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Preface

The thesis is organised in two parts: the first part puts into context the findings of the PhD in an introductive review, and the second part consists of the papers listed below. These will be referred to in the text by their paper number, written in the Roman numerals **I-IV**.

I. **Eriksen, M.K.**, Damgaard, A., Boldrin, A., Astrup, T.F., 2018. Quality assessment and circularity potential of recovery systems for household plastic waste. *Journal of Industrial Ecology*. Vol 23(1), pp. 156-168. DOI: 10.1111/jiec.12822

II. **Eriksen, M.K.**, Pivnenko, K., Olsson, M.E., Astrup, T.F., 2018. Contamination in plastic recycling: Influence of metals on the quality of reprocessed plastic. *Waste Management*. Vol 79, pp. 595-606. DOI: 10.1016/j.wasman.2018.08.007

III. **Eriksen, M.K.**, Astrup, T.F., 2019. Characterisation of source-separated, rigid plastic waste and evaluation of recycling initiatives: Effects of product design and source-separation system. *Waste Management*. Vol 87, pp. 161-172. DOI: 10.1016/j.wasman.2019.02.006

IV. **Eriksen, M.K.**, Christiansen, J.D., Daugaard, A.E., Astrup, T.F., 2019. Closing the loop for PET, PE and PP waste from households: Influence of material properties and product design for plastic recycling. *Submitted* to Waste Management.

In this online version of the thesis, papers **I-IV** are not included but can be obtained from electronic article databases, e.g. via www.orbit.dtu.dk, or on request from DTU Environment, Technical University of Denmark, Miljoevej, Building 113, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk.

In addition, five conference proceedings and the following publications, not included in this thesis, were also concluded during this PhD study:

I. Pivnenko, K., **Eriksen, M.K.**, Martín-Fernández, J.A., Astrup, T.F., 2016. Recycling of plastic waste: Presence of phthalates in plastics from households and industry. Waste Management. Vol 54, pp. 44-52. DOI: 10.1016/j.wasman.2016.05.014

II. Starostina, V., Damgaard, A., **Eriksen, M.K.**, Christensen, T.H., 2018. Waste management in the Irkutsk region, Siberia, Russia: An environmental assessment of alternative development scenarios. Waste Management and Research. Vol 36(4), pp. 373-385. DOI: 10.1177/0734242X18757627

III. Astrup, T.F., Pivnenko, K., **Eriksen, M.K.**, Boldrin, A., 2018. Life Cycle Assessment of Waste Management: Are We Addressing the Key Challenges Ahead of Us? Journal of Industrial Ecology. Vol 22(5), pp. 1000-1004. DOI: 10.1111/jiec.12811

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During this three-year PhD project, several people and organisations have played a crucial role in making the various research activities possible and turning the PhD journey into a great experience.

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The outcome of this project would not have been possible without practical guidance, support and help from **The Danish Environmental Protection Agency**, **København Kommune**, **Amager Ressource Center**, **Aalborg University**, **Aage Vestergaard Larsen**, **Færch Plast** and **Sinh Hy Nguyen** as well as collaboration with the co-authors not previously mentioned: **Anders Egede Daugaard**, **Jesper deClaville Christiansen**, **Kostyantyn Pivnenko** and **Mikael Emil Olsson**.

I would like to extend special thanks to **Giorgia Faraca**, my office mate during the entire PhD, with whom I shared and discussed the ups and downs of life. Thank you very much for elevating my days in the office. In general, the happy, inclusive and constructive atmosphere of the entire **Solid Waste Research Group**, through lunch meetings, social events, conferences, coffee breaks, etc., was essential for my PhD stay. Thanks to all of you. I am moreover greatly thankful for all my **friends outside work**, who make my life be about so much more than waste and Excel.

Finally, massive thanks go to **my family**. It goes without saying – *you are my rock*.

Summary

To mitigate growing environmental threats such as climate change and resource depletion, the circular economy concept has gained momentum. Traditionally, materials have been lost through incineration or landfilling; however, in a circular economy, materials are recirculated into society, ideally to the same quality levels as they had originally, so that all demands within the material loop are fulfilled and use of virgin material can be avoided. However, recycling of materials, especially from heterogeneous waste streams such as household waste (HHW), often leads to recycled material of reduced quality, which only leads to a partial closing of the material loop – and thereby only partial circularity. This aspect is currently not part of the environmental assessment of materials and is therefore yet to be quantified.

In the transition towards a circular economy, plastic is highlighted as a focus material, as it is produced in large amounts from fossil resources. Consequently, the European Union (EU) has defined mass-based recycling targets for packaging plastic and placed specific focus on plastic from HHW. However, considerable physical and quality-related losses are related to the recycling of plastic from HHW, due to the potential presence of 1) non-plastic material, 2) plastic made from several polymer types (the most common in HHW plastic are polyethylene terephthalate (PET), polyethylene (PE) and polypropylene (PP)) and 3) many different product types (bottles, trays, etc.) with different purposes and design. Moreover, recycled plastic might be contaminated by potentially harmful chemicals, all of which may limit the quality of recycled plastic and thus the circularity. Consequently, in order to identify the most circular plastic recycling systems, thorough knowledge related to the physical and chemical states of waste and recycled plastic from HHW is necessary.

The aim of this thesis was to quantitatively integrate the quality aspect of waste and recycled plastic into circularity assessment of plastic recycling systems, focusing particularly on plastic from HHW. This was achieved by 1) theoretically relating quantity and quality of recycled materials to the circularity of recycling systems, 2) providing selected chemical, physical and mechanical characteristics of waste and recycled plastic, including a detailed composition of source-separated plastic waste, and 3) quantitatively evaluating the performance of current and potential future plastic recycling systems, and on this basis recommend the most circular options.

From a circularity perspective, the quality of recycled materials is closely related to the applicability, i.e. how well the recycled materials can be turned into different products with different quality levels. This quality, together with recycled quantities and knowledge on the distribution of different applications in the specific material market, can be used to quantity the circularity potential of recycling systems. Between 18% and 57% of European PET, PE and PP markets rely on chemically high-quality material for the production of food packaging. As such, it is crucial for the circularity of plastic recycling systems to have the ability to recycle plastic into material that can be used for food contact applications.

Current plastic recycling practices experience substantial material losses and physical contamination. It is therefore recommended to implement state-of-the-art plastic sorting systems with high recovery efficiencies and low contamination levels, as this is the best way to limit these issues. However, even such best-performing systems achieve a circularity potential of only about 0.40 (1 indicates full circularity), and thus current plastic recycling systems are far from able to close the plastic loop. Due to elevated concentrations of metals in recycled plastic, the circularity might be reduced even further in the future, if plastic products are recycled multiple times. Finally, the composition of rigid source-separated plastic reveals a high degree of heterogeneity in regards to the purpose (food or non-food packaging), type (bottles, trays, etc.) and design of waste products within each of the three dominant polymers, i.e. PET, PE and PP, representing more than 90% of the waste.

In order to mitigate these issues and improve the circularity, it is crucial to increase the quantities of recycled plastic while maintaining the chemical, physical and mechanical quality. "Design for recycling" initiatives, where for example all products are produced in a single polymer, are highly recommended, as they can lead to increases in the quantities of recycled plastic of up to 23%. Moreover, separate recycling of food packaging is recommended, as it allows for the production of recycled plastic suitable for food packaging applications and thereby maintains chemical quality. However, from a mechanical and physical perspective, the high degree of heterogeneity of PP food packaging makes it unsuitable for closed loop recycling into new packaging. Thus, in order to create the conditions necessary for closed-loop plastic recycling from HHW, where all quality aspects are maintained, regulation is needed to limit plastic packaging to the polymer types PET, PE and PP while standardising product types within especially PE and PP.

More research is necessary in order to identify the most appropriate combinations of product design, polymer selection and waste collection systems, achieving the highest possible increases in quantity and quality, and thereby circularity. This includes research into 1) detailed compositions of soft and residual plastic, 2) performance of the sorting process, depending on the type and design of waste products, and 3) how and to what extend mechanical and physical properties limit the applicability of recycled plastic.

Dansk sammenfatning

I lyset af stigende miljørelaterede trusler, som klimaforandringer og ressourceknaphed, har særligt konceptet om cirkulær økonomi vundet indpas. Traditionelt set er materialer gået tabt via forbrænding eller deponi, men i en cirkulær økonomi skal materialerne recirkuleres i samfundet, ideelt set til de samme kvalitetsniveauer som de oprindeligt havde, så behovet for materiale i hele materialekredsløbet kan dækkes, og brug af nyt materiale dermed undgås. Genanvendelse af materialer, særligt fra forskelligartede affaldsstrømme som husholdningsaffald (HHA), fører dog ofte til genanvendte materialer med reduceret kvaltiet, hvilket kun medfører delvist lukkede materialekredsløb og dermed kun delvis cirkularitet. Et aspekt der på nuværende tidspunkt ikke indgår som del af miljøvurderingen af materialer og som ikke tidligere er blevet kvantificeret.

Plastik er lavet af fossile ressourcer, bliver produceret i store mængder og er derfor et vigtigt materiale i overgangen til en cirkulær økonomi. Dette er grunden til, at den Europæiske Union (EU) har fastsat masse-relaterede genanvendelsesmål for emballageplastik og har særligt fokus på plastik fra husholdninger. Betragtelige tab af både kvantitet og kvalitet er dog relateret til genanvendelse af plastik fra HHA, da det kan indeholde 1) andre materialer end plastik, 2) mange forskellige typer plastik (typiske for HHA er polyethylenterephthalat (PET), polyethylen (PE) og polypropylen (PP)), og 3) mange forskellige typer produkter (flasker, bakker, osv.) med forskelligt formål og design. Derudover kan den genanvendte plastik være forurenet med potentielt skadelige stoffer, hvilket ydereligere kan nedsætte kvaliteten og dermed cirkulariteten. Omfattende viden omkring den fysiske og kemiske tilstand af både affalds- og genanvendt plastik er derfor nødvendig, før de mest cirkulære genanvendelsessystemer kan identificeres.

Formålet med denne afhandling var kvantitativt at integrere kvaliteten af affalds- og genanvendt plastik i cirkularitetsvurderingen af plastikgenanvendelsessystemer, med særligt fokus på plastik fra HHA. Dette blev gjort ved 1) teoretisk at relatere kvaliteten af genanvendte materialer til cirkulariteten af genanvendelsessystemer, 2) at tilvejebringe udvalgte kemiske, fysiske og mekaniske karakteristika af affalds- og genanvendt plastik, deriblandt en detaljeret sammensætning af kildesorteret plastikaffald og 3) kvantitativt at vurdere hvordan nuværende og potentielt fremtidig plastikgenanvendelsessystemer præsterer og på den baggrund anbefale de mest cirkulære systemer.

I et cirkulært perspektiv er kvaliteten af genanvendte materialer tæt relateret til anvendelsesmulighederne i forskellige produkter og kvalitetsniveauer. Kvaliteten kan, sammen med kvantiteten af genavendt materiale, samt viden om hvordan brugen af materiale fordeler sig på forskellige anvendelsesmuligheder på det gældende materialemarked, bruges til at kvantificere cirkularitetspotentialet af genanvendelsessystemer. Mellem 18 og 57% af det europæiske PET, PE og PP marked er afhængig af kemisk set højkvalitetsplastik til produktion af mademballage. Det er derfor vigtigt for cirkulariteten, at genanvendt plastik kan bruges til ny mademballage.

I eksisterende plastikgenanvendelsessystemer ses betragtelig fysisk forurening samt tab af plastik. Det anbefales at implementere state-of-the-art plastiksortering, der kan opnå høje genindvindingseffektiviteter og lav forureningsgrad, da det er det mest effektive redskab til at minimere disse problematikker. Men, selv disse bedste systemer opnår kun et cirkularitetspotentiale på omkring 0.40 (fuld cirkularitet kræver 1) og eksisterende plastikgenanvendelsessystemer er derfor stadig langt fra i stand til, at lukke plastikkredsløbet. I fremtiden kan cirkulariteten potentielt blive reduceret endnu mere, på grund af forhøjede koncentrationer af metaller i genanvendt plastik, særligt hvis plastikprodukter bliver genanvendt mange gange. Endelig viste det sig, at kildesorteret hårdt plastikaffald er meget forskelligartet, når det kommer til både formål (mademballage eller andre formål), type (flaske, bakke, osv.) og design af produkter i affaldet, inden for hver af de tre hyppigst forekomne polymertyper, PET, PE og PP, der udgjorde mere end 90%.

For at imødegå disse udfordringer, og øge cirkulariteten, er det afgørende at øge mængderne af genanvendt plastik samtidig med, at den kemiske, fysiske og mekaniske kvalitet bevares. Initiativer relateret til ”design til genanvendelse”, hvor fx alle produkter produceres udelukkende af en enkelt polymer, anbefales kraftigt, da det kan føre til øgede mængder af genanvendt plastik med op til 23%. Derudover anbefales det, at genanvende mademballage separat, da det muliggør produktionen af genanvendt plastik, der kan bruges i ny mademballage, og dermed bevare den kemiske kvalitet. PP mademballageaffald indeholder dog særligt mange forskellige produkttyper, hvilket, ud fra et fysisk og mekanisk synspunkt, gør blandet PP affald uegnet til genanvendelse til ny emballage. For at skabe rammerne for potentiel genanvendelse af plastik fra HHA i lukkede kredsløb anbefales det derfor, at implementere regulering der begrænser plastikemballage til polymertyperne PET, PE og PP alt imens produkttyperne, inden for særligt PE og PP, standardiseres.

Mere forskning er nødvendigt for, at kunne udvikle regulering, der bidrager mest muligt til øget kvantitet *og* kvalitet af genanvendt plastik – og dermed til øget cirkularitet. Det inkluderer forskning relateret til 1) detaljeret sammensætningen af blød- og restplastik, 2) hvordan sorteringsprocessen præsterer, og kan optimeres, for forskellige produkttyper og designs samt 3) hvordan, og i hvilken udstrækning, fysiske og mekaniske egenskaber begrænser anvendeligheden af genanvendt plastik.

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Abbreviations

EU	European Union
IPCC	Intergovernmental Panel on Climate Change
IV	Intrinsic viscosity
FTIR	Fourier-transformed infrared spectroscopy
HDPE	High-density polyethylene
HHW	Household waste
IW	Industrial waste
LCA	Life cycle assessment
LDPE	Low-density polyethylene
MFA	Material flow analysis
MFI	Melt flow index
MRF	Material recovery facility
NIR	Near infrared spectroscopy
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
PS	Polystyrene
rHHW	Recycled plastic from household waste
rIW	Recycled plastic from industrial waste

Symbols

$\alpha^{rec:disp}$	Substitutability of recycled material
c^{rec}	Circularity potential of a recycling system
MS	Market share
MS_{high}	Market share in which high quality material can be used
MS_{low}	Market share in which low quality material can be used
MS_{medium}	Market share in which medium quality material can be used
η^{rec}	Resource recovery efficiency of a recycling system
Q	Quality
Q^{disp}	Quality of displaced material (assumed high for virgin plastic)
Q^{rec}	Quality of recycled material
Φ^{disp}	Functionality of displaced material
Φ^{rec}	Functionality of recycled material

1 Introduction

1.1 Material flows in a circular economy

Increasing global consumption of materials and energy, and the related increase in waste generation (Krausmann et al., 2009; Hoornweg et al., 2013), have created a massive pressure on the environment, leading potentially to damaging and irreversible changes, including climate changes and depletion of resources. In October 2018, the Intergovernmental Panel on Climate Change (IPCC) stated that limiting global warming to 1.5 ºC would require rapid, far-reaching and unprecedented changes in all aspects of society (IPCC, 2018). As a way to achieve such changes while mitigating resource depletion, reducing overall environmental impacts and reducing the dependence of external resources, the concept of circular economy has gained momentum. An ideal circular economy vision includes closed material loops, where all waste materials are reintroduced into society, often through recycling, thereby eliminating the need for virgin resources. Especially the potential to avoid virgin material production through recycling has been shown previously to result in substantial environmental benefits, when compared to alternative waste management options (Laurent et al., 2014). As a result, the European Union (EU) has placed great political focus on recycling and as a continuation thereof defined mass-based recycling targets for various materials, as a way to transition towards a circular economy (EU, 2018; EC, 2015).

However, the quantities of waste "collected" or "sent to recycling" do not say much per se about whether, or to what extent, the recycling of materials actually avoids the production of virgin materials and thereby contributes to the closing of material loops – first, because substantial physical losses of material can be related to the recycling pathway, thus reducing the quantities of waste material actually converted into recycled material, and second, because the quality of the recycled materials that are produced is often reduced compared to that of alternative virgin materials (Rigamonti et al., 2018). This is especially the case when the waste originates from heterogonous and contaminated streams, such as household waste (HHW), as most materials in HHW consist of different grades and qualities. In an ideal circular economy vision, materials of different qualities need to be recycled into the same quality levels, in order to close the material loop and prevent the use of virgin material at all quality levels (Figure 1, left). However, for many materials in HHW, the recycled material is often of lower quality as a consequence of a more limited application range, when compared to the virgin material. As a result, the recycled material

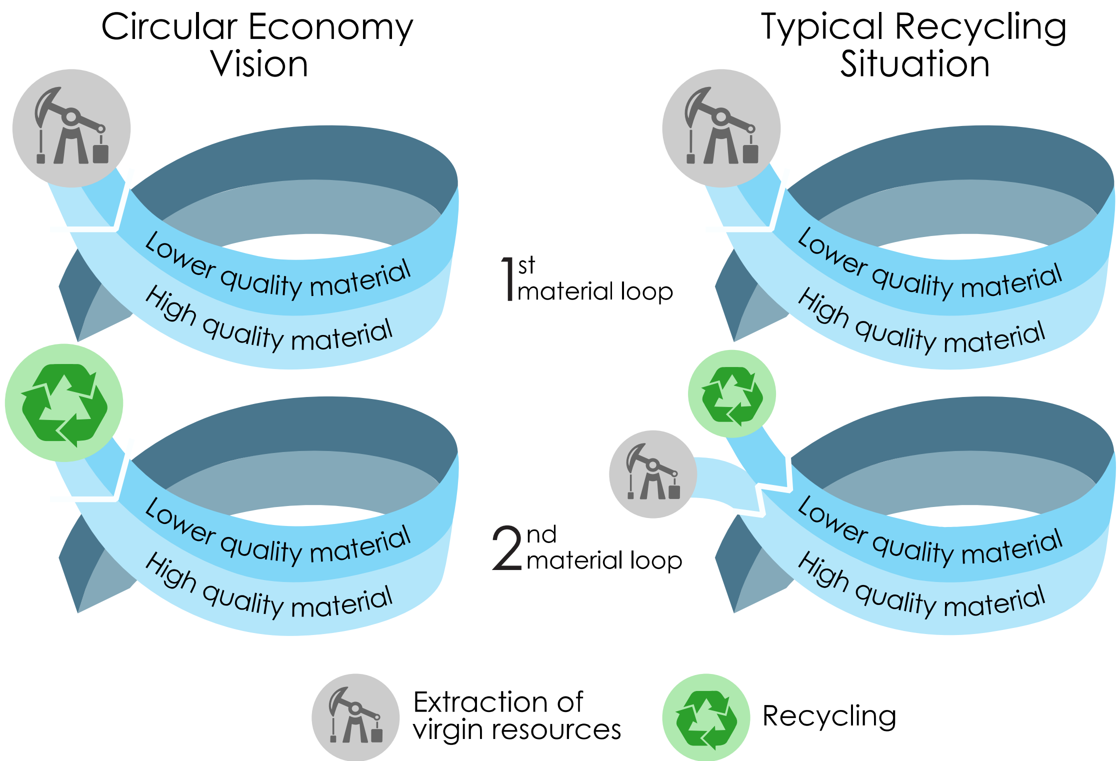

Figure 1 Conceptual illustration of material loops, assuming no physical loss of material, in an ideal circular economy vision and in a typical recycling situation, where material loops are only closed partially.

cannot substitute virgin material in the part of the material loop requiring high-quality material, the latter of which will therefore still rely on virgin material (Figure 1, right). Hence, this kind of recycling only closes the material loop partially. From a circularity perspective, potential applicability in the different parts of the loop is therefore linked closely to the quality of recycled materials.

Accordingly, in the transition towards circular economy, it is essential to understand the quality of recycled materials, how and to what degree quality is affected and how this quality can be linked quantitatively to the circularity of recycling systems, in order to identify the recycling systems with the highest potential to close material loops.

1.2 Plastic in a circular economy

In the transition towards a circular economy, plastic is an important material, due to several reasons. First, it is produced from fossil resources and accounts for 4-6% of global oil and gas consumption (PlasticsEurope and EPRO, 2017). Hence, to reduce the environmental impacts of plastic, especially when it comes to climate change and resource depletion, it is crucial to minimise emissions of fossil CO_2 from the incineration of plastic waste as well as minimise, or preferably eliminate, the production of virgin plastic – both theoretically achievable through recycling. Second, plastic is a very durable, inexpensive and versatile material, and as a result, the global consumption of plastic is expected to increase from 336 million tonnes in 2016 (PlasticsEurope and EPRO, 2017) to 1,124 million tonnes in 2050 (EMF, 2016). Thus, even though focus on plastic prevention has recently emerged in the public debate, and related reduction initiatives will be enforced in the EU (such as prohibition of selected single use plastic items [EC, 2018a]), it is still a crucial material for many purposes. For some, it even exhibits considerable environmental benefits when compared to alternative materials, due to its low weight and high food protection properties (Brandt and Pilz, 2011). A considerable demand for plastic is therefore assumed to prevail in the future.

On this basis, substantial focus has been placed on plastic and plastic recycling in the transition towards a circular economy. For example, the Ellen MacArthur Foundation has published three reports on a new plastic economy (EMF, 2019), and plastic is highlighted as a focus material in the EU's action plan towards a circular economy, which defines a recycling rate of 55% for plastic packaging waste in 2030 and places specific emphasis on plastic in HHW (EC, 2018b; EC, 2018c).

1.3 Challenges in plastic recycling from households

Recycling of plastic from HHW traditionally includes 1) source-separation of plastic in the household, 2) sorting of mixed plastic into individual polymer streams and 3) reprocessing of plastic waste into recycled plastic. However, several challenges are related to recycling of plastic from HHW, as it is a very heterogeneous and contaminated waste stream, due to:

- *Presence of non-plastic material*. Non-plastic material represents physical contamination and can originate from misplacements in the waste, i.e. non-plastic products erroneously sorted as plastic in the household, residues from the use phase, such as organic leftovers, or from products that are made of several materials, e.g. a plastic bag for chips, with a metal coating on the inside.
- *Presence of multiple polymers*. Plastic can be made of numerus polymers, the most common of which in HHW are polyethylene terephatalate (PET), polyethylene (PE), polypropylene (PP) and polystyrene (PS) (Edjabou et al., 2015; van Velzen et al., 2013). As their chemical structure is different,

these polymers cannot be recycled together, and they may therefore contaminate each other during recycling, if not effectively separated. Polymer cross-contamination of the plastic waste sent to reprocessing can originate from mis-sortings during the mechanical sorting process or from products that are made of several polymers, such as a PE bottle with a PET label.

- *Presence of multiple products*. Different product types (bottles, trays, etc.) with different purpose can have different properties, even when made of the same polymer (Cornell, 2016; Heinzl et al., 2015), which may change the original properties of the plastic, if different products are recycled together.
- *Chemical contamination*. Contamination of potentially harmful substances embedded in the matrix of the plastic can originate from physical contamination migrating into the plastic, recycling of plastic products containing different additives, degradation of additives into potentially harmful substances during reprocessing and the addition of new or extra additives during reprocessing.

Whereas the presence of non-plastic materials, multiple polymers and different products is related to the physical characteristics of the plastic waste, chemical contamination is related to the chemical characteristics of the recycled plastic. The common factor here is that they are all problematic for the recycling chain and can lead to a potential reduction in the quantity and quality of produced recycled plastic, and thereby a reduction of the circularity.

First, physical contamination can lead to material loss during sorting. The sorting of mixed plastic waste into polymers is done traditionally by using near infrared spectrometry (NIR), a technology that can identify the polymer of a waste plastic product by scanning the surface and then sorting it into the right polymer stream (Hopewell et al., 2009). However, due to plastic products of different shapes, with different designs and labels, the surfaces of which can be contaminated with residues from the use phase, substantial physical loss of plastic is often related to the sorting process (e.g. van Eygen et al., 2017). Moreover, no sorting process is ever 100% effective, and so reprocessing facilities define strict specifications, including physical contamination limits, for the plastic waste bales they receive. Consequently, if the levels of non-plastic products or products of unwanted polymers exceed these limits, the reprocessing facilities will not receive the plastic waste and convert it into recycled plastic, even though it was intended for recycling. Instead, the plastic waste will represent a loss from the recycling process. As such, the physical state of the plastic waste is crucial for the quantity of plastic being recycled.

Even if the level of physical contamination is below the limit values set by reprocessing facilities, the sorted polymer streams sent to reprocessing always include some contamination (Ærenlund, 2016; RRS, 2015; Luijsterburg and Goossens, 2014; Jansen et al., 2012; Enviros Consulting, 2009), which is known to have a negative effect on the quality of the recycled plastic (Ragaert et al., 2017; Villanueva and Eder, 2014). It is the common perception that recycled plastic from HHW exhibits lower mechanical and physical properties than virgin plastic (Rigamonti et al., 2018), due mainly to such physical contamination or to the heterogeneity of the waste. As an example, the processability of plastic, which is a measure of how it flows during reprocessing, is very important for the industry, as it determines what product types can be produced through conversion. Thus, when recycling different product types with different processability, the processability of the recycled plastic will most likely be different from that of the original products, influencing the applicability. Moreover, the degradation of polymer chains during recycling can lead to a change in processability (Ragaert et al., 2017), also influencing the applicability. In addition, Dahlbo et al. (2018) and Luijsterburg and Goossens (2014) reported that the tensile properties of recycled PE and especially PP were reduced when compared to a virgin control. Consequently, the physical state of the waste might be crucial for the physical and mechanical properties of the recycled plastic, and thereby potentially the applicability.

The applicability, however, might also be reduced from a chemical perspective. As an example, plastic used in food packaging needs to comply with strict European legislation regarding chemical composition and migration behaviour (EU, 2011) and therefore represents plastic with chemically high quality. Conversely, plastic used to make, for example, detergent bottles or flower pots does not have to comply with strict legislation, and does therefore represent plastic of chemically lower quality. If they are recycled together, the lower-quality plastic will contaminate the high-quality plastic, and the resulting recycled plastic will be of lower quality (see example in Figure 1, left). In fact, 95% of a PET waste stream, and 100% of a PE and PP waste stream, sent to recycling, has to consist of food packaging, in order to be recycled into recycled material that can be used once again to produce food packaging (EC, 2008; EFSA, 2011). Moreover, Camacho and Karlsson (2001) and Huber and Franz (1997) demonstrated that recycled HDPE and PP contained higher numbers of chemical compounds than virgin HDPE and PP. This was especially pronounced for

HDPE, where fragrance, flavour and cleaning agent compounds as well as phthalates were present in the recycled plastic but not in the virgin counterpart. This finding is in accordance with Pivnenko et al. (2016b), who found that the concentrations of selected phthalates in waste and recycled plastic from households were higher than in virgin plastic, thus highlighting that the use of recycled plastic in phthalate-sensitive applications needs to be monitored closely. Hence, even though the literature has focussed solely on organic compounds, it indicates that recycled plastic might be systematically, chemically contaminated. Consequently, both the physical state of the plastic waste and the chemical content of the recycled plastic might reduce the chemical quality.

In an attempt to limit losses of physical material and quality during recycling, different plastic recycling initiatives have emerged in the public debate, such as guidelines for the design of plastic products suitable for recycling (APR, 2018; FCP, 2018; Rethink Plastic, 2018), an extension to the Danish refund deposit system in 2020 to include bottles not only for mineral water and carbonated drinks, but also for juice and smoothies (MEFD, 2018), and the systematic revision of the requirements for placing plastic into the market in the EU, in order to make all plastic packaging reusable or easily recyclable in 2030 (EC, 2018d). While such initiatives are promising, their effects on recycling systems have not been assessed quantitatively, as this requires detailed plastic waste composition data. Even though several studies have reported compositions of plastic from HHW (e.g. Edjabou et al., 2015; Enviros Consulting, 2009), and some of them have even included the distribution of both polymers and product types (Brouwer et al., 2017; Petersen et al., 2015, van Velzen et al., 2013), information regarding polymer design and the separability of individual waste products has never been considered, and only few studies distinguished between food and non-food packaging. As many recycling initiatives deal with changes of these aspects, it is not possible, without detailed compositions, to assess quantitatively the potential effects of such initiatives.

In summary, both the physical and the chemical state of waste and recycled plastic are crucial for the applicability, which in turn again is crucial for the ability of plastic recycling systems to close individual polymer loops, i.e. the circularity. However, fundamental knowledge, systematically linking the physical and the chemical state of plastic waste and recycled plastic to the applicability, and therefore quality of recycled plastic, is still missing. As a result, no attempt has been made so far to quantify to what degree the quality aspect affects the circularity of current plastic recycling systems, or identify the most circular plastic recycling systems.

1.4 Aim and objectives

The overall aim of this PhD was to quantitatively integrate the quality aspect of waste and recycled plastic in the circularity assessment of plastic recycling systems, focusing on plastic waste from households. This was achieved through the following objectives:

1. Develop a comprehensive and transparent framework for evaluation of the quality of recycled plastic and the circularity of plastic recycling systems (Eriksen et al. **I)**.
2. Provide state-of-the-art characterisation of waste and recycled plastic from households, with a specific focus on parameters critical to quality, including selected chemical characteristics (Eriksen et al. **II**), detailed waste composition (Eriksen and Astrup **III**) and mechanical and physical properties (Eriksen et al. **IV**).
3. Provide recommendations on how to improve the overall circularity of recycling systems managing plastic waste from households (Eriksen et al. **I**, Eriksen and Astrup **III**, Eriksen et al. **IV**).

The remaining thesis is divided into seven chapters. **Chapter 2** provides an overview of the methodological approaches regarding the characterisation of recycled and waste plastic, as well as the assessment of current and potential future plastic recycling systems.

In Chapters 3, 4 and 5, the results of the PhD project are presented and the main outcomes of each chapter are summarised in the end of each chapter. In **Chapter 3**, the quantity and quality of recycled plastic are put into a theoretical context, linking them to the circularity potential of recycling systems. In **Chapter 4**, the performance of current plastic recycling systems is presented, including circularity potentials, physical and chemical contamination and composition of rigid source-separated plastic waste. In **Chapter 5**, potential ways to enhance the circularity potential of plastic recycling systems are assessed and discussed.

In **Chapter 6** and **Chapter 7**, the main conclusion and recommendations are presented, and the thesis ends with suggestions to further research in **Chapter 8**.

2 Methodology

In order to assess the performance and circularity of current and potential future plastic recycling systems, including information related to quality and recyclability, it is crucial to have detailed knowledge of the physical, chemical and mechanical characteristics of both waste and recycled plastic. Consequently, this work included sample preparation, analysis, waste characterisation and modelling activities, as presented in Table 1, and described in more details in the following sections.

Table 1 Overview of experimental campaigns included in the PhD, divided into focus on current recycling conditions and potential future recycling, and the associated papers. MFA: Material Flow Analysis, HHW: household waste.

Campaign	Current recycling	Future recycling
Sample preparation and analysis	Preparation of PET, PE, PP and PS samples from HHW. Analysis of the content of selected metals in these waste samples as well as externally collected samples of recycled and virgin plastic (Eriksen et al. **II**)	Preparation of reprocessed PET, PE and PP samples from mixed plastic and selected product groups from HHW. Analysis of selected physical and mechanical characteristics of these samples and externally collected recycled plastic samples from HHW (Eriksen et al. **IV**)
Waste character-risation	Composition of Danish rigid source-separated plastic from HHW (Eriksen and Astrup **III**)	
Modelling	MFA and circularity assessment of selected plastic recycling system configurations (Eriksen et al. **I**)	MFA of potential future management of rigid source-separated plastic, representing different recycling initiatives (Eriksen and Astrup **III**)

2.1 Characterisation of waste and recycled plastic

The chemical, physical and mechanical characteristics of waste and recycled plastic are essential for the performance of the entire recycling system. Consequently, the characterisation of waste and recycled plastic included analysis of chemical, physical and mechanical characteristics as well as characterisation of source-separated rigid plastic waste.

2.1.1 Chemical characteristics

To assess differences in the chemical nature of plastic from different polymers, as well as plastic appearing in different steps of the plastic chain, samples of PET, PE, PP and PS were analysed in Eriksen et al. **II**. These samples originated from HHW plastic, reprocessed plastic from households, reprocessed plastic from industry or virgin plastic, as presented in Table 2. While the samples of reprocessed and virgin plastic were collected from external sources in the form of pellets, granules or flakes, the waste samples were prepared as described in the following.

The waste samples were prepared from initial samples of 700 kg source-separated plastic waste and 930 kg residual waste from households. The rigid plastic in both samples was sorted into the polymers PET, PE, PP and PS and homogenised via coarse shredding. To reduce the sample size, while ensuring the representativeness of the sample, 1D splitting was performed on all samples, until a desired sample size of 1 kg was achieved. 1D splitting is a mass-reduction method that aims at reducing errors during sampling, ensuring the representativeness of the final sample (Dahlén and Lagerkvist, 2008; Lagerkvist et al., 2011), which was performed on a flat pile of coarsely shredded plastic, divided into equally sized increments, discarding every second increment until reaching the desired sample size. Finally, the samples were shredded finely into flakes < 10 mm.

Table 2 Sample overview, including polymer type, origin and number of samples. More details are provided in Eriksen et al. **II**.

Sample origin	**Number of samples**				
	PET	**PE**	**PP**	**PS**	**Total**
Waste plastic from households	10	10	10	6	**36**
Reprocessed plastic from households	2	5	3	0	**10**
Reprocessed plastic from industry	2	11	3	3	**19**
Virgin plastic	1	8	4	4	**17**
Total	**15**	**34**	**20**	**13**	**82**

Metals can be added intentionally to plastic as additives, be present as residues from catalysts in plastic production or sorb to the plastic from contamination during use or waste management (Hahladakis et al., 2018), and they are therefore expected to be present in plastic and thus also recycled plastic and plastic waste. Moreover, metals are in most cases expected to persist in plastic during recycling (Hansen, 2013). As a result, all samples were analysed for the total content of Al, As, Cd, Co, Cr, Cu, Fe, Hg, Li, Mn, Ni, Pb, Sb, Ti and Zn.

Differences in metal concentrations in the different sample groups, presented in Table 2, were analysed statistically, using compositional data analysis. All details related to the sample preparation procedure, the metal analysis and the statistical analysis are provided in Eriksen et al. **II**.

2.1.2 Mechanical and physical properties

In addition to the chemical characteristics of the recycled plastic, mechanical and physical properties, in particular the processability of the recycled plastic, are essential for the quality – and therefore the possibility of closed-loop recycling. Consequently, in Eriksen et al. **IV** samples of reprocessed plastic were prepared from different waste plastic configurations, representing a significant share of rigid Danish source-separated plastic waste, as well as collected from external sources. A sample overview is provided in Table 3, including information related to the waste product types and the polymer from which the samples were made of. As indicated in the table, the samples made from mixed products represented reprocessed plastic produced from current recycling systems, whereas the remaining samples, made of specific waste product types, were included in order to assess the potential quality of the reprocessed plastic,

Table 3 Overview of reprocessed samples regarding waste product types and polymer from which they were made. Additional details are provided in Eriksen et al. **IV**.

Waste product types	**PET**	**PE**	**PP**	**Comment**
Fruit and vegetable trays	✓		✓	
Meat trays			✓	Sealing foil was removed manually in one sample, to simulate a mono-polymer product design
Mixed food trays	✓		✓	
Dairy tubs			✓	
Beverage bottles	✓			Plastic lids and labels were removed manually in one sample, to simulate a mono-polymer product design
Soap related bottles		✓		
Mixed products	✓	✓	✓	Represents the current recycling situation

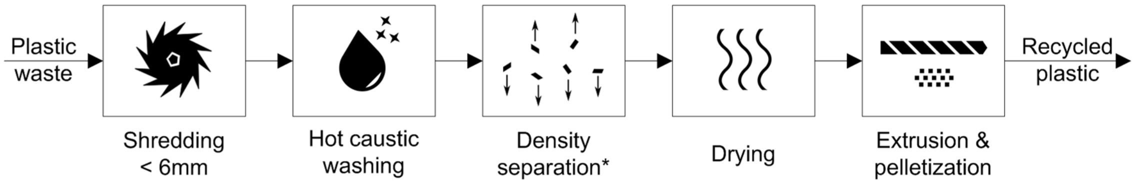

Figure 2 Sample preparation procedure (Eriksen et al. **IV**). * was only performed on selected samples.

and thereby closed-loop recycling potential, if these product types were separately managed and recycled. A large share of meat trays, beverage bottles and soap related bottles consists of multiple polymers, due to sealing foils, lids or labels being made of a different polymer than the main product component. Thus, an additional sample of meat trays, beverage bottles and soap-related bottles was prepared, in which plastic-sealing foil or lids and labels were removed manually, to simulate the effect of changing the design to a mono-polymer product.

The waste products were representatively collected as part of the waste characterisation campaign, presented in the following section. Waste for all samples was shredded, caustically hot-washed (representing an industrial washing process (APR, 2019)), dried at 60ºC for 24 hours, extruded and cut into pellets, as presented in Figure 2. In cases where the shredded waste partitioned into two fractions during the washing process (a floating and a sinking fraction), the undesired fraction was removed.

All samples were analysed for selected mechanical and physical properties:

- *Degradation pathway*, which was analysed using thermo-gravimetric analysis.
- *Mechanical properties*, specifically tensile strength and strain as well as impact strength.
- *Processability*, measured as melt mass-flow index (MFI).

Whereas the degradation pathway of the samples provides information related to chemical heterogeneity, mechanical properties provide information on how much stress and impact the plastic can take. The MFI is key when evaluating the potential for closed-loop recycling of PE and PP, as plastic with different MFIs are suitable for different processing methods, which in turn are suitable for the production of specific product types, as presented in Table 4. As it shows, not all plastic is suitable for production of packaging products. As an example, plastic with MFI between 5-50 are suitable for thick-walled applica-

Table 4 Overview of melt flow index (MFI), suitable processing methods and product types, based on practical experience from the recycling industry (Scholdan, 2018).

MFI	Processing method	Suitable product types
0-1	Extrusion	Plastic bags, low trays produced by thermoforming
0.3-5	Blow moulding	Bottles and flasks
5-50	Injection moulding	Thick-walled products, mostly non-packaging, such as outdoor fences
50→	Injection moulding	Thin-walled products, suitable for tubs, buckets, trays, etc.

tions, and thereby not most packaging. The MFI is often related to the length of the polymers in the plastic, i.e. the shorter the polymers, the higher the MFI, and thus, if the polymers in the plastic degrade during recycling, it will often lead to an increase in MFI, especially for PP (Kozlowski, 2015).

2.1.3 Plastic waste characterisation

The composition of plastic waste sent to recycling is crucial for the performance of plastic recycling systems, in regards to both the quantity and the quality of any recycled plastic potentially produced. As an example, if a waste plastic product consists of multiple polymers (such as a PET bottle with a PE lid), mechanical removal of the unwanted polymer during recycling (the PE lid) will often result in losing it to incineration (FCP, 2018). Similarly, black plastic cannot be recognised by traditional NIR sorting equipment (Turner, 2018) and is therefore directed to incineration. Moreover, it is crucial to know the distribution of food and non-food packaging in the waste, in order to estimate its distribution in the sorted waste streams sent to recycling, which is essential for the potential of recycling into food-grade recycled plastic. Hence, in Eriksen and Astrup **III**, rigid source-separated plastic waste was collected and characterised in details, as described in the following.

An initial sample of 3,700 kg source-separated plastic waste was collected from the municipality of Copenhagen in 2017 over four separate days, to ensure that waste from different areas, as well as from both multi- and single-family houses, was included. From the initial sample, 550 kg non-plastic items and 490 kg misplaced large objects, which should have been delivered to recycling stations, were removed. Moreover, the soft plastic fraction was removed, as it accounted for only 10% of the plastic items targeted by the source-separation scheme. The sample of rigid plastic waste was sorted into PET, PE, PP,

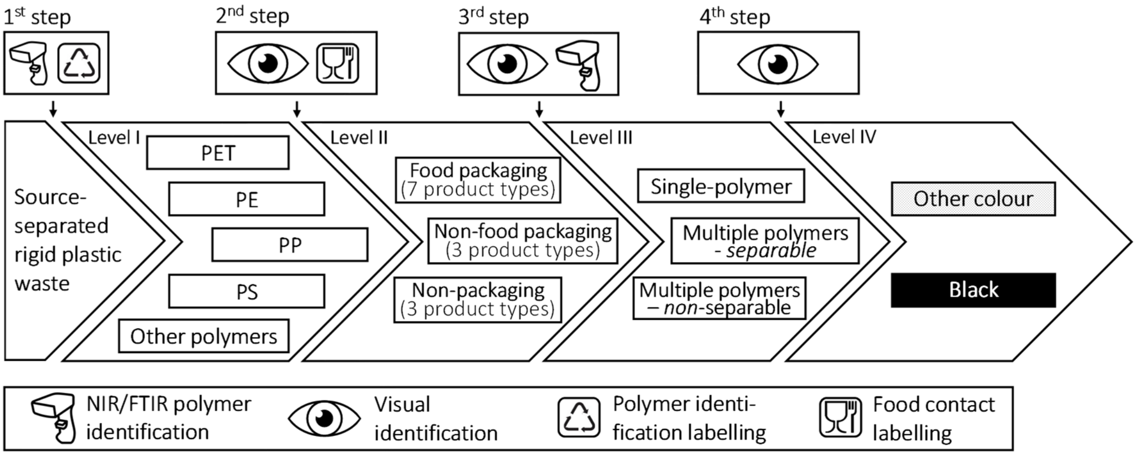

Figure 3 Illustration of the characterisation procedure and the four characterisation levels. More details are provided in Eriksen and Astrup **III**.

PS and residues, using a NIR scanner at a pilot-scale plastic sorting facility, and during this sorting process four subsamples of each fraction were collected for further characterisation. This resulted in a final sample of 460 kg source-separated rigid plastic waste, which was characterised in details, following the procedure presented in Figure 3, according to the four levels:

1. Characterisation into the five polymer types presented in Figure 3, by using an industrial-scale NIR scanner supplemented by identification via either the polymer identification label or a handheld FTIR.
2. Characterisation into product types, the main categories being food packaging, non-food packaging and non-packaging. This was done based on visual identification of either the label describing the previous content or the EU ‘cup fork’ symbol ensuring that the plastic was approved for food contact.
3. Characterisation of the waste plastic products according to their design. This included characterisation into single polymer products, products made of multiple polymers that can be mechanically separated and products made of multiple polymers that cannot be mechanically separated. This was done based on a combination of visual and FTIR identification.
4. Characterisation into black products and products of other colours, based on visual identification.

The characterisation scheme provided 390 theoretical combinations, albeit waste was only detected in 97 of these. More details about the characterisation is given in Eriksen and Astrup **III**.

2.2 Assessment of plastic recycling systems

In order to evaluate the performance of plastic recycling systems, an assessment of current recycling systems (Eriksen et al. **I**) as well as of potential future recycling systems (Eriksen and Astrup **III**) were included in the PhD. Both assessments were based on material flow analyses (MFAs), with the system boundaries presented in Figure 4, and included an evaluation of the quantity and quality of either the sorted plastic waste recovered from the material recovery facility (MRF) (Eriksen et al. **I**), or of the recycled plastic (Eriksen and Astrup **III**). The circularity potential was moreover assessed for current recycling systems, based on the method developed and presented in Chapter 3. Both assessments are described in more details in the following sections.

2.2.1 Current plastic recycling practices

When evaluating the performance of current plastic recycling systems, it is relevant to assess different configurations, as the configuration of the recycling system is expected to vary significantly. For example, the overall sorting scheme can go from including bottles only, to including all plastic generated in households or not having a sorting scheme at all. Similarly, source-separation and collection efficiencies can vary, depending on the maturity of the sorting scheme, the willingness of citizens to participate, collection schemes, etc. Moreover, the effectiveness of the sorting process, where the mixed plastic waste is sorted into individual polymer streams, can vary considerably, depending on the specific configuration of the facility, the technical performance of the equipment and the composition and degree of contamination of the plas-

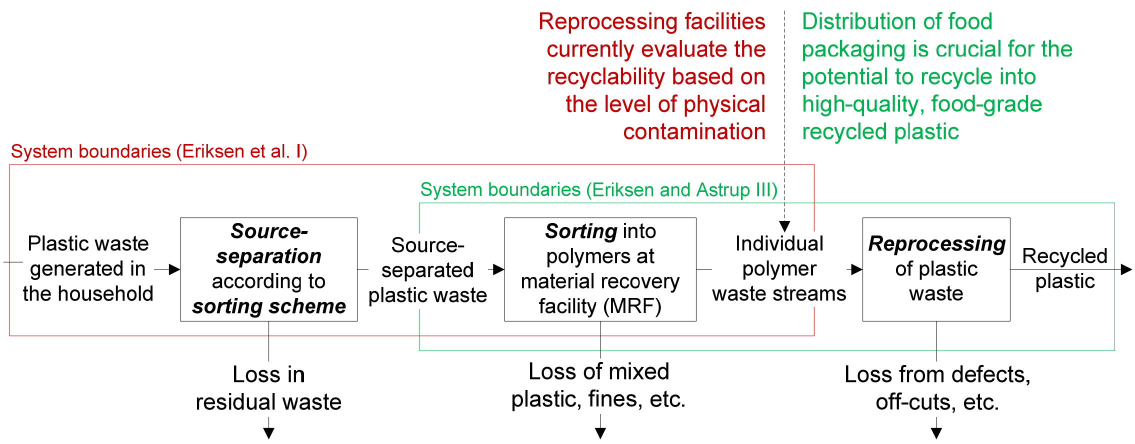

Figure 4 Simple flow diagram illustrating the most important processes in plastic recycling, potential losses and where the composition and contamination of the waste become crucial for the final applicability. System boundaries for the assessment of current system (red) and potential future systems (green) are illustrated.

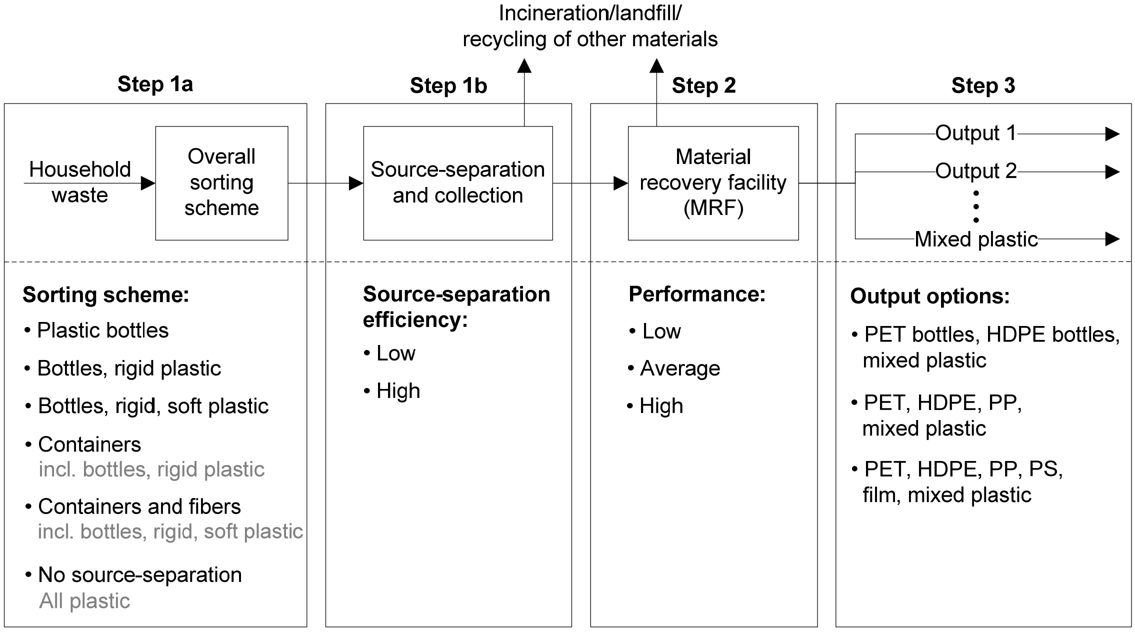

Figure 5 Overview of scenario configuration steps and options within each step (Eriksen et al. **I**). Performance of the MRF is equal to recovery efficiency of targeted material to the intended output, i.e. high recovery efficiencies means high performance.

tic waste. Consequently, key options in each step of the recycling chain included in the system boundaries were identified, as presented in Figure 5.

While the options for overall sorting scheme (step 1a) and MRF outputs (step 3) represented fundamental differences in approach, the options for source-separation (step 1b) and MRF (step 2), were modelled using efficiencies, ranging from the lowest to the highest efficiencies reported in literature for existing recycling systems, in order to capture the large variety in performance of current systems. As an example, it was assumed that the lowest performing MRF were able to route 10-40 % of the targeted plastic fractions to the intended outputs, depending on polymer and product type, whereas this was assumed to be as high as 70-95% for the high performing MRF (for details see Eriksen et al. **I**). Combinations of all options in the different steps resulted in 84 realistic scenarios.

Input into the system was similar in all scenarios and included all waste generated in the household, including non-plastic waste, as this allowed for the modelling of non-plastic impurities through the system. The plastic was assumed to represent 14% of the total amount of waste generated, with an estimated average European composition, as presented in Table 5, including knowledge related to polymers, as this allows for the additional modelling of polymer cross-contamination during plastic sorting.

Table 5 Composition of the plastic part of the generated HHW divided into plastic material fractions and polymer types [%]. The composition was estimated based on Rigamonti et al. (2014), Edjabou et al. (2015) and Petersen et al. (2015). (Eriksen et al. **I**).

Plastic fractions	PET	HDPE	LDPE	PP	PS	Others	Total
Bottles	23	7	0	0	0	0	30
Soft packaging	0	0	30	0	0	10	40
Hard packaging	4	3	0	7	1	5	20
Other plastic items	0	0	0	0	0	10	10
Total	26[a]	10	30	7	1	25	100

[a)] 23 and 4 are rounded and thus sum to 26

Based on the MFAs of all scenarios, it was possible to evaluate the recovered quantities of plastic sent to recycling in each scenario. Additionally, the potential quality grades into which the outputs from the MRF could potentially be recycled, were identified, based on the physical – but excluding influence of chemical – contamination of the sorted plastic waste. This was done by assessing if the presence of physical contamination, at the point where the waste leaves the MRF but before it enters reprocessing (see Figure 4), complies with contamination levels for different quality grades, developed by reprocessing facilities. Where contamination levels needed to be below 4-7%, depending on contaminant and polymer type, for the plastic waste to be recycled into high-quality recycled plastic, 7.5-18% contamination was acceptable in plastic waste recycled into low-quality plastic.

The circularity potential of all defined scenarios was assessed based on the method developed and presented in Chapter 3. The starting point of the circularity assessment was the quality of the different waste plastic streams recovered from the MRF in each scenario.

All details related to defining the scenarios, performing the MFA and assessing the quality and circularity potential are presented in Eriksen et al. **I**.

2.2.2 Potential future recycling initiatives

Several recycling initiatives have recently been developed and proposed in the public debate, each with the aim of increasing the quantity and/or quality of recycled plastic from households. Such initiatives mainly involve changes to product designs or changes in the configuration of the source-separation scheme (e.g. Rethink Plastic, 2018; MEFD, 2018).

To capture the potential effect from selected recycling initiatives, the following scenarios were defined and assessed in Eriksen and Astrup **III**:

- *Current recycling*: Represents the current recycling pathway for source-separated plastic from households.
- *Design for recycling (1)*: All plastic packaging is produced as single-polymer products in colours other than black.
- *Separate collection of food packaging (2)*: Two bins are introduced in the household, one for PET and PP food packaging and the other for any remaining plastic waste.
- *Alignment of polymers and products (3)*: All food packaging is produced in PET and PP, whereas all non-food packaging is produced in PE, while only packaging plastic is targeted in the source-separation scheme.
- *Combination 1+2*: Design for recycling is combined with separate collection of food packaging.
- *Combination 1+3:* Design for recycling is combined with alignment of polymer and products.

To quantitatively evaluate the effect of these recycling initiatives, an MFA was performed for all scenarios. The system boundaries were given as illustrated in green in Figure 4, and the quality of the recycled plastic waste was evaluated based on the distribution of food and non-food packaging in the sorted plastic waste streams sent to recycling; high-quality recycled plastic was potentially suitable for food packaging, low-quality recycled plastic was not. The flow diagrams for each scenario are presented in Figure 6.

As an assessment of these initiatives requires detailed knowledge regarding polymers, product types and product design, the input into the scenarios was similar to the composition found from the waste characterisation campaign presented in Chapter 4.2, or estimated based on this composition, in those scenarios where the initiatives involved changing the product design and thereby the waste composition.

All details related to the definition of scenarios and the performance of the MFA are presented in Eriksen and Astrup **III**.

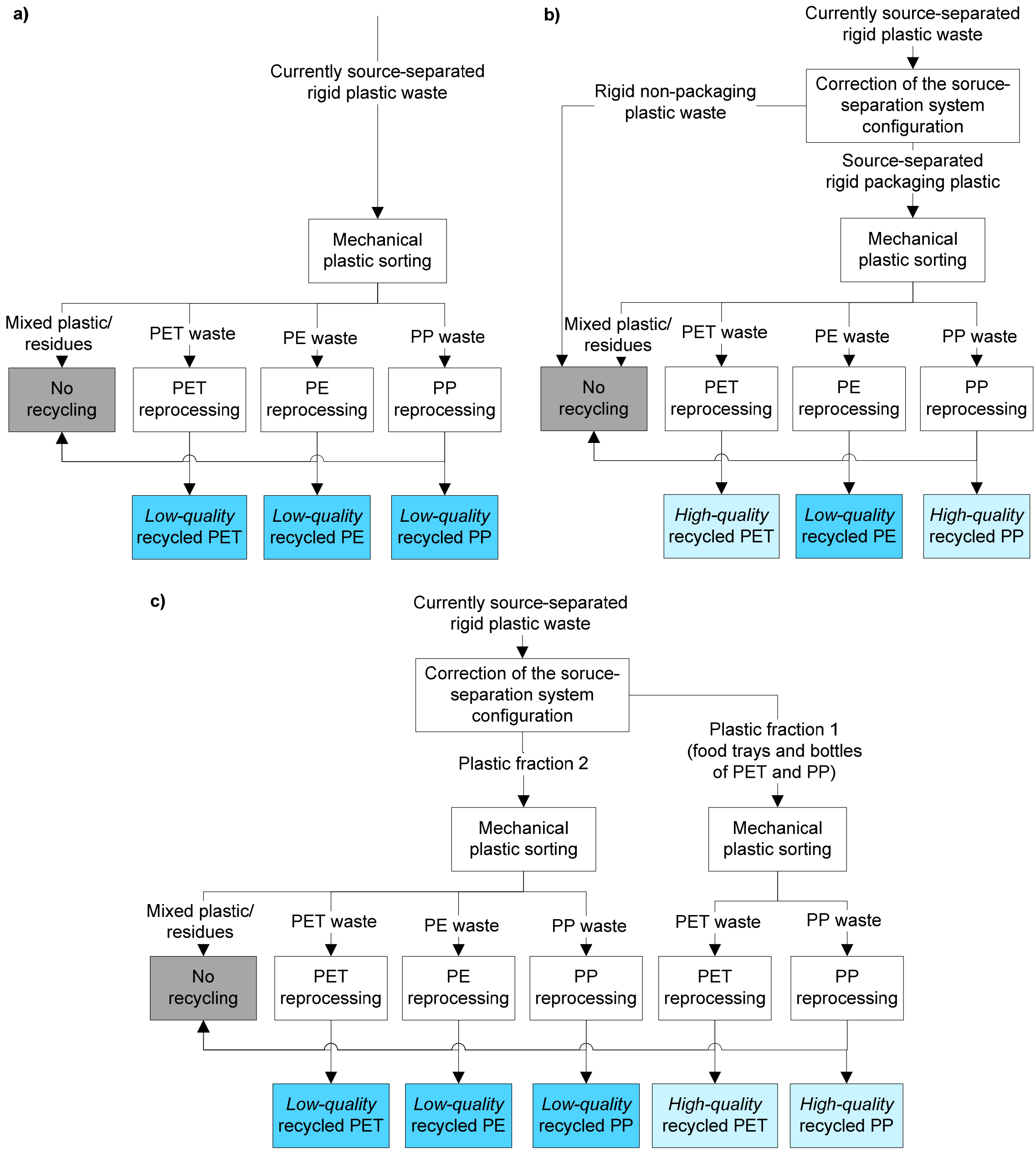

Figure 6 Flow diagram representing the recycling process associated with a) *current recycling* and *design for recycling* scenarios, b) *alignment of polymers and products* and *combination 1+3* scenarios and c) *separate collection of food packaging* and *combination 1+2* scenarios. More details in Eriksen and Astrup **III**.

3 Linking quantity, quality and circularity

In order to evaluate to what degree recycling systems contribute to the circular economy, we need to be able to evaluate the potential circularity of recycling systems on a large societal scale, i.e. recycling systems' ability to contribute to the closing of material loops. A prerequisite for a circular economy vision with closed material loops is steady-state material flows, where the amount of waste generated is equal to the demand of new materials, so that the entire material demand can ideally by supplied by recycled rather than virgin material. In such conditions, the circularity is intrinsically related to the potential to avoid virgin material production. Thus, if one part of the market cannot be fulfilled by recycled material, due to reduced quality, this part of the market ultimately has to rely on virgin resources, as presented in the introduction in Figure 1. Even though steady-state material flows for most materials are currently far from the reality (Fellner et al., 2017), such conditions represent the end goal of a circular economy, and it is important to be able to evaluate if we are moving towards this vision.

This chapter introduces the developed circularity potential and illustrates its applicability on plastic recycling systems. The framework was applied on recycling configurations representing current recycling systems and the results are presented in the following Chapter 4.

3.1 Substitution potential

When quantifying environmental impacts through life cycle assessment (LCA), quantity and quality reductions have traditionally been accounted for when calculating the substitution potential, i.e. the total mass of virgin material avoided, as a result of the recycled material produced from a waste management system (Vadenbo et al., 2016). The substitution potential has in practise been quantified as the product of the physical material losses throughout a recycling system and the quality related losses of the recycled material. Most studies have included material losses through efficiency coefficients (e.g. source-separation, sorting and reprocessing efficiencies) (e.g. Rigamonti et al., 2009; Bassi et al., 2017). As opposed, the quality loss of the recycled material has often been quantified relative to the quality of the virgin material assumed substituted (Rigamonti et al., 2009; van der Harst et al., 2016), which Vadenbo et al. (2016) has designated the substitutability, $\alpha^{rec:disp}$, quantified as in Eq. 1.

$$\alpha^{rec:disp} = \frac{\Phi^{rec}}{\Phi^{disp}} \qquad \textbf{Eq.1}$$

where Φ^{rec} and Φ^{disp} represents the functionality of the *rec*ycled material and the virgin material assumed *disp*laced, respectively.

The overall aim of the substitutability is to quantify mass-based losses during the production stage, triggered by a reduction in the quality of the recycled plastic. As an example, if the use of recycled plastic in a specific application, such as flower boxes, results in the production of a higher amount of defective flower boxes, due to the reduced quality of the recycled plastic, more recycled plastic is required to produce the same amount of marketable flower boxes, compared to if virgin plastic was used. Thus, the substitutability, $\alpha^{rec:disp}$, is intrinsically related to the physical losses of material happening during the production phase of a specific application (1-$\alpha^{rec:disp}$), as a result of reduced quality. This relation is illustrated in Figure 7, top, where application x could be flower boxes.

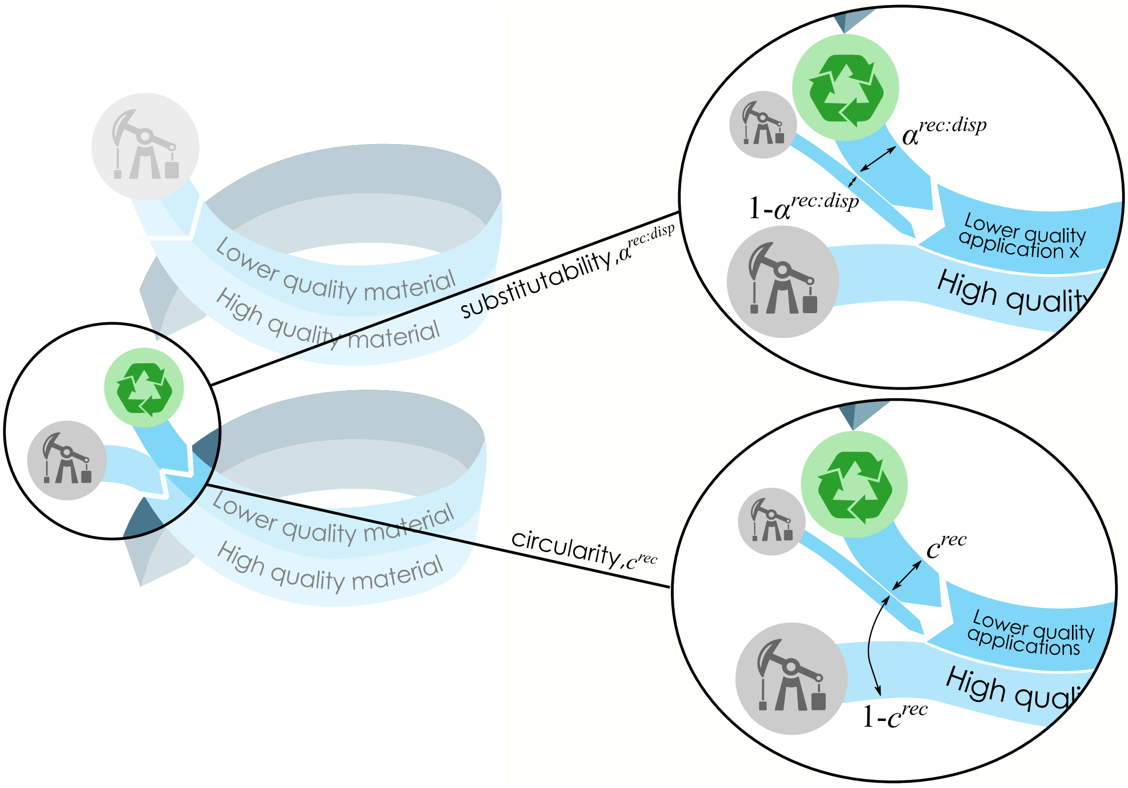

Figure 7 Illustration of the difference between substitutability, $\alpha^{rec:disp}$, traditionally used in a life cycle assessment, and long-term circularity potential, c^{rec}.

However, as the substitutability focuses on the substitution of virgin material in a specific application, applications might exist where a reduction in the quality of the recycled material does not influence the required functionality; for example, very low material requirements are related to items such as plastic poles with reflectors placed along a highway. Consequently, the recycled material might be able to substitute 100% virgin plastic in such specific applications, and the reduction in material quality will thereby not be reflected in the substitutability. In such cases, the substitutability does therefore not account for the system-related consequences that the quality of the recycled material might cause, i.e. if the recycled material can only be used in applications requiring low-quality material, applications requiring high-quality material will still have to rely on virgin material. As such, the substitutability, $\alpha^{rec:disp}$, of a material may be 1, although recycling of this material may not lead to closing of material loops from a quality aspect. Thus, LCA do currently not account for the circularity aspect, which is a crucial limitation in the transition towards a circular economy.

3.2 Circularity potential

To mitigate this limitation, the circularity potential of recycling systems, c^{rec}, is introduced in Eriksen et al. **I**. Just as the traditional substitution potential, it includes both physical material losses as well as a reduction in functionality. However, where the physical material losses, designated η^{rec} (Vadenbo et al., 2016), is similar to that used for the substitution potential, the functionality is based on overall system requirements, instead of requirements for use in a specific application. As such, the functionality of the recycled material is a measure of its ability to fulfil the requirements within a specific material market. Thus, the circularity potential, c^{rec}, is intrinsically related to the part of the market in which the recycled material cannot fulfil the demand (1-c^{rec}), collectively representing the entire material market (see Figure 7, bottom). The circularity potential, c^{rec}, is quantified as follows in Eq. 2:

$$c^{rec} = \eta^{rec} \cdot \frac{\Phi^{rec}}{\Phi^{disp}} = \eta^{rec} \cdot \frac{MS(Q^{rec})}{MS(Q^{disp})} \qquad \textbf{Eq.2}$$

$$\rightarrow \begin{cases} MS_{high} \text{ for } Q = \text{high}, \\ MS_{medium} \text{ for } Q = \text{medium}, \\ MS_{low} \text{ for } Q = \text{low} \end{cases}$$

where the functionality, Φ, of the *rec*ycled or *disp*laced material is defined as the market share, *MS*, in which the material can fulfil the demands, depending on its quality, *Q*. The market share, *MS*, for high-quality materials is always 100%, as high quality material can potentially fulfil the requirements for use in all applications in the market. However, if the quality of the recycled material is reduced (*Q* = medium or *Q* = low), it can only fulfil the requirements for applications in certain parts of the market, and thus it can only contribute to partial closing of the material loop. The quality of virgin material is always assumed to be high, as its production or synthesis can be controlled to accommodate requirements in all applications relevant for a specific material.

Recycling can ensure that the materials already introduced into society are recirculated in the best ways possible, thereby achieving a high circularity potential; however, it cannot in itself, even when 100% effective, ensure closed material loops, if the material flows are not in a steady state. On this basis, it is important to point out that growing material consumption, traditionally coupled with increasing economic growth (Krausmann et al., 2009), is a barrier for circular material flows, even if solely having optimal recycling systems with a circularity potential close to 1. Consequently, in the transition towards a circular economy, identifying recycling systems with high circularity potentials needs to be supplemented by measures ensuring steady-state material flows, in order to ultimately reach absolute material circularity.

3.3 The European plastic market

To apply the circularity potential assessment on plastic recycling systems, it is crucial to have detailed knowledge related to applications where plastic is used, the quality requirements for the plastic used in these applications and their market share.

Thus, based on legislation and plastic statistics, eight application groups were found relevant for plastic, as listed in Table 6. These were divided into three quality levels, based on legal requirements related to the chemical characteristics of plastic used in the applications. The strictest legislation was related to plastic used in food packaging; consequently, it is assumed to be of chemically high quality, and as such, the production of food packaging represents the high-quality part of the material loop in Figure 1.

Moreover, Table 6 presents the European market shares for the three quality levels in the PET, HDPE, LDPE, PP and PS markets. The table shows that food packaging, and thereby the high-quality part of the individual polymer loops, represents at least 15% for PS and up to 57% for PET. Thus, if recycling systems managing high-quality plastic are not able to produce new high-quality

Table 6 Share of the European PET, HDPE, LDPE, PP and PS markets [-] that currently rely on high-, medium- or low-quality material. More details are provided in Eriksen et al. **I**.

Applications	**Suitable qualities**	**European polymer markets**				
		PET	**HDPE**	**LDPE**	**PP**	**PS**
Food packaging	High	0.57	0.27	0.54	0.18	0.15
Toys, electrical and electronics, pharmaceuticals	High, Medium	0.00	0.03	0.04	0.06	0.11
Building and construction, non-food packaging, automotive, others.	High, Medium, Low	0.43	0.70	0.42	0.76	0.74
All applications	-	**1.00**	**1.00**	**1.00**	**1.00**	**1.00**

recycled plastic that can be used in food packaging applications, significant shares of the different polymer loops still need to rely on virgin resources. This in turn limits the circularity potential of such recycling systems, a tendency especially pronounced for PET and LDPE.

Chapter 3 – ***Linking quantity, quality and circularity***

- Focusing solely on increasing the quantities of recycled plastic is not sufficient in the transition towards a circular economy, as the degree to which material loops can be closed, i.e. the circularity potential, is a function of the quantity *and* quality, i.e. applicability, of recycled material.
- Identifying recycling systems with high circularity potential needs to be complemented by measures ensuring steady-state material flows, in order to potentially reach absolute material circularity.
- Between 15% and 57% of the European PET, HDPE, LDPE, PP and PS markets is used to produce food packaging made of chemically high-quality plastic, representing the parts of the polymer loops that cannot be closed, if plastic waste is recycled into low-quality material.

The circularity assessment methodology is applied on current recycling systems and the results are presented in section 4.1.

4 Current situation: Recycling of mixed plastic waste

Regarding plastic from HHW, current recycling pathways can have many different configurations. However, besides countries having a separate deposit system for PET beverage bottles, a common feature of European recycling systems is that the plastic collected for recycling is collected as mixed plastic waste, which subsequently needs to be sorted into polymers and then recycled. Consequently, when assessing the state of the current plastic recycling situation, it is crucial to identify material losses, contamination and waste compositions, related to collection, sorting and reprocessing of mixed plastic waste, all of which are presented in this chapter.

4.1 Material loss, physical contamination and circularity potential

The performance and circularity potential of current recycling systems was assessed in Eriksen et al. **I**, and it was found that the degrees of physical losses and contamination related to the recycling pathway, depend strongly on the configuration of the specific recycling system. This is illustrated in Figure 8, which presents the recovered quantities of plastic waste sent to recycling, i.e. η^{rec}, for selected recycling configurations, with varying overall sorting schemes and MRF efficiencies. The quality of the recovered plastic waste is indicated in the figure, based on the level of physical contamination. The height of the bars (■+■+□) represents the total share of plastic waste generated in the household that can potentially be recovered from plastic sorting and sent to reprocessing. However, due to physical contamination of the plastic waste, exceeding the requirements for recycling, only the high- and lower-quality parts of the bars (■+■) represent the share of plastic waste suitable for actual recycling; the high-quality part (■) contains physical contamination below 4-7% whereas the lower quality part (■) has higher contamination levels. Additionally, the overall circularity potential of each scenario is presented in Figure 8 (■).

It is clear from the figure that the more plastic fractions that are included in the overall sorting scheme, the more material is recovered from the sorting process (■+■+□). As an example, if, instead of source-separation in the household, plastic is mechanically recovered from residual waste, up to around 100% of the generated plastic mass can be recovered. However, for most systems, large

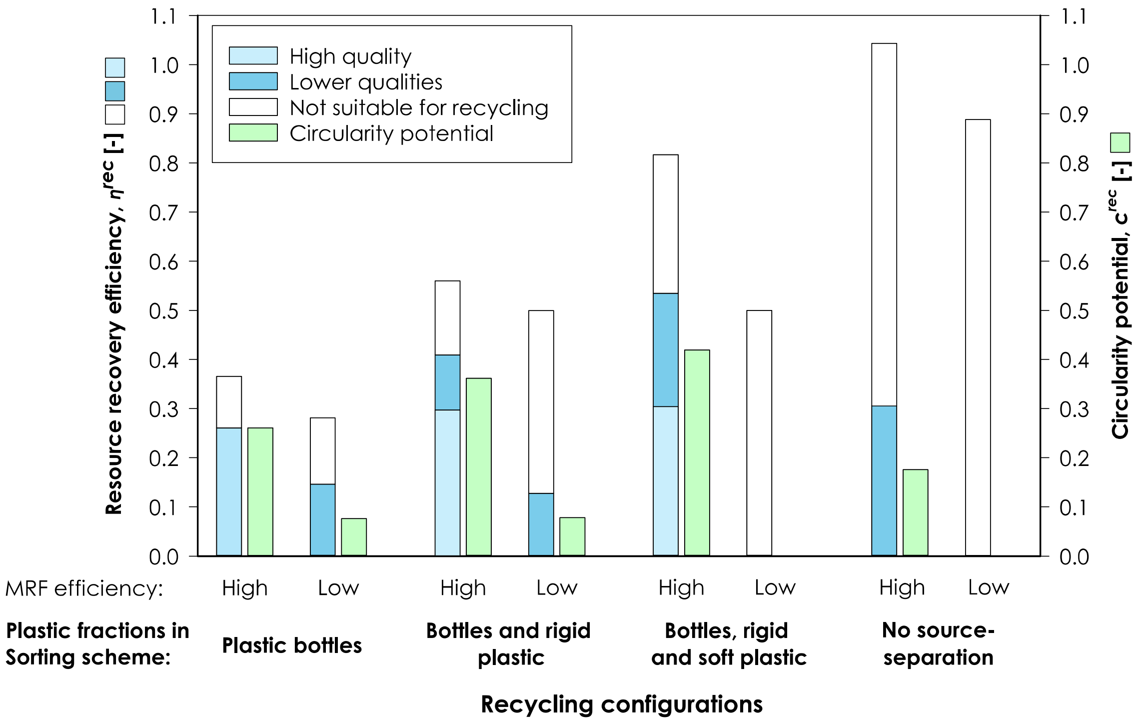

Figure 8 Resource recovery efficiency, η^{rec}, divided into qualities, and circularity potential, c^{rec}, of selected scenarios. All scenarios have high source-separation efficiencies + all possible MRF outputs and are ordered according to plastic fractions targeted in the sorting scheme and material recovery facility (MRF) efficiency. By adding the high and lower quality streams (■+■), the recovered *and* recyclable shares of the generated plastic waste are obtained. Results for all scenarios are presented in Eriksen et al. **I**.

shares of the recovered plastic are unsuitable for recycling (☐), which is especially pronounced for the recycling systems with no source-separation system. Thus, even though these systems perform the best in terms of the total recovered quantities, they represent some of the worst configurations, when it comes to recovering of *recyclable* fractions. The best performing recycling configuration can at best recover a recyclable fraction representing 53% of the generated plastic waste, which includes source-separation of bottles, rigid and soft plastic, aligned with high source-separation and MRF efficiencies.

Regarding the quality of the recyclable part, at best low-quality material could potentially be produced if the recycling system has a low-performing MRF, and consequently only configurations with high-performing MRFs could potentially produce high-quality plastic. As the potential to produce recycled material of high-quality is crucial for the overall circularity of the recycling system, MRF efficiency is moreover essential for the circularity potential. As a result, high MRF efficiency is a key element when designing recycling systems with the potential to produce large quantities of recyclable waste plastic streams, with low levels of physical contamination, thereby having the potential for recycling into high-quality recycled material.

In addition to the MRF efficiency, the number of plastic fractions targeted in the sorting scheme is important for the quantities of recyclable plastic waste recovered for recycling – and therefore the circularity potential. Even though the quantity of recovered high-quality material is almost identical for sorting schemes targeting bottles and rigid plastic, compared to those also targeting soft plastic, the quantities of soft plastic represent a substantial part of the total generated plastic waste and therefore result in an elevated circularity potential, even though it can at best be recycled into lower-quality recycled plastic. As such, recycling systems with high source-separation and MRF efficiencies, targeting both bottles, rigid and soft plastic in the sorting scheme, represent the kind of currently existing recycling system, performing the best in terms of both the recovery of recyclable quantities and overall circularity potential.

However, even for such recycling systems, only a circularity potential of 0.42 was identified, indicating that with current recycling systems we are still far from a circular economy vision for plastic, as this would require a circularity potential close to 1. It is moreover important to note that the potential production of high-quality recycled plastic, identified in these results, requires that plastic waste can be decontaminated to meet the chemical requirements for food contact materials (Rieckmann et al., 2011). Something that is currently only possible for PET beverage bottle waste with a maximum of 5% non-food items, often separately collected through refund deposit systems (Simon, 2010). Thus, the composition of plastic waste and the chemical nature of recycled and waste plastic are presented and discussed in the rest of the chapter.

4.2 Composition of source-separated plastic waste

The composition of the plastic waste sent to sorting and subsequent recycling is crucial in understanding more accurately what happens through the recycling system, what the current limitations are and how these might be addressed.

Consequently, in Eriksen and Astrup **III**, a detailed characterisation of source-separated rigid plastic from HHW in Copenhagen was performed. The composition, regarding polymers and product types, is presented in Table 7.

The table reveals several issues related to the recyclability of plastic waste. First, both food packaging and non-food packaging waste products were present within the three dominant polymers, namely PET, PE and PP, making up

Table 7 Composition of the rigid part of the source-separated plastic waste from the Municipality of Copenhagen, December 2017, divided into polymers and product types. The numbers are rounded. (Eriksen and Astrup **III**).

Product type / polymer	**PET**	**PE**	**PP**	**Others**	**Total**
Food packaging	***25±1***	***5***	***20±1***	***2***	***52±1***
Bottles for beverages	6	2	0	0	8
Bottles for food	2	2	0	0	5
Trays and tubs for fruit and vegetable	2	0	1±1	0±1	3±1
Trays and tubs for dairy	0	0	4±1	0	5±1
Trays and tubs for meat	4	0	6±1	0	10±1
Trays and tubs for other/unidentified food	9±1	0	9±1	1	18±1
Other rigid food packaging	1	1	0	0	2
Non-food packaging	***6***	***19±1***	***3***	***1±1***	***30±1***
Bottles for soap-related purposes	4±1	14	2	0	19±1
Bottles with hazardous labelling	0	4±1	0	0	4±1
Other rigid non-food packaging	2	1	1	1	6±1
Non-packaging	***0***	***2±1***	***11±1***	***5***	***18±1***
Toys	0	0	0	0	1
Flower pots	0	0	2	0	3
Others	0	2	9±1	4±1	15±1
Sum	***31±1***	***27±1***	***34±2***	***8±1***	***100***

more than 90% of the waste. Thus, even though the majority of food packaging was made of either PET or PP, and the majority of non-food packaging was made of PE, current sorting of the waste into these three polymers will still result in waste streams including both chemically high-quality food packaging and other products made of chemically lower-quality plastic. Consequently, rigid PET, PE and PP waste, sorted from source-separated mixed plastic, can currently only be recycled into lower-quality recycled plastic.

While homogeneity regarding product types was relatively high for the PE waste, as more than 80% consisted of bottles, it was low for the PET waste, consisting of a mixture of bottles and trays, and especially the PP waste, due to the presence of a variety of trays and tubs, some non-food bottles and a considerable share of non-packaging items.

Regarding product design and separability, 43% of the plastic consisted of multiple polymers. In one-third of these cases, the polymers were not mechanically separable, thereby inevitably leading to some degree of polymer cross-contamination. Most of the multi-polymer products that were mechanically separable were either PET or PE bottles, whereas the majority of multi-polymer products that were not separable were meat trays with sealing foils, primarily of PP.

Finally, 10-11% of the plastic waste was black and thereby not recoverable using current mechanical sorting technologies.

4.3 Chemical contamination

A distinctive property of plastic from HHW is that it has been through a use phase in which contamination, in the form of various chemical compounds, may sorb to the plastic waste as a result of direct use or misuse (Pivnenko and Astrup, 2016). Moreover, additives will most likely be added during reprocessing, to counteract the reduction of material properties during recycling (Gu et al., 2017). Consequently, the concentration of selected chemicals might be elevated in recycled plastic, potentially limiting the applicability and thereby quality when compared to virgin plastic.

In Eriksen et al. **II**, the total concentration of selected metals relevant for use in plastic, expected to persist during recycling, was analysed in samples of waste, reprocessed and virgin plastic. The results revealed that concentrations of some metals were polymer-specific, as seen, for example, in the case of Sb. As illustrated in Figure 9, its concentration was significantly higher in PET samples than in samples of any of the other polymers, most likely due to residues left in the PET plastic from the use of Sb_2O_3 as a catalyst during virgin PET plastic production (EU, 2008).

The analysis also showed that metal concentration was significantly higher in samples originating from households, including both waste and reprocessed samples, when compared to virgin plastic samples, and that the overall highest concentrations of especially Cu, Pb, and Zn were found in samples of reprocessed plastic from HHW (see Figure 9). As the concentrations of these metals were not similarly high in the waste samples, this may indicate that they originated from the addition of metal containing additives during reprocessing, rather than from contamination. Thus, by using the concentration of metals as an indicator of inorganic chemical contamination, the results suggest that accumulation might happen during recycling of mixed plastic from HHW.

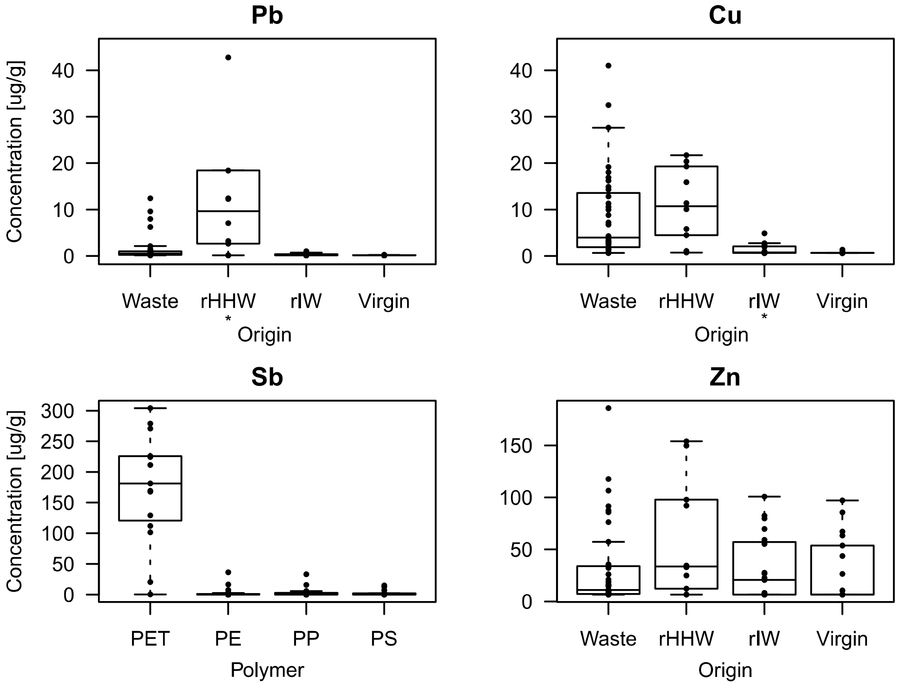

Figure 9 Concentration [µg/g] of Pb, Cu and Zn divided into origins and Sb divided into polymers. Lowest whisker = minimum concentration, bottom of box = 25th percentile, bold line in box = median, top of box = 75th percentile, top whisker = 95th percentile.
* Samples with values higher than twice the largest whisker are not illustrated. rHHW: recycled plastic from household waste, rIW: recycled plastic from industrial waste. Results for all analysed metals are presented in Eriksen et al. **II**.

These elevated metal concentrations did in general not limit the applicability of the reprocessed plastic from HHW directly, as the concentrations did not exceed the existing limit values for use. However, the elevated concentrations for several metals were higher than what has normally been reported in food packaging. Thus, the applicability might be indirectly limited. Moreover, as recycling rates for plastic are expected to increase in the future, concentrations of metals in recycled plastic from mixed HHW might increase further, as each plastic product might have to be recycled multiple times– a phenomenon already quantified for paper (Pivnenko et al., 2016a). Consequently, in such case the applicability might be directly limited in the future.

Chapter 4 – ***Current situation: Recycling of mixed plastic***

- Current recycling of mixed plastic from households can at best close around 40% of the plastic loop, due to large physical losses during source-separation and sorting, as well as physical contamination in the sorting process.
- The performance of the mechanical sorting process is by far the most important parameter when it comes to increasing potential quantities recovered for recycling, as well as eliminating physical impurities, allowing recycling into high-quality recycled plastic (when only accounting for physical and *not* chemical contamination). As such, state-of-the-art plastic sorting is key in reaching the highest possible circularity.
- Danish mixed source-separated plastic waste exhibits a high degree of heterogeneity, as it contains several polymers (mainly PET, PE and PP), many different product types (around 50% chemically *high-quality* food packaging) and complex product designs (around 45% is made of more than one polymer, and 10-11% is black).
- The content of potentially harmful metals was elevated in recycled plastic from households when compared to virgin plastic. This was not to an extent whereby it would directly limit the applicability, but it might become a limitation for the applicability in the future, when recycling rates increase and plastic has to be recycled multiple times.

5 Future potentials: Increasing circularity

As presented in the previous two chapters, the circularity of recycling systems depends on the quantity and quality of recycled plastic. As such, when suggesting solutions aiming at increasing the circularity, by mitigating limitations related to the current recycling practises, it is important to focus on increasing the quantity while preserving the quality, as well as understanding how to balance the two. Moreover, it is essential to understand that both the chemical, physical and mechanical quality of the plastic need to be preserved, in order to potentially achieve closed-loop recycling, where plastic waste can be recycled into the same type of applications with the same quality level.

Consequently, this chapter presents and discusses selected recycling initiatives aiming at 1) increasing recycled quantities (Eriksen and Astrup **III**), 2) preserving the chemical quality (Eriksen and Astrup **III**), and 3) preserving the mechanical and physical quality (Eriksen et al. **IV**).

5.1 Increasing recycled quantities

Several organisations have promoted design guidelines that the plastic industries can follow, in order to design and produce products suitable for recycling (APR, 2018; FCP, 2018; Rethink Plastic, 2018) and thereby improve recycling of plastic from HHW in general. Guidelines, amongst others, dictating that products should be produced in a single polymer (FCP, 2018) and that the production of products that cannot be detected by current sorting techniques, such as black plastic, should be avoided (APR, 2018). Consequently, the effect of changing plastic product designs, so that all products would be made from a single polymer only, in colours other than black, was assessed in Eriksen and Astrup **III**. Figure 10 presents the quantities and quality of recycled plastic produced from current recycling practises as well as selected recycling initiatives. From the figure, it becomes clear that “design for recycling” initiatives can lead to a potential increase in recycled plastic by up to 23%, compared to current recycling. The results moreover show that design for recycling is the only initiative, out of the assessed initiatives, that has the potential to increase the quantities of recycled plastic considerably, when involving state-of-the-art plastic sorting. However, as the results also show, design for recycling cannot by itself ensure that the high chemical quality of food packaging is preserved, as such initiatives will not change the distribution of food and non-food packaging in the three dominant polymers, PET, PE and PP.

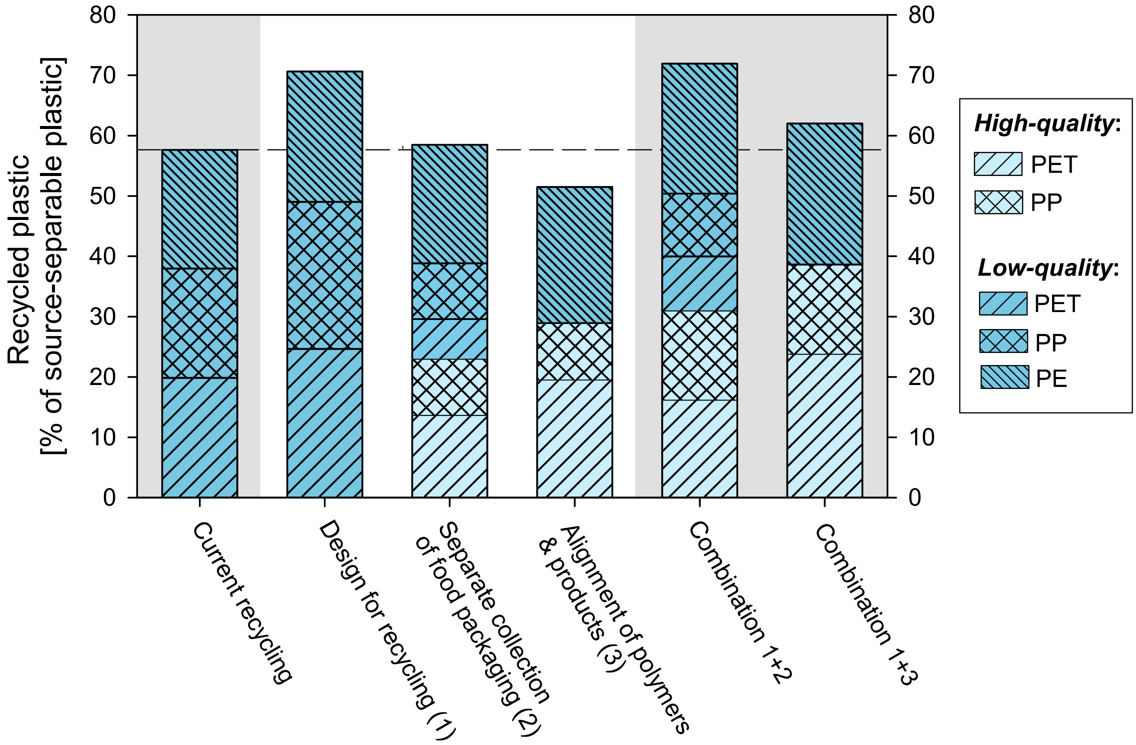

Figure 10 Percentage of source-separated plastic waste recycled into new recycled plastic, for current recycling, as well as selected recycling initiatives where product design or source-separation has been optimised. All scenarios are modelled with high performing plastic sorting. The quality of the recycled plastic is indicated, and a dashed line indicates the level of the current recycling. All results are presented in Eriksen and Astrup **III**.

5.2 Preserving chemical quality

Thus, additional initiatives, besides "design for recycling" initiatives, introducing separate recycling of chemically high-quality food packaging, are necessary, in order to preserve the chemical quality.

One way to achieve this would be to collect food packaging separately by extending the source-separation system from one to two plastic bins in the household, the first containing food packaging and the second containing any remaining plastic. Such initiative could potentially lead to between 20% and 30% of the source-separated plastic waste being recycled into high-quality recycled plastic, as illustrated in Figure 10. However, a drawback is that an extension of the source-separation system will result in an extra collection route, which might be economically expensive, as collection is known to represent a large share of the cost for waste management systems (Martinez-Sanchez et al., 2015).

Another option is to regulatory align or harmonise product types and polymers. This means that the use of specific polymers in specific products is controlled in the sense that food packaging would only be produced in PET or PP, whereas all non-food packaging would be produced in PE, which follows the pattern already in the waste. In this way, already existing systems with NIR technology could potentially be used to mechanically separate food packaging from non-food packaging, as long as only packaging plastic is targeted in the source-separation scheme. Such initiatives would make it possible to recycle 30-40% of the source-separated plastic waste into high-quality plastic, potentially recycling all of the recovered plastic in a closed loop (food packaging to high-quality and non-food packaging to low-quality). This initiative has the potential of producing the largest quantities of high-quality recycled plastic, and consequently a high circularity potential is expected to be achievable. However, the total quantities of recycled plastic will decrease compared to current recycling, as only packaging plastic is targeted in the source-separation scheme.

Common to both options is that the quantities of total as well as high-quality recycled plastic can be increased considerably, if combined with "design for recycling" initiatives.

5.3 Preserving physical and mechanical quality

In addition to establishing the necessary conditions for preserving the chemical quality of plastic, it is equally important to make sure that the physical and mechanical properties of the recycled plastic allow for actual closed-loop recycling into similar products. Selected physical and mechanical properties of recycled PET, PE and PP were therefore assessed in Eriksen et al. **IV**, with the aim of identifying the conditions necessary to facilitate closed-loop recycling from a physical and mechanical perspective. Since the different polymers were found to exhibit different behaviours during recycling, the following sections are divided into polymers with similar behaviours.

5.3.1 PET

For PET plastic, the single most important property is intrinsic viscosity (IV), which is a measure of the length of the polymers in the plastic (Kozlowski, 2015); essentially, the longer the polymers, and thereby the higher the IV, the better the quality (Kozlowski, 2015). In order to use PET for bottle production, the plastic needs to be of high quality, whereas for tray or film production, the quality can be lower (Lynggaard, 2018). As the polymers in PET plastic have been shown to degrade during recycling, thereby reducing its quality, it could be expected that recycled PET would be unsuitable for production into new bottles. However, when recycling PET waste into new PET suitable for food contact, the recycled plastic needs to be decontaminated. Due to the chemical nature of PET polymers, degradation of the polymers is a reversible process, and thus the IV and thereby quality of the recycled PET can be rebuilt during decontamination, to meet the requirements for both bottles and tray production (Kozlowski, 2015; Rieckmann et al., 2011). Hence, PET plastic is well-suited for recycling multiple times, and the current heterogeneity of PET food packaging waste is therefore not expected to be a limiting factor for material quality, if the system is designed correctly. Thus, by limiting the use of PET plastic to food-packaging applications, it is theoretically possible to preserve the chemical, physical and mechanical properties of mixed PET from HHW.

5.3.2 PE and PP

For PE and PP plastic, the processability and mechanical properties of the recycled plastic are crucial for the applicability; consequently, these properties were analysed for recycled PE and PP waste produced from specific waste product types and mixed products in Eriksen et al. **IV**.

Based on the processability, it was possible to evaluate for what kinds of products the recycled plastic samples were suitable for, as the processability is a determining factor in how plastic can be processed (see Table 4). Based on these results, the potential recycling pathways for selected recycled PE and PP samples, as well as their MFI, are presented in Figure 11.

The figure shows that separate recycling of PE soap bottles, as well as recycling of mixed rigid PE waste, produce recycled PE suitable for bottle production, allowing for closed-loop recycling of soap bottles. As different PE products are known to exhibit different processability (Kozlowski, 2015), this was somewhat unexpected for mixed PE waste. However, it might be because the mixed PE waste consisted of around 80% bottles, assumed to be almost exclusively made of high-density PE (HDPE), therefore representing a relatively homogeneous waste stream in terms of product type. Moreover, the mechanical properties of these recycled PE samples prepared from Danish source-separated HHW were particularly high compared to virgin HDPE, specifically regarding tensile strength, which further supports the possibility of closed-loop recycling of bottles. However, samples of recycled PE from mixed HHW, collected from external sources, exhibited significantly lower tensile strength. As low-density PE (LDPE) normally exhibits much lower tensile strength than HDPE (Crompton, 2012), the reduced mechanical properties in the externally

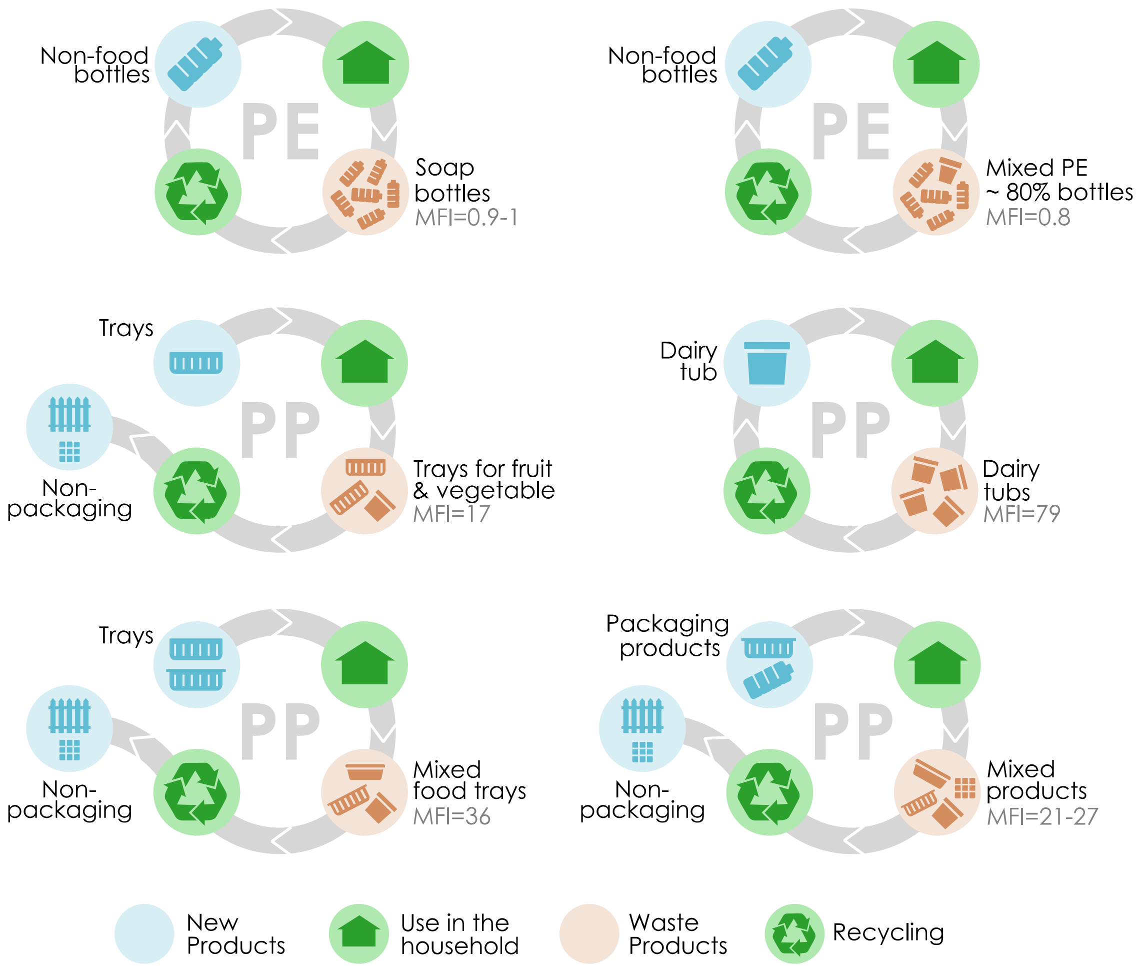

Figure 11 Illustration of potential recycling pathways for different waste products, based on the melt flow index (MFI) of the recycled plastic they can produce, determining the processing method and suitable products, as presented in Table 4. All results are presented in Eriksen et al. **IV**.

collected samples could be due to the recycling of rigid HDPE products together with flexible LDPE films, which highlights the importance of a homogeneous PE waste input, for closed-loop recycling of PE waste.

Regarding PP, Figure 11 shows that only closed-loop recycling of dairy tubs was physically possible, if recycled separately. Thus, as opposed to PE, it was not found physically possible to recycle mixed PP waste or even specific tray-related product types into new thin-walled packaging applications. For example, even though trays for fruit and vegetables are a relatively specific product group, they are still produced in several different ways, some most likely by extrusion and thermoforming, and some by injection moulding, making the recycled plastic entirely unsuitable for tray production. Thus, even if PP food-packaging is managed and recycled separately, which is necessary to potentially maintain the chemical quality, it is not enough to ensure closed-loop recycling into thin-walled packaging applications. Thus, the large degree of heterogeneity of the PP waste, when it comes to product type, is a major limitation for closed-loop recycling.

Moreover, the PP waste degraded during reprocessing, changing the processability of the recycled plastic and potentially reducing the mechanical properties, representing another limitation of multiple times closed-loop recycling of PP.

To solve these issues, initiatives maintaining the high degree of homogeneity of PE waste, increasing the homogeneity of PP waste and limiting the influence of PP degradation are necessary.

Solutions could include technical initiatives such as additional tracer-based sorting, chemical recycling and additional usage of additives. In chemical recycling, polymers in the plastic are broken down into their individual constituents, from where new plastic, suitable for food-contact purposes, can be produced (Ragaert et al., 2017). If such a solution were indeed applied, the heterogeneity of the plastic waste would not be an issue, and the influence of degradation during recycling would be eliminated. However, chemically recycled polymers are currently more expensive than the alternative virgin polymers (Ragaert et al., 2017) and might have elevated environmental impacts when compared to traditional recycling (Faraca et al., 2019).

Usage of specially designed additives in the production of PP plastic can stabilise the processability and thereby limit the effects of degradation during recycling (Kozlowski, 2015). As opposed to chemical recycling, such a solution would have to be complemented by additional initiatives, dealing with the heterogeneity of PP from HHW.

Advanced sorting of the plastic waste prior to reprocessing, i.e. separating different grades of the same polymer from one another, could represent such a solution. Currently, plastic sorting techniques are based on intrinsic properties such as polymer type or colour. However, substantial research has been done to develop new sorting methods whereby a tracer is added during production, e.g. based on florescence, which, with the right equipment, can then be recognised during sorting (Brunner et al., 2015). Even though several limitations are still related to such technologies, they nevertheless represent a unique opportunity for mechanically separating different product types of the same polymer from each other, thereby reducing the heterogeneity of the individual plastic waste streams sent to recycling.

Regulatory initiatives represent another kind of solution, aiming at reducing the heterogeneity of plastic waste. In addition to controlling, that food packaging should be made of PET and PP and non-food packaging of PE, necessary to ensure the chemical quality, the type of products being produced could be regulated as well. For example, the production of PP trays could be regulated so that all trays are produced using the same processing method, requiring PP plastic with a processability within a limited interval. Or, it could be to regulatory avoid the use of PP in short-lived packaging applications, thereby redirecting the use of PP to other sectors, where it can be used in products having a longer lifetime. Moreover, regulatory initiatives could be used to implement deposit systems for selected products and thereby control the composition of product types entering the system, as is currently done for PET beverage bottles in many European countries (Simon, 2010). Initiatives that could moreover be used to promote the reuse of products entering the system, rather than recycling, potentially reducing environmental impacts (Simon, 2010), while postposing the influence of polymer degradation over time.

Chapter 5 - ***Future potentials: Increasing circularity***

- Designing plastic products for recycling could increase the quantities of recycled plastic by up to 23%.
- Crucial to maintaining chemical quality is separate recycling of food packaging, which could be achieved, for example, by having two bins for plastic in the household (one for food packaging, one for the rest) or regulatory aligning polymers and product types (all food packaging in the same polymer(s) and all non-food packaging in a different polymer).
- To preserve physical and mechanical properties, the different recycling behaviours of PET, PE and PP need to be accounted for systematically.
  - Potential quality reductions of recycled PET from food packaging can be restored during decontamination, to meet food-grade standards, making PET suitable for multiple times closed-loop recycling.
  - Homogeneity of PE and PP waste is crucial for potential closed-loop recycling, as the processability of different PE and PP waste products (bottles, trays, etc.) differs significantly.
  - Degradation of PP is a limitation for recycling multiple times.
- Increasing the homogeneity of PE and PP products, and thereby facilitating recycling into new thin-walled packaging products in a closed loop, where physical and mechanical quality are preserved, might be achieved through additional tracer-based plastic sorting, regulatory harmonisation of product design, implementation of deposit systems and/or chemical recycling.

6 Conclusions

A framework relating the quality and quantity of recycled materials to the circularity potential of recycling systems was developed and applied to the case of plastic from household waste (HHW). The framework was accompanied by analyses of the chemical, mechanical and physical characteristics of waste and recycled plastic, a detailed characterisation of source-separated plastic waste and an assessment of potential performance of future recycling initiatives.

The *quality* of recycled material, closely related to its applicability, alongside *quantity* is crucial for the circularity of recycling systems. For plastic, between 18 and 57% of the European market for PET, HDPE, LDPE and PP relies on chemically high-quality plastic for food packaging production, and thus the production of recycled plastic suitable for food contact applications is key in achieving high circularity.

Considerable material losses and physical contamination currently occur when recycling plastic from HHW. State-of-the-art plastic sorting is an important element in limiting these issues; however, the best systems only have a circularity potential of around 0.40 (ideal circular economy requires 1) and are thus far from able to close the plastic loop. Moreover, the concentration of metals was elevated in recycled plastic, compared to virgin plastic, an aspect that might reduce the circularity further in the future, when plastic recycling rates increase. The composition of rigid, source-separated plastic was highly heterogeneous, as it contained both chemically high-quality food packaging and items of lower-quality, within each of the three dominant polymers, PET, PE and PP, representing more than 90% of the plastic waste. Additionally, 43% of the plastic waste was made of multiple polymers, and 10-11% was black.

To improve the circularity of plastic recycling systems, it is crucial to implement solutions that increase the quantity of recycled plastic while simultaneously preserving the chemical, physical and mechanical quality. Where the design of products for recycling can significantly increase the quantities of recycled plastic, separate recycling of high-quality food packaging is crucial, in order to preserve the chemical quality. However, for PE and especially PP waste, the homogeneity of the waste, when it comes to product type, is crucial in making recycling into new packaging products physically possible. Such homogeneity currently does not exist for PP food packaging, and solutions increasing the homogeneity of the PP waste sent to recycling are therefore additionally necessary, in order to reach the highest potential circularity.

7 Recommendations

In order to improve the circularity of current recycling systems managing plastic from household waste, it is necessary to adopt a holistic, system perspective and as such, actions related to all of the following solutions are recommended.

Solutions to increase the *quantity* of recycled material:

- Implement ***highly efficient plastic sorting,*** able to recover around 85-95% of the targeted plastic products into the intended waste stream (PET, PE, PP and film - *not* mixed plastic) sent to recycling, while limiting physical contamination to below around 5%.
- ***Design new plastic products for recycling***. This includes design measures such as producing products in a single polymer only and avoiding the production of black plastic products.
- Target both ***rigid and soft plastic*** in source-separation schemes, in order to collect and subsequently recycle the largest possible quantities of plastic. It is, however, important to stress that mixing rigid and soft plastic can reduce the efficiency of the sorting system, and it is therefore crucial that the source-separation and sorting processes are designed specifically to handle and separate both fractions.

Solutions to preserve the *quality* of the plastic during recycling:

- Systematic ***regulation of plastic products***, so that food packaging and non-food packaging can be recycled separately, using highly efficient, currently available sorting technology. This could include measures such as 1) limiting the production of packaging plastic to PET, PE and PP, 2) limiting the use of PET plastic to food packaging and 3) limiting the number of product types produced in PP. Such initiatives should be central to the "*essential requirements for placing packaging on the market[...] to ensure that, by 2030, all plastics packaging placed on the EU market is reusable or easily recycled.*" that the European Commission is currently revising, as part of its plastic strategy (EC, 2018d).
- Specifically ***target packaging plastic*** in the source-separation scheme, as this can be controlled regulatory via the EU's plastic strategy.

8 Perspectives

Based on the experience and knowledge gained during this PhD, several suggestions for future research are presented below:

- In addition to the detailed characterisation of rigid, source-separated plastic from households provided in this work, it is essential to obtain similarly ***detailed knowledge of the composition and quality of soft plastic***, as well as ***plastic in residual waste***, in order to quantitatively assess the effect of potential future recycling initiatives on the entire recycling pathway, including effects on waste generation and source-separation efficiencies.
- This study showed that the sorting process, whereby mixed plastic waste is sorted into individual waste polymer streams, is crucial for the performance and circularity potential of the entire recycling system. It is therefore suggested to ***improve understanding of the interaction between product design and the performance of the mechanical sorting process*** and on that basis establish how efficient these processes can be for different product types and product designs, allowing a more accurate assessment of the effect of changes to plastic product design and thereby, ultimately, waste composition.
- This PhD provides an indication of how the waste composition of different polymer waste affects the properties and potential applicability of recycled plastic. However, in order to regulatory align and harmonise polymers and products most effectively, it is important to ***gain further knowledge of how mechanical, physical and chemical properties of recycled plastic are influenced by different compositions of plastic waste*** sent to recycling, as well as ***how these properties influence the applicability of recycled plastic*** and thereby the possibility for closed-loop recycling. To obtain the most reliable results, and include the effects of contamination, it is important that such studies are based on actual waste, instead of imitated recycling processes with virgin plastic.

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In this online version of the thesis, **papers I-IV** are not included but can be obtained from electronic article databases, e.g. via www.orbit.dtu.dk, or on request from:

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