sealfilm**	0.98	0.78	1.00

a. Resource consumption



b. Global warming

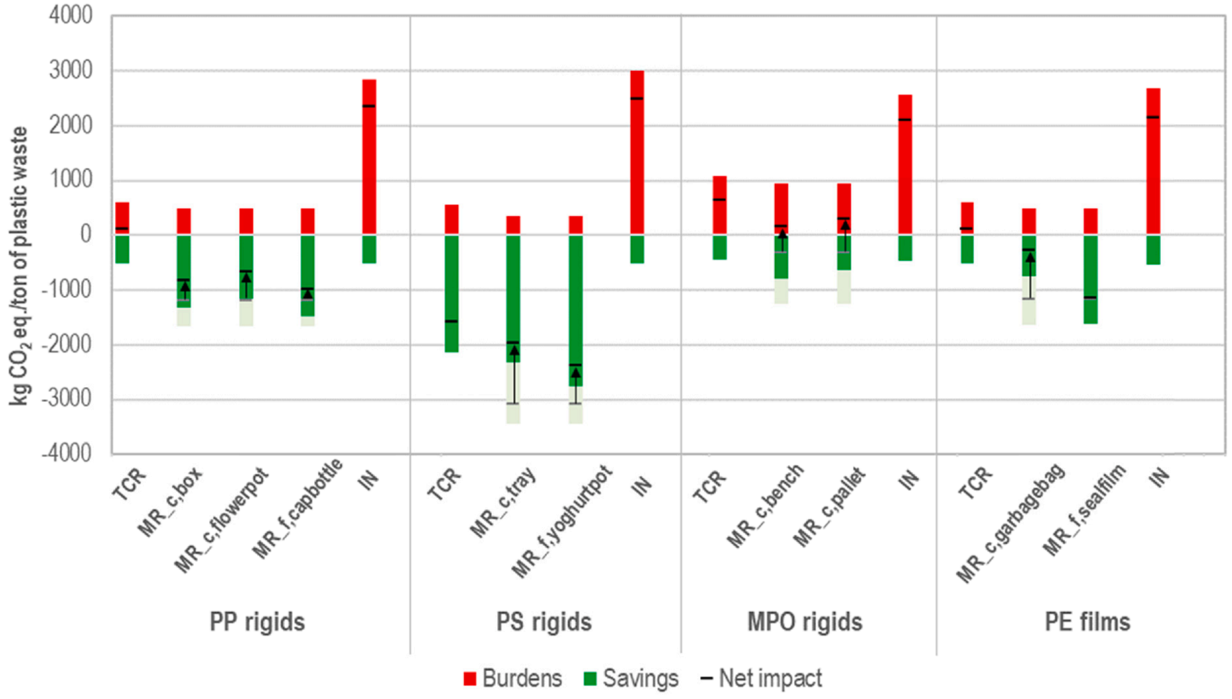


Fig. 2. Resource consumption (a) and global warming (b) impacts of thermochemical recycling (TCR) and mechanical recycling (MR) compared to the reference incineration with energy recovery (IN) for PP, PS, MPO rigids and PE films. In case of MR, for each plastic fraction, one or two current applications (MR_c,…) were defined, and one future FCM application (MR_f,…) if applicable. The black upwardly directed arrows show the reduction in savings (and net impact) by accounting for the average OS_{MR} values shown in Table 6 instead of OS_{MR} equal to 1. Note that the reduction in case of *MR_f,sealfilm* for PE films is too small to visualize with an arrow.

% better for **PP_f,capbottle** and 50 % better for **PE_f,sealfilm**.

As the number of applications analysed in this article was limited to nine, it is useful to define the TS_{MR} threshold for which the net environmental impacts of MR and TCR become equal. The TS_{MR} threshold was calculated by keeping the MS_{MR} of the scenario constant and looking at which TS_{MR} value the net impact of MR and TCR became equal. First, for currently possible non-FCM applications, the TS_{MR} threshold for which the net environmental impacts of MR and TCR are equal amounts to 0.87 for PP rigids. This explains why the net environmental impact of MR is better than TCR when PP regranulates are used for storage box production (**PP_c,box**) as the TS_{MR} equals 0.97 for that application, which is higher than 0.87 (Table 6). For the other plastic fractions, i.e. PS rigids, MPO rigids and PE films, the net environmental impact of TCR is always better than MR in case of currently possible non-FCM applications. In other words, for these non-FCM applications, an unrealistic TS_{MR} higher than 1 should be required to have a better net environmental impact of MR compared to TCR for PS rigids, MPO rigids and PE films. Second, for FCM applications potentially feasible in the future, the TS_{MR} threshold for which the net environmental impact of MR and TCR are equal amounts to 0.70, 0.95 and 0.68 for PP rigids, PS rigids and PE films, respectively. While the net environmental impact of MR is better than TCR in case of the potential future FCM applications for PP rigids (**PP_f,capbottle**) and PE films (**PE_f,sealfilm**), it is not the case for the potential future FCM application from PS rigids (**PS_f,yoghurtpot**) as the calculated TS_{MR} equals only 0.79 ($<$0.95) for the latter application (Table 6).

3.2.2. *Global warming*

Compared to IN, both MR and TCR obtain far better results for global warming for all defined applications. MR obtains a 86–196 % better impact than IN, while TCR obtains a 69–163 % better impact than IN. Compared to TCR, MR maintains the lowest net global warming impact for all scenarios (Fig. 2**b**). All differences in net impact between MR and TCR are larger than 25 %. The largest differences between MR and TCR results are obtained for: (i) PE films when LDPE regranulates are used for FCM film production in the future (**PE_f,sealfilm**): more than twelve times better (-1137 versus 101 kg CO_2 eq./metric tonne PE films for MR and TCR, respectively), (ii) PP rigids when PP regranulates are used for bottle cap production in the future (**PP_f,capbottle**): more than ten times better (-993 versus 100 kg CO_2 eq./metric tonne PP rigids for MR and TCR, respectively), and (iii) PP rigids when PP regranulates are used for the currently possible storage box production (**PP_c,box**): more than nine times better (-830 versus 100 kg CO_2 eq./metric tonne PP rigids for MR and TCR, respectively).

For currently possible non-FCM applications through MR, the TS_{MR} threshold for which the net environmental impacts of MR and TCR are equal amounts to 0.26, 0.66, 0.46 and 0.22 for PP rigids, PS rigids, PE films and MPO rigids, respectively. This explains why the net environmental impact of MR was found to be always better than TCR as the TS_{MR} in Table 6 are higher than the respective thresholds. For FCM applications potentially feasible in the future through MR, the TS_{MR} threshold for which the net environmental impact of MR and TCR are equal amounts to 0.21, 0.56 and 0.21 for PP rigids, PS rigids and PE films, respectively.

3.2.3. *Terrestrial acidification*

Compared to IN, both MR and TCR obtain far better results for terrestrial acidification for all defined applications (Fig. S.1 in supplementary material, section E). Compared to IN, MR obtains a 4–23 times better impact, and TCR a 10–19 times better impact. MR obtains a 19–57 % worse net terrestrial acidification impact than TCR in six out of nine scenarios. Highlighting the biggest differences, the net impact of MR is worse than TCR, about 57 %, for PE films when LDPE regranulates are used for, currently possible, non-FCM film production (**PE_c,garbagebag**), and about 54 %, for MPO rigids when MPO flakes are used for the currently possible pallet production (**MPO_c,pallet**). MR obtains a comparable or better net impact than TCR in three scenarios: **PS_c,tray** (negligible difference)**, PE_f,sealfilm** (negligible difference) and **PS_f, yoghurtpot** (MR 18 % better).

3.3. *Discussion*

In this study the overall substitutability (OS_{MR}) was defined as the multiplication of the technical (TS_{MR}) and market substitutability (MS_{MR}) of MR products. When assuming full substitution ($OS_{MR} = 1$), the environmental savings from the substitution of virgin materials by MR products are generally higher compared to the ones by TCR products. This can be explained by MR avoiding additionally the processes steam cracking, polymerisation and granulation compared to TCR which only avoids the virgin supply of the feedstock for polymer production (Civancik-Uslu et al., 2021).

Regarding the calculation of TS_{MR}, it must be acknowledged that the calculation of TS_{MR} could be further advanced by accounting, in addition to a set of mechanical material properties, for other criteria such as parameters reflecting processability of the recycled material (e.g. melt flow index), but also odour and colour, all in function of the envisaged application (Demets et al., 2021; Golkaram et al., 2022). Odour and colour can be a cause for recycled materials not being used in certain products, while they are not an issue for other applications. Golkaram et al. (2022) demonstrated their quality model resulting in a single substitutability value that accounts for colour, odour and safety requirements, in addition to mechanical and processability properties, for three applications. Vadenbo et al. (2017) proposed to subject the technical substitutability to additional constraints such as end user's perception constraints (e.g. personal preferences), but also institutionally prescribed constraints (e.g. regulations). However, in our approach, we accounted for legislative (safety) constraints through the market substitutability factor that excludes market sectors that cannot be targeted by the recycled material because of these constraints. The data used to quantify the potential market size are subject to necessary updates over time. Future research in expanding the set of criteria accounted for in the substitutability calculation in LCA studies should therefore avoid double counting in the TS_{MR} and MS_{MR} factors.

Calculation of the substitutability can be more accurate by accounting for a broader set of properties (Golkaram et al., 2022), but also through investigating the quality-property relationship or scoring function (e.g. Trapezoidal, Gaussian), which depends on the property-application combination (Demets et al., 2021). However, more accurate substitutability calculations are quite data demanding which can be a challenge in LCA studies.

In this study, MS_{MR} was defined for two sets of scenarios, i.e. currently possible scenarios and hypothetical future optimistic scenarios. In this way, the currently existing difference between TCR and MR regarding the possibility of closed-loop recycling into products with high and medium levels of safety requirements such as food packaging, toys and pharmaceutical applications could be reflected in the LCA results. However, these hypothetical scenarios will only be realized by simultaneously improving traceability, sorting (e.g. to clearly separate food from non-food packaging) and recycling processes, as well as a clear implementation of food safety regulations (De Tandt et al., 2021). As the additional efforts to make the hypothetical scenarios possible could not yet be estimated in the current study, these should be investigated and estimated in future research.

It is important to highlight that in our approach presented in this article only the calculation of TS_{MR} is specific to a particular application (e.g. flower pot or street bench), while MS_{MR} is not defined specifically for that (single) application. One could argue that it is better to align MS_{MR} with TS_{MR} and defining both of them in terms of a specific application. For example, when using the TS_{MR} for a flower pot made from mechanically recycled PP, one could use the MS_{MR} representing only the market share of PP flower pots in the total virgin PP market. When only focusing on a single application with a relatively small market size, the market could get saturated relatively soon leading eventually to zero substitution of virgin material. The latter approach has not been followed in our study, because of two reasons. First, it requests application-specific market data that were, and are usually, not available at such a detailed level. Second, when one would calculate the market substitutability for a specific application, the possibility of using the mechanically recycled material for other potential applications would be neglected and thus resulting in an underestimation of MS_{MR}. As this study is a waste perspective LCA where the functional unit is defined in terms of the quantity of waste treated through either TCR, MR or IN, it is important to take into account the entire market potential of the mechanically recycled material. In what follows, some guidance is given on how to reproduce the approach from this article. First, the MS_{MR} for a specific mechanically recycled material can be defined based on whether one wants to analyse a currently possible scenario including only applications with minimal legal requirements regarding chemical composition and/or migration behaviour ($MS_{MR} < 1$) or a hypothetical future scenario in which applications with high and medium levels of safety requirements such as food packaging, toys and pharmaceutical applications are also feasible for mechanically recycled materials ($MS_{MR} = 1$). Second, the effect of different TS_{MR} values for a set of specific applications on the net environmental impact results can be analysed (as a kind of sub-scenario analysis). This approach enables to evaluate the choice for a specific application on the resulting net environmental impact and can thus support decision-makers in choosing the best material-application combination. As said before, it should be possible, when for a large sample of applications data about mechanical properties are available, to calculate an average TS_{MR} for a group of applications that are all technically feasible for a particular mechanically recycled material in order to obtain a more general picture.

In this article, both the technical (TS_{TCR}) and market substitutability of TCR products (MS_{TCR}), and therefore also the overall substitutability (OS_{TCR}), were considered equal to 1. It should be highlighted that the point of substitution was after post-treatment of the pyrolysis oil, therefore, TCR experts supported the full substitutability of the TCR products. However, it should be noted that the operational knowledge of TCR of post-consumer plastic waste is still at an early stage and, therefore, several challenges before its mature implementation at industrial scale can be expected. Possibly required pre-treatment (e.g. dechlorination) and post-treatment (e.g. hydrotreatment) steps could be investigated in more detail. For example, regarding the use of naphtha in steam crackers, especially the effect of remaining contaminants (after post-treatment steps) on the occurrence of coke formation and fouling should be further investigated (Kusenberg et al., 2022). Furthermore, it should be noted that additional treatment steps always come with additional impact, which should be taken into account. In addition to the route in which pyrolysis is followed by steam cracking to produce new polymers, the production of syngas through gasification combined with methanol production can also lead to new polymers (Ragaert et al., 2017). Finally, one may also expect that TCR will undergo up-scaling and learning improvements. Future research could therefore further investigate the environmental performance of TCR and the substitutability of its products in relation to the performed treatment steps.

4. Conclusions and perspectives

This article presented a data-driven method for the calculation of the overall substitutability (OS) of recycled materials from plastic waste, composed of technical (TS) and market substitutability (MS), which can be used in the calculation of the avoided impacts from virgin plastics in LCA studies. This approach enables to account for a potential reduction in, through TS, the quality of the recycled material and, through MS, the potential fields of application (market sectors) for the recycled material because legislation may hinder the use of recyclates for products with higher levels of safety requirements such as food packaging.

In this study, more specifically, the quantification of the overall substitutability of mechanically recycled products (OS_{MR}) was elaborated, composed of technical (TS_{MR}) and market substitutability (MS_{MR}), followed by its application to six non-food contact material (non-FCM) applications and three hypothetical future FCM applications, and its use in a comparative LCA of MR and CR of several plastic waste fractions in Belgium.

TS_{MR} results for the studied non-FCM and FCM applications were comparable, but OS results varied between 0.35 and 0.79 for non-FCM applications and between 0.78 and 1 for FCM applications, reflecting the lower MS_{MR} results for the current situation. As TS_{MR} for the potential future FCM applications are not noticeably lower than the TS_{MR} for the current non-FCM applications, they show the potential of using mechanical recyclates for FCM applications from a pure technical point of view. It must however be acknowledged that the calculation of TS_{MR} should be further advanced by accounting for other criteria such as ease of processing, colour and odour.

While, by assuming a full substitution (equivalent to OS = 1) for both MR and TCR products, Civancik-Uslu et al. (2021) concluded that, for resource consumption, global warming and terrestrial acidification, MR was always favourable over TCR, this study has shown that for several scenarios the net impact of MR becomes worse than TCR. Out of nine scenarios, MR obtained a worse resource consumption and terrestrial acidification impact than TCR in six scenarios. MR maintained the lowest global warming impact in all scenarios. These results confirm that a proper quantification of the substitutability of recycled materials is key when comparing the environmental performance of TCR and MR of plastic waste.

As the additional efforts to make the FCM applications from mechanical recyclates possible in the future could not yet be estimated in the current LCA study, these should be investigated in future research. Similarly, the environmental performance of TCR at industrial scale should be further investigated in detail, as several challenges before its mature implementation can be expected.

Nevertheless, this study confirms that TCR is a promising recycling technology for household plastic waste from an environmental perspective. In order to achieve the increased European recycling targets by 2030, TCR can be a complementary technology to MR, which is the main recycling method in operation today but can currently not be used for all applications, such as the closed-loop recycling of food packaging, except PET bottles. This study, therefore, also shows the need to further investigate the technical and legislative bottlenecks that hinder the use of mechanical recyclates for products with higher levels of safety requirements such as food packaging.

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.

Data availability

The data that has been used is confidential.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2022.08.006.

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