Designing a Circular Economy for Plastics: The Role of Chemical Recycling in Germany

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Abstract

Greenhouse gas emissions from human economic activity are causing global warming, leading to numerous impacts, including sea level rise, biodiversity loss, and increases in extreme weather events. For this reason, parties involved in the Paris Climate Agreement agreed to limit global warming to reduce its impacts. The second largest global emitter of carbon dioxide is the industrial production of goods. Within industrial production, the chemical industry with the production of olefins and other high-value chemicals for, among other things, plastic production, has a significant impact. Therefore, the present dissertation addresses designing a circular economy for plastics employing chemical recycling, contributing to the decarbonization and defossilization of the German chemical industry.

Five studies published as companion articles address substantial aspects of the chemical recycling of plastic waste as well as barriers to establishing a circular economy. Study A assesses chemical recycling via pyrolysis for lightweight packaging waste and shows that combining the currently predominant mechanical recycling with chemical recycling has economic and environmental advantages over employing these technologies individually. At the same time, more carbon can be recycled, reducing the dependence on fossil resources. Study B shows the importance of integrating the quality of secondary materials in assessing recycling routes. The preferable recycling technology can change based on the quality metrics and their integration into the assessment. Study C conducts pyrolysis experiments for automotive plastic waste and includes the generated data in an economic and environmental assessment of a chemical recycling route. Different economic and environmentally preferable waste handling options are identified when comparing chemical recycling with waste incineration with energy recovery. Study D examines the economics of automotive plastic waste pyrolysis and identifies the minimum plant input capacity at which the pyrolysis is economically feasible in German framework conditions. Study E combines the collected findings in a facility location optimization model for pyrolysis plants treating lightweight packaging and automotive plastic waste in Germany's current waste treatment network. Political steering strategies are analyzed to align economic and environmental objectives in the waste treatment sector.

In addition to the detailed results of the individual studies, four overarching implications are derived: First, waste containing primarily polyolefins and engineering plastics can be technically pyrolyzed and are a suitable feedstock for chemical recycling. However, the most significant waste quantities studied are generated in short-lived lightweight packaging. Second, chemical recycling is environmentally preferable over waste incineration with energy recovery for all assessed waste streams. Economically, chemical recycling is not preferable compared to waste incineration with energy recovery for automotive plastic waste resulting in a conflict of economical and environmentally preferable waste handling options. Third, the quality of the secondary materials must be considered when assessing waste recycling options, as this strongly influences economic and environmental assessment. Fourth, political steering strategies like the extension of CO_2 certificate trading and introducing recycling rates for waste that is a feedstock for waste incineration with energy recovery can align economical and environmentally preferable waste treatment options.

Consequently, the present dissertation provides valuable insights into the role of chemical recycling when designing a circular economy for plastics. Therefore, it has the potential to significantly contribute to closing the circularity gap of plastics.

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Abbreviations and Symbols

Abbreviations

APW	Automotive Plastic Waste
CAPEX	Capital Expenditures
CCS	Carbon Capture and Storage
CCU	Carbon Capture and Utilization
CED	Cumulative Energy Demand
CO_2	Carbon Dioxide
E&I	Equipment and Infrastructure
EoL	End-of-Life
EU ETS	EU Emission Trading System
GDP	Gross Domestic Product
GHG	Grennhouse Gases
GWP	Global Warming Potential
HDPE	High-Density Polyethylene
kt	Kilo Tonnes
LCA	Life Cycle Analysis
LCI	Life Cycle Inventory
LDPE	Low-Density Polyethylene
LWP	Lightweight Packaging Waste
MFCA	Material Flow Cost Accounting
MSWI	Municipal Solid Waste Incineration
Mt	Million Tonnes
OPEX	Operational Expenditures
PET	Polyethylene Terephthalate

PP Polypropylene

PS Polystyrene

PVC Polyvinyl Chloride

RDF Refuse Derived Fuel

TEA Techno-Economic Assessment

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Part I

Framework, Foundations and Implications

1 Introduction and Motivation

The rise in global welfare due to sustained economic growth in recent decades is associated with increased resource consumption and the emission of greenhouse gases (GHG) such as carbon dioxide (CO_2) (Friedlingstein et al., 2022). Figure 1.1 shows the relationship between CO_2 emissions and welfare measured in the gross domestic product (GDP). Wealthy countries emit more CO_2 per capita than less wealthy countries.

GHG emissions from human economic activity are causing global warming, which leads to numerous impacts, including impacts on agricultural yields, health, sea level rise, biodiversity loss, and increases in extreme weather events (IPCC, 2022). For this reason, 196 parties agreed to limit global warming in the Paris Climate Agreement (UNFCCC, 2015).

While the energy sector is responsible for the largest share of global CO_2 emissions, industrial production is the second largest emitter, responsible for 24% of global GHG emissions (IPCC, 2022). The chemical industry alone accounts for 10% of the total (IPCC, 2022), 18% of the European (Agora Industry, 2022), and 19% of the German (Agora Energiewende and Wuppertal Institut, 2019) industrial GHG emissions.

Therefore, decarbonizing industrial production, especially the chemical industry, is necessary to limit global warming and its impacts. Concerning the chemical industry, steam cracking of naphtha and natural gas liquids to produce olefins and other high-value chemicals is a high-CO_2-emitting process that accounts for nearly 20% of the chemical subsector's GHG emissions (IEA, 2018). Since high-value chemicals are used, among other things, in the production of plastics, plastic production is significant for decarbonizing the chemical industry.

The production of plastics is expected to increase further due to their material properties and importance in various value chains (Braun et al., 2021; IEA, 2018). In the plastic value chain, fossil carbon is a raw material and also a fuel providing heat and steam for production (IPCC, 2022). Accordingly, strategies must be identified for decarbonizing the energy supply and defossilizing the raw material supply.

With increasing plastic production, the increasing volume of plastic waste is another environmental challenge (Braun et al., 2021). About 22% of global plastic waste is mismanaged and ends up uncontrolled in nature, while 49% is landfilled and 19% is incinerated (OECD, 2022). In Europe, 23% of post-consumer plastic waste is landfilled, and 42% incinerated (PlasticsEurope, 2022). In Germany, there is basically no landfilling, but 66% of post-consumer plastic waste is incinerated, leading to fossil-based GHG emissions (Conversio, 2022). Mismanagement, landfilling, and incineration of plastic waste result in environmental pollution and additional GHG emissions at the

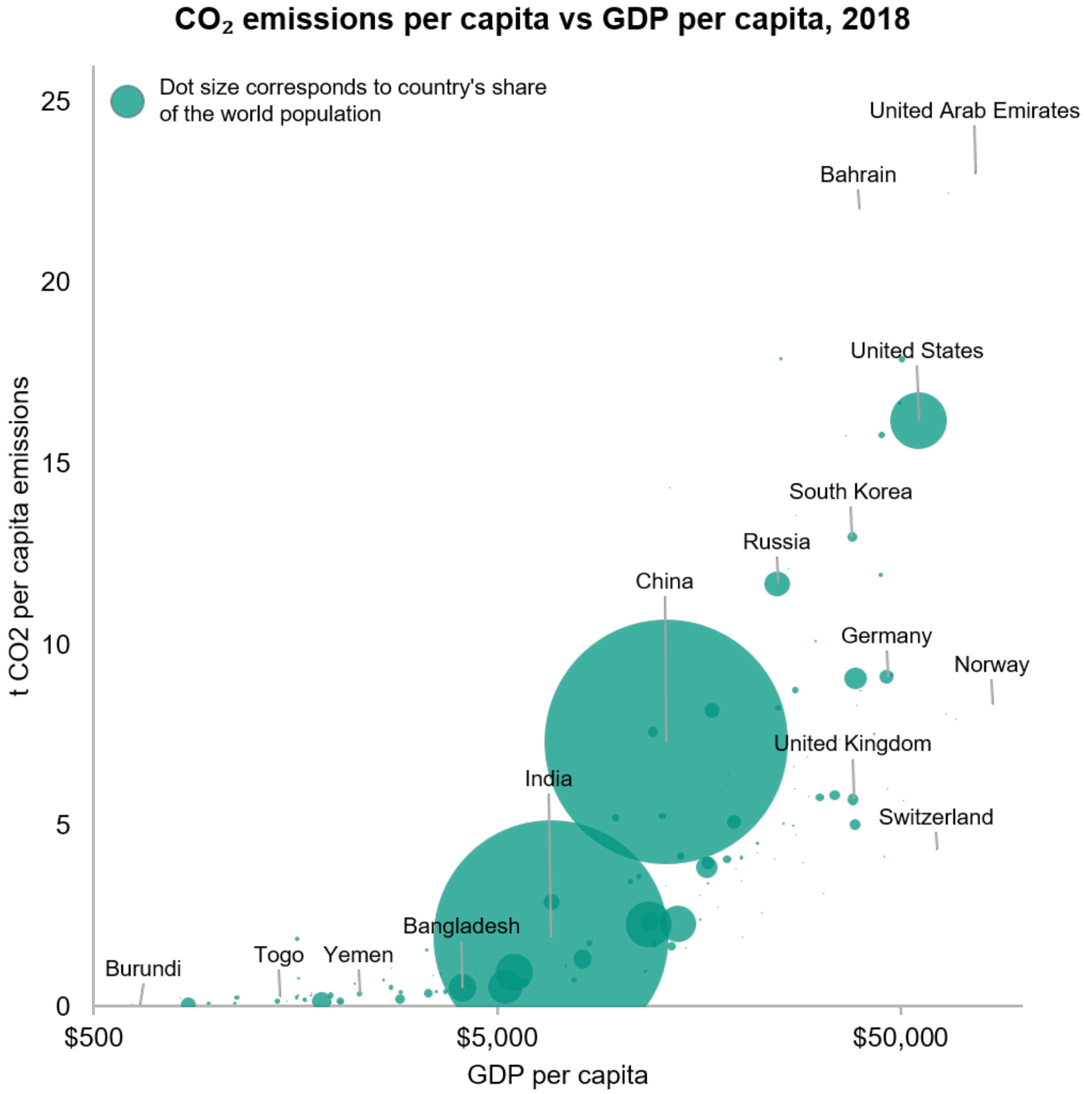


Figure 1.1: Breakdown of countries and their CO_2 emissions per capita and GDP per capita in 2018. The size of the dots corresponds to the country's share of the world population. This measures CO_2 emissions from fossil fuels and industry only; land use change is not included. The figure is based on Our World in Data and the Global Carbon Project; Maddison Project Database 2020 (Bolt and van Zanden, 2020; Friedlingstein et al., 2022).

plastics' end-of-life (EoL) (OECD, 2022). Therefore, it is necessary to aim for a more sustainable plastic production.

In addition to environmental impacts, economic and political risks in the plastics supply chain must also be considered. The war in Ukraine has exposed Europe's and Germany's dependence on fossil resources from politically unstable regions in the energy supply and industrial production. In 2022, 44% of Germany's crude oil imports came from Russia (BAFA, 2023), with 15% of the crude oil being used as feedstock for the chemical industry and, thus, also for the production of plastics (German Bundestag, 2019).

One approach to reducing political risk in the plastic supply chain is to close material loops using post-consumer plastics as a secondary resource, reducing the need for fossil raw materials (Agora Industry, 2022). The use of alternatives to fossil raw materials can be referred to as defossilization. By defossilization, steps can be taken toward strategic autonomy and mitigating political risks (IPCC, 2022; Agora Industry, 2022). From an economic perspective, plastic production costs are also dominated by crude oil's availability and price (Braun et al., 2021). Therefore, closing material loops also has the potential to reduce economic risks by becoming more independent from crude oil. Finally, recycling plastic waste and closing material loops are associated with environmental advantages supporting the decarbonization of the EoL of plastics (IPCC, 2022; Agora Industry, 2022; EEA, 2020). Thus, developing and enhancing circular and resource-efficient value chains can be a strategy to address multiple challenges simultaneously.

Improving EoL recycling of plastic can be achieved by establishing chemical recycling complementing existing mechanical recycling processes. Chemical recycling includes technologies that dissolve or decompose plastics into basic chemicals or hydrocarbons that can be used for synthesizing new chemicals or materials (Davidson et al., 2021). Therefore, the chemical recycling of plastics is an opportunity to close the material cycle and to make additional carbon sources available (IPCC, 2022).

However, due to its high energy demand, it is essential to compare chemical recycling options with other EoL options (e.g., mechanical recycling or waste incineration with energy recovery) (IPCC, 2022). Chemical recycling must also be assessed from an economic perspective, as it is expected to be an expensive waste-handling option compared to established processes. Commercialization requires political support and appropriate regulations (IPCC, 2022).

In summary, a circular economy of plastics, with strategies like chemical recycling, can support environmental objectives, strengthening autonomy from fossil carbon sources, and thus reduce economic and political dependence on supplies of fossil raw materials from politically unstable regions (Agora Industry, 2022). In this context, the *"Circular Economy for Plastics"* project of the *THINKTANK Industrial Resource Strategies* funded by the Ministry of the Environment, Climate Protection, and the Energy Sector of the state of Baden-Württemberg in Germany and industry partners was initiated. The project focused on pyrolysis's economic and environmental performance as an innovative chemical recycling option for plastic waste streams. The pyrolysis process was assessed as part of a potential EoL path and compared to other EoL options. This dissertation and the related studies were developed in the project context.

This dissertation researches chemical recycling as a sub-strategy of a German circular economy for plastics. The potential for using plastic waste as an alternative carbon source and secondary resource for the chemical industry is estimated. In addition, the environmental impacts in the waste treatment sector are studied. Five studies address the economic and environmental performance of chemical recycling via pyrolysis and compare it to established EoL options for plastics. Based on the findings, an optimized waste treatment system for specific plastic waste streams is developed for Germany. Conclusions for a political framework supporting the strategic objective of becoming more independent from fossil resource imports are derived. Consequently, the dissertation supports the development of a German circular economy for plastics.

The dissertation is structured as follows: Chapter 2 provides an overview of the theoretical foundations. Chapter 3 outlines the dissertation's research objectives addressed in the related studies. Their results are highlighted in Chapter 4. Implications of the studies for supporting a circular economy for plastics are discussed in Chapter 5. Finally, the dissertation is summarized and critically examined in Chapter 6. The companion articles are attached at the end of the dissertation.

2 Theoretical Foundation

The theoretical foundations discuss defossilizing the plastic life cycle and associated strategies (cf. Section 2.1). Conclusions for a German circular economy for plastics are drawn by presenting the European and German circularity gaps, discussing causes for the existing circularity gaps, and examining the chemical recycling of plastics to close these gaps (cf. Section 2.2).

Chemical recycling must be assessed economically and environmentally to compare it to established EoL paths. For this purpose, the necessary foundations are presented (cf. Section 2.3). Also, foundations for the design of reverse logistic networks are introduced to integrate chemical recycling into the existing waste management infrastructure (cf. Section 2.4).

2.1 Defossilization of the Plastic Life Cycle

Forecasts indicate that annual global production of plastics will increase from 400 million tons (Mt)[1] in 2020 to around 600 Mt by 2050 (cf. Figure 2.1) (IEA, 2018). As plastic production increases, so does the carbon demand for production, mainly met by fossil sources (EEA, 2020). The high energy demand of the production process, in combination with the predominant energy mixes, also increases associated GHG emissions (Cabernard et al., 2021).

Strategies are being pursued to reduce GHG emissions throughout the life cycle of plastics. The chemical industry's electrification strategy aims to provide process heat electrically, substituting heat and steam from the combustion of fossil resources (Schiffer and Manthiram, 2017). Electricity from renewable resources is expected to replace fossil fuels throughout the production process (Cabernard et al., 2021). Renewable hydrogen can support the transformation of the chemical industry and plastics production by being used where processes' electrification is impossible (Rambhujun et al., 2020). Remaining carbon emissions can be reduced through carbon capture and storage (CCS) or carbon capture and utilization (CCU) technologies (Kätelhön et al., 2019).

However, complete decarbonization of the chemical industry and plastics production is impossible. Carbon cannot be replaced as raw material for production processes, as it is a crucial building block for chemical products. Nevertheless, strategies for defossilization, substituting fossil carbon in production, can be pursued. Biomass and recycling carbon establishing a circular economy can reduce the need for fossil carbon (IEA, 2018; Zheng and Suh, 2019; Meys et al., 2021).

[1] tons refer to metric tons throughout this dissertation

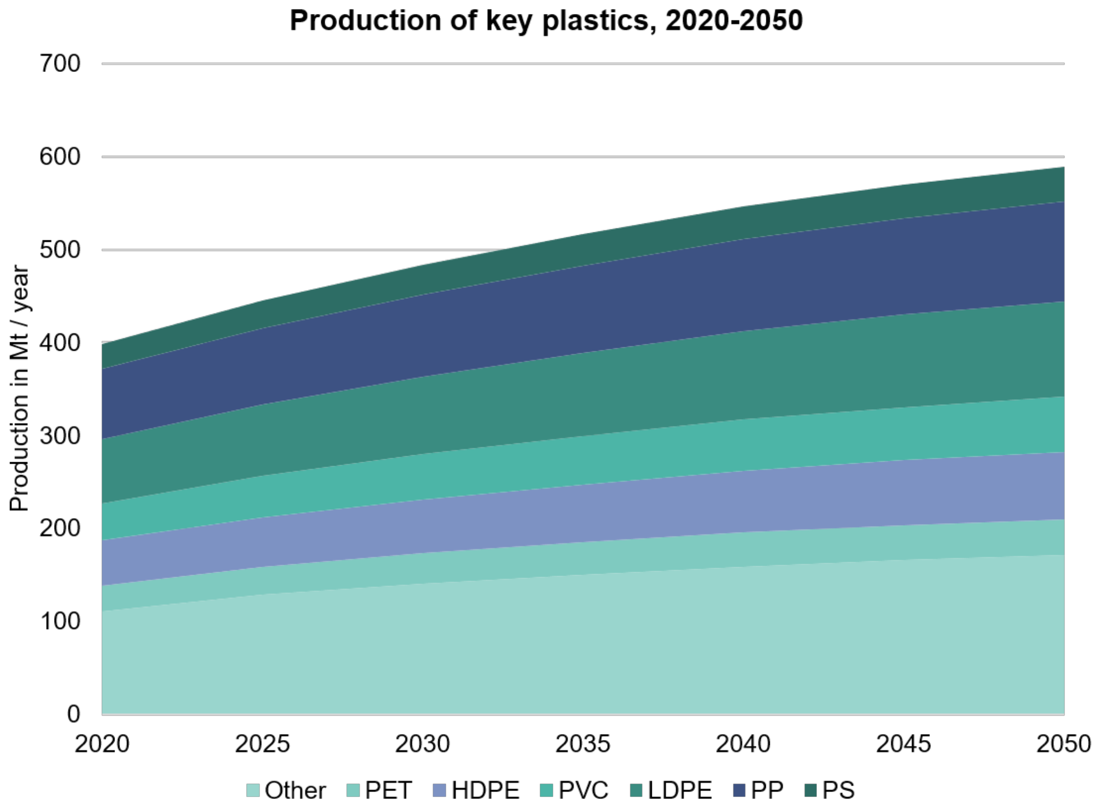


Figure 2.1: Production of key plastics starting 2020 and forecasted until 2050 (IEA, 2018). Key plastics are polyethylene terephthalate (PET), high-density polyethylene (HDPE), polyvinyl chloride (PVC), low-density polyethylene (LDPE), polypropylene (PP), and polystyrene (PS). Plastic selection is limited to thermoplastics that can be effectively recycled and have a resin identification code for distinction.

The EoL is an additional source of GHG emissions in the plastics life cycle. Incineration of plastic waste accounts for 5% of the global plastic life cycle emissions and 13% of the plastic life cycle emissions in Europe (Vanderreydt et al., 2021). Even though plastic recycling has environmental advantages over other EoL options (EEA, 2020), it is not established. Only 9% of global plastic waste (OECD, 2022), 23% of European (Agora Industry, 2022) and 33% of German plastic waste is recycled (Conversio, 2022).

Low recycling rates result in a circularity gap, where the material leaves the economy's material cycle and enters the environment in a controlled or uncontrolled way. The carbon that leaves the economic cycle is no longer available for production processes and must be replaced by carbon from other sources. Accordingly, establishing a plastic circular economy with more resolute plastic waste recycling supports decarbonizing and defossilizing the plastic life cycle (Meys et al., 2021).

In summary, a transformation in the chemical industry, plastic production, and plastic waste management need to take place to reduce the GHG emissions of plastics throughout their life cycle and to become more independent of fossil carbon sources. Besides electrification (Schiffer and Manthiram, 2017), energy transition (Cabernard et al., 2021), CCS and CCU (Kätelhön et al., 2019), and biomass utilization (Meys et al., 2021) expanding the circular economy for plastics, with a focus on increasing recycling rates, is a valuable strategy for achieving a decarbonized and defossilized plastics life cycle (Meys et al., 2021).

2.2 Circular Economy for Plastics

The circular economy is a concept that focuses on closing material and energy loops by combining strategies to use energy and materials more efficiently while minimizing waste (Geng et al., 2012). Strategies within a circular economy are the design and production of goods that enable repairs, consist of components that can be reused or repurposed, and support an EoL recycling (Wiebe et al., 2019). The strategies can reduce the need for virgin materials and reduce carbon emissions (IPCC, 2022).

2.2.1 Legal Framework for the Circular Economy for Plastics

Given the increasing amount of plastic waste and the circular economy's potential to decarbonize and defossilize the plastic life cycle, the EU and Germany support the circular economy for plastics. Measures for this can be found in laws, strategies, and plans.

Many laws affecting the design of the circular economy originate from the waste management sector. At the European level, this is particularly the EU Waste Framework Directive 2008/98/EC. According to this directive, waste must be treated according to the waste hierarchy (European Parliament and Council of the European Union, 2008). The primary goal is waste prevention (cf. Figure 2.2). If the waste cannot be avoided, reuse should be considered before using recycling procedures. Recycling is reprocessing waste to fulfill its original purpose, while recovery processes put waste into a useful purpose replacing other materials (European Parliament and Council of the European Union, 2008). On the lowest level of the hierarchy, waste disposal includes landfilling or waste incineration without energy recovery. Thus, the waste hierarchy supports the circular economy strategy by specifying an order in which waste treatment methods should be applied and emphasizing the desirability of recycling.

The German Circular Economy Act transfers the requirements of the EU Directive 2008/98/EC to the national level. Here, the circular economy is defined as avoiding and recycling the waste in the sense of the waste hierarchy (German Bundestag, 2012). When considering the entire life cycle, protecting people and the environment must be prioritized (German Bundestag, 2012). Technical possibilities, economic viability, and social issues must be considered (German Bundestag, 2012). Also, the German Circular Economy Act establishes fixed rates for the reuse and recycling of municipal waste, thus directly regulating materials recycling (German Bundestag, 2012).

In addition to the general guidelines and laws, there are more specific drafts for individual waste streams. The EU Directive 94/62/EC and the German Packaging Act set specific rates for recycling packaging made from different materials. The EU Directive calls for 55% of plastic packaging

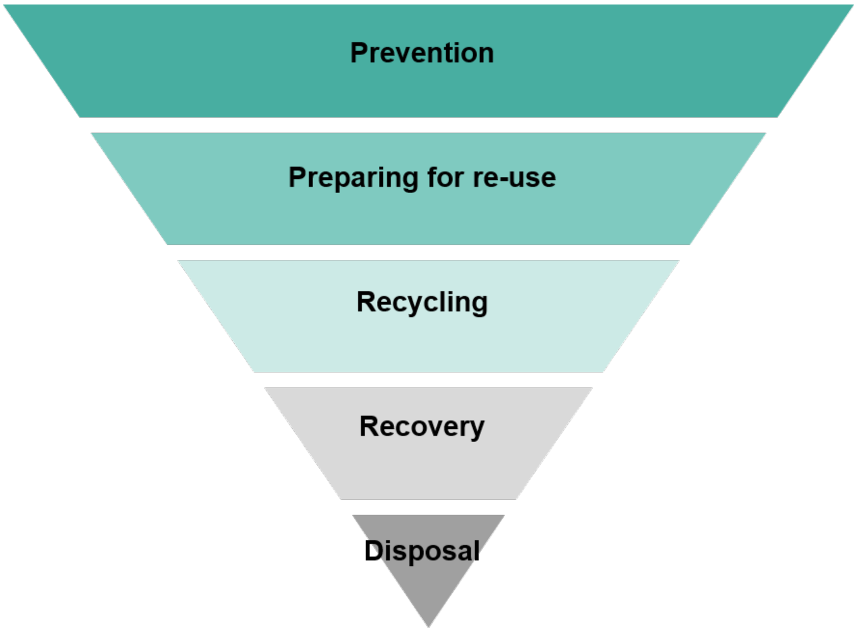


Figure 2.2: Waste hierarchy for waste prevention and management.

to be recycled in 2030 (European Parliament and Council of the European Union, 1994). Germany requires that 90% of plastic packaging must be recovered and 63% must be recycled using mechanical recycling (German Bundestag, 2017). Demanding mechanical recycling is a German specification that requires that the recycled material replaces new material of the same substance or remains available for further use (German Bundestag, 2017).

The EU Circular Economy Action Plan emphasizes the change of focus from waste treatment to the circular economy. As part of the European Green Deal, this plan includes several policy initiatives to move the EU towards a more circular economy. In particular, the design of sustainable and durable products, the circular principle in production processes, the design for recycling, and value recovery models are emphasized (EEA, 2020; European Commission et al., 2020). Here, the ban of landfilling plastic waste and the further development of existing and new recycling processes are of significant importance (EEA, 2020). The objective is to create a system where plastics never end up as waste. The 9 R strategies provide starting points for circular economy solutions (European Commission et al., 2020):

1. Refuse: Make a product redundant by abandoning its function or offering the same function through a radically different (e.g., digital) product or service.

2. Rethink: Make product use more intensive (e.g., through product-as-a-service, reuse and sharing models, or by putting multi-functional products on the market).

3. Reduce: Increase product manufacture or use efficiency by consuming fewer natural resources and materials.

4. Reuse: Reuse a product that is still in good condition and fulfills its original function (and is not waste) for the same purpose for which it was conceived.

5. Repair: Repair and maintain a defective product so it can be used with its original function.

6. Refurbish: Restore an old product and bring it up to date (to a specified quality level).
7. Remanufacture: Use parts of a discarded product in a new product with the same function (and as-new condition).
8. Repurpose: Use a redundant product or its parts in a new product with a different function.
9. Recycle: Recover materials from waste to be reprocessed into new products, materials, or substances, whether for the original or other purposes. Recycling includes reprocessing organic material but does not include waste incineration with energy recovery and reprocessing into materials to be used as fuels or for backfilling operations.

Details of the general EU Circular Economy Action Plan and specific plastics strategies are described in the EU Plastics Strategy. The strategy focuses on the recycling-friendly design of plastic products, supporting the demand for recycled plastics and creating viable markets for recycled and renewable plastics, as well as an improved collection of plastic waste to ensure high-quality feedstock for the recycling industry (European Commission, 2018). In addition, the EU aims to drive innovation and investment in plastics recycling, focusing on developing circular solutions to expand and modernize separation and recycling capacities (European Commission, 2018).

2.2.2 Strategies for the Circular Economy for Plastics

Looking at the plastic life cycle under the present legislation and the European strategies on the circular economy and plastics, one can see how the circular economy can decarbonize and defossilize the life cycle. Figure 2.3 shows the plastic life cycle: Fossil raw materials are extracted and processed in refineries before producing chemicals and polymers. The plastics are formulated and compounded and then used to make plastic products. The products are sold to customers by retailers and enter the use phase. Following the 9 R Strategies, products that have reached their EoL can be repaired, reused, or collected as waste, depending on their condition. After sorting, it is possible to refurbish products in good condition or repurpose them. Parts of the product can be remanufactured and used in new products. Products that do not have the appropriate quality end up in EoL management: Plastic waste unsuitable for recycling is either landfilled or incinerated, while plastic waste suitable for current mechanical recycling processes is recycled.

Recycling plastic waste can avoid waste incineration and landfilling and associated emissions. The need for primary materials can be reduced, lowering the amount of material in the energy-intensive resin-production stages that account for 61% of conventional plastic production's global GHG emissions (Zheng and Suh, 2019). The other R strategies also reduce the need for primary material and, thus, the material in the energy-intensive production stages.

Reuse, repair, refurbishment, repurposing, and remanufacturing are suitable strategies for good-quality waste. However, for plastic waste that does not meet these conditions, recycling remains the only option for keeping the material in the material cycle. Research is currently focused on plastic recycling technologies to close the existing gap in the material cycle, as the other R-strategies are

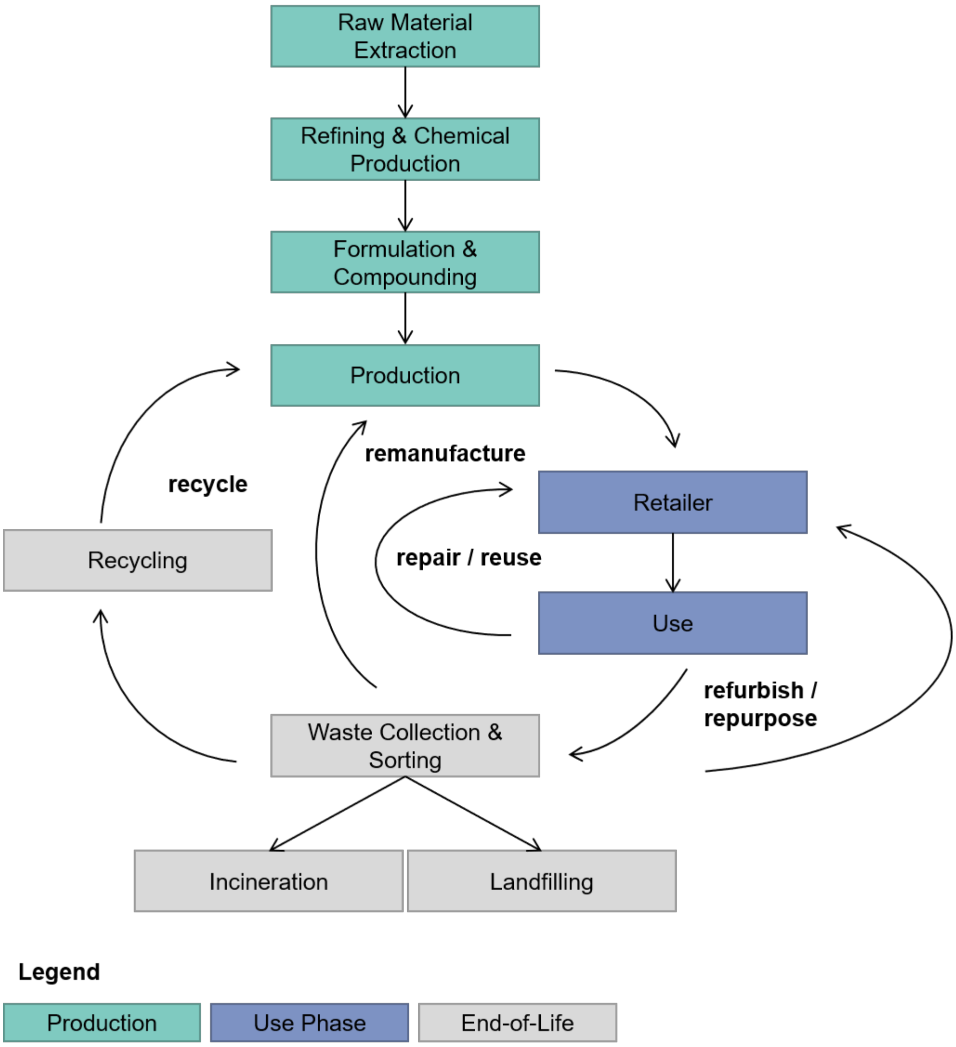


Figure 2.3: Plastic life cycle and circular economy strategies.

less about specific materials and more about products (Johansen et al., 2022; King and Locock, 2022). Accordingly, this dissertation also limits its consideration to the recycling of plastics.

2.2.3 Circularity Gap of EoL Plastics

In 2020, 29.5 Mt of plastic waste was produced in the EU (Agora Industry, 2022). About 42% of the plastic waste was incinerated while 23% was landfilled (Agora Industry, 2022). Only 35% was input for recycling processes (Agora Industry, 2022). Here, additional recycling losses occurred, so only 23% of the EoL plastics were finally recycled (Agora Industry, 2022). In Germany, only some post-consumer plastic waste was landfilled. At the same time, 61% was incinerated, 6% was lost during recycling and were incinerated, while 33% was recycled (Conversio, 2022). Figure 2.4 shows these circularity gaps for Germany and the EU.

The circularity gaps partly result from the mechanical recycling technologies available. A requirement for mechanical recycling is separated plastic waste streams containing only one polymer

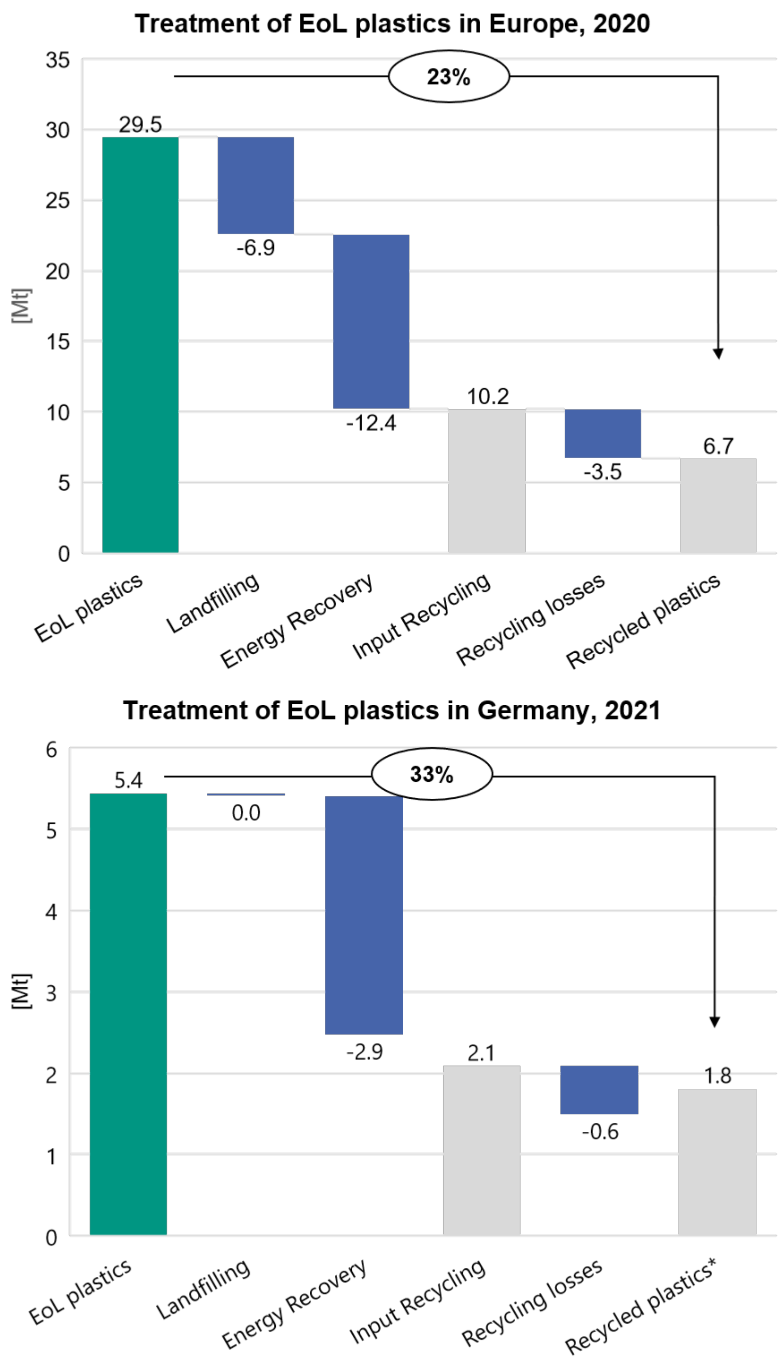


Figure 2.4: European and German plastic circularity gap. Data from Agora Industry (2022) and Conversio (2022). Numbers marked with * include exports.

(Ragaert et al., 2017). Accordingly, the waste must be sorted and separated into different polymer streams before these are mechanically recycled (Ragaert et al., 2017). Waste contamination complicates the sorting (Pivnenko et al., 2015). Plastic composites, e.g., in the form of multi-layer packaging, are difficult to be recycled since the plastics can hardly be separated, resulting in additional material losses (Pivnenko et al., 2015). Plastics that are not collected separately are also not available for mechanical recycling. These plastics are then landfilled or incinerated (with energy recovery).

In addition, mechanically recycled plastics are not the same quality as primary plastics and cannot be used in all applications (Klotz et al., 2022). Plastics degrade over time, with thermal-mechanical degradation occurring through the recycling process, leading to a loss of material properties over their lifetime (Ragaert et al., 2017). The quality losses increase with the contamination of the waste streams (Eriksen et al., 2019). Therefore, with current mechanical recycling processes, recycling these contaminated waste streams is not economical as there is no market for the lower quality secondary plastics (Eriksen et al., 2019). Thus, the loss of material properties also contributes to the circularity gap. Chemical recycling of plastics has the potential to close these circularity gaps.

2.2.4 Chemical Recycling of Plastics

Chemical recycling can be described as applying thermochemical or chemical technologies to break down plastic waste into its components, such as monomers or hydrocarbons (Davidson et al., 2021). The different technologies have specific requirements and yield various recycled feedstock that substitutes virgin feedstock for chemical processes (European Commission, 2019; Rahimi and García, 2017). Figure 2.5 shows a selection of available plastic recycling technologies and their products.

In solvent-based plastics recycling or dissolution, the polymer composition of the plastic remains unaltered (Schlummer et al., 2020). The solvent molecules interact with the polymer macromolecules to form a purified polymer solution where the solvent and polymer can be separated (Schlummer et al., 2020; Martinez Sanz et al., 2022). Since the solvents are polymer-specific, dissolution can only be used for sorted plastic waste.

In depolymerization, polymer chemistry reverses the polymerization process and recovers the plastic monomers (Davidson et al., 2021). Depolymerization is also referred to as solvolysis, whereby a distinction is made between hydrolysis, acidolysis, glycolysis, and methanolysis, depending on the used reactant (Schlummer et al., 2020). These recovered monomers can then be used in the polymerization process to produce virgin polymers. Again, sorted plastic waste streams are required (Schlummer et al., 2020).

Thermochemical processes include pyrolysis and gasification. Pyrolysis uses heat in a low-oxygen atmosphere to break down the carbon chains of plastic and produce solid, liquid, and gaseous hydrocarbon chains (Davidson et al., 2021). Pyrolysis can be used for heterogeneous plastic mixtures and contaminated waste streams (Ragaert et al., 2017). However, some specific plastics and contaminants must not enter the process (Solis and Silveira, 2020). The quality of the main

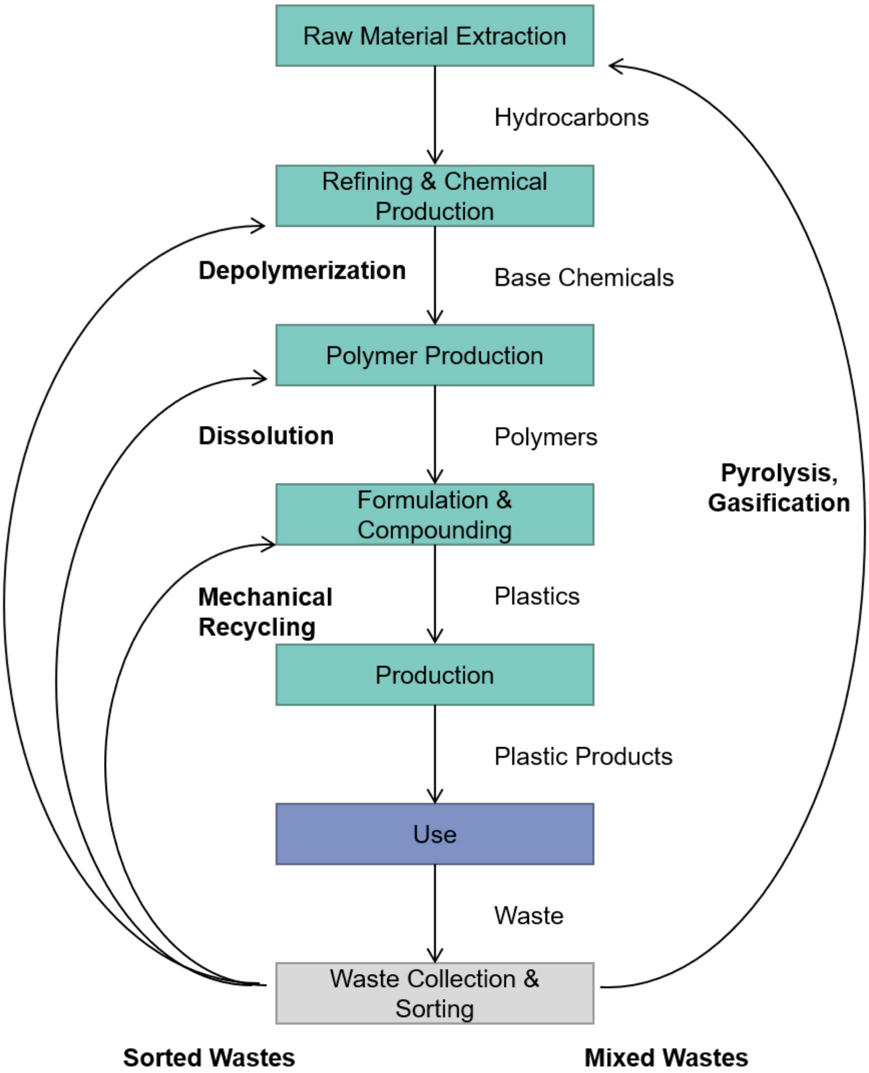


Figure 2.5: Overview of mechanical and chemical plastic recycling technologies and their products (based on Lee (2021)).

product, pyrolysis oil, highly depends on the quality of the waste feedstock. Further pyrolysis oil processing is usually necessary before it can be fed into steam crackers to replace crude oil derivatives and be reprocessed into primary plastic (Kusenberg, Roosen, et al., 2022; Kusenberg, Eschenbacher, et al., 2022).

Gasification involves the controlled application of heat, steam, and oxygen to break down the plastic and produce synthesis gas, a mixture of hydrogen and carbon monoxide (Davidson et al., 2021). Gasification is also flexible concerning feedstock, making it suitable for mixed plastic waste (Solis and Silveira, 2020). In addition, the product of the synthesis gas can be widely used, although the product gas usually requires upgrading to increase its quality (Solis and Silveira, 2020).

Pyrolysis and gasification are suitable for commercial use (Solis and Silveira, 2020). However, compared to the products of gasification, pyrolysis oil has the potential to replace crude oil derivatives in the production process of plastics, thus directly closing the plastic material cycle (IPCC, 2022; Kusenberg, Roosen, et al., 2022; Kusenberg, Eschenbacher, et al., 2022). Therefore, this dissertation is limited to analyzing chemical recycling utilizing pyrolysis. Unless otherwise indicated, chemical recycling and pyrolysis are used synonymously in the following.

The German Circular Economy Act and the EU Waste Directive 2008/98/EC define chemical recycling as a recycling technology, which means it is generally preferable to waste incineration with energy recovery according to the waste hierarchy (Vogel et al., 2020). However, the products of chemical recycling can also be used as fuel in waste incineration with energy recovery. Therefore, the German authorities demand proof of chemical recycling routes' environmental benefits and economic viability when closing the material cycle compared to waste incineration with energy recovery (Vogel et al., 2020).

2.2.5 Critiques of the Circular Economy

In addition to the potential of the circular economy, criticisms must be mentioned. The idea of a closed economic cycle with no need for new primary materials stands in contrast to material losses at each loop through the economic cycle. Each loop is associated with losses in quantity (material losses, by-products) and quality (material mixing, degradation) (Corvellec et al., 2022). As a result, new materials and energy must be used in each material loop to overcome dissipation losses (Cullen, 2017). Also, quality losses in material properties and limitations in manufacturing and reprocessing technologies are often neglected (Velis and Vrancken, 2015). This is especially the case for substituting primary material with secondary material (Zink and Geyer, 2017). The complexity of waste and the fact that it cannot be avoided entirely is not fully considered when discussing circular economy strategies (Mavropoulos and Nilsen, 2020).

Challenges developing circular economy models include technical, economic, and regulatory barriers (de Jesus and Mendonça, 2018). Secondary resources from waste are associated with supply constraints, price volatility (Babbitt et al., 2018), quality limitations (Zink and Geyer, 2017), contamination (Baxter et al., 2017), and other inherent uncertainties (Linder and Williander, 2017). These challenges must be addressed to establish waste as a resource in a circular economy.

Another criticism of the circular economy is that no distinction is made between the circular economy and sustainability (Corvellec et al., 2022). Circular systems are not necessarily more environmentally sustainable than linear systems (Brandão et al., 2020; Panchal et al., 2021), so circular economy strategies and environmental impacts should always be considered together (Moraga et al., 2022).

This includes new recycling technologies such as chemical recycling. A detailed assessment from a life cycle perspective is required, as recycling activities can be energy and emissions-intensive (IPCC, 2022). Chemical recycling processes are also associated with carbon losses in the form of exhaust gases and solid residues (Dogu et al., 2021; Davidson et al., 2021). They are therefore associated with CO_2 emissions, which can be even more significant for systems with chemical recycling than for waste incineration with energy recovery (Meys et al., 2020). Case-specific analyses are needed.

Costs define the economic limits of chemical recycling compared to primary material prices. In the past, external conditions (e.g., availability and price of fossil feedstocks) have not provided the necessary incentives to pursue chemical recycling options and avoid combustion- and process-related CO_2 emissions (IPCC, 2022). Under current market conditions, using fossil fuels is likely cheaper than producing secondary materials, and policy support is needed for commercialization (Wyns et al., 2019; Material Economics, 2019; Wesseling et al., 2017; Bataille, 2020).

From these criticisms follows that the chemical recycling of plastics needs to be environmentally assessed to compare it to other EoL options and establish its potential to close the plastic circularity gap. An economic assessment must also be conducted to research the economic viability of chemical recycling and identify policy frameworks in which it is economical to realize any potential environmental benefits.

2.3 Economic and Environmental Assessment

In order to be able to compare different EoL options for plastics, these must be assessed economically and environmentally. This enables highlighting the advantages of individual options and identifying potential economic and environmental trade-offs. Life cycle assessment (LCA) and material flow cost accounting (MFCA) can be combined for the joint assessment of environmental and economic performance (Rieckhof and Guenther, 2018).

Life cycle management with LCA is a non-monetary tool to visualize the potential environmental impacts of a product system over its entire life cycle (ISO 14040: 2006). Thus, LCA takes an engineering perspective and the perspective of a sustainability manager (Rieckhof and Guenther, 2018). MFCA categorizes all product system-related input and output flows and identifies cost drivers (ISO 14051: 2011). Thus it can increase the transparency of resource flows and associated costs. It acts as the interface between production engineering, environmental and cost accounting, and management (Rieckhof and Guenther, 2018).

LCA and MFCA build on a similar understanding of materials and methods (Viere et al., 2011; Kokubu et al., 2009; Sygulla et al., 2014) and are therefore well suited to determine economic and environmental key figures (Rieckhof and Guenther, 2018). LCA helps to identify hotspots of resource use and associated environmental impacts, while MFCA visualizes resource flows and monetizes resource uses.

The common understanding of the methods and the parallel application of the two methods is shown in Figure 2.6 (Rieckhof and Guenther, 2018). In line with the LCA approach (ISO 14040: 2006), four different implementation steps can be distinguished (Rieckhof and Guenther, 2018):

- Goal and scope definition: In the goal and scope definition, the system under study and the function of the system are described. The system boundaries are clearly defined, and the functional unit of the assessment is determined.

- Inventory analysis: After the scope of the assessment is defined, the relevant data for the assessment are compiled. In LCA, this includes physical information on material flows. In MFCA, cost data are added to this data.

- Assessment: The compiled data are used to calculate the corresponding key indicators in the assessment. In LCA, a choice can be made between different environmental indicators and environmental impacts, while in MFCA, the costs of the material flows are calculated.

- Evaluation and interpretation: Finally, the key indicators and underlying information can determine cost drivers and drivers of environmental impacts. The data basis can be discussed, and sensitivities for uncertain data can be calculated.

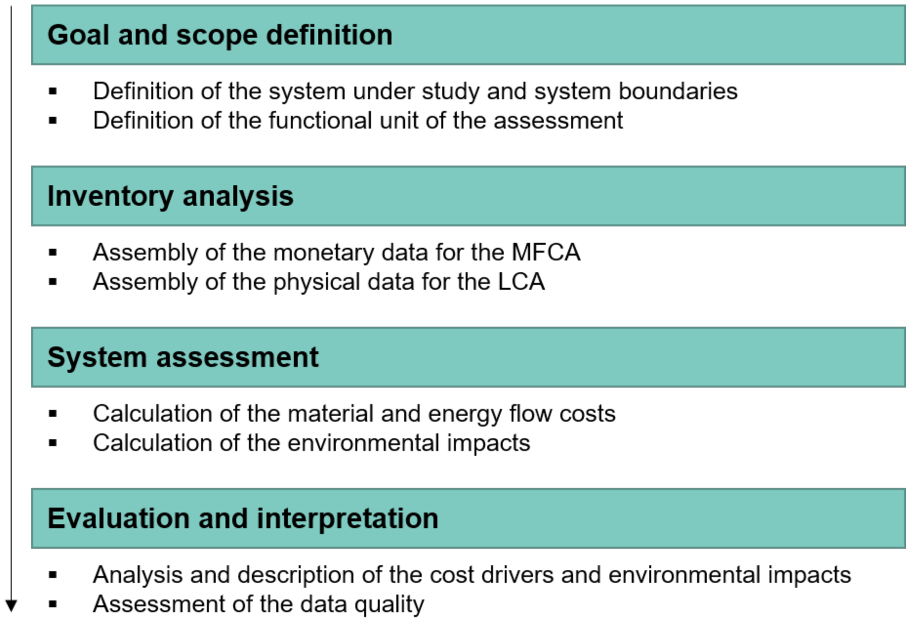


Figure 2.6: Integrated LCA and MFCA implementation steps based on Rieckhof and Guenther (2018).

Additionally, techno-economic assessments (TEA) are frequently performed for a more detailed economic assessment of technologies. Trippe (2013) describes the methodological procedure and the required data basis for conducting the TEA (cf. Figure 2.7).

First, the solution space for the concept under investigation is described, and the system boundaries are defined. Process variants can extend a basic configuration of the process. Material and energy balances are established and used to design the system. Based on the plant design, investment estimation can be made using manufacturers' data, business reports, and literature data. With the analysis of the raw material supply and the market prices of the input, the conversion and manufacturing costs can be determined, and the minimum selling price can be derived. The minimum selling price can be compared with the expected product selling price to make statements about the expected profitability. The data basis and assumptions in the input parameters can be examined with scenario and sensitivity analyses.

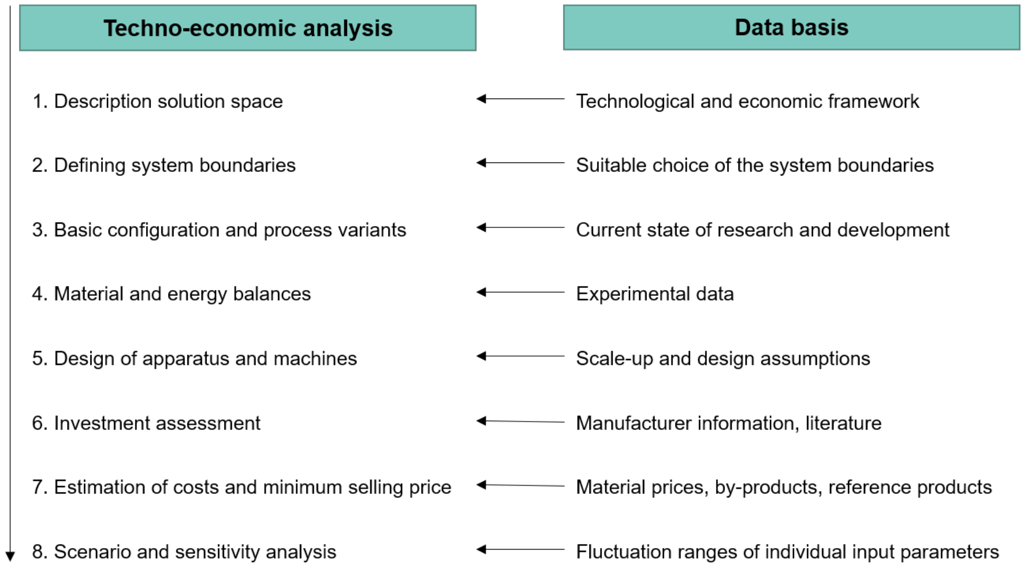


Figure 2.7: Assessment evaluation approach of techno-economic studies based on Trippe (2013).

Based on the environmental assessment using LCA and the economic assessment using MFCA and TEA, the advantageousness of individual recycling options for plastic waste and possible economic and environmental trade-offs can be established. The findings can also be incorporated into modeling a reverse logistic network or a waste management network to research political steering strategies in a policy framework to influence environmental and economic trade-offs.

2.4 Modeling Reverse Logistic Networks

Reverse logistics networks optimize the recovery or reuse of EoL resources in supply chains (Egri et al., 2021). Waste treatment and recycling is typically a complex recovery process consisting of various collecting, sorting, and processing stages. Reverse logistics network planning involves facility location and network flow problems. Facility location problems involve finding the optimal location for facilities to fulfill a given demand while minimizing costs, environmental impacts, or multiple objectives (Laporte et al., 2019). Network flow problems consist of supply and demand points connected by multiple routes to transfer the supply to the demand (Bertsekas, 1998). The route to be chosen is the one that minimizes cost, environmental impact, or multiple objectives depending on the decision maker's preference function.

Different decisions can be modeled and analyzed depending on the layout of the recycling network and the number and type of facilities considered. A review of reverse logistics in the context of plastics recycling shows no uniform level of detail in modeling plastic waste recycling networks (Valenzuela et al., 2021). Most studies include transportation between or placement of transfer points and recycling centers, considering potential customers (Valenzuela et al., 2021). Uniform modeling of plastic recycling networks is challenging because, depending on the country or region modeled, there are different waste compositions and qualities due to different collection systems. Furthermore, different infrastructures exist, and different technologies for waste treatment can be considered. Relocations of network facilities (Bing et al., 2015, 2014) or multi-objective formulations to redesign waste treatment while minimizing costs and GHG emissions (De la Hoz et al., 2017; Langarudi et al., 2019) can be explored. Santander et al. (2022) review the existing literature considering the evaluation of the social, political, and technological dimensions of sustainability in the context of recycling networks.

From the literature reviews, it becomes clear that additional studies can strengthen the understanding regarding the modeling and design of recycling networks, especially by incorporating innovative recycling processes considering multiple objective functions.

Chapter 3 presents this dissertation's research objectives. The study results addressing these research objectives are summarized in Chapter 4.

3 Research Objectives

The present dissertation studies the role of chemical recycling in designing a circular economy for plastics contributing to the decarbonization and defossilization of the German chemical industry. The focus is on the chemical recycling of plastics using pyrolysis to close the current circularity gap.

For chemical recycling to be a suitable strategy for a circular economy for plastics, the technical feasibility of pyrolysis must be given. The technical feasibility differs depending on the waste feedstock used. Feedstock availability is relevant for the scalability of pyrolysis. The economic viability of pyrolysis should be compared with established waste treatment processes to estimate its economic success. The environmental comparison with other waste treatment processes is also relevant, identifying the environmentally preferable waste treatment option. When comparing the waste treatment processes, the quality of the secondary products must be considered.

Suppose the availability of the feedstock and the economic and environmental assessment of the pyrolysis of plastic waste shows that chemical recycling is a suitable strategy for a circular economy for plastics. In that case, political steering strategies supporting the circular economy for plastics and establishing chemical recycling must be examined.

In summary, the following cross-study research objectives are considered in this dissertation and described in more detail in the following sections:

1. Analysis of potential feedstock for chemical recycling (cf. Section 3.1)
2. Economic and environmental assessment of chemical recycling (cf. Section 3.2)
3. Discussion of the quality of products from different recycling processes (cf. Section 3.3)
4. Analysis of plastic waste treatment networks and derivation of political steering strategies (cf. Section 3.4)

Five scientific studies improve understanding chemical plastics recycling as a circular economy strategy. Figure 3.1 shows an overview of the studies and their interrelationships, while their contributions to the cross-study research objectives are summarized in Table 3.1.

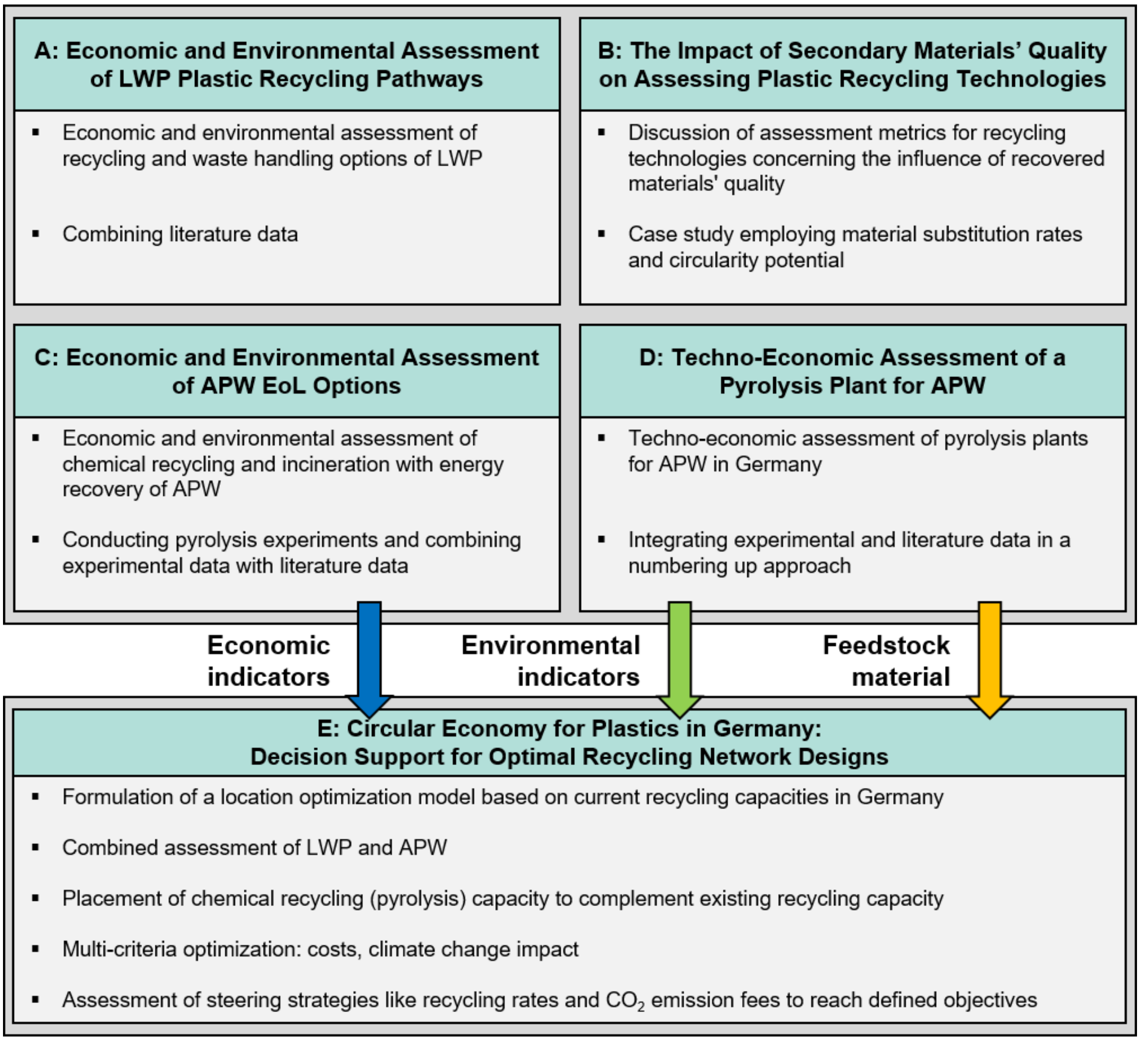


Figure 3.1: Overview of the dissertation and the conducted studies.

3.1 Analysis of potential Feedstock for Chemical Recycling

As described in Section 2.1, introducing and expanding a circular economy can reduce emissions along the entire life cycle of plastics (Meys et al., 2021). Thereby, chemical recycling of plastics can be one option for an overall circular economy strategy. The potential of chemical recycling to close the circularity gap can be assessed by identifying waste streams currently contributing to the circularity gap, establishing their volume, and the technical feasibility of their chemical recycling.

Research focuses on mixed plastic waste from packaging waste (Jeswani et al., 2021; Kusenberg, Roosen, et al., 2022; Meys et al., 2020; Bergsma, 2019a, 2019b) and recycling of mixed household waste (Lim et al., 2022; Gao et al., 2022; Zhao et al., 2022). These are high-volume waste streams. Only some studies consider chemical recycling for treating more demanding waste streams with challenging contaminants or plastic mixtures (Cardamone et al., 2022; Gracida-Alvarez et al., 2019a, 2019b). Due to high shares of non-standard functionalized engineering thermoplastics,

Table 3.1: Overview of the studies and their contribution to the cross-study research objectives.

Cross-study Research Objective	Study A	Study B	Study C	Study D	Study E
Analysis of potential feedstock for chemical recycling	X		X		X
Economic and environmental assessment of chemical recycling	X	X	X	X	
Discussion of the quality of products from different recycling processes	X	X			
Analysis of the plastic waste treatment network and political steering strategies					X

thermosets, and elastomers, automotive plastic waste (APW) can be considered a demanding waste stream.

Also, studies rarely transparently report the mass and energy balances of pyrolysis experiments and rely on data obtained from the pyrolysis of waste fractions not assessed in the study. Transparently reporting the mass and energy balances for chemical recycling expands the available database in the literature. Mass and energy balances can be used to calculate possible increases in recycling rates, the potential for reducing primary plastic production, and the reduced need for fossil raw materials through chemical recycling.

Study A contributes to the literature by assessing and transparently presenting the pyrolysis of lightweight packaging waste (LWP). In Germany, LWP waste accounts for 59% of post-consumer plastic waste (Conversio, 2020), with 34% of plastic packaging not being mechanically recyclable (Christiani and Beckamp, 2020). Study A relies on literature data to establish mass and energy balances for the chemical recycling of LWP waste and determine the contribution of chemical recycling to increasing recycling rates. Study C takes a similar approach, focusing on engineering plastics from the automotive sector. The technical feasibility of pyrolysis of APW is demonstrated, and the mass and energy balances from the conducted experiments are reported. Study E estimates the German domestic feedstock potential for chemical recycling for the waste streams in Studies A and C.

The studies contribute to the literature by estimating the waste potential in Germany for pyrolysis and demonstrating the technical feasibility of pyrolysis for demanding waste fractions such as APW.

3.2 Economic and Environmental Assessment of Chemical Recycling

The concept of circular economy is criticized because it is often assumed to be synonymous with sustainability (cf. Section 2.2.5). However, energy-intensive technologies such as chemical recycling need to be assessed more closely regarding their environmental impact (IPCC, 2022) to support the development of a sustainable economy.

For this reason, Studies A and C assess the climate change impact of chemical recycling routes and compare these with the climate change impact of the currently established EoL options. Potential environmental advantages can only be realized if chemical recycling is economically competitive. Therefore, Studies A and C also calculate the waste treatment costs of the EoL options. Based on the mass and energy balances of Study C, Study D provides a detailed TEA of a plant design for the pyrolysis of APW in Germany. Thereby, the costs of chemical recycling are established in more detail and are compared to fossil reference products.

The studies stand out from the existing literature on the environmental assessment of chemical recycling (Civancik-Uslu et al., 2021; Jeswani et al., 2021; Keller et al., 2022) by additionally considering the process costs of the investigated recycling routes (Study A and C) and the quality of the data basis of the assessment (Study C and D). Study D distinguishes itself from the literature (Riedewald et al., 2021; Larrain et al., 2021) by the technology used and the transparency of the assessment.

3.3 Discussion of the Quality of Products from different Recycling Processes

When assessing the economics and environmental impacts of recycling options, it is crucial to consider the quality of the secondary material produced. As Klotz et al. (2022) describe, established mechanical recycling options often result in downcycling, which means that not all plastic applications can be supplied with the produced secondary material due to quality problems. For this reason, potential quality losses of secondary plastics from chemical recycling processes should be addressed.

Even though pyrolysis oil must be upgraded to substitute naphtha, it can be used in steam crackers resulting in similar yields of high-value chemicals compared to a naphtha feedstock (Kusenberg, Eschenbacher, et al., 2022; Kusenberg, Roosen, et al., 2022). Thus, chemical recycling can prevent downcycling if the high-value chemicals are further processed into plastics.

Downcycling and differences in secondary plastics' quality must be considered when assessing recycling routes. Study B compiles options of how the quality differences can be considered when assessing the substitution of primary materials and uses the case study from Study A to determine the circularity potential of chemical recycling compared to mechanical recycling.

The study complements the existing literature by critically discussing the influence of secondary materials' quality and how it can be measured. The influence of secondary materials' quality on the assessment of recycling technologies is also demonstrated using different assessment metrics.

3.4 Analysis of Plastic Waste Treatment Networks and Political Steering Strategies

Extending the economic and environmental assessment from individual EoL paths to waste treatment networks enables investigating the extension of existing recycling networks with additional waste treatment capacities employing different technologies. The network analysis shows potential barriers implementing individual technologies while considering local conditions (Sommer et al., 2022). For a detailed investigation of steering approaches, Study E models a part of the German recycling network for plastics. Based on the data and findings from studies A, C, and D, political steering strategies are investigated to integrate chemical recycling into the German waste treatment infrastructure.

The developed model contributes to the literature by focusing on extending an existing network with additional waste treatment capacities employing a chemical recycling process. Even though the model focuses on Germany and the German plastic waste treatment network and infrastructure, valuable insights can be derived and applied to other countries. The model has a high level of detail in modeling the waste treatment starting with plastic waste generation and including possible customers for pyrolysis oil. In addition, a multi-objective formulation allows for addressing the impact of steering approaches on GHG emissions and network costs.

A description of each study and a summary of their findings answering the outlined research objectives follow in the next Chapter 4. Implications for a German plastic circular economy defossilizing its chemical industry are discussed in Chapter 5.

4 Summary of Studies and Results

This chapter provides an overview of the studies conducted, their context, and their contribution. The results of the studies are summarized and discussed. Each study is attached in the second part of this dissertation.

4.1 Study A: Assessment of LWP Plastic Recycling

This section refers to the article "Techno-economic assessment and comparison of different plastic recycling pathways: A German case study." This article was written in collaboration with Rebekka Volk, Justus Steins, Savina Yogish, Richard Müller, Dieter Stapf, and Frank Schultmann. It was published in the Journal of Industrial Ecology as Volk et al. (2021).

Study Context and Contributions

The study compares the following recycling routes for LWP: (1) Mechanical recycling in combination with waste incineration with energy recovery of the sorting residues, (2) chemical recycling via pyrolysis, and (3) a recycling approach combining mechanical recycling of the LWP waste and chemical recycling of the sorting residues (cf. Figure 4.1). The recycling routes are compared based on their process costs, Global Warming Potential (GWP)[1], Cumulative Energy Demand (CED), and carbon recycling rate. Quality differences in the secondary plastics produced are considered and qualitatively discussed.

The study adds the assessment of the described recycling routes for plastics to existing literature. Mechanical recycling has already been compared to waste incineration with energy recovery and landfilling from an environmental perspective and identified as beneficial (Chen et al., 2011; Turner et al., 2015; Gu et al., 2016a, 2016b, 2017; Van Eygen et al., 2018, 2018b). Few studies compare mechanical recycling with chemical recycling processes (Perugini et al., 2005; Bergsma, 2019a, 2019b; Jeswani et al., 2021). They focus on environmental indicators and do not include an economic comparison of the technologies. Also, some of the assessment data are not publicly available, so the reproducibility of the calculations is limited. Concluding from this, the study contributes to the literature in three aspects:

[1] In Study A, GWP and climate change impact of the processes were used synonymously. In order to be consistent with the formulation of the study, this summary also refers to GWP instead of climate change impact.

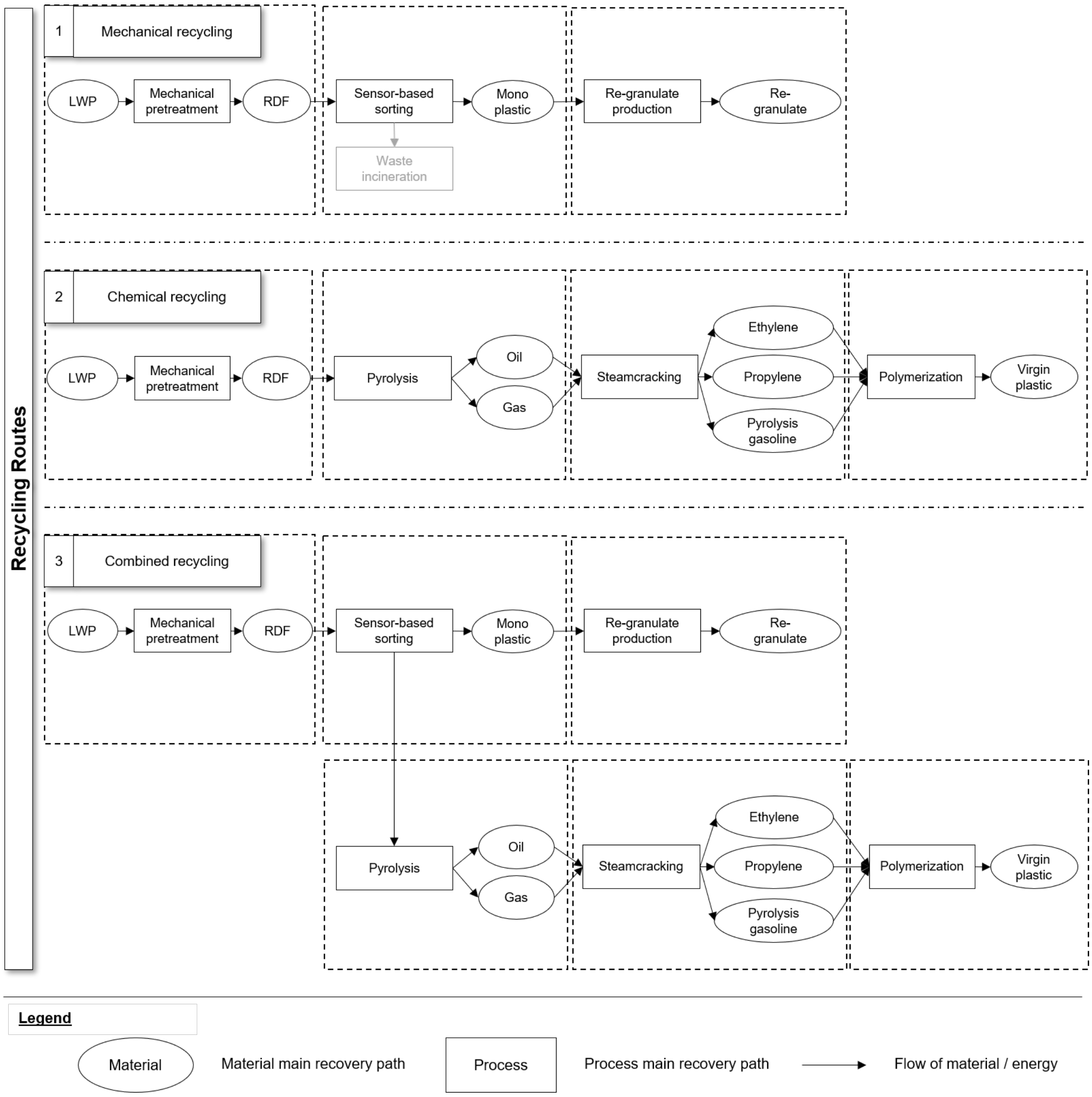


Figure 4.1: System boundaries for the assessed LWP recycling routes.

1. The techno-economic and environmental assessment of the described recycling options for plastics concerning the indicators of processing costs, GWP, CED, and carbon efficiency is presented transparently. The assessment is based on different scenarios, and the underlying data is published.

2. Recycling options are considered using literature data in the overall context of industrial-scale recycling routes.

3. The assessment is based on a waste composition containing interfering materials such as minerals, metals, and organic materials.

Results and Discussion

Economic and environmental assessments were conducted for the scenarios in Table 4.1. The scenarios are grouped based on the assessed recycling route. The chemical recycling data set is not sufficient enough to investigate different scenarios.

Table 4.1: Scenario overview of recycling routes assessment for LWP waste. The scenarios are grouped according to the recycling route: Mechanical recycling (MR), chemical recycling (CR), and the combined (CO) recycling route.

Scenario No.	Recycling route	Sorting yield	Incineration paths of sorting residues
1.1.1	MR	42%	100% MSWI plant
1.1.2	MR	42%	25% MSWI plant, 75% RDF power plant
1.1.3	MR	42%	18% MSWI plant, 58% RDF power plant, 13% cement plant, 11% coal-powered plant
1.2.1	MR	22%	100% MSWI plant
1.2.2	MR	22%	25% MSWI plant, 75% RDF power plant
1.2.3	MR	22%	18% MSWI plant, 58% RDF power plant, 13% cement plant, 11% coal-powered plant
2	CR	-	-
3.1	CO	42%	-
3.2	CO	22%	-

The economic comparison of the recycling routes (cf. Figure 4.2) shows that chemical recycling is economically favorable over mechanical recycling with waste incineration of the sorting residues if revenues for substituting primary material and energy are considered. The gross costs of chemical recycling are higher than those of the mechanical recycling route. However, chemical recycling produces more secondary plastics, generating higher revenues than mechanical recycling. These higher revenues offset the higher processing costs. The recycling route combining mechanical and chemical recycling generates even higher revenues as the yield of secondary plastics can be maximized. This results in the best economic performance, even though the gross costs of the combined approach are higher than for the mechanical recycling route.

The environmentally preferable option depends on the waste sorting success. With a high waste sorting success[2] and, respectively, a low amount of sorting residues that need to be incinerated, the mechanical recycling route is associated with a lower GWP than chemical recycling. The environmental performance is subject to the waste incineration facilities and their efficiency. Figure 4.3 provides an overview of the GWP assessment. It shows that the combined approach is, in any case, associated with a lower GWP than the individual technologies since emissions from waste incineration are avoided, and credits for substituting primary plastics can be maximized. The same dependencies and results appear for the indicators of CED (cf. Figure 4.4) and carbon recycling rate.

[2] The high waste sorting success corresponds to the current sorting success in Germany.

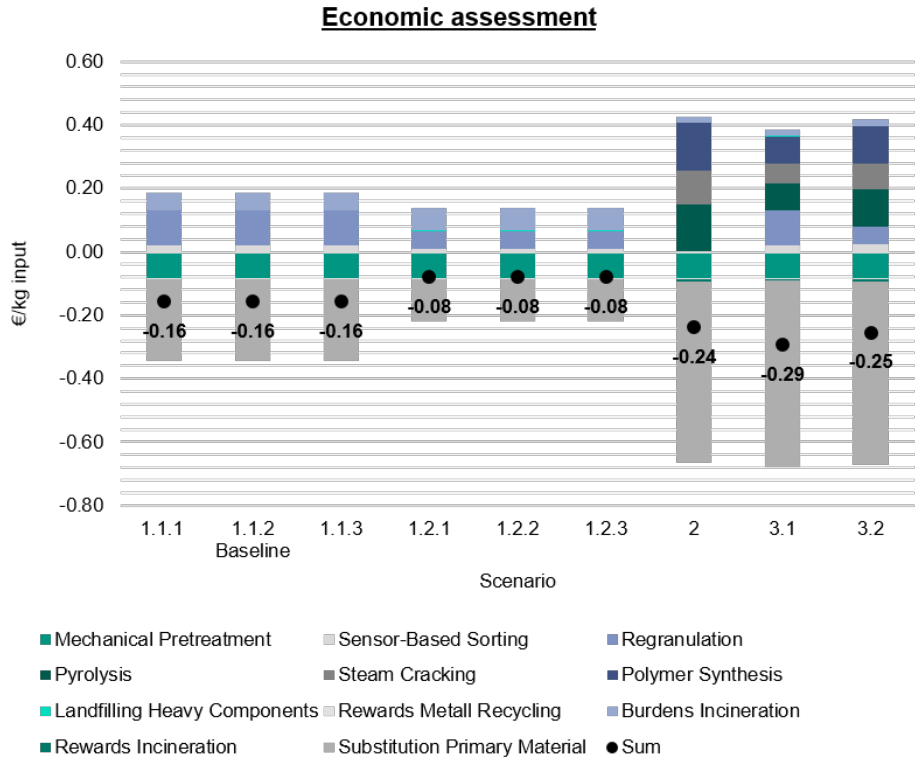


Figure 4.2: Economic assessment of EoL paths and scenarios with costs above the x-axis and revenues beneath it. Assessment for 1 kg of input waste.

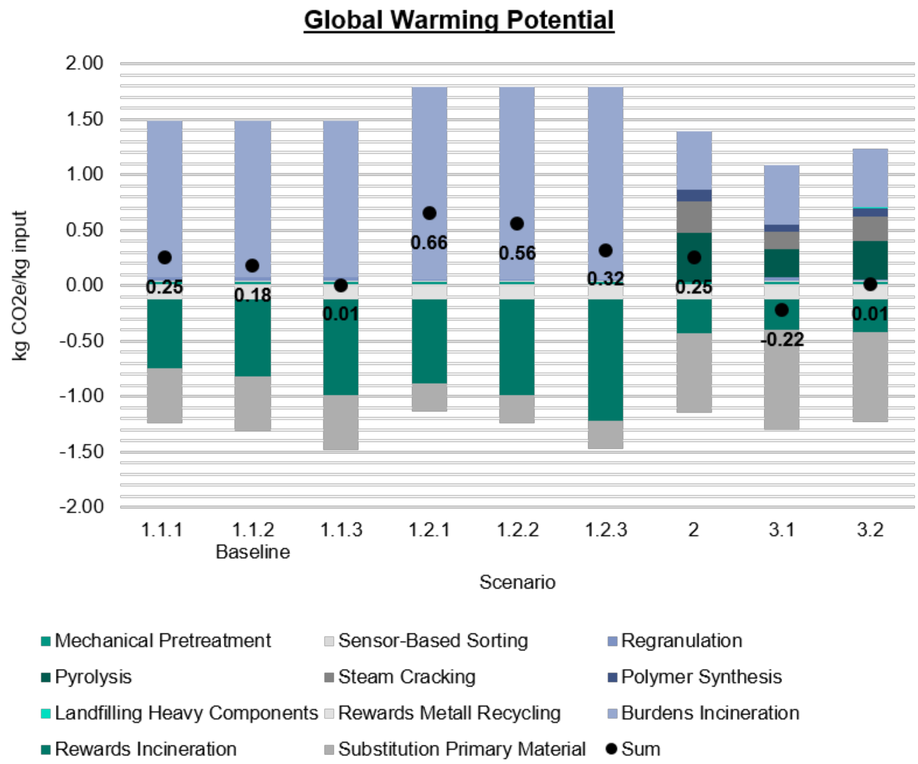


Figure 4.3: Comparison of considered EoL paths and scenarios regarding their net GWP impact. Assessment for 1 kg of input waste. Burdens are above the x-axis, and credits are beneath it.

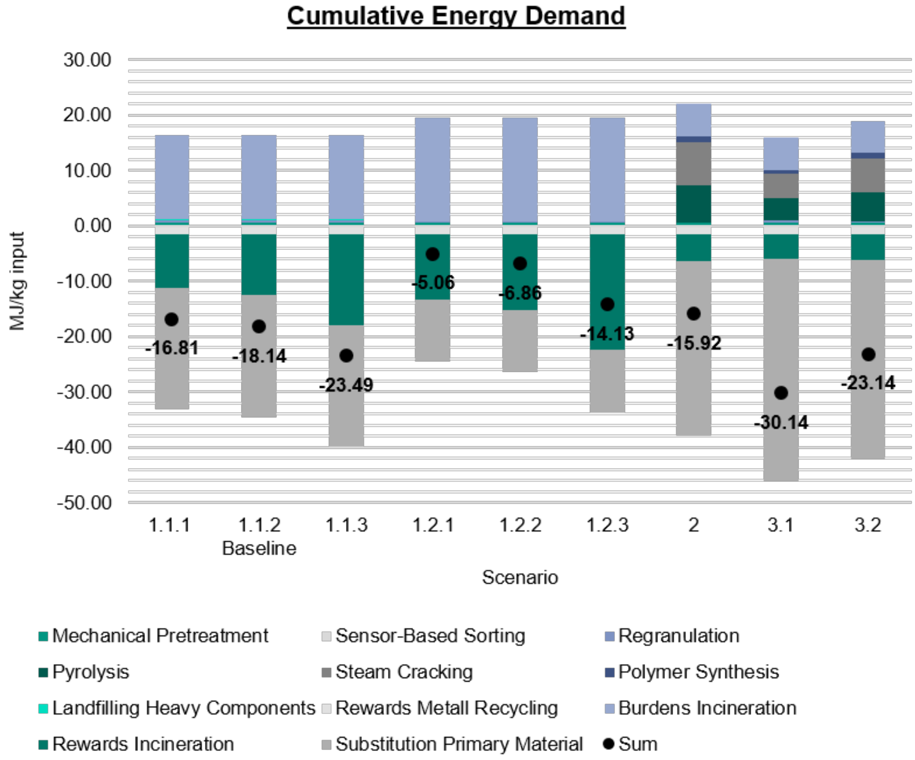


Figure 4.4: Comparison of considered EoL paths and scenarios regarding their net CED impact. Assessment for 1 kg of input waste. Burdens are above the x-axis, and credits are beneath it.

The calculations were rerun with a correction factor to account for differences in the quality of secondary plastics from mechanical and chemical recycling. The credits for substituting primary material for mechanical recycling were reduced.

Chemical recycling is associated with lower GHG emissions than all mechanical recycling scenarios if the initial material substitution credits are reduced by more than 50%. However, the combined recycling approach is still associated with the lowest emissions. For the CED, the threshold is a reduction of more than 35% of the initial credits. Chemical recycling performs better than the combined recycling approach if the initial material substitution credits are reduced by more than 65%.

The results show the advantageousness of combining the different technologies to recycle waste currently recovered as energy. The result is consistent with the existing literature (Chen et al., 2011; Perugini et al., 2005).

There are some limitations to the study. The study is based on literature that needs validation through experiments. Furthermore, the assessment refers to the reference year 2019 and thus does not consider recent price developments in the energy markets. The assessment is a case study for Germany, which means that input parameters from Germany were used. Assumptions like the sorting yield at the sensor-based sorting plant or the assessed energy recovery routes introduce uncertainties into the calculations, which are only partially addressed in scenarios and the sensitivity analysis performed.

Despite these limitations, the study provides a transparent economic and environmental assessment of different recycling routes for LWP and highlights the critical parameters of the assessment. This is essential to understanding the role of chemical recycling in the circular economy of plastics.

4.2 Study B: The Impact of Product Quality on assessing Recycling Technologies

This section refers to the article "The impact of secondary materials' quality on assessing plastic recycling technologies." This article was written in collaboration with Rebekka Volk and Frank Schultmann and was published in the Conference Proceedings of the Life Cycle Management Conference 2021 as Stallkamp et al. (2022).

Study Context and Contributions

Studies comparing recycling processes for plastics (Volk et al., 2021; Jeswani et al., 2021) expiring difficulties assessing the quality of the secondary materials. Current mechanical recycling processes face challenges such as non-polymer impurities, polymer cross-contamination, degradation, and additives affecting the material (Pivnenko et al., 2015). A result is a possible harm to the quality of the secondary material (Hahladakis et al., 2018) and downcycling. Chemical recycling can prevent downcycling and produce secondary plastics with primary quality (Davidson et al., 2021). When comparing recycling technologies, these quality differences between their secondary products must be considered.

Therefore, this study discusses approaches to assess material qualities. It also presents two approaches to integrate material quality into the assessment of recycling processes in more detail: the material substitution rate (European Commission, 2018) in LCAs and a circularity potential developed by Eriksen et al. (2019).

The material substitution rate within the avoided burden approach of LCAs considers that the amount of primary material that can be substituted by secondary material depends on the quality of the secondary material, thus capturing downcycling (European Commission, 2018). For this purpose, the quality ratio of the secondary and primary materials is defined (European Commission, 2018).

The circularity potential of Eriksen et al. (2019) also quantifies the quality of the secondary material. The market share of the secondary material is established and compared to the market share of the primary material. The circular economy potential is the ratio of market shares multiplied by the material losses of the recycling process.

The assessment approaches are compared, and the effect of secondary materials' quality is discussed in a German case study and the mechanical and chemical recycling of HDPE from LWP.

Results and Discussion

A literature search shows no standardized definition of material quality and how it is assessed. Figure 4.5 provides an overview of properties and indicators for assessing the material quality of secondary plastics. Technical properties and technical functionality are accurate regarding the possible application field of the material but also require the most significant amount of data to be determined. Economic indicators, such as the market values of primary and secondary materials, can be used to estimate material quality. However, economic indicators are subject to fluctuations and might integrate possible rejections of secondary material due to assumed quality issues. If no data is available, the impact of secondary materials' quality can be discussed qualitatively. An appropriate quality level must be defined for this purpose, and the effects of increasing or decreasing this level must be discussed. This demonstrates the range of the results and their uncertainty. All assessment approaches face the challenge of missing standardization when considering the quality of secondary materials.

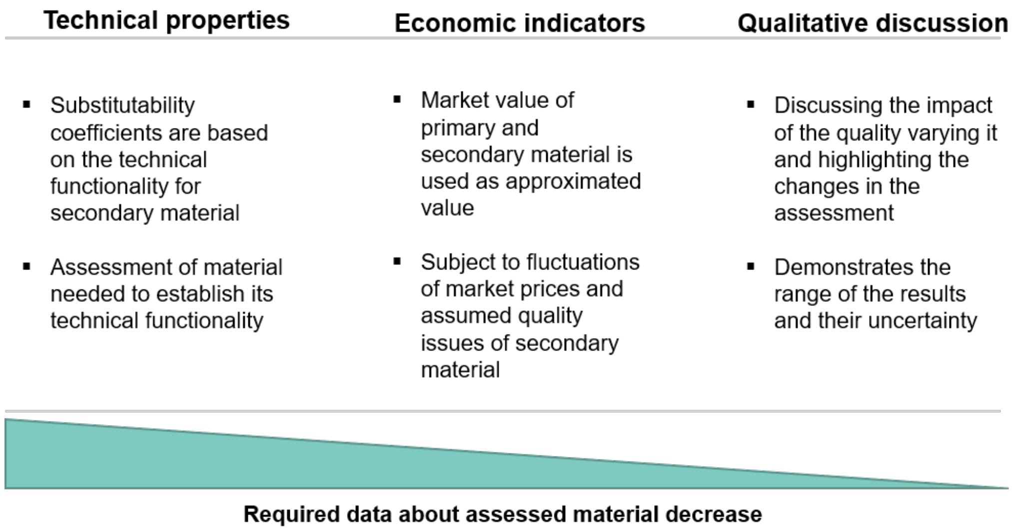
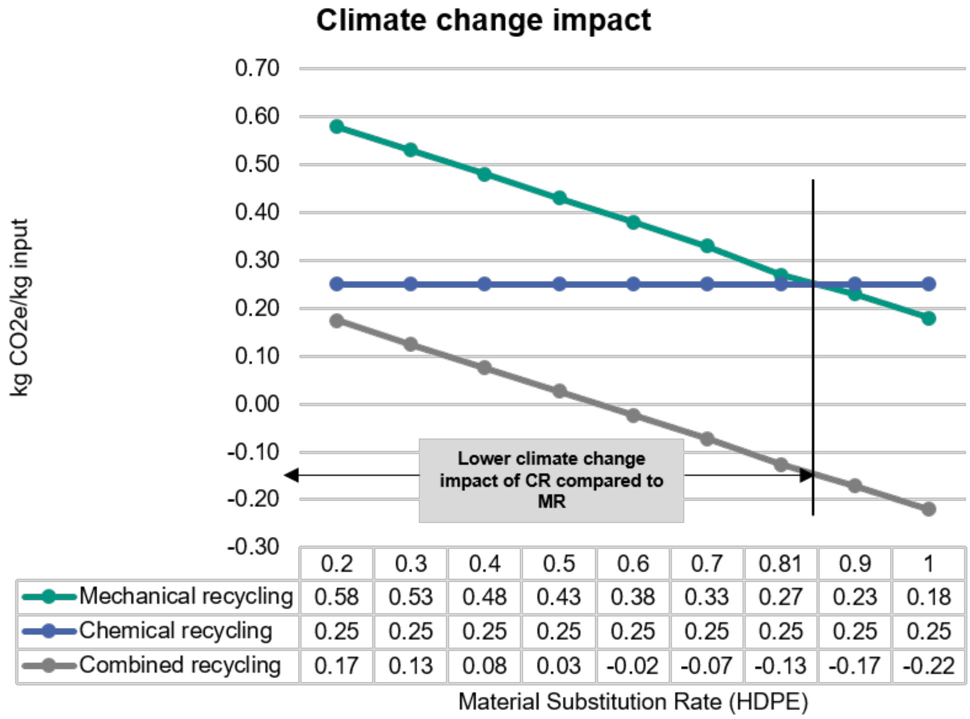


Figure 4.5: Indicators for assessing the material quality of secondary plastics.

The case study with the two indicators of the material substitution rate and the circularity potential shows this difficulty with integrating material quality into the assessment of recycling technologies and expressing quality with a single key figure. The case study compares the mechanical and chemical recycling of HDPE from LWP waste in Germany based on a qualitative discussion. Data for a quality comparison based on technical properties and economic indicators are unavailable.

In the case of quality losses of the secondary material, the environmental assessment of mechanical recycling deteriorates in the LCA with the avoided burden approach. It can change the environmentally preferable recycling option (cf. Figure 4.6). Assuming that 1 kg of mechanically recycled HDPE can only replace 0.81 kg of virgin HDPE due to quality issues, chemical recycling has a lower climate change impact than mechanical recycling. This is due to lower rewards for substituting virgin-like primary HDPE. Combining mechanical and chemical recycling is, in every case, environmentally preferable.



	0.2	0.3	0.4	0.5	0.6	0.7	0.81	0.9	1
Mechanical recycling	0.58	0.53	0.48	0.43	0.38	0.33	0.27	0.23	0.18
Chemical recycling	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Combined recycling	0.17	0.13	0.08	0.03	-0.02	-0.07	-0.13	-0.17	-0.22

Figure 4.6: The climate change impact of the assessed recycling routes depends on the material substitution rate. Results are displayed for HDPE. Based on Volk et al. (2021) (assumptions: $A = 0, R2 = 1, LrecEol = const.$).

Regarding the circularity potential, chemical recycling is preferred in any case (cf. Figure 4.7). The higher resource efficiency[3] and producing virgin-like primary plastic results in a higher circularity potential. This circularity potential is even higher than the potential of the recycling approach combining mechanical and chemical recycling. This demonstrates that the recycling option favorable for the circular economy must not be the environmentally preferable recycling option.

Ultimately, the case study shows the difficulty in assessing the quality of secondary materials through a single indicator and that various indicators must be used to assess recycling options, as these can lead to different conclusions regarding the recycling option to be selected. The study thus emphasizes the need for a standardized view of quality and the use of multiple indicators to assess recycling options. Since publication, further studies have addressed this fundamental challenge and developed a model for the quality assessment of recycled plastics (Golkaram et al., 2022) or examined the technical and market substitutability of mechanically and chemically recycled plastics in more detail (Huysveld et al., 2022).

[3] More material is chemically recyclable, as no plastic mono streams are needed.

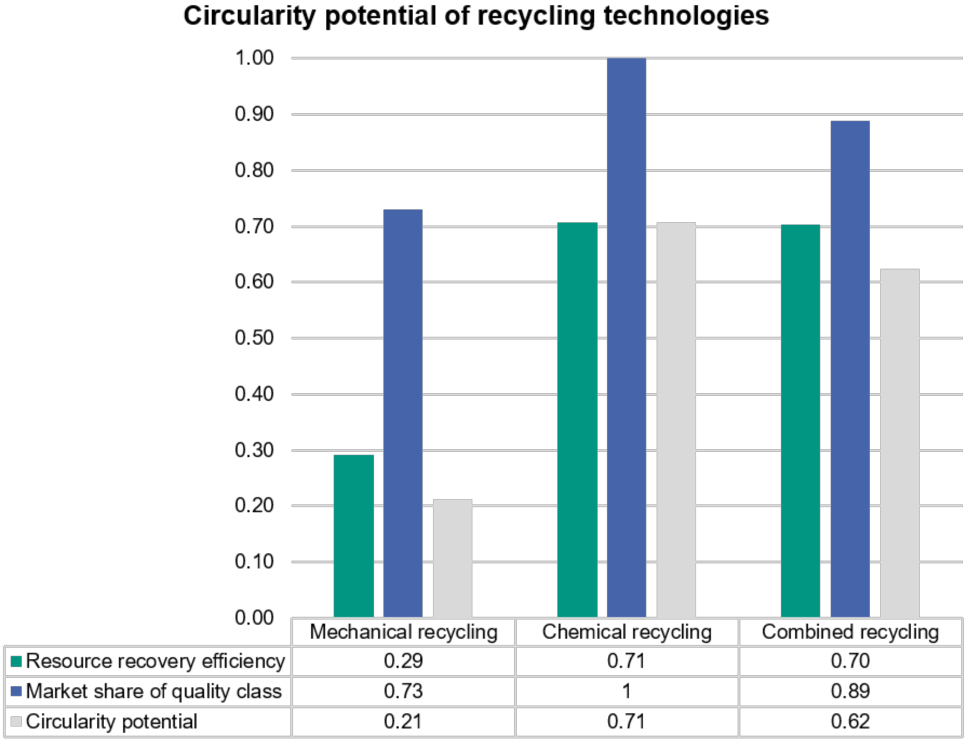


Figure 4.7: Circularity potential of the assessed recycling technologies for HDPE in Volk et al. (2021).

4.3 Study C: Economic and Environmental Assessment of APW EoL Options

The following section refers to the article "Economic and environmental assessment of automotive plastic waste EoL options — energy recovery versus chemical recycling." This article was written in collaboration with Malte Hennig, Rebekka Volk, Frank Richter, Britta Bergfeldt, Salar Tavakkol, Frank Schultmann, and Dieter Stapf. It was published in the Journal of Industrial Ecology as Stallkamp, Hennig, Volk, Richter, et al. (2023).

Study Context and Contributions

The study researches the technical feasibility of the pyrolysis of engineering plastics from the automotive sector by conducting pyrolysis experiments with a sample from actual APW. Thus, the study addresses the suitability of chemical recycling to close the material loop for the heterogeneous waste stream of APW. Waste characterization, mass balances, and product composition of pyrolysis gas and pyrolysis oil are reported for transparency.

In a second step, the study integrates the data from the conducted pyrolysis experiments into the economic and environmental assessment of a chemical recycling route for APW. It compares the chemical recycling route to the current waste-handling practice of waste incineration with energy recovery. The chemical recycling route includes upstream mechanical pretreatment of the APW, pyrolysis, and downstream upgrading of the liquid pyrolysis product to the specifications of steam cracker feedstock, followed by high-value chemical production through steam cracking. For the up and downstream processes, literature data are used. Figure 4.8 and Figure 4.9 outline the system boundaries of both assessed waste treatment options.

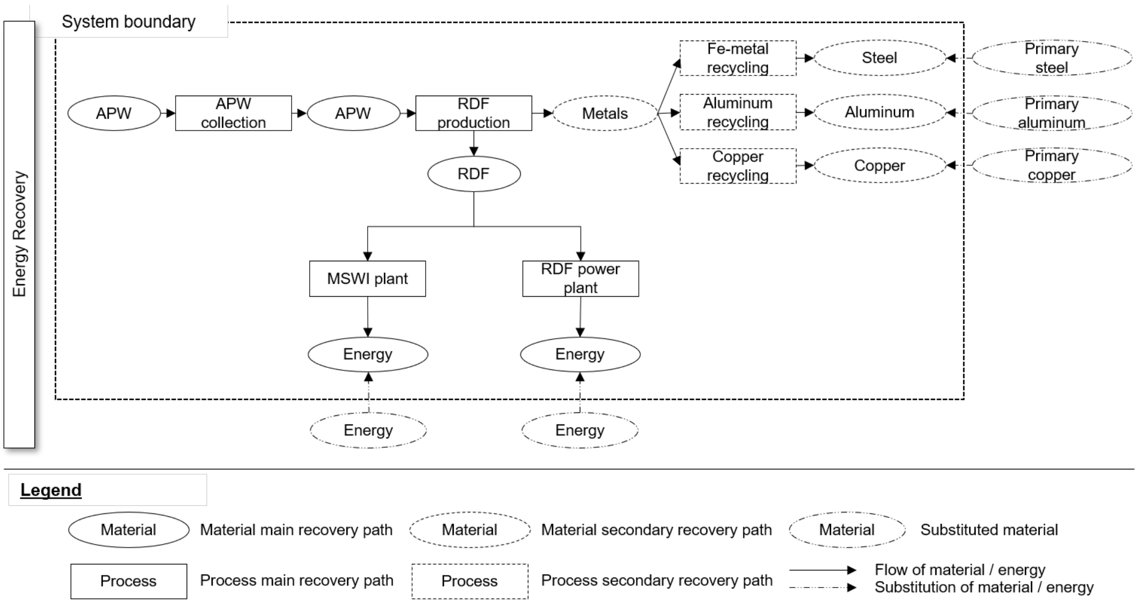


Figure 4.8: System boundaries for the energy recovery route, including primary material and energy substitution.

The economic and environmental assessment combines MFCA with LCA methods (Rieckhof and Guenther, 2018). Due to missing data, e.g., elemental flows, the LCA is streamlined by narrowing the considered environmental impacts to climate change and CED (Gradin and Björklund, 2021). Scenarios are used to analyze the influence of underlying data and assumptions on the assessment (cf. Table 4.2).

Table 4.2: Scenario overview of the assessment of waste treatment options for APW.

Scenario No.	Scenario Description	Description
1.1	Energy recovery (baseline)	Incineration path of produced RDF: 30% MSWI plant, 70% RDF combustion plant.
1.2	Energy recovery (optimized)	Incineration path of produced RDF: 100% RDF combustion plant.
2.1	Chemical recycling (baseline)	Yield of pyrolysis products according to conducted experiments[1]: 50% pyrolysis oil, 20% pyrolysis gas, 28% pyrolysis residue, 2% aqueous condensate.
2.2	Chemical recycling (lower yield)	Yield of pyrolysis products adapted: 45% pyrolysis oil, 20% pyrolysis gas, 34% pyrolysis residue, 2% aqueous condensate.[1]

[1]: Pyrolysis product distribution converted to a feedstock free of metals.

Consequently, this study contributes to understanding different waste treatment options for automotive plastics and their contribution to a circular economy. The potential of the chemical recycling of

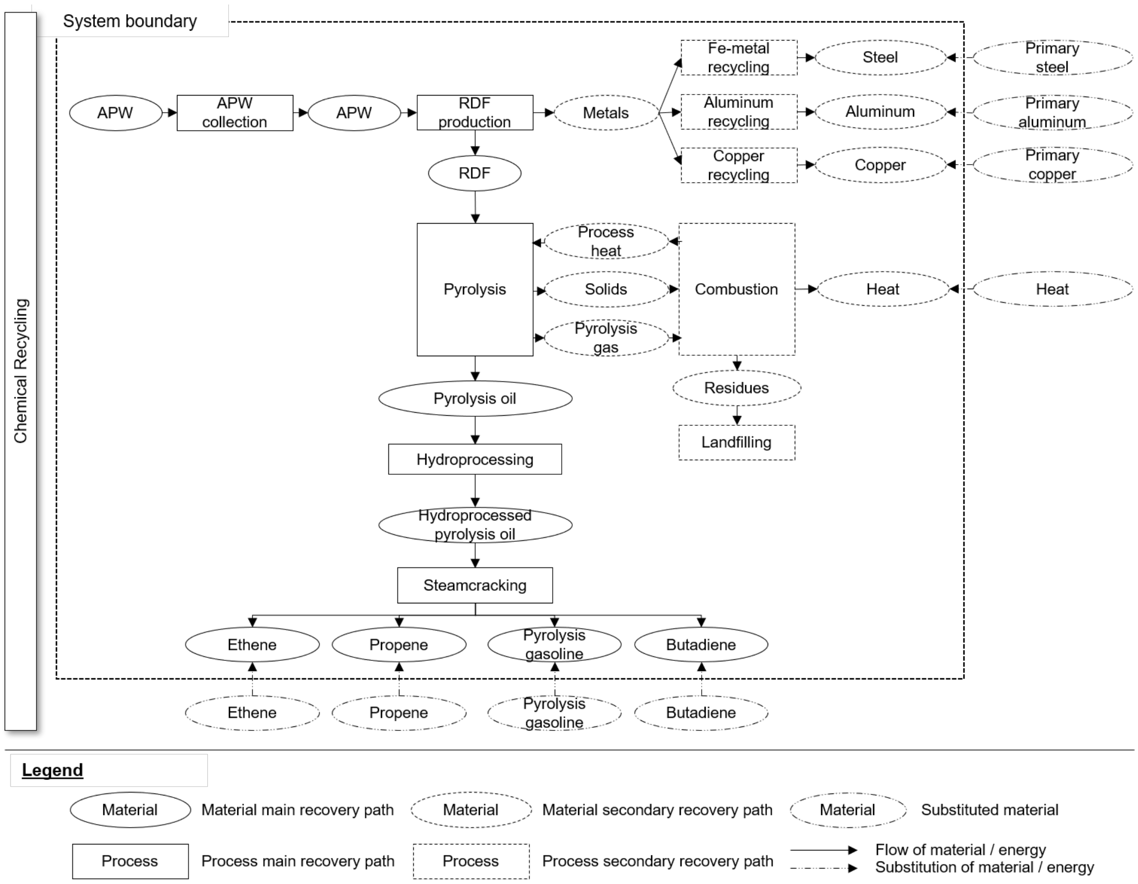


Figure 4.9: System boundaries for the chemical recycling route, including primary material and energy substitution.

APW to keep carbon in the economic material cycle and contribute to achieving climate neutrality is established.

Results and Discussion

Five experiments were run with actual APW. Respective mass distributions of the pyrolysis products can be taken from Figure 4.10. The average shows that about 45 wt% of the waste was converted into pyrolysis oil, while 2 wt% was converted into an aqueous condensate. Pyrolysis gas comprised 18 wt%, while 31 wt% was retained as pyrolysis residue, consisting of mineral fillers, glass fibers, and metals. On average, 4 wt% of the sample weight was lost (balance loss) due to encrustations within the reactor and measuring inaccuracies. Elemental analysis of condensates showed that additional upgrading steps are needed to reach steam cracker specifications.

A preliminary life cycle inventory (LCI) is derived from the pyrolysis experiments and is used to assess the chemical recycling route (cf. Table 4.3). The LCI comprises the conversion of RDF to pyrolysis oil, pyrolysis gas, and pyrolysis residues. While the pyrolysis oil is sent to a hydroprocessing unit for upgrading to steam cracker specifications, the pyrolysis by-products are incinerated. The incineration of by-products supplies the heat demand of the pyrolysis unit. Excess heat is used to generate district heating. The remaining ashes are landfilled.

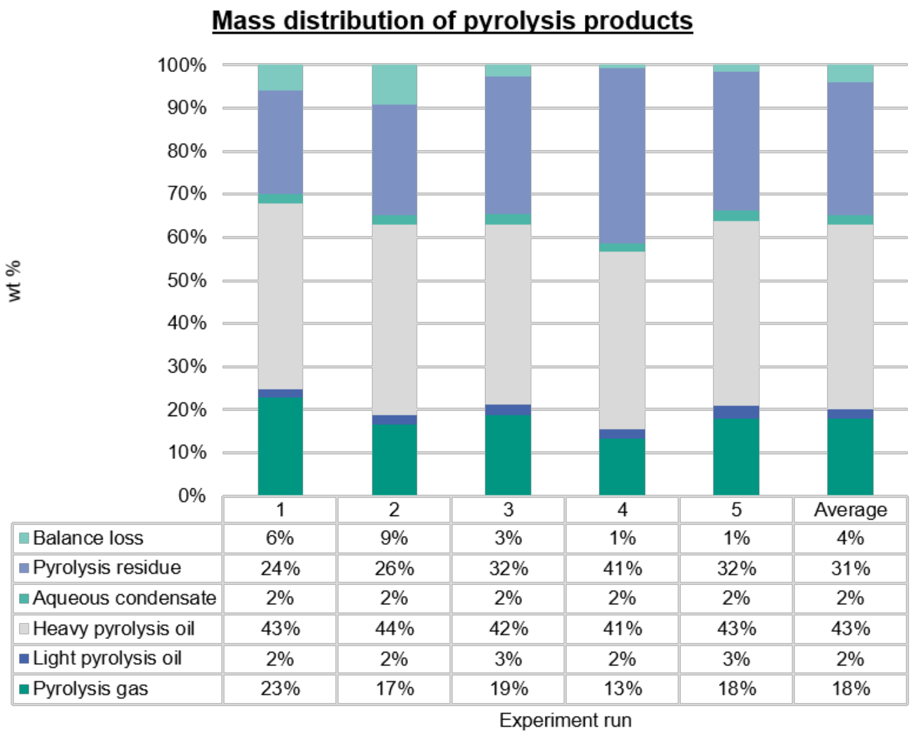


Figure 4.10: Mass distribution of pyrolysis products in experimental runs under similar conditions.

Table 4.3: LCI Data of APW pyrolysis and coupled energy recovery from pyrolysis by-products.

Input		Ouput	
Item	**Quantity**	**Item**	**Quantity**
Pyrolysis			
RDF (t)	0.91	Pyrolysis oil (t)	0.45
Process heat for pyrolysis (kWh)	997	Pyrolysis by-products (t)	0.46
Energy recovery from pyrolysis by-products			
Pyrolysis by-products (t)	0.46	Ashes (t)	0.14
Combustion air (t)	2.30	CO_2 in flue fas (t)	0.63
Pyrolysis by-products (kWh)	1806	Flue gas (others) (t)	1.97
		Process heat for pyrolysis (kWh)	997
		District heat (kWh)	610
		Heat losses (kWh)	199

Including the generated data in the chemical recycling route process assessment enables the comparison with waste incineration with energy recovery. The indicators are shown in Figures 4.11, 4.12, and 4.13 for the different scenarios. 4.14 provides an overview of the material and carbon flow of the chemical recycling route.

The economic comparison (cf. Figure 4.11) shows that waste incineration with energy recovery performs economically better than chemical recycling. This is due to high rewards for producing and substituting energy and lower gross processing costs due to fewer processing steps along the value chain. The efficiency of the incineration influences the result. Economically, a lower pyrolysis oil yield results in lower net processing costs along the chemical recycling route. High energy prices result in higher revenues from recovered energy from waste incineration than from produced HVCs.

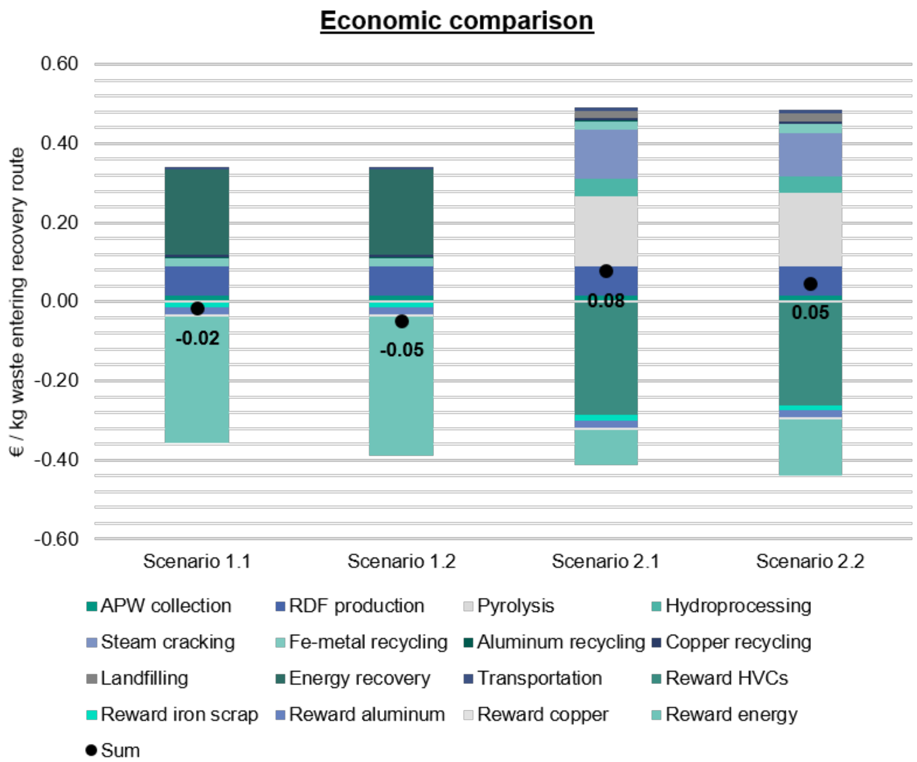


Figure 4.11: Economic assessment of EoL paths and scenarios with costs above the x-axis and revenues beneath it. Assessment for 1 kg of input waste.

The waste incineration paths and their efficiencies influence the climate change impact of the energy recovery scenarios (cf. Figure 4.12). Regarding climate change, chemical recycling performs considerably better than waste incineration with energy recovery. Rewards for substituting primary high-value chemicals counterbalance the high impacts of steam cracking and pyrolysis.

The CED impact of waste incineration with energy recovery decreases with more efficient waste incineration paths as more electricity and heat can be recovered (cf. Figure 4.13). Both chemical recycling scenarios show lower CED impacts than waste incineration with energy recovery.

The CED savings decrease with decreasing pyrolysis oil yield as fewer high-value chemicals are produced.

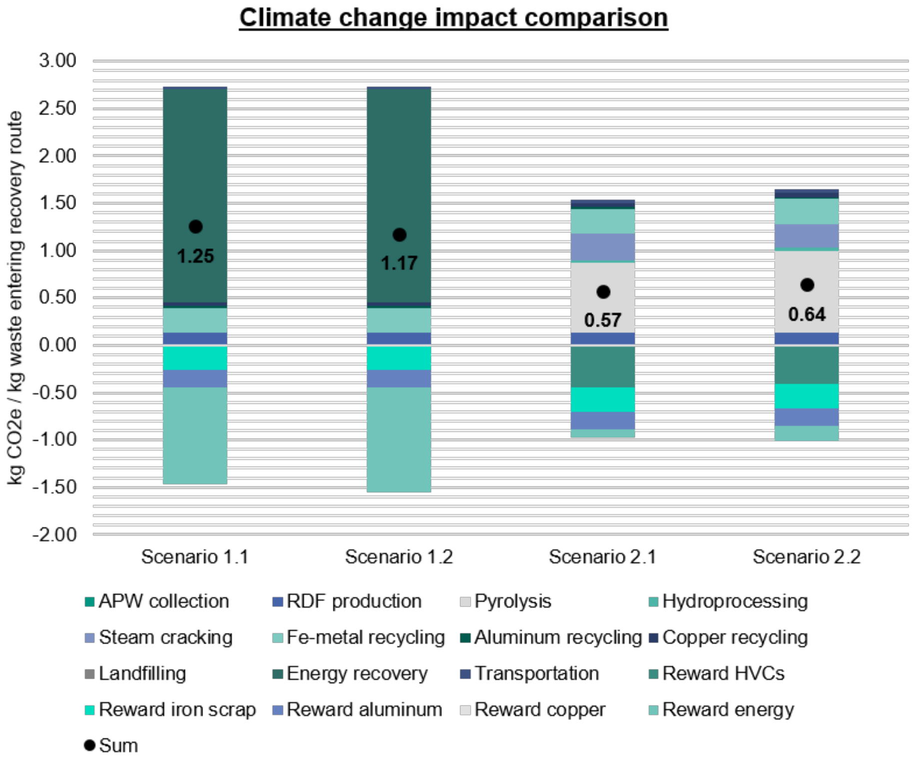


Figure 4.12: Comparison of EoL paths and scenarios regarding their net climate change impact. Assessment for 1 kg of input waste. Burdens are above the x-axis, and credits are beneath it.

In addition to the environmental benefits, chemical recycling can contribute to a circular economy closing the carbon cycle (cf. Figure 4.14). Mass and elemental balances for carbon recovery demonstrate that a high pyrolysis oil yield is advantageous when contributing to a circular economy.

There are some limitations to this study. The study assesses a defined waste stream of APW from workshop repair jobs with a specific composition to which the results are limited. This waste fraction has a low volume compared to automotive shredder residues. Mechanical recycling options and a combined mechanical and chemical recycling approach are excluded from the study. Data for the hydroprocessing of the pyrolysis oil is unavailable yet, and very general assumptions had to be used. Other assumptions also introduce uncertainties into the calculations, which are only partially addressed in scenarios and the sensitivity analysis performed.

Despite these limitations, the study provides transparent documentation of pyrolysis experiments that demonstrate the technical feasibility of the pyrolysis of APW containing engineering thermoplastics. Additionally, it provides a transparent economic and environmental assessment of different recycling routes for APW and highlights the critical parameters of the assessment. It identifies a conflict between the economic and environmentally preferable EoL options for APW.

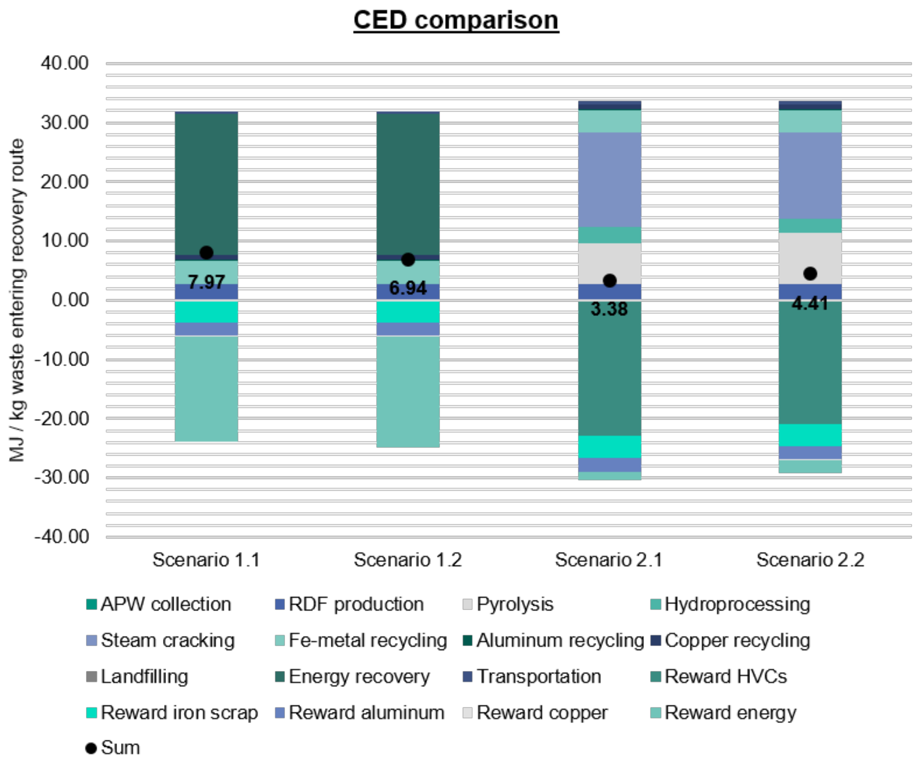


Figure 4.13: Comparison of EoL paths and scenarios regarding their net CED impact. Assessment for 1 kg of input waste. Burdens are above the x-axis, and credits are beneath it.

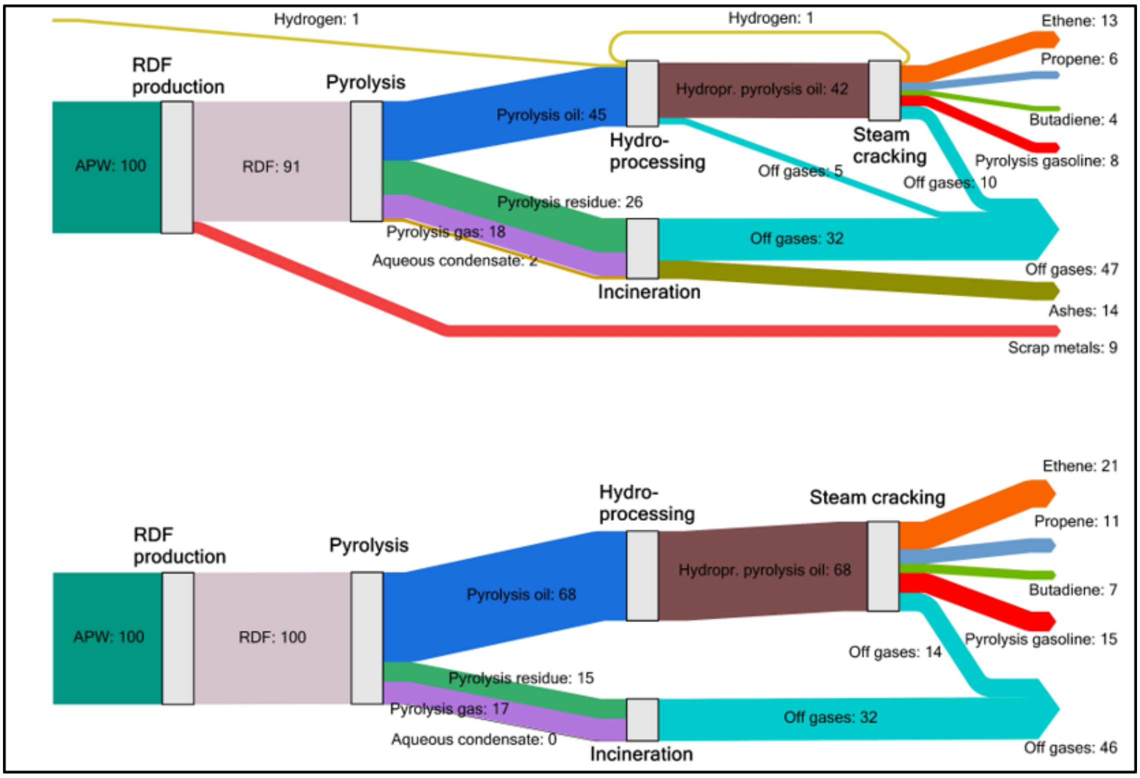


Figure 4.14: Material flow (top) and carbon flow (bottom) of the chemical recycling (scenario 2.1). Numbers in percent (rounded) of APW input.

4.4 Study D: Techno-economic Assessment of a Pyrolysis Plant for Automotive Plastic Waste

This section refers to the article "Techno-economic assessment of pyrolysis plants for automotive plastic waste." This article was written in collaboration with Malte Hennig, Rebekka Volk, Dieter Stapf, and Frank Schultmann and is submitted to a scientific journal as Stallkamp, Hennig, Volk, Stapf, and Schultmann (2023).

Study Context and Contributions

The study investigates the economics of APW pyrolysis using a TEA. Few studies assess the economics of plastic waste recycling via pyrolysis for mixed plastic waste containing a high share of polyolefins (Westerhout et al., 1998; Sahu et al., 2014; Fivga and Dimitriou, 2018; Jiang et al., 2020; Larrain et al., 2020; Riedewald et al., 2021). The economic assessment of the pyrolysis of more demanding plastic waste streams like APW has yet to be considered.

APW is generated during repairs of vehicles and at their EoL. Due to the lack of repair data, the volume of APW in Germany is estimated based on EoL vehicles. A conservative estimation based on UBA (2022) results in an APW volume of around 1,380 t in 2019. This low waste amount can increase to 8,280 t if the dismantling of large plastic components becomes part of automobiles' EoL treatment processes (Wilts et al., 2016). Also, mixed plastic waste fractions separated from ASR by post-shredder-treatment processes have shown similar behavior in pyrolysis (Zeller et al., 2021). They increased the potential waste feedstock to around 10,000 t in 2019.

In Germany, the APW from workshops is currently used for energy recovery purposes (Cossu and Lai, 2015; Mehlhart et al., 2018), and, therefore, its chemical recycling does not compete with mechanical recycling. The considered pyrolysis plant (cf. Figure 4.15) employs a twin screw reactor concept that has previously been used for the pyrolysis of biomass in the production of green fuels (Campuzano et al., 2019; Henrich et al., 2016; Kapoor et al., 2020). The concept can be adapted for recycling plastic waste (Zeller et al., 2021).

The main product of the recycling process is a pyrolysis oil, while the pyrolysis gas is used in a combined heat and power unit to recover heat and electricity. The generated electricity is used to provide the electrical energy demand of the plant. Surplus electricity exceeding the plant's demand is sold to the grid operator. If electricity production is insufficient, additional electricity is sourced from the grid. The by-products aqueous condensate and solid residues are considered waste that must be disposed of. However, there might be future use cases for the solid residues. Therefore, two scenarios are calculated: solid residual disposal by co-incineration (1) in waste incineration plants associated with costs, and (2) industrial co-incineration, assuming the generation of a profit from selling the solid residue as fuel.

The equipment and infrastructure (E&I) investment for the pyrolysis plant is calculated based on the plant design and a list of equipment needed, following the capacity estimate approach for all

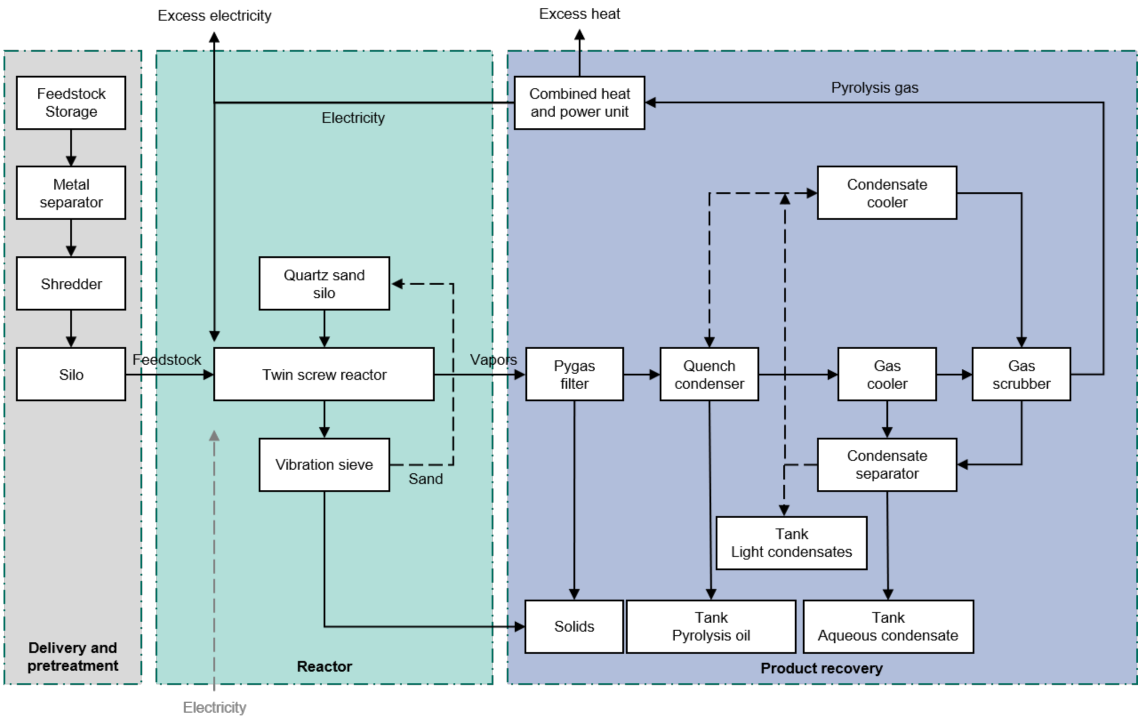


Figure 4.15: Assessed pyrolysis plant with three modules: (1) delivery and pretreatment, (2) reactor, and (3) product recovery. Plant design and figure are based on Trippe et al. (2010).

standard mechanical and process engineering components (Humphreys, 2004). The components' investment is scaled based on the capacity and component-specific cost-capacity factors. However, reactors like the twin screw reactor have mechanical limitations that do not allow limitless scaling. Therefore, for higher throughputs, it is assumed that additional reactors must be operated following a numbering-up approach. It is assumed that a maximum of four reactors are connected to one product recovery unit to reduce the complexity of the plant design. This results in a second numbering-up stage. The E&I investment is used to calculate the plant's capital expenditures (CAPEX) using an equipment factor method (Peters et al., 2003).

The operational expenditures (OPEX) are partly based on the E&I investment[4] and also on mass and energy balances from pilot-scale pyrolysis experiments published by Stallkamp, Hennig, Volk, Richter, et al. (2023) (Study C) conducted with an actual APW.

Thus, this study examines the economics of the pyrolysis of a technically demanding waste stream that cannot be mechanically recycled. A transparent TEA establishes the minimum selling price for pyrolysis oil and the minimum capacity for a pyrolysis plant at which economic operation is possible. The feedstock supply is analyzed, and possible prices for pyrolysis oil are estimated.

[4] This refers to the maintenance and yearly insurance.

Table 4.4: Overview of the assessed capacity classes of the APW pyrolysis plant.

Capacity class	Capacity [t/year]	Number of reactors
1	3,750	1
2	7,500	2
3	11,250	3
4	15,00	4
5	18,750	5
6	22,500	6
7	26,250	7
8	30,000	8
9	33,750	9
10	37,500	10

Results and Discussion

The CAPEX for ten different capacities are assessed corresponding to the number of reactor modules operated at full load and 7,500 operating hours (cf. Table 4.4). With the capacity increase, the total CAPEX increase disproportionately. When comparing the total CAPEX of capacity 1 with the total CAPEX of capacity 10, a 10-fold increase in capacity results in a roughly 4.5-fold increase in CAPEX (cf. Figure 4.16).

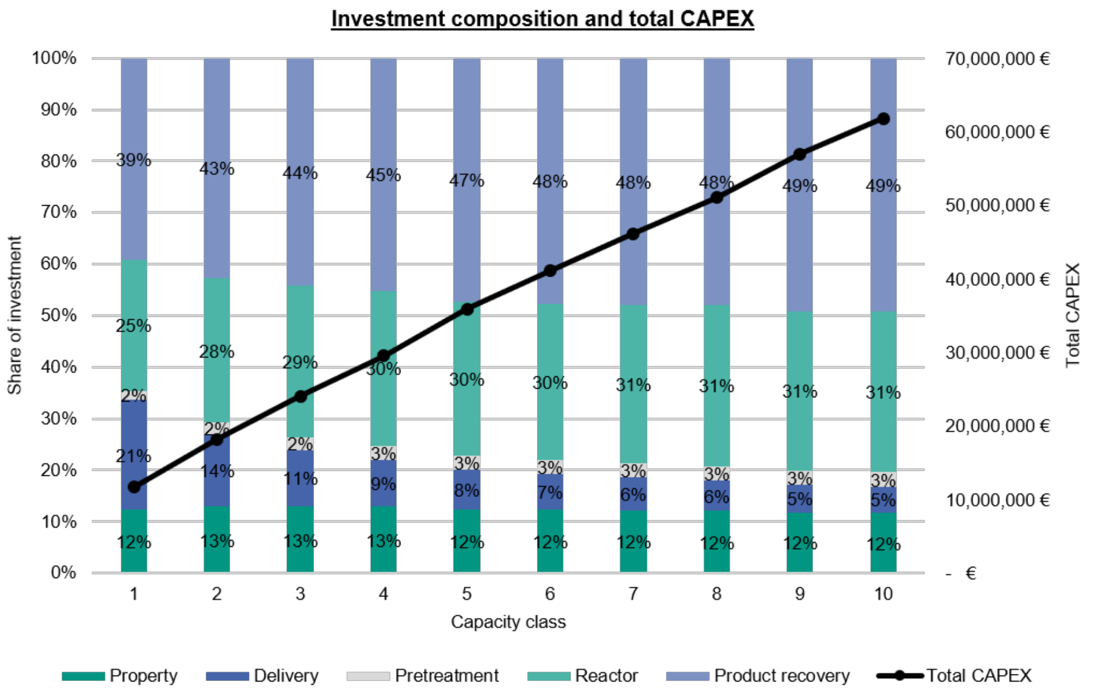


Figure 4.16: Investment composition and total CAPEX of different capacity classes for a pyrolysis plant for APW.

The OPEX are separated into fixed and variable OPEX. The fixed OPEX are independent of the amount of feedstock handled and are based on the size and capacity of the plant. The variable OPEX depend on the amount of feedstock handled and are calculated based on the process flows and mass and energy balances. In scenario 1, the OPEX include the disposal costs for solid residues, while in scenario 2, the payments from selling the solid residues as fuel are considered. There is a decreasing trend in the OPEX for both assessed scenarios (cf. Figure 4.17). The increase in OPEX from capacity 5 to capacity 6 is due to additional personnel needed with the increased plant capacity.

Capital payments and personnel costs dominate the OPEX, while with increasing capacity, the impact of the personnel costs decreases. In scenario 1, the share of disposal costs for the solid residues increases with increased capacity. In scenario 2, the percentage of revenues generated with the solids is consistent. Increasing revenues from feedstock gate fees and generated heat reduce the OPEX with increasing plant capacity.

The minimum sales price for pyrolysis oil (cf. Figure 4.18) is derived from the production costs[5] and a target margin of 15% resulting from the deployment of a new technology and a new product in an existing market (Peters et al., 2003; Riedewald et al., 2021). The minimum sales prices are compared to the average U.S. Residual Fuel Oil price of 462 €/t (EIA, 2022) in 2021.

With increasing plant capacity and production, the production costs and minimum sales prices decrease as relative costs fall due to economies of scale. In scenario 1, the minimum sales price never undercuts the reference price of U.S. Residual Fuel Oil (grey line). However, falling minimum sales prices indicate that higher plant capacities can (almost) economically compete with the reference product. The production costs are below the reference price of U.S. Residual Fuel Oil in capacity 10 with an input of 37,500 t/year. A profit can, therefore, already be made if a part of the 15% margin is sacrificed. In scenario 2, the production costs for the pyrolysis oil fall below the reference price, starting at capacity 8. With a target margin of 15%, the minimum sales price falls below the reference price, starting at capacity 9 with an input of 33,750 t/year.

The TEA demonstrates that higher plant capacities reduce the minimum sales price, enabling a more economical operation. Also, waste should be prevented, and value streams should be generated. This is in line with other economic assessments like Larrain et al. (2020) and Riedewald et al. (2021). However, current pyrolysis experiments run on pilot-scale reactors, and mass and energy balances may be affected by scale-up to a commercial scale. Also, due to the project maturity of the assessed plant, a deviation from the calculated investment is possible. Nevertheless, compared to other studies, the accuracy of the TEA is high due to the numbering-up approach for critical parts of the pyrolysis plant, like the reactor or the product recovery module. A final limitation of the study is the feedstock volume of APW. The results outline that, depending on the use of the by-products, the economic operation of the pyrolysis plants could start at 33,750 t input/year. Therefore, the capacity for an economic operation is greater than the yearly APW estimation for Germany. However, the APW estimation is conservative and does not include APW from workshop repair jobs. There is also the potential that this waste fraction becomes more relevant if the dismantling of plastic components becomes part of automobiles' EoL treatment processes. Also, mixed plastic waste fractions separated from ASR by post-shredder-treatment processes have shown similar behavior

[5] Allocation of the OPEX to the produced quantity of the desired product (pyrolysis oil).

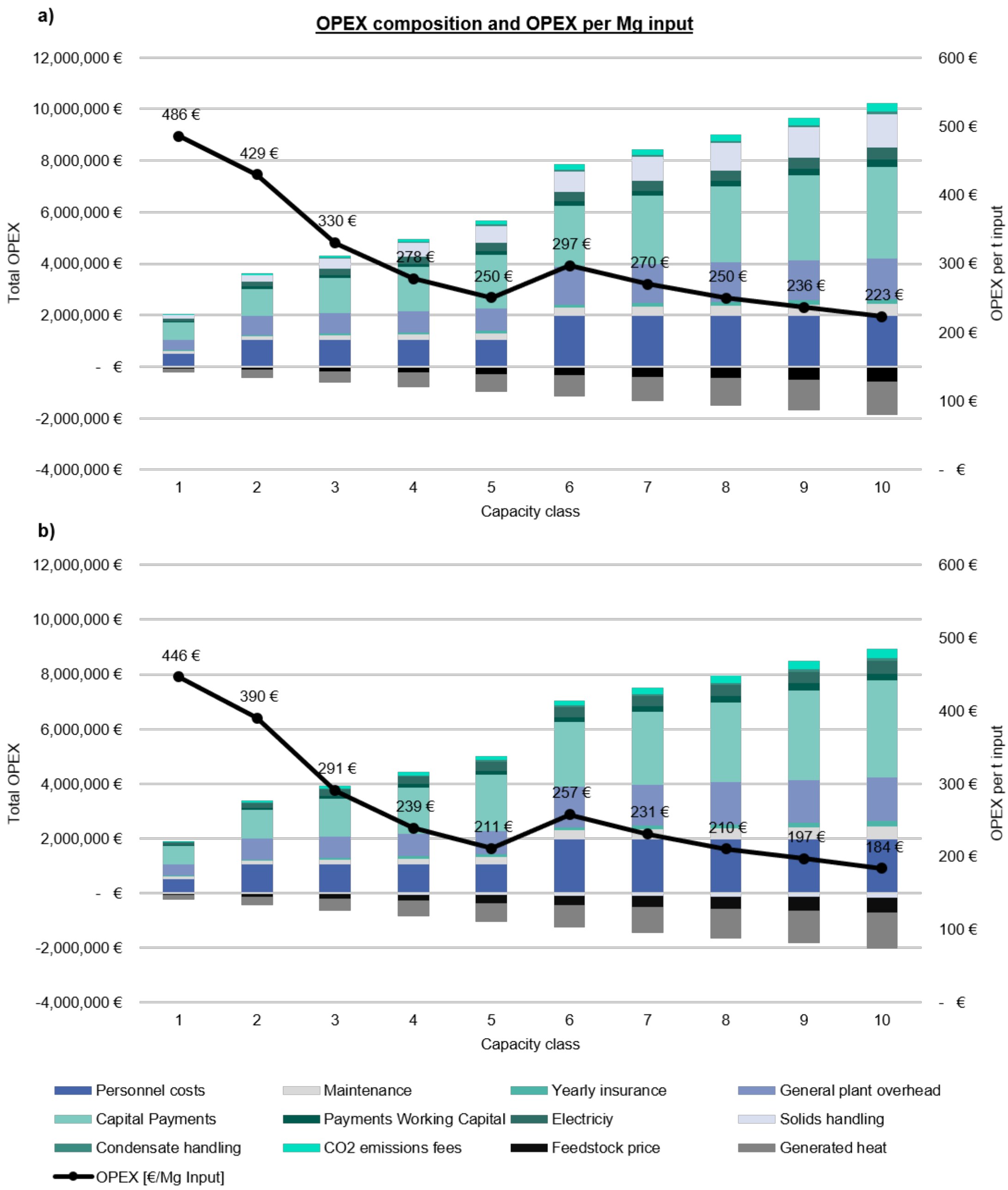


Figure 4.17: OPEX composition and total OPEX per t input for different scales of a pyrolysis plant for APW in scenario 1 (a) and scenario 2 (b).

in pyrolysis as APW. They might be a future feedstock of such plants making their operation more economically attractive. In addition, the assessed reactor technology has proven to be very robust regarding different feedstocks.

Despite these limitations, the study provides a transparent TEA of a pyrolysis plant for APW. The critical parameters for the economic assessment are highlighted, and the minimum sales prices of pyrolysis oil and multiple plant capacities are derived.

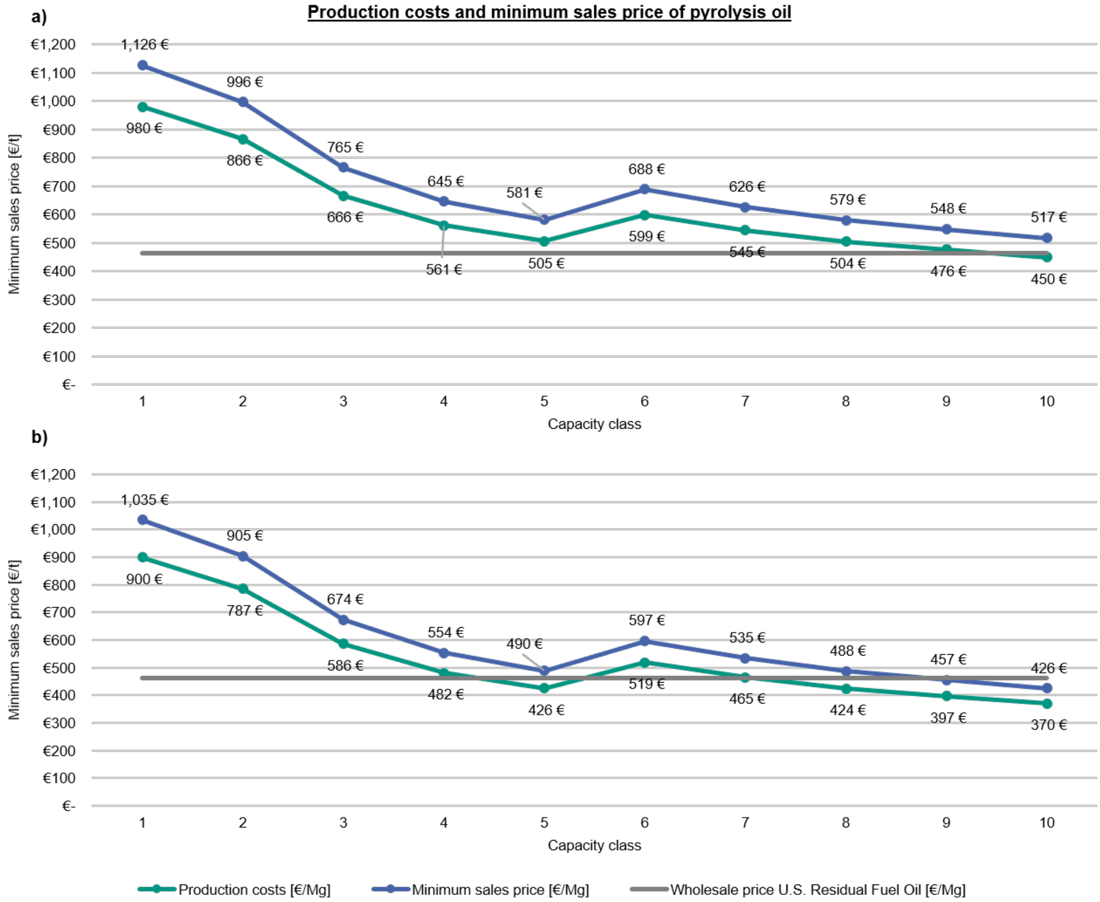


Figure 4.18: Production costs and minimum sales price of pyrolysis oil for different capacity classes of a pyrolysis plant for APW in scenario 1 (a) and scenario 2 (b).

4.5 Study E: Decision Support for Recycling Network Designs in a Plastic Circular Economy

The following section refers to the article "Circular Economy for Plastics in Germany: Decision Support for Optimal Recycling Network Designs." This article was written in collaboration with Rebekka Volk and Frank Schultmann and is submitted to a scientific journal as Stallkamp, Volk, and Schultmann (2023).

Study Context and Contributions

The study's objective is to combine environmental and economic analyses of the chemical recycling of plastic waste and integrate them into a location decision model within the German plastic waste treatment network. Modeling the waste treatment network allows the support of decision-makers in choosing steering strategies and designing regulatory frameworks supporting a plastic circular economy and decarbonizing waste treatment (Sommer et al., 2022).

Valenzuela et al. (2021) provide an overview of waste treatment models in plastics recycling, including models that relocate network facilities or multi-objective formulations minimizing costs and climate change impact. However, no studies include extensions of waste handling options of chemical recycling using pyrolysis.

Volk et al. (2021) (Study A) and Stallkamp, Hennig, Volk, Richter, et al. (2023) (Study C) compare the economic and environmental performance of the pyrolysis of LWP sorting residues and APW to other EoL options. This study combines these economic and environmental assessments with methods from Operations Research to model a part of the current German waste treatment network for LWP sorting residues and APW focusing on the energy recovery infrastructure (Model 1). Moreover, it compares its processing costs and climate change impact to an optimized network design integrating chemical recycling plants (Model 2). Model 2 considers infrastructural preconditions, potential economies of scale, and the material flow on a national level in 2021. It is extended to a multi-objective location model minimizing total network costs and climate change impact to assess how the network structure changes when both objectives are included in the optimization (Model 3). For this purpose, a goal programming approach minimizes the distance between the optima and the individual optimizations with equal weighting.

In addition, different political steering strategies to establish chemical recycling by pyrolysis are analyzed and discussed. This supports political decision-makers aligning environmental and economic interests and supporting the development of a circular economy for plastics.

The locations, numbers, and capacities of the pyrolysis plants are variables that the model determines. The placement of the pyrolysis plants minimizes costs while transport distances between plants and costs of waste conversion are considered. Discrete plant capacity classes are assumed since upscaling is usually done by adding single reactors to the plant design (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023) (Study D). In addition to material flow conservation constraints, the capacities of the individual plants must not be exceeded (cf. Figure 4.19).

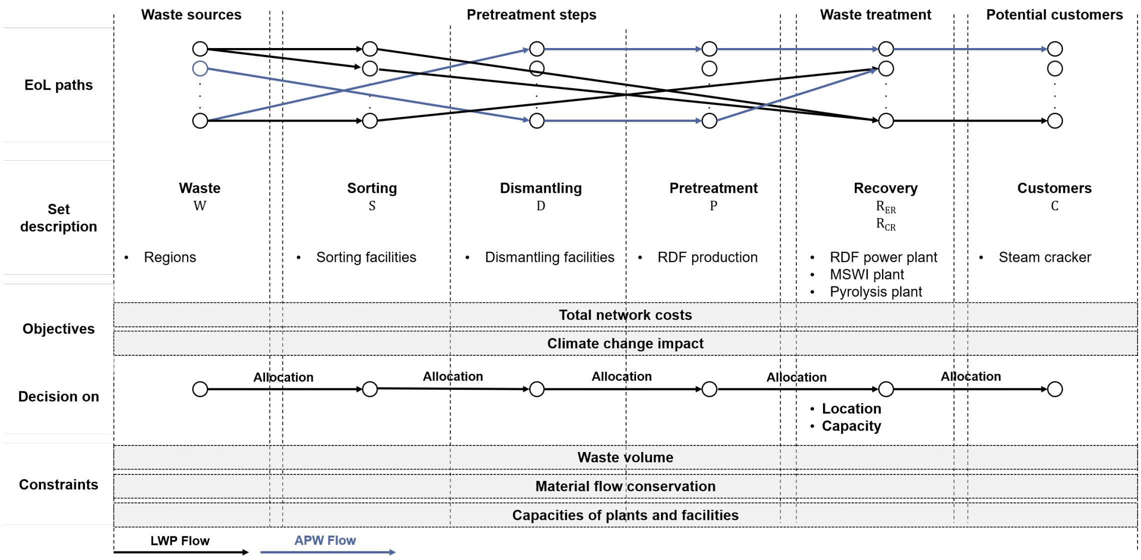


Figure 4.19: Overview of the mathematical optimization model of plastics EoL paths.

Results and Discussion

The cost-minimal plastic waste treatment network consists of the existing waste incineration infrastructure and using the plastic waste in energy recovery processes (cf. Figure 4.20 a and b). Even though Model 2 allows the placement of pyrolysis plants for chemical recycling, no plants are placed. Due to the high profits associated with waste incineration with energy recovery, the waste is exclusively incinerated and not chemically recycled. The waste is exclusively delivered to RDF incineration plants taking advantage of their higher efficiency and higher revenues. Here, longer transport distances are accepted to exploit the associated economic advantage.

Minimizing the climate change impact of Model 2, it is noticeable that all waste is chemically recycled due to the lower climate change impact associated with the pyrolysis process compared to waste incineration with energy recovery (cf. Figure 4.20 c and d). For this purpose, 58 pyrolysis plants are placed close to LWP sorting plants. The pyrolysis plants have an average utilization of 57%.

The total climate change impact is 39% of the impact of the baseline network (Model 1). So, by shifting the network design and allowing for chemical recycling, 61% of the total CO_2 emissions could be saved, demonstrating the environmental contribution that chemical recycling can make.

In order to consider the competing economic and environmental objectives, Model 3, a multi-objective decision model, is employed, determining a balanced solution for both objectives. In the resulting network (cf. Figure 4.21), 47% of the available waste feedstock is incinerated, and 53% is chemically recycled. Five pyrolysis plants with a capacity of 120,000 t input/year were opened in 2021 and are fully utilized. The total annual costs of the waste handling network are approximately double the costs of the cost-minimizing solution. At the same time, the CO_2 emissions are 32% lower than the emissions in the baseline model.

By integrating the climate change impact into the optimization of the waste management system, a balanced solution between economic and environmental objectives is found. Nevertheless, the waste management system is not designed top-down by governmental bodies but is cost and technology-based. For this reason, political steering strategies are discussed to assert the environmental advantages of chemical recycling in practice.

The assessed steering strategies include (1) an extension of the EU emission trading system (ETS) to the waste treatment sector and (2) increasing or implementing recycling rates for the assessed waste streams that are currently incinerated.

The extension of the EU ETS to waste treatment has a steering effect toward a lower environmental impact of the waste treatment system aligning economic and environmental objectives. The break-even price for the gross processing costs of waste incineration with energy recovery and pyrolysis regarding CO_2 certificates is 46 €/t CO_2. A higher certificate price leads to the economic competitiveness of pyrolysis plants for LWP sorting residues with 120,000 t input/year due to lower CO_2 emissions than waste incineration. Therefore, the average CO_2 certificate price in 2021 of 55 €/t and the historical certificate price of about 100 €/t (reached in 2023) lead to changes in the network design.

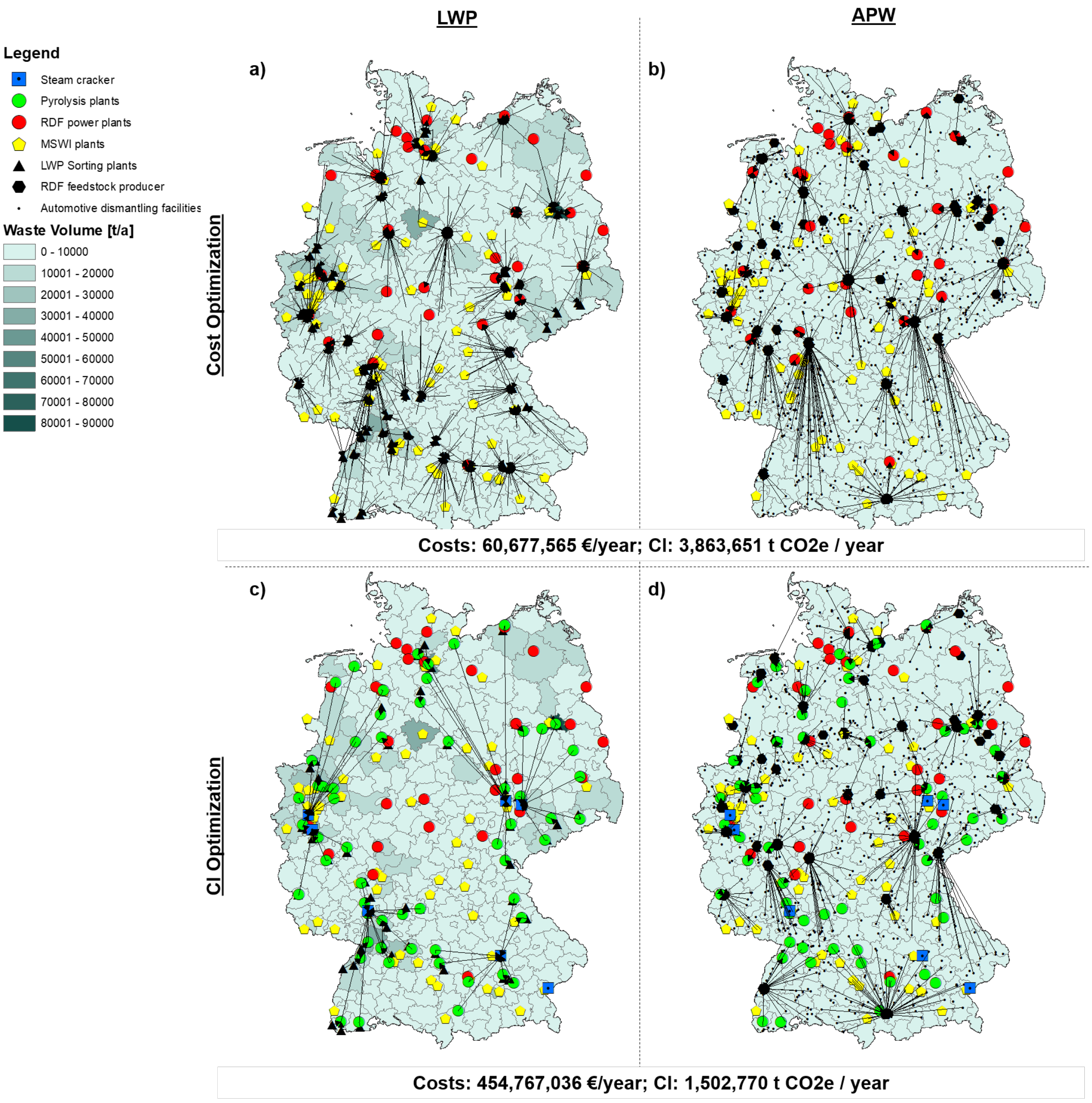


Figure 4.20: Map of the optimization results for the waste network design employing waste incineration with energy recovery and chemical recycling considering the different waste streams and objectives: a) LWP sorting residues for minimizing costs, b) automotive plastic waste for minimizing costs, c) LWP sorting residues for minimizing climate change impact, d) automotive plastic waste for minimizing climate change impact.

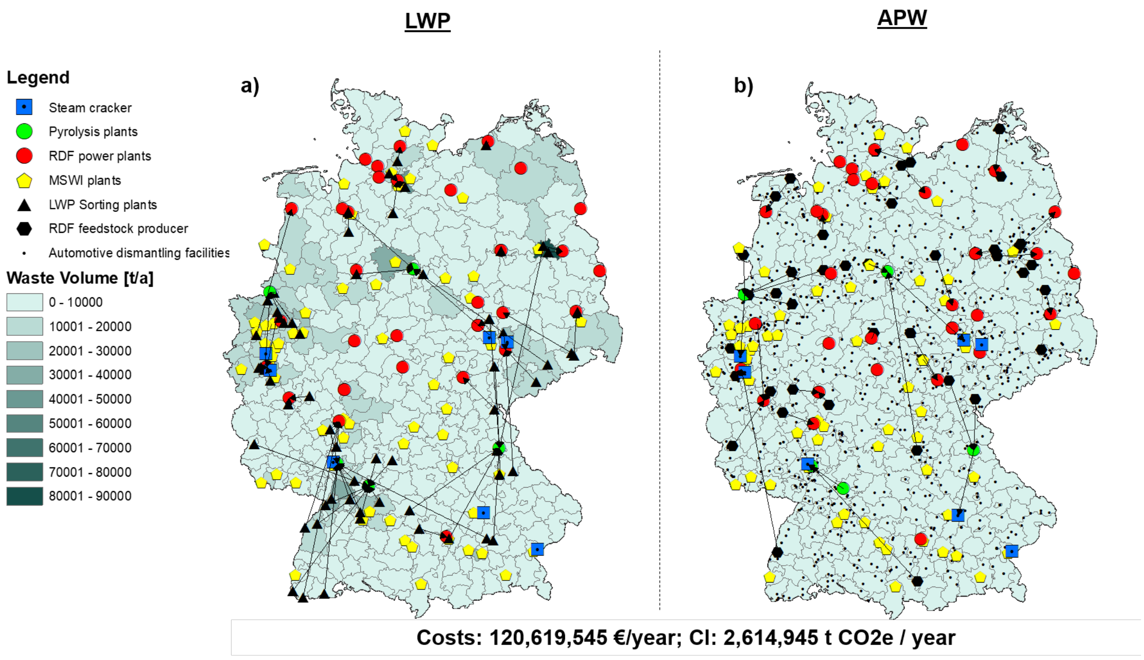


Figure 4.21: Map of the goal programming results of the waste network design employing waste incineration with energy recovery and chemical recycling for a) LWP sorting residues and b) automotive plastic waste.

The average certificate price in 2021 lead to the majority of the waste (96%) being chemically recycled due to the lower CO_2 emissions compared to waste incineration with energy recovery and, therefore, fewer CO_2 emission fees that must be paid. The total costs associated with the waste treatment network increased by 207%, while the network's CO_2 emissions decreased by 59% compared to the baseline network. The maximum historical CO_2 certificate price increases the network costs by 243% compared to the baseline network while all waste is chemically recycled. Here, the cost increase leads to a climate change impact reduction of 61% compared to the baseline, matching the climate change impact minimizing solution.

Specifying a recycling rate (APW) or its increase (LWP) ensures that the amount of plastics required to meet the set recycling rates is chemically recycled. With recycling rates of 55% for LWP matching EU recycling targets[6] and 35% for APW, the total waste treatment system costs increase 76% compared to baseline. Adjusting the recycling rates reduces the CO_2 emissions by 26% compared to the baseline.

The scenario analysis shows that the simple steering strategy of adjusted recycling rates generates a steering effect towards the environmentally advantageous EoL alternative. However, the steering strategy is based on the condition that there is legal certainty regarding the classification of chemical recycling as a recycling technology and crediting the treated waste to the recycling rates. This classification would support developing chemical recycling and a circular economy for plastics. However, in contrast to CO_2 emission fees, recycling rates are a regulatory measure that does not align the environmental and economic interests.

[6] Currently, 43% of LWP waste is mechanically recycled (UBA, 2022b).

There are some limitations to the study. The study is based on data that refers statically to the reference year 2021 and thus does not consider waste, market, or regulatory developments. These can influence the waste treatment network and the placement of pyrolysis plants. Parameters and assumptions also introduce uncertainties into the calculations, which should be addressed in scenarios and sensitivity analysis. Also, only a part of the waste treatment network is modeled, allowing only a relative comparison of waste incineration with energy recovery and chemical recycling of the assessed waste streams.

Despite these limitations, the study provides a model to assess the network effects of potential political steering strategies to support chemical recycling. The environmental impact and the impact of the circular economy for plastics integrating chemical recycling in the waste treatment network can be assessed.

5 Implications

After presenting the main contributions and findings of Studies A - E, this dissertation's implications and overall contributions are discussed below. The research objectives from Chapter 3 are addressed, and conclusions for the role of chemical recycling in a German circular economy for plastics are drawn.

5.1 Analysis of potential Feedstock for Chemical Recycling

This dissertation shows that wastes containing a large number of polyolefins (LWP) and wastes containing a large number of engineering plastics (APW) are suitable waste feedstock for pyrolysis. For the pyrolysis of APW, mass and energy balances were published transparently. The mass and energy balances expand the existing literature on plastic waste pyrolysis. The sorting residues from the mechanical recycling of LWP (Study A) and the APW (Study C) are feedstock for waste incineration with energy recovery and thus do not compete with mechanical recycling. The plastic circularity gap can be further closed by redirecting these waste streams to chemical recycling.

Figure 5.1 shows the waste generation of the assessed waste streams regionalized by German districts developed in Study E. In addition, it shows the current waste treatment infrastructure in which the waste is generated. Significantly more waste is generated from LWP than APW. In 2021, 2,681 kt of LWP waste was collected from German households. If only the sorting residues from mechanical recycling are considered a possible chemical recycling feedstock, this results in an input potential of 1,126 kt. In 2021, there were 1,011 t of APW in Germany. However, this is a conservative estimation based only on waste generated from dismantling components at the end of a vehicle's life. It does not include waste from repair jobs, additional waste streams with a similar waste composition, and an enhanced dismantling of EoL vehicles.

Based on the mass balances of the pyrolysis with the respective waste input (Zeller et al., 2021; Hennig et al., 2022; Stallkamp, Hennig, Volk, Richter, et al., 2023), the potential to substitute fossil carbon and produce secondary plastics through chemical recycling can be determined. The mass balances show that 337 kt of additional carbon can be recycled through chemical recycling and pyrolysis oil production. This corresponds to 451 kt of pyrolysis oil, which can be used as a naphtha substitute after further upgrading. Assuming that there are no significant material losses during pyrolysis oil processing, this covers 2% of the total naphtha demand of the chemical industry in Germany (21,500 kt/year; Geres et al. (2019)).

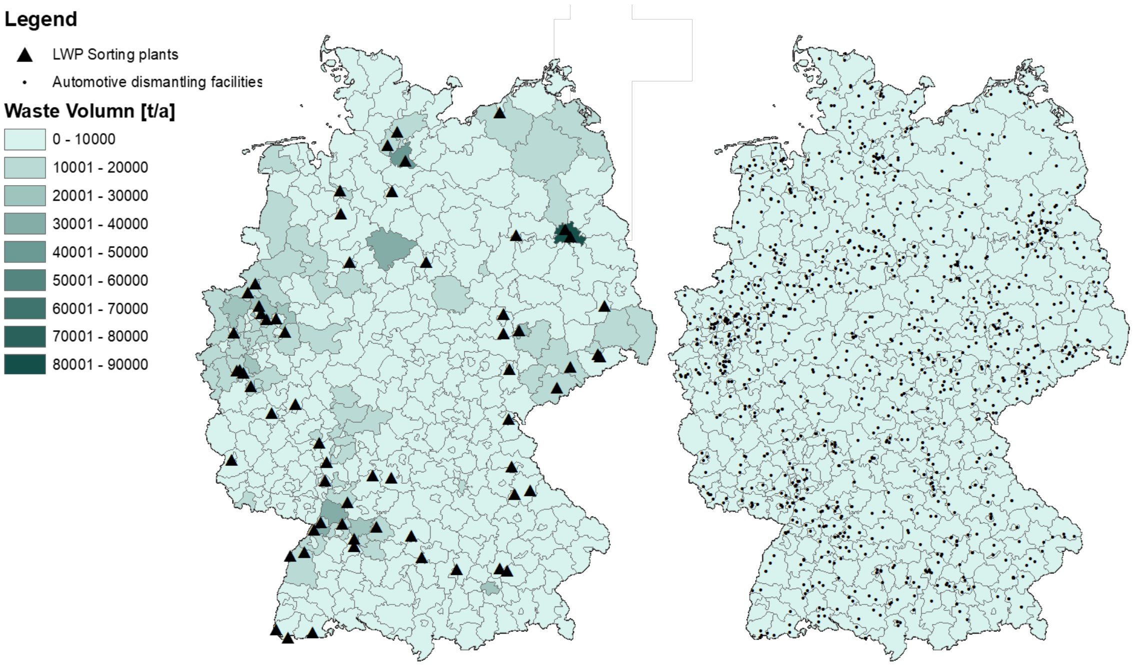


Figure 5.1: Waste potential in Germany in 2021 of LWP waste (left) and APW (right). The waste is accumulated at LWP sorting facilities (black triangles) or automotive dismantling facilities (black dots).

Assuming further processing of the high-value chemicals from pyrolysis oil to polyethylene[1], 3% of the German polyethylene production can be covered with the pyrolysis oil and the share of plastic products produced from recyclate could increase from 16% to 17%[2].

The estimation shows that the waste streams studied can already have a small impact on reducing the need for fossil raw materials by recycling them. Other potential waste streams suitable for chemical recycling include plastics from construction, electric and electronic equipment waste, and shredded EoL vehicles.

In addition to recycling carbon and defossilizing the plastic life cycle, shifting waste streams to chemical recycling can decarbonize waste treatment. Chemical recycling of the assessed waste streams is associated with a lower climate change impact than their incineration. For the waste streams in Germany, this adds up to CO_2 savings of 1,364 kt and thus a percentage reduction of 59% compared to incineration of the waste[3]. Here, the GHG emission reduction only refers to the savings potential for the waste incineration of the studied waste and thus has only a limited information value regarding the emission reduction in the entire waste management sector.

1 Polyethylene is the plastic with the highest production volume in Germany (Conversio, 2022).
2 Data: Conversio (2022); Zimmermann and Walzl (2009); Volk et al. (2021)
3 Data: Stallkamp, Hennig, Volk, Richter, et al. (2023); Volk et al. (2021); Zeller et al. (2021)

5.2 Economic and Environmental Assessment of Chemical Recycling

In order to establish chemical recycling as a waste management option, a classification of its environmental impacts and economic feasibility compared to mechanical recycling and waste incineration with energy recovery (i.e., the established process routes) is required. The conducted studies prove that chemical recycling is more environmentally advantageous than waste incineration with energy recovery. The chemical recycling of LWP sorting residues and APW is associated with a lower climate change impact than their incineration with energy recovery. For the recycling of LWP, the assessment shows the environmental and economic advantages of combining mechanical and chemical recycling. The defossilization of the plastic life cycle is also supported by chemical recycling since feedstock carbon can be kept within the economic cycle.

The conducted TEA demonstrates that the economic feasibility of chemical recycling depends on the capacity of the pyrolysis plant, the degree of its utilization, and the use cases for the by-products. With an input capacity of 33,750 t/year or higher and full capacity utilization, economical operation of the plants is possible in Germany. The main cost drivers are capital payments for the investment made and personnel costs. A comparison with waste incineration with energy recovery shows that the economic advantage of energy recovery depends on energy prices. High energy prices mean energy recovery is preferable over chemical recycling since the energy recovered (electricity and heat) generates corresponding revenues. The economic comparison between chemical and mechanical recycling shows that chemical recycling is economically competitive with comparable production costs along the recycling route. Here, high revenues for the secondary plastics produced and higher material yields compensate for higher production costs.

5.3 Discussion of the Quality of Products from different Recycling Processes

When comparing recycling routes for plastic waste, the quality of the secondary material produced must be included in the assessment. The literature proposes different approaches, but they all face difficulties in assessing secondary material quality. This dissertation provides an overview of the approaches for including the materials' quality in the assessment. Depending on the available data, different approaches can be chosen for assessing the material qualities:

- Technical properties of the material,
- economic evaluations in the form of prices,
- and a general qualitative discussion of the impact based on assumptions.

In the case of plastics, the quality of the secondary material produced directly impacts the defossilization of the plastic life cycle. With lower material quality, secondary plastics cannot replace primary plastic in all applications, and thus to some extent, primary plastics from fossil feedstocks are still needed. Study B shows this by the lower circularity potential of mechanical recycling compared to chemical recycling. The higher circularity potential of chemical recycling is due to the possibility of creating plastics from pyrolysis oil applicable for all use cases.

The quality of the secondary material also influences decarbonization since lower material quality results in a lower substitution of primary material. Thus, emission-intensive production steps still have to be performed. The quality of the secondary materials thus significantly influences the recycling option that is to be preferred environmentally and economically.

In addition, Study B shows that the recycling option most likely to contribute to a circular economy is not necessarily the most environmentally sustainable. This is mainly due to the quality of the secondary materials produced. In the specific case of Study B, the quality of the secondary plastic from mechanical recycling is not equal to that of virgin plastic. For this reason, it is essential to include the quality of the secondary plastic produced in every assessment and every comparison of recycling technologies.

5.4 Analysis of Plastic Waste Treatment Networks and Political Steering Strategies

The conflict between economic and environmental objectives when treating plastic waste is also evident when extending the research to the waste treatment network. In order to align objectives and support the chemical recycling of plastic waste, political decision-makers can draw on different steering strategies.

The developed model proves that by extending European or national CO_2 certificate trading to the waste management sector, the costs of CO_2 process emissions can be internalized, increasing the costs of waste incineration with energy recovery more than the costs of chemical recycling. Thus, the environmental advantages of chemical recycling are associated with economic advantages. However, the price level for the CO_2 certificates is crucial to achieving a steering effect. If the price is too low, only the system's total costs will increase, and no shift in waste streams from waste incineration with energy recovery to chemical recycling will occur. The break-even price for a steering effect is calculated to be 46 €/t CO_2.

Another political steering strategy derived from the model is to demand recycling rates for plastic waste sent to waste incineration. The rates must be technically achievable. However, the recycling rates ultimately lead to waste streams being shifted to the extent that they meet the demanded rates. This political steering strategy is a regulatory measure that does not align economic and environmental objectives.

The steering strategies can support decarbonizing plastic waste recycling and the defossilization of the plastic life cycle. Integrating chemical recycling into the waste management system can unlock the potential to close the plastics loop further and reduce dependence on fossil carbon sources.

6 Conclusions

The results of the studies and the implications for the cross-study research objectives of this dissertation are summarized in the following (cf. Section 6.1). In the outlook, the limitations of the dissertation are stated to outline future research approaches (cf. Section 6.2).

6.1 Summary

This dissertation contributes to the literature with five studies that examine the role of chemical plastic recycling in Germany and assist decision-makers in identifying appropriate steering strategies to close the plastics' material loop. Consequently, valuable insights are gained into decarbonizing and defossilizing the plastics sector of the chemical industry.

Study A assesses chemical recycling for the LWP waste stream and shows that combining the currently predominant mechanical recycling with chemical recycling has economic and environmental advantages over employing these technologies individually. At the same time, recycling instead of waste incineration allows more carbon to be recycled and reduces dependence on fossil resources. This study combines MFCA with a streamlined LCA for the assessment.

Study B shows the importance of integrating the quality of secondary materials in assessing recycling routes. Generally, there are three approaches to assessing secondary materials' quality: technical properties, economic evaluations as an approximation, and qualitative discussion. In the case study, chemical recycling has a higher circularity potential than mechanical recycling when considering the quality of secondary materials in a qualitative discussion.

Study C combines MFCA with a streamlined LCA assessing the chemical recycling of APW. Based on pyrolysis experiments, a trade-off between economic and environmentally preferable waste treatment options is identified when comparing chemical recycling with waste incineration with energy recovery. The study proves that chemical recycling is environmentally advantageous over waste incineration with energy recovery but economically disadvantageous. The assessment data is based on pyrolysis experiments explicitly conducted for this purpose and published transparently. This contributes to a more solid data basis compared to other studies.

Study D examines the economics of chemical recycling with a detailed TEA of APW pyrolysis. Economical operation of a pyrolysis plant in Germany is possible at full utilization, starting at 33,750 t input/year capacity. The main cost drivers are capital payments for the investment made and personnel costs. The economic operation of the plants also depends on the use of the by-products, in particular, whether they have to be disposed of with costs or whether they still represent a minor value stream for which revenue can be generated.

Study E combines the collected findings in a location optimization model for pyrolysis plants in Germany's current waste treatment network. Political steering strategies are identified to support political decision-makers in resolving the conflict between economic and environmentally favorable waste treatment options and to integrate chemical recycling into waste management. The analysis of these strategies shows that expanding CO_2 certificate trading to the waste management sector and recycling rates can contribute to the decarbonization and defossilization of the plastic life cycle.

Implications for four cross-study research objectives — feedstock potential, quality assessment, economic and environmental assessment, and steering strategies — were derived. Wastes containing primarily polyolefins but also engineering plastics can be technically pyrolyzed. However, the most significant waste quantities studied are generated in short-lived lightweight packaging. Environmentally, chemical recycling is preferable over waste incineration with energy recovery for all waste streams considered. For LWP, combining mechanical and chemical recycling results in the lowest environmental impact of the assessed EoL options. Economically, chemical recycling is not preferable over waste incineration with energy recovery due to the high revenues from energy recovery. The quality of the secondary materials must be considered when assessing waste recycling options, as these strongly influence economic and environmental assessment. The political decision to extend the CO_2 certificate trading to the waste treatment sector can align economic and environmental objectives. Introducing recycling rates for waste that is a feedstock for energy recovery is an additional useful policy in the regulatory framework to support chemical recycling. However, the recycling rate steering strategy requires legal certainty regarding the classification of chemical recycling as a recycling technology and the crediting of treated waste towards recycling rates.

6.2 Outlook

Future studies contributing to understanding the role of chemical recycling in a German circular economy for plastics could address the limitations of this dissertation. This dissertation limits its research to pyrolysis and does not examine other chemical recycling processes, such as gasification, depolymerization, or dissolution. Expanding the assessment of recycling processes would allow for a more comprehensive understanding of which processes are economically and environmentally preferable. The same applies to expanding the waste streams, like plastic waste from the construction sector or electronic waste.

Also, chemical recycling is not combined with other circular economy strategies. In particular, the potential of repair, refurbishment, or remanufacturing strategies has to be quantified and analyzed. In addition, the strategy of the circular economy should be considered along with other strategies for decarbonization and defossilization of plastics and chemical production, e.g., concerning goal conflicts. That is, combining the circular economy with the integration of renewable feedstock and CCS and CCU technology is a desirable subject of future research.

Future research can build upon the comprehensive analytical approach and data collected in this dissertation. Especially experiments on industrial-scale pyrolysis plants should be conducted to generate more comprehensive data on the pyrolysis process. Based on this, complete LCAs for the pyrolysis of various plastic waste in industrial-scale processes should be conducted to support the comparison of recycling technologies and extend it with additional environmental impact indicators. Due to the limited data availability, the assessments here are streamlined to the environmental indicators of climate change impact, cumulative energy demand, and carbon efficiency.

The limited data basis also leads to the fact that the political steering strategies for supporting the circular economy are only assessed concerning the costs and the climate change impact. Nevertheless, the mathematical optimization model can be expanded regarding additional environmental and social criteria. Also, the model could be extended by a more comprehensive data basis, uncertainties, and dynamics. The implementation of such approaches would enable deriving more robust implications. However, the analysis approach developed here provides a starting point for further research.

This dissertation substantially contributes to understanding the role of chemical recycling in a German circular economy for plastics. The knowledge gained supports the defossilization and decarbonization of the plastic life cycle.

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Part II

Companion Articles

Overview of Related Publications

Study A

Volk, R., Stallkamp, C., Steins, J. J., Yogish, S. P., Müller, R. C., Stapf, D., & Schultmann, F. (2021). Techno-economic assessment and comparison of different plastic recycling pathways: A German case study. Journal of Industrial Ecology, 25(5), 1318–1337. `https://doi.org/10.1111/jiec.13145`.

Study B

Stallkamp, C., Volk, R., & Schultmann, F. (2022). The impact of secondary materials' quality on assessing plastic recycling technologies. E3S Web of Conferences, 349, 05001. `https://doi.org/10.1051/e3sconf/202234905001`.

Study C

Stallkamp, C., Hennig, M., Volk, R., Richter, F., Bergfeldt, B., Tavakkol, S., Schultmann, Frank, F., & Stapf, D. (2023). Economic and environmental assessment of automotive plastic waste end-of-life options – energy recovery versus chemical recycling. Journal of Industrial Ecology. `https://doi.org/10.1111/jiec.13416`.

Study D

Stallkamp, C., Hennig, M., Volk, R., Stapf, D., & Schultmann, F. (2023). Techno-economic assessment of pyrolysis plants for automotive plastic waste. Submitted to a scientific journal.

Study E

Stallkamp, C., Volk, R., & Schultmann, F. (2023). Decision support for designing a recycling network closing the German plastic loop. Submitted to a scientific journal.

A Techno-Economic Assessment and Comparison of different Plastic Recycling Pathways: A German Case Study

Abstract[1]

Greenhouse gas (GHG) emissions need to be reduced to limit global warming. Plastic production requires carbon raw materials and energy that are associated today with predominantly fossil raw materials and fossil GHG emissions. Worldwide, the plastic demand is increasing annually by 4%. Recycling technologies can help saving or reducing GHG emissions, but they require comparative assessment. Thus, we assess mechanical recycling, chemical recycling by means of pyrolysis and a consecutive, complementary combination of both concerning Global Warming Potential (GWP) [CO2e], Cumulative Energy Demand (CED) [MJ/kg], carbon efficiency [%] and product costs [€] in a process-oriented approach and within defined system boundaries. The developed techno-economic and environmental assessment approach is demonstrated in a case study on recycling of separately collected mixed lightweight packaging (LWP) waste in Germany. In the recycling paths, the bulk materials polypropylene (PP), polyethylene (PE), polyvinylchloride (PVC) and polystyrene (PS) are assessed. The combined mechanical and chemical recycling (pyrolysis) of LWP waste shows considerable saving potentials in GWP (0.48 kg CO2e/kg input), CED (13.32 MJ/kg input), and cost (0.14 €/kg input) and a 16% higher carbon efficiency compared to the baseline scenario with state-of-the-art mechanical recycling in Germany. This leads to a combined recycling potential between 2.5 and 2.8 million metric tons/year that could keep between 0.8 and 2 million metric tons/year additionally in the (circular) economy instead of incinerating them. This would be sufficient to reach both EU and German recycling rate targets.

[1] This chapter includes the final version of the article "Techno-economic assessment and comparison of different plastic recycling pathways: A German case study" by Rebekka Volk, Justus Steins, Savina Yogish, Richard Müller, Dieter Stapf, Frank Schultmann, and myself. The article was published in the *Journal of Industrial Ecology* as (Volk et al., 2021). The supplementary material can be found on the journal website.

A.1 Introduction

Plastic production requires predominantly crude oil and energy while emitting GHG emissions that need to be reduced to combat climate change (IPCC, 2013). The German chemical sector, the largest in Europe, accounts for 6% of annual GHG emissions (UBA, 2018b; Destatis, 2018; Wyns et al., 2018; VCI, 2019). In Europe, 49 million tons[2] of plastics are produced annually for packaging (40%), construction (20%), automotive parts (9%) and electronics (6%). 25.8 million tons of plastic waste are generated annually (European Parliament and Council of the European Union, 1994). Plastic waste can be recycled mechanically or chemically, or processed to chemical or physical energy carriers. Currently, less than 30% is collected for recycling; a significant part is exported to non-EU countries with lower environmental standards (European Commission, 2018). In Germany, 46 wt.-% of the plastic waste (incl. production and processing wastes) is recycled mechanically; 1 wt.-% is recycled chemically (Conversio, 2018).

The EU demands a recycling rate for plastics packaging waste of 55% in 2030, while Germany demands 63% until 2022 (European Commission, 2018; European Parliament and Council of the European Union, 1994). But, mechanical recycling only can hardly fulfil the imposed recycling rates due to technical restrictions and economic limitations (Pivnenko et al., 2015; Ragaert et al., 2017).

Mechanical recycling processes plastic waste fractions without significant changes to their chemical composition, while chemical recycling processes it to intermediate chemicals by changing its chemical structure (Conversio, 2018; Stapf et al., 2019). The latter can be used as secondary or renewable feedstock (Meran et al., 2008; Sommerhuber et al., 2016; Gu et al., 2016a, 2016b) in plastics' or other chemicals' production as and can significantly reduce GHG emissions (Makuta et al., 2000; Dormer et al., 2013). Mechanical recycling cannot produce high-quality or virgin-quality plastics, but up to 20–50% cheaper plastics compared to virgin plastics (Gu et al., 2016a, 2016b, 2017). Thus, it is challenging to identify the environmentally or economically best recycling option, depending on locally available technologies, capacities, process efficiencies, specific waste compositions and conditions (Van Eygen et al., 2018b).

Life Cycle Assessment (LCA) is widespread to assess environmental impacts during a products' lifecycle (Rieckhof and Guenther, 2018; Rebitzer, 2002; Klöpffer and Renner, 2008). Multiple LCA and techno-economic analyses of olefin production from oil, coal, methane and ethane (Ren et al., 2006, 2009, 2008; Xiang, Yang, et al., 2014; Xiang, Qian, et al., 2014; Xiang et al., 2015; Amghizar et al., 2017; Z. Zhao et al., 2017) and plant-specific approaches (Patel, 2003; Pereira et al., 2013; Kanchanapiya et al., 2014) were conducted. LCA was also applied to mechanical recycling, incineration with energy recovery and landfilling of plastic waste to compare disposal alternatives (Lazarevic et al., 2010; Wäger et al., 2011; Al-Maaded et al., 2012; Turner et al., 2015; Wäger and Hischier, 2015; Gu et al., 2017; Van Eygen et al., 2018). Separately collected waste fractions (Perugini et al., 2005; D. S. Achilias et al., 2007; Turner et al., 2015; Van Eygen et al., 2018b), post-industrial plastic waste (Huysman et al., 2017) and post-consumer electronic waste

[2] Tons refer to metric tons throughout the article.

(D. Achilias et al., 2009; Wäger and Hischier, 2015; Wäger et al., 2011) were assessed. Mechanical plastics recycling and re-granulate performance was extensively researched (Chen et al., 2011; Turner et al., 2015; Gu et al., 2016a, 2016b, 2017; Van Eygen et al., 2018, 2018b). Few works considered chemical recycling and only recent works address mixed post-consumer packaging waste (Perugini et al., 2005; D. S. Achilias et al., 2007; D. Achilias et al., 2009; Lazarevic et al., 2010; BKV and Plastics Europe, 2019; Bergsma, 2019a, 2019b; Meys et al., 2020; Jeswani et al., 2021). However, there is not enough pyrolysis data for other use cases than mixed household waste (Vogel et al., 2020).

Some works assess national waste management systems or compliance to future regulations (Chen et al., 2011; Van Eygen et al., 2018, 2018b; Bergsma, 2019a, 2019b). Van Eygen et al. (2018, 2018b) assessed single-polymer and mixed-polymer recycling of Austrian plastic packaging waste. They highlight the importance of high-quality single-polymer recycling and lower environmental benefits of mixed-polymer recycling compared to the status quo. Chen et al. (2011) assessed (LCA) plastic waste recycling and energy-recovery technologies versus landfilling in China and find highest GHG reductions in low-grade plastics production from mechanical recycling and highest fossil fuel-savings in refuse-derived fuel (RDF) production and combustion. Huysman et al. (2017) developed a quality indicator to measure circular economy performance of post-industrial plastic waste.

Perugini et al. (2005) assessed and compared mechanical and chemical recycling (low-temperature fluidized bed pyrolysis and high-pressure hydrogenation) of plastic containers and highlight the good environmental performance of coupling feedstock and mechanical recycling. D. S. Achilias et al. (2007) assessed chemical recycling (dissolution/reprecipitation and catalytic pyrolysis on laboratory fixed bed reactors) of single-polymer model plastics, commercial plastics and plastic wastes and receive a polymer recovery of >90%. BKV and Plastics Europe (2019) analysed the technology readiness of chemical recycling processes (pyrolysis, gasification) of plastic waste focusing on data availability, necessary pretreatments and economic competitiveness. Bergsma (2019a, 2019b) assessed potential material inputs for chemical recycling from unrecycled Dutch waste (e.g. recycling losses, PET-trays, mixed plastics) and compared chemical (pyrolysis, hydropyrolysis, gasification) and mechanical recycling. Meys et al. (2020) developed a theoretical chemical recycling model to identify the best possible performance of five environmental impacts in 75 scenarios compared to existing recycling processes. Jeswani et al. (2021) assessed pyrolysis of mixed plastic waste (LCA) to understand favourable conditions and influencing factors.

Sensor-based sorting of plastic wastes is not new (Allen et al., 1999; Feldhoff et al., 1997; Murase and Sato, 1999; Scott, 1995; Wan et al., 1994). Recent publications address hyperspectral (Serranti et al., 2011; Habich and Beel, 2014) or black plastics sorting (Huang et al., 2017). Today, near-infrared sensors are widespread, but can neither separate HDPE and LDPE nor extract coloured/black plastics.

Literature gaps exist regarding the assessment of single-/dual-commingled waste plastics from collection systems (Turner et al., 2015), real-waste fractions with minerals, metals, and other contaminations and new recycling technologies (e.g. pyrolysis). Notably, there is a lack of high-quality Life Cycle Inventory (LCI) data on material reprocessing/recycling and more case-specific

LCA studies on recycled plastics are highly desirable (Turner et al., 2015; Gu et al., 2017). Most works investigate new sorting and recycling technologies under lab conditions only (D. S. Achilias et al., 2007). Assessments and primary data of pilot or industrial-scale chemical recycling plants (pyrolysis, gasification, solvolysis) are often missing and transparent LCI data is missing almost entirely (Gu et al., 2017), except for recent works (BKV and Plastics Europe, 2019; Meys et al., 2020). Also, a comparative study of sorting and recycling technologies for mixed plastic packaging waste is missing.

This study develops a method to assess primary plastics production (see Annex A3 of supporting information S2), post-consumer plastic packaging waste sorting (Section A.2.2.1) and recycling (mechanical, chemical and combined) (A.2.3) concerning costs, carbon efficiency, cumulative energy demand (CED) and global warming potential (GWP) (Section A.2.4) for polyethylene (PE), polypropylene (PP), polyvinylchloride (PVC) and general purpose polystyrene (GPPS). We assess packaging waste provided by the German packaging collection systems (see Section A.2.3 for its composition). This study differs from previous studies in three main aspects:

1. It provides a transparent and comparative techno-economic and environmental assessment of primary plastic production and different plastic recycling paths comprising GWP, CED, carbon efficiency, and costs. The underlying data is provided and includes details for a theoretical sensor-based sorting plant. Therefore, we address the lacking high-quality LCI data on bespoke processes.

2. It develops a techno-economic assessment of recycling paths on industrial scale based on literature data.

3. It considers a real waste composition from separately collected mixed-polymer lightweight packaging (LWP) waste instead of mono-fractions (Perugini et al., 2005), contaminated mono-fractions (Meys et al., 2020) or specified mixed plastic fractions (Jeswani et al., 2021). The assessment of real mixed plastic waste complex since multiple materials' treatments (of minerals, metals, fine fraction or organic material) have to be allocated.

Our approach enables producers and customers of plastic packaging to integrate GWP, CED, cost and carbon efficiency into multi-objective procurement and investment decision-making. This is particularly valuable when facing CO2e prices or tax, stricter regulation and volatile landfilling, incineration and co-combustion prices. This study also supports policymakers regarding the promotion and regulation of favorable recycling options for mixed plastics packaging waste.

A.2 Methodology

A.2.1 Goal and Scope

This study combined mass flow analysis (MFA) with LCA data of the considered recycling technologies (mechanical, chemical and combined recycling) of mixed LWP waste. Mass and energy balances for the assessment of sensor-based sorting, mechanical recycling, chemical (feedstock) and combined recycling of mixed LWP were established in a partial LCA (attributional approach). The model data is literature-based and process-oriented where data was not available, i.e. value chains were disaggregated into relevant unit processes that are assessed (i) based on simulation or (ii) measured data where physic-chemical models do not exist.

The assessment covers a mechanical pretreatment step to separate metals and minerals from refuse-derived fuel (RDF) as recycling feedstock, the state-of-the-art re-granulate production from sorted plastics for medium- and low-quality plastic products (like waste bins, formwork panels or park benches) and the chemical feedstock production via chemical recycling (here: pyrolysis). Other chemical recycling processes are not assessed (see Section A.4.2). The mass balances (incl. amounts and material types) and the impact assessment of CED, GWP, carbon efficiency and cost of each recycling path were calculated (Section A.3). Then, the different recycling paths were compared including compensation for substituting primary material and for energy co-generation through incineration of by-products.

The functional unit was 1 kg of mixed LWP waste that is collected separately and recycled in Germany. In the assessment, input waste was not associated with any GWP, CED, or cost.

Mechanical recycling includes mechanical pretreatment, sensor-based sorting and regranulation[3]. The main product is plastic re-granulate, often with a reduced quality compared to the original plastics (input) and with few possible recycling cycles (Ellen MacArthur Foundation, 2016). Main fields of application are road construction (100-150 kilotons/a), window and door profiles (100-150 kilotons/a), pipes (50-70 kilotons/a), landscaping, agriculture, electronics, packaging and plastic sheets (Conversio, 2018).

Chemical recycling processes mechanically pre-treated waste to chemical intermediates. In both recycling paths, the same mechanical pretreatment process was applied that is suitable to provide feedstock for maximum sensor-based plastics sorting yield and that is also standard for producing RDF from mixed waste (Stapf et al., 2019). In the combined recycling assessment, the recovered pure plastics from sensor-based sorting were mechanically regranulated, while the sorting residues were chemically recycled.

For all recycling paths, all economic and environmental burdens and rewards associated with the process steps, the by-products and their handling were assigned to the respective main products as the treatment of plastic waste is the focus of this study. The substitution of primary plastic material was rewarded in all paths by multiplying the amount of produced re-granulate or virgin plastic by the impact of the substituted primary plastic (Section A.2.4).

The system boundaries (Table S2-1 Annex A1 (S2)) exclude energy inputs for plant construction and machine production, the plastics use phase, the transportation[4], and cleaning/washing[5] processes. Likewise, transportation emissions are excluded, as they are expected to be relatively low (Chen

[3] Costs of all regranulation substeps (post-sorting, cleaning/washing, melting) are considered. Regarding CED and GWP, only melting is considered due to its dominance and missing data for the other substeps.

[4] Transportation between production gate and customer, post-consumer collection and transport to sorting, recycling, re-processing or incineration plants should be included in future research.

[5] Contamination usually does not impair the sorting result (Safavi et al., 2010)

et al., 2011). In the following subsections, technologies and recycling paths are assessed in detail. Different scenarios (Section A.2.5) and sensitivities (Section A.3.4) demonstrate the variability of the results.

A.2.2 Technnology Assessment

A.2.2.1 Mechanical Recycling and Sensor-Based Sorting Assessment

Prior to sensor-based sorting or pyrolysis, LWP waste is sorted in a mechanical pretreatment step using conventional technologies, including comminution, classification, sifting and metal separation and it is processed to RDF (Stapf et al., 2019). Then, the produced RDF is sorted in a sensor-based sorting plant to separate fractions with distinct qualities for further recycling and processing. Plastic types often cannot be separated by conventional sorting technologies, and mixed fractions cannot be recycled to high-quality products as small ratios of cross-contamination can lead to unusable batches (Masoumi et al., 2012; Ellen MacArthur Foundation, 2016). Since data on sensor-based sorting is not publicly available, we assessed different sorting technologies and modelled a theoretical sorting plant (Annex A2 (S2)).

Sensor-based sorting produces sorted plastics and sorting residues. Sorted plastics are the input for a regranulation process, while sorting residues are valuable fuel for different thermal recycling paths due to their high calorific value. As the modelled mechanical pretreatment is more refined than usual mechanical steps in mechanical-biological treatment plants, further treatments of sorting residues are omitted. Burdens associated with incineration of the sorting residues and rewards for substituting other fuels were included and allocated to the sensor-based sorted plastics. Within the scenario analysis, sorting residues were allocated to different thermal recycling paths (Section A.2.5).

The sorted plastic types are regranulated in an extruder. Additional sorting processes and associated mass reductions at the re-granulator were considered in the sorting plant yield. A separate assessment was not carried out.

A.2.2.2 Chemical Recycling

Chemical recycling processes mechanically pretreated waste to monomers or other chemicals (Conversio, 2018) that can be used as secondary feedstock in plastics' or other chemicals' production. In the assessed process, the produced RDF is fed into an integrated pyrolysis unit producing two useful co-products (pyrolysis liquid and pyrolysis gas), and a solid fraction (by-product). The pyrolysis liquid consists of oily and watery parts that are separated by condensation for further use. Gas and liquid fractions consist of hydrocarbons mainly; the solid fraction consists of minerals, char and hydrocarbons. Pyrolysis oil replaces naphtha as the feedstock of the hot part of the steam cracking process, where its components are thermally cracked under the presence of steam. In contrast, the pyrolysis gas enters only the cold downstream part of the steam cracking process for separation of the individual gaseous components. The solid fraction from the pyrolysis is combusted to provide its process heat and to condition it for deposit. The excess heat is sold for district heating purposes and is considered in all impact categories. In the steam cracking process, monomers are produced that are subsequently polymerized. The produced plastics have virgin quality and are rewarded with primary plastics' substitution.

A.2.3 Inventory Data of Technologies and other Data

The assessment of the mechanical, chemical and combined recycling paths was demonstrated for LWP waste in a case study for Germany. The separately collected German LWP sums up to ca. 2.5 million tons/year. Its average composition is shown in Figure S2-5 (Annex A4 (S2)). The environmental impact factors (Table S2-5 Annex A4 (S2, S3), specific to Germany) were used to calculate the GWP [CO2e/kg] and CED [MJ/kg] per process step based on its energy demand. Together with energy efficiencies of the combustion systems, they were also used to calculate associated rewards and burdens. Uncertainties and their impacts on the results are considered in a sensitivity analysis (Annex A7 (S2)). For the calculation of the substitution effects of primary material, LCAs based on the "cradle-to-gate" approach for primary plastic production in Europe were used (PlasticsEurope, 2018a) (Annex A3 (S2)).

A.2.3.1 Mechanical Pretreatment

LWP waste is sorted with conventional technologies and can be separated into metals (3% with 1.2% ferrous and 1.8% non-ferrous metals), heavy components (10%), a low-calorific fine components (20%) and water (2%) (Stapf et al., 2019). The variable sorting costs include the gate fee of incoming waste, compensation for metal recycling, landfilling of heavy components and the gate fees for energetic utilization (Annex A4 (S2)). The fixed costs for a waste pretreatment plant with a capacity of 100,000 tons/year and operating 8,000 h/year added up to 2.07 million €/year.

The environmental impact was derived from the electrical energy demand of 0.40 MJ/kg treated waste. This was measured at an exemplary treatment process and is higher than literature values (Bilitewski et al., 2018) due to higher processing demands for the RDF production (see Annex A9 (S2)). The energy demand of the mechanical pretreatment had a net[6] impacts of 1.84 MJ/kg (CED) and 0.32 kg CO2/kg (GWP). We reward electricity and heat production from incineration of the low-calorific fine components based on their calorific value, the emission-factor for household waste and the efficiency of waste incineration plants. Ferrous metals replace primary metal in cast iron production; non-ferrous metals substitute primary raw aluminium (as a non-ferrous representative).

A.2.3.2 Mechanical Recycling and Sensor-Based Sorting

A theoretical sorting plant (Figure A.1) was assessed to quantify its impact on the mechanical recycling path regarding costs, CED, and GWP. The combination of sorting technologies results in

[6] Net environmental impacts, carbon efficiencies and costs include rewards; their gross values only include burdens.

operational costs of 31.84 €/(ton input), in 0.09 MJ/(kg input) (CED), and in 0.006 kg CO2e/(kg input) (GWP) (Annexes A2, A4 (S2)).

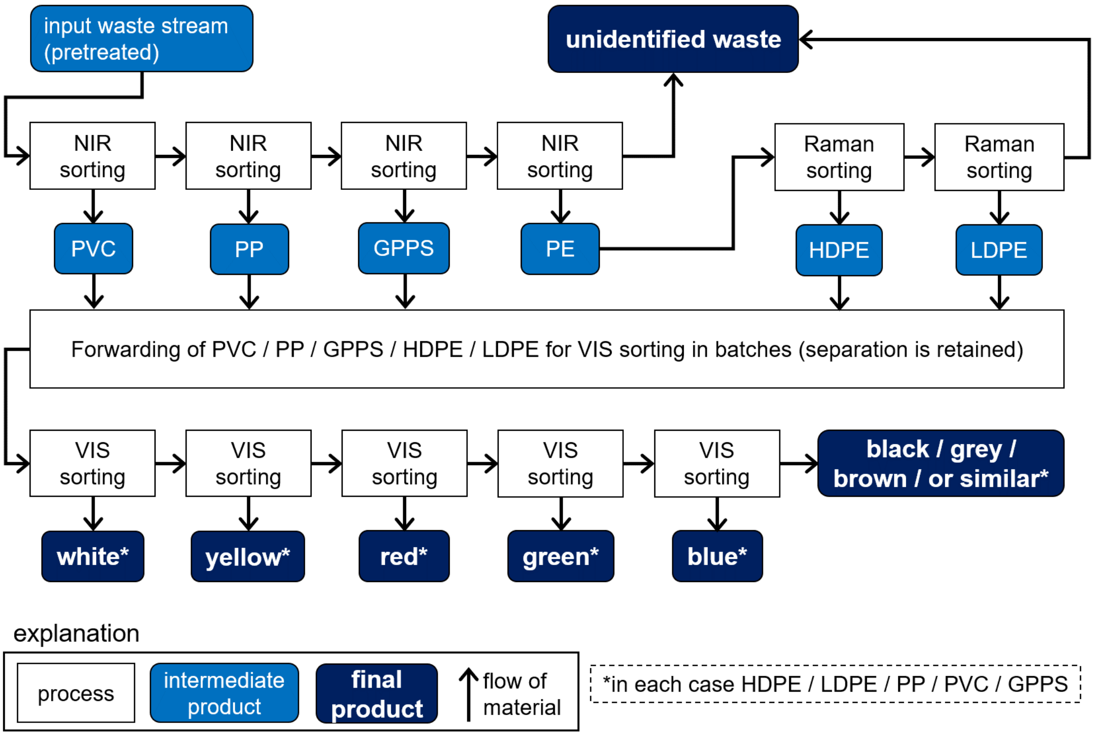


Figure A.1: Material flow in a theoretical integrated sensor-based sorting plant, designed for maximum colour-wise material recovery of plastic fractions PE, PP, PVC, and GPPS, respectively. Sorting residues are unidentified waste and non-sorted plastic colours. The input waste stream is conventionally sorted and mechanically pretreated.

In mechanical recycling, sensor-based sorting residues are incinerated[7]. Thus, incineration gate fees and emissions, as well as compensation for produced heat and electricity were included. In the baseline scenario, all sorting residues were incinerated in RDF power plants and municipal solid waste incineration (MSWI) plants (Section A.3.1). Other thermal recycling paths were calculated and discussed in further scenarios (Sections A.2.5, A.3.2). The efficiency rates and environmental impact factors used for the impact assessment are displayed in Table S2-5 (Annex A4 (S2) and available as data tables (S3).

In the regranulation, a carbon efficiency of 98% was assumed due to additional sorting steps and material losses (Dehoust et al., 2016). For different carbon efficiencies see Section A.2.5. As regranulation costs are not available in literature, the cumulated anterior processing costs were subtracted from the available re-granulates' market prices resulting in 0.4 €/kg[8]. Additionally,

[7] In chemical recycling, the sensor-based sorting residues are chemically recycled.

[8] We compare bale and re-granulate prices to assess regranulation costs, since the regranulation is the primary process executed between these qualities. Price differences are 0.33 €/kg (PE-HD), 0.36 €/kg (PE-LD), 0.54 €/kg (PP), and 0.24 €/kg (PET) (Plasticker, 2019) leading to 0.4 €/kg on average.

we assumed an energy consumption of the regranulation of 0.21 kWh per kg re-granulate that is ranging between 0.18 to 0.24 kWh/kg specified by Großmann (2011).

A.2.3.3 Chemical Recycling

Adequate experimental data of reasonable quality for scalable mixed plastics pyrolysis is only provided by Andreas et al. (1981). They pyrolyzed a waste composition similar to LWP sorting residues in a rotary kiln reactor at 650°C and yielded gas (38 wt.-%), liquids (30 wt.-%) and solids (32 wt.-%). Applying this data, we integrate the pyrolysis process into the process route given in Figure A.2. The underlying data of Figure A.2 and all following figures can be found in the supporting information S1. RDF from mechanical pretreatment is fed into the integrated pyrolysis unit and its impacts are allocated to the desired pyrolysis products (gas, oil) by mass. Pyrolysis oil replaces naphtha as steam cracking feedstock, while the pyrolysis gas components are separated in the cold part of the steam cracking process. The integrated pyrolysis process consists of the pyrolysis reactor, consecutive oil and gas upgrading steps, the solids' incineration and heat recovery system, solids' transport and mixing, and necessary auxiliary units. It is designed to separate all additives, pollutants, and impurities from pyrolysis oil and gas to meet the feedstock specifications of downstream chemical processing of virgin quality material. Process simulation determines both GWP and CED of the pyrolysis process of German LWP waste with 0.993 kg CO2/kg (GWP) and 14.99 MJ/kg (CED).

The solid fraction of the pyrolysis unit is combusted; the minerals are deposited. Excess heat is sold for district heating and considered in all assessment categories; it was credited with 0.155 kg CO2e/kg (GWP) and with 2.63 MJ/kg (CED).

Following BKV and Plastics Europe (2019), the integrated process scheme was developed for an 8.1 ton/h pyrolysis process operating 8,000 h/year. The scale-up is derived from the mass and energy balances from Andreas et al. (1981) and operational and capital investment cost were calculated. The cost assessment is based on the investment of a rotary kiln waste treatment plant of similar complexity, which was scaled down to the considered pyrolysis plant size according to (BKV and Plastics Europe, 2019). Fixed costs added up to 14.2 million €/year for this plant size which fits to the mechanical pretreatment process specified in Section A.2.3.1. Variable costs are 8.21 €/ton of the main product and include electricity of the combustion air-compressor (4 bar at 5 €/1,000 m3), landfill fees for combustion residues (100 €/ton), and the compensations generated from district heating (0.03 €/kWh). Our calculations led to total pyrolysis costs of around 320 €/ton of mixed plastic waste. For steam cracking, existing data on primary ethylene and propylene production is used (PlasticsEurope, 2012). Therefore, no further detailed steam cracking assessment is made; however, the impact of the inputs is altered.

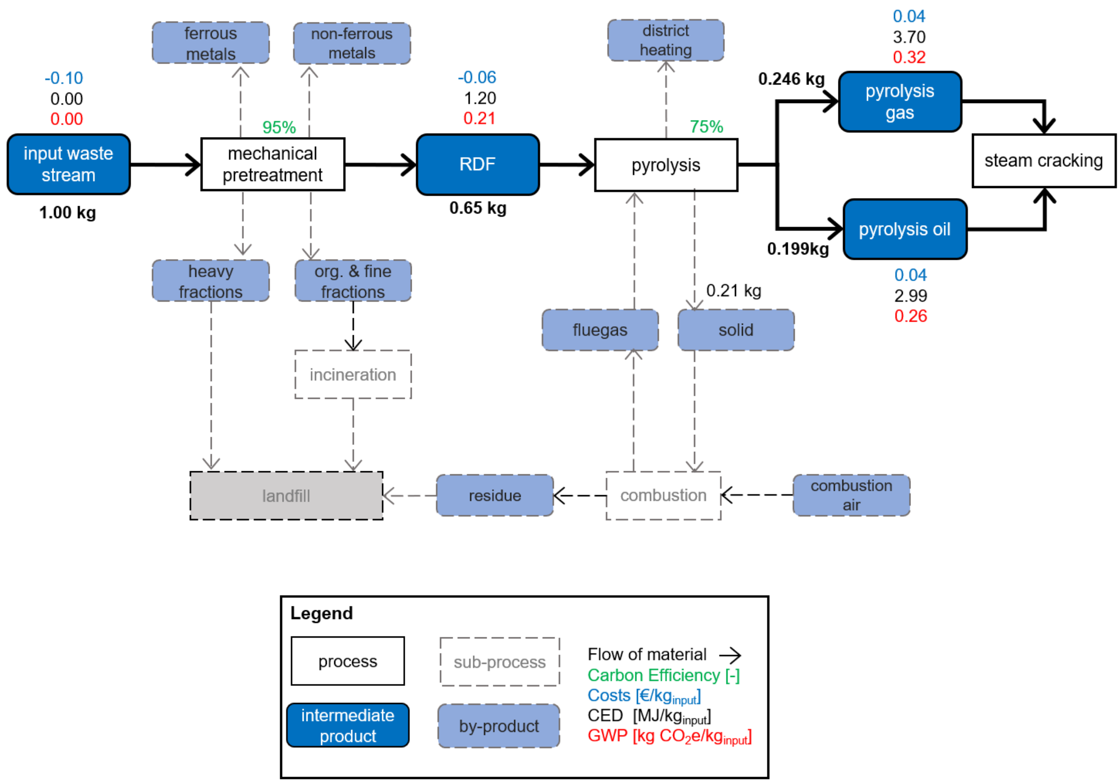


Figure A.2: Detailed pyrolysis route of mixed plastics per 1 kg of input waste. Refuse-derived fuel (RDF) is the intermediate product of mechanical pretreatment. Underlying data used to create this figure can be found in supporting information S1.

A.2.4 Impact Assessment

GWP[9], CED[10], cost and carbon efficiency are assessed. The carbon efficiency is the recycling rate in terms of the ratio of total carbon mass of the desired products divided by the total carbon mass of the feed(s) per conversion or separation step or per recycling path. Finally, the recycling paths are compared concerning their product costs. Each process step was assessed individually; the recycling paths sum up all process steps along the path. Also, above mentioned rewards and burdens were considered. Downstream process steps include the impact of upstream processes of the value chain. Equations (A.1) to (A.3) show the exemplary calculation of CED, GWP, and cost of mechanical pretreatment to produce RDF:

[9] Here, GWP100 is assessed as defined in the Kyoto Protocol (IPCC, 2013)

[10] CED is the "total quantity of primary energy which is necessary to produce, use and dispose of a product" (VDI, 2012)

$$\begin{aligned}
CED_{RDF} = {} & CED_{inputwaste} \\
& + CED_{mechanicalpretreatment} \\
& + CED_{landfillingofheavycontent} \\
& + CED_{incinerationoffinefraction} \\
& - CED_{compensationforelectricitygenerationfromincineration} \\
& - CED_{compensationforheatgenerationfromincineration} \\
& - CED_{compensationforrecyclingofferrousmetals} \\
& - CED_{compensationforrecyclingofnon-ferrousmetals}
\end{aligned} \tag{A.1}$$

$$\begin{aligned}
GWP_{RDF} = {} & GWP_{inputwaste} \\
& + GWP_{mechanicalpretreatment} \\
& + GWP_{landfillingofheavycontent} \\
& + GWP_{incinerationoffinefraction} \\
& - GWP_{compensationforelectricitygenerationfromincineration} \\
& - GWP_{compensationforheatgenerationfromincineration} \\
& - GWP_{compensationforrecyclingofferrousmetals} \\
& - GWP_{compensationforrecyclingofnon-ferrousmetals}
\end{aligned} \tag{A.2}$$

$$\begin{aligned}
costs_{RDF} = {} & costs_{inputwaste} \\
& + costs_{mechanicalpretreatment} \\
& + costs_{landfillingofheavycontent} \\
& + costs_{incinerationoffinefraction} \\
& - costs_{compensationforelectricitygenerationfromincineration} \\
& - costs_{compensationforheatgenerationfromincineration} \\
& - costs_{compensationforrecyclingofferrousmetals} \\
& - costs_{compensationforrecyclingofnon-ferrousmetals}
\end{aligned} \tag{A.3}$$

The carbon efficiency is based on stoichiometric mass balances. Only in the mechanical pretreatment, we assume 5% carbon in the fine fraction so that it has to be combusted (Stapf et al., 2019).

A.2.5 Scenario Definition

Multiple scenarios were developed on varying sensor-based sorting yields and different incineration paths for sorting residues to analyse the influence of underlying data and assumptions (Table A.1). Also, a combination of mechanical and chemical recycling is considered. Further assumptions and parameters are varied in a sensitivity analysis (Section A.3.4, Annex A7 (S2)).

Christiani and Beckamp (2020) state that 32% of the LWP provided by the German collection systems are high-grade recyclable plastics. After conventional sorting, 35% of the mass is discarded and the relative amount of high-grade recyclable plastics rises to 49%. A buffer of 6% for non-identifiable plastics leads to a resulting sensor-based sorting yield of 43% for mechanical recycling. This buffer results from the share of other polyolefins in the high-quality recyclable plastic composition (Figure S2-5 Annex A4 (S2)). Furthermore, additional material losses between 2% and 50% for sorting steps at the regranulation plant are assigned to the sorting plant (Dehoust et al., 2016). The best and worst-case scenarios for mechanical recycling the sensor-based sorting yield is multiplied by the extremes of additional regranulation losses. Thus, we distinguished a sorting yield of 42% (scenarios1.1) and 22% (scenarios1.2). Due to missing data, we assume sorting yields only per polymer type that is then further processed. So, after the sensor-based sorting, we assess a mono stream of one specific polymer type that is then processed in a regranulation facility. In this step, we do not consider a mix of polymers.

Sorting residues are incinerated or co-combusted in cement kilns, coal-fired or RDF combustion plants or MSWI plants (in decreasing efficiency order) depending on its calorific value and chlorine content.

Scenario 1.1.1 reflects the worst case where sorting residues are incinerated in MSWI plants. Scenario 1.2.1 differs from it in a lower sorting yield and thus a higher sorting residue incineration. Scenario 1.1.2 is the baseline scenario with co-combustion of sorting residues in efficient RDF combustion plants (75%) and MSWI plants (25%). This reflects the current practice in Germany and other European countries (Ketelsen and Kanningen, 2016; Van Eygen et al., 2018; Bilitewski et al., 2018; Jeswani et al., 2021). Scenario 1.2.2 has a lower sorting yield than the baseline scenario. Scenarios1.1.3 and 1.2.3 include all four thermal recycling paths following current German combustion shares for RDF (Ketelsen and Kanningen, 2016) assuming that the sorting residues undergo further treatment (e.g. sensor-based sorting steps) to separate PVC and meet the strict chlorine limits for co-combustion in coal and cement power plants (5-15 g/kg in coal-powered plants and <10g/kg in cement kilns) (UBA, 2015).

Scenario 2 examines the chemical recycling (Section A.2.3.3). As a variation of the process parameters would result in unknown pyrolysis gas, oil and solid yields and compositions, sub-scenarios can neither be defined nor analysed. Scenarios3.1 and 3.2 combine mechanical and chemical recycling and examine high and low sensor-based sorting yields. Here, sorting residues are fed into the chemical recycling process[11].

[11] Pyrolysis inputs differ in chemical recycling (scenario 2) (RDF) and combined recycling (scenarios 3.1 and 3.2) (sorting residues). Therefore, the pyrolysis products could be different.

Table A.1: Overview of scenarios (-: parameter does not apply).

Scenario No.	Description	Sorting yield	Incineration paths of sorting residues
1.1.1	MR	42%	100% MSWI plant
1.1.2	MR	42%	25% MSWI plant, 75% RDF power plant
1.1.3	MR	42%	18% MSWI plant, 58% RDF power plant, 13% cement plant, 11% coal-powered plant
1.2.1	MR	22%	100% MSWI plant
1.2.2	MR	22%	25% MSWI plant, 75% RDF power plant
1.2.3	MR	22%	18% MSWI plant, 58% RDF power plant, 13% cement plant, 11% coal-powered plant
2	CR	-	-
3.1	CO	42%	-
3.2	CO	22%	-

A.3 Results

The impacts of the recycling paths were calculated based on the collected data and the defined scenarios in a case study for Germany. The results are illustrated for HDPE, but are also available for LDPE, PP, PVC, and GPPS (Annex A6 (S2, S3)). The impacts are described per kilogram of waste input (Figure S2-6, Annex A5 (S2)).

A.3.1 Baseline Scenario

The mechanical recycling results in gross sorting and regranulation costs of 0.10 €/kg and induces gross values of 0.67 kg CO2e/kg (GWP) and 3.83 MJ/kg (CED). The net values take full (100%) substitution of primary material into account (Figure S2-1 to Figure S2-4, Annex A3 (S2)) and result in costs of -0.16 €/kg (=revenues), 0.18 CO2e/kg (GWP) and -18.14 MJ/kg (CED). The differences between net and gross values result from avoided costs and avoided energy for primary material production and highlight the calculatory impact of primary material substitution on costs, GWP and CED. However, in this scenario, the substitution cannot compensate for the high GWP burdens that result mainly from incineration of sorting residues.

In the whole recycling path, incineration accounts for 95% of the GWP and 93% of the CED impact. Incineration of the sensor-based sorting residues accounts for around 60% of the CED and GWP. The regranulation process and its electrical energy demand has a high impact on CED and costs.

The results presume that the sensor-based sorting process' outcome is a single plastic fraction that is processed further (Section A.2.5). We do not break down the sensor-based sorted plastic by type due to missing data regarding the plastic composition after the sorting.

A.3.2 Other Scenarios

The other scenarios of mechanical recycling consider different thermal recycling paths for sorting residues and higher material losses. The results (Figure A.3 and Figure A.4) show that including additional industrial co-combustion (scenario 1.1.3) leads to net reductions by 95% (GWP) and 30% (CED) compared to the baseline (scenario 1.1.2). In the worst-case (scenario 1.1.1), the net GWP increases by 15% and the net CED increases by 7%. This is also valid for the lower yield scenarios. It results from the higher efficiency of industrial co-combustion compared to MSWI and highlights the impact of the considered thermal recycling. Scenarios with efficient cement kilns and coal-powered plants particularly show better environmental performance than the baseline scenario.

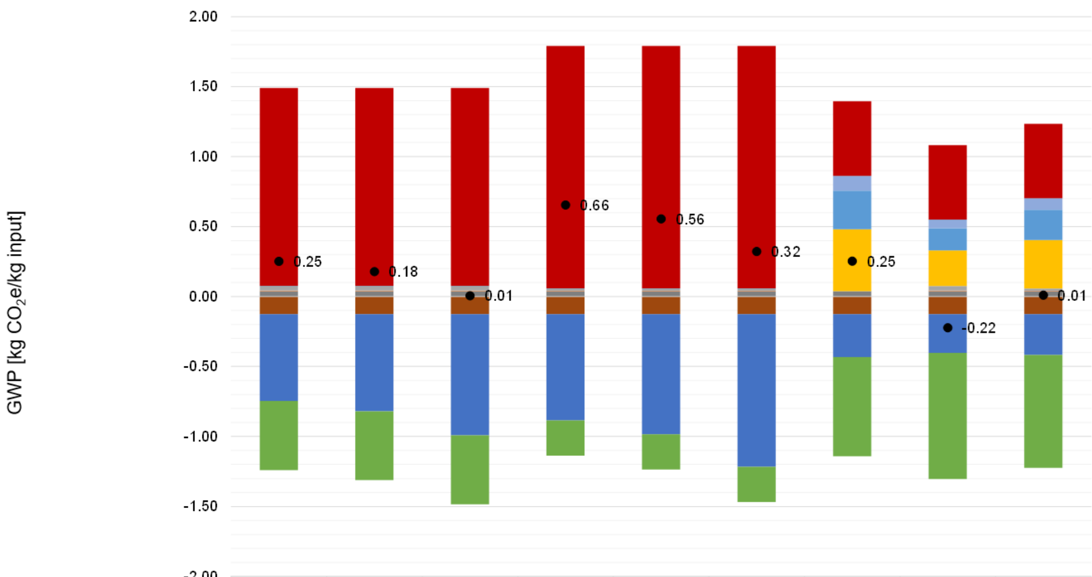
Higher material losses lead to higher GWP and CED impacts regardless of the thermal recycling mix. Additional sorting residues are incinerated, and less primary material is substituted and rewarded. Regarding costs, there are no differences between the incineration mixes due to the same assumed gate fees. However, additional material losses lead to rising costs of mechanical recycling due to additional gate fees for incineration. Also, the amount of produced re-granulate decreases and reduces the substitution effect.

Scenario 2 (chemical recycling; Figure S2-7, Annex A5 (S2)) results in -0.24 €/kg net sorting and reprocessing costs (=revenues) and induces -15.92 MJ/kg (CED) and 0.25 kg CO2e/kg (GWP). Steam cracking has a significant impact on all impact categories, mainly because of its high energy demand. Gross values of scenario 2 are 0.33 €/kg sorting and recycling costs, 15.66 MJ/kg (CED) and 0.96 kg CO2e/kg (GWP). The high yield of recycled material leads to a high reward for primary material substitution. The combined approach (scenario 3.1; Figure S2-8, Annex A5 (S2)) results in net values of -0,29 €/kg sorting and reprocessing costs (=revenues), -30.14 MJ/kg (CED) and -0.22 kg CO2e/kg (GWP) in a combined assessment of virgin plastics and re-granulate output. With higher material losses at the regranulation plant (scenario 3.2), GWP and CED impacts increase because more sorting residues are chemically recycled. Costs also increase due to a lower total yield. In both scenarios, a high yield leads to high rewards for primary plastic substitution.

A.3.3 Comparison of Recycling Processes and Scenarios

The assessed recycling paths are compared for HDPE, LDPE, PP, PVC, and GPPS (Figure A.6 and Annex A6 (S2, S3)). The study results of HDPE discussed in the following are representative for the other assessed plastic types (except PVC). The results for PVC are different, although the trend is consistent.

GWP impacts (Figure A.3) of mechanical recycling are influenced by the incineration paths and their efficiencies (Section A.3.2). More efficient industrial co-combustion reduces the impacts due to higher substitution rewards for generated heat and electricity. GWP is also influenced by material yield, as higher material losses lead to additional sorting residues that are incinerated, and less primary material is substituted.



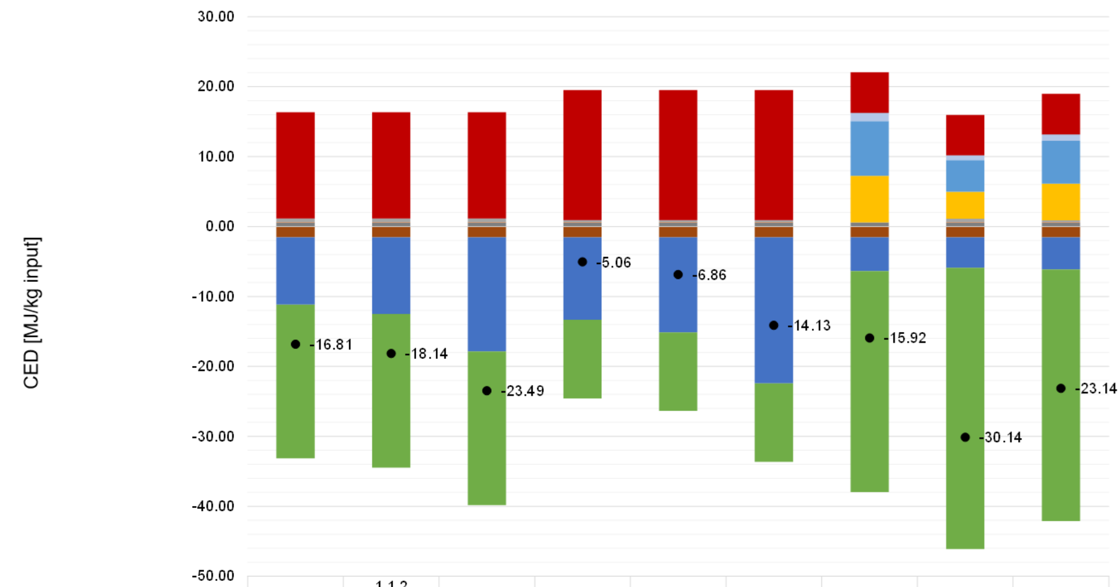
	1.1.1	1.1.2 Baseline	1.1.3	1.2.1	1.2.2	1.2.3	2	3.1	3.2
Substitution Primary Material	-0.49	-0.49	-0.49	-0.25	-0.25	-0.25	-0.71	-0.90	-0.81
Rewards Incineration	-0.62	-0.69	-0.87	-0.76	-0.86	-1.09	-0.31	-0.28	-0.29
Burdens Incineration	1.41	1.41	1.41	1.73	1.73	1.73	0.53	0.53	0.53
Rewards Metall Recycling	-0.13	-0.13	-0.13	-0.13	-0.13	-0.13	-0.13	-0.13	-0.13
Landfilling Heavy Components	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Polymer Synthesis	0.00	0.00	0.00	0.00	0.00	0.00	0.11	0.06	0.08
Steam Cracking	0.00	0.00	0.00	0.00	0.00	0.00	0.28	0.16	0.22
Pyrolysis	0.00	0.00	0.00	0.00	0.00	0.00	0.44	0.26	0.35
Regranulation	0.03	0.03	0.03	0.02	0.02	0.02	0.00	0.03	0.02
Sensor-Based Sorting	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mechanical Pretreatment	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04
Sum	0.25	0.18	0.01	0.66	0.56	0.32	0.25	-0.22	0.01

Scenario

Figure A.3: Comparison of considered recycling processes and scenarios regarding their net GWP impact [kg CO2e/kg input]. Assessment for 1 kg of input waste. In scenarios2, 3.1 and 3.2, „Burdens Incineration" results from the incineration of the fine fraction of the mechanical pretreatment. Underlying data used to create this figure can be found in the supporting information S1.

Chemical recycling performs slightly worse compared to the baseline scenario for most fractions (except for GPPS, where it is significantly worse). Reduced incineration burdens compensate the GWP impact of the steam cracking and polymer synthesis as no sorting residues are incinerated. The non-existent incineration rewards from the combustion of sorting residues counterbalance higher rewards for substituting primary material. Even though chemical recycling performs slightly worse than mechanical recycling regarding GWP, differences in impurities, additives and quality aspects of the final products are not included. Including these has a substantial effect on the recycling paths' performance (Section A.3.4)). In the combined approaches, lower material losses at the regranulation plants lead to considerably lower GWP and net GWP credits resulting from omitted incineration burdens from sorting residues and higher total yields due to additional virgin plastic produced by chemical recycling.

All assessed scenarios show CED savings (Figure A.4). Within mechanical recycling, CED savings increase with a decrease in material losses due to decreasing incineration burdens and increasing primary material substitution rewards. CED savings of mechanical and chemical recycling are comparable. Although chemical recycling has a higher energy demand than mechanical recycling,



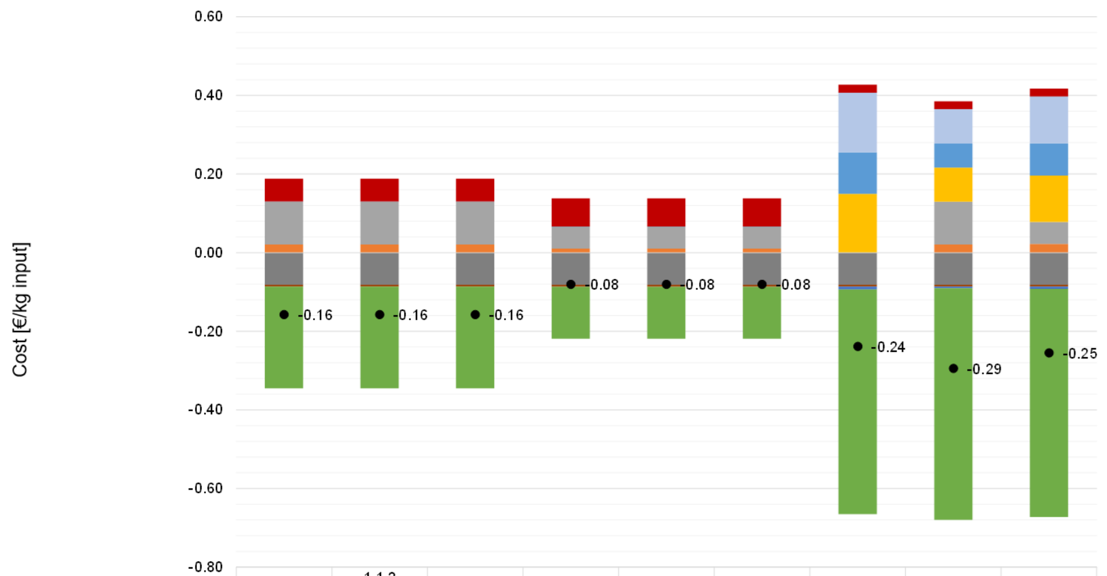
	1.1.1	1.1.2 Baseline	1.1.3	1.2.1	1.2.2	1.2.3	2	3.1	3.2
Substitution Primary Material	-21.96	-21.96	-21.96	-11.22	-11.22	-11.22	-31.58	-40.22	-35.98
Rewards Incineration	-9.66	-10.98	-16.33	-11.80	-13.60	-20.87	-4.86	-4.36	-4.60
Burdens Incineration	15.21	15.21	15.21	18.59	18.59	18.59	5.80	5.80	5.80
Rewards Metall Recycling	-1.53	-1.53	-1.53	-1.53	-1.53	-1.53	-1.53	-1.53	-1.53
Landfilling Heavy Components	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Polymer Synthesis	0.00	0.00	0.00	0.00	0.00	0.00	1.14	0.66	0.90
Steam Cracking	0.00	0.00	0.00	0.00	0.00	0.00	7.82	4.52	6.14
Pyrolysis	0.00	0.00	0.00	0.00	0.00	0.00	6.67	3.86	5.23
Regranulation	0.46	0.46	0.46	0.24	0.24	0.24	0.00	0.46	0.24
Sensor-Based Sorting	0.06	0.06	0.06	0.06	0.06	0.06	0.00	0.06	0.06
Mechanical Pretreatment	0.58	0.58	0.58	0.59	0.59	0.59	0.58	0.58	0.58
Sum	-16.81	-18.14	-23.49	-5.06	-6.86	-14.13	-15.92	-30.14	-23.14

Scenario

Figure A.4: Comparison of considered recycling processes and scenarios regarding their net CED impact [MJ/kg input]. Assessment for 1 kg of input waste. Underlying data used to create this figure can be found in the supporting information S1.

this is compensated by higher rewards for substituting primary material. Thus, the CED advantageousness of specific mechanical and chemical recycling processes cannot be clearly stated, but depends on different factors e.g. sorting yield, incineration of sorting residues and specific plant efficiencies (Sections A.3.4 and A.4). Combined recycling has the highest CED savings, combining low incineration burdens and high rewards for substituting primary material. With less yield in mechanical recycling, scenarios1.1.3 and 3.2 are comparable regarding GWP and CED, but with considerably higher revenues in scenario 3.2.

For all considered plastics, mechanical recycling obtains net revenues (Figure A.5). For HDPE, the revenues are 0.16 €/kg waste input decreasing to 0.08 €/kg with lower sorting yield. Chemical recycling has higher revenues (0.24 €/kg waste input) and the combined recycling yield maximum revenues (between 0.16 to 0.29 €/kg waste input depending on the plastics type (see Annex A6 (S2))). The reason for this are higher (virgin) product qualities and market prices, and a higher overall yield. The revenue amount depends on the re-granulate price, the substituted amount, cost of primary plastic production and the specific polymer synthesis cost. Combined recycling shows the highest carbon efficiency (74%) in scenario 3.1, outperforming chemical (59%) and mechanical recycling (20-40%) (Figure A.6).



	1.1.1	1.1.2 Baseline	1.1.3	1.2.1	1.2.2	1.2.3	2	3.1	3.2
Substitution Primary Material	-0.26	-0.26	-0.26	-0.13	-0.13	-0.13	-0.57	-0.59	-0.58
Rewards Incineration	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	-0.01
Burdens Incineration	0.06	0.06	0.06	0.07	0.07	0.07	0.02	0.02	0.02
Rewards Metall Recycling	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Landfilling Heavy Components	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Polymer Synthesis	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.09	0.12
Steam Cracking	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.06	0.08
Pyrolysis	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.09	0.12
Regranulation	0.11	0.11	0.11	0.06	0.06	0.06	0.00	0.11	0.06
Sensor-Based Sorting	0.02	0.02	0.02	0.01	0.01	0.01	0.00	0.02	0.02
Mechanical Pretreatment	-0.08	-0.08	-0.08	-0.08	-0.08	-0.08	-0.08	-0.08	-0.08
Sum	-0.16	-0.16	-0.16	-0.08	-0.08	-0.08	-0.24	-0.29	-0.25

Scenario

Figure A.5: Comparison of considered recycling processes and scenarios regarding their net product costs [€/kg input]. Assessment for 1 kg of input waste. Underlying data used to create this figure can be found in the supporting information S1.

In Germany, 5.2 million tons of post-consumer plastic wastes waste from packaging, construction, vehicles, electrical and electronic equipment, household, agriculture and others are collected annually (Conversio, 2018), including 3.8 million tons of the considered plastics. Depending on the material losses, between 0.8 (scenario 1.2.1) and 1.5 (scenario 1.1.1) million tons re-granulate per year could be kept in use. Assuming combined recycling, between 2.5 (scenario 3.2) and 2.8 (scenario 3.1) million tons/year could be recycled. Thus, 1-2 million tons/year could be kept in the economy additionally, instead of incinerating them. This would suffice to achieve both the EU and German plastics packaging recycling targets (Section A.1). Moreover, the additional plastic yield from chemical recycling would be of virgin quality.

Today, around 1.9 million tons/year re-granulates are reclaimed from the total plastic waste in Germany (=ca. 30%) annually by mechanical recycling (Conversio, 2018). However, re-granulates often have lower quality because of limited material purity, degradation of material properties, or colour impurities. Thus, they cannot be used for specific applications (e.g. food packing, medical products) (UBA, 2018a). This is discussed within the sensitivity analysis (Section A.3.4). In other EU countries, mechanical recycling rates are lower than in Germany (PlasticsEurope, 2018b),

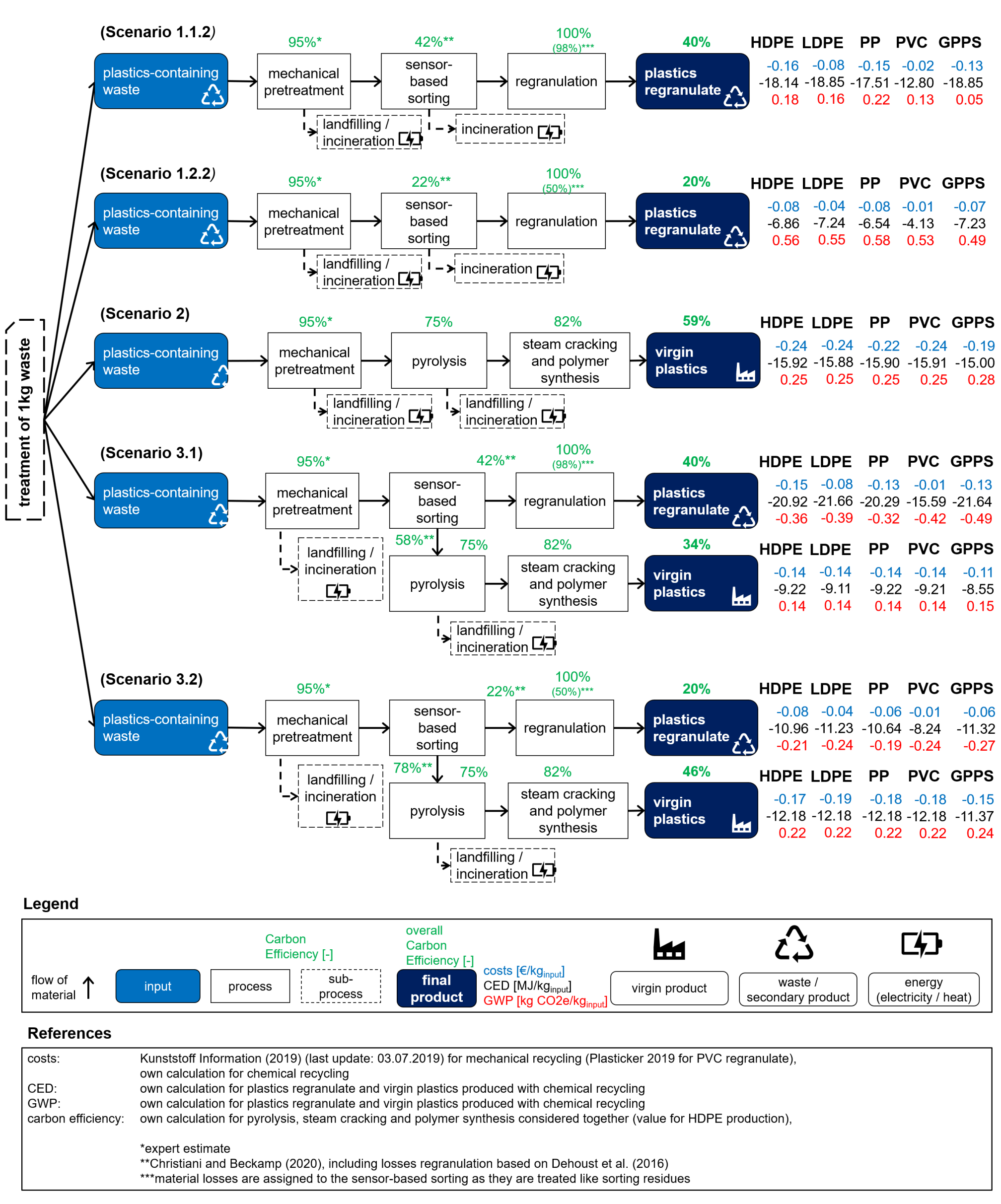


Figure A.6: Comparison of different plastic recycling paths including rewards for substituted primary plastic production (assessed in Annex A3 (S2)), power and heat gains, as well as burdens or rewards respectively for metal byproducts, incineration and landfilling. Values are given for the treatment of 1 kg of waste (see Figure S2-5 (S2) for its composition). The percentages above the process boxes indicate the carbon efficiency of the respective process while those at the final products indicate the carbon efficiency of the whole recycling option. Underlying data used to create this figure can be found in the supporting information S1.

and application of the assessed recycling technologies could realize significant GWP and CED reductions.

A.3.4 Quality of Re-granulate from Mechanical Recycling

Virgin material substitution ratio and organic contamination can significantly influence assessments of mechanical plastics recycling (Lazarevic et al., 2010; Turner et al., 2015; Gu et al., 2017;

Van Eygen et al., 2018, 2018b). Thus, a substitution ratio of 1:0.81 is recommended by Rigamonti et al. (2009) and (Turner et al., 2015) for mechanical plastics recycling to reflect lower material qualities of re-granulates. The baseline scenario does not consider reduced re-granulate qualities; these are subject to the following sensitivity analysis (see Annex A8 (S2) for details).

The sensitivity analysis shows that lower substitution ratios lead to significantly higher environmental impacts of mechanical recycling due to lower rewards for substituting primary material (Figure A.7). If more re-granulate than virgin material is needed to produce a specific product, chemical recycling is more advantageous concerning CED and GWP. For substitution ratios of 1:0.4 and higher, the combined mechanical and chemical recycling is advantageous concerning CED. For GWP, combined recycling is advantageous independently of the substitution ratio. Below a substitution ratio of 1:0.4, chemical recycling leads to higher CED savings than combined recycling. Associated costs do not change due to assumed constant market prices and market clearance (all re-granulates are sold for the given price).

The sensitivity analysis emphasizes the need to assess the re-granulate quality. In chemical recycling, all or most additives, pollutants and impurities are captured in the solid fraction and extracted from further processing. Thus, it is particularly useful to handle "difficult" plastic wastes.

A.4 Discussion

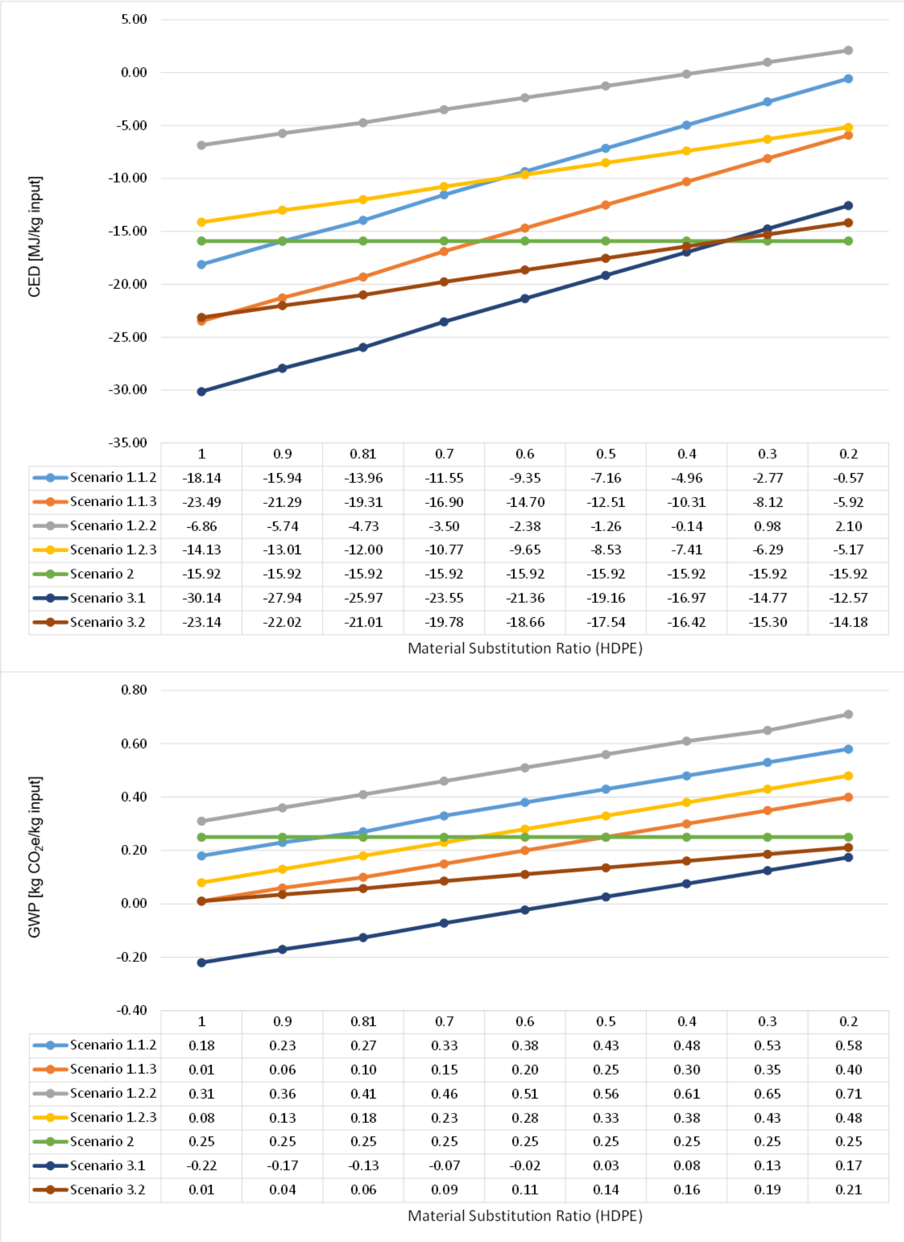
Here, the study results are discussed regarding distinctions to existing studies (Section 4.1) and limitations of this study are specified (Section 4.2).

A.4.1 Comparison of Results with Literature

For mixed plastics, Turner et al. (2015) calculated lower GWP impacts than this study. However, their data quality is rather poor and comparability can be questioned due to different assessed plastics (source-segregated plastics versus real waste with mixed plastics, paper, metals and other materials) and due to other national energy mixes. A higher carbon intensity of the energy mix results in higher rewards for energy generation, and better performance of mechanical recycling. (Gu et al., 2017) found that mechanical PE recycling has only around 76% of the GWP impact of its virgin production. We can confirm these results for PE. In this study, the average GWP impact of all assessed plastics constitutes 70% of their virgin production.

Both Chen et al. (2011) and Perugini et al. (2005) highlight synergies between various technologies, especially a cascade utilization of mechanical and chemical recycling regarding a good environmental performance. These advantages of combining mechanical and chemical recycling can be confirmed. However, a direct comparison with Chen et al. (2011) is impossible, due to differences in the methodology.

Bergsma (2019a), pyrolysis of the waste fraction led to lower GWP savings than mechanical recycling, which is consistent with our results. Meys et al. (2020) favor mechanical recycling



CED	1	0.9	0.81	0.7	0.6	0.5	0.4	0.3	0.2
Scenario 1.1.2	-18.14	-15.94	-13.96	-11.55	-9.35	-7.16	-4.96	-2.77	-0.57
Scenario 1.1.3	-23.49	-21.29	-19.31	-16.90	-14.70	-12.51	-10.31	-8.12	-5.92
Scenario 1.2.2	-6.86	-5.74	-4.73	-3.50	-2.38	-1.26	-0.14	0.98	2.10
Scenario 1.2.3	-14.13	-13.01	-12.00	-10.77	-9.65	-8.53	-7.41	-6.29	-5.17
Scenario 2	-15.92	-15.92	-15.92	-15.92	-15.92	-15.92	-15.92	-15.92	-15.92
Scenario 3.1	-30.14	-27.94	-25.97	-23.55	-21.36	-19.16	-16.97	-14.77	-12.57
Scenario 3.2	-23.14	-22.02	-21.01	-19.78	-18.66	-17.54	-16.42	-15.30	-14.18

GWP	1	0.9	0.81	0.7	0.6	0.5	0.4	0.3	0.2
Scenario 1.1.2	0.18	0.23	0.27	0.33	0.38	0.43	0.48	0.53	0.58
Scenario 1.1.3	0.01	0.06	0.10	0.15	0.20	0.25	0.30	0.35	0.40
Scenario 1.2.2	0.31	0.36	0.41	0.46	0.51	0.56	0.61	0.65	0.71
Scenario 1.2.3	0.08	0.13	0.18	0.23	0.28	0.33	0.38	0.43	0.48
Scenario 2	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Scenario 3.1	-0.22	-0.17	-0.13	-0.07	-0.02	0.03	0.08	0.13	0.17
Scenario 3.2	0.01	0.04	0.06	0.09	0.11	0.14	0.16	0.19	0.21

Figure A.7: CED and GWP impact of the material substitution ratio of HDPE re-granulate and virgin material. The material substitution rate does not change for virgin material produced by chemical recycling (scenario 2). Underlying data used to create this figure can be found in the supporting information S1.

over refinery feedstock production for the mono waste streams of PET, HDPE, LDPE, PP, and GPPS regarding GWP impact, which is also consistent with our results. Jeswani et al. (2021) found no significant difference regarding GWP and CED impact between mechanical and chemical recycling (pyrolysis), but state a high impact of thermal recycling paths and carbon intensities of the electricity mixes on the results, due to their implications for the incineration rewards/burdens. This is consistent with our results. A combined recycling path is not considered in Jeswani et al. (2021).

Although similar results are found, the comparison with other studies is difficult as the considered plastic fractions differ. Particularly, different input waste leads to different pyrolysis results and impacts the assessment of chemical recycling.

A.4.2 Shortcomings/Limitations

Despite reasonable results, this study has limitations and the results face uncertainties due to (1) data and methodological limits, (2) model limits, or (3) assumptions:

(1) First, the measured LWP composition is quite old and might not reflect the current LWP composition. PET is excluded as it is separately collected and recycled in Germany. Furthermore, pyrolysis data is limited to specific pilot plants. The theoretical pyrolysis modelling (Section A.2.3.3) based on this data requires future validation in experiments with current waste compositions. Potential changes in the plants' operational modes are not covered.

Polymer deterioration and additives (Gu et al., 2017) usually reduce material quality in mechanical recycling (Turner et al., 2015). Like Van Eygen et al. (2018b), our mechanical recycling assessment neither covers fillers/additives nor hazardous/interfering materials like phthalates (Pivnenko et al., 2016); only different substitution ratios are addressed in the sensitivity analysis (Section A.3.4). Furthermore, pollutants or impurities enter the material flow (into and onto the plastics) due to contamination during use or by other wastes. Particularly, additives like bromine, chlorine and phthalates need to be included in LCA datasets following EU regulation 1907/2006 (REACH). Compliance with this regulation is problematic for mechanical recycling, but less challenging for chemical recycling.

Furthermore, this study does not consider chemical recycling processes other than pyrolysis, such as gasification, or partial solvolytic recycling. It should be emphasized that pyrolysis results strongly depend on the applied technology, process temperature and process pressure. We used data of rotary kiln pyrolysis technology; different pyrolysis technologies might lead to varying results. This is particularly important when comparing different studies. Also, we only assessed the pyrolysis of LWP waste and transferred the results to the pyrolysis of LWP waste sorting residues. A more detailed analysis of the examined scenario must be carried out, since the data of (Andreas et al., 1981) does not allow data variation and scenario analysis. Providing this data is part of ongoing research by the authors to provide pyrolysis LCA data and enable sensitivity analysis of chemical recycling. Detailed investigations of the pyrolysis of different waste compositions is necessary and will be part of future research.

Chemical recycling technologies are less mature than mechanical recycling technologies. The lower technology readiness level of chemical recycling has not been considered in the sensitivity analysis of this study. Moreover, transportation was excluded, as its impact is relatively low; 900 km account for 10% of the treatments (Chen et al., 2011).

(2) Second, this study assesses a defined, representative LWP waste in Germany. However, it's a snapshot and thus not reflecting carbon emission-factors of other national energy mixes or its timely

change in Germany (e.g. coal exit). Thus, the study results are specific for Germany. Furthermore, the study is static and does not cover dynamics, e.g. of feedback loops, changing LWP waste compositions[12], changing market demand, cost/price variability, trends, or changing substitution ratios over time.

(3) Third, assumptions introduce uncertainties that were partly covered by scenarios (Section A.2.5) and sensitivity analyses (Section A.3.4, Annex A7 (S2)). However, assumptions might not reflect real market behaviour or re-granulate and secondary plastics applications, but might impact the assessment (particularly the calculated rewards). Market effects have been discussed and considered in a reduced substitution ratio (Section A.3.4). In reality, a mix of scenarios or sensitivities is likely, due to differing effectiveness of collection and recycling networks, plant efficiencies, and variable waste compositions.

A similar approach to assess plastics production, sorting, and recycling paths is not known to the authors. It can support decision-makers from academia, industry, and politics to make better-informed choices for optimal recycling and treatment strategies of mixed plastic wastes. Furthermore, it provides performance benchmarks for existing and new processes.

Finally, it should be noted that numerous limitations listed here are not unique to this study, but are ubiquitous in LCA studies on waste recycling (Turner et al., 2015; Chen et al., 2011).

A.5 Conclusion

Mechanical, chemical and a combined recycling of mixed plastics waste is transparently assessed, compared and analysed in different scenarios with respect to GWP, CED, carbon efficiency, and costs. Rewards and burdens are integrated for primary plastics substitution, metal by-products, incineration of non-recyclable residues, and landfilling of mineral non-combustible recycling residues, as well as GWP and CED gains from heat and power recovery. The developed assessment model is applied in a case study on separately collected LWP waste in Germany.

The results show that mechanically or chemically recycled plastics are advantageous compared to virgin plastics produced from fossil feedstock. Mechanical and chemical recycling perform similarly regarding GWP and CED, depending on sorting yield, thermal recycling paths of sorting residues and substitution ratio. Chemical recycling performs better than mechanical recycling concerning cost and carbon efficiency. Both chemical recycling and mechanical recycling are outperformed by a combination of both in all four assessed categories; particularly for GWP and CED this is valid for substitution ratios as low as 1:0.4. In the baseline, the costs are insensitive to differing substitution ratios.

Recycling potentials of LWP waste in Germany are 2.8 million tons/year when considering a combined mechanical and chemical recycling of the considered plastics. This would be sufficient to

[12] Real waste might vary in composition and amount across regions, seasons, and due to other factors, e.g. design-for-recycling or regulations (European Commission, 2019a, 2019b)

reach both EU and German recycling targets (European Commission, 2018; European Parliament and Council of the European Union, 1994). However, additional measures like design-for-recycling, CO2-taxes, higher incineration prices (European Commission, 2018), improved packaging performance (Ellen MacArthur Foundation, 2016) or management (German Bundestag, 2017) are required to reach mechanical recycling targets (Conversio, 2018; Joachim Christiani, 2017).

The study results strongly depend on local or national circumstances such as waste composition, local processing plants or energy mix (carbon intensity). Thus, they are only partly transferable. Calculated economic benefits might also not be generated, if organisational barriers, lacking stakeholder cooperation or market aloofness for recycled products persist.

Research is necessary to provide experimentally validated data specifically for chemical recycling of real mixed waste. Standardization efforts[13] aim at including mass balancing, allocation rules and chemical characteristics (e.g. calorific value) to improve data consistency, comparability and traceability of recycled feedstock into new and certified recycled products (Ellen MacArthur Foundation, 2016; PlasticsEurope, 2017). Future research should address the quality assessment within mechanical recycling. And, a detailed study of the pyrolysis process and process control for different waste compositions (beyond household waste) is needed (Vogel et al., 2020). The developed assessment model could be applied to other plastic waste or chemical recycling technologies such as gasification (Seidl et al., 2019) or solvolysis (Y.-B. Zhao et al., 2018; Schlummer et al., 2020).

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[13] E.g. for mechanical recycling (DIN EN 13430:2004), labelling (DIN EN 6120:2019), tracing (DIN EN 15343:2008, EUCertPlast certificate), technical capabilities (DIN EN 15347:2008; DIN EN 15342:2008 (GPPS); DIN EN 15344:2008 (PE); DIN EN 15345:2008 (PP); DIN EN 15346:2015 (PVC) and DIN EN 15348:2015: (PET)) and testing and quality control (DIN CEN/TS 16010/16011:2013).

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B The Impact of Secondary Materials' Quality on Assessing Plastic Recycling Technologies

Abstract[1]

Global plastic production reached a new high in 2019. The high use of plastic leads to a high amount of plastic waste. Thereof, only 33% was collected for recycling in Europe. Plastic production depends on crude oil and energy and has high environmental impacts such as greenhouse gas emissions. The recycling of plastic waste can reduce dependency on fossil resources, help reduce environmental impacts, and achieve sustainability goals. Currently, the chemical recycling of plastic is discussed to complement the existing mechanical recycling. Comparing the recycling technologies is needed to identify and establish the most environmentally and economically promising technology for each waste stream. However, the quality of the recovered material has a high impact on assessment results. This study discusses different assessment metrics for recycling technologies concerning the influence of recovered materials' quality by material substitution rates and circularity potential. In a case study, mechanical and chemical recycling via pyrolysis of HDPE from lightweight packaging waste from Germany is assessed. Mechanical recycling has a lower climate change impact than chemical recycling for material substitution rates above 0.85. On the other hand, chemical recycling has a higher potential to close the plastic loop and retain plastics within the economy due to the higher secondary material quality. The assessment allows evaluating recycling options for the considered plastics from the German collection systems for packaging.

B.1 Introduction

The amount of plastic produced globally reached a new high in 2019 (PlasticsEurope, 2020). The increase in plastic production results in an increasing amount of plastic waste that waste management systems have to handle. In Europe, plastic packaging is the most significant waste fraction, of which 58% is either landfilled or used for energy recovery (PlasticsEurope, 2020).

[1] This chapter includes the final version of the article "The impact of secondary materials' quality on assessing plastic recycling technologies" by Rebekka Volk, Frank Schultmann, and myself. The paper was published in the Conference Proceedings of the Life Cycle Management Conference 2021 as Stallkamp et al. (2022).

Accordingly, there is still great potential for closing the plastic loop. Increased recycling could support achieving sustainability goals within the chemical industry and reduce the dependency on crude oil.

Currently, mechanical recycling is the dominant way of plastic recycling. However, it faces challenges such as non-polymer impurities, polymer cross-contamination, degradation, and additives affecting the material (Pivnenko et al., 2015). These challenges negatively impact the quality of the secondary material resulting in downcycling (Hahladakis et al., 2018). Therefore, alternative recycling options are investigated to complement mechanical recycling. This includes chemical recycling options like depolymerization, gasification, hydrocracking, and pyrolysis that convert plastic waste into feedstocks for the chemical industry to produce potentially primary plastics avoiding downcycling (Davidson et al., 2021).

Mechanical and chemical recycling options have to be compared to identify the most promising technology for each waste stream. The quality of the secondary material impacts comparison. As the material quality of the secondary plastic from mechanical and chemical recycling differs, comparison should include quality assessment. Therefore, this study discusses different assessment metrics for recycling technologies concerning the influence of materials’ quality. It focuses on material substitution rates (MSR) within life cycle assessments (LCA) and a circularity potential (CP) defined by Eriksen et al. (2019). Additional performance indicators including the assessment of secondary materials’ quality can be found in Huysman et al. (2017).

B.2 Material Quality in Recycling Technologies’ Assessment

LCAs are a methodology to assess different recycling technologies. With the avoided burden approach (Nakatani, 2014), the MSR comparing primary and secondary materials’ quality is included when assessing the environmental impacts of recycling routes (Jeswani et al., 2021; Volk et al., 2021). Eriksen et al. (2019) introduce the assessment metric CP for recycling technologies to address downcycling, meaning that secondary plastics cannot be used in every application. The MSR (section B.2.1) and the CP are described (section B.2.2).

B.2.1 Material Substitution Rate

Within LCAs, the avoided burden approach rewards the secondary material with burdens associated with a respective primary material production that is avoided using the secondary material instead (Nakatani, 2014). The amount of the primary material or the field of application where the primary material can be substituted depends on the secondary material’s quality. The quality assessment in MSR captures downcycling of a material compared to the original primary material (European Commission, 2018):

$$LCI_{rec} = (1 - A) * R_2 * (LCI_{recEoL} - LCI_V^* * MSR) \tag{B.1}$$

$$MSR = Q_{Sout}/Q_P \tag{B.2}$$

Table B.1: Overview of the nomenclature of the formulas for calculating the material substitution rate.

LCI_{rec}	Life cycle inventory of recycling with credits for avoided primary material [-]
A	Factor for allocation of burdens and credits between supplier and user of the material [-]
R_2	Proportion of the material in the product that will be recycled in a subsequent system [-]
LCI_{recEoL}	Specific emissions and consumed resources arising from recycling [-]
LCI_V^*	Specific emissions and consumed resources arising from acquisition and pre-processing of primary material [-]
Q_{Sout}	Quality of the ongoing secondary material at the point of substitution [-]
Q_P	Quality of primary material [-]

Equation B.1 calculates the life cycle inventory of a recycling option. The recycling process's emissions and environmental impacts (LCI_{recEoL}) are reduced by environmental impacts arising from primary material production (LCI_*^V). The reduction is determined by the MSR comparing the secondary material's (Q_{Sout}) with the primary material's quality (Q_P) (see Equation B.2). With an $MSR = 1$, secondary and primary materials' quality are identical, and the secondary material can replace the primary material in all applications. With an $MSR = 0$, no primary material can be substituted.

B.2.2 Circularity Potential

The CP assesses the potential of recycling systems to contribute to a specific material's circularity in the long term (Eriksen et al., 2019). It includes the impact of downcycling and highlights applications where the secondary material can be employed. Therefore, physical losses (Equation B.3) and quality losses (Equation B.4) are considered, and the market sizes for secondary material and primary material applications are compared (Eriksen et al., 2019):

$$n^{rec} = M^{rec}/U^{rec} \tag{B.3}$$

$$c^{rec} = n^{rec} * (MS(Q^{rec})/MS(Q^{disp})) \tag{B.4}$$

The quality of the secondary material (Q_{rec}) is defined by the amount of non-plastic items and non-targeted polymers in the waste stream (Eriksen et al., 2019). Eight key applications for plastic in Europe are identified. They are assigned to three quality groups (low, medium, high) according to acceptable impurity levels based on legislation and quality criteria defined by plastic

Table B.2: Overview of the nomenclature of the formulas for calculating the circularity potential.

n^{rec}	Resource recovery efficiency [-]
M^{rec}	Amount of material recovered [kg]
U^{rec}	Resource potential in the waste stream [kg]
c^{rec}	Circularity potential [-]
MS	Market share where materials with quality level Q or lower can be applied [%]
Q^{rec}	Quality of secondary material [low, medium, high]
Q^{disp}	Quality of potentially displaced primary material [low, medium, high]

reprocessing facilities. Low-quality material can be used for building and construction, automotive applications, or other applications with minimal legal restrictions (Eriksen et al., 2019). Medium quality material can be additionally applied in toys, pharmaceutical packaging, and electronics. A high-quality material is also suitable for food packaging (Eriksen et al., 2019). The assigned quality class of the secondary material obtained from a recycling technology and the market share of applications of that quality class determines the CP of the recycling technology.

B.3 Applying Assessment Methods in a Case Study

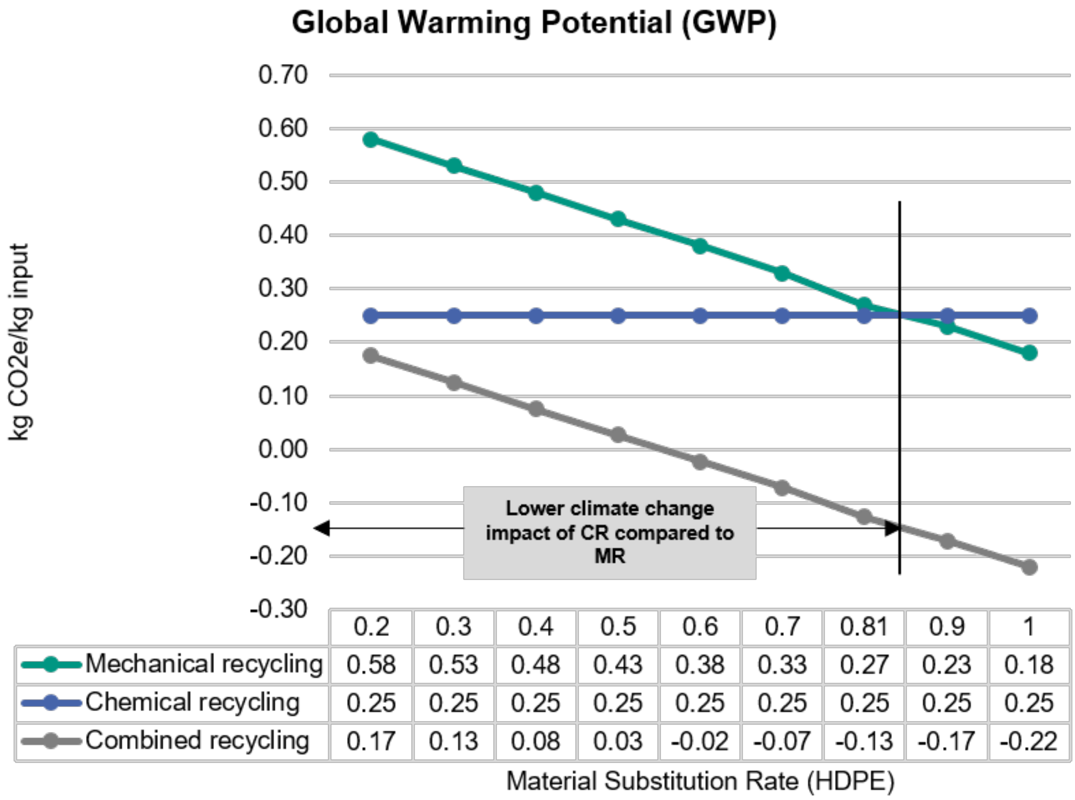
Volk et al. (2021) assess three recycling routes for plastics from lightweight packaging (LWP) waste in Germany. They assess a mechanical recycling route producing secondary plastics, a chemical recycling path producing primary-like plastics, and a combination of mechanical and chemical recycling where a share is processed to the secondary plastic and to the primary plastic, respectively. Due to data availability, MSR and CP are discussed based on this case study.

B.3.1 Material Substitution Rate

The impact of the MSR on the global warming potential (GWP) assessment results is demonstrated, as it influences the avoided burden for the substitution of primary plastics in mechanical recycling (see Figure B.1) (Volk et al., 2021). With a lower MSR, the secondary material has a lower quality than the primary material and potentially replaces less initial primary material. Thus, the reward for associated GWP decreases. This increases the net environmental impact of the mechanical recycling routes. Regarding GWP, the environmentally favoured recycling path changes at an MSR of 0.85, where 1 kg of mechanically recycled secondary plastics substitutes 0.85 kg of primary plastics (Figure B.1).

B.3.2 Circularity Potential

In addition to Volk et al. (2021), the CP for the recycling paths is calculated for HDPE (Figure B.2). The resource recovery efficiency on the mechanical recycling route is lower ($n^{rec} = 0.29$) than on the chemical recycling route ($n^{rec} = 0.71$) due to higher material losses. For mechanical recycling,



	0.2	0.3	0.4	0.5	0.6	0.7	0.81	0.9	1
Mechanical recycling	0.58	0.53	0.48	0.43	0.38	0.33	0.27	0.23	0.18
Chemical recycling	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Combined recycling	0.17	0.13	0.08	0.03	-0.02	-0.07	-0.13	-0.17	-0.22

Figure B.1: Climate change impact of the assessed recycling paths depending on the material substitution rate. Results are displayed for HDPE. Based on Volk et al. (2021) (assumptions: $A = 0$, $R2 = 1$, $LrecEol = const.$).

homogenous waste streams are needed, and, e.g., mixed plastics or multi-layer packaging cannot be recycled. It is assumed that HDPE from miscellaneous plastic packaging can be recovered on the chemical recycling route. In the combined approach, the resource recovery efficiency is nearly as high as on the chemical recycling route ($n^{rec} = 0.70$) due to the chemical recycling of sorting residues from the mechanical recycling route.

Per definition, the primary plastic is considered to be of high quality (Eriksen et al., 2019). Therefore, the market share for the potentially replaced primary material is 100% ($MS(Q_{disp}) = 1$). The application of the mechanically recycled secondary plastic within food packaging is limited due to required high-quality standards in legislation (WG PE, 2020). Therefore, it is assumed that the mechanically recycled secondary plastic falls within the medium quality class. Based on an overview of the European polymer market (Eriksen et al., 2019), the market share of the mechanically recycled secondary plastic (market share of medium or low-quality HDPE) is 73% ($MS(Q_{rec}) = 0.73$). It is assumed that chemically recycled secondary plastics are of high quality and can be applied in all application classes (Volk et al., 2021). The market share of chemically recycled secondary plastic is 100% ($MS(Q_{rec}) = 1$). In the combined recycling, 42% of the waste is recycled mechanically, and 58% is recycled chemically. Thus, the weighted market share of the combined approach is 89% ($MS(Q_{rec}) = 0.89$). This results in a CP for the mechanical recycling of HDPE of 21%, assuming a steady-state HDPE market and no material losses. Chemical recycling has a CP of 71%, and the combined recycling approach has a CP of

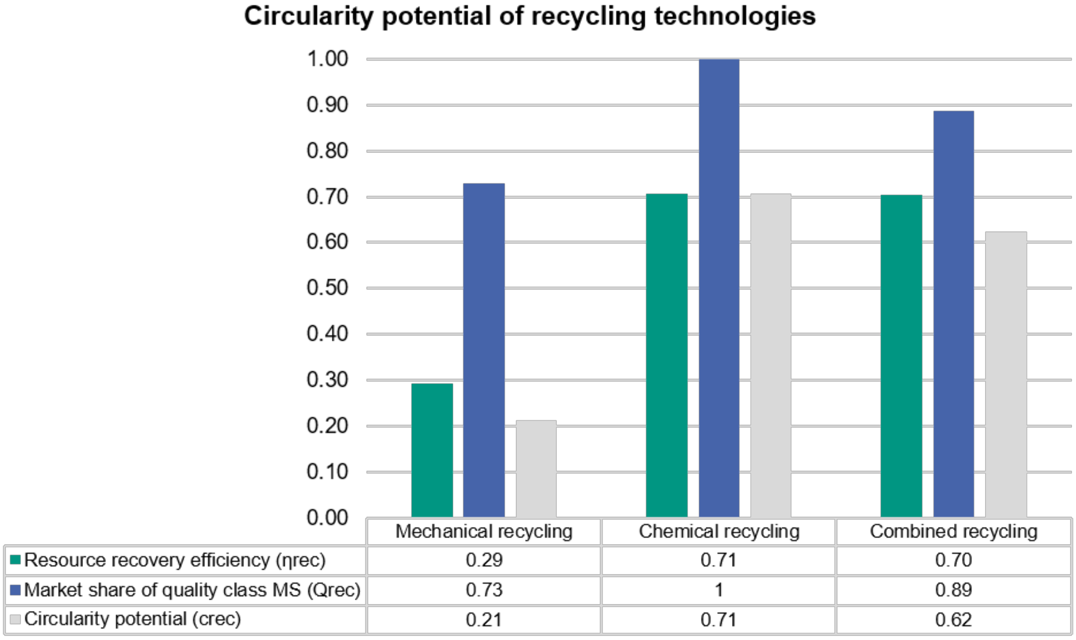


Figure B.2: Circularity potential of the assessed recycling technologies for HDPE in Volk et al. (2021).

62%. Besides different recovery efficiencies, the different qualities of mechanically and chemically recycled plastics lead to the different circularity potentials of the recycling technologies.

B.3.3 Comparing and Discussing MSR and CP

MSR and CP are not directly comparable, as the CP does not impact the LCA results but is a separate performance indicator. However, both MSR and CP assess secondary materials' quality facing the challenge that quality is not further defined, and there is no consistent methodology to assess it. The European Commission (2012) formulates three approaches to compare material quality based on (1) material analysis and physical indicators, (2) economic indicators, or (3) qualitative discussions. However, they do not establish a uniform definition of a material's quality. A material analysis allows the establishment of its quality based on technical properties, such as molecular weight, tensile strength, or density (Rigamonti et al., 2020). A possible economic indicator would be the market price of the secondary material (Jeswani et al., 2021), where the differences between the market value of primary and secondary materials are an approximation for material quality differences. Alternatively, a qualitative discussion of the material quality and a sensitivity analysis is possible (Volk et al., 2021). Often, the available data dictate the indicator used to compare material qualities.

The methodology of the CP highlights the challenges in quantitatively evaluating material qualities. Secondary materials' quality is assessed by defining three quality classes focusing on non-plastic items and non-targeted polymers in the waste stream (Eriksen et al., 2019). Degradation and the presence of additives (Pivnenko et al., 2015) are excluded due to few data (Eriksen et al., 2019). Furthermore, a single indicator cannot represent the quality for all possible application types (Eriksen et al., 2019) as the quality of plastic depends on a wide range of properties such as physical and chemical composition. Moreover, three quality classes might not capture the wide range of plastic applications.

Regardless of how secondary materials' quality is measured, it impacts the assessment results of environmental indicators and CP. A decrease in materials' quality results in a decreasing performance of the assessed recycling systems. All in all, MSR and CP are not comparable but deal with the same challenges assessing secondary materials' quality.

B.4 Conclusion

Two approaches were introduced and compared that integrate the quality of the secondary material into recycling options' comparison. The MSR is part of the avoided burden approach within LCAs. The CP is a performance indicator for recycling systems focusing on the potential to close the material loop. Although the approaches are not directly comparable, they face the same challenge of determining and assessing the quality of secondary materials. However, there is no single or standard definition for plastic material quality and, therefore, there are multiple ways to determine it. A standardized approach to assess secondary material quality is lacking to ensure comparability of assessments, and approaches depend significantly on available data. Before utilizing economic indicators, a material analysis should be done, and qualitative discussions can be conducted when no data are available. In general, using a single indicator to represent secondary material quality seems insufficient, as the quality of plastic depends on a wide range of properties. Additionally, multiple metrics should be used to assess plastic recycling technologies. This is highlighted by the inconsistent results for MSR and CP indicating the lowest global warming potential is achieved combining mechanical and chemical recycling, however, outlining that the highest circularity potential provides the chemical recycling of LWP waste.

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C Economic and Environmental Assessment of Automotive Plastic Waste End-of-Life Options – Energy Recovery versus Chemical Recycling

Abstract[1]

Most automotive plastic waste is landfilled or used in energy recovery as it is unsuitable for high-quality product mechanical recycling. Chemical recycling via pyrolysis offers a pathway toward closing the material loop by handling this heterogeneous waste and providing feedstock for producing virgin plastics. This study compares chemical recycling and energy recovery scenarios for automotive plastic waste regarding climate change impact and cumulative energy demand (CED), assessing potential environmental advantages. In addition, an economic assessment is conducted. In contrast to other studies, the assessments are based on pyrolysis experiments conducted with an actual waste fraction. Mass balances and product composition are reported. The experimental data is combined with literature data for up- and downstream processes for the assessment. Chemical recycling shows a lower net climate change impact (0.57 to 0.64 kg CO_2e/kg waste input) and CED (3.38 to 4.41 MJ/kg waste input) than energy recovery (climate change impact: 1.17 to 1.25 kg CO_2e/kg waste input; CED: 6.94 to 7.97 MJ/kg waste input), while energy recovery performs better economically (net processing cost of -0.05 to -0.02 €/kg waste input) compared to chemical recycling (0.05 to 0.08 €/kg waste input). However, chemical recycling keeps carbon in the material cycle contributing to a circular economy and reducing the dependence on fossil feedstocks. Therefore, an increasing circularity of automotive plastic waste through chemical recycling shows a conflict between economic and environmental objectives.

[1] This chapter includes the preprint version of the article "Economic and environmental assessment of automotive plastic waste end-of-life options – energy recovery versus chemical recycling" by Malte Hennig, Rebekka Volk, Frank Richter, Britta Bergfeldt, Salar Tavakkol, Frank Schultmann, Dieter Stapf and myself. The paper is accepted for publication in the Journal of Industrial Ecology as Stallkamp, Hennig, Volk, Richter, et al. (2023). The supplementary material can be found on the journal website.

C.1 Introduction

The amount of plastic in automobiles increased considerably in the last decades to realize the advantages of plastic components, such as lower costs and weight reduction (Wilts et al., 2016). Therefore, more automotive plastic waste will have to be handled in the future. Defect plastic components are replaced during the use phase generating mixed automotive plastic waste (APW). At the end-of-life (EoL), a highly heterogeneous automotive shredder residue (ASR) contains mixed plastics of different origins. Currently, landfilling and energy recovery are the dominating waste-handling options for APW and ASR in Europe (Cossu and Lai, 2015; Mehlhart et al., 2018), resulting in high environmental burdens and a loss of valuable resources likewise. Enhancing the circular economy with advanced recycling technologies is a strategy to keep carbon from waste available as feedstock for the chemical industry and reduce its dependency on fossil carbon feedstock (Meys et al., 2021).

Mechanical recyclers already face the challenge of complex waste mixtures and composite materials handling, e.g., lightweight packaging waste. However, automotive plastic waste additionally contains high shares of non-standard functionalized engineering thermoplastics, thermosets, and elastomers resulting in polymer cross-contamination and non-polymer impurities within recovered waste fractions (Cossu and Lai, 2015; Pivnenko et al., 2015) that pose an additional challenge for mechanical recycling processes. Therefore, mechanical recycling of automotive plastics can alter the recyclates' mechanical properties (Bernasconi et al., 2007; Colucci et al., 2017; Pietroluongo et al., 2020; Yang et al., 2018). New waste handling options are needed to reduce environmental impacts, decrease the dependence on fossil feedstocks by increasing circularity while at the same time being cost-competitive in comparison to current waste handling practices.

Chemical recycling processes can close the material and carbon loop by handling heterogeneous waste streams unsuitable for mechanical recycling (Dogu et al., 2021) and providing feedstock for virgin plastics production (Meys et al., 2020; Solis and Silveira, 2020). Pyrolysis as a chemical recycling option is currently assessed for multiple waste streams (Davidson et al., 2021; Kusenberg, Faussone, et al., 2022; Volk et al., 2021; Zeller et al., 2021), with promising results for waste streams primarily consisting of polyolefins.

The literature on pyrolysis can be divided into technology research and technology assessments. Technology-focused research investigates the pyrolysis process in detail regarding feedstock material, reactor type, product composition, and process parameters. This includes studies on the pyrolysis of ASR (Cossu et al., 2014; Galvagno et al., 2001; Notarnicola et al., 2017). However, no studies on the pyrolysis of APW are known. Studies of the second category focus on economic feasibility and environmental impact. Due to the lack of data from established processes or a low technology readiness level (TRL), assessments are made either based on thermodynamic considerations (Meys et al., 2020), experiments with different waste fractions (Cardamone et al., 2022) or laboratory scale experiments of virgin instead of post-consumer material (Gracida-Alvarez et al., 2019a, 2019b). Jeswani et al. (2021) used experimental data from larger-scale experiments. However, the data is not publicly available due to confidentiality and cannot be validated. While these studies can provide valuable input regarding the potential of different chemical recycling processes, there is the potential for misleading results when experimental data from technology research-oriented studies not specifically designed for process assessment are used. This may be the case if data from a waste stream is used to approximate another waste fraction that behaves differently in pyrolysis.

Therefore, this study follows an integrated approach by conducting pyrolysis experiments according to the subsequent process assessment's needs. A sample from an actual APW is taken, thoroughly characterized, and pyrolyzed in a pilot-scale continuous reactor with a throughput of approximately 1 kg/h. Mass balances and pyrolysis gas and oil product composition are reported for transparency. The chemical recycling process chain includes upstream mechanical pre-treatment of the APW, pyrolysis, and downstream upgrading of the liquid pyrolysis product to the specifications of steam cracker feedstock (naphtha substitute) followed by high-value chemical (HVC) production through steam cracking. Literature data for up- and downstream processes are combined with experimental data from the pyrolysis of APW to create a life cycle inventory (LCI) and enable life cycle assessment (LCA) before the commercialization of pyrolysis technology at a large scale. The economic indicator of process costs and the environmental indicators of climate change impact and cumulative energy demand (CED) are compared with the current waste-handling practice of energy recovery. Consequently, this study contributes to understanding different waste treatment options for automotive plastics and their contribution to a sustainable circular economy.

C.2 Methodology

Besides the experimental analyses, this study combines material flow cost analysis (MFCA) with LCA methods (Rieckhof and Guenther, 2018). Due to missing data, e.g., elemental flows, the LCA is streamlined by narrowing the considered environmental impacts to climate change impact and CED (Gradin and Björklund, 2021).

C.2.1 Scope

The assessed chemical recycling path is derived from the current energy recovery route for mixed APW collected in automotive workshops. The system boundaries of both considered processes include metal recycling and energy recovery (Figure C.1). Table S1-1 in the supporting information (S1) summarizes them. The functional unit of the assessment is the treatment of 1 kg of APW collected from automotive workshops.

C.2.1.1 Energy Recovery Path

APW is sent to refuse-derived fuel (RDF) production. In RDF production, the APW is shredded, and steel, aluminum, and copper are separated and sent to respective recycling processes. The RDF is incinerated in municipal solid waste incineration (MSWI) and RDF power plants. Energy

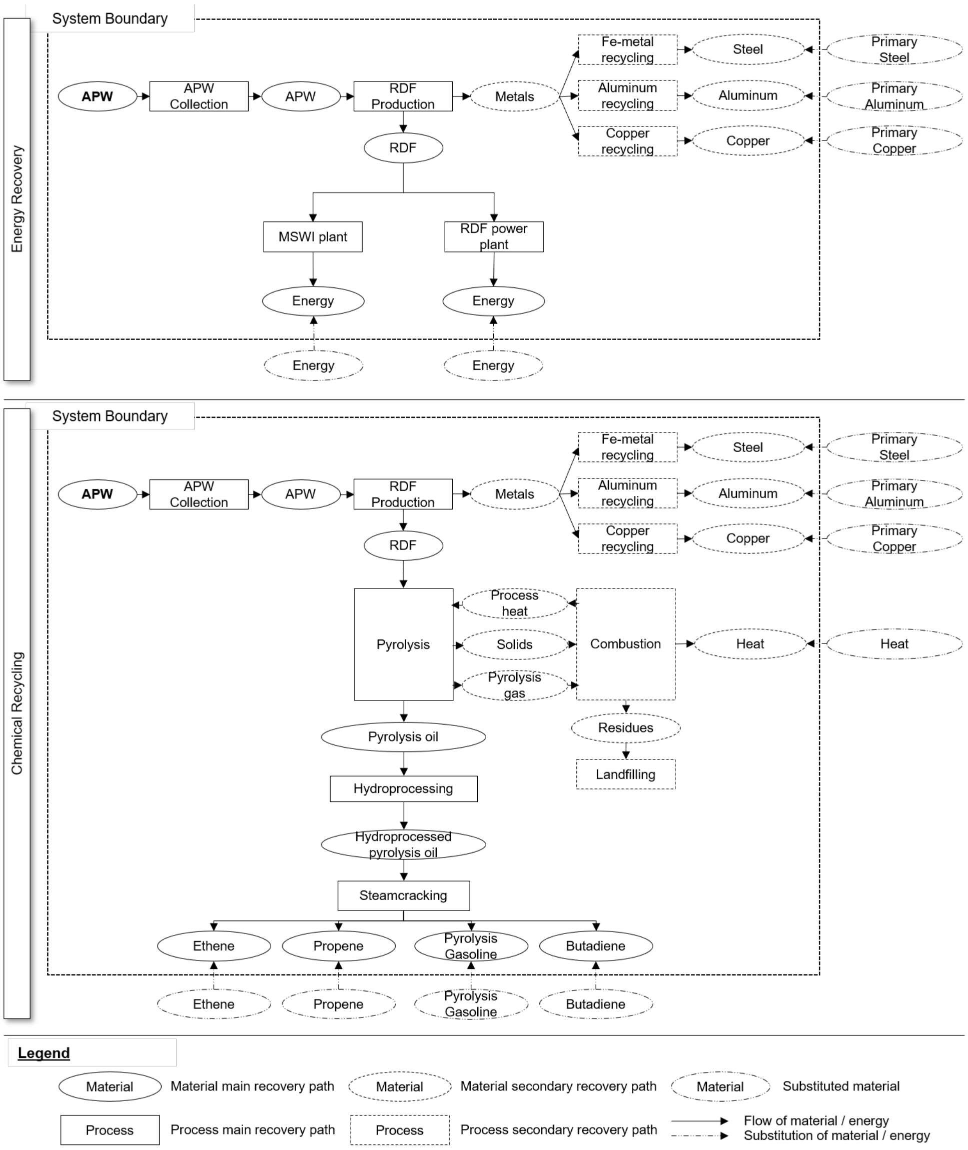


Figure C.1: System boundaries for the energy recovery and chemical recycling paths, including primary material and energy substitution.

is recovered through heat and electricity based on average efficiencies for MSWI and RDF power plants.

C.2.1.2 Chemical Recycling (Pyrolysis) Path

Following RDF production, RDF is fed into an integrated pyrolysis unit producing pyrolysis oil as the main product and pyrolysis gas, an aqueous condensate, and a solid pyrolysis residue fraction as by-products. Gas and oil fractions mainly consist of hydrocarbons; the pyrolysis residue fraction consists of mineral residues and carbonaceous char. Based on their calculated net calorific values, it is assumed that high-temperature process heat for the pyrolysis can be provided by the incineration of pyrolysis by-products, i.e., pyrolysis residues, pyrolysis gas, and aqueous condensate. The plant design assumes excess low-temperature heat can be sold to customers, such as a district heating network. Ashes are landfilled. Furthermore, the pyrolysis oil is assumed to be upgraded in a hydroprocessing step, replacing naphtha as a steam-cracking feedstock. By steam cracking, high-value chemicals (HVC) ethene, propene, butadiene, and pyrolysis gasoline are produced, which are raw materials for synthesizing chemical products, including virgin plastics.

C.2.2 Impact Assessment

An economic and environmental assessment of both recovery paths (Figure C.1) is performed where each process step is assessed individually, and all process steps along the recycling path are summed up. The APW is not associated with production or use phase burdens following the zero-burden approach (Nakatani, 2014). Products and marketable by-products are rewarded with the impact of primary material production following the avoided burden approach (Nakatani, 2014). Impacts are allocated based on mass. Intermediate products are associated with the burdens of the previous treatment steps.

The economic assessment includes fixed operational costs[2] based on the plant investments and variable utility operating costs. The base year of the economic assessment is 2021. Products and marketable by-products result in revenues that are realized. Costs of CO_2 emissions are internalized based on direct CO_2 emissions of each process step and CO_2 emission prices of the EU emission trading system. The avoided CO_2 emissions from the primary production of products and by-products are rewarded with a negative emission price.

The streamlined LCA assesses the climate change impact over 100 years as defined by the IPCC (2013). The CED, as defined by VDI (2012), is evaluated to assess the energy impacts of the recovery paths (Iacovidou et al., 2017).

C.2.3 Scenario Definition

Scenarios analyze the influence of underlying data and assumptions on the assessment (Table C.1). The energy recovery baseline scenario (scenario 1.1) is the incineration of RDF in MSWI plants (30%) and RDF power plants (70%). The incineration shares reflect the current practice in European countries (Jeswani et al., 2021; Van Eygen et al., 2018). Scenario 1.2 assesses the RDF combustion in more efficient RDF power plants only.

The baseline scenario for chemical recycling (scenario 2.1) uses the conducted pyrolysis experiments' data (section C.3). Pyrolysis oil is used to produce HVC, while pyrolysis gas and solids are used for energy recovery. Scenario 2.2 considers a lower pyrolysis oil yield than scenario 2.1, assigning 10% of pyrolysis oil yield to the pyrolysis residue.

[2] Maintenance, insurance, and general plant overhead including human resources, research and development, information technology, finance, and legal (Larrain et al., 2020).

Table C.1: Scenario overview of process chain assessment for APW waste.

Scenario No.	Scenario Description	Description
1.1	Energy recovery (baseline)	Incineration path of produced RDF: 30% MSWI plant, 70% RDF combustion plant
1.2	Energy recovery (optimized)	Incineration path of produced RDF: 100% RDF combustion plant
2.1	Chemical recycling (baseline)	Yield of pyrolysis products according to conducted experiments[1]: 50% pyrolysis oil, 20% pyrolysis gas, 29% pyrolysis residue, 2% aqueous condensate.
2.2	Chemical recycling (lower yield)	Yield of pyrolysis products adapted: 45% pyrolysis oil, 20% pyrolysis gas, 34% pyrolysis residue, 2% aqueous condensate[1]

[1]: Pyrolysis product distribution converted to a feedstock free of metals.

C.3 Experimental Assessment of Pyrolysis

The pyrolysis technology assessment and LCI data for the pyrolysis are based on pilot-scale experiments using collected APW material. The material was thoroughly characterized. Mass balances and energy consumption for pyrolysis were determined from experiments, and pyrolysis oil product quality was assessed based on heteroatom content. The pyrolysis experiments included metals in the feedstock. Therefore, this section reports pyrolysis product distribution based on the full feedstock. For process assessment, the product distribution is converted to metal-free feedstock based on a metal content of 9% in the feedstock material as the metal content is recovered in RDF production in both paths. Details are provided in the SI (A3, S1).

C.3.1 Characterization of Automotive Plastic Waste Sample

The automotive plastic waste sample was obtained from waste collected at automotive workshops around Stuttgart, Germany. First, all parts were inventoried. Large metal parts, such as bumper cross beams, were excluded from the sample. Smaller metal parts remained in the sample and were not removed before the experiments. From the automobile manufacturer's database, sample composition was estimated to be polyolefins (57%), polycarbonates and blends thereof (19%), polyamides (5%), other polymers (12%), and other non-polymer materials (e. g., metals, minerals, biomass) (7%). However, manual sorting of a shredded sample showed a significant deviation of the metal content (3.5% ferrous and 5.5% non-ferrous metals), making deviations of other components' shares likely. For elemental balances of the pyrolysis experiments, the pre-sorted material's elemental composition and ash content were determined (Table S1-4, S1).

C.3.2 Pyrolysis Experimental Set-up

The sample was shredded to a particle size of 10 mm. The experiments were conducted in an electrically heated continuous pilot-scale screw reactor with a throughput of plastic waste of approximately 1 kg/h and a solids residence time of 45 minutes at a temperature of 450 °C. A moderator medium (sand) improves heat and mass transfer properties at a 1:4 mass ratio (feedstock: moderator). The reactor has already been described elsewhere (Tomasi Morgano et al., 2018; Zeller et al., 2021). The supporting information describes analytical techniques used for material and product characterization (A2, S1). Five repetitive experiments with 5 kg of waste material each determined the mean pyrolysis product distribution.

C.3.3 Pyrolysis Products

Approximately 45 wt% of the waste is converted to pyrolysis oil (cf. Figure C.2). 2 wt% is converted to an aqueous condensate. Pyrolysis gas makes up 18 wt%, while 31 wt% is retained as pyrolysis residue, consisting of mineral fillers, glass fibers, and metals. On average, 4 wt% of the sample weight is lost (balance loss) due to encrustations within the reactor and measuring inaccuracies. Elemental analysis of condensates shows that pyrolysis oil contains approximately 80 wt% carbon and 12 wt% hydrogen (cf. Table S1-7, S1). The pyrolysis oil contains 8 wt% of heteroatoms, the main share being oxygen and 1 wt% water. The chlorine content in the pyrolysis oil is below 0.1 wt%. Pyrolysis gas consists of carbon dioxide as a principal component. Additionally, it contains significant amounts of C1 to C4 hydrocarbons and a minor amount of hydrogen. A detailed overview of pyrolysis product distribution, composition, and element mass balances of pyrolysis is given in section A2 of the supporting information.

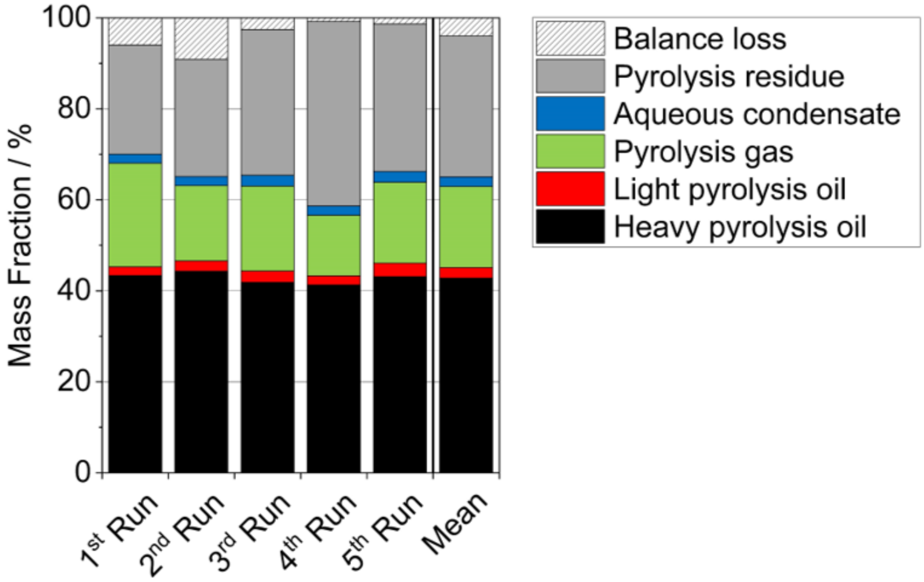


Figure C.2: Mass distribution of pyrolysis products in repetitive experimental runs under equal conditions. Underlying data used to create this figure can be found in Supporting Information S2.

C.3.4 Utilization of Pyrolysis Products

Pyrolysis oil can be used as feedstock for steam cracking to produce ethene and propene (Kusenberg, Roosen, et al., 2022). However, steam cracker specifications regarding heteroatom and olefin content require an additional hydroprocessing step for upgrading pyrolysis oil (Kusenberg, Eschenbacher, et al., 2022). The gas from the pyrolysis of APW contains valuable C3 and C4 olefins, but the low gas yield and a high share of carbon dioxide prevent the material usage. Instead, it can be used as fuel for the pyrolysis process heat required having a net caloric value of 19 MJ/kg. Pyrolysis residue (9 MJ/kg) and aqueous condensate (-2 MJ/kg) must be disposed of and can also serve as fuel for process heat generation.

C.4 Life Cycle Inventory Data

General assumptions and parameters for the assessments are summarized in Table S1-2 in the SI, and additional detailed LCI data is also provided in the SI (A3, S1). The following subsections describe the subprocesses and LCI data for the different end-of-life options.

C.4.1 APW Collection

A disposal company collects APW from automotive workshops and supplies it to RDF producers. The economic assessment of the disposal company is limited to the material-specific treatment costs considered in the MFCA. The cost associated with a transfer station is up to 0.02 €/kg input (Bilitewski et al., 2018). Environmental impacts are not assessed as they are assigned to transport burdens only (section C.4.8).

C.4.2 RDF Production

APW is shredded and sorted. A share of 9% metals can be recovered and separated into ferrous (39%) and non-ferrous metals (61%). An analysis of the non-ferrous metals in the feedstock sample for the pyrolysis experiments demonstrates the domination of aluminum and copper (>90%) with a balanced mass ratio between them. Thus, we calculate with a share of 50% aluminum and 50% copper for all non-ferrous metal by-products. Process costs include electricity costs and fixed operating costs of 10% (Larrain et al., 2020; Riedewald et al., 2021) of the investment. Investments, electricity costs, and environmental impacts for an RDF production plant with a capacity of 20,000 Mg/year and operating 7,500 h/year are derived from an exemplary production process based on manufacturer specifications that are scaled up to the assessed capacity, and by employing a plant factor (Stapf et al., 2019). The investment adds up to 880,000 €/year; operation expenses result in 0.06 €/kg input. Environmental impacts are derived from the process's electrical energy demand of 1.17 MJ/kg input multiplied by the German CO_2- and CED-factors of the electricity grid (Table S1-2, SI). The CO_2-factor describes the CO_2 emissions associated with producing 1 kWh of electricity. In contrast, the CED-factor describes the primary energy needed to generate 1 kWh of usable energy. Both factors also exist for the German heat mix. The mechanical pre-treatment has environmental impacts of 2.80 MJ/kg input (CED) and 0.14 kg CO_2/kg input (climate change impact). The LCI data is summarized in Table S1-8 (S1).

C.4.3 Metal Recycling

Separated metals are sent to conventional recycling processes. Their environmental impacts are derived from ecoinvent datasets[3], and processing costs are based on energy demand. The environmental rewards for substituting iron scrap in steel production are also based on an ecoinvent dataset[4]; the financial compensation is assumed with 0.025 €/kg (Stapf et al., 2019). For aluminum and copper, a compensation of 0.25 €/kg (Stapf et al., 2019) is assumed, and environmental rewards are derived from respective ecoinvent datasets[5].

C.4.4 Energy Recovery

Relevant data for the incineration in MSWI plants and RDF power plants is provided in Table S1-9 (S1). Processing costs are assumed to be 0.12 €/kg input (Bilitewski et al., 2018), and the environmental impacts are calculated to be 26.3 MJ/kg input (CED) and 2.47 kg CO_2/kg input (climate change impact) based on the APW's elemental composition. Rewards are calculated based on the efficiency of the energy recovery facilities, the recovered energy (electricity and heat), and respective CO_2- and CED-factors of the recovered energy.

C.4.5 Pyrolysis

The pyrolysis plant converts RDF to pyrolysis oil, pyrolysis gas, and pyrolysis residues. Mass and elemental balances are closed by adding balance losses to the pyrolysis residue fraction. Adjusted experimental data for a metal-free feedstock is used for product distribution and composition (A3.3, S1).

Pyrolysis oil is sent to a hydroprocessing unit for upgrading to steam cracker specifications (section C.4.6). The incineration of pyrolysis by-products supplies the high-temperature heat demand of the pyrolysis unit. Excess low-temperature heat is supplied to a district heating network. The remaining ashes are landfilled. Climate change impact and CED of pyrolysis are calculated based on the amount of carbon dioxide released from the incineration of pyrolysis by-products (i.e., pyrolysis gas, pyrolysis residue, and aqueous condensates) (climate change impact) and their respective net

[3] For the aluminum recycling the datasets from Müller (2020) and Lesage (2020) are used while Classen (2020) is applied for copper recycling.

[4] Ecoinvent dataset from Wernet (2020).

[5] For aluminum production the dataset from Jungbluth (2020) and for copper production the dataset from (Turner, 2020) is applied.

calorific value (CED). Rewards are granted for excess heat substituting conventionally derived district heating. This results in a net climate change impact of 0.51 kg CO_2/kg input (A3.3, S1) and a net CED of 4.49 MJ/kg input. Total processing costs are estimated to be 0.16 €/kg input.

C.4.6 Hydroprocessing

Raw pyrolysis oil has to be upgraded to fulfill the demands of the petrochemistry processes designed for fossil feedstocks (Kusenberg, Eschenbacher, et al., 2022). This includes removing heteroatoms and saturation of double bonds by hydrotreating and adjusting the pyrolysis oil boiling curve by hydrocracking.

Pyrolysis oil differs from conventional feedstocks mainly in terms of oxygen content and the occurrence of double bonds. Only little research has been conducted on the hydroprocessing of pyrolysis oil from plastic waste. However, for pyrolysis oil from polyolefinic plastic waste, it has been demonstrated on a laboratory scale that upgrading pyrolysis oil is feasible (Neuner et al., 2022). Therefore, the composition of hydroprocessed pyrolysis oil and the demand for hydrogen is estimated based on general assumptions (A3.4, S1). Nevertheless, further investigation of the hydroprocessing process for pyrolysis oil is necessary to validate the assumptions made.

A part of the estimated amount of hydrogen needed for hydroprocessing can be separated from steam cracker product gas C.4.7). Required additional hydrogen is assumed to be purchased. Here, we calculate with hydrogen produced as a by-product in an oil refinery. Associated environmental impacts are based on an ecoinvent dataset (Brunner, 2021), and prices are assumed to be 1575 €/Mg H2 (German Bundestag, 2020). The hydroprocessed pyrolysis oil is sent to the steam cracker. Due to lacking information, a detailed hydroprocessing simulation is impossible. Thus, no energy demands for pumps and gas compression can be calculated, and estimating climate change impact and CED is not possible. In addition, no credits or burdens are considered for treating hydroprocessing by-products and utilization of excess hydrogen.

Hydroprocessing costs are considered in terms of fixed operating expenses and costs for hydrogen, as no energy demands are known. The investment for hydroprocessing is based on a hydroprocessing unit for vegetable oil (Marker, 2005) with costs for hydroprocessing of 0.10 €/kg input.

C.4.7 Steam Cracking

It is assumed that hydroprocessed pyrolysis oil replaces naphtha as steam-cracking feedstock. The life cycle inventory for producing primary HVC is used (PlasticsEurope, 2012); no further detailed assessment of steam cracking is performed. The assumed product yield is based on the steam cracking process with naphtha feedstock (Figure S1-2, S1).

C.4.8 Transportation

Assumptions and references for the transport distances are summarized in Table S1-12 (S1). Based on a spatial analysis for Germany, total transportation distances between automotive workshops, disposal companies, RDF production, and energy recovery are 115 km. For chemical recycling, instead of MSWI or RDF power plants, the average distance between RDF production and chemical plants with German steam crackers is considered. The spatial analysis results in an average distance of 247 km. Transportation for pyrolysis products or hydroprocessed pyrolysis oil is not considered since we assume the integration of pyrolysis and hydroprocessing into existing chemical plants. Environmental impacts and costs are provided in the SI (A3.6, S1).

C.5 Results and Discussion

The environmental impacts and economic assessment of energy recovery and chemical recycling are calculated based on the experimental and LCI data collected.

C.5.1 Energy Recovery Scenarios

The baseline scenario of the energy recovery results in gross costs of 0.34 €/kg input and induces gross values of 2.73 kg CO_2e/kg input (climate change impact) and 31.80 MJ/kg input (CED). All RDF is incinerated in either an MSWI or RDF power plant. Incineration accounts for 83% of the climate change impact and 75% of the CED impact. The net values consider substitution rewards for energy and primary metals, resulting in an economic assessment of -0.02 €/kg input (Figure C.3), 1.25 CO_2e/kg input (climate change impact, Figure C.4), and 7.97 MJ/kg input (CED, Figure C.5).

The results (Figure C.4, Figure C.5) show that the energy recovery in RDF power plants (scenario 1.2) leads to net reductions of 7% (climate change impact) and 13% (CED) compared to the baseline scenario 1.1. The economic assessment improves by 89% (Figure C.3). The higher energy efficiency of RDF power plants and associated substitution rewards achieve the improvements.

C.5.2 Chemical Recycling (Pyrolysis) Scenarios

Chemical recycling of APW (scenario 2.1) results in gross processing costs of 0.49 €/kg input and induces gross values of 1.54 kg CO_2e/kg input (climate change impact) and 33.69 MJ/kg input (CED). It leads to HVCs production of 0.13 kg ethene, 0.06 kg propene, 0.04 kg butadiene, and 0.08 kg pyrolysis gasoline (Figure C.6). Figure C.6 also outlines the carbon flow of the chemical recycling route, indicating the share of carbon that can potentially be recovered and contribute to a circular economy. Steam cracking significantly impacts all indicators[6] due to its high energy

[6] Steam cracking accounts for 29% of the cost, 19% of the climate change impact and 50% of the CED.

demand and emissions. Pyrolysis substantially influences the cost of the chemical recycling path (36%) due to increased investments and a high impact on the environmental indicators[7] due to the incineration of the pyrolysis by-products providing process heat. The net values of scenario 2.1 are an economic assessment of 0.08 €/kg input, 0.57 kg CO_2e/kg input (climate change impact), and a CED of 3.38 MJ/kg input.

In scenario 2.2, a decrease in pyrolysis oil yield is assumed, and the mass balance difference is allocated to the solid fraction. Thus, more solids are incinerated, increasing climate change impact and CED. Higher rewards for recovered energy do not fully compensate for this increase, leading to a net climate change impact of 0.64 kg CO_2e/kg input. The CED impact increases to 4.41 MJ/kg input. The net processing costs along the value chain decrease to 0.05 €/kg input because the high energy prices make the revenues from recovered heat higher than those from produced HVCs. The reduced pyrolysis oil yield results in fewer produced HVCs and less carbon potentially available for a circular economy.

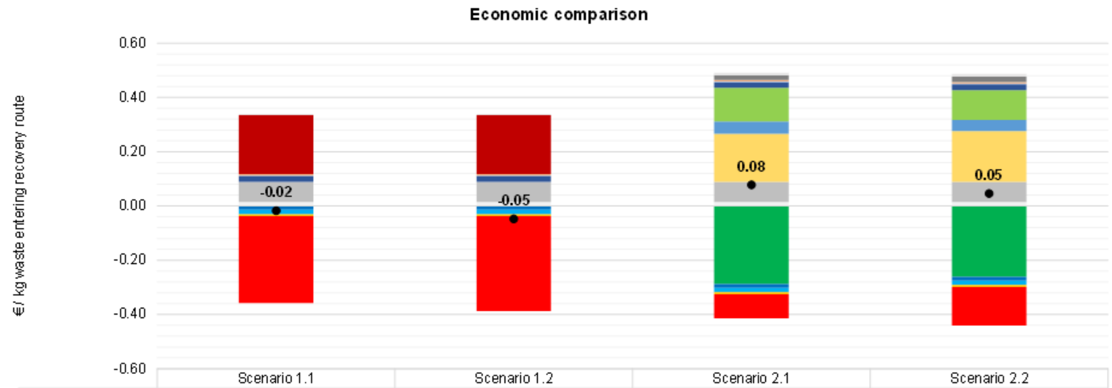
C.5.3 Comparison of Energy Recovery and Chemical Recycling (Pyrolysis)

Economically, energy recovery performs better than chemical recycling. This is due to high rewards for producing and substituting energy and lower gross processing costs due to fewer processing steps along the value chain. Energy recovery performs even better using more efficient RDF power plants with higher rewards for recovered energy and lower emissions. Economically, chemical recycling performs better with a lower pyrolysis oil yield due to an increased share of incinerated material and associated rewards for providing excess heat. Lower pyrolysis oil yields also result in lower processing costs at the steam cracking plant, and the higher rewards for substitute district heating compensate for lower rewards for the avoided primary production of HVCs. This is an effect of the high energy prices in 2021 used in the assessment.

The climate change impact of the energy recovery scenarios is influenced by the incineration paths and their efficiencies (Figure C.4). Incineration in efficient RDF power plants reduces the climate change impact compared to MSWI plants due to higher substitution rewards for generated heat and electricity. Regarding climate change impact, chemical recycling performs considerably better than energy recovery. Rewards for substituting primary HVCs counterbalance the high impacts of steam cracking and pyrolysis. In the chemical recycling scenarios, the impact of climate change increases with decreasing pyrolysis oil yield.

The CED impact of energy recovery decreases with more efficient incineration paths as more electricity and heat can be recovered (Figure C.5). Both chemical recycling scenarios show lower CED impacts compared to the energy recovery. The CED savings decrease with decreasing pyrolysis oil yield as fewer HVCs are produced. This leads to lower rewards for the avoided primary production that cannot be compensated by increasing rewards for recovered energy.

[7] climate change impact: 48% and CED: 21%



	Scenario 1.1	Scenario 1.2	Scenario 2.1	Scenario 2.2
Reward energy	-0.32	-0.35	-0.09	-0.14
Reward copper	-0.01	-0.01	-0.01	-0.01
Reward aluminum	-0.02	-0.02	-0.02	-0.02
Reward iron scrap	-0.01	-0.01	-0.01	-0.01
Reward HVCs	0.00	0.00	-0.29	-0.26
Transportation	0.00	0.00	0.01	0.01
Energy recovery	0.22	0.22	0.00	0.00
Landfilling	0.00	0.00	0.02	0.02
Copper recycling	0.01	0.01	0.01	0.01
Aluminum recycling	0.00	0.00	0.00	0.00
Fe-metal recycling	0.02	0.02	0.02	0.02
Steam cracking	0.00	0.00	0.12	0.11
Hydroprocessing	0.00	0.00	0.04	0.04
Pyrolysis	0.00	0.00	0.18	0.19
RDF production	0.07	0.07	0.07	0.07
APW collection	0.02	0.02	0.02	0.02
Sum	-0.02	-0.05	0.08	0.05

Figure C.3: Economic assessment of end-of-life paths and scenarios with costs above the x-axis and revenues beneath it. Assessment for 1 kg of input waste. Underlying data used to create this figure can be found in S2.

In addition to the environmental benefits, chemical recycling can potentially contribute to a circular economy closing the carbon cycle. However, mass and elemental balances for carbon recovery demonstrate that a high pyrolysis oil yield is advantageous when the goal is to contribute to a circular economy.

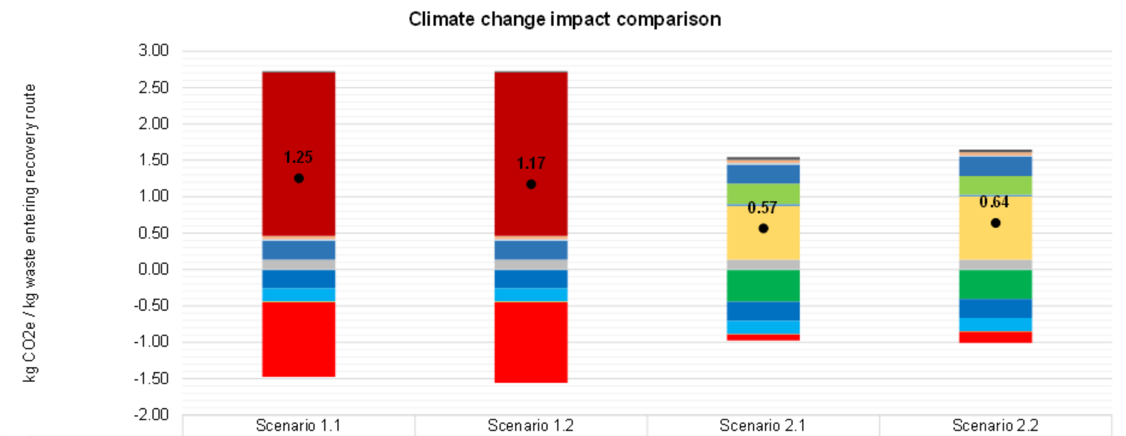
C.5.4 Sensitivity Analysis

The sensitivity analysis focuses on processing costs and the global warming potential and was conducted for both baseline scenarios. An overview of the analyses is provided in Figure C.7.

C.5.4.1 Energy Costs and Electricity Mix

Sensitivity analysis Sen1 and Sen2 analyze the effect of electricity prices and electricity revenues with an increase (Sen1) and a decrease of 10% (Sen2). Sen1 results in a 15% decrease in the total costs of energy recovery (Figure C.7) as revenues for recovered energy from incineration are higher than increased electricity costs at the RDF producer. The electricity cost increase of RDF production increases the total costs for chemical recycling by 9%. Sen2 leads to a 15% increase in the total costs of energy recovery and a 9% decrease in chemical recycling.

Sen3 assumes an electric power supply mix that includes more fossil energy sources by a 10% increased CO_2-factor of the electricity mix. Sen4 considers a more decarbonized electricity supply

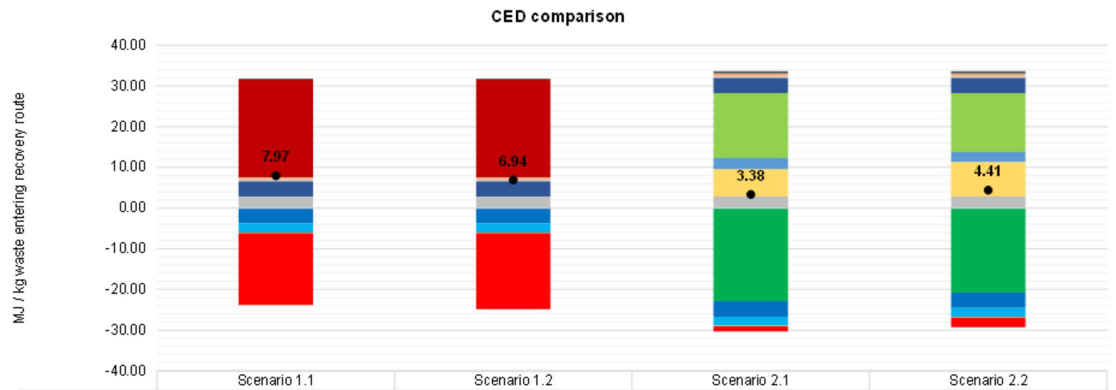


	Scenario 1.1	Scenario 1.2	Scenario 2.1	Scenario 2.2
Reward energy	-1.02	-1.11	-0.08	-0.16
Reward copper	-0.01	-0.01	-0.01	-0.01
Reward aluminum	-0.18	-0.18	-0.18	-0.18
Reward iron scrap	-0.26	-0.26	-0.26	-0.26
Reward HVCs	0.00	0.00	-0.44	-0.40
Transportation	0.02	0.02	0.04	0.04
Energy recovery	2.25	2.25	0.00	0.00
Landfilling	0.00	0.00	0.00	0.00
Copper recycling	0.04	0.04	0.04	0.04
Aluminum recycling	0.02	0.02	0.02	0.02
Fe-metal recycling	0.26	0.26	0.26	0.26
Steam cracking	0.00	0.00	0.28	0.26
Hydroprocessing	0.00	0.00	0.03	0.03
Pyrolysis	0.00	0.00	0.73	0.87
RDF production	0.14	0.14	0.14	0.14
APW collection	0.00	0.00	0.00	0.00
● Sum	1.25	1.17	0.57	0.64

Figure C.4: Comparison of end-of-life paths and scenarios regarding their net climate change impact. Assessment for 1 kg of input waste. Underlying data used to create this figure can be found in S2.

by a 10% decreased CO_2-factor. While the carbon dioxide emissions from APW incineration remain unaltered, financial rewards for substituting electricity change due to the CO_2 emission fees associated with conventional electricity generation. In Sen3, this causes a decrease in the total net cost by 13%. The total cost of chemical recycling is not impacted, as only heat and no electricity is substituted. Higher emissions associated with the electricity used for RDF production lead to a 3% higher climate change impact on energy recovery and a 2% higher climate change impact for chemical recycling compared to their respective baselines. Sen4 results in a 13% increase in the costs of energy recovery, while the costs for chemical recycling do not change. The climate change impact of energy recovery decreases by 3%, while the climate change impact for chemical recycling decreases by 2%.

Sen5 and Sen6 analyze the influence of a 10% increase and decrease in the CO_2 emission price. A 10% increase (Sen5) causes a 31% increase in energy recovery costs since incineration costs increase more than the rewards for the recovered energy. For chemical recycling, a cost increase for CO_2 emissions from steam cracking is compensated by increasing rewards for recovered energy resulting in a 2% increase in total processing costs. The variations are symmetrical and do not influence the climate change impact.



	Scenario 1.1	Scenario 1.2	Scenario 2.1	Scenario 2.2
Reward energy	-17.68	-18.71	-1.23	-2.31
Reward copper	-0.14	-0.14	-0.14	-0.14
Reward aluminum	-2.22	-2.22	-2.22	-2.22
Reward iron scrap	-3.78	-3.78	-3.78	-3.78
Reward HVCs	0.00	0.00	-22.93	-20.81
Transportation	0.30	0.30	0.62	0.62
Energy recovery	23.93	23.93	0.00	0.00
Landfilling	0.00	0.00	0.04	0.04
Copper recycling	0.74	0.74	0.74	0.74
Aluminum recycling	0.25	0.25	0.25	0.25
Fe-metal recycling	3.78	3.78	3.78	3.78
Steam cracking	0.00	0.00	15.98	14.51
Hydroprocessing	0.00	0.00	2.71	2.44
Pyrolysis	0.00	0.00	6.77	8.51
RDF production	2.80	2.80	2.80	2.80
APW collection	0.00	0.00	0.00	0.00
Sum	7.97	6.94	3.38	4.41

Figure C.5: Comparison of considered end-of-life paths and scenarios regarding their net CED impact [MJ/kg input]. Assessment for 1 kg of input waste. Underlying data used to create this figure can be found in S2.

C.5.4.2 Chemical Recycling

Sen7 and Sen8 estimate the effects of changing the energy demand of the pyrolysis process that may occur when it is scaled up to an industrial scale. The energy demand is increased by 10% (Sen7) and decreased by 10% (Sen8). The variations impact the processing costs of chemical recycling that increase or decrease by 10% as revenues from district heat vary. The climate change impact increases (Sen7) and decreases (Sen8) by 1% due to variations in the amount of heat substituted and associated burdens.

In Sen9 and Sen10, the avoided costs of the primary production of HVCs are varied. When conventional HVC production cost increase by 10% (Sen9), the chemical recycling cost decrease by 6% due to higher financial rewards for HVC. A 10% decrease in conventional HVC production costs (Sen10) leads to a 6% increase in chemical recycling costs.

C.5.5 Comparison with other Studies

There is no literature on APW pyrolysis. Therefore, the established data and results cannot be directly compared with other experimental data. The best comparison is the ASR pyrolysis on a similar experimental scale conducted by Notarnicola et al. (2017) and Galvagno et al. (2001). At similar temperatures (450-500 °C), ASR yields almost double the amount of char (ca. 50%) and considerably less pyrolysis oil (20-30%) in comparison to the pyrolysis of APW (31% char, 49% pyrolysis oil). The same is observed in laboratory-scale experiments (Joung et al., 2007; Santini

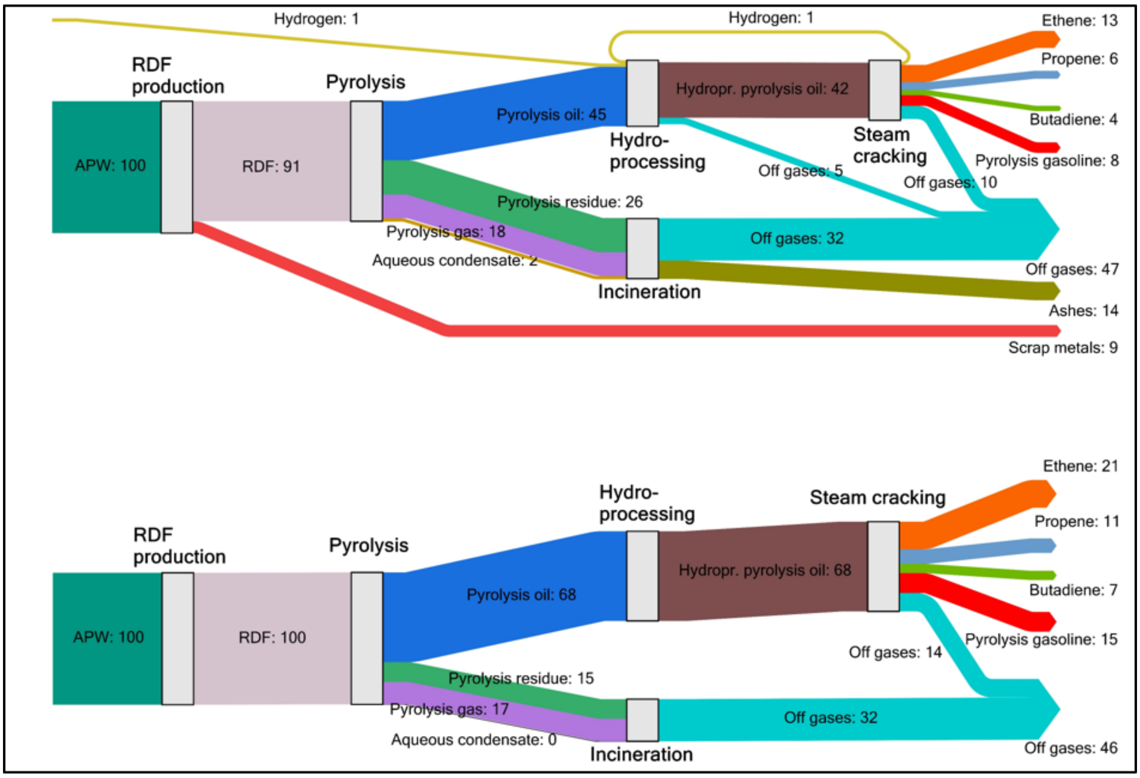
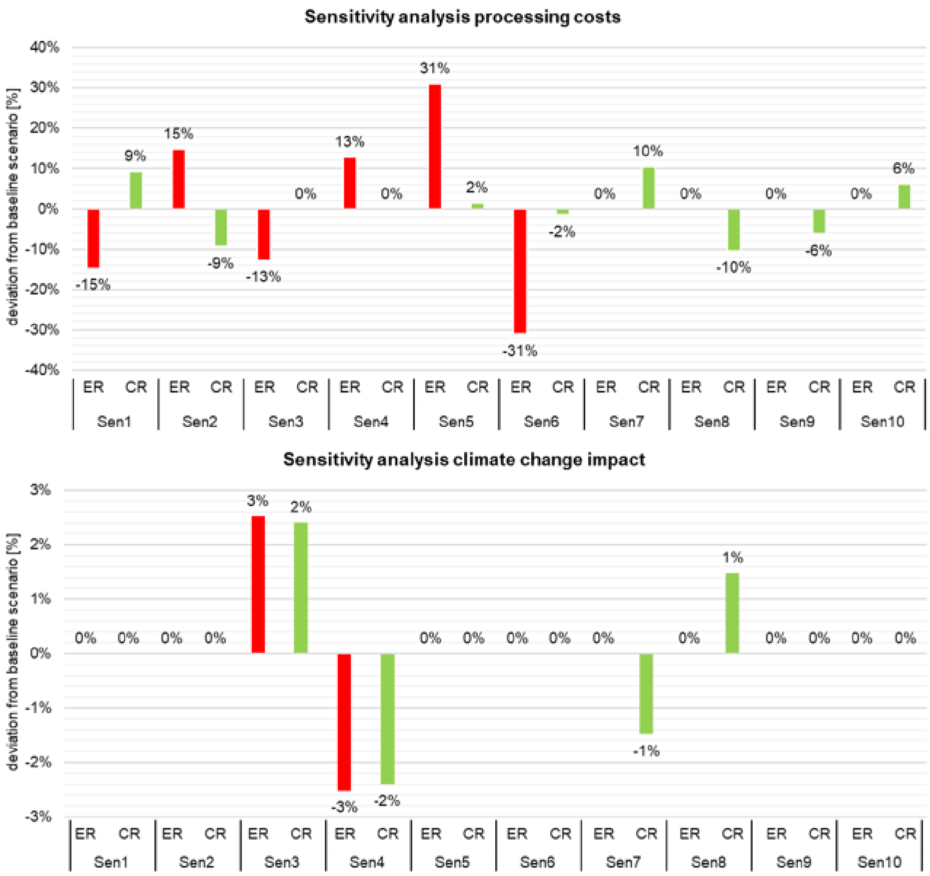


Figure C.6: Material flow (top) and carbon flow (bottom) of the chemical recycling (scenario 2.1). Numbers in percent (rounded) of APW input. Underlying data used to create this figure can be found in S2.

et al., 2012; Zolezzi et al., 2004). Despite variations in the reported composition of ASR and pyrolysis product distribution in different studies, it can still be concluded that APW generally yields higher amounts of pyrolysis oil due to a higher share of polymers in the feedstock material. However, ASR is available in much higher quantities.

Only a few studies assess recycling paths for automotive plastics regarding environmental or economic indicators. Ciacci et al. (2010) and Passarini et al. (2012) compare ASR treatment processes in an LCA. TThey both include chemical recycling or feedstock recycling. However, their results are incomparable since they assess an open-loop gasification scenario. The thermal treatment with energy recovery is designed as co-combustion with municipal solid waste (Ciacci et al., 2010; Passarini et al., 2012).T herefore, the results are also not comparable because of the different characteristics of the waste streams in MSWI plants.

However, (Ciacci et al., 2010) and (Passarini et al., 2012) indicate that advanced recycling technologies, such as chemical recycling, show better environmental performance than energy recovery. This is consistent with this study. Arena and Ardolino (2022) analyze the recovery of polymers from hard-to-handle plastic waste streams such as EoL vehicles. In their environmental assessment, they also include catalytic pyrolysis and energy recovery. Their assessment is based on a feedstock of single polymers and not an actual waste fraction. Therefore, the results are not entirely comparable, but they also indicate that catalytic pyrolysis performs better than energy recovery regarding the climate change impact. Cardamone et al. (2022) environmentally assess plastic recycling options for ASR with an extruder and pyrolysis combination. This waste-handling option is



Sensitivity	Area of Variation	Description
Sen1	Electricity	Increase of prices/revenues for electricity by 10 %
Sen2	Electricity	Decrease of prices/revenues for electricity by 10 %
Sen3	Electricity	Increase of the CO_2-factor of the electricity mix by 10 %
Sen4	Electricity	Decrease of the CO_2-factor of the electricity mix by 10 %
Sen5	Carbon price	Increase of carbon emissions price by 10 %
Sen6	Carbon price	Decrease of carbon emissions price by 10 %
Sen7	Chemical recycling	Increase of energy demand of pyrolysis by 10 %
Sen8	Chemical recycling	Decrease of the energy demand of pyrolysis by 10 %
Sen9	Chemical recycling	Increase of avoided costs for primary production of HVCs by 10 %
Sen10	Chemical recycling	Decrease of avoided costs for primary production of HVCs by 10 %

Figure C.7: Results of sensitivity analysis for energy recovery (ER) and chemical recycling (CR). Underlying data used to create this figure can be found in S2.

part of a recycling scheme that outperforms the current energy recovery option (Cardamone et al., 2022). They indicate that pyrolysis could be more environmentally beneficial than energy recovery, assuming a very optimistic pyrolysis oil yield. However, due to the chosen feedstock and process design of the chemical recycling, the results are only partially comparable to the results of this study. Other studies, e.g., Li et al. (2016) and Chen et al. (2019), conduct an LCA for automobile recycling but do not include the chemical recycling of ASR.

C.5.6 Limitations

This study has limitations and faces uncertainties due to (1) data and methodology, (2) model limits, and (3) assumptions. First, regarding (1) data and methodology: This study analyses a waste fraction with a comparatively low volume compared to ASR. However, the waste fraction of APW can become highly relevant if the dismantling of large plastic components becomes part of automobiles' EoL treatment processes. According to Wilts et al. (2016), dismantling could increase the amount of separated plastics available for recycling by a factor of six. Also, mixed plastic waste fractions separated from ASR by post-shredder-treatment processes have shown similar behavior in pyrolysis as the APW used in this study (Zeller et al., 2021).

The experiments demonstrate that the pyrolysis of engineering thermoplastics is technically feasible, and there is an environmentally beneficial alternative to their incineration. However, data for the subsequent hydroprocessing is unavailable yet, and very general assumptions had to be used. Also, this study does not consider other chemical recycling technologies besides pyrolysis. Mechanical recycling options and a combined mechanical and chemical recycling approach are also excluded. However, mechanical recycling processes for automotive plastics, like Sparenberg (2021) or the VW Sicon Process (Krinke et al., 2008), focus on separating standard thermoplastics and cannot handle engineering thermoplastics.

Second, this study has model limitations (2): The study assesses a defined waste stream of APW from workshop repair jobs with a specific composition to which the results are limited. The assessment is based on calculations for Germany. However, they are generic and can be transferred to countries with similar conditions. Nevertheless, this study does not consider dynamics such as changing waste compositions.

Third, assumptions (3) introduce uncertainties that are partly covered by scenarios and sensitivity analysis. However, assumptions regarding hydroprocessing (section 4.6) are not covered due to missing data. All processing facilities with direct CO_2 emissions are assumed to be covered within the EU emission trading system. No national emission trading systems are included in the assessment, indicating a possible extension. In reality, a mix of scenarios and sensitivities is likely, due to differing plant efficiencies, variable waste compositions, yields, and qualities of the pyrolysis products.

C.6 Conclusions and Outlook

The pyrolysis experiments demonstrate the technical feasibility of the pyrolysis of automotive plastic waste containing engineering thermoplastics. Based on the experimental results, energy recovery and chemical recycling of APW to HVCs via pyrolysis are assessed, compared, and analyzed in different scenarios regarding their climate change impact, CED, and processing costs. The results show that chemical recycling has lower net environmental impacts than energy recovery, while, under current market conditions, energy recovery performs better economically. Therefore, this study identifies a conflict between the economic and environmental objectives of EoL options for APW. The sensitivity analysis shows that the price and revenue associated with electricity and the carbon emission price significantly impact the assessment. Here, the assessed indicators for energy recovery correlate stronger with the varied parameters than for chemical recycling.

Chemical recycling also has the potential to keep carbon in the material cycle. The gained pyrolysis oil is a valuable petrochemical feedstock and can be used in different processes, e.g., to produce new primary plastics. Therefore, the chemical recycling of APW can contribute to a circular economy by closing the carbon and the automotive plastic loop. Additional research is needed to provide experimentally validated data for the hydroprocessing of the produced pyrolysis oil. Further research should also address the potential of mechanical recycling processes for engineering thermoplastics, alternative feedstock utilization paths of pyrolysis oil with lower quality demands than steam cracking, and scenarios when technologies are combined. This can contribute to designing and optimizing recycling systems for complex mixed plastic wastes.

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D Techno-Economic Assessment of Pyrolysis Plants for Automotive Plastic Waste

Abstract[1]

Strengthening a circular economy for plastics can reduce the need for fossil resources and limit environmental impacts, e.g., greenhouse gas emissions. Chemical recycling via pyrolysis is an option for recycling plastic waste unsuitable for mechanical recycling, like mixed automotive plastic waste (APW). However, pyrolysis has to be economically feasible or government-sponsored to be realized. Therefore, this study conducts a techno-economic assessment of pyrolysis plants with capacities between 3,750 and 37,500 Mg input/year for APW. The study uses experimental data for the named waste type and screw-reactor technology. The resulting minimum sales prices for pyrolysis oil indicate that plants with a capacity exceeding 33,750 Mg/year can economically operate in Germany under current framework conditions. The results are mainly sensitive to full load hours and capital investment. The study shows that technically suitable and scalable pyrolysis technology can enable a circular economy for mixed plastic wastes.

D.1 Introduction

Global plastic production reached 367 million metric tons in 2020 (PlasticsEurope, 2022), and forecasts predict its further increase (IEA, 2018). Production primarily relies on fossil carbon sources (Ellen MacArthur Foundation, 2016).. Concepts of the circular economy, e.g., recycling, could reduce the need for fossil resources, reduce the energy demand for plastic production, and limit corresponding greenhouse gas emissions (Agora Industry, 2022; IPCC, 2022).

In Europe, however, the current end-of-life (EoL) management of post-consumer plastic waste is dominated by energy recovery (42%), while 35% of the waste is recycled mechanically and 23% is landfilled (PlasticsEurope, 2022). Mechanical plastic recycling technology is particularly suitable for homogenous and contaminant-free plastic waste streams (Punkkinen et al., 2017), while

[1] This chapter includes the preprint version of the article "Techno-economic assessment of pyrolysis plants for automotive plastic waste" by Malte Hennig, Rebekka Volk, Dieter Stapf, Frank Schultmann, and myself. The paper was submitted to a scientific journal. The supporting information will be published with the journal article.

mechanical degradation, contamination, and additives impact secondary plastics' material quality, resulting in downcycling (Pivnenko et al., 2015). Alternative chemical recycling technologies like pyrolysis are being explored to address these challenges. In pyrolysis, plastics are decomposed at high temperatures and in an inert atmosphere resulting in solid, liquid, and gaseous products (Dogu et al., 2021). Especially liquid products have the potential to replace fossil hydrocarbon feedstock in petrochemistry to produce high-value chemicals closing the carbon cycle (Lechleitner et al., 2020).

Pyrolysis is complementary to mechanical recycling and is designed for recycling waste streams that previously could not be recycled. Multiple studies highlight the environmental benefits of pyrolysis compared to energy recovery and landfilling of plastic waste unsuitable for mechanical recycling (Stallkamp, Hennig, Volk, Richter, et al., 2023; Jeswani et al., 2021; Meys et al., 2020; Schwarz et al., 2021). Combining different recycling technologies can result in high recycling rates and low environmental impacts (Volk et al., 2021). However, chemical recycling via pyrolysis must be economically feasible to realize these advantages.

Few studies assess the economics of plastic waste recycling via pyrolysis for mixed plastic waste (MPW). Westerhout et al. (1998) evaluate different reactor types in a concept screening to identify the reactor design with the highest financial returns. Sahu et al. (2014) simulate a catalytic fluidized bed reactor to analyze its economic performance in producing fuel from MPW. Fivga and Dimitriou (2018) assess the economics of a fluidized bed reactor using an ASPEN simulation model. Jiang et al. (2020) assess the economics of a molten salt pyrolysis plant with MPW feedstock employing an ASPEN simulation model with an internal rate of return (IRR) of 33%. Larrain et al. (2020) determined that a plant capacity between 70,000 and 115,000 Mg input/year is required to economically operate a pyrolysis plant in Belgium, depending on the product mix. Riedewald et al. (2021) assess the economic performance of a pyrolysis plant employing the PlastPyro process handling MPW in Belgium. They outline that a plant with 40,000 Mg input/year capacity is economically viable, with higher throughputs increasing the financial returns.

Even though all studies assess the handling of MPW, they do not provide a detailed overview of the waste composition, making comparing the results difficult. Comparability is also hampered due to the different pyrolysis reactor types assessed. In addition, most studies focus on wastes with a high share of polyolefins. More challenging plastic waste streams like APW containing considerable amounts of engineering plastics, e.g., highly functionalized thermoplastics, elastomers, and thermosets, are not considered in the literature yet. Therefore, this study conducts a techno-economic assessment (TEA) for a pyrolysis plant handling APW from workshops in Germany, a challenging waste stream currently used for energy recovery (Cossu and Lai, 2015; Mehlhart et al., 2018). The assessed plant employs a twin-screw reactor as a scale-up from a single-screw reactor design described by Zeller et al. (2021) and Tomasi Morgano et al. (2015). The conducted TEA is based on mass and energy balances from pilot-scale pyrolysis experiments published Stallkamp, Hennig, Volk, Richter, et al. (2023) that were carried out with an actual waste fraction of APW collected from car workshops.

Thus, in contrast to the studies mentioned, this study examines the cost of pyrolysis of a technically challenging waste stream that cannot be mechanically recycled. A transparent TEA is performed, and the minimum selling price of the products and the minimum capacity for a pyrolysis plant at which economic operation is possible are determined

D.2 Materials and Methods

Following Van Dael et al. (2015)., the TEA analyzes feedstock supply and product sales prices (section D.2.1). It establishes a process flow diagram of the plant design with associated mass and energy balances (section D.2.2). Based on this, the general approach of the economic assessment is described (section D.2.2).

D.2.1 Feedstock Supply and Product Sales Prices

D.2.1.1 Automotive plastic Waste in Germany

Within this study, we focus on APW in Germany, as Germany is assumed to be the location of the pyrolysis plant. APW and automotive shredder residues (ASR) are primarily landfilled or used for energy recovery (Cossu and Lai, 2015; Mehlhart et al., 2018). Therefore, this feedstock does not compete with mechanical recycling but with energy recovery. Energy recovery is associated with lower costs than chemical recycling and is consequently economically preferable (Stallkamp, Hennig, Volk, Richter, et al., 2023). However, it is assumed that upcoming legislation demands the recycling of APW and, therefore, will resolve this competition.

In Germany, the waste treatment of APW from workshops starts with its collection and transport to refuse-derived fuel (RDF) production. Metals are separated and sent to established recycling processes, while all other materials are processed to an RDF (Stallkamp, Hennig, Volk, Richter, et al., 2023). Stallkamp, Hennig, Volk, Richter, et al. (2023) provide a characterization of an RDF from APW of a premium car manufacturer in Germany that is used here.

A conservative estimate of the waste volume of APW can be based on dismantling plastic components from EoL vehicles. UBA (2022), states that around 3 kg of plastic parts are nowadays dismantled from an EoL vehicle. Germany had 461,266 EoL vehicles in 2019 (UBA, 2022), resulting in an APW volume of around 1,380 Mg. This volume is very low compared to ASR (345,000 Mg (UBA, 2022)) or plastic packaging waste (3,180,000 Mg (UBA, 2022b)). However, this waste estimation does not include APW from workshop repair jobs. Also, the waste amount can increase in the future if the dismantling of large plastic components becomes part of automobiles' EoL treatment processes. According to Wilts et al. (2016), dismantling could increase the amount of separated plastics available for recycling by a factor of six, resulting in 8,280 Mg for 2019. Also, mixed plastic waste fractions separated from ASR by post-shredder-treatment processes have shown similar behavior in pyrolysis (Zeller et al., 2021) and are a potential feedstock. Based on the waste volume of 2019, the mid-plastic fraction of the ARN process (ARN, 2016) could result in an additional feedstock of 1,622 Mg (ARN, 2016), resulting in a total feedstock of around 10,000 Mg for 2019 in Germany.

As the pyrolysis plant handles waste, a gate fee for waste handling is assumed, resulting in a negative feedstock price. According to Bilitewski et al. (2018), the price for handling and co-incineration of waste-derived fuels in power plants is 15 €/Mg.

D.2.1.2 Pyrolysis Oil and Heat

Pyrolysis oil is the desired main product of the pyrolysis process (Zeller et al., 2021). Multiple studies highlight its economic value (Sharuddin et al., 2016; Larrain et al., 2020; Punkkinen et al., 2017). However, the price of pyrolysis oil is hard to predict as different feedstocks and process parameters result in different oil qualities (Kusenberg, Eschenbacher, et al., 2022). Depending on the oil quality, additional purification steps are needed to provide a valuable feedstock for the petrochemical industry (Kusenberg, Eschenbacher, et al., 2022), which may lower its market value. At the same time, the demand for pyrolysis oils from plastic waste is high, and supply is limited as plastic waste pyrolysis is not yet industrialized at a large scale (Tullo, 2022). This may lead to higher realizable prices. As a conservative compromise, it is assumed that the unpurified pyrolysis oil is sold at the price of U.S. Residual Fuel Oil, accounting for potential quality issues. This assumption is consistent with (Riedewald et al., 2021). In 2021, the average U.S. Residual Fuel Oil price was around 462 €/Mg, with increasing prices at the end of the year (EIA, 2022). This price increase continued in 2022 (EIA, 2022).

Pyrolysis gas can be used to generate electricity and district heating by utilization in a combined heat and power unit (CHPU). Depending on the amount of electricity generated, the energy demand of the pyrolysis plant can be covered. Excess electrical energy can be fed to the grid (0.10 €/kWh; (Fraunhofer ISE, 2023)). If the electricity demand of the pyrolysis plant surpasses the amount generated by the CHPU, electricity can also be taken from the grid (0.21 €/kWh; (Bundesnetzagentur, 2022)). A district heating network can supply excess heat recovered from the CHPU. In 2021, the average price for district heating in Germany was 81 €/MWh (AGFW, 2022). The by-products aqueous condensate and solid residues are considered waste that must be disposed of. However, there might be future use cases for solid residues. Therefore, two scenarios are calculated: solid residual disposal by co-incineration (1) in waste incineration associated with costs, and (2) industrial co-incineration, assuming the generation of a profit from selling the solid residue as fuel. The aqueous condensate is co-incinerated in a hazardous waste incineration plant in every case.

D.2.2 Pyrolysis Process

The reactor design assessed in this study was derived from the single-screw plastic pyrolysis reactor described by Zeller et al. (2021). The product recovery section was adapted based on the product recovery of the biomass-to-liquid (BtL) pyrolysis plant described by Trippe et al. (2010). Using a similar reactor concept and reaction conditions in the assessment as used in pilot-scale APW pyrolysis experiments assures that the mass and energy balance reported by Stallkamp, Hennig, Volk, Richter, et al. (2023) can be transferred to the process layout assessed here (Figure 1). However, the pyrolysis plant has not yet been set up and used to validate pilot scale experiments.

In a delivery and pretreatment module of the plant, the feedstock is pretreated and stored to ensure the supply of the plant (Trippe et al., 2010). For the pyrolysis, feedstock and quartz sand are separately fed into a twin-screw reactor (Trippe et al., 2010). The sand is used for improved heat transfer within the electrically heated pyrolysis reactor, providing the temperature of 450°C required for pyrolysis (Zeller et al., 2021). The feedstock decomposes into pyrolysis vapors that are extracted from the reactor and fed to a condensation module for product recovery (Zeller et al., 2021). Sand and pyrolysis residue are discharged from the reactor and separated in a vibration sieve (Trippe et al., 2010). The sand is then fed back to the reactor while the pyrolysis residues are discharged.

The pyrolysis vapors are cleaned in a filter before entering a quench condenser (Trippe et al., 2010). The recovered pyrolysis oil is collected in a tank. Non-condensed gases and vapors are cooled in a gas cooler and purified in a gas scrubber (Trippe et al., 2010). Lighter condensing fractions and water are separated in the process and collected in tanks. Parts of the light condensates are used in the quench condenser and the gas scrubber to recover the pyrolysis condensates (Trippe et al., 2010). The aqueous condensate is collected for co-incineration. The remaining incondensable pyrolysis gas is incinerated in a CHPU to generate electricity and heat. The generated heat is sold, e.g., to a district heating network. The generated electricity is used to provide the electrical energy demand of the plant. Surplus electricity exceeding the plant's demand is sold to the grid operator. If electricity production is insufficient, additional electricity is sourced from the grid.

D.2.2.1 Mass Balances and Process Flows

The underlying mass and process flow for the pyrolysis of pretreated APW stem from Stallkamp, Hennig, Volk, Richter, et al. (2023). The main product of the pyrolysis process is the pyrolysis oil (50%), with the potential to replace naphtha as feedstock in the petrochemistry industry. However, additional oil purification steps must be performed (Kusenberg, Eschenbacher, et al., 2022; Stallkamp, Hennig, Volk, Richter, et al., 2023). The pyrolysis gas (20%) is incinerated to generate electricity and heat. The remaining pyrolysis residues (28%) must either be disposed of (scenario 1) or can be used as fuel in industrial co-combustion (scenario 2). The aqueous condensate (2%) must be disposed of.

D.2.2.2 Energy Balances

Current experiments with APW (Hennig et al., 2022; Stallkamp, Hennig, Volk, Richter, et al., 2023) and comparable waste fractions from treated ASR (Zeller et al., 2021) were performed on a pilot-scale reactor. The energy demand for electric heating of the pyrolysis reactor was determined at 6% of the net calorific value (Stallkamp, Hennig, Volk, Richter, et al., 2023). This results in an electricity demand of 1,578 MJ/Mg input for the pyrolysis plant. Electricity demands for, e.g.,

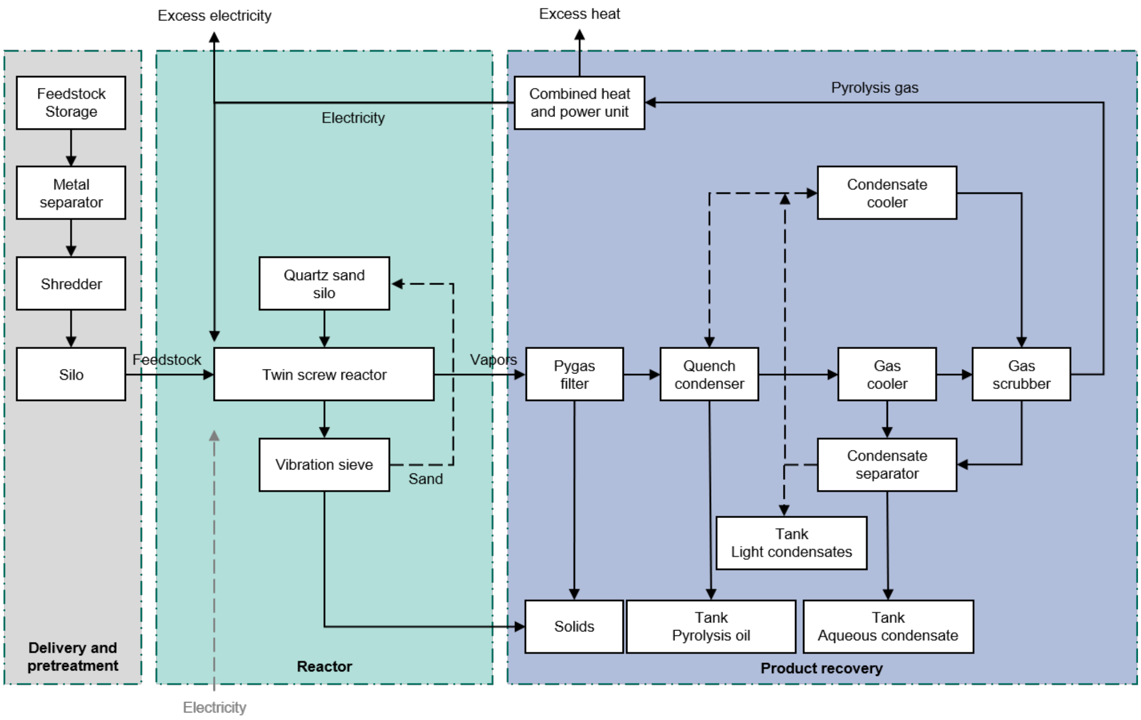

Figure D.1: Pyrolysis plant design with three modules: (1) delivery and pretreatment, (2) reactor, and (3) product recovery. The feedstock is delivered and pretreated, the reactors run with a quartz sand heating cycle, and products are recovered in a condensation system. Plant design and figure are based on Trippe et al. (2010).

pumps and electric drives, are neglected, but a sensitivity analysis analyzes the effects of a higher electricity demand.

D.2.3 Economic Assessment

D.2.3.1 Equipment and Infrastructure Investment

The E&I investment for the pyrolysis plant is calculated based on the plant design and a list of equipment needed, following the capacity estimate approach for all standard mechanical and process engineering components (Humphreys, 2004). The components' investment is scaled based on the capacity and component-specific cost-capacity factors (Equation D.1).

$$I_2 = I_1(\frac{C_2}{C_1})^x \tag{D.1}$$

However, reactors like the twin-screw reactor have mechanical limitations that do not allow limitless scaling. Due to the reactor filling level and screw design, a maximum reactor capacity of 0.5 Mg/h is assumed. This capacity corresponds to the reactor installed in the BtL plant (IKFT, 2018) and a current commercial pyrolysis plant construction (KIT Technology, 2021). For higher throughputs, it is assumed that additional reactors must be operated following a numbering-up approach. Scaling single reactors following the capacity estimate approach (Equation D.1) is

Table D.1: Overview of the nomenclature of the investment scaling.

I_2	Investment for scaled capacity 2
I_1	Investment for baseline capacity 1
C_2	Scaled capacity 2
C_1	Baseline capacity 1
x	component-specific cost-capacity factor

possible. It is assumed that a maximum of four reactors are connected to one product recovery unit[2] to reduce the complexity of the plant design.

All component prices were adapted to 2021, accounting for inflation using the ProcessNet Chemical Plant Index Germany (PCD) (DECHEMA and VDI, 2022). Based on Dysert et al. (2016) and Towler and Sinnott (2012) and the chosen capacity estimate approach, the conducted study is classified as a project screening or feasibility study (class 4). For this level, the classification matrix for estimating costs in the process industry assumes an accuracy interval for the investment between -30% and +50% (Dysert et al., 2016; Towler and Sinnott, 2012).

D.2.3.2 CAPEX

The CAPEX is based on the required E&I investment and calculated using an equipment factor method. FollowingPeters et al. (2003), the CAPEX is divided and calculated by applying defined percentages of the E&I investment (Table D.2). To calculate the CAPEX, we assume a brownfield setting to enable the integration of product streams into existing production or district heating networks.

D.2.3.3 OPEX

The OPEX is separated into fixed and variable OPEX. The fixed OPEX is independent of the amount of feedstock handled and is based on the size and capacity of the plant. They include personnel costs, maintenance, yearly insurance, and general plant overhead calculated based on percentages of the E&I investment (Table D.2) (Larrain et al., 2020). Personnel costs depend on the wages paid and are not influenced by the investment. The plant's capacity determines the number of workers needed. The fixed OPEX also includes depreciation and costs for financing the plant.

The variable OPEX depends on the amount of feedstock handled and is calculated based on the process flows and mass and energy balances. Here, material and energy streams are associated with costs, and the OPEX can be calculated by multiplying these cost rates (Table D.2) with the actual streams within the plant.

[2] excluding the CHP unit

D.2.3.4 Scale-Up

Ten different capacity classes are assessed, starting at an input capacity of 3,750 Mg/year (class 1), increasing by 3,750 Mg input/year, and resulting in a maximum input capacity of 37,500 Mg/year (class 10). The scaling of the plant corresponds to the number of reactor modules operated at full load with 7,500 hours of annual operation.

The plant concept is scaled up based on the baseline cost estimation for a class 1 plant. Here, the numbering-up approach for the reactor and the product recovery module is combined with gradual scaling and specific cost-capacity factors for individual equipment (sectionD.2.3.1)). So, an additional reactor module is added for each capacity class, and another product recovery module is added with every fifth reactor. Individual equipment and components are scaled based on Equation D.1.

D.3 Techno-Economic Assessment

The TEA is conducted following the steps outlined in section D.2.3. All general assumptions for the TEA are summarized in Table D.2. The supporting information (SI) provides additional parameters, the E&I investment, CAPEX, and OPEX for the pyrolysis plant and all assessed capacities (Table S-1 to Table S-5, SI).

D.3.1 Equipment and Infrastructure Investment and CAPEX

The CAPEX of the pyrolysis plant is calculated based on investment data (Larrain et al., 2020; Stapf et al., 2019; Trippe, 2013), which is adapted to the assessed capacity and accounted for inflation (sectionD.2.3). The investment (Table S-2, SI) and CAPEX (Table S-3, SI) of all capacity classes are provided in the SI. Here, the E&I investment for class1 is exemplarily presented.

The investment in the land needed results in 276,000 €. The delivery and pretreatment module of the plant includes crane systems for unloading, conveyor belts, metal separators, and a shredder. In total, it is associated with an estimated investment of 518,000 €. In class 1, one twin-screw reactor is operated. Vibration sieves, heat cycle components, and the reactor add up to 569,000 €. The product recovery module results in an investment of 879,000 €. This includes cleaning steps for the pyrolysis gas and vapors, condensation steps, the separation of condensates, and the CHP unit. In total, the E&I investment results in 2,241,000 €.

CAPEX is calculated based on this E&I investment using the investment percentages in Table 1 (Peters et al., 2003). For class 1, the total CAPEX result in 11,723,000 € (Table S-3). Total CAPEX and the composition of the investment for different plant sizes are presented in section D.3.3.

Table D.2: Assumptions and parameters for the techno-economic assessment of the pyrolysis plant.

Technical parameters	
Yield pyrolysis oil	50%
Pyrolysis gas	20%
Pyrolysis residues	30%
Energy demand of pyrolysis plant	15% of the feedstock's calorific value
Calorific value of feedstock	26 MJ/kg
Calorific value of pyrolysis oil	37 MJ/kg
Calorific value of pyrolysis gas	19 MJ/kg
Calorific value of pyrolysis residues	12 MJ/kg
Operational parameters	
Scale 1	3,750 Mg input/year
Scale 10	37,500 Mg input/year
Operating time	7500 h/year
Shifts	3 per day
Financial parameters	
Reference year	2021
Method of financing	Bank loan
Calculation interest rate	3%
Operating life	20 years
CAPEX (specified as % of E&I investment)	
Equipment installation	39%
Instrumentation and controls (installed)	26%
Piping (installed)	31%
Electrical system (installed)	10%
Buildings (including services)	29%
Yard improvements	12%
Service facilities (installed)	55%
Engineering and supervision	32%
Construction expenses	34%
Project Management	20%
Legal expenses	4%
Contractor's fee	19%
Contingency	37%
Working capital	75%
OPEX	
Labor expenses	Compare SI, Table S-4, and S-5
Maintenance	4% of EI investment
Yearly insurance	2% of EI investment
General plant overhead	65% of labor and maintenance expenses
Ash disposal	110 €/Mg
CO_2 emission fees	53 €/Mg
Feedstock costs	-15 €/Mg

D.3.2 OPEX

The OPEX for all capacity classes and both scenarios are summarized in the SI (Table S-4, S-5). Here, the OPEX for class 1 is exemplarily presented. Fixed OPEX depend on the capacity and the total investment for the pyrolysis plant. Based on the parameters shown in Table 1, maintenance results in 90,000 €, yearly insurance amounts to 34,000 €, and general plant overhead results in 393,000 €. Personnel costs are calculated based on the staffing of the plant and associated wages (Table S-6, S-7) and result in 515,000 €.

Annualization is used to calculate the cost of capital (Smith, 2016). It aims to transform non-periodic and periodic payments of a period into regular periodic payments (annuity). This annuity reflects the interest and repayment of capital. The annuity of the investment-linked payments corresponds to the cost of capital, which is the product of the fixed capital investment and the annuity factor (Equation D.2). The direct and indirect costs of the investment add up to the fixed capital investment (Table S-3, SI). The annuity factor depends on the lifetime of the plant and the interest rate (Equation D.3).

$$C_{capital} = \text{Fixed capital investment} * f_A \tag{D.2}$$

$$f_A = \frac{(1+i)^n * i}{(1+i)^n - 1} \tag{D.3}$$

Table D.3: Overview of the nomenclature of the annualization of the investment.

$C_{capital}$	Cost of capital
f_A	Annuity factor
i	interest rate
n	lifetime of the plant

This study uses an interest rate of 3% (Götz et al., 2022). The assumed lifetime of the plant is 20 years (German Federal Ministry of Finance, 1995). This results in an annual annuity factor of 0.067 and cost of capital of 675,000 €/year for the total investment. Additional capital costs for the working capital result from the multiplication with the calculation interest rate and sum up to 50,000 €/year.

The variable OPEX includes CO_2 net emission fees, feedstock costs, electricity costs, disposal costs for aqueous condensate, and revenues for generated heat. Additionally, in the baseline scenario (scenario 1), the OPEX includes the disposal costs for solid residues. In scenario 2, the payments from selling the solid residues as fuel are considered in the OPEX instead of disposal costs. Tables S-4 and S-5 (SI) provide an overview of the costs and revenues associated with each item. The total OPEX of the baseline scenario (scenario 1) results in 1,821,000 € if full capacity is utilized, corresponding to 486 €/Mg feedstock input.

D.3.3 Scale-up

The assessment results of all ten capacity classes are summarized in Figure D.2. With the capacity increase, the total CAPEX increases disproportionately due to the economies of scale in process engineering (Turton et al., 2008). Comparing the total CAPEX of class1 with the total CAPEX of class10, a 10-fold increase in capacity results roughly in a 5.3-fold increase in CAPEX. However, the numbering up approach slightly increases the investment shares of the reactor and product recovery unit.

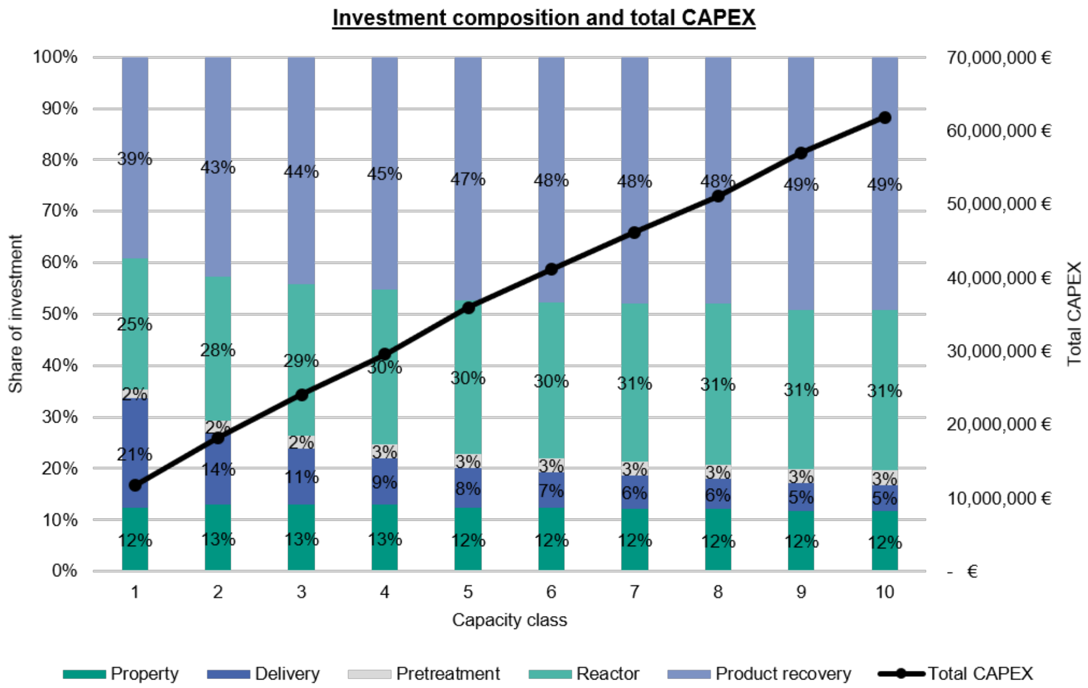

Figure D.2: Investment composition and total CAPEX of different capacity classes of a pyrolysis plant for APW.

Figure D.3 shows a decreasing trend in the OPEX with increasing plant capacity for both scenarios. The increase in the OPEX from class 5 to class 6 is due to additional personnel needed with the increased plant capacity (Table S-6, SI). The OPEX is dominated by capital payments and personnel costs, while with increasing capacity, the impact of personnel costs decreases. In scenario 1, the share of disposal costs for the solid residues in the total OPEX increases with increasing capacity, while in scenario 2, the percentage of revenues generated with the solids is consistent. The costs of handling the aqueous condensates are low and have no significant impact. Increasing revenues from feedstock gate fees and generated heat reduce the OPEX, and their reducing effect increases with the plant capacity.

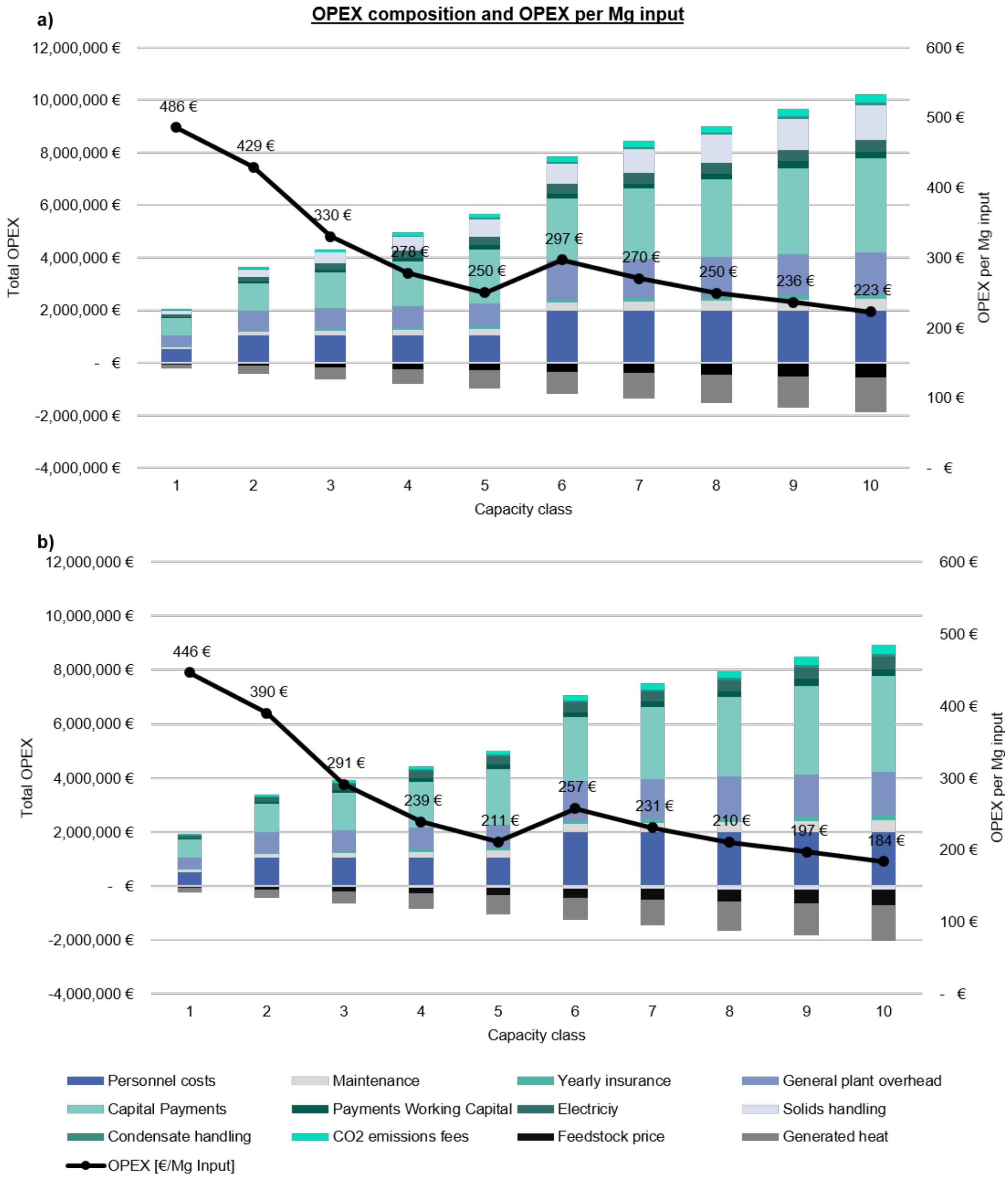

Figure D.3: OPEX composition and total OPEX per Mg input for different scales of a pyrolysis plant for APW in scenario 1 (a) and scenario 2 (b).

D.3.4 Minimum Sales Price

The minimum sales price for pyrolysis oil (Figure D.4) is derived from the production costs[3] and a target margin of 15% resulting from deploying a new technology and a new product in an existing market (Peters et al., 2003; Riedewald et al., 2021). The minimum sales price is compared to the average U.S. Residual Fuel Oil price of 462 €/Mg (EIA, 2022) in 2021. With increasing plant

[3] Allocation of the OPEX to the produced quantity of the desired product (pyrolysis oil).

capacity and production, the production costs and minimum sales prices decrease as relative costs fall due to economies of scale.

In scenario1, the minimum sales price never undercuts the reference price of U.S. Residual Fuel Oil (grey line). However, falling minimum sales prices indicate that higher plant capacities can (almost) economically compete with the reference product. The production costs are below the reference price of U.S. Residual Fuel Oil in class10. A profit can, therefore, already be made if a part of the 15% margin is sacrificed.

Looking at scenario2, the production costs for the pyrolysis oil are already below the reference price, starting at class8. With a target margin of 15%, the minimum sales price is below the reference price, starting at class9. Therefore, from an economic perspective, it should be aimed at selling the solid residues as fuel.

Both scenarios show that higher plant capacities reduce the minimum sales price, enabling a more economical operation. Also, waste should be prevented, and value streams should be generated.

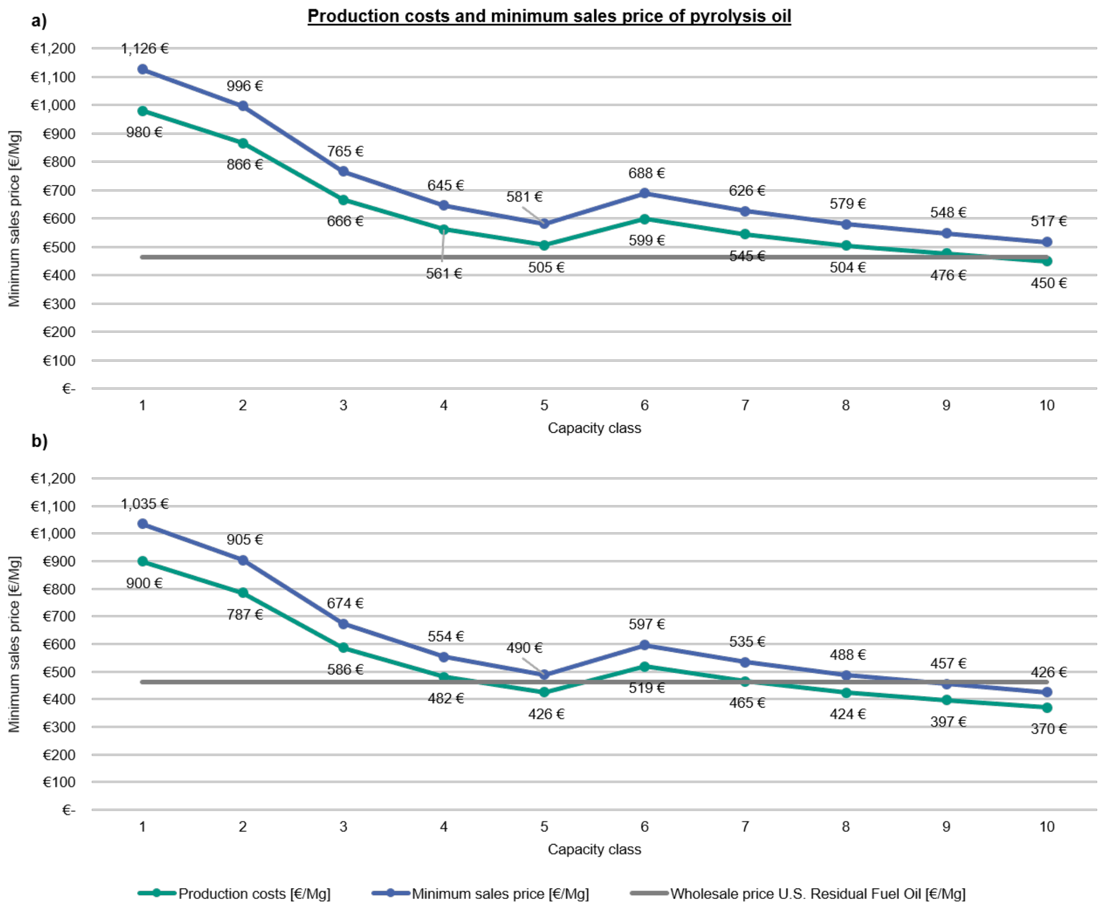

Figure D.4: Production costs and minimum sales price of pyrolysis oil for different capacity classes of a pyrolysis plant for APW in scenario 1 (a) and scenario 2 (b).

D.3.5 Sensitivity Analysis

A sensitivity analysis is conducted to identify the impact of single parameters on the assessment results. Parameters are varied individually in a range of ±25%, while all other calculation parameters are kept constant. The larger the slope of a curve in Figure D.5, the higher the influence of the associated parameter on the minimum sales price of the pyrolysis oil.

The minimum sales price of pyrolysis oil varies most with the full load hours of the plant. A 25% decrease in utilization results in a 26% increase in the minimum sales price. The calculated investment results in the second-largest variation. A 25% reduction in the investment is reflected in a 14% decrease in the minimum sales price. Reducing the plant's electricity demand by 25% results in a 12% decrease, while the same reduction of the general plant overhead causes a decrease of 5%. A 25% decrease in maintenance results in a reduction of 2%, while reducing the electricity price results in a decline of 1%. Reducing the CO_2 emission fees results in a 1% decrease while lowering the feedstock price (gate fee) increases the price by 2%. The heat price increases the minimum sales price by 2% when decreased by 25%.

All parameter variations except the plant's full load hours and electricity demand show a symmetrical behavior. The full load hours are only reduced but not extended beyond the maximal capacity of the plant. The plant's electricity demand results in either excess electricity that can be sold to the grid or the need to buy additional electricity. An increase in the electricity demand by 25% results in extra electricity that must be sourced, resulting in a rise in the minimum sales price of 15%. Therefore, the impact is not symmetrical to a 25% decrease in the electricity demand. In all other cases, a corresponding positive parameter variation of 25% influences the minimum selling price in the opposing direction to its 25% decrease.

D.4 Discussion

The TEA results indicate an economical operation of the assessed pyrolysis plants with APW feedstock for capacities greater than 33,750 Mg input/year if the solid residues can be sold as fuel. If the solid residues must be disposed of, no assessed plant capacity results in a minimum sales price below the price of the reference product. However, production costs are below the reference price; therefore, a profit can be generated if a margin below 15% is accepted.

In general, increasing plant capacities and throughputs reduce the minimum sales price and, therefore, can increase the financial return of the plant. This aligns with other economic assessments like (Larrain et al., 2020) and (Riedewald et al., 2021). However, current experiments run on pilot-scale reactors, and mass and energy balances may be affected by scale-up to a commercial scale of such a pyrolysis plant. Experiments on larger reactors and demonstration plants must be conducted to confirm mass and energy balances at industrial scale.

Capital payments and personnel costs dominate the OPEX. This dominance is due to the relatively low plant throughputs, limited by its technology and the amount of waste available at the site.

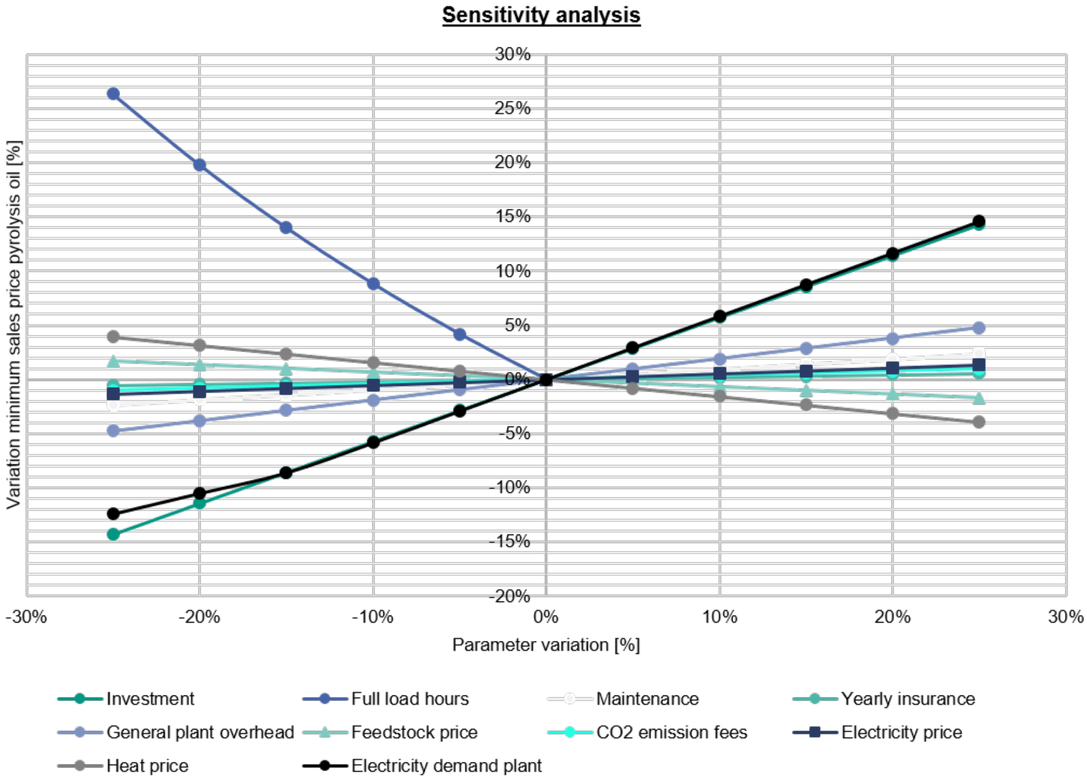

Figure D.5: Sensitivity analysis with a parameter variation of ±25% and its impact on the minimum sales price of pyrolysis oil as the main product of the pyrolysis of APW. The sensitivity analysis is conducted for capacity class 10 and the baseline scenario (scenario 1).

The sensitivity analysis highlights the impact of the full load hours, investment, and the plant's electricity demand on the minimum sales prices. Due to the project maturity of the assessed plant, a deviation from the calculated investment is possible even though, compared to other studies, the accuracy of the TEA is high. Other studies (Fivga and Dimitriou, 2018; Jiang et al., 2020; Sahu et al., 2014; Westerhout et al., 1998) primarily use the less precise factored cost estimate method. The conducted TEA combines the factored cost estimate method with a numbering-up approach for critical parts of the pyrolysis plant, like the reactor or the product recovery module. Therefore, a more accurate estimation is possible, as technical limitations in the capacity of components are considered.

A comparison with other TEA of pyrolysis plants for recycling plastics is hardly possible due to different reactor technologies, plant designs, and feedstock materials. Larrain et al. (2020) provide no details regarding their cost estimation of a waste plastic pyrolysis plant, as they base their assessment on classified data supplied by a waste treatment company. Moreover, the plant design and the employed technologies, especially the type of pyrolysis reactor, are not disclosed. Therefore, the structure and main parameters of the economic assessment are unclear, and the results cannot be reproduced. However, the trend of higher plant throughputs resulting in increasing financial returns can be confirmed. Riedewald et al. (2021) provide a comprehensive economic assessment of a pyrolysis plant based on the PlastPyro process for mixed plastic waste pyrolysis. They provide a detailed overview of their CAPEX, OPEX, and cost accuracy. However, they use both a different pyrolysis technology and feedstock. So, it is only comparable to a limited extent.

Finally, this TEA is conducted with the feedstock of APW from workshops. The results outline the economic operation of the pyrolysis plants starting at 33,750 Mg input/year capacity. Therefore, the capacity for an economic operation is greater than the expected total annual APW for Germany. However, the APW estimation is conservative and does not include APW from workshop repair jobs. There is also the potential that this waste fraction becomes highly relevant if the dismantling of plastic components becomes part of automobiles' EoL treatment processes. Also, mixed plastic waste fractions separated from ASR by post-shredder-treatment processes have shown similar behavior in pyrolysis as APW (Zeller et al., 2021). They might be a future feedstock of such plants making their operation more economically attractive. In addition, the assessed reactor technology has proven to be very robust regarding different feedstocks. Zeller et al. (2021) pyrolyzed a broad range of plastic waste samples in a screw-type pyrolysis reactor, including lightweight packaging sorting residues and residue from the mechanical recycling of electrical and electronic waste recycling.

D.5 Conclusion

Chemical recycling via pyrolysis can complement the current mechanical recycling of plastics and thus provide a recycling alternative to the incineration of these waste streams. Here, mixed engineering plastics from automobiles represent these demanding waste streams.

Several studies have shown the environmental advantages of pyrolysis over energy recovery and landfilling. However, pyrolysis also needs to be economically viable to succeed. The conducted TEA of pyrolysis plants employing twin-screw reactors shows that plant capacities starting at 33,750 Mg input/year can achieve minimum sales prices for pyrolysis oil that can economically compete with a reference product. It is assumed that solid pyrolysis residues can be sold as fuel. If the residues must be disposed of, the assessed plant capacities are not financially viable. However, production costs of higher capacity classes are below the reference price of residual fuel oil, indicating possible revenues. In addition, higher prices than for the reference price for pyrolysis oil might be achievable, improving the assessed business case.

In the present TEA, pyrolysis plant scale-up occurs through the numbering-up of the reactor and sizing-up of the product recovery system and auxiliary units to estimate the investment more accurately. The sensitivity analysis shows that the full load hours and the investment significantly influence the minimum selling price. Accordingly, as this study is a theoretical analysis, more accurate cost estimations must be conducted based on detailed project implementation and commissioning plans. This should include pyrolysis technologies with scale-up constraints different from the relatively limited screw-reactor technology.

The chosen reactor technology has proven robust with challenging plastic waste feedstocks. Mass and energy balances of the pyrolysis plant are based on pilot scale experiments with APW, a waste stream with currently relatively low volume in Germany. An increased dismantling of plastic components in the EoL treatment processes of automobiles can increase the amount of available waste. Additionally, the larger fraction of automotive shredder residues is similar to APW, so the pyrolysis plant could be used for other complex feedstocks. Moreover, for Europe, the needed waste feedstock for an economical operation of the pyrolysis plant might be reached. Economic viability ultimately depends on feedstock availability and full load hours.

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E Circular Economy for Plastics in Germany: Decision Support for Optimal Recycling Network Designs

Abstract[1]

Plastic production increases worldwide, and so does the amount of plastic waste. Plastic waste unsuitable for mechanical recycling is usually incinerated. Here, chemical plastic recycling offers an alternative for closing the plastic loop and decarbonizing plastic waste treatment. Therefore, this study optimizes waste treatment networks employing energy recovery and chemical recycling using pyrolysis. The waste treatment network for sorting residues from lightweight packaging and automotive plastic waste is optimized for both network costs and climate change impact. A multi-objective formulation using a goal programming approach optimizes both objectives simultaneously. Additionally, political steering strategies for aligning environmental and economic interests in the waste treatment sector are quantified and discussed. Extending the EU emission trading system to the waste management sector and introducing a CO_2 emission fee can reduce the network's climate change impact by up to 61%. Increasing recycling rates to meet EU recycling targets (55% for plastic packaging in 2030) can also support CO_2 emission reduction assuming legal certainty regarding the classification of chemical recycling as recycling technology and crediting the such treated waste to the recycling rates. With the EU recycling targets for plastic packaging and the technical potential of chemical recycling of automotive plastic waste, an emission reduction of up to 26% is possible. The associated changes in network costs are also outlined in this study for all political steering strategies.

[1] This chapter includes the final version of the article "Circular Economy for Plastics in Germany: Decision Support for Optimal Recycling Network Designs" by Rebekka Volk, Frank Schultmann, and myself. The paper is submitted to a scientific journal as (Stallkamp, Volk, and Schultmann, 2023). The supporting information will be published with the journal article.

E.1 Introduction

In 2020, global plastic production reached 367 million tons[2] (PlasticsEurope, 2022), and current forecasts indicate an increase in annual global production (IEA, 2018). Fossil raw materials and the energy-demanding production process contribute to plastics' considerable environmental footprint (IPCC, 2022; Cabernard et al., 2021). Multiple approaches to decarbonizing and reducing the environmental footprint of plastic production are pursued. One of them is the implementation of a circular economy via the so-called "R"-strategies such as recycling (Agora Industry, 2022).

Current mechanical recycling processes face challenges, such as treating mixed plastic waste and mechanical degradation, resulting in low recycling rates or downcycling (Pivnenko et al., 2015; Cossu and Lai, 2015). Therefore, alternative waste treatment options are researched, such as chemical recycling using pyrolysis. In pyrolysis, plastic is decomposed into solid, liquid, and gaseous products (Zeller et al., 2021). The liquid pyrolysis oil can potentially replace fossil hydrocarbon feedstock in petrochemistry to produce high-value chemicals and close the plastic cycle (Lechleitner et al., 2020). Pyrolysis is a complementary technology for the treatment of wastes that are not suitable for mechanical recycling (Qureshi et al., 2020).

The technical feasibility of pyrolysis is demonstrated for sorting residues from the mechanical recycling process of lightweight packaging (LWP) by Zeller et al. (2021) and for automotive plastic waste (APW) by Stallkamp, Hennig, Volk, Richter, et al. (2023), and Hennig et al. (2022). Volk et al. (2021) and Stallkamp, Hennig, Volk, Richter, et al. (2023) compare the economic and environmental performance of the pyrolysis of LWP sorting residues and APW to their current waste treatment routes employing energy recovery. Volk et al. (2021) highlight that combining mechanical and chemical recycling processes is economically and environmentally favorable for handling LWP waste. Stallkamp, Hennig, Volk, Richter, et al. (2023) show that energy recovery is economically favorable over pyrolysis for APW but is associated with higher environmental burdens and, thereby, identifies a conflict of objectives.

However, more than environmental and economic analyses are needed to help decision-makers choose conducive steering strategies to design regulatory frameworks that support a plastic circular economy and decarbonize waste treatment (Sommer et al., 2022). Sommer et al. (2022) argue that the waste treatment system with its interrelations of technologies and material flows must be modeled to understand and analyze system behavior and to draw respective conclusions and recommendations for action. Specific regional and logistical aspects, i.e., regional waste volume, waste treatment capacities, and economic and environmental assessment criteria, must be considered to identify the most beneficial end-of-life (EoL) route per each waste stream and to assess the potential impact of current and novel regulatory frameworks (Sommer et al., 2022).

Valenzuela et al. (2021) provide an overview of waste treatment models in the context of plastics recycling, including models that relocate network facilities or that have multi-objective formulations

[2] Tons refer to metric tons throughout the article.

minimizing costs and climate change impact (CI) jointly. However, none of the existing studies include waste handling options of chemical recycling using pyrolysis.

Therefore, this study models the current German waste treatment network for LWP sorting residues and APW employing energy recovery. Moreover, it compares its processing costs and CI to an network design integrating chemical recycling[3] plants with optimal capacity and location. The model considers infrastructural preconditions, potential economies of scale, and the material flow on a national level in 2021. For the said purpose, the model is extended to a multi-objective location model minimizing total network costs and CI to assess how the network structure changes when both objectives are included in the optimization. For this purpose, a goal programming approach was chosen to minimize the distance between the optima and the individual optimizations with equal weighting.

In addition, different potential political steering strategies to establish chemical recycling by pyrolysis are analyzed and discussed. This supports political decision-makers in aligning environmental and economic interests and supporting the development of a circular economy for plastics. The approach is fully transferable to other study areas.

E.2 Materials

This section describes the plastic waste feedstock sources for LWP sorting residues and APW throughout Germany and the costs and CI associated with the assessed EoL options. Due to data availability, the reference year for the modeling is 2021.

E.2.1 Plastic waste feedstock sources

Pyrolysis is a complementary technology to mechanical recycling (Qureshi et al., 2020). So, only waste streams unsuitable for mechanical recycling are included in this model. Due to data availability, the included waste streams are limited to LWP sorting residues and APW from dismantling facilities.

E.2.1.1 LWP sorting residues

In Germany, LWP is separately collected and sorted in LWP sorting facilities. Recyclable metals[4] and fiber-based materials[5] account for 26% of the input of the sorting plant (Christiani and Beckamp, 2020). They are separated for individual treatment. The remaining packaging waste can be divided into mechanically high-grade recyclable packaging (32%) and miscellaneous plastic composites (17%) that are mechanically hard to recycle (Christiani and Beckamp, 2020). Sensor-based sorting

[3] This paper uses chemical recycling and pyrolysis synonymously in the following.
[4] tin, aluminum
[5] paper, cardboard, liquid board

assigns identifiable individual plastics to plastic mono streams that are further used in recycling processes. The miscellaneous plastic composites are not assigned to these plastic mono streams and are a possible feedstock for the pyrolysis process. When adding the waste fraction of sorting residues (25%), 42% of the total input of LWP sorting plants (Christiani and Beckamp, 2020) is a possible feedstock for pyrolysis. These fractions are currently incinerated.

In Germany, around 2,681 kt LWP waste from households were sorted within sorting facilities in 2021[6]. This results in a waste feedstock of 1,126 kt for energy recovery or chemical recycling. Figure 5 (cf. Appendix A) provides an overview of the spatial resolution of this waste. Additionally, the initial treatment plants for the considered waste are mapped: LWP waste is treated in LWP sorting facilities, and the APW is generated in automotive dismantling facilities.

E.2.1.2 Automotive plastic waste from EoL vehicles

The APW is a waste fraction of plastic parts dismantled from vehicles (Stallkamp, Hennig, Volk, Richter, et al., 2023). The recycling of this waste fraction is challenged by highly functionalized engineering thermoplastics and a wide variety of polymers and compounds. For this reason, APW is mostly incinerated (Cossu and Lai, 2015). APW is generated during the repair of vehicles and the dismantling of EoL vehicles. A conservative waste volume estimation can be established by focusing on EoL vehicles and on the dismantling of significant plastic components. According to UBA (2022), currently around 3 kg of plastic parts are dismantled from an EoL vehicle before shredding in Germany. In 2021, 337,135 EoL vehicles remained in Germany for recycling (Kraftfahrtbundesamt, 2022), resulting in an APW volume of around 1011 tons.

The dismantled APW is delivered to a refuse-derived fuel (RDF) producer. Here, metals are separated (9%; Stallkamp, Hennig, Volk, Richter, et al. (2023); Hennig et al. (2022)), and the remaining plastics are processed into a RDF. RDF is a feedstock for energy recovery or chemical recycling. The amount of APW is low compared to the LWP sorting residues. However, the above estimation is conservative since it does not account for APW from repair jobs but only for current EoL dismantling. The APW can also become more relevant if dismantling large plastic components become part of automobiles' EoL treatment processes (e.g., via regulation) (Wilts et al., 2016).

E.2.2 EoL options for the assessed plastic waste

The current waste treatment option for the assessed plastic waste stream is waste incineration with energy recovery. Therefore, the scenario introducing chemical recycling is compared to a network with energy recovery only.

[6] Based on waste statistics of the federal states (DESTATIS, 2022).

E.2.2.1 Energy recovery

The LWP waste is separately collected in German households. The waste is transported to sorting facilities with lorries associated with 1.40 kg CO_2e emissions per ton and kilometer and transport costs of 0.18 € per ton and kilometer (Doka, 2022). We calculate the Euclidean distance for all transportation distances and apply a tortuosity factor of $\sqrt{2}$ to embrace actual road conditions (Delivand, 2011; Diehlmann et al., 2019).

LWP sorting facilities separate metals, fiber-based materials, and other non-plastic material fractions using conventional technologies including comminution, classification, sifting, and metal separation (Stapf et al., 2019). Sensor-based sorting separates the plastic mono streams for individual recycling. Miscellaneous plastic composites and sorting residues remain as a waste stream. The model focuses on this waste stream, comparing its incineration with energy recovery and chemical recycling. It is assumed that all other material streams[7] are handled the same way in all network designs and are therefore irrelevant for the relative comparison of the waste treatment network designs. In Germany, LWP sorting is associated with processing costs of 84 € and emissions of 335 kg CO_2e per ton input (Volk et al., 2021).

The sorting residues are a valuable feedstock for energy recovery due to their high calorific value. The transport to RDF and municipal solid waste incineration (MSWI) plants with lorries with a capacity of 16-32 tons freight is associated with transportation costs of 0.04 € per ton and kilometer and 0.19 kg CO_2e emissions per ton and kilometer (Valsasina, 2022a). As the pretreatment performed at the LWP waste sorting facility is more refined than the RDF producer's usual steps, further treatment of sorting residues is omitted (Volk et al., 2021).

The sorting residues are incinerated at RDF and MSWI plants, and energy (heat and electricity) is recovered. The energy recovery at MSWI and RDF plants is associated with costs of 123 €/ton input (Bilitewski et al., 2018) and 2,350 kg CO_2e/ton input. The CI of the incineration is calculated based on the inputs' heating value, and an emission factor for RDF[8]. The processing costs are credited with revenues from selling the produced heat (0.08 €/kWh) and electricity (0.10 €/kWh), assuming the average selling price in 2021. Based on their electricity and heat efficiencies[9], the net processing costs accounting for revenues result in -185 €/ton input for RDF and -94 €/ton input for MSWI plants. Negative net processing costs mean that revenues exceed material treatment costs, and the facility generates a profit.

For APW, the process chain starts with EoL vehicles and their transportation to dismantling facilities. Transportation with lorries with 32 tons freight is assumed, resulting in costs of 0.02 € per ton and kilometer and 0.10 kg CO_2e emissions per ton and kilometer (Valsasina, 2022b). Dismantling large plastic parts requires equipment for dry laying, a lifting platform, mechanical equipment, and

[7] metals, fiber-based materials, mono plastic streams

[8] The heating value of LWP sorting residues is 25 MJ/kg (Zeller et al., 2021) and the emission factor is 0.09 kg CO_2e/MJ (Flamme et al., 2018)

[9] The average German RDF power plant has an electricity efficiency of 0.15 and a heat efficiency of 0.37, resulting in a total efficiency of 0.52, while the average German MSWI plant has an electricity efficiency of 0.19 and a heat efficiency of 0.16 resulting in a total efficiency of 0.35 (Flamme et al., 2018).

appropriate personnel (Lander, 2004). The process costs add up to 4 €/ton, and the emissions amount to 9 kg CO_2e/ton (Del Duce, 2022).

The dismantled plastic parts are forwarded to a RDF producer assuming transportation with 16-32 tons freight lorries (Valsasina, 2022a). Here, metals are separated, and the remaining plastic waste is shredded (Stallkamp, Hennig, Volk, Richter, et al., 2023). The process is associated with 60 €/ton input costs and a CI of 140 kg CO_2e/ton input (Stallkamp, Hennig, Volk, Richter, et al., 2023). Afterward, the RDF is delivered to energy recovery facilities using 16-32 tons freight lorries. Its incineration is associated with net processing costs of -197 €/ton input and 2,472 kg CO_2e/ton input for RDF power plants and net processing costs of -103 €/ton input and 2,472 kg CO_2e/ton input for MSWI plants. Revenues for the recovered energy and emissions are calculated based on the material's heating value, the RDF emission factor, and the plant efficiencies[10].

E.2.2.2 Chemical recycling via pyrolysis

The chemical recycling route differs from the energy recovery route in handling the RDF from APW and LWP sorting residues. In the pyrolysis, both waste feedstocks yield pyrolysis oil and the by-products of pyrolysis gas, aqueous condensate, and solid residues (Stallkamp, Hennig, Volk, Richter, et al., 2023; Hennig et al., 2022). Pyrolysis gas can generate electricity and district heating by utilizing a combined heat and power unit (CHPU) (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023). According to Stallkamp, Hennig, Volk, Stapf, and Schultmann (2023), depending on the amount of electricity generated, the energy demand of the pyrolysis plant can be covered, and excess electricity can be fed to the grid. If the electricity demand of the pyrolysis plant surpasses the electricity generated by the CHPU, electricity is supplied from the grid (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023). Excess heat recovered from the CHPU is supplied to a district heating network (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023). The by-products of aqueous condensate and solid residues are considered waste that must be disposed of (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023).

Pyrolysis plants are not yet an established part of the waste treatment infrastructure in Germany or worldwide. So, their placement involves an investment where their optimal input capacity and location has to be determined. Due to economies of scale, no single cost metric and CI can be provided. However, Stallkamp, Hennig, Volk, Stapf, and Schultmann (2023) outlines the cost structure of APW pyrolysis plants of different capacities. Based on this modeling, capacity classes are established, and the associated operating costs and CI for waste handling are calculated accordingly. For each capacity class, the fix operating costs are independent from the handled material[11]. The variable operating costs for handling the input, the fix CI[12] for placing the plant,

[10] The material's heating value is 26 MJ/kg (Stallkamp, Hennig, Volk, Richter, et al., 2023) and the RDF emission factor is 0.09 kg CO_2e/MJ (Flamme et al., 2018)

[11] Include the cost of capital based on the fixed capital investment and an annuity factor (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023)

[12] Based on a reference plant and scaling (Dauriat, 2023) with linear annualization (20 years) according to German Federal Ministry of Finance (1995)

and the variable CI for handling the input are calculated based on the model of Stallkamp, Hennig, Volk, Stapf, and Schultmann (2023).

Ten different capacity classes are introduced, ranging from an input capacity of 3,750 tons input/year up to 120,000 tons input/year. Related data, cost functions, and a description are provided in the supporting information (cf. Appendix B). Here, costs and CI for a pyrolysis plant with a 20,000 tons input/year capacity are outlined.

For LWP sorting residues, variable operating costs of -35 €/ton input[13] and a variable CI of 255 kg CO_2e/ton input are calculated, while for APW, the variable operating costs sum up to 11 €/ton input and the variable CI is 166 kg CO_2e/ton input (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023). The associated fix operating costs for a 20,000 tons input/year capacity are calculated to be 4.6 Mio. €/year, while the construction process results in annualized[14] emissions of 42 tons CO_2e. The operating costs are credited with revenues from sold pyrolysis oil. Here, the market price for heavy fuel oil[15] is used to account for the lower quality of the pyrolysis oil compared to naphtha (Riedewald et al., 2021). The pyrolysis of LWP sorting residues yields 40% pyrolysis oil, 30% pyrolysis gas, 1% aqueous condensate, and 29% solid residues (Zeller et al., 2021). With APW as feedstock, the pyrolysis process yields 50% pyrolysis oil, 20% pyrolysis gas, 1% aqueous condensate, and 29% solid residues (Stallkamp, Hennig, Volk, Richter, et al., 2023). Variable operating costs, the fix operating costs, and the credits for the pyrolysis oil result in processing costs of 12 €/ton LWP sorting residues and 12 €/ton APW. Fix and variable CI result in an impact of 257 kg CO_2e/ton input for LWP sorting residues and 169 kg CO_2e/ton input for APW.

After performing an additional hydroprocessing step (Kusenberg, Eschenbacher, et al., 2022), pyrolysis oil is a potential feedstock for steam cracking (Kusenberg, Roosen, et al., 2022). Thus, refineries with steam crackers are integrated into the model to consider them when deciding on the location and capacity of pyrolysis plants. It is assumed that the necessary hydroprocessing steps for the steam cracker application can be performed on-site at the refinery. No decision will be made about the capacity and placement of hydroprocessing plants.

Costs and CI of all facilities and transportation are summarized in Table E.1 for both waste fractions. Due to the discussion of whether net values are permissible when considering EoL options, the case study was also calculated with gross input data not considering revenues for generated products (cf. Appendix C).

E.3 Methodological approach

The material introduced is input for multiple optimization models addressing plastic waste treatment networks. Single optimization models are formulated comparing the waste treatment network

13 For the explanation of negative operating costs cf. Appendix B
14 Linear annualization over 20 years according to German Federal Ministry of Finance (1995)
15 The market price is 462 €/ton in 2021 (EIA, 2022).

Table E.1: Net costs and global warming potential of energy recovery and chemical recycling route.

	LWP			APW			Reference
	Costs* [€/Mg]**	**Yield** [-]	**GWP** [kg CO2eq/Mg]	**Costs*** [€/Mg]**	**Yield** [-]	**GWP** [kg CO2eq/Mg]	
Facilities							
LWP sorting facility	84	0.42	335	-	-	-	[1]
RDF power plants	-185	-	2350	-197	-	2472	[1,2]
MSWI plants	-94	-	2350	-103	-	2472	[1,2]
Dismantling facilities	-	-	-	4	0.003	9	[3]
RDF producer	-	-	-	60	0.91	140	[2]
Pyrolysis plant*	12	0.40	257	12	0.50	169	[4,5,6]
Transport							
LWP waste**	0.18	-	1.40	-	-	-	[8]
EoL vehicles**	-	-	-	0.02	-	0.10	[8]
Intermediates**	0.04	-	0.19	0.04	-	0.19	[9]
Pyrolysis oil**	-	-	-	0.02	-	0.10	[8]

[1]: Volk et al. (2021), [2]: Stallkamp, Hennig, Volk, Richter, et al. (2023), [3]: Del Duce (2022),
[4]: Stallkamp, Hennig, Volk, Stapf, and Schultmann (2023), [5]: Zeller et al. (2021), [6]: Brunner (2022),
[7]: Doka (2022), [8]: Valsasina (2022b) , [9]: Valsasina (2022a)
*: for a input capacity of 20,000 Mg per year
**: per Mg and kilometer
***: include revenues from selling produced products

options of energy recovery (Model 1) with an approach combining energy recovery and chemical recycling (Model 2). In addition, a multi-objective decision model examines the optimal network configuration when energy recovery and chemical recycling are combined, and the network is optimized for cost and CI simultaneously (Model 3). The models enable decision-makers to assess political steering strategies to align the economically and environmentally favorable plastic waste treatment options. The models are applied in a case study of Germany.

E.3.1 Optimization approach

Waste treatment networks consist of collection, sorting, and processing stages that must be coordinated, focusing on optimizing the recovery of EoL resources and costs (Egri et al., 2021). The networks can be designed by solving a facility location problem to establish optimal locations of waste handling facilities while minimizing costs, environmental burdens, or both. Facility location planning has been extensively studied (Dekker, 2004), and extensions include capacity restrictions, considering existing facilities, or assessing multiple material streams (Nickel et al., 2014). An overview of waste treatment network models in the context of plastics recycling shows that there are no studies on expanding the waste treatment options with facilities for chemical recycling by pyrolysis (Valenzuela et al., 2021). Therefore, in this paper, we use different location decision models to compare the EoL options of energy recovery and chemical recycling in the overall context of a national waste treatment network.

Model 1 models the national plastic waste treatment in plants for energy recovery only. The waste is directed through the network such that the total network costs or CI are minimized. This optimization demands the handling of all LWP waste and APW from EoL vehicles generated. LWP and EoL vehicles accumulate in the centers of districts and are treated in primary treatment plants. For the primary treatment plants, the existing infrastructure of LWP waste sorting and dismantling facilities in Germany is used (cf. Figure 5, Appendix A). After its dismantling from EoL vehicles, the APW is transported to RDF producers. RDF made from APW, and the LWP sorting residues are feedstock for the energy recovery facilities. Here, existing infrastructure is used.

Model 2 expands the formulation of Model 1 and includes the construction of pyrolysis plants for chemical recycling. Therefore, the minimization of total network costs or CI includes constructing the pyrolysis plants, transports between facilities, and waste handling costs. For the pyrolysis plants, discrete plant capacity classes are assumed due to adding single reactors to the plant design to increase capacity (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023). The annualized plant investment is part of the fix operating costs and calculated with an interest rate of 3% and a plant operating life of 20 years (Stallkamp, Hennig, Volk, Stapf, and Schultmann, 2023). In addition to the constraints of material flow conservation, the capacities of the individual facilities must not be exceeded.

Model 3 extends the single-objective optimization to a multi-objective decision-making (MODM) model. Here, both objectives of network costs and CI are simultaneously optimized. There are several approaches to determining the solution to such optimization problems. Stallkamp et al. (2022) compared goal programming and lexicographic optimization regarding their suitability to model recycling networks, considering conflicting objectives. They highlight the advantage of goal programming in establishing a balanced solution between competing objectives (Stallkamp et al., 2022). Therefore, Model 3 implements a goal programming approach extending the single objective optimization to an MODM optimization.

An overview of the EoL paths, objectives, decisions to make, and constraints for the decisions within the three models is provided in Figure E.1.

E.3.2 Mathematical model description

In the following, the general model descriptions of Model 1 to Model 3 are translated into mixed integer linear programming (MILP) models. Table E.2 provides an overview of the notation of the problems' sets, parameters, and variables.

Table E.2: Notation of sets, parameters, and variables used in the optimization models.

Sets	
W	Set of districts with waste sources ($w \in W$)
S	Set of sorting facilities for LWP waste ($s \in S$)
D	Set of dismantling facilities for EoL vehicles ($d \in D$)

P	Set of facilities treating APW waste for the recovery stages ($p \in P$)
R_{ER}	Set of facilities for energy recovery ($rer \in R_E R$)
R_{CR}	Set of districts with potential locations for chemical recycling facilities ($rcr \in R_C R$)
C	Set of customers with demand for pyrolysis oil ($c \in C$)
K	Set of input capacity classes of pyrolysis plants ($k \in K$)
M	Set of material waste streams ($m \in M$)
Parameters	
$d_{w,s}$	Distance between districts w and facilities s in kilometer
$d_{w,d}$	Distance between districts w and facilities d in kilometer
$d_{d,p}$	Distance between facilities d and p in kilometer
$d_{s,rer}$	Distance between facilities s and rer in kilometer
$d_{p,rer}$	Distance between facilities p and rer in kilometer
$d_{s,rcr}$	Distance between facilities s and rcr in kilometer
$d_{p,rcr}$	Distance between facilities p and rcr in kilometer
d_{rc}	Distance between facilities rcr and c in kilometer
$waste_{w,m}$	Waste material of material m at location w
$demand_c$	Demand of pyrolysis oil at location c
$c^t_{w,s}$, $c^t_{w,d}$, $c^t_{d,p}$, $c^t_{s,rer}$, $c^t_{p,rer}$,$c^t_{s,rcr}$, $c^t_{p,rcr}$, c^t_{rc}	Transportation costs for 1 ton material per kilometer
$ci^t_{w,s}$, $ci^t_{w,d}$, $ci^t_{d,p}$, $ci^t_{s,rer}$, $ci^t_{p,rer}$,$ci^t_{s,rcr}$, $ci^t_{p,rcr}$, ci^t_{rc}	Transportation CI for 1 ton material per kilometer
$c_{s,m}$, $c_{d,m}$, $c_{p,m}$, $c_{rer,m}$	Processing costs per ton input material m at facilities s, d, p, rer
$ci_{s,m}$, $ci_{d,m}$, $ci_{p,m}$, $ci_{rer,m}$	Processing CI per ton input material m facilities s, d, p, rer
$\gamma_{s,m}$, $\gamma_{d,m}$, $\gamma_{p,m}$, $\gamma_{rer,m}$	Product yield per ton input material m in facilities s, d, p, rer
$capacity_{s,m}$, $capacity_{d,m}$, $capacity_{p,m}$	Input capacity for material m at facilities s, d, p
$capacity_{rer}$	Input capacity at facilities rer
$c_{k,m}$	Variable operating costs per ton material for material m at pyrolysis plant of capacity k
c^{fix}_k	Fix operating costs for pyrolysis plant of capacity k
$ci_{k,m}$	Processing CI per ton material for material m at pyrolysis plant of capacity k
ci^{fix}_k	Annualized CI for placing a pyrolysis plant of capacity k
$\gamma_{k,m}$	Product yield per ton material for material m at pyrolysis plant of capacity k

$capacity_{rcr,k}$	Input capacity at facilities rcr depending on capacity class k
F_1^E	Cost single objective optimum, relevant for the maximum deviation in the goal programming approach
F_2^E	CI single objective optimum, relevant for the maximum deviation in the goal programming approach
Variables	
$x_{ws,m}$, $x_{wd,m}$, $x_{dp,m}$, $x_{srer,m}$, $x_{prer,m}$, $x_{srcr,m,k}$, $x_{prcr,m,k}$, x_{rc}	Amount of transported material m between facilities
$y_{r,k}$	$\begin{cases} 0, \text{no plant of capacity class } k \text{ is opened in district } r \\ 1, \text{a plant of capacity class } k \text{ is opened in district } r \end{cases}$
$dist$	Maximum deviation from each normalized single objective optimum in the goal programming approach
$F_1(x, y)$	Cost minimization as a function of the decision variables x and y in the goal programming approach
$F_2(x, y)$	CI minimization as a function of the decision variables x and y in the goal programming approach

Set W contains all districts where waste is generated. Collection facilities are grouped in the sets S (sorting facilities for LWP waste) and D (dismantling facilities for EoL vehicles). Existing pretreatment facilities (RDF producer) for APW are collected in set P. Set R_{ER} consists of all existing energy recovery facilities. Set R_{CR} contains all districts where pyrolysis plants can be placed. Set C includes all existing customers for pyrolysis oil, such as refineries with steam crackers. Set K contains all potential capacities for pyrolysis plants and set M the waste streams LWP sorting residues and APW.

Parameters for calculation include distances between facilities, regional waste generation, and customer demand for pyrolysis oil. Material-specific transport costs and emissions, costs and emissions of processing the materials at the different facilities, and the specific process yields are also input parameters. The material handling capacities are provided by facility and material. There is no distinction between materials for the energy recovery facilities' capacities as they handle both LWP sorting residues and APW equally. For the pyrolysis plants, variable operating costs, process CI and process yield depend on the capacity class and the input material. In contrast, fixed operating costs and CI only depend on the capacity class.

Decision variables determine the amount of transported material between districts and facilities. If pyrolysis plants are constructed, then the binary variables $y_{r,k}$ indicate if a pyrolysis plant of capacity k is placed in the district r.

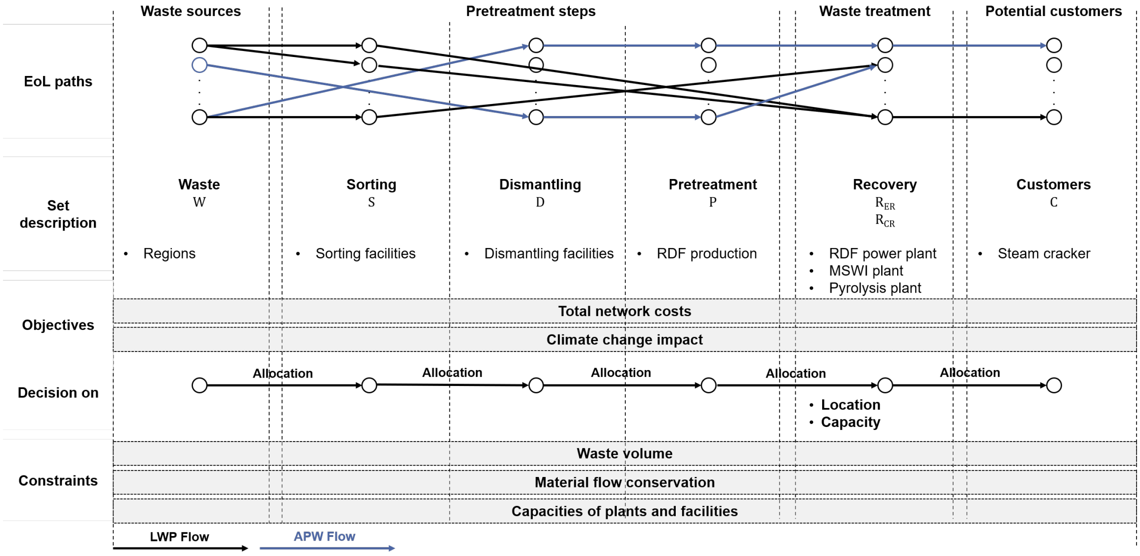

Figure E.1: Overview mathematical optimization model.

E.3.2.1 Model 1: Existing energy recovery network

Model 1 replicates the existing waste treatment system in Germany and optimizes its material flows. The model's objective function (1) minimizes the transport and processing costs. Decision variables are the material flows between facilities. Minimization is performed under the constraints of flow conservation (Constraints (2) to (6)). Materials entering a facility must exit it considering process yields. Plant-specific capacities must not be exceeded (Constraints (7) to (10)). Constraints (11) and (12) define the value range of the decision variables.

$$\begin{aligned}
\min \quad & \sum_{w\in W}\sum_{s\in S}\sum_{m\in M} x_{ws,m}\cdot c^{t}_{ws}\cdot d_{ws} \\
& + \sum_{w\in W}\sum_{d\in D}\sum_{m\in M} x_{wd,m}\cdot c^{t}_{wd}\cdot d_{wd} \\
& + \sum_{d\in D}\sum_{p\in P}\sum_{m\in M} x_{dp,m}\cdot c^{t}_{dp}\cdot d_{dp} \\
& + \sum_{p\in P}\sum_{r\in R_{ER}}\sum_{m\in M} x_{prer,m}\cdot c^{t}_{prer}\cdot d_{prer} \\
& + \sum_{s\in S}\sum_{r\in R_{ER}}\sum_{m\in M} x_{srer,m}\cdot c^{t}_{srer}\cdot d_{srer} \\
& + \sum_{w\in W}\sum_{s\in S}\sum_{m\in M} x_{ws,m}\cdot c_{s,m} \\
& + \sum_{w\in W}\sum_{d\in D}\sum_{m\in M} x_{wd,m}\cdot c_{d,m} \\
& + \sum_{d\in D}\sum_{p\in P}\sum_{m\in M} x_{dp,m}\cdot c_{p,m} \\
& + \sum_{p\in P}\sum_{r\in R_{ER}}\sum_{m\in M} x_{pre,m}\cdot c_{rer,m} \\
& + \sum_{s\in S}\sum_{r\in R_{ER}}\sum_{m\in M} x_{srer,m}\cdot c_{rer,m}
\end{aligned} \tag{1}$$

s.t.

$$\sum_{s\in S} x_{ws,m} = waste_{w,m}, \qquad \forall w \in W, \forall m \in 0, \tag{2}$$

$$\sum_{d\in D} x_{wd,m} = waste_{w,m}, \qquad \forall w \in W, \forall m \in 1, \tag{3}$$

$$\sum_{r\in R_{ER}} x_{srer,m} = \gamma_{s,m} \cdot \sum_{w\in W} x_{ws,m}, \qquad \forall s \in S, \forall m \in 0, \tag{4}$$

$$\sum_{p\in P} x_{dp,m} = \gamma_{d,m} \cdot \sum_{w\in W} x_{wd,m}, \qquad \forall d \in D, \forall m \in 1, \tag{5}$$

$$\sum_{r\in R_{ER}} x_{prer,m} = \gamma_{p,m} \cdot \sum_{s\in S} x_{sp,m}, \qquad \forall p \in P, \forall m \in 1, \tag{6}$$

$$\sum_{w\in W} x_{ws,m} \leq capacity_{s,m}, \qquad \forall s \in S, \forall m \in 0, \tag{7}$$

$$\sum_{w\in W} x_{wd,m} \leq capacity_{d,m}, \qquad \forall d \in D, \forall m \in 1, \tag{8}$$

$$sum_{d\in D} x_{dp,m} \leq capacity_{p,m}, \qquad \forall p \in P, \forall m \in 1, \tag{9}$$

$$\sum_{s\in S}\sum_{m\in M} x_{srcr,m} + \sum_{p\in P}\sum_{m\in M} x_{prcr,m} \leq capacity_{rer}, \forall r \in R_{ER}, \tag{10}$$

$$x_{ws,m}, x_{wd,m} \geq 0, \qquad \forall w \in W, \quad \forall s \in S, \quad \forall d \in D,, \tag{11}$$

$$x_{dp,m}, x_{srer,m}, x_{prer,m} \geq 0, \qquad \forall d \in D, \forall s \in S, \forall p \in P, \forall r \in R_{ER} \tag{12}$$

E.3.2.2 Model 2: Combining energy recovery and pyrolysis

Model 2 extends the existing energy recovery infrastructure by placing pyrolysis plants combining waste treatment technologies. The objective function (13) reflects this by adding the fix operating costs for the placed pyrolysis plants depending on their respective capacities. Additional transportation costs between pyrolysis plants and potential customers for pyrolysis oil are modeled. Waste treatment costs include now material processing at the energy recovery and pyrolysis plants.

This minimization is also subject to flow conservation (Constraints (14) to (19)). Flows from sorting and pretreatment facilities are adjusted to allow transports to chemical recycling and energy recovery facilities. Customers may be supplied with up to a maximum of their demand (Constraint 20). The processing plants' capacities must not be exceeded (Constraints (21) to (25)). For the pyrolysis plants, this maximum capacity is determined by the capacity class selected. Constraint (26) specifies that only one pyrolysis plant can be placed in each district. Constraints (27) to (30) define the range of values of the decision variables.

$$\begin{aligned}
\min \quad & \sum_{w\in W}\sum_{s\in S}\sum_{m\in M} x_{ws,m} \cdot c^t_{ws} \cdot d_{ws} \\
& + \sum_{w\in W}\sum_{d\in D}\sum_{m\in M} x_{wd,m} \cdot c^t_{wd} \cdot d_{wd} \\
& + \sum_{d\in D}\sum_{p\in P}\sum_{m\in M} x_{dp,m} \cdot c^t_{dp} \cdot d_{dp} \\
& + \sum_{p\in P}\sum_{r\in R_{cr}}\sum_{m\in M}\sum_{k\in K} x_{prcr,m,k} \cdot c^t_{prcr} \cdot d_{prcr}
\end{aligned}$$

$$+\sum_{s\in S}\sum_{r\in R_{cr}}\sum_{m\in M}\sum_{k\in K} x_{srcr,m,k}\cdot c^{t}_{srcr}\cdot d_{srcr}$$

$$+\sum_{r\in R_{cr}}\sum_{c\in C} x_{rc}\cdot c^{t}_{rc}\cdot d_{rc}$$

$$+\sum_{p\in P}\sum_{r\in R_{ER}}\sum_{m\in M} x_{prer,m}\cdot c^{t}_{prer}\cdot d_{prer}$$

$$+\sum_{s\in S}\sum_{r\in R_{ER}}\sum_{m\in M} x_{srer,m}\cdot c^{t}_{srer}\cdot d_{srer}$$

$$+\sum_{w\in W}\sum_{s\in S}\sum_{m\in M} x_{ws,m}\cdot c_{s,m}$$

$$+\sum_{w\in W}\sum_{d\in D}\sum_{m\in M} x_{wd,m}\cdot c_{d,m}$$

$$+\sum_{d\in D}\sum_{p\in P}\sum_{m\in M} x_{dp,m}\cdot c_{p,m}$$

$$+\sum_{p\in P}\sum_{r\in R_{ER}}\sum_{m\in M} x_{pre,m}\cdot c_{rer,m}$$

$$+\sum_{s\in S}\sum_{r\in R_{ER}}\sum_{m\in M} x_{srer,m}\cdot c_{rer,m}$$

$$+\sum_{p\in P}\sum_{r\in R_{cr}}\sum_{m\in M}\sum_{k\in K} x_{prc,m,k}\cdot c_{k,m}$$

$$+\sum_{s\in S}\sum_{r\in R_{cr}}\sum_{m\in M}\sum_{k\in K} x_{src,m,k}\cdot c_{k,m}$$

$$+\sum_{r\in R_{CR}}\sum_{k\in K} y_{r,k}\cdot c^{fix}_{k} \tag{13}$$

s.t.

$$\sum_{s\in S} x_{ws,m} = waste_{w,m}, \qquad \forall w\in W, \forall m\in 0, \tag{14}$$

$$\sum_{d\in D} x_{wd,m} = waste_{w,m}, \qquad \forall w\in W, \forall m\in 1, \tag{15}$$

$$\sum_{r\in R_{RC}}\sum_{k\in K} x_{srcr,m,k} + \sum_{r\in R_{ER}} x_{srer,m} = \gamma_{s,m}\cdot\sum_{w\in W} x_{ws,m}, \qquad \forall s\in S, \forall m\in 0, \tag{16}$$

$$\sum_{p\in P} x_{dp,m} = \gamma_{d,m}\cdot\sum_{w\in W} x_{wd,m}, \qquad \forall d\in D, \forall m\in 1, \tag{17}$$

$$\sum_{r\in R_{RC}}\sum_{k\in K} x_{prcr,m,k} + \sum_{r\in R_{ER}} x_{prcr,m} = \gamma_{p,m}\cdot\sum_{d\in D} x_{dp,m}, \qquad \forall p\in P, \forall m\in 1, \tag{18}$$

$$\sum_{c\in C} x_{rc} = \sum_{s\in S}\sum_{k\in K} x_{srcr,m,k}\cdot\gamma_{k,m} + \sum_{p\in P}\sum_{k\in K} x_{prcr,m,k}\cdot\gamma_{k,m}, \forall r\in R_{CR}, \forall m\in M, \tag{19}$$

$$\sum_{r\in R_{CR}} x_{rc} \le demand_{c}, \qquad \forall c\in C, \tag{20}$$

$$\sum_{w\in W} x_{ws,m} \le capacity_{s,m}, \qquad \forall s\in S, \forall m\in 0, \tag{21}$$

$$\sum_{w\in W} x_{wd,m} \le capacity_{d,m}, \qquad \forall d\in D, \forall m\in 1, \tag{22}$$

$$sum_{d\in D} x_{dp,m} \le capacity_{p,m}, \qquad \forall p\in P, \forall m\in 1, \tag{23}$$

$$\sum_{s\in S}\sum_{m\in M} x_{srcr,m} + \sum_{p\in P}\sum_{m\in M} x_{prcr,m} \le capacity_{rer}, \qquad \forall r\in R_{ER}, \tag{24}$$

$$\sum_{s\in S}\sum_{m\in M} x_{srcr,m,k} + \sum_{p\in P}\sum_{m\in M} x_{prcr,m,k} \le capacity_{rcr,k}\cdot y_{r,k}, \forall r\in R_{CR}, \forall k\in K, \tag{25}$$

$$\sum_{k\in K} y_{r,k} \le 1, \qquad \forall r\in R_{CR}, \tag{26}$$

$$x_{ws,m}, x_{wd,m}, x_{dp,m} \ge 0, \qquad \forall w\in W, \forall s\in S, \forall d\in D, \forall p\in P, \forall m\in M, \tag{27}$$

$$x_{prer,m}, x_{srer,m}, x_{rc,m} \geq 0, \qquad \forall p \in P, \forall s \in S, \forall r \in R_{ER}, \forall c \in C, \forall m \in M, \tag{28}$$

$$x_{srcr,m,k}, x_{prcr,m,k} \geq 0, \qquad \forall s \in S, \forall p \in P, \forall r \in R_{CR}, \forall m \in M, \forall k \in K, \tag{29}$$

$$y_{r,k} \in \{0,1\}, \qquad \forall r \in R_{CR}, \forall k \in K \tag{30}$$

E.3.2.3 Model 3: Multi objective approach optimizing cost and CI simultaneously

Model 3 is an MODM model and optimizes both objective functions simultaneously. The optimization is run in a defined solution space restricted by the problem's constraint (Walther, 2010) and quantifiable objective functions describe the decision-maker's objective system (Geldermann and Lerche, 2014). The goal programming approach implemented in Model 3 is a min-max approach using a maximum norm minimizing the maximum deviation from each objective (Flavell, 1976). Thereby, the approach leads to a good balance between all objectives.

The continuous, non-negative auxiliary variable $dist$ is introduced for the problem formulation. It measures the distance of each objective to its respective single-criteria optimum. The variable $dist$ should be minimized, establishing the objective function (31).

Constraints (32) to (44) are equal to the constraint (14) to (26) of Model 2. Constraints (45) and (46) represent the distance measurement, where the distances between objective values are normalized using a percentage normalization and the single-criteria optima F_1^E (total network costs) and F_2^E (total network CI). Consequently, the variable $dist$ is interpreted as the relative deviation (in percent) of the objective value from its optimal value. Constraints (47) to (50) define the range of the decision variables, including $dist$.

$$\min \quad dist \tag{31}$$

s.t.

$$\sum_{s \in S} x_{ws,m} = waste_{w,m}, \qquad \forall w \in W, \forall m \in 0, \tag{32}$$

$$\sum_{d \in D} x_{wd,m} = waste_{w,m}, \qquad \forall w \in W, \forall m \in 1, \tag{33}$$

$$\sum_{r \in R_{RC}} \sum_{k \in K} x_{srcr,m,k} + \sum_{r \in R_{ER}} x_{srer,m} = \gamma_{s,m} \cdot \sum_{w \in W} x_{ws,m}, \qquad \forall s \in S, \forall m \in 0, \tag{34}$$

$$\sum_{p \in P} x_{dp,m} = \gamma_{d,m} \cdot \sum_{w \in W} x_{wd,m}, \qquad \forall d \in D, \forall m \in 1, \tag{35}$$

$$\sum_{r \in R_{RC}} \sum_{k \in K} x_{prcr,m,k} + \sum_{r \in R_{ER}} x_{prcr,m} = \gamma_{p,m} \cdot \sum_{d \in D} x_{dp,m}, \qquad \forall p \in P, \forall m \in 1, \tag{36}$$

$$\sum_{c \in C} x_{rc} = \sum_{s \in S} \sum_{k \in K} x_{srcr,m,k} \cdot \gamma_{k,m} + \sum_{p \in P} \sum_{k \in K} x_{prcr,m,k} \cdot \gamma_{k,m}, \forall r \in R_{CR}, \forall m \in M, \tag{37}$$

$$\sum_{r \in R_{CR}} x_{rc} \leq demand_c, \qquad \forall c \in C, \tag{38}$$

$$\sum_{w \in W} x_{ws,m} \leq capacity_{s,m}, \qquad \forall s \in S, \forall m \in 0, \tag{39}$$

$$\sum_{w \in W} x_{wd,m} \leq capacity_{d,m}, \qquad \forall d \in D, \forall m \in 1, \tag{40}$$

$$sum_{d\in D} x_{dp,m} \leq capacity_{p,m}, \qquad \forall p \in P, \forall m \in 1, \tag{41}$$

$$\sum_{s\in S}\sum_{m\in M} x_{srcr,m} + \sum_{p\in P}\sum_{m\in M} x_{prcr,m} \leq capacity_{rer}, \qquad \forall r \in R_{ER}, \tag{42}$$

$$\sum_{s\in S}\sum_{m\in M} x_{srcr,m,k} + \sum_{p\in P}\sum_{m\in M} x_{prcr,m,k} \leq capacity_{rcr,k} \cdot y_{r,k}, \forall r \in R_{CR}, \forall k \in K, \tag{43}$$

$$\sum_{k\in K} y_{r,k} \leq 1, \qquad \forall r \in R_{CR}, \tag{44}$$

$$\frac{F_1(x,y) - F_1^E}{F_1^E} - dist \leq 0,, \tag{45}$$

$$\frac{F_2(x,y) - F_2^E}{F_2^E} - dist \leq 0,, \tag{46}$$

$$x_{ws,m}, x_{wd,m}, x_{dp,m} \geq 0, \qquad \forall w \in W, \forall s \in S, \forall d \in D, \forall p \in P, \forall m \in M, \tag{47}$$

$$x_{prer,m}, x_{srer,m}, x_{rc,m} \geq 0, \qquad \forall p \in P, \forall s \in S, \forall r \in R_{ER}, \forall c \in C, \forall m \in M, \tag{48}$$

$$x_{srcr,m,k}, x_{prcr,m,k}, dist \geq 0, \qquad \forall s \in S, \forall p \in P, \forall r \in R_{CR}, \forall m \in M, \forall k \in K, \tag{49}$$

$$y_{r,k} \in \{0,1\}, \qquad \forall r \in R_{CR}, \forall k \in K \tag{50}$$

E.4 Results of the case study for Germany

All three models are applied to a case study of Germany and the two waste streams of LWP sorting residues and APW. As a baseline, the costs and CI associated with the current energy recovery waste treatment network are minimized (Model 1). Then, network designs combining energy recovery and chemical recycling are compared to the baseline network minimizing cost as well (Model 2). Finally, the goal programming approach is applied to minimize both costs and CI simultaneously to design a wholistic waste management network for both waste streams (Model 3).

E.4.1 Model 1: Existing energy recovery network

In the first run of Model 1, the optimum material flow through the existing energy recovery infrastructure that minimizes its total costs is identified. The shortest distances between facilities are generally chosen for transportation. RDF plants are preferred over MSWI plants for incineration since more energy can be recovered due to their higher efficiency. Here, longer transport distances are accepted to exploit the associated cost advantage for waste incineration.

Table 3 (cf. Appendix A) provides an overview of the costs at each stage of the waste treatment network as well as each transport connection. The numbers result from running Model 1 with the introduced input data for the German case study. Negative costs can occur at the final stages of the network and mean that the revenues for the product are higher than the processing costs at that stage. Thus, such negative costs equal profit and a positive business case. The total waste management system is associated with costs of 61 Mio. €/year. The processing cost account for 18 Mio. €/year (30%) while 43 Mio. €/year (70%) transportation costs will occur in the optimized case. Most processing costs are accounted for by sorting LWP (225 Mio. €/year), while energy recovery provides a profit (208 Mio. €/year).

When minimizing the CI of the existing energy recovery infrastructure, the shortest distance between districts and plants is chosen, and RDF and MSWI plants are addressed equally. Waste incineration in both plant types is associated with the same emissions since no CO_2 credits for substituting electricity and heat from other sources are issued. The minimization of CO_2 emissions results in total emissions of 3,852 kt CO_2e/year. In the cost-optimal network, the total CO_2 emissions are also 3,852 kt CO_2e/year, so no environmentally advantageous network configuration can be chosen here (cf. Table 4, Appendix A). The highest share of emissions is from waste processing or treatment (92%). Incineration accounts for 75% of the waste processing emissions, while pretreatment steps only account for 25%. Here, the highest share is from sorting LWP waste due to its high volume. Waste transportation only accounts for 8% of the total network CI.

Figure E.2 displays maps of the optimization result for both objectives individually and for both considered waste streams, including the existing German waste treatment infrastructure and the waste volume of LWP and APW. Arrows indicate the allocation of waste and intermediate products to facilities.

E.4.2 Model 2: Combining energy recovery and pyrolysis

Even though Model 2 allows pyrolysis plants for chemical recycling, no plants are placed in the cost-minimizing solution. Due to the high profits associated with energy recovery under current framework conditions, the waste is exclusively incinerated and not chemically recycled. The waste is exclusively delivered to RDF incineration plants taking advantage of their higher efficiency and higher revenues. The cost-minimizing solution of Model 2 is equal to the cost-minimizing solution of Model 1 (cf. Table 5, Appendix A and Figure E.3a and b).

Minimizing the CI of Model 2, it is noticeable that all waste is chemically recycled due to the lower CI associated with the pyrolysis process compared to incineration (cf. Table 6, Appendix A and Figure E.3). For this purpose, in the optimal case 58 pyrolysis plants are placed close to LWP sorting plants. The pyrolysis plants have an average utilization of 57%. The total network emissions are 1,503 kt CO_2e/year, of which 79% are associated with waste treatment, and 21% are related to waste transportation. This equals 39% of the baseline network (Model 1) and the cost-optimal network configuration of Model 2. So, shifting the network design and allowing for chemical recycling, 61% of the total CO_2 emissions could be saved, demonstrating the environmental contribution that chemical recycling can make. The annualized CO_2 emissions of constructing the pyrolysis plants are neglectable. Waste treatment emissions dominate the objective value and are mainly associated with LWP sorting (76%), while pyrolysis causes 24% of the waste treatment emissions.

Model 2 highlights the required trade-off between economic and environmental objectives. Under current framework conditions, energy recovery minimizes costs, while pyrolysis has environmental advantages. In order to consider competing economic and environmental objectives, an MODM model is employed in the following, minimizing them both simultaneously (cf. section E.4.3).

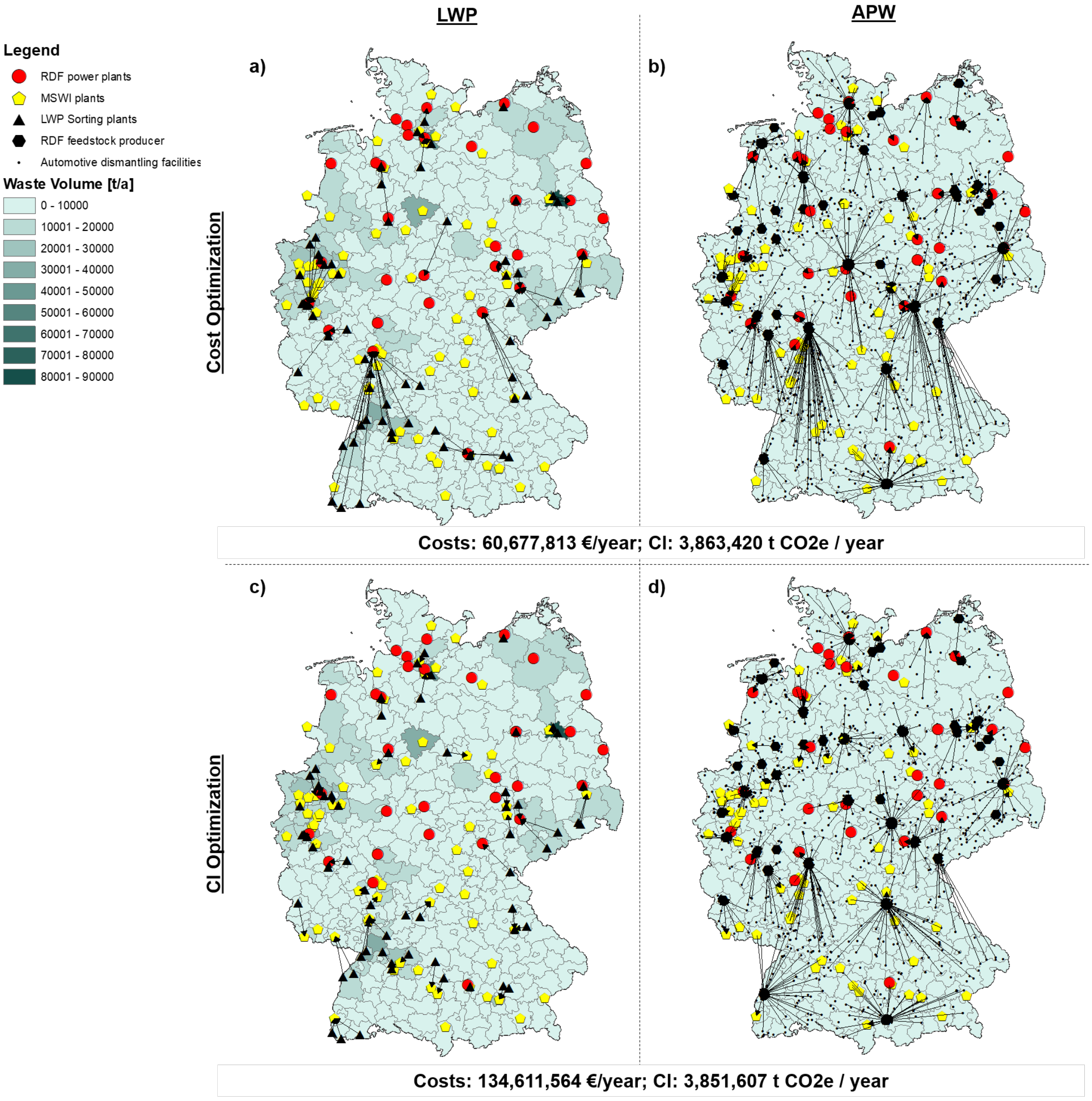

Figure E.2: Map of the Model 1 results for the existing waste network design in Germany employing energy recovery only considering the different waste streams and objectives: a) LWP sorting residues for minimizing costs, b) APW for minimizing costs, c) LWP sorting residues for minimizing CI, d) APW for minimizing CI.

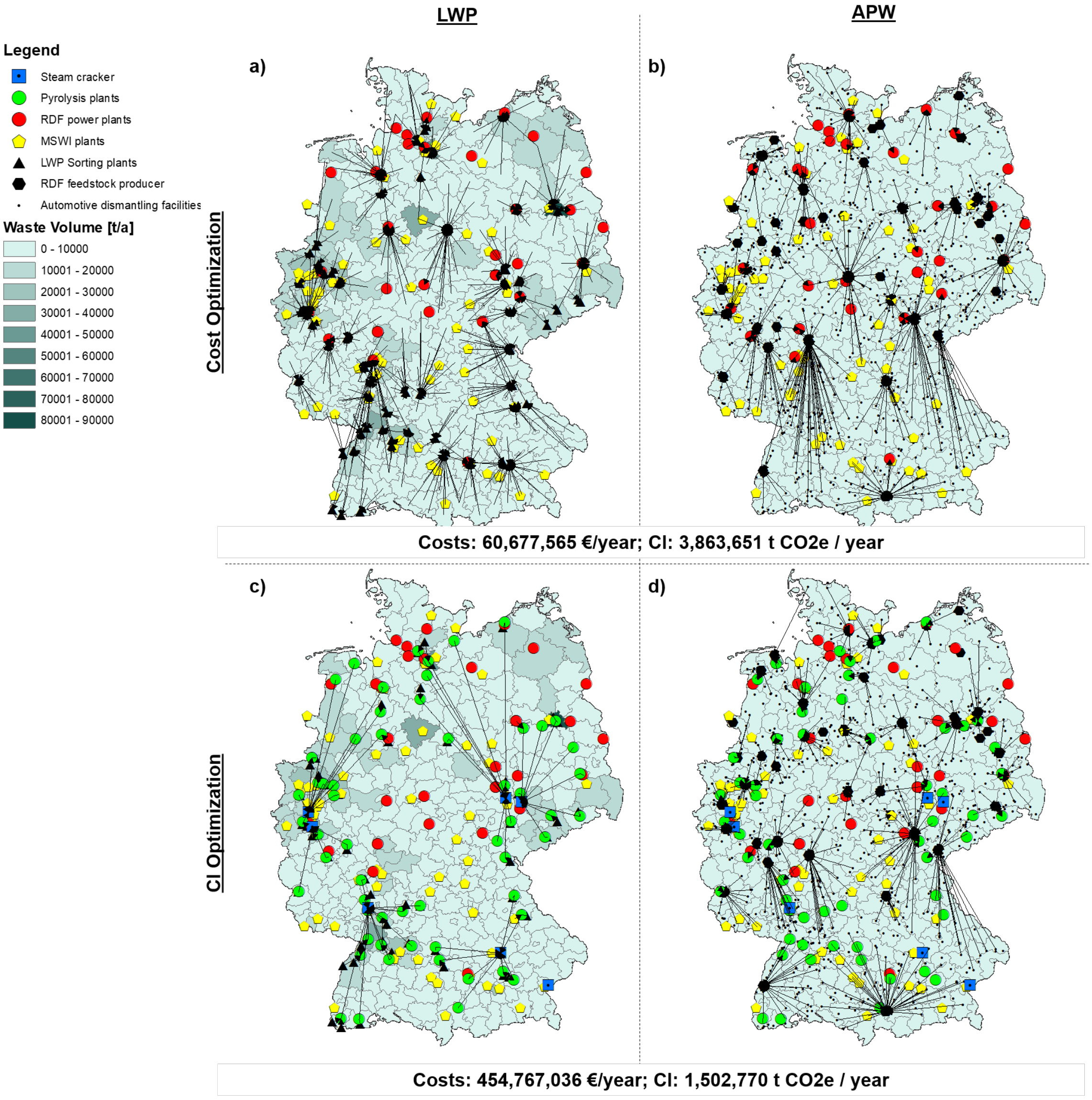

Figure E.3: Map of the Model 2 results for the new waste network design employing energy recovery and chemical recycling considering the different waste streams and objectives: a) LWP sorting residues for minimizing costs, b) APW for minimizing costs, c) LWP sorting residues for minimizing CI, d) APW for minimizing CI.

E.4.3 Model 3: Multi-objective approach optimizing cost and CI simultaneously

Model 3 considers both networking costs and CI simultaneously and determines a network design that minimizes the distance to the respective single-objective optimal solutions employing energy recovery and chemical recycling. In the optimal multi-objective network (cf. Figure E.4), 47% of the available waste feedstock is incinerated, and 53% is chemically recycled. Five pyrolysis plants with a capacity of 120,000 tons input/year are opened around the center of Germany and are fully utilized. Promising locations are in Peine, Borken, Heilbronn, Mannheim, and Weiden/Oberpfalz.

In 2021, the total costs of the waste handling system are 120 Mio. €. These are thereby approximately double the costs of the cost-minimizing solution of Model 1 and 2 (cf. Table 7, Appendix A). The annualized investment in the placed pyrolysis plants is 50 Mio. € accounting for 42% of the total costs. The waste treatment costs are 25 Mio. €/year (21%) while generating revenues at the energy recovery and chemical recycling stage. The transportation costs amount to 45 Mio. €/year (37%).

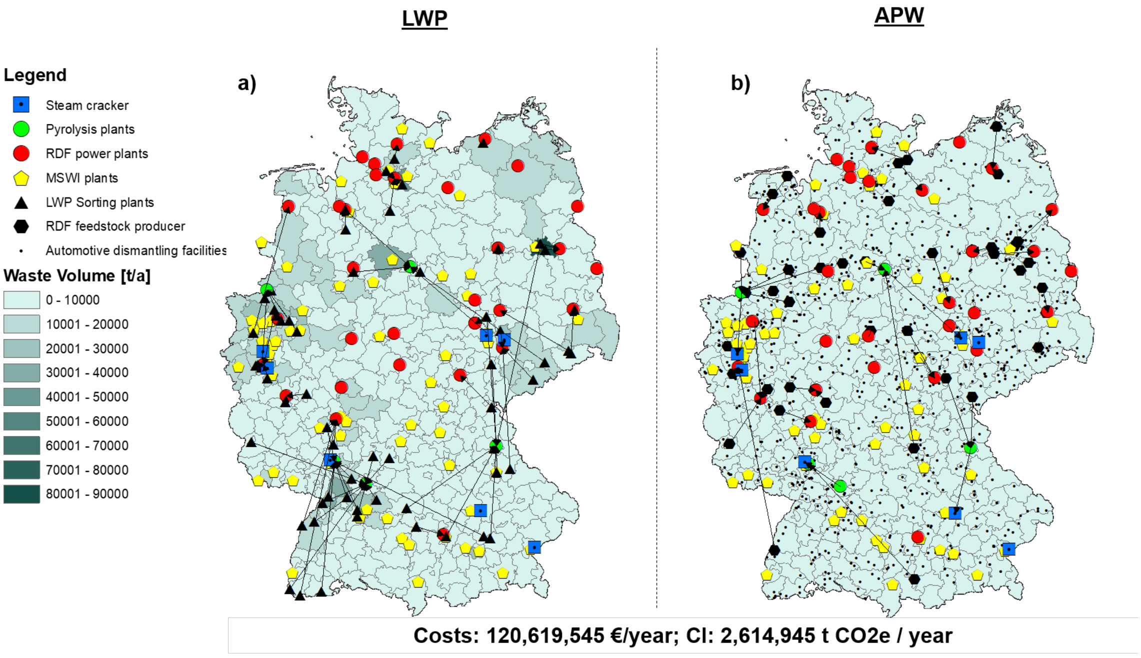

Figure E.4: Map of the model 3 multi-objective optimization results of the waste network design employing energy recovery and chemical recycling for a) LWP sorting residues and b) APW.

The total emissions of the optimum multi-objective solution amount to 2,615 kt CO_2e/year (cf. Table 7, Appendix A). The solution, thereby, deviates from the emission-minimal solution achieved in Model 2 by 74%. The construction of new pyrolysis plants only contributes 0.6 kt CO_2e/year and is, therefore, neglectable. Waste treatment emits 2,289 kt CO_2e/year (88%), of which the main part is associated with waste incineration. The transportation of waste and materials emits 326 kt CO_2e/year (12%).

By integrating the CI into the optimization of the waste management system, a balanced solution between economic and environmental objectives is found. However, this also results in higher costs for the waste treatment system. Based on the cost-optimal solution of the baseline scenario (Model 1) and combining the cost increase and the CI reduction, for every avoided ton of CO_2 emissions, the network costs increase by 48 €.

Nevertheless, the waste management system is not designed top-down by governmental bodies but is cost and technology-based. For this reason, the following section discusses political steering strategies to assert the environmental advantages of chemical recycling in practice.

E.4.4 Political steering strategies

In the following different political steering strategies are discussed to make the environmental advantages of pyrolysis over waste incineration also economically relevant, and thus aligning economic and environmental interests. The alignment also supports the development of a circular economy for plastics by promoting an additional recycling option. The assessed steering strategies include (1) an extension of the EU emission trading system (ETS) to the waste treatment sector and (2) increasing or implementing recycling rates for the assessed waste streams that are currently incinerated.

E.4.4.1 Extension EU ETS to the Waste Treatment Sector

The extension of the EU ETS to waste treatment does change the cost-optimal configuration of Model 2, assuming the average certificate price of 55 €/ton in 2021. In this case, the majority of the waste (96%) is chemically recycled due to the lower CO_2 emissions compared to energy recovery and, therefore, fewer CO_2 emission fees that have to be be paid. For chemical recycling, nine pyrolysis plants with an input capacity of 120,000 tons/year are built in the optimum network design, benefiting from economies of scale. The total costs associated with the waste treatment network increase by 207% resulting in costs of 187 Mio. €/year (cf. Table 8, Appendix A). The annualized investment in the placed pyrolysis plants is 90 Mio. € accounting for 48% of the total costs. The waste treatment costs are 32 Mio. €/year (17%) and are thereby 81% higher compared to the cost-efficient solution. This is due to the lower revenues associated with chemical recycling than energy recovery. Another additional cost driver is the CO_2 emissions fee accounting for 11% of the total network costs. The transportation costs do not change compared to the cost-efficient network configuration.

The network cost increase is associated with a 59% decrease in the CI of the network compared to the cost-efficient solution of Model 2. The total network emissions are 1,601 kt CO_2e/year. This is due to the lower emissions of chemical recycling compared to energy recovery and the high amount of chemically recycled waste. The CI associated with waste transportation remains the same compared to the cost-efficient solution, and the CI associated with placing the pyrolysis plants is neglectable. Combining the cost increase and the CI reduction, for every avoided ton of CO_2 emissions the network costs increase by 56 €.

With the historical maximum CO_2 certificate price of about 100 €/ton (reached in 2023), all waste is chemically recycled. The waste treatment network costs sum up to 208 Mio. €/year, which is an increase of 243% compared to the cost-efficient network configuration. Compared to the solution with an CO_2 emission fee of 55 €, an additional pyrolysis plant with an input of 120,000 tons/year is placed (cf. Table 9, Appendix A). Therefore, waste processing costs and annualized investment increase resulting in higher network costs. Additionally, the sum of CO_2 emission fees are higher due to the higher certificate price. The cost increase leads to a CI reduction of 61% compared to the cost-efficient solution of Model 2, matching the CI-minimizing solution of Model 2. The additional CI reduction is due to the total chemical recycling of the waste. The total network costs increase by 63 € for every ton of avoided CO_2 emissions.

Concluding, the extension of the EU ETS to waste treatment has a steering effect toward a lower environmental impact of the waste treatment system. The break-even price for the gross processing costs of energy recovery and pyrolysis regarding CO_2 certificates is 46 €/tons CO_2e. A higher certificate price leads to the economic competitiveness of pyrolysis plants for LWP sorting residues with 120,000 tons input/year capacity due to lower CO_2 emissions of waste handling.

E.4.4.2 Tightening of required recycling rates

Specifying a recycling rate (APW) or its increase (LWP) for the network design ensures that a defined amount of plastics is chemically recycled. In the assessed scenario, the recycling rate for LWP[16] was set to 55% to meet recycling rates demanded by the EU in 2030 (European Commission, 2018). For APW, a recycling target of 35% was assumed to stay within the technical limits of the assessed process.

With these more ambitious recycling targets, the total waste treatment system costs are 107 Mio. €/year, which is an increase of 76% compared with the cost-efficient solution of Model 2 (cf. Table 10, Appendix A). The waste processing costs increase by 36% due to the lower revenues associated with chemical recycling. The annualized investment for four placed pyrolysis plants with an input of 120,000 tons/year also increases the total network costs by 40 Mio. €. Transportation costs do not change with the tightened recycling rates.

Adjusting the recycling rates reduces the CO_2 emissions by 26% compared to the cost-efficient solution of Model 2 resulting in emissions of 2,859 kt CO_2e/year. The CI reduction is achieved by the lower CI of chemical recycling compared to energy recovery. Additional CO_2 emissions are neglectable, while transport emissions do not change. Combining costs increase and CI decrease, the network costs increase by 46 € for every ton of CO_2 emissions avoided.

The scenario analysis shows that the steering strategy of adjusted recycling rates generates a steering effect towards the environmentally advantageous EoL alternative. However, the steering strategy is based on the condition that there is legal certainty regarding the classification of chemical recycling as a recycling technology and a crediting of the treated waste to the legally binding recycling rates.

[16] Currently, 43% of LWP waste is mechanically recycled ((UBA, 2023)).

This classification would support the development of chemical recycling and, ultimately, a circular economy for plastics. However, in contrast to CO_2 emission fees, recycling rates are a regulatory measure that does not align the environmental and economic interests.

E.5 Discussion

The study results indicate which network designs and political steering strategies support decarbonizing plastic waste treatment from LWP sorting residues and APW by supporting chemical recycling. It can be differentiated between single and multi-objective considerations, alignment of environmental and economic objectives and setting more demanding recycling rates. In addition to the environmental assessment of the CI, the processing costs associated with the respective network configurations are considered. Consequently, the costs of decarbonizing plastic waste treatment and supporting a circular economy can be identified.

Despite the valuable insights gained from the developed models, the presented modeling has limitations. Currently, the models are static models that identify optimal decisions for the year 2021. However, the amount of waste in the waste streams considered, the composition, and the political framework can change over time, impacting the decision making for an optimal network design. Here a possible extension is dynamic modeling.

Additionally, the models are limited to comparing energy recovery and chemical recycling and focus on the material flows needed for this comparison. The treatment of by-products and waste streams is excluded. The model also excludes the hydrotreatment of pyrolysis oil, assuming that refineries with steam crackers have the needed equipment and infrastructure to upgrade the pyrolysis oil to make it a suitable cracker feedstock. Here, the potential quality issues of the pyrolysis oil are accounted for by choosing heavy fuel oil as a reference product. As a result of this model limitations, only a relative comparison of the waste handling methods and no overall statement regarding the waste treatment network can be made. Despite these limitations, the model is a sound basis for modeling extensions to describe a more comprehensive or even complete plastic waste treatment network.

The assessed waste streams can also be expanded to include plastics from other sectors for the network design, such plastics from as the construction sector or used electrical equipment. However, joint processing of waste streams in the considered pyrolysis plants is hypothetical and has yet to be technically tested. Should the technical feasibility of, e.g. batch operation be demonstrated, then the developed models are suitable as a starting point for more far-reaching modelling. Since two waste streams are already considered, the procedure for the extension by additional waste streams is already presented and can easily be carried out with the required data.

Individual constraints of the modeling can be very strict. Placing only one pyrolysis plant per district would be an example. However, the maximum capacity of the largest capacity class is sufficient to pyrolyze 9% of the feedstock from LWP sorting residues and APW in Germany in 2021.

Concerning the input data, it should be noted that the is data for Germany. However, this data can be replaced by values for other districts to employ the models in other settings.

In addition to the political steering strategies examined, other policy measures could be investigated. This includes the German Federal Climate Protection Act, which defines the permissible annual CO2 equivalents emissions for individual sectors, including the waste management sector (German Bundestag, 2019). The law regulates that the sector "waste management and others" may emit only 44% of the annual emissions in 2030 compared to 2020. Since the present waste management model is limited to the waste treatment of selected plastic waste, this political control strategy is not considered.

Despite these limitations, valuable insights can be gained for a circular economy for plastics and the decarbonization of plastic waste treatment. The models analyze and identify political steering strategies to support chemical recycling to support the closing of the plastic loop.

E.6 Conclusion

When considering the net costs of energy recovery and chemical recycling, a conflict of objectives arises between economically and environmentally favorable treatment options for plastic waste. Under Germany's current political, economic, and technical conditions, energy recovery is economically favorable, while chemical recycling has environmental advantages.

The conflict between economic and environmental objectives is reflected in the modeled waste treatment network for Germany. The cost-efficient solution is to send the plastic waste to energy recovery facilities, while the waste is chemically recycled to minimize the networks' CI.

In the multi-objective optimization, the balanced solution shows that pyrolysis plants with a high input capacity tend to be placed to take advantage of economies of scale. Here, a network is established that is associated with almost double the costs of the cost-efficient solution but also a 32% lower CI. Two different political steering strategies were investigated since top-down optimization is only a theoretical construct that legislation cannot impose. The assessed strategies show that the extension of the EU ETS to the waste treatment sector has a steering effect towards the environmentally more favorable waste treatment alternative. The all-time high emission fee would lead to a waste management system in which all waste is sent to chemical recycling. This steering effect starts at a CO_2 emission certificate price of 46 €/ton. Therefore the current price of over 80 €/ton already has a steering effect. Recycling rates higher than current rates also have a direct steering effect; corresponding waste quantities are chemically recycled to meet the rates. However, this requires legal certainty regarding the classification of chemical recycling as a recycling technology and a crediting of the treated waste to the recycling rates.

This study identifies opportunities for action by German policymakers in shaping the regulatory framework to realize the environmental benefits of chemical recycling over energy recovery. The alignment of economic and environmental interests and the associated support for chemical recycling is also beneficial to support the implementation of a circular economy for plastics.

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