

# DETAILED CHARACTERIZATION OF POST-CONSUMER PLASTIC PACKAGING WASTE AND ITS DEPENDENCE ON COLLECTION SCHEMES: A DUTCH CASE STUDY

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Kortrijk, May 2024

The promotor, The author, The author, Prof. Dr. Ir. Steven De Meester Seppe Claeys



## <span id="page-3-0"></span>**PREFACE**

As the final part of my four-year course at the University of Ghent at Campus Kortijk, I present to you my master's dissertation "Detailed characterization of post-consumer plastic packaging waste and its dependence on collection schemes: a Dutch case study". This dissertation investigates the composition of plastic waste and whether there is a difference in composition between source separated and post-sorted plastic waste. This dissertation was written for obtaining the degree of Master of Science in Bioindustrial Sciences: Circular Bioprocesstechnology.

The reason I chose this course is because I have always been interested in the topic of climate change. Especially the technologies and strategies we can use to have an impact. Therefore, my ambition has been to learn about new sustainable and environmentally friendly developments that enable to deliver a positive impact. During my course, I was introduced to the concept of the "Circular Economy". It became evident that transitioning to a circular economy is of great importance. This together with my personal interest in recycling, especially plastics, inspired me to write my master's dissertation on this topic. I am convinced that an efficient waste management strategy for plastics will be an essential part of making the change to this circular economy. My goal is to further specialize in this field, paving the way for a professional career in this area of expertise.

I would like to express my gratitude to my tutor, Ph.D. student Alexandra Schmuck, for her excellent guidance throughout this journey. Your feedback, advice and motivation were instrumental in the successful completion of this dissertation. It was a genuinely great experience to work together on this project. I also extend my thanks to my promotor, Prof. Dr. Ir. Steven De Meester, for providing me with the opportunity to be part of this project and learn more about plastic recycling. Next, I would also like to thank Lies Harinck, Jung Suk Coene, and Noa Collier from the technical and scientific staff for the guidance and assistance during the many hours of practical lab work.

I would also like to acknowledge my friends from withing the course for the wonderful four years we had the pleasure of sharing. I will not forget the countless games of "manillen" as break from the dissertation and our other assignments. I am also grateful for the support we found in each other during this journey. I am equally grateful to my friends outside the course and my family. Then I would especially like to thank my girlfriend Britt for the unconditional support during these four years. Then I also want to thank my parents. They provided me with the opportunity to pursue this course, motivated me, and stood by me with unwavering confidence over these four years. They ensured that I could undertake this course under ideal conditions.

Lastly, I thank you, the reader, for your time and interest in this dissertation. I hope it provides you insightful perspectives within the plastics recycling sector.

Seppe Claeys

Kortrijk, May 19, 2024



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# <span id="page-7-0"></span>**ABSTRACT**

This master's dissertation provides an in-depth analysis of the composition of post-consumer plastic packaging waste for a case study in the Netherlands, with a particular focus on the influence of different collection schemes, namely source separation and post-separation. The large heterogeneity and the presence of various contaminants in plastic waste can cause difficulties in current mechanical recycling or chemical recycling processes. Ideally, the optimization of collection schemes, sorting infrastructure and recycling technologies starts with knowledge on the composition of the plastic waste and the efficiency of each unit operation in removing certain contaminants.

Therefore, in this dissertation, a major analytical campaign was organized in collaboration with a Dutch sorting company. The main objective is to quantify the polymer and elemental composition, which included metals, halogens, and elemental (CHNSO). This was performed using the following analytical instruments: Fourier-transform infrared spectroscopy (FTIR), inductively coupled plasma (ICP), ion-exchange chromatography (IC), and an elemental analyzer. This approach aimed to obtain deeper insights into the final quality of the material based on sorting efficiency in a typical sorting plant and the influence of the collection method. The obtained data can be used in decision making context to select which waste stream is most suitable for which recycling technology. The study is completed with a material flow analysis including the illustration of material flows through a sorting plant by a Sankey diagram.

A key finding of this study is that post-separation performs no worse than source separation in terms of bale quality. However, post-separation consistently results in higher residue levels, and attention should be paid to the elemental composition, particularly concerning certain prohibited metals in terms of migration from food contact materials. This is of great importance because there is a demand for mandatory recycled content in plastic packaging. To meet this demand, the high quality of recycled plastics must be guaranteed.



## <span id="page-8-0"></span>SAMENVATTING

Deze masterscriptie biedt een diepgaande analyse van de samenstelling van post-consument kunststof verpakkingsafval voor een casestudy in Nederland, met speciale aandacht voor de invloed van verschillende inzamelsystemen, namelijk bronscheiding en nascheiding. De grote heterogeniteit en de aanwezigheid van verschillende verontreinigingen in kunststofafval kan problemen veroorzaken in de huidige mechanische of chemische recyclageprocessen. Idealiter begint de optimalisatie van inzamelschema's, sorteerinfrastructuur en recyclagetechnologieën met kennis over de samenstelling van het kunststofafval en de efficiëntie van elke processtap om bepaalde contaminanten te verwijderen.

Daarom werd in dit proefschrift een grote analytische campagne georganiseerd in samenwerking met een Nederlands sorteerbedrijf. Het hoofddoel is het kwantificeren van de polymeer- en elementaire samenstelling, waaronder metalen, halogenen en elementair (CHNSO). Dit werd uitgevoerd met behulp van de volgende analytische instrumenten: Fouriertransform infraroodspectroscopie (FTIR), inductief gekoppeld plasma (ICP), ionenwisselingschromatografie (IC) en een elemental analyzer. Het doel van deze aanpak was om meer inzicht te krijgen in de uiteindelijke kwaliteit van het materiaal op basis van de sorteerefficiëntie in een typische sorteerinstallatie en de invloed van de inzamelmethode. De verkregen gegevens kunnen gebruikt worden bij het nemen van beslissingen om te selecteren welke afvalstroom het meest geschikt is voor welke recyclingtechnologie. De studie wordt aangevuld met een materiaalstroomanalyse inclusief de illustratie van materiaalstromen door een sorteerinstallatie aan de hand van een Sankey-diagram.

Een belangrijke bevinding van uit deze studie is dat nascheiding niet slechter presteert dan bronscheiding op het gebied van baalkwaliteit. Nascheiding leidt echter consistent tot hogere residugehalten en er moet ook aandacht worden besteed aan de elementaire samenstelling, met name wat betreft bepaalde verboden metalen met betrekking tot migratie uit materialen die in contact komen met voedingsmiddelen. Dit is van groot belang omdat er vraag is naar verplichte recycled content in plastic verpakkingen. Om aan deze vraag te voldoen, moet de hoge kwaliteit van gerecyclede kunststoffen worden gegarandeerd.



# <span id="page-9-0"></span>1 INTRODUCTION

Plastics, due to their versatile properties, have become an integral part of our daily lives, contributing significantly to the enhancement of our living standards. It is almost inconceivable to imagine a world devoid of plastics. Their desirable characteristics have led to their widespread use in various sectors, including packaging, electronics, textiles and clothing, healthcare, automotive, construction, and several others (Letcher, 2020).

The history of plastics dates back to the 1850s when the first man-made polymer, nitrocellulose, was created by English metallurgist and inventor Alexander Parkes and patented under the name Parkesine. In 1869, American inventor John Wesley Hyatt developed a practical method for producing solid, stable nitrocellulose, leading to the creation of the first commercially successful man-made polymer, Celluloid. The first synthetic polymer, or plastic, known as Bakelite, was created by Belgian-American chemist Leo Baekeland in 1907. The inventions of Celluloid and Bakelite marked the dawn of the plastic era. However, it was in the post-war decades following World War II that plastic producers expanded into a wide range of sectors, leading to the steady growth of the plastic industry (Geyer, 2020). As of 2021, global plastic production had reached 390.7 million tons (Plastics Europe, 2023a).

The great properties plastics possess that make the material so attractive have also led serious environmental problematics. Due to its non-degradable nature, plastic waste has accumulated in various ecosystems where it negatively effects the environment (Sharma et al., 2023; Welden, 2020). Preventing and resolving plastic pollution forms one of the most important environmental challenges for the coming years (Parker, 2019).

An effective waste management strategy for plastic packaging waste (PPW) through recycling can reduce the impact of plastic waste on the environment (Hahladakis & Iacovidou, 2019). Therefore, the European Union (EU) has set some ambitious targets: the target is to recycle 50% of PPW by 2025, and this target increases to 55% by 2030 (Directive 94/62/EC, 2018).

The first steps in the recycling chain are collection and sorting. The collection scheme is of great importance because they directly influence the composition of the waste stream and thereby affect the further downstream procedures, like the pretreatment, sorting, and recovery operations. The waste streams contain mixed plastics with a complex composition even in collection systems that require at home sorting such as common practice in Belgium, Germany and part of the Netherlands. A crucial step in obtaining high-quality plastic products after reprocessing is the sorting process. This is typically carried out in a material recovery facility (MRF) and involves separating plastics from contaminants and categorizing them by polymer type. (Delva et al., 2019).

Several studies have already been conducted characterizing certain fractions of waste streams. Dimitrakakis et al. (2009a) determined the presence of heavy metals and halogens in small waste electrical and electronic equipment found in residual household waste (HHW) in Germany. Strangl et al. (2017) characterized odorous contaminants in postconsumer (PC) PPW in Germany. Eriksen & Astrup (2019) characterized source-separated rigid plastic waste on the level of product design in Denmark. Gala et al. (2020) characterized the chemical composition, heating values, metal, and halogen contents of plastic film waste from mixed municipal solid waste (MSW) in Spain. Roosen et al. (2020) performed a detailed analysis of polymer and elemental composition of selected PPW products. It is on the latter study that this dissertation is based.The added value of the research in this dissertation is that samples were taken from two collection



schemes, source- and post-separation, at the same sorting plant. This provides a unique view on the differences in composition between the two schemes. Only in the recent work of Klingenberg et al. (2024) a similar study was conducted with the focus on the polymer composition and an assessment of material performances for both collection schemes. While the EU is striving to improve the source-separation efficiencies, post-separation is gaining a lot of interest in the Netherlands (Dijkgraaf & Gradus, 2020).

The first main goal of this master's dissertation is to expand on the work of Roosen et al. (2020) by preforming a detailed analysis of the polymer and elemental composition of target PPW items out of three material categories supplemented by random samples out of four categories. The target samples include PE bottles, PP trays, and mixed foils. The four waste categories of the random samples are PE rigids, PP rigids, PE flexibles, and mixed plastics waste. The second main goal is to determine if there is a difference between PPW collected and sorted through a sourceseparation and a post-separation system. This comparison is investigated by focusing on the random samples and the influence of special items (e.g., toys, CD case, and blister packaging), that are regularly found in post-separation and less in source-separation. The third main goal is to perform a material flow analysis (MFA) by developing a model that represents the material flows of the sorting plant in matter. The model is used to simulate the bale quality that will be compared with the findings of the analysis of the polymer composition.

This study starts with chapter [2](#page-11-0) [Literature Review,](#page-11-0) existing out of three parts. Firstly, a general introduction to plastics and addressing the importance of a circular economy (CE) for plastics. Secondly, framing the current waste management strategy for PPW.This section goes through the life cycle of plastics looking at the various end of life options and finally the challenges associated with them. Thirdly, the impact of the collection method, source separated or post-sorted, is discussed in more detail. Chapter [3](#page-30-0) [Materials and Methods,](#page-30-0) describes in detail the analysis techniques applied and the instruments used. The analysis performed are divided into three parts. The first part exists out the quantification of the polymer composition. The second part involves the quantification of the elemental composition, including the metal contents, halogen contents, and CHNSO analysis. The third part comprises a MFA. In chapter [4](#page-37-0) [Results,](#page-37-0) the results are presented and these are discussed in chapter [5](#page-47-0) [Discussion.](#page-47-0) The final conclusion of this master's dissertation can be found in chapter [6](#page-53-0) [Conclusion.](#page-53-0)



# <span id="page-11-0"></span>2 LITERATURE REVIEW

### <span id="page-11-1"></span>2.1 Introduction to Plastics

#### <span id="page-11-2"></span>2.1.1 Plastics the Basics

Plastics were a revolutionary material during the 20<sup>th</sup> century. Today, plastics have become an essential material in our daily life. Plastics are found in all kinds of modern applications that might not have been possible to make without the unique properties of these materials (Crawford & Martin, 2020).

Plastics are group of materials made of synthetically produced polymers to which additives are incorporated. Polymers are large molecules made byconnecting multiple small molecular units, called monomers, to each other by the process of polymerization. The number of those units is called the degree of polymerization. The terms polymers and plastics are often used interchangeably but do not mean the same. A polymer is a pure material without additives present existing out of a long chain-like molecule. Polymers can also be found in nature in such as rubber, cellulose, or silk. On the other hand, plastics are a man-made material. "Plastic" is a general term used to describe man-made or synthetic polymers that use additives to enhance their properties(Crawford & Martin, 2020; Geyer, 2020).

Today's plastics are made up of a wide range of polymers with variety of chemical additives added to them to change and improve their properties. A key distinction for plastics can be made by their melting behaviour. The most wellknown and frequently used plastic types are thermoplastics, including polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polystyrene (PS), and polyamide (PA). Because of their ability to melt, harden, and re-melt, thermoplastics are potentially recyclable. Thermosets are another class of plastics. This type of plastic is formed out of an irreversible three-dimensional network. Therefore, thermosets cannot be remelted and reformed, which makes recycling them difficult. Examples of common thermosets are polyurethanes (PUR), unsaturated polyester, silicone, epoxy, melamine, phenolic, and acrylic resins (Geyer, 2020).

[Figure 2-1](#page-11-3) illustrates the global annual production of primary plastics from 1950 to 2019. Primary plastics are manufactured using primary feedstock, which includes crude oil and natural gas (Geyer, 2020). The graph reveals the persistent year-on-year increase in plastic production. Projections indicate that by 2060, plastic production will reach to approximately 1,231 Mt tons annually (OECD, 2022).



<span id="page-11-3"></span>Figure 2-1: Global annual primary plastics production (in Mt) by material type from 1950 to 2019 (Geyer et al., 2017; OECD, 2022)



In 2022, global plastics production reached 400.3 million tonnes. Whereas 90.6% is fossil-based, 8.9% mechanically recycled, 0.5% bio-based, < 0.1% chemically recycled, and < 0.1% carbon-captured. Europe was responsible for 58.7 million tonnes of the plastic produced worldwide in 2022 (Plastics Europe, 2023b). In 2021, global production was 390.7 million tonnes, with Europe producing 57.2 million tonnes. [Figure 2-2](#page-12-0) shows the distribution of global plastics production by type. Circular plastics make up roughly 9.8% of all plastics produced worldwide. [Figure 2-3](#page-12-1) shows the distribution of global plastics use by application. Plastic packaging is clearly the largest market followed by building and construction, with 44% and 18% respectively (Plastics Europe, 2023a).



<span id="page-12-0"></span>There are differences in the lifespans of plastic parts and products that enter the market. Certain products, such as cars, cables, insulation boards, and electrical and electronic devices, stay in use for many years. This multi-year lifespan entails that these products do not become waste in the same year that they entered the market. For example, plastics used in building and construction applications stay in use for around 50 years, while plastic packaging, on the other hand, is mostly collected in the same years as it was introduced to the market. The differing lifespans of various plastic products and parts contribute to the explanation of why waste quantities for a given year are lower than the total number of plastic products and parts that are introduced to the market in that same year (Plastics Europe, 2022).

This also means that the plastics introduced to the market [\(Figure 2-4\)](#page-12-2) are not comparable with the plastic waste collected [\(Figure 2-5\)](#page-12-3). [Figure 2-4](#page-12-2) and [Figure 2-5](#page-12-3) show the consumption of plastic products and collection of postconsumer (PC) plastic waste by application in 2020 for the EU27+3, respectively. The total of PC plastic waste amounts to 29.5 million tonnes compared to 53.6 tonnes introduced to the market the same year (Plastics Europe, 2022).



<span id="page-12-1"></span>

<span id="page-12-3"></span>Figure 2-5: Post-consumer plastic waste collection by application (Plastics Europe, 2022)

<span id="page-12-2"></span>

#### <span id="page-13-0"></span>2.1.2 Environmental Consequences of Plastic Pollution

Plastic pollution has emerged as one of the most important environmental challenges (Parker, 2019). Plastic wastes have accumulated in ecosystems due to the non-degradable nature of these polymeric materials, their growing use, and limited waste management regulations. Some of the actions making the situation of plastic pollution worse include the increased usage of single-use plastic items and global low recycling rates (Sharma et al., 2023). In 2020, in the EU27+3, of the 29.5 million tons of PC plastic waste collected, 35% were sent for recycling. The remaining waste is mostly used for energy recovery (42%) and otherwise landfilled (23%) (Plastics Europe, 2022).

When discussing plastic pollution, the first thought is often marine plastic pollution. It is important not to forget that all plastic is produced on land, and most of the plastic products that do not find their way to a desired end-of-life (EOL) management solution are disposed of on land (Hurley et al., 2020). Since 2016, it is estimated that for the marine environment, the global emission of plastic waste is within a range of 9 to 23 million metric tons, and for the terrestrial environment, 13 to 25 million metric tons (MacLeod et al., 2021).

Discarded plastic waste is commonly seen in a variety of ecosystems and frequently interacts negatively with wildlife (Welden, 2020). Plastic pollution can be found everywhere, from deserts to farms, from mountaintops to the deep ocean, in tropical landfills, and in Arctic snow (MacLeod et al., 2021). Therefore, extensive research about plastic pollution in the environment is being carried out (Avio et al., 2017; Chae & An, 2018; Schmaltz et al., 2020). Even with variations in the polymer structure and the combination of additives, the majority of plastics are highly resistant to degradation. So, most plastics have significantly longer lifespans than individual organisms. Therefore, separating out specific polymers to evaluate their influence on the environment is not highly effective. Potential environmental effects are often determined by the product's size, point of loss, and how the material is transported through the environment (Welden, 2020). For example, micro- and macro-plastics will interact differently with the environment. The point of loss can be on land or in water environments either intentionally or unintentionally. Plastic waste can be transported by being blown away by the wind or following the flow of a river.

Biota, especially marine wildlife, is regularly faced with either the ingestion or entanglement of plastic debris. In the case of entanglement, species get wrapped in plastic debris. As a result of these interactions, the organism may encounter harm, have lower mobility from carrying a piece of debris, or perish directly or indirectly due to starvation, exposure, or drowning. Ingestion of plastic debris is a serious concern for primarily filter feeders and predators of planktonic organisms (Welden, 2020). Kühn & van Franeker (2020) found that 914 species, including marine birds, mammals, turtles, fish, and invertebrate species, are affected by entanglement and/or ingestion of plastic debris. Wherefrom, 701 species were recorded to have ingested plastics, and 354 species were entangled.

Microplastic pollution is a complicated issue with serious consequences for environmental and public health. This pollution problem is a classic transboundary example of how land-based pollution may spread so far that it even reaches isolated locations (Bank & Hansson, 2022). Microplastics are plastic particles in a size range from 1 to 5 µm. When plastic particles are smaller than 1 µm, they are classified as nanoplastics (Kiefer et al., 2023). Microplastics can originate from a variety of sources, like shedding fibers from clothing in the washing machine, tire wear, industrial processes, and agricultural activities. (Kiefer et al., 2023). Another possibility is that when plastic waste is directly polluted in the environment, microplastics originate from the fragmentation process of larger particles. This degradation process is induced by mechanical stress, photo-oxidation, hydrolysis, and interference with biota (Sorasan



et al., 2022). Microplastics can spread from rivers to seas or oceans, where they may cause damage to marine life. Sometimes, sea fish eat these microplastics, and humans consume this seafood without knowing that it is contaminated with microplastics. These microplastics may contain toxins that can enter the bloodstream, thereby affecting human health (Laskar & Kumar, 2019).

The greenhouse gas (GHG) emissions from various processes in the lifespan of plastic-related products pose a serious danger to the environment and contribute to the rise in global temperatures. Fossil fuels, including coal, gas, and crude oil, are the raw materials that make up plastic building blocks, and during the plastic's lifespan, GHG is released. The release of GHG emissions takes place from the extraction and transportation of fossil fuels all the way to the refining and manufacturing of the products and later during the EOL management (Sharma et al., 2023). The work of Zheng & Suh (2019) determined that the GHG emissions over the life cycle of conventional plastics produced in 2015 account for 1.8 gigatons (Gt) of CO<sub>2</sub> equivalent, excluding the contribution of carbon credits from the recycling process. This amount is equal to 3.8% of the 47 Gt  $CO<sub>2</sub>$  equivalent that was released into the atmosphere that year. I[n Figure 2-6,](#page-14-0) an overview is given for the contribution of each life cycle stage and polymer type. In the EOLstage, recycling produced 49 megatons (Mt) of CO<sub>2</sub> equivalent, which is a -67 Mt CO<sub>2</sub> equivalent reduction compared to the production of an equal amount of virgin polymers. Thus, more polymer recycling instead of virgin plastic production could lower the total global GHG emissions (Zheng & Suh, 2019).



Figure 2-6: GHG emissions from the conventional plastics life cycle worldwide in 2015, by polymer type and life cycle stage (Zheng & Suh, 2019)

<span id="page-14-0"></span>With the development of circular technologies, there are increasingly more alternatives for lowering GHG emissions from plastics, including plastic recycling, bio-based manufacturing, and carbon capture and utilization (Bachmann et al., 2023).



#### <span id="page-15-0"></span>2.1.3 Drivers to Transition to a Circular Economy

The idea of an ideal CE is that materials never become waste and nature is restored. Products and materials are kept in circulation through processes like maintenance, reuse, refurbishment, remanufacture, recycling, and composting. In the current linear economy, products are made out of the materials available on Earth, and when the product has lost its value or is not needed any more, it is disposed of (Ellen MacArthur Foundation, n.d.).

Years of research have shown that effective waste management of PPW through recycling has reduced the impact on various aspects of the environment. Recycling results in a net reduction in the emissions of carbon dioxide, carbon monoxide, acid gases, particulate matter, heavy metals, and dioxins. Recycling also leads to net energy savings. Finally, it also contributes to less acidification and nitrification of surface waters (Chilton et al., 2010; Craighill & Powell, 1996; Wenisch et al., 2004). Life-cycle assessment (LCA) is a widely used tool to determine the environmental impacts and sustainability of a given process, like EOL management. Recycling, which is key in the transition to a CE in general, is preferred above landfilling and incineration with energy recovery (Hou et al., 2018). A recent study from Civancik-Uslu et al. (2021) compared mechanical recycling, thermochemical recycling, and incineration with energy recovery for four newly collected subfractions in Belgium: PP, PS, mixed polyolefin (PO) rigids, and PE films. The results of this study clearly showed that recycling is the better option than incineration and mechanical recycling has a lower environmental impact than thermochemical recycling. It is clear that recycling is an essential step in the transition to a CE and closed material loops (Eriksen & Astrup, 2019).

To make the transition to a CE, the European Commission adopted the new circular economy action plan (CEAP) in March 2020 (European Commission & Directorate-General for Communication, 2020). The action plan is one of the main building blocks of the European Green Deal and has the objective of making the EU transform into a modern, resourceefficient, and competitive economy. The ambitious target is to make the EU net of GHG emissions neutral by 2050 (European Commission & Directorate-General for Communication, 2021). As part of the CEAP, the EU adopted the "plastic strategy". The aim of this strategy is to reduce marine pollution, GHG emissions, and our dependence on fossil fuels. To achieve these goals, the EU wants to change how plastic products are designed, produced, used, and recycled (European Commission et al., 2018). In Directive 94/62/EC, the EU has set ambitious targets for the recycling rates of its member states. By 2025, a minimum of 65% by weight of all packaging waste must be recycled, and for PPW, 50%. Against 2030, it is 65% for all packaging waste and 55% for PPW (Directive 94/62/EC, 2018).



## <span id="page-16-0"></span>2.2 Current Waste Management Strategy for Plastic Packaging Waste

## <span id="page-16-1"></span>2.2.1 Plastic Life Cycle

The plastic life cycle [\(Figure 2-7\)](#page-16-2) consists of several steps. Beginning with the start-of-life phase (Ragaert et al., 2017). This phase involves the extraction of raw materials, refinement and cracking, processing, and manufacturing (Debris Free Oceans, 2023). During this phase, the raw materials, such as fossil fuels or recycled materials, are converted into plastic products using various processing techniques. During the start-of-life phase, the materials and products have not been in contact with the consumer. While processing, the first type of plastic waste is generated, namely postindustrial (PI) waste. This PI waste originates from runners from injection molding, waste from production changeovers, fall-out products, cuttings, and trimmings. PI waste has several advantages regarding the quality of the waste stream. PI is most often clean and of known composition. Also, it is sometimes mono stream, which means that there is no contamination from other polymers or non-polymers(Ragaert et al., 2017).The second phase is the use phase. Here, the plastic products are used by the consumer (Ragaert et al., 2017). As seen in [Figure 2-3,](#page-12-1) plastic can be used in several applications in of a variety of different sectors. The last phase is when the plastic products are disposed of and become PC waste; this is the EOL phase. PC waste is a more complex stream than PI waste due to the contamination that occurs during the use phase and the EOL phase itself. As a result, the exact composition of this waste stream is, in most cases, unknown (Ragaert et al., 2017). There are several EOL options for plastic products available that will be discussed in the following sections.



Figure 2-7: Plastic life cycle with circular strategies implemented (Schwarz et al., 2021)

<span id="page-16-2"></span>

#### <span id="page-17-0"></span>2.2.2 End-of-life Management

A major global challenge is the EOL management of MSW that is both sustainable and efficient. Circularity and EOL treatment are becoming increasingly popular, particularly with regard to plastics (Roosen et al., 2022). The majority of packaging materials, such as paper and cardboard, glass, and metal, have already seen significant development in the CE. However, the PC PPW collection and recycling system is still less advanced (Brouwer et al., 2019). The processes of collecting and recycling plastic waste showcase regional variances, supported by diverse infrastructure that varies greatly among nations, regions, and cultural backgrounds. While the Global North has sophisticated waste management infrastructure and a formal collection system to support plastic waste management, many Global South countries have undeveloped or uncontrolled recycling programs with lax enforcement of environmental and human protection (Hahladakis & Iacovidou, 2019).

In order to fulfill waste prevention and management legislation and policies, the following waste hierarchy [\(Figure](#page-17-2)  [2-8\)](#page-17-2) should be applied. Prevention is the most preferred method, and disposal is only acceptable if no other option is possible. Prevention is the action performed prior to a product becoming waste. "Preparing for re-use" refers to the process of checking, cleaning, or fixing recovery operations, which is how waste-derived products or their component parts are made ready for reuse without the need for additional pre-processing. Recycling consists of any recovery process that turns waste materials back into materials, products, or substances for use in different applications or for the original purpose. Recovery means any process where the main outcome is waste being prepared to fulfill a specific function for a useful purpose by substituting other materials. Disposal is any operation that is not recovery (Directive 2008/98/EC, 2018).



Figure 2-8: Five-tiered waste hierarchy as established in Directive 2008/98/EC (European Commission, 2023)

<span id="page-17-2"></span>Out of the waste hierarchy [\(Figure 2-8\)](#page-17-2) recycling is the preferred EOL option for plastic waste. But in 2020, only 35% of all PC plastic waste in the EU is recycled. Most of the plastic waste, 42%, is sent to energy recovery, and the remaining 23% is landfilled. Energy recovery is the process where combustible plastic isincinerated to generate energy in the form of electricity and/or heat conversion (Plastics Europe, 2022).

#### <span id="page-17-1"></span>2.2.3 Mechanical Recycling

Plastic recycling is a process where secondary materials are used for the production of new products and components. These secondary materials are manufactured out of plastic waste that has been recovered and reprocessed. There are four recycling processes: primary recycling (PI), secondary recycling (mechanical recycling), tertiary recycling (chemical recycling), and quaternary recycling (energy recovery) (Hahladakis & Iacovidou, 2019). Today, if only the first three recycling processes are considered, the majority of PPW is recycled through mechanical recycling (Lange, 2021).



Based on the quality of the secondary material produced after recycling, a difference can be made between closed- and open-loop recycling. Closed loop, or upcycling, is the process where secondary materials of equal or close to similar quality are made. This makes it possible to use the recycled material for the production of the same product completely, or sometimes in a mixture with its virgin equivalent, to create a form of dilution. Open-loop, or downcycling, is the process where secondary materials are manufactured that may have lower quality than virgin materials. This last process can be seen as a form of cascading (Hahladakis & Iacovidou, 2019; Ragaert et al., 2017). The recycled materials are not of sufficient quality to be used in high-end recycling routes but mostly find destinations in mixed-polymer bulk applications like plant trays, compost bins, and street or garden benches (Roosen et al., 2020).

The mechanical recycling process consists of a complex chain of activities that can be separated into three parts. First there is the collection of the PPW, followed by sorting, and finally reprocessing (Jansen et al., 2015).

#### <span id="page-18-0"></span>2.2.3.1 Collection System

The first step in the plastic recycling value chain is the collection system. Collection is an important step to convert waste raw materials into new plastic products; it is quite often also the yield-limiting step in the complete recycling process (Delva et al., 2019; Thoden van Velzen et al., 2019). "Collection" refers to the process of gathering waste, which included preliminary sorting and storage in preparation for transportation to a waste treatment facility (Directive 2008/98/EC, 2018). Collection systems should be optimized because they define the composition of the waste streams and, in turn, the downstream processes, such as the operations for pre-treatment, separation, and recovery operations. In general, there are four main collection methods used for PPW:

- Curbside collection is the most widely accessible collection method. Because it is the most practical procedure for civilians to undertake, this reflects in high recovery rates. It is requested of inhabitants to separate potentially valuable recyclables (e.g., plastic, paper, carboard, and metals) from their household waste (HHW).
- Drop-off collection involves the placement of various containers for specific materials that are to be recycled at key community locations.
- Buy-back centers, which are primarily owned by private businesses, buy recyclables from customers. The materials collected in the centers have a low level of contamination due to the specifications these centers impose.
- Deposit/refund programs provide for the refund of a deposit upon the return of plastic containers to either the original seller or the relevant redemption center (Delva et al., 2019).

For the first two collection methods, there may also be a difference in how recycled resources are collected and later recovered. PPW may either be collected separately from the other waste streams, or co-mingled with metals and drinking cartons (PMD scheme) or PPW might be collected together with all other waste streams. These two collection methods are also referred to as : source-separated or separate waste collection systems, and pre-treatment in a MRF of mixed MSW or post-separation (Cimpan et al., 2015; Lederer et al., 2022).

Source-separation is often the collection method of choice which historically can be explained by the early attempts made in 1970 in Europe to physically process mixed MSW in order to recover valuable resources (e.g., compost, recyclable materials, and alternative fuels) including the development of specific technologies. However, the output product



quality was unsuitable for recycling applications. The technologies also brought the risk of spreading hazardous substances into the environment. This made source separation the preferred method of collection system, both technically and environmentally, which is still strongly reflected in European waste legislation today (Cimpan et al., 2015).

In the 1980s, the main focus was on separate collection programs for a few materials, such as glass and paper. The materials could be easily collected through a curbside collection system. It was in the 1990s that separate collections quickly evolved, and the portfolio of collected mono-material streams expanded significantly. Source separation began to show its weakness, particularly the public's willingness to participate. Also, the increasing number of mono-material fractions collected caused an increase in the complexity and cost of the collection system. Mixed collection systems were seen as a solution to simplify the complexity and reduce the requirement for public engagement. This led to the development of source separation programs where recyclables were partly or fully commingled. The types of materials accepted for collection in these commingled recyclable collection programs differ between countries and regions (Cimpan et al., 2015).

For example, Belgium makes use of a commingled source-separated collection system with the PMD bag. PMD stands for plastic packaging, such as bottles, flasks, containers, jars, trays, lids, bags, and foils which are collected together with other recyclable materials like metal packaging and drinking or beverage cartons. In households, these packaging materials are separated in a separate blue bag or PMD bag from the residual MSW. The bags are retrieved via a curbside collection system (Fost Plus, 2023).

Research from Belgium has shown that the expansion of the collection system to include more PPW definitely has a positive effect on the quantity of PC plastic waste collected, but the waste stream becomes more complex compared to the previously limited system (Roosen et al., 2022).

Recyclable materials can also be recovered from MSW in addition to source separation programs. Their quality might be comparable to materials from separate collection systems, depending on how much cross-contamination takes place between the materials. MSW in general has a higher moisture content and degree of surface contamination. That is why these materials are traded on secondary material markets. Especially in urban areas, where source separation is challenging, post-sorting MSW is taking interest. The system can be applied as a complementary to or as a substitute of source separation collection system (Cimpan et al., 2015).

In the Netherlands three systems exist for the separation of PPW from households. The first system is similar to the one used in Belgium, a source-separated system where PMD bags are used to collect PPW together with metal packaging and beverage cartons. In the second system, plastic, metal, and beverage cartons are collected together with all residual waste. Later sorting will take place in a MRF; this is post-separation of mixed MSW. Then there is the third option, the deposit refund system, where large PET bottles and crates of beer bottles can be returned to supermarkets to get their deposit back (Kennisinstituut Duurzaam Verpakken, n.d.). The focus of this dissertation is on these first two collection schemes, source separation and post-separation, which will be discussed further in section [2.3.2.](#page-24-0)

#### 2.2.3.2 Sorting

The sorting of the incoming waste stream is the second step in the process of mechanical recycling. In general, the composition of the incoming waste stream is heterogeneous, so its exact composition is not entirely known. Most waste



streams consist of mixed plastics that are likely contaminated with either organic fractions (e.g., food residues) or nonplastic organic fractions (e.g., metals, wood, and paper) (Delva et al., 2019). Typically, a series of sorting steps are used to separate plastic waste into the desired fractions (Lange, 2021). The steps in the sorting process are dependent on the incoming waste stream and the quality and quantity requirements of the output product (Serranti & Bonifazi, 2019). These include size sorting, removal of foreign materials, sorting according to polymer type, and sizing and granulation into plastic recyclate (Li et al., 2022).

A lot of the separation methods are based on gravimetric principles. Air classifiers separate lighter materials from heavier ones by using air as a medium. The waste stream enters a column with a rising air current sideways. Light materials are blown upward, and heavier ones drop downward. The fundamental idea behind ballistic separation is the disparate motion characteristics of particles with varying weights, sizes, and shapes. In sink-float separation, the material is introduced into a tank containing a fluid with a certain density. Materials with a lighter density than the fluid will float, and those with a higher density will sink. Then there is the hydrocyclone, which is based on the centrifugal forces and fluid resistance of different particles (Serranti & Bonifazi, 2019).

Most sorting facilities make use of sensor-based sorting technologies. These exist out of three components: a conveyer belt for the material feed, a sensor that is connected to a computer analyzing the collected data, and a pneumatic system to mechanically separate materials (Serranti & Bonifazi, 2019). Near-infrared (NIR) spectroscopy is the most commonly used sensor-based automatic sorting technique (Ragaert et al., 2017). This technology does have some limitations. The sensor can only read what it sees. For example, for a PET bottle with a PP label, it is possible that the sensor only reads the label that is made out of PP instead of the bottle that is made of PET. Additional challenges are multilayer products, such as PP trays, for which the NIR can only read the top or bottom layer. The tray may, however, consist of multiple other layers which have a significant share in the total mass of the product (Roosen et al., 2020). Also, spectroscopic scanners used as sensors are not able to read carbon black pigments. This results in the rejection of these materials. To optimize the sorting process, this technology is used in combination with other separation steps (Hahladakis & Iacovidou, 2019; Ragaert et al., 2017). Other sensor-based techniques are visible spectroscopy, hyperspectral imaging , X-ray fluorescence, and laser-induced breakdown spectroscopy (Serranti & Bonifazi, 2019).

Other commonly used separation technologies are magnetic separation for the removal of ferrous metals and an Eddy current separator for the removal of non-ferrous compounds (Serranti & Bonifazi, 2019). The linkage of the various unit operations may differ from one sorting plant to another.

It is important that the plastic waste after the sorting process must be of good quality to be accepted for reprocessing. There are several factors affecting the quality of plastic waste: application, polymer type, multiplicity of collection schemes, additives and other materials, impurities introduced at the consumption level, and degradation during its service life (Hahladakis & Iacovidou, 2019). In Germany and the Netherlands, as a minimum set of quality standards the sorted fractions must meet the DKR specifications. Targeted products for the Dutch situation are the following: PET (DKR 328-1), PE (DKR 329), PP (DKR 324), Film (DKR 310) and mixed plastics (DKR 350) (Jansen et al., 2015).

#### 2.2.3.3 Reprocessing

After the sorting and pre-treatment for mechanical recycling that only involved mechanical processes (e.g., grinding, washing, separating, drying, re-granulating, and compounding) follows the reprocessing of the sorted fraction consisting out of plastic flakes or pellets. Frequently used reprocessing techniques are re-melting, injection molding,



extrusion, rotational molding, and heat pressing. The final product or recycled plastics can then replace virgin plastics for the production of plastic products (Delva et al., 2019).

#### <span id="page-21-0"></span>2.2.4 Chemical Recycling

Chemical recycling, also known as thermochemical or feedstock recycling, is an alternative method to mechanical recycling. In this method, the plastic waste stream is degraded into its monomers or other useful basic chemicals. Typical processes used are hydrolysis, pyrolysis, hydrocracking, and gasification. The products from chemical recycling can either be used again for polymerization into new plastics or for the production of valuable chemicals and alternative fuels (Delva et al., 2019).

This recycling technique shows great potential for handling highly heterogeneous and contaminated waste streams that are difficult to separate economically viable or are completely technically unfeasible with mechanical recycling process. For example, pyrolysis is an interesting technology for heterogeneous waste streams like mixed PE/PP/PS, multilayer packaging, fiber-reinforced composites, polyurethane construction and demolishing waste (Ragaert et al., 2017).

#### <span id="page-21-1"></span>2.2.5 Challenges for Creating an Effective Waste Management System

One of the challenges of creating an effective waste management system is the heterogeneity of current waste management systems, infrastructure, and legislation that vary a lot amongst different regions, countries, and cultures. This makes it tough to get a broad view of the different systems (Di Foggia & Beccarello, 2022; Hahladakis & Iacovidou, 2019). Different collection systems can make it confusing for people to dispose of their waste in the correct recycling receptacles. This results in compromising the quality of the plastic waste stream (Hahladakis & Iacovidou, 2019).

Another challenge is the composition of the waste steam that can be highly heterogeneous. This is due to the different product types, the product design and the contamination of the material (Eriksen & Astrup, 2019). This complexity of the waste stream makes it a difficult task for sorting facilities to separate the incoming waste into the desired fractions.

At the moment most of the plastic are recycled through mechanical recycling. The issue here is that plastic degrades under certain conditions over time. Factors affecting the plastic material are heat, oxidation, light ionic radiation, hydrolysis, and mechanical shear. Plastic degradation both takes place during the lifetime of the plastic product and during the reprocessing phase mainly caused by thermal-mechanical degradation (Ragaert et al., 2017). Another issue is that mechanical recycling can cause accumulation of unwanted constituents containing metals and halogens that can potentially form health risks. Because of this its possible that recycled plastic does not meet the required legislative migration limits for food contact materials (Regulation 10/2011, 2023). Therefore, recycled plastics are used in other lower value applications then food packaging (Eriksen et al., 2018; Roosen et al., 2020).

Optimizing new technologies to increase the quantity and improve the quality of recycled PPW begins with knowledge of the composition of the collected waste streams. Nonetheless, only limited systematic data on the complete composition of PPW streams is currently available (Gala et al., 2020; Roosen et al., 2020).



## <span id="page-22-0"></span>2.3 Plastic Packaging Waste Composition and the Influence of the Collection System

#### <span id="page-22-1"></span>2.3.1 Contamination in the Plastic Packaging Waste Stream

Contamination can affect polymer reprocessing activities (Hahladakis & Iacovidou, 2019). Based on rheological behavior it was shown that contaminants act as catalyst to degrade the material. The combination of contaminants in the plastic and the high shear rates during extrusion processes lead to a decrease in molar mass (Garcia et al., 2013). First, there are the designed contaminants that are either embedded in the plastic (e.g., additives, coatings, inks) or non-embedded but attached to the plastic (e.g., caps, labels, adhesives) (Hahladakis & Iacovidou, 2019). This group of contaminants is merged with plastics on purpose to control the plastics properties and ensure that it meets the requirements for use in particular applications (Eriksen et al., 2018). Secondly, there are the contaminants or impurities that are created with the plastics during the production phase, their service life, and EOL management in collection and sorting processes (Eriksen et al., 2018; Hahladakis & Iacovidou, 2019). In the production phase, contaminants can come from catalyst residues or metal impurities (Eriksen et al., 2018). Created contaminants at use phase and collection are dust, soil, organic residues, grease, etc., and at the sorting stage, non-targeted polymeric materials can cause contamination in the desired stream (Hahladakis & Iacovidou, 2019).Some of the crucial heteroatom and metal contaminants in the PPW stream and their main origin are listed in [Table 2-1.](#page-22-2)

<span id="page-22-2"></span>

Table 2-1: Main origins of heteroatoms and metals in PPW (Kusenberg et al., 2022)



#### 2.3.1.1 Multilayer Plastic Films

Polymer contamination is occurrence of polymers other than the wanted polymer in the sorted waste category. Even with perfect sorting of all packaging items, various types of polymers can still be found in each waste category. The degree of polymer contamination relies on the complexity of the packaging products that enter the waste stream (Roosen et al., 2020).

Multilayer plastic films are frequently used in the packaging industry for the versatile benefits they offer. They are composed of various polymers (e.g., PE, PP, PET, PVC, PLA) or other materials (e.g., aluminum foil, paper). Through the use of different materials, various technical properties can be combined, like barriers, mechanical strength, and heat tolerance (Soares et al., 2022). For example, a PET layer is commonly utilized for its effective water and gas barrier in multilayer films, while PE and PP are chosen primarily for their superior sealing properties. PE, due to its frost resistance, is well-suited for packaging frozen foods. To enhance specific properties, poly(vinyl chloride) (PVC) or poly(vinylidene chloride) (PVDC) functions as odor and gas barriers (ideal for smoked food), PA adds rigidity or puncture resistance (beneficial for cheese packaging), and EVOH serves as an oxygen barrier (useful in meat and fish packaging). To achieve enhanced barrier capabilities, packaging may incorporate a metallized layer, as observed in chip packaging (Roosen et al., 2020).

The use of multilayers results in diminishing material uses and costs by reducing the average thickness of the packaging, insufficient use, and product losses (Soares et al., 2022). The difficulty multilayer plastics bring is that they cannot be recycled with traditional recycling technologies, like mechanical recycling, because of their complexity (Walker et al., 2020). Other recycling techniques are necessary to process multilayer plastics, such as compatibilization, delamination, and dissolution precipitation (Cabrera et al., 2022). When multilayer plastic is recycled together with single-material plastics, it is possible that this will result in low-quality recycled materials with inferior mechanical performance and physical appearance (Mahmood et al., 2021).

#### 2.3.1.2 Odorous Compounds

The presence of odorant compounds and volatile organic compounds (VOCs) is a critical concern related to PC plastics (Cecon et al., 2021; Zeng et al., 2023) that hinder its opportunities for high-end recycling (Roosen et al., 2022). These odorous compounds can originate from multiple sources. Odorants and other contaminants can migrate into the packaging material during the use phase, and thereby contaminating the waste stream (Strangl et al., 2017). Even after disposal, odorous components are created through microbial activity (Roosen et al., 2022). Also, the degradation products from the polymer material and additives present a source of odorous compounds (Zeng et al., 2023). More than 400 different VOCs have been identified and can be classified based on their molecular structure as hydrocarbons, aromatic compounds, flavor and fragrance compounds, alcohols, aldehydes, carboxylic acids, ketones, esters, ethers, amines, amides, sulfur, and halogenated compounds (Cabanes et al., 2020). Industrial washing processes that mostly just make use of cold or hot water or caustic solutions are not always able to remove these smelling fragments completely (Ügdüler et al., 2020). As a result, recycled materials are mainly used in low-grade outdoor applications (Strangl et al., 2020) rather than high-grade applications such as food packaging materials.

#### 2.3.1.3 Metals

Metals are expected to be very persistent in the plastic material after recycling, although lesser amounts might migrate during the use phase. Nowadays, a series of metals are purposefully added during the plastic production process. These



metals can come from colorants (e.g., Ti, Cr, Co, Cd, Pb, Zn, Fe, Al, Cu), stabilizers and antioxidants (e.g., Cd, Pb, Zn), or other additives (e.g., As, Li, Pb, Cd, Zn, Sb, Al). It is also possible that metals originate from catalysts used in the production phase (e.g., Sb, Ti, Cr, Hg, Mn), or are sorbed during the production, use, and waste management phases (e.g., Fe, Al, Cu, Mn, Zn, Ni). Because many of these metals are known to have toxic effects and/or are persistent and bioaccumulate, it is preferable to minimize the metals present in recycled plastic in order to reduce potential health risks and material quality loss. This finally results in lower applicability of the recycled plastic and thus less circularity (Eriksen et al., 2018).

#### 2.3.1.4 Halogens

Halogens, including fluorine, chlorine, bromine, and iodine, are elements frequently used in a variety of industrial and commercial applications to enhance material performance. In recent decades, synthetic halogenated compounds, particularly those containing fluoride, bromide, and chloride, have been used in all kinds of household items like plastic toys, waste electric and electronic equipment (WEEE), textiles, plastic packaging, and more (Zhang et al., 2013). One of the problems with halogens like chlorine is the corrosion of equipment. The primary source of chlorine in PPWoriginates from polymers that contain chlorine, such as PVC and PVDC (poly(vinylidene chloride)) (Roosen et al., 2022). The halogen bromine is often used in brominated flame retardants (BFRs). These BFRs are typically used in polymers for the construction sector, electric and electronic equipment, textiles, and furniture. BFRs are persistent by nature and have the trait of accumulating. They also show toxic effects and are carcinogenic (Pivnenko et al., 2017). The presence of these BFRs in WEEE has raised concerns about their waste management (Schlummer et al., 2007). Because not all WEEE is separately collected, some fraction ends up with the other HHW in the residual bin (Dimitrakakis et al., 2009b). Certain BFRs have been found in children's toys and food-contact plastics from the European market (Pivnenko et al., 2017). The presence of BFRs in food-contact plastics may be due to contamination from polymers derived from recycled WEEE plastics (Schlummer et al., 2007).

#### <span id="page-24-0"></span>2.3.2 Plastic Collection Scheme in the Netherlands: Source Separation vs. Post-separation

As discussed in section [2.2.3.1,](#page-18-0) there are two commonly used pathways for the collection of plastic packaging materials: source separation and post-separation. Each Dutch municipality may choose how plastic waste is separated (de Bruin et al., 2021), so the question increasingly arises if a switch from source separation to post-separation would be better in terms of circularity, costs, environmental pressure, and service level (de Bruin et al., 2021; Urgert IMC, 2021). An increase is already visible in the number of municipalities actually implementing post-separation. Since 2014, the share of post-separation has increased from 7 percent to 20 percent by 2021 (Gradus, 2021). In 2021, there were 31 municipalities that performed post-separation, and 101 partially used post-separation (NVRD, 2021). Since each of the two systems has its advantages and disadvantages, this brings some form of public discussion. This section will further compare the two systems.

In source separation, plastic waste is separated manually from other waste streams in households before collection (Groot et al., 2014; Nillesen et al., 2022). Due to variations in inner-city complexity, service quality, and financial incentives, source separation rates differ significantly between municipalities and even between neighborhoods (Nillesen et al., 2022). In post-separation, machines mechanically separate the plastic from MSW in a waste treatment center (Groot et al., 2014; Nillesen et al., 2022). Another possibility is to make use of a hybrid route that makes use of both source- and post-separation routes. A first separation takes place at the household level, and then the remaining residual waste is also sorted by machines to recover the small fraction of recyclable material present in this waste stream (Nillesen et al., 2022).



[Figure 2-9](#page-25-0) illustrates a simplified model of the PC plastic packaging network for the Netherlands in 2017 with net masses flows represented. The figure shows the path that plastic material follows and the amount of the specific fraction that is recovered.



Figure 2-9: Sankey diagram of the PC PPW recycling network in the Netherlands in 2017 (Brouwer et al., 2019)

#### <span id="page-25-0"></span>2.3.2.1 Recycling Rate

In [Table 2-2](#page-25-1) from Nillesen et al.(2022), the recycling rates for post-separation, source separation, and the hybrid pathway are given with their intermediate steps. For source separation and hybrid pathways, a distinction has been made between urban and rural, as this drastically affects recycling rates. For instance, for post-separation, 64% of plastic is separated from the MSW. The sorting plant has an efficiency of 90%, and at the recycling stage, it is 56%. This results in an average recycling rate of 32%.For source separation, there is a clear difference between an urban and rural area. Source separation in an urban area (16%) is only half the average recycling rate of post-separation, and in rural areas it is slightly higher at 35%. Overall, the hybrid, where the two pathways are complementary to each other, results in the highest average recycling rate.

<span id="page-25-1"></span>

#### Table 2-2: Recycling rates per plastic pathway (Nillesen et al., 2022)

These results [\(Table 2-2\)](#page-25-1) are in line with those of Dijkgraaf & Gradus (2020), who found that municipalities in 2014 only applied post-separation 8.39 kg of plastic waste per inhabitant was separated. For source separation, 6.88 kg of plastic waste per inhabitant was separated, and a hybrid system also resulted in the best performance with 9.76 kg per inhabitant.



The determining factor for the recycling rate of a source separation system is the participation rate of residents in the given area. In rural areas that use a curbside collection system, the participation rate can be higher than 70%. In urban areas with high-rise buildings, it is more difficult to get a high participation rate, and this remains around 30-40%. For a post-separation system, the determining factor is the post-separation installation in relation to the composition of the MSW and the implementation method. The greatest losses here occur in the sieving steps, where the smallest sieve size is between 35 and 65 mm. So smaller plastic packaging materials are lost. The sorting yield can be up to 70%. The sorting yield for post-separation is higher than for source separation, because it already went through a NIR-sorter. Then, for the recycling yield, source separation performance is better because there is more attached moisture and dirt on post-separation materials. Overall, both systems result in quite similar recycling rates depending on the collection area (Maaskant-Reilink et al., 2020).

#### 2.3.2.2 Quality

The more complexity and heterogeneity of the collected waste stream increases, the more difficult it is for the MRF to sort the plastic waste into fractions of high purity. This is due to more contaminants being present in the waste stream and because more cross-contamination can possibly occur. This dilemma is called 'the quantity-quality trade-off' where there is either more plastic waste collected and recycled of lesser quality, therefore perhaps not suitable for high-end applications, or vice versa (Brouwer et al., 2019).

On request of the municipality, Utrecht Maaskant-Reilink et al. (n.d.) wrote a report on the quality of recycled plastic material from source separation and post-separation and whether there is a difference between the two systems. In [Table 2-3](#page-26-0) an overview of five quality parameters is given of how both systems were compared to each other and what those parameters affected. The main findings are that post-separation performs better on polymeric contamination, and source separation results in less particle contamination.

<span id="page-26-0"></span>

Table 2-3: Differences and similarities between source and post-separation with regard to material quality (Maaskant-Reilink et al., 2020)

#### 2.3.2.3 Cost Model

In the work of Groot et al.(2014), the cost model for source- and post-separation was compared, and two collection schemes, curbside and drop-off collection, were also taken into account. It was found that for municipalities that



perform source separation, on average, the total costs per ton of plastic waste collected were twice as high as for postseparation municipalities.

A more recent study from Nillesen et al.(2022) also calculated the cost per ton of plastic waste collected [\(Table 2-4\)](#page-27-0). The gross cost includes those directly related to the specific pathway, and the opportunity costs are those related to the cost of using recycled plastic instead of virgin plastic or a shortage of energy if plastic is not incinerated. Those two together result in the total cost. It is clear that the hybrid pathway is the most expensive. This makes sense since both source- and post-separation steps are necessary. The source- and post-separation have a similar cost, but in general, source-separation has a lower gross cost. The additional cost of post-separation outweighs the higher collection costs associated with source separation. It is important to note that the recycling rate significantly impacts the level of costs at each step. Collecting, transporting, and processing costs are dependent on the mass of plastic at that stage of the process. If large amounts of unrecyclable materials are collected, then these will be sorted out but not processed further. These unrecyclable materials will either be incinerated or landfilled, which is cheaper than recycling. So, if the recycling rate is low, then the gross cost will also be more likely to be lower. But the goal is to recycle and not incinerate or landfill, so it might be more interesting to use another metric like cost per ton of recycled material [\(Figure 2-10\)](#page-27-1). In this situation, source separation in an urban area is almost twice as expensive as the other pathways.

<span id="page-27-0"></span>





<span id="page-27-1"></span>Figure 2-10: Cost per plastic pathway (*€* per ton recycled plastic waste) (Nillesen et al., 2022)



#### 2.3.2.4 Carbon Emissions

In the study of Nillesen et al.(2022), the carbon emissions per plastic pathway were also calculated. The gross emissions include the carbon dioxide emitted directly from the process, and the opportunity effect consists of two parts. First, the carbon emissions of virgin plastic that substitutes non-recycled material (positive value), and second, emissions from generating alternative energy instead of plastic incineration. The factor that influences carbon emissions the most is the recycling rate. A low recycling rate leads to a larger amount of plastic that ends up being incinerated and more production of replacement virgin plastics.



#### Table 2-5: Carbon emissions per plastic pathway (ton CO<sub>2</sub>/ton plastic waste) (Nillesen et al., 2022)

#### 2.3.2.5 When Source Separation and when Post-Separation?

When deciding which collection pathway is best, it involves some thoughtful considerations. It is a complex matrix in which the following facts must be taken into account: separation yields, recycling rates, costs, carbon emissions, area, compliance rates, and much more.

Source separation generally operates best in rural areas and when compliance rates are high. In this setting, there is more space for an extra waste bin, and it is less difficult to participate. It can be seen that source separation performance decreases in more urbanized areas. This setting becomes more interesting for post-separation, this both in terms of environment and economy(Dijkgraaf & Gradus, 2020; Nillesen et al., 2022). That is why large cities with lots of high-rise buildings like Amsterdam, Rotterdam, and Utrecht have decided to implement post-separation. But also, some more rural municipalities have changed to post-separation because there is a MRF nearby (Milieu centraal, n.d.).



# <span id="page-29-0"></span>2.4 Conclusion

The three most commonly used EOL options for PPW are landfilling, incineration with energy recovery, and recycling. Of the latter, it is predominantly mechanical recycling. There is great heterogeneity between different recycling strategies, but in general, each system consists of three steps: collection, sorting, and reprocessing. The focus of this dissertation is on the Dutch recycling model. In the Netherlands, three systems exist. First, there is the most commonly used source separation. Where waste is separately sorted at home and collected either by curbside collection or a drop-off point. Then there is post-separation, where sorting takes place after collection in a MRF. The third system is a deposit-refund system for large PET bottles.

The standard in the Netherlands is source separation via a PMD system. But there are quite a few municipalities that have already switched and are looking at switching to post-separation. It is difficult to say which system is best, as each has its advantages and disadvantages. Different aspects, like quantity, quality, and economics have to be taken into account. From a quantitative perspective, it largely depends on the area and the level of participation. In more rural areas where the participation level is high, source separation performs well. In more urban areas, the participation level is lower; therefore, a post-separation system is more suitable. In terms of quality, it is suggested that post-separation delivers a product of similar purity and quality.

In this master's dissertation, a detailed analysis will be performed on the waste stream composition and the elemental composition of PPW. This will help to have a better understanding of the possible contaminants present in the waste streams. It will be examined whether differences can be observed between source separation and post-separation. Also, specific target products from both pathways will be investigated.This will be complemented with an MFA of the sorting plant, for which a Sankey diagram will be established. V



# <span id="page-30-0"></span>3 MATERIALS AND METHODS

#### <span id="page-30-1"></span>3.1 Material Flow Analysis

The MFA was made .using a model in Microsoft Excel developed by researchers at Ghent University. The first step was making a flowsheet based on the 'De Wijster' sorting plant. The flowsheet in [Figure 3-1](#page-30-2) represents the sorting process for waste collected through source separation. To make the flowsheet first all the sorting equipment must be defined and connections between the sorting equipment must be determined.



Figure 3-1: Flowsheet Attero sorting plant with process steps (blue), sorted bales (green), other fractions (yellow), and residual (red)

<span id="page-30-3"></span><span id="page-30-2"></span>The second step was to define the incoming waste stream or feedstock. A total input of 50,000 tons per year was chosen. I[n Table 3-1](#page-30-3) a list of all the different waste categories that occur in the stream are listed with the composition and the relative mass each waste category represents.



#### Table 3-1: Composition incoming waste stream





The third step was creating a sorting matrix. In this matrix for each waste stream the sorting efficiencies are filled in per sorting equipment. For example, for the waste stream HDPE Bottles goes through the NIR PET (+) then 1.0% is send to NIR PET Bottles and 99.0% is send to NIR PE Rigids (+). So, most of the mono PE goes further down in the cascade of NIR's. When it passes the NIR PE Rigids (+) 10.0% is sent to the NIR PP Rigids (+) and 90.0% to NIR PE Rigids (-). Here the largest fraction goes out the cascade and goes through a negative NIR sorter for PE and finally along a manual sorting conveyer to end up in the PE Rigids bale. This process is done for all the waste streams with all the sorting equipment. To determine the sorting efficiencies used for developing this matrix a sorting database and expert judgement was used. This sorting database was not built in this study but provided out of previous work among (Brouwer et al., 2018; Cimpan et al., 2015; Kleinhans et al., 2021, 2022; Roosen et al., 2022).

When all the sorting efficiencies are filled in the model calculates the mass balance of which the mass flows were visualized in a Sankey diagram.



# <span id="page-32-0"></span>3.2 Sample Taking and Preparation

The sorted plastic waste samples were obtained at the sorting plant 'De Wijster' of Attero located in the Netherlands. Attero is a leader in the field of resource recovery and renewable energy. The company was founded in 1929 under the name Vuil Afvoer Maatschappij. Today, Attero is specialized in the processing of household, organic, and mineral waste. Attero counts 718 employees, 18 locations, processed 3.5 million tonnes of waste, produced 840 Gigawatts-hours, and in 2022 the turnover was €340 million. Some of the core activities of Attero Wijster is waste sorting and recycling (Attero // Energiek Met Milieu, n.d.).

At 'De Wijster', Attero lightweight PPW is sorted out of commingled source separated and mixed post-sorted PPW streams. Samples were taken from both streams. In April 2023, samples were taken from plastic waste that was separately collected, i.e. source separated. In June 2023, samples were taken from plastic waste that was collected in a mixed collection system, i.e. post-sorted. A major part of the collected samples were PPW made out polyolefins (PO).

During both sampling campaigns, target and random samples were collected from four different collection points. Three collection points, HDPE/PE rigids, PP rigids, and mixed plastic waste (i.e. DKR350) were taken at the manual sorting belt prior to baling. LDPE, also known as PE flexibles, samples were collected by Attero employees because for LDPE there is no manual sorting belt at the sorting plant. Therefore, there is only a random sample of this stream and no target samples. Selection of target samples was based on their frequency of occurrence on the sorting belt prior to baling. These samples included various products such as shampoo bottles, butter trays, yogurt trays, packaging foils, among others. Selection aimed to create a representative fraction of each waste stream. For each waste stream category, a minimum of 10 products were collected, comprising 5 from the source separated collection system and 5 of the same products from the post-separated system. This was possible for the HDPE and PP rigid stream, for mixed plastic waste samples similar items were selected. In a later stage each item was decomposed into itsindividual parts(e.g. a shampoo bottle in bottle, cap, and label). For some products specific parts were absent in one of the two collection systems.

In addition to the target items, random samples were also obtained from the same three waste stream categories and also from LDPE. For random samples, one 60-liter bag of waste was collected from each of the HDPE, PP rigid, and mixed plastic waste belts prior to baling, from both the source separated and post-sorted collection systems. The collection of the LDPE samples was done by the Attero employees. The random samples were taken to represent the entire waste stream category.

Finally, five special samples (e.g. plastic toys, CD case, blister packaging,…) were collected that occurred mainly at postseparated waste. These items were analyzed to determine potential disparities with standard PPW found in source separated systems, thereby possibly affecting the composition of the waste stream.

In total 73 samples were obtained, comprising 15 random samples, 53 target samples, and 5 special samples. A comprehensive list of all samples is provided in [Appendix A.](#page-63-1)

Before further sample preparation, all samples underwent a washing process in order to remove all residues. However, a fraction of unwashed random samples waskept. Very dirty samples were manuallycleaned prior to the washing cycle. For washing a standard washing machine was used with tap water at a temperature of 60 °C. The residue level was determined by weighing the samples before and after washing and subsequent drying [\(1\)](#page-33-0).



<span id="page-33-0"></span>The final step of the sample preparation involved size reduction of all samples, this applies to both washed and unwashed samples. Initially, products were shredded using the SH Granulator fromShini. Between each shredding cycle, the shredder was cleaned to remove any potential sample residue. Subsequently, certain items underwent further size reduction using the Pulverisette 19 from Fritsch, utilizing a sieve cassette with 4 mm square perforation. Again, cleaning was performed after each cycle. For further size reduction cryogenic grinding was applied using a ball mill from Anton Paar BM500 with liquid nitrogen for the cooling. This obtained a homogeneous powder from the samples.



## <span id="page-34-0"></span>3.3 Quantification of the Polymer Composition

For polymer identification, plastics were analyzed using attenuated total reflectance fourier-transform infrared spectroscopy (ATR-FTIR). ATR-FTIR analyses were carried out on a Nicolet™ iS20 FTIR Spectrometer. Each sample was analyzed on both sides. If both sides of the packaging material were identified to be the same polymer, then the material was assumed to be mono material. Otherwise, if different polymers were identified, the material was assumed to be multi-material.

The analysis of the target samples consisted of 32 scans with a resolution of 4 cm<sup>-1</sup>, and spanning the wavenumber region from 400 to 4000 cm<sup>-1</sup>. Each target sample underwent one analysis. The weight of each individual product part was measured, which was utilized to calculate the mass percentage of the respective product in question. To determine the composition representing the three waste stream categories, the average mass percentage was calculated based on the five selected products that represent each waste stream category.

The random samples were analyzed using the preview function. Within each category of random samples, 100 flakes were analyzed. After scanning, the material was segregated by polymer type so that the weight of each polymer type can be determined. This process was carried out in triplicate to calculate the average weight of each polymer type (mpolymer) by which the polymer composition can be determined and the bale quality.

The mass fraction ( $w_i$ ) of a polymer type within a item or waste stream is the ration  $w_i$  of the mass  $m_i$  of that substance to the total mass  $m_{tot}$  of that item or waste stream (equation [2\)](#page-34-1). The polymer composition was obtained by calculating the mass fraction of each polymer type within the item or waste stream. The bale quality equals the mass fraction of the desired polymer type in the corresponding waste stream category of the random samples.

$$
w_i = \frac{m_i}{m_{tot}}
$$

<span id="page-34-1"></span>2

# <span id="page-35-0"></span>3.4 Quantification of the Elemental Composition

#### <span id="page-35-1"></span>3.4.1 Metal Contents

The metal content was determined by performing inductively coupled plasma optical emission spectroscopy (ICP-OES). This analysis comprised all samples, including random samples, target samples, and special samples. Sample preparation involved acid digestion utilizing the Multiwave 5000 microwave from Anton Paar. First, 0.2±0.01 g of sample was weighed into Teflon vessels, followed by the addition of 2 mL of double-distilled water and 8 mL of 65% nitric acid (HNO<sub>3</sub>). Subsequently, the samples were heated in the microwave for 40 minutes up to 200 °C maintaining this temperature for 20 minutes. The resulting liquid was diluted with double-distilled water to a volume of 25 mL. The samples were then filtered using a CHROMAFIL syringe filter with a regenerated cellulose membrane with a 0.45 µm pore size.

Prior to measurement, a standard series was prepared ranging from 0 to 50 ppm. For this, the CERTIPUR multi-element standard solution IV was used complemented with single element standard solutions for As, Sb, Se, Ti, and V. The ICP-OES analyses were performed using a Thermo Scientific iCAP 7000 model, equipped with a CELTAC AXP-560 autosampler and Qtegra Software. [Appendix B](#page-65-0) provides a comprehensive list of the analyzed elements, the emission lines, and viewing modes. Each sample was prepared in triplicate and analyzed after which the average could be calculated. Due to the large number of samples to be analyzed the measurement was spread across eight days.

For the metals, a Mann-Whitney U test was performed to determine if there is a significant difference in metal concentration between the two collection methods for each metal separately per bale category [\(Appendix H\)](#page-73-0). The Mann-Whitney U test, also known as the Wilcoxon rank-sum test, is a nonparametric statistical test that is used to determine if there are differences between two groups that are not normally distributed.

#### <span id="page-35-2"></span>3.4.2 Halogen Contents

The halogen content was determined by ion chromatography (IC). The halogens in question are fluorine, chlorine, and bromide. Prior to the IC analysis, the samples were pretreated through microwave-induced oxygen combustion using the Multiwave 5000 from Anton Paar. Initially, the pulverised samples were pelletised into pellets of 0.02±0.01 g each. This was done to ensure that the sample remained intact within the vessels on the carrier glass where the combustion takes place, facilitating controlled reaction processes. An ashless filter paper was placed on the carrier glass, and 50 µL of 50 m% ammonium nitrate was added to initiate the combustion. Into the vessels 8 mL of the aqueous solution of 1  $m$ M NaHCO<sub>3</sub> and 8 mM Na<sub>2</sub>CO<sub>3</sub> was added to collect the vapours.

Before starting the microwave-induced oxygen combustion the vessels containing the samples were put on a pressure of 20 bar that builds up to a maximum of 65 bar and 225 °C within 10 minutes during the combustion process. The resulting solution after combustion was diluted with double-distilled water to 25 mL. Subsequently, the samples were filtered using a CHROMAFIL syringe filter with a regenerated cellulose membrane with a 0.45 µm pore size.

The IC analysis were performed on an Eco IC system from Metrohm equipped with the 863 Compact IC Autosampler. All samples were analysed once. Measuring in triplicate was not possible due to time constraints caused by a failure of the oxygen combustion equipment.


#### 3.4.3 CHNSO Analysis

For the analysis of CHNSO a ThermoScientific Flash 2000 Organic Element Analyzer was used. Before measurement a standard curve wase prepared by using a CHNS Elemental MicroAnalysis Standard (B2044). Analysis was done for each sample in three repetitions for homogeneous samples and five repetitions for heterogeneous samples. The element oxygen was not analysed because for the measurement of oxygen other settings for the instrument are necessary. Instead, the concentration of oxygen was calculated using a subtraction method. The instrument gives the mass percentage of each element, so the sum of carbon, hydrogen, nitrogen, sulphur, and oxygen should be a hundred percent. It may be assumed that the mass percentage of oxygen equals a hundred minus the sum of the measured mass percentages of the other elements.

For each sample run between 2 and 3 mg of sample was used. Sample preparation included pulverisation of the plastic and sieving with a 425 µm sieve.



## 4 RESULTS

### 4.1 Quantification of the Polymer Composition

In total, 73 samples were analyzed. Of these samples, 15 items were target items but these target items included items with several parts, e.g. a bottle with a cap and label equaled 3 items of one sample. Of those composite samples, seven items showed a heterogeneous polymer composition and eight had a homogeneous polymer composition. A heterogeneous polymer composition usually occurred in HDPE bottles where the separable parts (e.g. caps and labels) were made from a different polymer as shown i[n Figure 4-1](#page-37-0) for a shampoo bottle. Another common situation is for the films from the mixed plastic stream, where heterogeneity occurs in the form of multilayer plastics.

[Figure 4-1](#page-37-0) exemplifies an ATR-FTIR spectrum, which shows the distinctive composition of the three constituent parts comprising a shampoo bottle. Each part consists of a unique polymer type, namely PE constituting the bottle, PP comprising the cap, and polyester for the label.



Figure 4-1: ATR-FTIR spectrum parts shampoo bottle with bottle (red), cap (green), and label (blue)

<span id="page-37-0"></span>[Figure 4-2](#page-38-0) illustrates the polymer composition and residue levels for each waste category (e.g., HDPE Bottle, PP Tray, and Mixed Foil). This was determined by first calculating the average mass of each individual product part per polymer type of the five respective target samples for each waste stream category. Then, the total masses including the residue of the product items were 68.70 g for HDPE Bottle, 49.81 g for PP Tray, and 27.96 for Mixed Foil. The composition was obtained by applying equatio[n 2.](#page-34-0) For example, the mass fraction of PP within the HDPE bottles category was calculated as followed.

$$
w_{PP} = \frac{5.04 \, g + 4.24 \, g + 3.70 \, g + 3.70 \, g}{4}}{68.70 \, g} = 6.07\%
$$



There was no distinction made between the source separated and post-sorted streams for this analysis, because it is assumed that the same product items would result in the same polymer composition for both streams. The graph in [Figure 4-2](#page-38-0) is made with data collected from mainly source separated waste complemented with data from the postsorted waste stream for product parts that were missing in the source separated stream (e.g., caps, labels). Detailed polymer composition data for the target samples is provided i[n Appendix C.](#page-66-0) Based on the results of this study the level of homogeneity for HDPE bottles is 82.63% and for PP trays 72.08%. In case of mixed foils out the mixed plastic waste stream the largest share is a multilayer plastic with PE and PA on the outside with 51.86%, followed by a multilayer plastic of PE and PP with 25.39%, and 18.49% PP foils.

In examining the residue levels of the target samples, it was observed that PP trays exhibited the highest residue percentage at 15.15%, followed by HDPE bottles at 8.11%, while mixed foils registered the lowest at 4.26%. This analysis primarily utilized source separated waste to ascertain the composition of the target samples. Consequently, when comparing target samples to random samples, the focus should be on those that were source separated. However, the residue levels of the random samples do not fall in line with the target samples. In the mixed plastic waste stream, the residue level was the highest at 12.79%, followed by PP at 12.09%, then PE at 9.90%, and at LDPE with 9.20%.



<span id="page-38-0"></span>Figure 4-2: Polymer composition and residue level for the target items separated for each part of the product in m% (se[e Appendix C](#page-66-0) for detailed data for each product part)

[Figure 4-3](#page-39-0) displaying the four waste categories of the random samples: PE, LDPE, PP, and Mixed. For each waste category the source separated and post-sorted streams were compared. Plastic flakes were categorized as mono- or multimaterial based on whether both sides of the flake were the same polymer or different polymers, respectively. The identified mono-materials included PET, PE, PP, PS, PA, PUR, Al foil, and Polybutadiene. For comparison multi-materials were defined as a 'main' polymer group, i.e. PET, PE, PP, PS, and an 'other' group which differed for each main polymer group. The materials that form a multi-material with PET include PE, PP, PS, and PUR; with PE, it includes PP, PS, Cellulose, PA, PEA, PEVA, Polyacrylate, and PUR; with PP, it includes PS, PUR, and Polyacrylate; and with PS, it includes Polyacrylate and PA. Detailed polymer composition data for the random samples is provided i[n Appendix D.](#page-67-0)



From the results presented i[n Figure 4-3,](#page-39-0) the following things can be noted. For each bale, the main polymer type is the most abundant, e.g. PE in the PE bale. The mixed plastic waste bale is the most heterogeneous one but exists mostly out PO. For every waste category the residue level is always higher when post-sorted than source separated.



<span id="page-39-0"></span>Figure 4-3: Polymer composition and residue level for the random samples of the four waste stream categories for both source separated (S) and post-sorted (P) schemes

The Fisher's Exact Test was conducted to determine if there was a significant association between the polymer composition of the waste stream categories and the sorting method [\(Appendix E\)](#page-68-0). This is a non-parametric test to determine if there are differences between two or more groups with categorical data. There was chosen for a Fisher's Exact Test instead of a chi-quadrate test because for some frequenciesthe values are less then five. Therefore, a Fisher's exact test is more reliable.The amount of polymer types from in [Figure 4-3](#page-39-0) was further reduced to the largest fractions i.e. PET, PE, PE/other, PP, PP/other, residue, and other.

For PE the test resulted in a p-value of 0.07209. This is greater than the significance level of 0.05, indicating that the null hypothesis must be accepted. This means there is not sufficient evidence to conclude that there is a significant association between the polymer composition of the bales and the sorting method. The p-value for PP is 0.09608 and for Mixed 0.09608. For these waste streams there is also not a significant association between the polymer composition and the sorting method. For LDPE the test resulted in a p-value of 0.01116. Which is smaller than the significance level of 0.05, so the null hypothesis must be rejected and the alternative hypothesis accepted. This means that for the LDPE waste stream there is a significant difference in composition between the two sorting methods.



### 4.2 Quantification of the Elemental Composition

The elemental composition was determined for the target samples, the random samples, and the special samples. The elements analyzed include organic elements ( C, H, N, S, and O) in mass percentages (wt%), metals ( Al, AS, Ca, Cd, Co, Cu, Fe, K, Li, Na, Ni, Pb, Sb, Se, Ti, and V) in mg/kg, and halogens ( F, Cl, and Br) also in mg/kg.

In [Figure 4-4,](#page-41-0) [Figure 4-5,](#page-42-0) and [Figure 4-6](#page-42-1) the average composition of the three waste streams of the source separated target samples are presented. The table underneath each chart gives the total element concentration. The detailed elemental composition on product part level can be found in [Appendix F.](#page-70-0)

For target samples the CHNSO-analysis the element S was never detected, this will also be the case for the random samples and the special items. The total metal contents are 2244 mg/kg for HDPE bottles, 10018 mg/kg for PP trays, and 2910 mg/kg for mixed foils. The metals that make up more than 90% of the total metal content for each waste stream are Al, Ca, Fe, Na, and Ti.

The element composition of the random samples is presented i[n Table 4-1.](#page-43-0) The rows give the specific samples, and the columns give the elements. The analysis was both done for the samples that were washed and those that were unwashed. The later one does still contain residue. There was no mixed source separated unwashed sample available. For all other source separated and post-samples there was a washed and unwashed sample available. In line with the target samples, the metals Al, Ca, Fe, Na, and Ti take up most of the total metal content. The total metal contents for the source separated washed samples are 1616 mg/kg for PE, 6561 mg/kg for PP, 9964 mg/kg for LDPE, and 8754 mg/kg for mixed plastic waste. For mixed plastic waste the concentration Ca extended the upper limit of quantification (ULOQ), therefore a value of 7500 mg/kg was added to compensate. Then for the post-sorted washed samples the total metal contents are 6745 mg/kg for PE, 2375 mg/kg for PP, 9192 mg/kg for LDPE with the concentration of Ca extending the ULOQ so 7500 mg/kg was added to compensate, and 14265 mg/kg for mixed plastic waste. So, the metal content is higher in the case of the post-sorted samples, except for the PP bale. Overall, the bales of LDPE and mixed plastic waste contain the highest metal content. For some samples the concentration Ca exceeded the ULOQ, for those instances a value of 7500 mg/kg was added because this was the highest value measured in other samples.

I[n Figure 4-7](#page-44-0) a detailed bar chart is presented for As, Cd, and Pb visualizing the average concentration comparing source separated and post-sorted plastics for the four categories. These three metals are prohibited in terms of migration in food contact materials. I[n Appendix G](#page-71-0) these charts are presented for all the analyzed metals.

The results of these tests are presented in [Table 8-10,](#page-73-0) listing the p-values. For the PE bale the metals As and Sb only were observed in the post-sorted waste. For the metals Ca, Cd, Cu, and V there was a significant difference, with the highest concentrations for post-sorted waste. In contrast to Li, where a higher concentration was set for source separation. For the PP bale the metal As was only observed in the post-sorted waste. For the metals Cd, and Li there was a significant difference, with the highest concentration for post-sorted waste, in contrast to Ca, Co, Cu, Fe, and V. For the LDPE bale the metals As and Sb were only observed in the post-sorted waste. For the metals Al, Cu, and Fe there was a significant difference, with the highest concentration for post-sorted waste. For the mixed plastic waste bale, the metals Sb and Se were only observed in the post-sorted waste. For the metals Cu, Fe, Ni, and V there was a significant difference, with the highest concentration for post-sorted waste.



For the five special samples the elemental composition can be found in [Table 4-2.](#page-45-0) These special samples originate from the post-sorted plastic waste stream, namely the PP and mixed plastic belt. In the CHNSO-analysis for the blister packaging the oxygen concentration56.61 wt%, is remarkable higher compared with the samples of the PP and mixed plastic waste samples. The blister packaging, as could be expected has a high concentration of 51240 mg/kg Al. When looking at the metals that are totally unwanted like As, Cd, and Pb. For As, it occurs more often in the special items at higher concentrations compared to PP and mixed random samples. Cd concentrations are relatively similar. The metal Pb can be found in both the random samples and the special items but the concentrations in the special items are much higher.



<span id="page-41-0"></span>Figure 4-4: Elemental composition PE bottle







<span id="page-42-0"></span>

Figure 4-6: Elemental composition foil

<span id="page-42-1"></span>

<span id="page-43-0"></span>

|          | nr. | Sample                 |       |       | Concentration (in wt%) |      |       | Concentration (in mg/kg) |         |       |       |      |     |       |      |       |      |     |       |           |         |     |      |     |     |     |
|----------|-----|------------------------|-------|-------|------------------------|------|-------|--------------------------|---------|-------|-------|------|-----|-------|------|-------|------|-----|-------|-----------|---------|-----|------|-----|-----|-----|
|          |     |                        | C     | H     | N                      | S    | 0     | Al                       | As      | Ca    | Cd    | Co   | Cu  | Fe    | K    | П     | Na   | Ni  | Pb    | <b>Sb</b> | Se      | Ti  | v    | F   | Cl  | Br  |
|          |     | <b>PE (S)</b>          | 84.22 | 15.39 | 0.00                   | 0.00 | 0.39  | 134                      | $<$ LOD | 815   | 0.18  | 0.71 | 2.4 | 276   | 19   | 0.22  | 257  | 1.2 | 1.5   | $<$ LOD   | 2.8     | 106 | 0.30 | 0.6 | 0.5 | 0.0 |
|          |     | PP(S)                  | 82.94 | 15.26 | 0.03                   | 0.00 | 1.77  | 2170                     | $<$ LOD | 2694  | 0.05  | 0.81 | 16  | 1305  | 15   | 0.74  | 264  | 1.3 | 3.0   | $<$ LOD   | $<$ LOD | 90  | 2.5  | 0.7 | 0.5 | 0.0 |
|          |     | LDPE (S)               | 79.83 | 14.50 | 0.08                   | 0.00 | 5.59  | 1001                     | $<$ LOD | 7507  | 0.29  | 0.59 | 22  | 547   | 89   | 0.25  | 676  | 2.2 | 6.6   | $<$ LOD   | 3.8     | 107 | 1.6  | 1.0 | 1.6 | 0.0 |
|          |     | Mixed (S)              | 72.81 | 12.34 | 0.01                   | 0.00 | 14.84 | 590                      | $<$ LOD | >ULOO | 0.14  | 1.2  | 27  | 343   | 76   | 0.19  | 78   | 1.1 | 1.3   | $<$ LOD   | $<$ LOD | 135 | 0.92 | 0.3 | 0.6 | 0.0 |
| Washed   |     | PE(P)                  | 84.12 | 15.59 | 0.00                   | 0.00 | 0.30  | 154                      | 0.76    | 5978  | 4.4   | 0.67 | 7.2 | 354   | 16   | 0.045 | 73   | 6.2 | 1.2   | 0.63      | 11      | 137 | 0.68 | 0.3 | 0.5 | 0.0 |
|          | b   | PP <sub>(P)</sub>      | 81.72 | 15.11 | 0.00                   | 0.00 | 3.17  | 163                      | 0.54    | 1737  | 0.37  | 0.45 | 5   | 256   | 25   | 1.8   | 67   | 1.1 | 2.3   | $<$ LOD   | $<$ LOD | 114 | 0.84 | 1.5 | 0.5 | 0.0 |
|          |     | LDPE (P)               | 81.13 | 14.80 | 0.00                   | 0.00 | 4.07  | 448                      | 0.24    | >ULOQ | 0.10  | 0.53 | 43  | 826   | 100  | 0.24  | 121  | 2.5 | 5.4   | 0.49      | 7.4     | 136 | 1.3  | 1.1 | 0.7 | 0.0 |
|          | 8   | Mixed (P)              | 80.83 | 14.82 | 0.09                   | 0.00 | 4.26  | 521                      | $<$ LOD | 2126  | 0.05  | 1.3  | 49  | 11012 | 82   | 0.18  | 328  | 6.9 | 13    | 1.7       | 15      | 106 | 2.5  | 0.8 | 0.7 | 0.0 |
|          |     |                        |       |       |                        |      |       |                          |         |       |       |      |     |       |      |       |      |     |       |           |         |     |      |     |     |     |
|          | 9   | <b>PE (S)</b>          | 81.10 | 14.41 | 0.00                   | 0.00 | 4.49  | 303                      | $<$ LOD | 5247  | 6.3   | 1.2  | 22  | 1209  | 155  | 0.29  | 229  | 4.4 | 3.1   | 0.61      | 16      | 20  | 4.2  | 0.2 | 0.7 | 0.0 |
|          | 10  | PP(S)                  | 82.82 | 14.99 | 0.00                   | 0.00 | 2.19  | 301                      | $<$ LOD | 1849  | 0.16  | 1.4  | 8.1 | 1159  | 158  | 4.1   | 249  | 3.2 | 1.9   | 0.50      | 12      | 29  | 1.6  | 0.2 | 0.6 | 0.0 |
|          |     | LDPE (S)               | 64.65 | 11.33 | 0.46                   | 0.00 | 23.55 | 3040                     | $<$ LOD | >ULOQ | 0.24  | 3.1  | 195 | 6901  | 2283 | 2.7   | 1909 | 20  | 82    | 5.5       | 94      | 111 | 11   | 0.6 | 2.4 | 0.0 |
| Unwashed | 12  | <b>PE</b> ( <b>P</b> ) | 73.51 | 12.84 | 0.27                   | 0.00 | 13.37 | 1846                     | $<$ LOD | 5851  | 0.10  | 1.4  | 64  | 4556  | 860  | 1.3   | 868  | 8.2 | 18    | 1.8       | 27      | 75  | 4.9  | 0.4 | 1.4 | 0.0 |
|          | 13  | PP(P)                  | 82.08 | 14.78 | 0.00                   | 0.00 | 3.14  | 230                      | $<$ LOD | 1493  | 0.028 | 0.38 | 22  | 263   | 88   | 0.13  | 365  | 0.5 | 0.37  | $<$ LOD   | $<$ LOD | 111 | 0.28 | 0.1 | 2.3 | 0.0 |
|          | 14  | LDFE(P)                | 82.95 | 14.99 | 0.00                   | 0.00 | 2.06  | 384                      | $<$ LOD | 2633  | 0.030 | 0.35 | 10  | 314   | 31   | 1.1   | 84   | 0.6 | 0.082 | $<$ LOD   | $<$ LOD | 53  | 0.75 | 0.0 | 0.9 | 0.0 |
|          | 15  | Mixed (P)              | 72.39 | 12.32 | 0.30                   | 0.00 | 15.00 | 1631                     | $<$ LOD | >ULOQ | 0.16  | 1.2  | 38  | 2128  | 1080 | 1.8   | 1123 | 6.3 | 15    | 0.39      | 14      | 70  | 5.0  | 0.2 | 1.6 | 0.0 |
|          |     |                        |       |       |                        |      |       |                          |         |       |       |      |     |       |      |       |      |     |       |           |         |     |      |     |     |     |

Table 4-1: Elemental composition random samples





<span id="page-44-0"></span>Figure 4-7: Column chart visualizing the average concentration of the prohibited metals in terms of migration in food contact materials for both the source separated and post-sorted plastic waste per waste categories. The average concentration, the blue dots represent each repetition, and error bars represent the standard deviation. (A) Arsenic, (B) Cadmium, and (C) Lead

<span id="page-45-0"></span>

#### Table 4-2: Elemental composition special samples

### 4.3 Material Flow Analysis

The results of the MFA were plotted in a Sankey Diagram [\(Figure 4-8\)](#page-46-0). The Sankey Diagram illustrates how the different materials behave in each sorting step. For each bale category the total output, bale quality and recovery rate are included. A detailed overview of the mass balance is provided in [Appendix I.](#page-74-0)



<span id="page-46-1"></span><span id="page-46-0"></span>Figure 4-8: Sankey Diagram Material Flow Analysis Attero Sorting Facility

## 5 DISCUSSION

### 5.1 Quantification of the Polymer Composition

In this study the polymer composition of target plastic packaging products was determined through FTIR-spectroscopy complemented with random samples of the same waste stream categories and a random sample of LDPE additionally. [Figure 4-2](#page-38-0) shows the average polymer composition for the target samples including the residue level. This average is a unique representation for the selected products. Only five products per waste category were analyzed. Because of the limited selection, this result cannot be considered as a representation of the entire bale. In [Appendix C](#page-66-0) the composition can be found per representative product and product part. I[n Figure 4-3](#page-39-0) the polymer composition is given for the random samples. For these samples there was a distinction made between source separated and post-sorted plastic waste. In [Appendix D](#page-67-0) the masses for each material type that occurs in the waste stream is presented. It is important to note that there exists a disparity in the management of waste between the two collection schemes by Attero at 'De Wijster'. Source separated waste is in general sorted shortly after arrival, while post-sorted waste is gathered in a storage facility until sufficient quantity is present to change the sorting operations from source separated too post-sorted. This disparity could possibly affect the residue that is left on the plastic material. It was visually observed that source separated items generally contained a higher moister content compared to post-sorted, possibly due to prolonged storage leading to residue release. Contrarily, post-sorted items tended to be dirtier and harder to clean, potentially because they are in a further stage of spoilage and mold formation. While one washing cycle sufficed for sourceseparated waste, one to three cycles were necessary for post-sorted waste, depending on the level of contamination present.

It should be noted that only FTIR-spectroscopy was performed to determine the polymer composition of the samples. So, there is only information about the material on the inner and outer layer of the samples. In the case of multilayers, the separate layers between were not identified. Therefore, there is no information of the mass of each polymer type in the multilayer plastic. This could be determined by the layer thickness, surface area of the product, and the density of the specific polymer type. To obtain this information FTIR analysis could be expanded with polarized optical microscopy (POM) and differential scanning calorimetry (DSC).

The polymer compositions represented i[n Figure 4-2](#page-38-0) are in line with the results of Roosen et al., (2020) for HDPE bottles. The proportion of each product part in the total mass of the respective product remains consistent. The most notable difference for HDPE bottles lies in the caps material. In the study of Roosen et al., (2020), the cap is almost exclusively made of PP, whereas in this study, the cap in two out of five times is made of PE. Utilizing the same polymer for the cap as the bottle simplifies the recycling process. This might indicate that some companies are beginning to implement the principles of design for recycling by minimizing the use of different materials within their product. Due to this the overall bale quality is slightly higher. For the PP trays in the study of Roosen et al., (2020) more different materials were observed then in this study. In this study the bale quality for the PP trays is 72.08%, 12.77% is a combination of PP with cellulose propionate and the remaining 15.15% is residue. Cellulose propionate is an organic cellulose ester (Gilbert, 2017). Cellulose esters have good film forming and binding properties. These cellulose derivates are utilized for practical applications in food packaging, such as membranes, edible films or paper coatings (Nechita & Iana-Roman, 2020). What was also remarkable is that for sample numbers 32, 34, 36, and 37 PS was identified on the outside of the PP trays but only where printing was present [\(Appendix A\)](#page-63-0). A multilayer packaging material made out of PP and PS is uncommon combination. The use of PS is probably not to enhance the barrier properties of the PP tray, but for the printable outside



layer of PS (European Commission et al., 2016). The last waste stream category mixed foils, mainly containing plastic films, is the most complex stream. In [Figure 4-2](#page-38-0) there are three material combinations found, namely PP, PE/PP, and PE/PA. There is a great possibility that for all these films there are other materials between those identified outer layers. Three of the five mixed foils samples have both PP on the in- and outside layer, one sample has PA on the inside layer and PE on the outside layer, and one sample has PP on the outside layer and PE on the inside layer. In the study of (Roosen et al., 2020) some other layers often seen in packaging materials were identified, including PET, ethylene vinyl alcohol (EVOH), PV(D)C (polyvinylidene dichloride), aluminum (Al), polyurethane (PUR), ethylene-vinyl acetate (EVA), and paper. Layers of EVOH, PV(D)C, EVA, and Al are mostly used as functional layers to improve the barrier functions. EVOH has good oxygen barrier properties but needs to be protected against moisture therefore it often sandwiched between PE or PP. PV(D)C has gas, aroma, moisture barrier functionalities, protects surfaces against scratches and abrasion, and is used for heat-sealable food contact layer. EVA is used for its moisture barrier properties, heat-sealable food contact layers, or as adhesion layer between polar and non-polar polymers. Al is often used in packaging materials for its effective barrier against UV light, moisture, air, and odors. Polyurethane is mostly used as adhesive in a tie layer (Anukiruthika et al., 2020; European Commission et al., 2016). In the results of the random samples some of these materials are found, namely PET, PUR, PEVA, and Al. Also, polyacrylate is regularly identified. Acrylates are used as adhesive and in printings or coatings (European Commission et al., 2016). All these different layers of polymers and other materials provide ideal functional properties, but also bring challenges in terms of recycling multilayer plastics. Chemical recycling is the most promising pathway to breakdown these complex materials. This is through different techniques, such as delamination, hydrolysis, glycolysis, aminolysis, methanolysis, catalytic depolymerizations, and enzymatic catalysis (Tamizhdurai et al., 2024).

There was a notably lower residue level in target mixed foils, compared to the high residue level in the random mixed stream, this may be attributed to the weak adhesive nature of the residue, which on a two-dimensional structure such as most foils facilitate in the detachment of residue while taking specific product from the stream. In contrast, the three-dimensional structures of PE bottles and PP trays trap residue within. While handling the samples this was visually observed, e.g. shampoo bottles contained excess shampoo and butter trays still contained butter. Therefore, when taking a product from these streams, still a substantial amount of residue is concurrently collected. This mechanism may also explain the generally higher residue levels in random samples compared to target samples, except PP trays, as loosely attached residue is not accounted for in the target samples.

When the polymer purity of the target samples is compared with the bale quality of the random samples there is no clear trend between the two. In case of the HDPE bottles the polymer purity is 82.63% and 69.26% for the random PE sample bale quality. For PP trays it is 72.08% and for the random PP sample 84.29%. The random samples bale quality is lower then mandated by the DKR specifications where for PE and PP bottles, cups, and trays of a volume smaller than 5 liters the bale purity must be equal or higher than 94.0% (Name Rohstofffraktionsspezifikation 324-0 Polypropylen, n.d.; Name Rohstofffraktionsspezifikation 329 Polyethylen, n.d.). The DKR will not separate, for example a PE bottle in its separate parts like in this study but counts the whole bottle including cap and label as PE. This is why the bale quality according to the DKR specifications might be an overestimation.

The polymer composition of the random samples is presented in [Figure 4-3.](#page-39-0) To identify if there was a significant association between the polymer composition of the waste stream categories and the sorting method a Fisher's Exact test was conducted. The test results states that there is no sufficient evidence to conclude that there is a significant



association between the two variables for the categories PE, PP and mixed plastic waste. For the LDPE streams there is a significant association between polymer composition and sorting method.

For each waste category i[n Figure 4-3,](#page-39-0) the polymer type that is most abundant is the main polymer type, e.g. PE in the PE bale. The mixed plastic waste stream shows the most heterogeneity but is mostly composed out of PO. In the cases of PE and LDPE the bale quality is higher for post-sorted plastics compared to source separated. The difference in bale quality between the two sorting methods for the PE bale is 4.10% and for the LDPE bale 3.86%. For PP it is the other way where source separation results in the highest bale quality. Here the difference is 10.75%, what is larger than for the other categories. So overall for the bale quality the sorting method that presents the best results varies for each category. It can be concluded that the MRF is able to deliver outgoing sorted bales of similar bale quality for some categories. For this, it is necessary that the sorting operations are changed against the standard practice of handling source-separated plastic waste. For handling post-sorted plastic waste, the sorting operations involvemore steps. There was no information provided about the specifics of these extra sorting steps.

The residues levels i[n Figure 4-3](#page-39-0) do show a pattern. For all categories post-sorted waste result in a higher residue level than source separated, even for the categories where the bale quality of post-sorted is higher. This was also observed during the preparation of the samples. Post-sorted samples were dirtier and harder to clean. The cause of this higher residue level for post-sorted plastic waste can be dedicated to the high complexity of the incoming waste stream, because for post-separation, plastic packaging materials are collected together with residual waste. The mixing of this variety of wastes causes contamination of the plastics during their EOL phase. The MRF is not able to bring down the residue level of the post-sorted plastic waste to the same level as for source separated plastic waste.The higher residue level of the sorted plastic bales shall put more pressure on the further reprocessing operations, especially the washing process.



### 5.2 Quantification of the Elemental Composition

The visualization of results relating to the source separated target samples is presented in [Figure 4-4,](#page-41-0) [Figure 4-5,](#page-42-0) and [Figure 4-6.](#page-42-1) This figure outlines the distribution of elements within the part of the respective products. Notably, neither sulfur nor bromine is observed in any of the three product categories. Furthermore, the comparative analysis of the CHNSO elemental rations aligns closely with the findings reported by Roosen et al. (2020).

The halogen concentrations of fluorine, chlorine, and bromine in this study appear exceptionally low.Zhang et al (2013) previously investigated halogen concentration in various household products, including plastic packaging material. This study indicated that fluorine was either undetectable or fell below the limit of detection (LOD), for chlorine the concentration was 420 ± 13 mg/kg, and for bromine 37.792 ± 79 mg/kg. Similarly, Roosen et al. (2020) corroborated these observations, revealing that fluorine concentration in certain plastic waste categories remained undetectable or exhibited comparably low levels. Moreover, the findings in this study for all samples yielded consistent low halogen concentrations, this suggests that further optimization of the applied analytical method in this study may be necessary. Also due to time limitations the halogen content was calculated out of one repetition. In future studies multiple repetitions are recommended.

In the assessment of the metal concentrations in plastic materials, these metals can be categorized into three distinct groups. When recycled plastics are intended for use in the production of new food packaging, stringent legislative regulations govern the migration of metals from plastic packaging material to the food product (Regulation 10/2011, 2023). Firstly, there are metals that are allowed or no specific requirements are listed (e.g., Ca, K, Na, Se, Ti, and V). Secondly, certain metals are strictly prohibited (e.g., As, Cd, and Pb). Lastly, specific migration limits apply to other metals (e.g., Al, Co, Cu, Fe, Li, Ni, and Sb). This study did not analyze all metals that are not allowed (e.g., Cr and Hg) or subject to specific migration limits. Generally, the concentration of allowed metals exhibits higher measured concentration, while concentrations of prohibited metals result in lower concentrations.

[Table 4-1](#page-43-0) presents the elemental composition for both washed and unwashed random samples.The unwashed samples still contain residue. In certain cases, the concentration of a metal is higher in the washed samples (e.g. concentration K for source separated PE), while at other times, it is higher in the unwashed samples (e.g. concentration Li for postsorted PP). In cases like the latter, this could suggest that some sort of metal contamination occurred. This occurs for multiple metals in source separated PP (e.g., Al, Ca, Cu, Fe, Na, Pb, Ti, V) , post-sorted PP (e.g., As, Ca, Cd, Co, Li, Ni, Pb, Ti, V), and post-sorted LDPE (Al, As, Ca, Cd, Co, Cu, Fe, K, Na, Ni, Pb, Ti, V). For the other samples there is a clear reduction for most metals after washing. For source separated PE there is a large reduction in Al, Ca, Cd, Cu, Fe, K, Ni, Pb, Se, and V concentration, a small reduction in Co and Li, and an increase in Na and Ti. For source separated LDPE there is a large reduction in Al, Ca, Co, Cu, Fe, K, Li, Na, Ni, Pb, and Se, a small reduction in Ti, and a small increase for Cd and Li. For postsorted PE there is a large reduction in Al, Co, Cu, Fe, K, Li, Na, Ni, Pb, Sb, Se, and V, a small increase for As and Ca, and an increase for Cd and Ti. For post-sorted mixed plastic waste there is a large reduction in Al, Ca, Cd, K, Li, Na, and V, a small reduction in Pb, a small increase for Co, Cu, Ni, and Se, and an increase for Fe, Sb, and Ti. A large reduction in metal concentrations is probably due to the effect of the washing process. By removing the residue, a large fraction of these metals is also removed. When there is either a small reduction or increase in metal concentration it most likely that those metals are part of the plastic. The reason why for some samples the washing step decreases the metal concentration and for other increases is possibly by contamination. This contamination could have occurred during the washing process or in later phases while handling the samples, for example during size reduction steps or while



weighing the samples. Another reason could be due to the small amount of sample needed for analysis and the large heterogeneity of the waste samples, even though efforts were made to maximize the homogeneity of the sample. Still some samples (e.g. Co in post-sorted PP) show large standard deviations ( [Appendix G\)](#page-71-0). Thus, for some samples, the precautions were still insufficient to provide an accurate concentration. To improve this, the number of repetitions per sample conducted could be increased.

This study aims to have a better understanding of the disparities between source separated and post sorted plastic waste. To achieve this, column charts were constructed for each metal of the washed random samples [\(Appendix G\)](#page-71-0). To determine if there is a significant difference between the sorting methods in metal concentration for each bale category four Mann-Whitney U tests were conducted [\(Appendix H\)](#page-73-1). For most of the metals, there is no significant difference in concentration between the two sorting methods for each bale category. When there does appear to be a significant difference for a particular metal for the bales of PE, LDPE and mixed. Then those concentrations are in almost all cases higher for post-sorted plastic waste. But for the PP bale, this is not the case. What is remarkable, is that the metals As and Sb if they are measured in a bale category, they are only observed in post-sorted waste. This implicates that these metals are most likely to enter the sorted plastic bale if post-sorting is applied. Therefore, there must be a pathway for these metals to enter the post-sorted stream that is not possible when performing source separation. The residue from these samples was rinsed off, so this is not an option. So, these metals must come from the plastic material. One possible route identified in this study is that plastic products not used as food packaging have a different elemental composition. Therefor, the elemental composition was determined for five special samples [\(Table 4-2\)](#page-45-0) that occur in the post-sorted plastic waste streams. These special samples were obtained from PP and mixed plastic waste streams. The metal As is observed in three out of the five samples. When As was observed, the concentration is plurality of the concentrations found in the random samples. Then the metal Sb, is observed in two out of five special items. It is in one case, for the PE foam, that the concentration 13 mg/kg largely exceeds the concentration of the random samples. So, the assumption is that other plastic household items normally thrown in the residual waste and thus not collected together with plastic food packaging in the case of source separation, form a potential source of elemental contamination. For further research on post-separation of plastics, it could be investigated which plastic products occur in the stream that cannot be found in source separation.

For the other prohibited metals Cd and Pb in terms of migration from food packaging materials the following was established. Cd appears in both source separated and post-sorted plastic waste. However, in the case of post-sorted PE, its concentration surpasses that observed in all other waste stream categories. In case of Pb, source separated PE, PP, and LDPE exhibit higher concentration of Pb compared to their post-sorted counterparts. However, the highest Pb concentration for the washed samples occurs in post-sorted mixed plastics. The presence of these metals underscores the complexities associated with recycling plastics into high-end applications, such as food packaging.



## 5.3 Material Flow Analysis

The material flow model, as shown in the Sankey diagram presented in [Figure 4-8,](#page-46-1) provides a visual representation of how each material type behaves during the sorting process. When critically evaluating the model, several considerations come to light. Firstly, it is essential to acknowledge that there was limited information regarding the sorting process line. For making this flowsheet in [Figure 3-1](#page-30-0) limited information was provided due to trade secrets about the installation. Consequently, assumptions were made based on other similar flowsheets of material recovery facilities tailored to improve the sorting efficiencies. Furthermore, certain material types lacked specific sorting efficiency data for particular sorting steps. In these instances, the sorting efficiencies were based on the behavior of similar materials. In general, while the model serves as a valuable tool, it is crucial to recognize that it represents an approximation rather than an exact reflection of reality. Interpretation should be approached with a critical view.

When comparing the bale quality derived from the model with the results obtained through FTIR analysis the model aligns in some cases well with the observations in this thesis while in other cases it has a wider divergence. The predicted bale qualities of the MFA of PE Rigids is 83.3%, PP Rigids 83.3%, and PE Flexibles 98.5%. For the source separated random samples out [Figure 4-3](#page-39-0) is the bale quality of PE 69.3%, PP 84.3%, and LDPE 56.2%. In case of the PE and LDPE bale the model gives an exaggerated bale quality compared to the measured values. In the sorting mass balance overview in [Table 8-11](#page-74-1) there is no residue present in these bale categories. This is because the model considers residue as an individual material. Allowing the sorting plant to have high separation efficiencies for individual residue. But in reality, residue occurs both loose in the waste stream and attached to the plastic material. Where the solid residue still remains partly present in the bale after the sorting process. For PP, the model provides relatively accurate prediction.



## 6 CONCLUSION

This master dissertation consisted of a major analytic campaign of PPW. A detailed characterization was made about the polymer composition and the elemental composition. The elemental composition included metals content, halogen content, and CHNSO analysis. The analyzed samples were PE rigids, PP rigids, mixed plastic waste, and PE flexibles. The analysis included both specific target samples (e.g., shampoo bottle, butter trays, packaging film), and random samples that represent the whole stream. The added value of this study was that samples were derived from a source separated collection scheme and post-sorted collection scheme.

The polymer composition was determined through FTIR of both sides of the plastic sample. This method is limited in case of multilayer plastic packaging, because no information is provided of the possible inside layers of the plastic waste sample. Therefor this study could be expanded with polarized optical microscopy and differential scanning calorimetry. Next, the elemental composition was split into three parts. Firstly, the elemental content was determined with ICP-OES. Secondly, the halogen content was measured with IC. Thirdly, the CHNSO analysis was performed with an organic element analyzer. For the halogen content the results were lower than expected. Therefore, it would be better to reevaluate the technique used and perform multiple repetitions for each sample.The latter was not possible for this study due to time limitations. Lastly, a MFA was conducted. For this, a model was used that simulates the sorting line process with data derived from previous research.

A MFA was performed based on the sorting plant and data about the sorting efficiencies out of previous research. A Sankey diagram is drawn in [Figure 4-8](#page-46-1) and detailed data about the mass balance is provided i[n Appendix I.](#page-74-2) The Sankey diagram provides insights into how the different materials would behave in the sorting plant and provides an indication about the recovery rates. Also, the bale quality is represented in the diagram. The bale quality for PE and LDPE is exaggerated compared to the measured values from the determination of the polymer composition. This is due how the model handles residue as an individual material and has no regard for residue attached to plastic materials. Because the sorting equipment is able to separate loose residue from the stream with high efficiencies of up to about 99%, this leads to very low residue levels in certain bale categories. Due to the limitations of the model, the results should be interpreted as indication and not an accurate representation of reality. There was only limited information known about the sorting process itself. For some materials, assumptions had to be made for the sorting efficiency based on the behavior of similar materials. This part of the study could be further elaborated to both sorting systems to compare this as well. If more information is available on the sorting processes, it would be possible to also make a comparison in recovery rate.

The composition of target samples was determined for three product categories, namely HDPE bottles, PP trays, and Mixed foils. For each product category five products were selected based on their occurrence on the manual sorting line. Due to the small number of products per category, these items are not representative for the full waste stream. The polymer composition per category is visualized in [Figure 4-2](#page-38-0) and detailed data is provided in [Appendix C.](#page-66-0) For each category the largest fraction is of the main polymer (e.g. PE in HDPE bottles). But also, other polymers next to the main polymer were identified. This addresses the fact that already from the product level there is a large heterogeneity in polymer composition. This heterogeneity results in complex waste streams that present certain challenges for sorting centers. It is most likely that, especially for Mixed foils, there is even more diversity in polymers, e.g. in form of multilayer plastics. These multilayer plastics are challenging to recycle with the current mechanical recycling techniques. Certain chemical recycling techniques are promising for handling these complex materials. Applying design-for-recycling



during the design phase of plastic packaging could contribute to reducing the polymer complexity. In general, for the target samples the residue is lower than for the random samples. For the target samples, there is probably only attached residue and no loose residue. The elemental composition for HDPE bottles is presented in [Figure 4-4,](#page-41-0) for PP trays i[n Figure 4-5,](#page-42-0) and for Mixed foils in [Figure 4-6.](#page-42-1)

The composition of the random samples was determined for four bale categories, namely PE rigids, PP rigids, LDPE, and Mixed plastic waste. These samples can be considered as a representation of the bale. For this, however, it must be taken into account that these samples were only taken at one point in time. To improve accuracy, it is recommended to take samples at different moments. For the random samples, comparison was made between plastic waste that was source separated and post-sorted for each bale category. I[n Figure 4-3](#page-39-0) the polymer composition is presented. Here also, the polymer that is most abundant per category is the main polymer of the respective bale. From [Figure 4-3,](#page-39-0) it is clear that when plastics are collected and sorted through a post-separation system, this leads to a higher residue level. This does not mean that the bale quality is therefore lower for post-sorted plastic waste. Only for the LDPE bale, a significant difference was observed in polymer composition between source separation and post-separation, with the highest bale quality for post-sorted LDPE. So, in terms of polymer purity, a post-separation collection method with a more advanced sorting process is able to produce sorted plastics bales of equal or in some cases higher bale quality. With the remark that instead of polymer contamination, more residue is still present in the sorted bale. Consequently, this will put more pressure on the washing process during reprocessing phase. The impact what this will have on water consumption and wastewater production could be investigated in further research.

The elemental composition of the random samples is presented in [Table 4-1.](#page-43-0) This also includes unwashed samples. The effect of sample washing on elemental composition can be divided into three situations. In the first scenario, washing provided the reduction in metal concentration. This means that the metals in question were present in the residue and were removed. This was the case for source separated PE, source separated LDPE, post-sorted PE, and post sorted mixed plastic waste. In a second scenario, there is only a small difference in metal concentration between with or without washing. This suggests that the metal is mainly present in the plastic material itself. In a third scenario, the metal concentration increases after washing. This is the case for source separated PP, post-sorted PP, and post-sorted LDPE. The consideration here is that contamination may have occurred during the washing process or at later stage during sample handling. Due to the differences between the bale categories studied, no conclusion can be made of the effect of the washing on elemental composition. For comparing source separated plastics with post-sorted the focus was on the washed samples. In general, post-sorted plastics have the highest total metal content. Interesting are the metals As and Sb were only observed in small concentrations in plastic waste that was post-sorted for different bale categories. Considering that As is a prohibited metal in terms of migration in food contact material and for Sb there are specific migration limits. This means that these metals possibly enter the bale through a pathway that is not optional in the case of a source separation system. One possible route considered in this study is that other plastic products that are not for food packaging purposes have a different elemental composition. As a result, more or other metals may be present in post-sorted plastic waste than in source separated. This study therefore included elemental composition mapping for five special samples. In three of these samples the metal As was observed in higher concentrations and the metal is observed in two special samples. This is a first indication that products that do not occur in source-sorted plastic waste but do occur in post-sorted, bring unwanted metals into the sorted stream. Further research is needed to identify these products and determine whether there is a correlation between them and the elemental composition.



This thesis succeeded in conducting a major analytic campaign on the composition of sorted plastic waste. One of the biggest challenges here is the limited time for conducting such study. This was mainly because of equipment failures which caused limitations in the number of repetitions that could be performed.

One of the key findings of this dissertation is that post-separation performs no worse than source separation in terms of bale quality. For both polymer and elemental composition, the best performing system differs by bale category. However, for post-sorting, the residue level is always higher. Here it should be examined whether this will have strong influence in washing during the reprocessing phase. Further in terms of elemental composition, the impact of non-food contact materials in the post-separation stream should be investigated. This is of great importance as there is ongoing political discussion about introducing a mandatory amount of recycled content in plastic packaging materials. At the same time there is strict legislation for food contact materials in terms of migration of specific metals from the material to the food product. Therefore, recycled plastics must meet high quality requirements to achieve this.





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# **APPENDICES**

## <span id="page-63-0"></span>8.1 Appendix A: Sample List



Table 8-1: Sample List







Special items

## 8.2 Appendix B: Analyzed Elements During ICP Analysis, and Emission Line



Table 8-2: Analyzed elements during ICP analysis, and emission line



## <span id="page-66-0"></span>8.3 Appendix C: Data Polymer Composition of Target Samples



Table 8-3: Data Polymer Composition of Target samples

The "\*" indicates the samples that were post-sorted instead of source separated



## <span id="page-67-0"></span>8.4 Appendix D: Data Polymer Composition Random Samples



Table 8-4: Data Polymer Composition Random Samples

The residue level was not derived via FTIR, by the difference in weight of the sample before and after the washing step. From this difference the residue level could be calculated in percentages. This was redirected to the corresponding mass shown in this table



### <span id="page-68-0"></span>8.5 Appendix E: Fisher's Exact Test Polymer Composition

The assumptions that must be met for the Fisher's Exact Test are:

- Independence: The observations must be independent of each other. This means that the presence of an item in one category does not influence the presence of another item in the same or a different category.
- Categorical Data: The test can only be used for data that can be categorized.
- Fixed Margins: The totals of the rows and columns must be fixed, i.e., they should not change as the data are collected.

For the Fisher's Exact Test, the null hypothesis and the alternative hypothesis are as follows:

- Null hypothesis (H0): There is no association between the polymer composition of the waste stream categories and the sorting method. In other words, the polymer composition is independent of the sorting method.
- Alternative Hypothesis (H1): There is an association between the polymer composition of the waste stream categories and the sorting method. This means that the polymer composition is dependent on the sorting method.



#### Table 8-5: Data input PE, in percentages (%)



#### Table 8-6: Data input LDPE, in percentages (%)

#### Table 8-7: Data input PP, in percentages (%)



#### Table 8-8: Data input Mixed, in percentages (%)



To conduct the Fisher's Exact Test the "fisher.test()" function in RStudio was used. The significance level used is 0.05. The r-script was written using Microsoft Copilot. The results of the Fisher's Exact Test for each waste stream category are presented i[n Table 8-9.](#page-69-0)



#### Table 8-9: Results Fisher's Exact Test polymer composition for each waste stream category

<span id="page-69-0"></span>



# 8.6 Appendix F: Elemental Composition Target Samples

<span id="page-70-0"></span>



## <span id="page-71-0"></span>8.7 Appendix G: Concentrations of Metals in the Random Washed Samples




Figure 8-1: Column chart visualizing the average concentration of the sixteen metals each for both the source separated and post-sorted plastic waste per waste categories. The column indicates the average concentration, the blue dots represent each repetition, and error bars represent the standard deviation

## 8.8 Appendix H: Mann-Whitney U Test Elemental Composition

The assumptions that must be met for the Mann-Whitney U test are:

- All the observations from both groups are independent of each other
- The responses are at least ordinal

For the Mann-Whitney U test, the null hypothesis and the alternative hypothesis are as follows:

- Null hypothesis (H0): There is no significant difference between source separated and post-sorted plastic waste. In other words, there is no significant difference in the distribution of metal concentration between the two collection methods.
- Alternative hypothesis (H1): There is a significant difference between source separated and post-sorted plastic waste. In other words, there is a significant difference in the distribution of metal concentration between the two collection methods.

To conduct the Mann-Whitney U test the "wilcox\_test" from the "coin" package in RStudio was used. The significance level used is 0.05. The r-script was written using Microsoft Copilot.

<span id="page-73-0"></span>The p-values of the Mann-Whitney U test for each waste stream category and metal are presented in [Table 8-10.](#page-73-0) The rows represent the respective metal and the columns represent the bale categories. If no observations were done of a metal for one or both collection methods, then the test could not be conducted for that element resulting in no p-value (e.g. arsenic for all bale categories).



## Table 8-10: P-values Mann-Whitney U test



## 8.9 Appendix I: Sorting Mass Balance Overview

Table 8-11: Material Flow Analysis sorting mass balance overview in ton per year



