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LIFE CYCLE ASSESSMENT STUDY ON PYROLYSIS OF POST-CONSUMER PLASTIC WASTE

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ABSTRACT

Plastic polymers have become an essential part of our lives and industries due to their low production cost, widespread application possibilities, and versatility. Driven by economic growth and development, plastic production has been increasing. With the increased production and plastic variety, challenges related to the disposal of plastics have developed. Alternatives to the established end-of-life solutions are investigated to lessen the impacts and manage the challenges of these technologies. One such alternative option for plastic waste management is chemical recycling via pyrolysis. This study uses life cycle assessment (LCA) to compare climate change impacts of pyrolysis, reporting only the global warming potential in kg CO_2-equivalent, with the widely practiced end-of-life solutions: mechanical recycling and incineration with energy recovery. Three scenarios were modeled using the GaBi software: incineration with energy recovery (Scenario 1), mechanical recycling (Scenario 2), and pyrolysis (Scenario 3). The pyrolysis temperatures considered in this study were 400°C, 500°C, and 600°C, to observe the effects of temperature on pyrolysis reaction and their impacts. Impacts were estimated using the IPCC AR5 GWP100, excluding biogenic carbon impact assessment method. A sensitivity analysis on mechanical recycling product substitution ratio was carried out. A 1:1 substitution ratio was assumed for the LCA and a 1:0,5 ratio for the sensitivity analysis. The total climate change impacts were 1125 kg CO_2-eq. for Scenario 1, -475 kg CO_2-eq. for Scenario 2, -238 kg CO_2-eq. for Scenario 3 at 400°C, -198 kg CO_2-eq. for Scenario 3 at 500°C and -159 kg CO_2-eq. for Scenario 3 at 600°C. Pyrolysis has higher climate change impacts compared to mechanical recycling but can recover more of the plastic waste stream. Sensitivity analysis showed that the climate change impacts of mechanical recycling depend on the recyclate quality, and its substitution ratio.

ABBREVIATIONS

CO_2	Carbon dioxide
ECS	Eddy current separator
HDPE	High-density polyethylene
LDPE	Low-density polyethylene
NaOH	Sodium hydroxide
NIR sensor	Near-infrared sensor
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride

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1. Introduction

Plastics as we know them, have been in our lives since the first polyvinyl chloride (PVC) was produced in 1907. Plastic products are low-cost, durable, light, and relatively easy to process. They can be molded into complex shapes, for various usage purposes. These facts, coupled with the fast development and modernization of society and industry, have resulted in the production of plastics vastly increasing. They have replaced glass, wood, metal, and ceramic in the production of consumer goods. Today, plastics are utilized from the construction sector to automotive, electronic, and agriculture sectors and used in products for packaging and leisure activities.

Despite its vast advantages, the disposal of plastics has become a problem. Especially with the demand increasing each year. Just the single-use facemasks used during the Covid-19 pandemic present a significant increase in the amount of plastic waste to be disposed of. It is unclear how much plastic waste is not collected each year as the lifetime of every produced plastic is not the same. For example, the lifetime of a yogurt cup is less than one year whereas automotive parts have a service life on average of 15 years. Therefore, it is difficult to compare produced and collected plastic waste year by year, but it is logical to assume there is a fraction of plastic waste that ends up directly in the environment and is not collected.

From the collected waste, 25% is landfilled (Plastics Europe, 2019). Plastics that are landfilled or not collected in the first place, degrade over time in the environment and end up in marine ecosystems. Microplastics as these are called, cannot be filtered with our current technologies as they are too small (less than 5 mm in length), pollute the rivers and oceans and all the living creatures depend on those water sources. The full effects of microplastics on the environment and health are still not known but research so far shows ingested particles accumulate in organisms, and microplastics reduce viability in the soil and penetrate deep into the ocean floor sediments. All these points to possible health and environmental problems in the future, caused by plastics seeping into our environment.

Plastic products have invaded the lives of humans in a wide range from industrial materials to daily consumption items, they have become vital to human life and our societies. Appropriate disposal technologies are needed to cope with the increasing use of plastic products. For the discussion of the sustainability of disposal technologies of plastic wastes, a life cycle assessment (LCA) study will be conducted. LCA methodology was developed to analyze scientific information about the environmental impacts of a process or product. Environmental impacts of selected plastic waste disposal technologies will be analyzed

1.1. Objectives

This study focuses on pyrolysis as an alternative end-of-life solution to the existing plastic recycling and disposal technologies currently in practice. However, pyrolysis is not widely used in practice for plastic waste management and the environmental impacts of using it as an end-of-life solution for plastic waste are not clear.

The objectives of this study are as follows:

(i)   To analyze the environmental impacts of chemical recycling via pyrolysis through life cycle thinking and to compare them to established mechanical recycling and incineration with energy recovery options.

(ii) To understand the pyrolysis and mechanical recycling technologies and their challenges by studying previous works in literature.

(iii) To identify the steps leading to the biggest emissions in chemical recycling via pyrolysis.

(iv) To compare the environmental impacts of different reaction temperatures for pyrolysis.

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2. Literature Review

2.1. Plastics and importance of suitable disposal methods

In 2018, 61.8 million tonnes of plastics[1] were produced in Europe and 359 million tonnes globally (Plastics Europe, 2019). This amount of production, coupled with the fact that most plastics are not biodegradable (which can stay in the environment for up to 400 years), results in a problem of plastic waste. It is estimated that from 1.15 to 2.41 Mt of plastic ends up in oceans every year (Plastics Europe, 2019). Plastics disintegrating in the sea cause microplastic contaminations. Microplastics are also found in freshwater resources due to the littering of riversides. Aquaculture, littering near water sources, shipping, and fishing activities are some of the causes of plastic contamination (Lebreton et al. 2017). A shift in the global trend of using single-use containers instead of reusable ones, coupled with the increase in the generation of global solid waste due to an increase of GDP in countries worldwide, increased the share of plastics in municipal solid waste from 1% to 10% in 45 years since 1960 (Geyer, 2016).

Plastics can be produced from fossil fuel derivatives, starch, cellulose, or soy. However, most of the plastics are produced from derivatives of fossil fuels, at present. These petroleum-based plastics are further divided into two as *thermoplastics* and *thermosets* (Worrell and Reuter 2014, p. 180). Thermoplastics do not experience a chemical change when reheated and can be remolded, whereas thermosets can only be molded once and once they are formed, the chemical reaction is irreversible. Almost 85% of plastic production is thermoplastics and of the thermoplastics, 70% are polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and polyvinyl chloride (PVC) (McDougall, pg.322, 2001). PE can be further divided into three as high-density polyethylene (HDPE), low-density polyethylene (LDPE), and linear low-density

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[1] Includes Thermoplastics, Polyurethanes, Thermosets, Elastomers, Adhesives, Coatings and Sealants and PP-Fibers. Not included: PET-fibers, PA-fibers and Polyacryl-fibers.

polyethylene (LLDPE). Polyurethane (PUR) is the most common thermoset used (Worrell and Reuter 2014, p. 180). **Table 1** shows a more comprehensive list. According to Worrell et al. (2014), PE, PP, PET, and PS are mostly used to produce packaging plastics, PVC, PUR, and PS are mostly used in the construction sector.

Thermoplastics	Thermosets
Polyethylene (high-density polyethylene (HDPE), Low-density polyethylene (LDPE), Linear low-density polyethylene (LLDPE)), Polypropylene (PP), Polyvinyl chloride (PVC), Polyethylene terephthalate (PET), Polystyrene (PS), Expanded polystyrene (EPS), Polyamides, Polycarbonate (PC), Polymethyl carbonate, Thermoplastic elastomers, Polysulfone, fluoropolymers, Acrylonitrile butadiene styrene, Ethylene-vinyl alcohol etc.	Polyurethane (PUR), Unsaturated polyesters, Silicone, Phenol-formaldehyde resins, Urea-formaldehyde resins, Epoxy resins, Melamine resin, Phenolic resins, Acrylic resins, Vinyl esters etc.

Table 1. Types of plastics are separated into two categories as thermoplastics, which can be remelted, and thermosets, which cannot be melted (PlasticsEurope, 2018).

Natural gas, coal, and crude oil are used to produce petroleum-based plastics (Plastics Europe, 2018). It is reported that annually, around 4% of the produced petroleum is used for producing plastics; with the addition of the energy required for the plastic production process itself, this consumption of fossil fuels rises around to 8% of the available petroleum resource (Hopewell et al. 2017). Moreover, we discard most of our yearly plastic production in the form of single-use plastics after a year of production. Recycling would reduce oil usage, CO_2 emissions, and the amount of waste that needs to be disposed of.

Additives are chemical substances added into a plastic to affect their properties. Additives can be used to improve mechanical properties (e.g., more resilient) or make them more heat or weather resistant. For recyclate to meet the technical requirements in the industry, additives such as talcum powder, calcium carbonate, titanium dioxide or antioxidants, and UV stabilizers are used (Gu et al., 2017). Types of additives that are used depend on the plastics that are produced, but almost all commercial plastics have some type of it (Deanin, 1975). The benefits of additives are obvious for manufacturers. However, those additives increase the difficulty of processing for mechanical recycling technologies. On average, it was found that PET, PE, PS, PP, PVC plastics (not including fibers) contain additives at least 7% by mass, in PE that can go up as high as 36% (Geyer et al., 2016).

Plastics can be petro-based polymers (petroplastics) or bio-based polymers (bioplastics), which can also be biodegradable. Currently, commonly used plastics are all petroplastic and an end-of-life solution that is not landfilling, is needed. Besides thermal treatments, there is no way of completely removing plastic waste. Permanent contamination of nature by plastics is a growing concern; Geyer et al. (2016) predict at least 4 million tonnes of plastic waste and synthetic fibers ended up in oceans in 2010 alone. This waste also threatens freshwater supplies and land habitats. As common as plastic waste has become, it is suggested in Geyer et al. (2016) to be used as a geological indicator of the era we live in, Anthropocene- the era of men.

According to a report by OECD (2018), plastic recycling is a much less energy-intensive, and thus less greenhouse gas-emitting method compared to the production of virgin plastics. A good quality waste management system prevents leakage of plastics into the environment and should be considered an investment for the future.

Plastic waste is separated into two as post-consumer and post-industrial. Post-industrial plastic waste streams are clean, homogenous, consistent in quality, and would usually be recycled at the site. Post-consumer plastic waste streams are contaminated with organic material, wood, glass, metals, etc., and are heterogeneous. Current end-of-life options for plastic waste include landfilling, incineration with or without energy recovery, mechanical recycling, and chemical recycling. Even in European countries, the average rate of plastic waste landfilling was 25% in 2018 (Plastics Europe, 2019). This percentage climbs higher in Asia and Africa. Incineration with energy recovery is a relatively good option with well-known and industrialized technologies; but since plastics are petroleum-based products, the path of incineration results in a need for new fossil fuel extraction. Especially in developing countries, plastic generation (and thus, waste) is increasing, whereas the waste management infrastructure is not extensive enough (Worrell and Reuters 2014, p. 3).

Plastics are made from different polymers and additives depending on the requirements of their intended task, which affects their recyclability. Some plastics cannot resist the mechanical or thermal stress of the recycling process. Properties and uses of some of the most common plastics are summarized in **Table 2** (Ritchie, 2018). The information is based on general guidelines and not specific local practices. There might be some variations depending on local waste management infrastructure.

Abb.	Polymer	Most Common Uses	Properties	Recyclability
PET	Polyethylene terephthalate	Plastic bottles	Strong and lightweight	Widely recycled
HDPE	High-density polyethylene	Chemical bottles (cleaning agents, shampoos, bleach)	Stiff, hard to breakdown	Widely recycled
PVC	Polyvinyl chloride	Plastic piping and flooring, cable insulation, window frames	Can be rigid or soft, depending on the additives	Generally not recycled, due to the chlorine it contains
LDPE	Low-density polyethylene	Plastic bags, food wrappings	Light, cheap and versatile; susceptible to mechanical force and heat	Not recycled, due to its low-stress tolerance
PP	Polypropylene	Bottle lids, food containers (not single-use), plastic furniture, automobile parts, medical equipment	Tough and long-lasting; does not react with chemicals or water	Depends on the local availability
PS	Polystyrene	Single-use food containers and cutlery	Light and structurally weak	Rarely recycled
OTHER	Other plastics	Fiberglass	Diverse, with various properties	Not recycled. Diversity of materials prevent proper sorting

Table 2. Most common plastics and their properties, usages, and recyclability by mechanical recycling technology are listed in the table (Ritchie, 2018).

As widespread the plastic usage is, unfortunately, problems related to their disposal are as well. Pollution is most visible in countries with weak or nonexistent waste collection schemes (Parker, 2019b). Plastic pollution has become such a global problem for

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countries, disregarding developed or non-developed, United Nations is attempting to form an international treaty regulating the production and use of single-use plastics by 2030 (Parker, 2019a). Although the discussions about a total "phasing-out" have failed so far, countries have managed to agree on "reducing" single-use plastics by 2030 (Parker, 2019a).

Packaging plastics are coded according to the polymer resin it is made from (**Table 2**). The main ones are LDPE (includes LLDPE), HDPE, PP, PS, PET, and PVC (Ragaert et al., 2017). Any material made from a polymer resin different from those six or a combination of those six is coded under "other". "Other" may include acrylonitrile butadiene styrene (ABS), polylactic acid (PLA), polycarbonate (PC), or multi-layer packaging. Anything recognized as "other", or PVC is sorted out from the feed and not recycled.

Practiced methods of dealing with disposed plastics are landfilling, incineration with or without energy recovery, mechanical recycling, and chemical recycling. According to a report by Boston Consulting Group (2019), 80% of all produced plastics are now in landfills or the environment, 12% is incinerated and the remaining 8% is either recycled or in-use.

Landfilling is still a popular choice in countries where space is available (BCG, 2019). The average annual rate of landfilling is 25% in EU28+NO/CH countries (Plastics Europe, 2018). Although this is more preferable than the leakage of plastics into the environment, depositing plastic waste in a landfill means permanent loss of that resource. Another problem with landfilling is the gradual seepage into the soil and water sources. Landfilling will not be included in the end-of-life options in this study. Instead, incineration with energy recovery will be taken as the base case.

Incineration with energy recovery is the conversion of material into heat, electricity, or fuel. Of the 29.1 Mt of post-consumer plastic waste collected in EU28+NO/CH countries, 43% is sent to waste-to-energy plants (Plastics Europe, 2018), making combustion the most widely applied recovery option. However, since combustion

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destroys valuable material, the search for alternative end-of-life options continues (Letcher, 2020, p. 502). It also releases dangerous toxins and dioxins into the atmosphere and so its emissions are highly regulated (Ragaert et al., 2017). Due to strict conditions on the feedstock for recycling, recycling might not be a viable option in every case, decreasing the end-of-life options to either landfilling or incineration. Between the two options, incineration can be the approach that is more ecological and economical, especially in underdeveloped countries (Letcher, 2020, p. 502).

Combustion of plastics requires emissions from the process to be under strict control. During incineration, harmful substances including volatile organic compounds, particulates, toxic metals, and dioxins may be released into the atmosphere (Letcher, 2020, p. 502). Our current technologies can deal with these problems and combined heat and power plants that are under strict regulations are considered safe for human health and the environment (Letcher, 2020, p. 503).

2.2. Mechanical Recycling

Mechanical recycling (or material recovery) is a method to recover the material of a disposed of product, in which the basic structure of the material is not changed throughout. In simple terms, mechanical recycling is the process of shredding, melting, and remolding plastic waste. Ideally, recycling feed should be homogenous and non-contaminated to produce high-quality recycled plastics. In this context, "homogenous" is used to describe a feed containing a single type of plastic (such as only PET or HDPE), and "contamination" refers to organic remains on the product (such as food remains). A recycling company constantly balances the economics of obtaining a high purity feed and the energy requirement of that. It is a well-known, economically viable, and industrialized method (Gu et al., 2017). Studies show that substituting virgin materials with recycled material allows the manufacturer to save up to 50% in costs (Gu et al., 2017). It is the method with the smallest environmental footprint currently available and is economically viable. Mechanical recycling has the disadvantage of not tolerating mixed streams or blended materials. The quality of the final product depends on the level

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of organic contamination of the feedstock, and materials cannot be recycled indefinitely due to loss of value.

Steps that can take place during mechanical recycling are called separation, grinding, washing, sorting, contaminant separation, granulation, and compounding (European bioplastics, 2016). Some of these steps might be skipped or occur multiple times depending on the composition of the waste fraction.

Petroplastics are sourced from fossil fuels, which increases humanity's dependency on the natural fossil fuel resources of Earth. Current mechanical recycling methods require a certain amount of virgin plastic to prevent a decrease in quality. As mechanical recycling and incineration with energy recovery end up decreasing the plastics available for reproduction, neither technology can eliminate the need for virgin plastics. Therefore, the raw material extraction and the burden on the environment continue. Also, incineration has the problem of high emissions of CO_2 and hazardous gases. There is a need for a technology that would allow plastics to be produced without virgin plastics and a solution for end-of-life processing. This need led to the research of chemical recycling technologies.

2.2.1. Importance and challenges

Mechanical recycling allows the recovery of material from an otherwise discarded product. The quality of the output depends on the purity and contamination of the feedstock; post-consumer plastic waste can be extremely contaminated and diverse. Sorting and washing technologies are needed to get high-quality plastic material. Otherwise, product value can be lowered, or products can even be directly landfilled.

The advantages of this technology are that it has a very small environmental impact and being able to substitute virgin (material that was never recycled) material lowers the environmental footprint even further. However, this technology requires strict conditions from the feedstock and does not tolerate contamination or multi-layering. Such materials either undergo multiple steps of washing and sorting or are rejected from recycling and treated via incineration with energy recovery. The substitution ratios of virgin material

can be dependent on the intended usage purposes of a product, the required mechanical properties (Rubel et al., 2019). Virgin plastics are also added in to prevent a drastic decrease in product quality or for lowering its costs. This is because the price of utilizing or manufacturing virgin plastics does not represent the external costs and virgin plastics remain competitive against recycled plastics due to market failures and policy misjudgments by the government (e.g. supporting hydrocarbon inputs to plastics production) (OECD, 2018).

Especially HDPE, PE, and PET are recycled with mechanical recycling. Low-value plastics or plastics that require multiple steps of treatment are not economical to recycle with this technology due to the resource-intensive process.

Post-consumer plastics wastes require sorting and cleaning before being utilized in mechanical recycling, which can be energy-intensive. Otherwise, the recyclate quality would be low and cannot be utilized in recycling, supported by adding virgin polymer or need to be combined with different polymers or compatibilizers to increase the durability and quality of the product. Materials formed with combinations of polymers or compatibilizers are very hard to mechanically recycle again and usually incinerated.

Manufacturers add different chemicals or dyes to their products, which complicates their treatment. They are hard to remove from a mixed waste fraction and require additional steps in the process, which increases the cost. It is also a problem that there is no single standard manufacturing process even for the same type of product (some materials, for example, ones that are in contact with food, have some standardization).

A preparatory separation process to purify the waste stream is needed especially, for municipal solid wastes. Dry recyclables are separated from the organic fraction of the waste. Then non-plastic impurities (such as labels and little pieces of metal or paper) are separated to improve source quality by using machine sorting technologies (Worrell and Reuter, 2014, p. 184). Next, plastics are sorted further into polymer types (PP, PS, HDPE, LDPE, PET, etc.). After these steps, manual sorting of each waste fraction to increase the purity of the final product is a common practice. Sorted streams are

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shredded, washed, and dried to prepare for extrusion, as a clean and homogenous fraction is best for mechanical recycling. Shredding also increases the bulk density of plastics and makes each trip of transportation more profitable (Gu et al., 2016). Whatever remains after sorting out and washing, is rejected from the process and they are deemed not fit to be recycled mechanically. These are either landfilled or incinerated.

Commonly used sorting technologies are eddy current separator, sink-float separator, drum separator, induction sorting, x-ray technology, and NIR (near-infrared) sensors. Sorting facilities use different combinations of such technologies to obtain the best capital or energy to purity rate. In theory, mechanical recycling needs 98% pure fractions to produce high-quality output. However, in practice 94-95% purity rate is already the maximum achievable amount (Worrell and Reuter, 2014, p. 184). This difference is based on the market requirements. A 98% purity rate and manageable market prices cannot both be accomplished. To achieve high purity rates, more energy and more capital are needed. After 95%, market prices cannot cover the initial investment and a monthly fee for energy (Worrell and Reuter, 2014, p. 184).

The availability of plastic recycling gives a false impression that plastics can be recycled without limit, when in fact the cycle of material recovery is not infinite. There is a limit to how many times a product can be recycled before it eventually loses some of its structural characteristics. This situation is called "downcycling" and happens due to the mechanical and thermal stress involved in mechanical recycling, and downcycled materials end up in landfills or are incinerated (Ritchie, 2018). To guarantee a certain threshold of quality to be met, recycled plastics are usually mixed with virgin plastics in the final product. Additives are another way of improving the properties of recyclate. However, the material becomes more difficult to process via mechanical recycling in the next cycle by adding additives (Geyer et al., 2016).

Geyer et al. (2016) are pessimistic in their view of mechanical recycling, describing mechanical recycling as a delaying mechanism for disposal instead of a solution. The study argues that for mechanical recycling to be a dependable solution, the end-products

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of mechanical recycling need to displace virgin plastic production and should be sustainable. Geyer et al. (2016) do not foresee such a sustainable mechanical recycling strategy, as with each recycling cycle, product quality decreases due to thermal stress.

2.2.2. Steps in mechanical recycling

Mechanical recycling companies try to obtain the highest quality pellet from their process by cleaning and sorting the feedstock. There are various technologies developed for lowering the process costs to improve the economic viability of mechanical recycling. There is no single recycling route followed globally. Steps are optimized for specific needs of feedstock and facilities. One or more of these can be applied multiple times depending on needs. Mechanical recycling steps can be summarized into collection, separation, sorting, grinding, washing. drying and reprocessing. Detailed information on each step can be found below.

Collection step is the collection of plastic waste to be utilized as a resource. The plastic fraction can be collected with other recyclables, such as in Belgium where only certain plastics are collected and mixed with metals and carton liquid packages, or as a single fraction for all plastic packing, such as in Netherlands and Denmark (Ragaert et al., 2017). According to Ragaert et al. (2017), if a country handles post-consumer plastic packaging separately, they are also likely to implement a separate system to collect PET bottles. When all packaging plastics are in a single stream, they are mixed with undesired polymers (especially non-packaging polymers such as ABS, polyamide, and polycarbonate) or organic material (paper or food remnants). In such cases separating the waste into its different fractions is necessary.

Sorting of the waste stream needs to be fast and accurate since a recycling business earns money through how much waste it processes. Product size, colorants, or coverings all affect the analysis. Therefore, there have been many technologies developed to improve this step. Not all sorting technologies need to be applied in a sorting facility. The ratio of the weight of the material that ends up in the product (e.g., Recycled PET) to the weight of the material fed into the sorter gives the efficiency percentage of that sorter.

One of the main issues is the dyes coating the materials. The quality of the recycled plastic can be compromised because of them, or the optical sorter may have a problem due to a dark color. Recyclers might be able to remove the coating through an abrasive method, but the separation of plastic particles from the dye is problematic (Al-Salem et al., 2009).

Sorting technologies rely on the density, color, solubility, and electrostatic charge of plastic materials (Rubel et al., 2019). For example, UV rays can assist during manual sorting to identify PVC from PET. Technologies are separated into two as wet and dry sorting; the ones that require a medium are classified as wet technologies.

The most precise sorting machines currently used utilize infrared light. A NIR-optical sorting machine has a sensor that sends or receives thermal patterns and then can determine the polymer type. Afterward, air jets remove the object from the conveyor (Letcher 2020, p. 150). Recycling facilities line up several NIR-sorting machines to separate different polymers. Since this technology relies on infrared light, conditions such as chemical structure or colorants can affect the sorting efficiency.

Automated sack opener cuts open trash bags. Contents are fed into a **progressive rotating sieve**, which sorts out the smallest and largest (over 220 mm) objects. This usually indicates bottle caps (PP) and the trash bags (LDPE) themselves. The fraction, now left with medium and large objects, is usually fed to a **wind sifter** to sort out any loose paper and plastic bags if there are any left. Next, the ferrous and non-ferrous metals and cartons are removed from the fraction. For the removal of ferrous metals, the fraction goes through an overhead **magnet**. Next, it goes through an optical sensor for the removal of cartons, and lastly an **eddy current separator (ECS)** for the removal of non-ferrous metals. Any plastic foils within the fraction are removed using a **ballistic separator**, leaving only hard plastics. (Ragaert et al., 2017)

Induction sorting technology locates and separates, with the assistance of air jets, different metals with sensors located under the conveyor. **Drum separator** separates materials relying on particle size. Particles with diameters smaller than the holes on the

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drum separator are extruded from the drum, whereas the ones with bigger diameters remain in the drum (Worrell and Reuter, 2014, p. 185).

Optical surface technologies such as FT-NIR (near-infrared thermography) and X-ray transmission imaging do not require a liquid medium and are well-known. **NIR Sensor** works by distinguishing different materials based on how they reflect light when illuminated. FT-NIR sensor separates PET and HDPE, but it does not work for multilayer packaging; it cannot identify products with dark colors or that are inside of each other or on top of each other. Optical color recognition sorters further separate the PET feed into different fractions according to color (feeds with additives and dyes need to be treated differently). **X-ray detection** is for PVC detection by taking advantage of the high chlorine content in PVC polymers (Ragaert et al. 2017).

Other dry sorting technologies include **melt-filtration,** which separates higher melting polymers from non-plastics such as wood, paper, sand, rubber, glass, and aluminum (Al-Salem et al. 2009); **hydrocyclone**, which separates according to weight differences and **tribo-electrostatic separation** which separates non-ferrous materials by colliding polymer flakes in a charged unit and running them through an electric field. To obtain the best results, this technology is better used mixed with a maximum of two polymers like ABS/PC, PET/PVC, and PP/PE (Ragaert et al., 2017).

Flotation, froth flotation and magnetic density separation technologies require a liquid medium to assist or complete the task. **Flotation (or float-sink separator)** separates by density differences; it usually uses water as a medium and is a relatively cheap technology. PP and PE (densities below 1 g/cm^3) float in water whereas PS, PET, PVC, ABS, and all other common polymers sink (Al-Salem et al., 2016). **Froth floatation** methods have air bubbles that adhere/not adhere to a polymer surface and cause it to float/sink (Ragaert et al., 2017). A magnetic fluid with iron oxide that changes density according to the magnetic field is used as a separation agent in **magnetic density separation** technology (Ragaert et al., 2017). The problem with these wet technologies

is how to further separate these fractions into single fractions since many of the polymer types do not have a single density value but have a possible range (Ragaert et al., 2017).

After separation and sorting, waste is **ground** into smaller pieces. The reason for grinding is to reduce the size of scrap, obtain more efficient transportation size and ease the storage of material. Shredders have rotating blades along connected to a motor and a grid, ending at a collection point (Worrell and Reuter, 2014, p. 185). There can be several steps of grinding throughout the mechanical recycling process. First grinding in which the waste is reduced to fist-size and then the second grinding after some steps (usually after washing and removal of non-polymers from the feed). The secondary grinding shreds fist-sized particles into 1-12 mm-sized flakes (Ragaert et al., 2017). Flakes being small allow a better washing and sorting efficiency due to higher surface area.

Post-consumer waste can be very contaminated, **washing** to remove contaminants is needed before any recycling. Washing is usually done with cold or hot water, which can be up to 60ºC (Worrell and Reuter, 2014, p. 185). A chemical washing can be applied if there is a paint coating on the material or a need to remove glue from plastic flakes. The wastewater resulting from this process can be treated and reused internally (Worrell and Reuter, 2014, p.185).

Washing is done multiple times throughout the mechanical recycling process. There would be a prewash (in a rotation drum wash) to separate rocks, metals, and glass after a coarse grinding; then a second washing (in friction washers) to remove organic contaminants, the third washing after a secondary grinding, and a final fourth washing in the float-sink separation installation (this is a sorting step but as it is a wet technology, fractions receive an automatic wash). (Ragaert et al., 2017)

After the washing steps are all completed, the stream goes through a mechanical drier to remove the remaining water. **Drying** continues until the fraction contains less than 0.1wt% moisture (Worrell and Reuter, 2014, p. 185).

Soft materials in this dried waste fraction are sorted with a wind sifter and **reprocessed**. The rest of the stream goes through melt-filtration first, since any material that does not

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melt that might have been left in this fraction would lower the quality of the final product. (Ragaert et al., 2017)

For plastic films, this step of reprocessing in recycling is called **agglomeration.** Plastic films are shredded, heated, and quickly cooled to form agglomerates. The agglomeration process is very energy-intensive (300-700 kWh/t) and most of the time avoided. (Worrell and Reuter 2014, p. 185)

Extrusion is the other technique for reprocessing, and it is more commonly used than agglomeration. It is a process used to produce pellets from virgin plastics and recycled plastics. Extruder mixes the two types of pellets under pressure and temperature, degasses, removes impurities, and cools and pelletizes (Worrell and Reuter, 2014, p. 185).

2.2.3. Practiced mechanical sorting examples

Ragaert et al. (2017) describe how a sorting facility in Belgium operates. In the sorting facility, waste first passes through a crude shredder to reduce the material size and form a homogenous stream. After that is the first washing in a rotating drum washer to remove rocks, glass, and metals and a secondary washing with friction washers to remove organic contaminants on the plastics. Then a second grinding further minimizes the size of particles down to flakes of 1-12 mm and the waste fraction is washed once more in the friction washer. Smaller particle size means more surface area and better washing efficiency. After this third washing, flakes are taken into the float-sink separation installation to be sorted for the first time. As mentioned above, float-sink separators use water as a medium, and thus particles with more than 1 g/cm^3 density sink, giving us a float fraction, and a sink fraction. To remove any remaining ferrous metals, a strong magnet is passed over the sink fraction. After dried by a mechanical drier, the sink fraction is ready as secondary raw material. The float fraction is dried by the same method, using a mechanical drier, but taken into a wind sifter to further separate lighter (soft) plastics from heavy (hard) plastics in the fraction. Softer particles (mostly foils made from LDPE and PP) fly up with an upward airflow in the wind sifter, whereas

heavier particles fall (remaining PP and HDPE). The hard fraction does not need any more processing and is ready as a secondary raw material, but the soft fraction has a low bulk density to be processed in converters, so it goes through a step of melt-filtration. This step removes any remaining contaminants that do not melt and would lower the quality of the final product.

Ragaert et al. (2017) also describe a case in which floating fraction (PP, HDPE, and LDPE) in the sink-float fraction is mechanically recycled whereas the sink fraction will be incinerated. The sink fraction, in this case, would have PET (bottles are separately collected), multilayer plastics, talc-filled PP, PS, PVC, small amounts of ABS, PMMA, PC, and PA. According to Ragaert et al. (2017), the float fraction equals only 16% of the total plastic waste mixture (there is also an additional 4% from fibers), whereas 69% of total plastics in the mixture (and the 8% metals) are incinerated.

Schonfield et al. (2008) provide several mechanical sorting strategies applied in the UK and analyze the impacts from each scenario. System does not include the collection of waste but assumes a mixed plastic feed was obtained and will be processed for disposal. After bags are shredded and waste is on the conveyor, the first step is to separate rigid plastics from plastic films. This is important for optical separators to work efficiently. Then a NIR sorter is used to sort plastics into fractions for PET, PVC, PE, and PP (PS can be sorted through NIR sensors. However, it is not utilized in recycling in this study and is therefore incinerated). Sorted fractions are shredded, cleaned, and extruded into profiles. Any loss from recycling along with residues (films, labels, fibers, unsorted plastics, and rejects) from sorting are either incinerated and/or landfilled. NIR sorters and density separators were both used in various scenarios in this study. According to Schonfield et al. (2008), density separators tend to have a higher sorting efficiency but are also less flexible and sometimes have high energy requirements. Therefore, it was concluded NIR sorters have a similar performance to density separation technologies.

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2.3. Chemical Recycling

Chemical recycling methods cause a change in the structure of the material and produce a liquid - oil. It is a relatively newer technology compared to previously mentioned ones. This technology changes the chemical composition of the material, hence the name. Produced liquid oil can be used for energy or as a monomer for polymerization depending on the technique and polymer. Chemical recycling is not widely industrialized, methods require large capital and waste stream to be profitable. In theory, depolymerization can convert mixed waste plastic streams into monomers, but there is still no depolymerization technology able to handle all types of plastic polymers. Chemical recycling methods that convert end-of-life plastics into fuel are less selective in the feedstock but cannot solve the raw material need for new plastic production as fuel is burned for energy. Technologies of chemical recycling still have many challenges, including but not limited to, economics. Some of the technologies invented that are under chemical recycling are chemolysis, catalytic cracking, gasification, and pyrolysis are few of them and the ones mentioned in this document. These technologies were selected to be mentioned in this document because of their relatively wider application prospects. There are other technologies in development kept as proprietary knowledge.

Chemical recycling is, despite being promising, still a relatively new technology with few industrial implementations. An LCA study, investigating environmental impacts and comparing them to existing technology would allow us to comment on prospects of chemical recycling.

The current interest regarding plastic waste is not to just recover energy or material but to have an innovative end-of-life solution that also preserves Earth's resources. The difference between chemical recycling from mechanical recycling or incineration with energy recovery is that it produces valuable monomers or petrochemical feedstock. The technologies in this segment can be separated into two based on their outputs as monomer recycling or plastic-to-fuel recycling (Rubel et al., 2019). Monomer recycling technologies specialize in the decomposition of a polymer back to its monomers.

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Aminolysis, glycolysis, hydrolysis, methanolysis, ammonolysis, and chemolysis methods are for chemically recycling PET. Feedstock recycling has greater flexibility over composition and is more tolerant to impurities than mechanical recycling, although it is capital intensive and requires very large quantities of used plastic for reprocessing to be economically viable (e.g., 50,000 tonnes per year) (Wong, 2010).
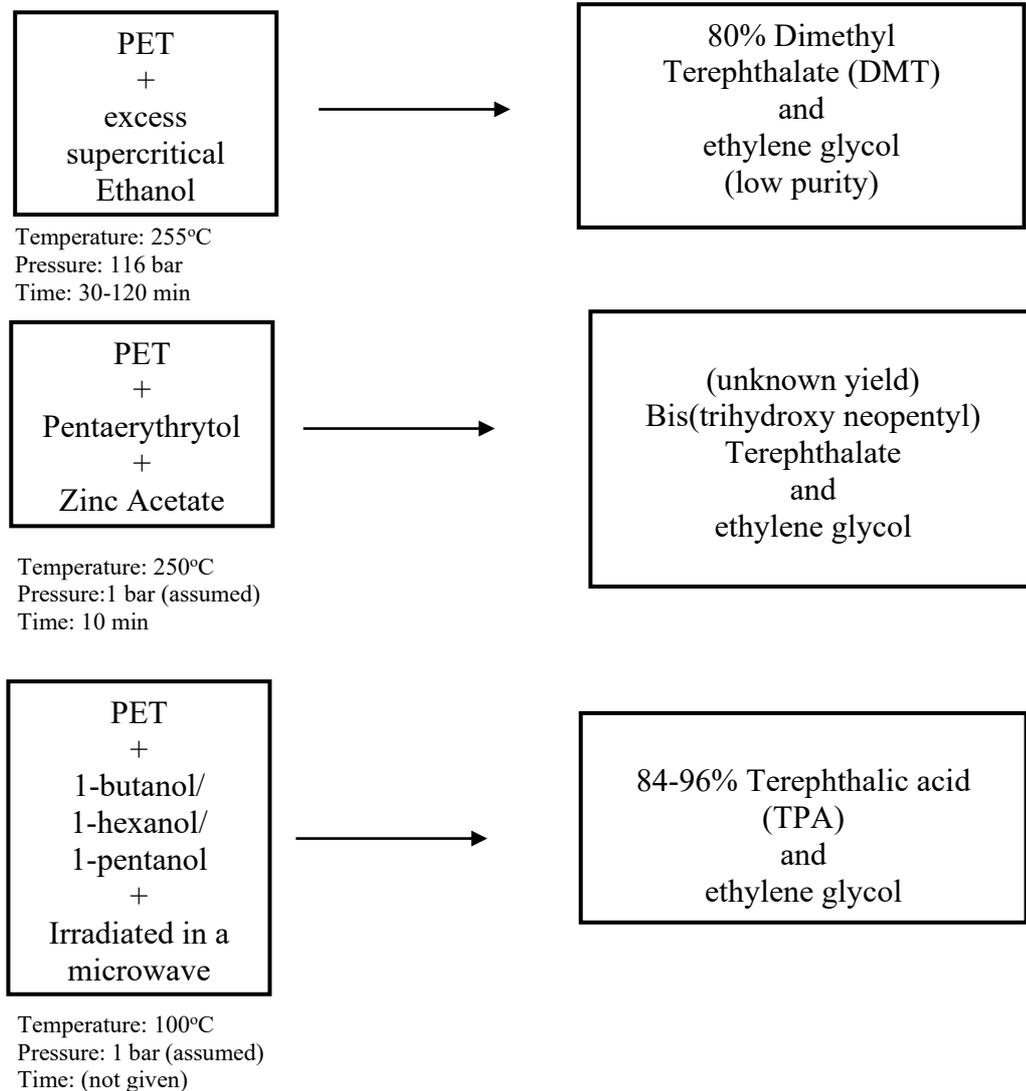
For chemical recycling methods to be common practice, they should be economically sustainable besides being environmentally sustainable; currently, chemically recycled polymers are still more expensive than virgin materials (Ragaert et al., 2017). The reason is the production of recycled monomer has many more steps that it passes through in its lifetime and needs large capital investment.

Plastic waste feeds such as mixed PE/PP/PS, multi-layered packaging, fiber-reinforced composites, etc. are not mechanically recycled and are difficult to depolymerize. These materials are traditionally either incinerated in combined heat and power plants for energy recovery or landfilled. A technology to allow us to recover the materials from such products is required in today's plastic waste management system.

2.3.1. Chemolysis of PET

PET can be completely depolymerized into its monomers or partially depolymerized into oligomers depending on the chosen depolymerization route. Possible monomer outputs from chemical recycling are terephthalic acid (TPA), dimethyl terephthalate (DMT), bis(hydroxyl ethylene) terephthalate (BHET), and ethylene glycol (EG) (Ragaert et al., 2017). The route of the reaction and outputs depend on the chemical agent used in the process. PET can be chemically depolymerized through alcoholysis, hydrolysis, methanolysis, ammonolysis, aminolysis, or glycolysis. Among these, the most common and commercially applied method of recycling PET is glycolysis (from PI scrap). These methods of chemical recycling all require either high pressure (hydrolysis and methanolysis) or chemicals (such as ethanol, methanol, sulfuric acid, or ethanolamine). Glycolysis requires precise control over the reaction to prevent further reactions between the end-products (Rageart et al., 2017).

Alcoholysis is the depolymerization of PET in the presence of excess alcohol to produce esters of terephthalic acid and ethylene glycol (Geyer B., 2016). The alcoholysis method was developed to avoid the problems of glycolysis (too much variation in the yield) and hydrolysis (releases pollutants) methods. Reaction products and yield amount differ greatly depending on the reagent used in the process and the conditions it was completed at. Some reaction examples from Geyer et al. (2016) of alcoholysis of PET can be found in **Figure 1.**

Temperature: 255°C
Pressure: 116 bar
Time: 30-120 min

Temperature: 250°C
Pressure: 1 bar (assumed)
Time: 10 min

Temperature: 100°C
Pressure: 1 bar (assumed)
Time: (not given)

Figure 1. Examples of alcoholysis reaction conditions of PET and the outputs in each case (Geyer et al., 2016).
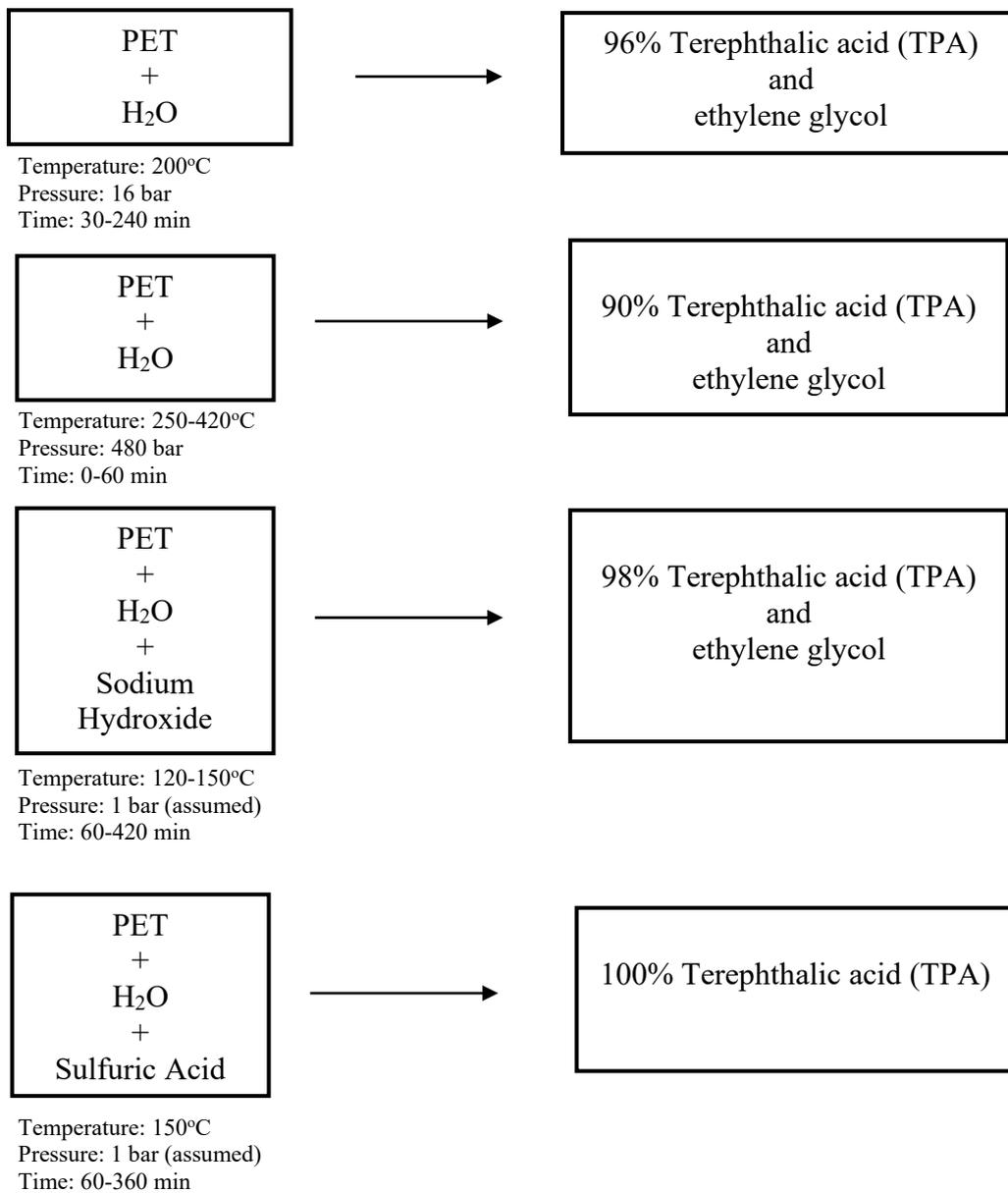
Figure 2. Examples of conditions and outputs from PET hydrolysis (Geyer et al., 2016).

Hydrolysis happens in the presence of excess neutral, acidic, or basic water concentrations between the temperature ranges of 115-420°C and under high pressure of, 10-480 bar (Ragaert et al. 2017; Geyer et al., 2016). Reaction times for this process are generally over 7 hours (Geyer et al., 2016). Monomer produced by hydrolysis is mainly terephthalic acid, with ethylene glycol as a byproduct **(Figure 2)**. Disadvantages

of hydrolysis are its low purity terephthalic acid output and the slow speed of the reaction.

Methanolysis can be classified as a type of alcoholysis. It has garnered special recognition due to its low price point and availability. It was observed that as the pressure increases, dimethyl terephthalate yield increases (from 80% to 98%) and reaction time decreases (Geyer et al., 2016). Generally, depolymerization occurs between the temperatures 160-350°C and pressure 9-20 bar (**Figure 3**).


```
┌──────────────┐
│     PET      │              98% Dimethyl
│      +       │   ────▶      Terephthalate (DMT),
│   Methanol   │              methyl-2-hydroxy ethylene
└──────────────┘              terephthalate (MHET)
Temperature: 300°C            and
Pressure: 147 bar             Ethylene Glycol
Time: 0-90 min

┌──────────────┐
│     PET      │              60% Dimethyl
│      +       │   ────▶      Terephthalate (DMT)
│   Methanol   │              and
└──────────────┘              Ethylene Glycol
Temperature: 300°C
Pressure: 9 bar
Time: 0-90 min
```
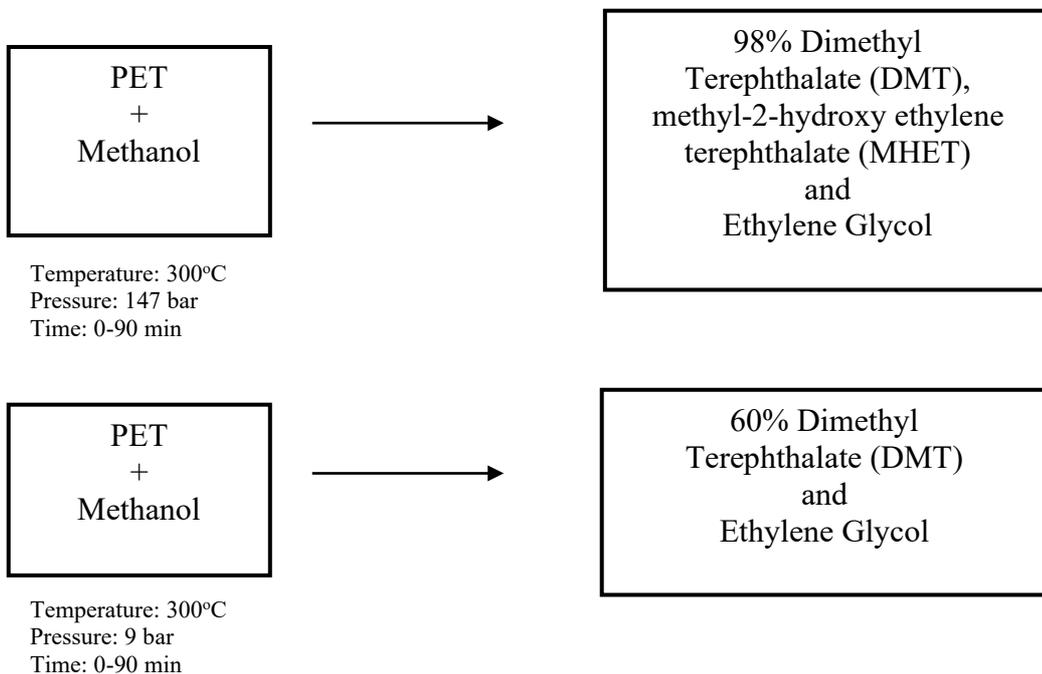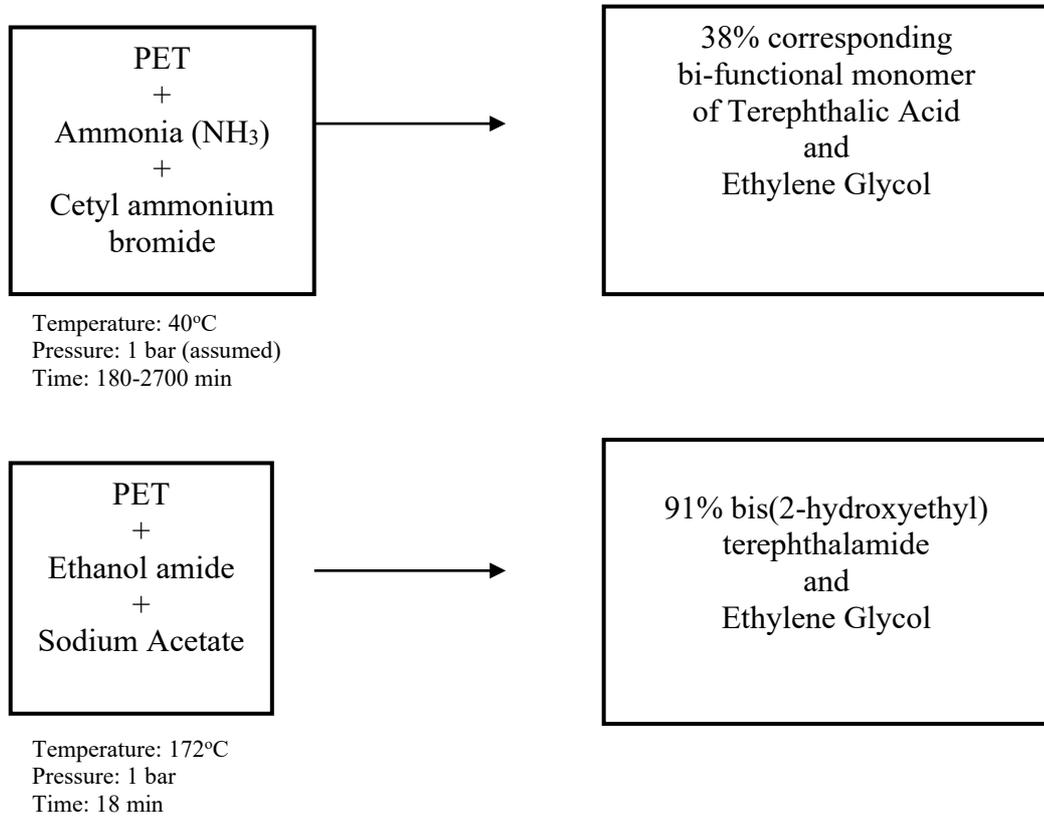
Figure 3. Examples of reaction conditions and outputs from PET methanolysis (Genta et al., 2005).

Ammonolysis and **Aminolysis** were developed to avoid the high temperature and pressure requirements of methanolysis and hydrolysis. Temperatures required for ammonolysis or aminolysis are between 25-190°C, pressure is generally low and reaction times have a wide range from a few hours to a few days (**Figure 4**). The yield is mainly monomeric amides of terephthalic acid such as bis(2-hydroxy-ethylene) terephthalamide, and some form of the catalyst is utilized to increase the yield. Products

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of these reactions can be used as plasticizers, epoxy curing agents, or polyurethane synthesis. (Geyer et al., 2016)

Figure 4. Examples of reaction conditions and outputs of PET ammonolysis and aminolysis (Geyer et al., 2016).

Glycolysis is the depolymerization of PET in the presence of ethylene glycol and sometimes a catalyst. It leads to the formation of PET-oligomers and monomers (Geyer B, 2016). Temperature ranges for this process are between 150-270°C and reaction times can be up to 15 hours, whereas the pressure has little effect on the process (Chen et al. 1991). The most common agent used in glycolysis is ethylene glycol. More detail on glycolysis reactions can be found in **Figure 5**.

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```
┌─────────────────────┐                          ┌─────────────────────────┐
│        PET          │                          │  98% Bis(hydroxyethyl)  │
│         +           │                          │   terephthalate (BHET)  │
│   Ethylene glycol   │  ──────────────────▶     │          and            │
│         +           │                          │        oligomers        │
│    Zinc Acetate     │                          │                         │
└─────────────────────┘                          └─────────────────────────┘
```

Temperature: 198°C
Pressure: 1 bar (assumed)
Time: 30-180 min

```
┌─────────────────────┐                          ┌─────────────────────────┐
│        PET          │                          │  98% Bis(hydroxyethyl)  │
│         +           │                          │   terephthalate (BHET)  │
│   Ethylene glycol   │  ──────────────────▶     │          and            │
│         +           │                          │        oligomers        │
│     Manganese       │                          │                         │
│      Acetate        │                          │                         │
└─────────────────────┘                          └─────────────────────────┘
```

Temperature: 198°C
Pressure: 1 bar (assumed)
Time: 30-180 min
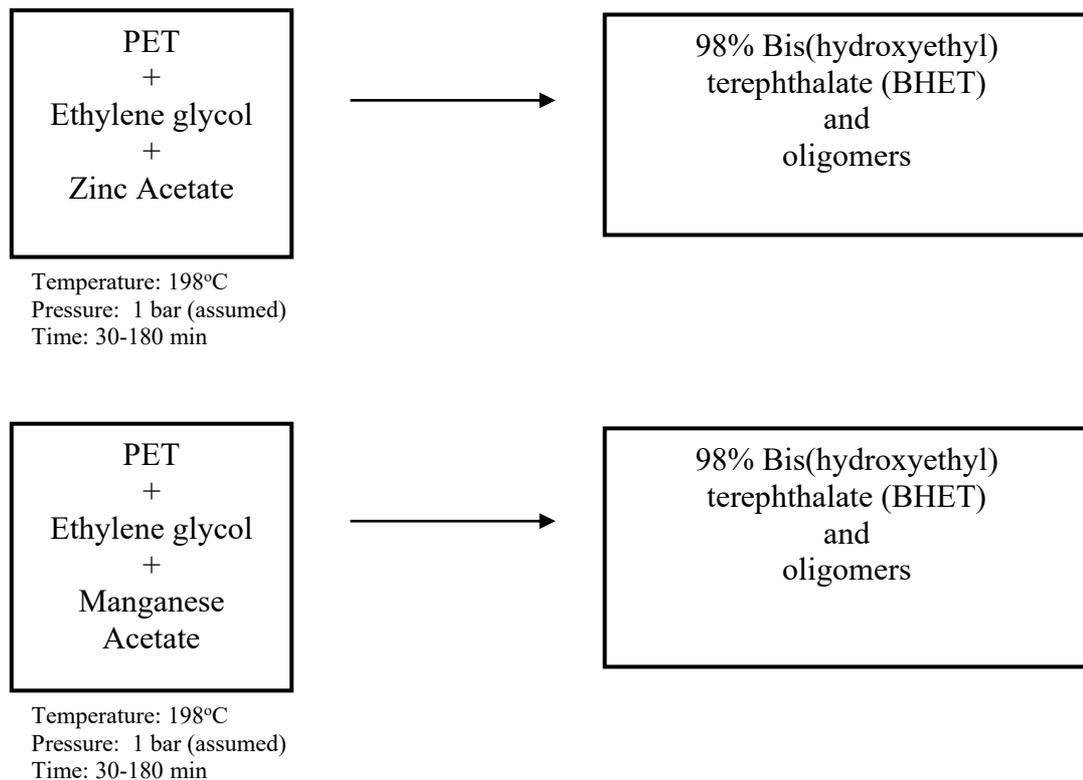
Figure 5. Examples of reaction conditions and outputs of PET glycolysis (Chen et al., 1991; Geyer et al., 2016).

2.3.2. *Plastic-to-Fuel Recycling*

Catalytic Cracking degrades materials in the presence of a catalyst and produces high-quality fuels, gaseous olefins, and compounds (hydrocarbons) to be used as raw material (Villanueva and Eder, 2014) Compared to pyrolysis, catalytic cracking has a lower degradation temperature (thus lower energy requirements), higher conversion rates and less variation in the output. The presence of a catalyst allows a selective degradation of plastic fuel, which results in lighter fuel fractions (Brems et al., 2012).

Gasification is the partial oxidization of organic material at high temperatures and produces synthetic gas (syngas). It is different from incineration as the oxidizing conditions are controlled and kept low. Syngas is a product that can replace fuel directly or after treatment. This technology might produce other gaseous compounds with a high

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molecular weight that lower the conversion rate, depending on the polymers in the plastic mixture, and as such, should be removed. This extra gaseous stream can be used to produce energy for the continuation of the gasification process or as fuel (Brems et al., 2012; Villanueva and Eder, 2012). Depending on the usage, the gasification process can be classified as recycling or energy recovery.

In a European Commission report by Villanueva and Eder in 2012, some problems related to this technology in Europe were summarized. As mentioned before, there is a possibility of producing compounds with high molecular weight. These compounds lower the yield, but they also cause blockage in the reactor pipes. Gasification has been known and used for more than 50 years, but the plants still have heavy legislative requirements. The only gasification plant in Finland reports having heavy permit costs and requirements for operation. Within Europe, only Germany and Austria have large-scale operations as of the European Commission report in 2012.

2.3.3. Pyrolysis

Pyrolysis (or thermolysis) is a thermo-chemical recycling technology for end-of-life plastic waste. Pyrolysis technology operates under the conditions of high heat and no oxygen and releases gas, liquid, and solid residue (char). Operating under oxygen-free conditions means there is no oxidation occurring. Therefore, pyrolysis has lower CO (carbon monoxide) and CO_2 (carbon dioxide) emissions compared to incineration (Rubel et al., 2019). The main benefit of this technology is its ability to handle mixed and/or contaminated streams of plastics. Multilayer films, mixed plastic (PE/PP/PS) streams, or products with dyes/additives which cannot be handled through mechanical recycling pose no problems in pyrolysis (Al-Salem et al., 2017). The main product of pyrolysis is liquid - oil, whereas the gases and char can be used internally as an energy source to sustain the pyrolysis reaction. The liquid oil can substitute diesel oil after treatment or can be used as raw material for plastic production (Al-Salem et al., 2017).

As an endothermic reaction, pyrolysis requires a source of energy source to start. However, after the first cycle, the reaction can be kept sustainable by utilizing the energy

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within the char and gaseous streams (Fivga et al., 2018). The reaction can occur between the range of 450-800ºC (Ragaert et al., 2017). Temperature directly affects the yield from the reaction (Oliveira, 2019).

Pyrolyzing plastics release the long polymer chains back to shorter hydrocarbons suitable for polymer or fuel production. This liquid is the main product of pyrolysis. Although the percentages differ depending on the input composition, reactor design, and conditions, on average pyrolysis yield is 5% char, 15% gas, and 80% liquid oil. Char and gas can be reused onsite, and liquid oil can be used as fuel after treatment or as raw material (Al-Salem et al., 2017).

The net calorific value of the pyrolysis oil depends on the feedstock composition and changes depending on that. However, it is generally above 40 MJ/kg which is very close to diesel's calorific value (43 MJ/kg). In an article by Fivga and Dimitrou (2018), pyrolysis oil was considered to have 44,6 MJ/kg net calorific value. Char has around 10 MJ/kg net calorific value (Jeswani et al., 2021) and a gaseous stream between 22-30 MJ/m^3 (Al-Salem et al., 2009). The energy from the gaseous stream can be utilized to cover the thermal energy needs of the system but electricity needs to be from the grid (Oliveira, 2018).

Emissions from pyrolysis reaction can be calculated per kg waste treated via pyrolysis. However, the total amount of emissions will be affected significantly by the product pyrolysis oil is used to substitute with (Jeswani et al., 2021). The main product of the pyrolysis reaction is the pyrolysis oil and it can be used in various ways depending on the quality. Different utilizations of the pyrolysis products lead to decreases in different environmental impacts. For example, substituting wax lowers climate change impact, light fuel oil lowers resource depletion, and heavy fuel oil lowers acidification potential (Jeswani et al., 2021).

Emissions to air from pyrolysis are usually similar to or slightly higher than mechanical recycling, with a good quality recyclate (Civancik-Uslu et al., 2021; Jeswani et al., 2021). LCA studies Jeswani et al. (2021) and Khoo (2019) predict total emission values

between 238-740 kg CO_2 eq. from pyrolysis. Compared to incineration with energy recovery, the climate change impact of pyrolysis was 50% less (Jeswani et al., 2021). This significant decrease in emissions of pyrolysis compared to incineration is a part of the reason for the interest in pyrolysis technology and chemical recycling (Jeswani et al., 2021).

According to a review by Al-Salem et al. (2017), fixed bed reactors, fluidized bed reactors, and rotary kilns are the most common reactor types used in pyrolysis studies. Reactor units affect the yield, polymer interactions, heat, and mass transfer. Fixed bed reactors are shown to have less complex emission systems compared to others, high efficiency, and lower corrosion in the reactor due to easier removal of ash (Al-Salem et al., 2017). They are easier to set up and operate and therefore the best option for pyrolysis according to Al-Salem et al. (2017). However, Butler et al. (2011) and Ragaert et al. (2017) support the usage of fluidized bed reactors as their yield is more uniform and provides higher conversion rates. From these articles, it can be concluded that between these three types of reactors, they each have their benefits and operators choose the one that fits their needs best.

Compared to mechanical recycling, which is a highly selective technology, pyrolysis can be simpler as pyrolysis reaction can handle most mixed and contaminated streams of feedstock (Al-Salem et al., 2009). For pyrolyzing municipal plastic waste, there is no need for separation, except for PVC and PET. These two types of plastics, plus steel, are not ideal to have in the stream and better be sorted out. PET produces a substance that turns solid in the reaction tank; whereas PVC is not recommended for pyrolysis since it produces hydrochloric acid, which is toxic and can result in acid rains, and has a very low yield of liquid oil (Sharuddin et al., 2018). According to Sharuddin et al. (2018), pyrolysis oil produced from feed that has PVC has chlorinated compounds, decreasing oil quality, and harming the environment. If PVC is included in the mix, chlorine removal by scrubbing is necessary before the gas can be combusted onsite (Oliveira, 2019).

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Al-Salem et al. (2009), Gu et al. (2017), and Lazarevic et al. (2010) are in agreement that obtaining high-quality recyclate via mechanical recycling is difficult because of its inability to handle mixed waste streams and pyrolysis presents a clear advantage on that point.   Compared to that, mechanical recycling has lower emissions and the infrastructure and regulations for it already exist. The establishment of an interconnected waste management system was mentioned as the next step in the development of waste management systems by Al-Salem et al. (2009), Gu et al. (2017), Lazarevic et al. (2010), and Ragaert et al. (2017). In such a system, pyrolysis would be used to treat plastic waste that cannot be mechanically recycled. For example, the plastic product could be heavily contaminated, have additives, have reached the limit of recycling cycles it can endure, or be a multilayer product.

3. Life cycle assessment

Life cycle assessment (LCA) methodology was used to analyze the impacts of chemical recycling processes. LCA is a scientific method, developed to analyze information on the environmental impacts of a product or process for a decision-making process(ISO 14040, 2006). Material flows related to a product are analyzed to define the environmental impacts of the whole system and to figure out which are most significant. A life cycle can generally be summarized in material procurement, manufacturing, transportation, use, and disposal phases. An LCA study can include some, or all these phases depending on its scope (ISO 14040, 2006).

For this LCA study, GaBi LCA modeling software was used, linked with the Professional + Extensions database linked. Results obtained were analyzed via IPCC AR5 GWP100, excl biogenic carbon impact assessment method.

3.1. Goal and scope

The goal of this study is to assess the environmental impacts of selected end-of-life practices for plastics. To compare different plastic waste treatment scenarios and identify what are the environmental benefits of pyrolysis technology compared to common practices. The main technologies included in this study are incineration with energy recovery, mechanical recycling, and pyrolysis as feedstock recycling technology. The study aims at analyzing the environmental impacts of these technologies focusing on the climate change impacts reporting only the global warming potential in kg CO_2-equivalent and discussing the observed benefits.

The study of each of these technologies would require different and very detailed observations depending on countries, regulations, desirable outputs, and available technologies. Therefore, some assumptions have been made in this study, which are listed in Chapter 3.3 and **Table 3**.

3.2. Functional Unit

The functional unit is a reference point for the quantifiable performance of a system/product. This analysis is for comparing the various end-of-life options for plastics and the functional unit chosen for this is the disposal of 1000 kg of plastics.

3.3. Scenarios and System Boundaries

The system boundary is vital for an LCA study. It determines the scope of the study and what functional unit to be used. The system boundary for this study starts at the waste treatment plant. The collection or transportation of the waste is not considered in any of the scenarios. These plastic wastes undergo a waste treatment process, incineration, mechanical recycling, or pyrolysis, depending on the scenario. The system boundary ends with the utilization of waste as a valuable material: fuel, raw material, or energy. Substitution of the products on the market with those obtained during plastics disposal was included.

The scenarios in this study are:

(i) Scenario 1: Plastic waste is treated via incineration with energy recovery.
(ii) Scenario 2: Plastic waste is mechanically recycled and rejects are treated via incineration with energy recovery.
(iii) Scenario 3: Solid plastic waste is recycled with pyrolysis. PET is mechanically recycled and PVC is incinerated with energy recovery.

In this study, Scenario 1 will form the baseline. In Scenario 2, the plastic waste stream is sorted and recycled, and rejected plastics are incinerated. Recycled plastics (recyclate) displace some of the virgin plastics and lower the need for raw material. In Scenario 3, plastic waste is processed with thermal pyrolysis to produce oil for external use and gas for internal use; liquid oil is used to substitute naphtha.

Types of plastics investigated in this study are the main packaging plastics (HDPE, LDPE, PET, PS, and PP). PVC is still occasionally used in packaging, but it is mostly

sorted out at recycling facilities due to economics and not recycled (Worrell and Reuter 2014, p. 304).

Scenario 1 is the incineration of all plastic waste without any separation or pretreatment. Thermal and electrical energy is recovered from the processed waste. This scenario is expected to have a high amount of CO_2 emissions since organic material is burned, which releases carbon into the atmosphere.

Scenario 2 is focused on recycling the plastic material mechanically as much as possible. The waste is separated and sorted as pretreatment and HDPE, LDPE, PP, PET, and PS are recycled. PVC is incinerated in a CHP plant. To achieve this, plastic waste was first processed in a drum feeder. Next, a vacuum removed all those bags that were now opened and the plastic films. A magnet and an ECS remove any steel or aluminum present in the stream so that finally, waste is shredded and separated through optical sorting. NIR sensors identify and separate the plastics that will be recycled later. Whatever remains is now rejected and incinerated. Plastics that will be recycled are washed, dried, and processed through extrusion. The recycled material is used to substitute virgin plastic. Due to lack of data, volatile organic compound emission from the extrusion process in the mechanical recycling is assumed to be zero.

In a real production facility, recycled plastics would be utilized by mixing with virgin material. The ratio of virgin plastic to recyclate depends on the type of plastic, the product being produced (a bottle may be turned into fiber in the next recycling cycle), and the required mechanical properties of said product. It is not a set ratio. Therefore, for easier handling, %100 virgin material substitution will be assumed in scenarios when any plastic is mechanically recycled.

In scenario 3, which is the pyrolysis scenario, HDPE, LDPE, PP, and PS were pyrolyzed whereas PET was mechanically recycled, and PVC was incinerated. As pretreatment, sacks are opened by a drum feeder, steel and aluminum are sorted out to be recycled and the plastic waste is shredded and homogenized. PET and PVC are sorted out by a NIR sensor and respectively recycled and incinerated. The recycling of PET is conducted with

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the same steps and data as in Scenario 2. After pyrolyzing the remaining waste in the pyrolysis tank, vapors and char are produced. These vapors pass through condenser(s) and condensable gases turn into liquid oil, later used in substituting naphtha. What remains after the condensation (non-condensable gases and char) is incinerated to produce thermal energy and used to sustain the reaction. In this LCA study, pyrolysis temperatures considered are 400°C, 500°C, and 600°C. Assumed yield fractions can be found in **Table 3.** As the pyrolysis temperature increases, liquid oil yield increases and gas yield decreases.

Fractions (right) Temperature (below)	Gas	Liquid oil	Char
400°C	20 %	75 %	5 %
500°C	15 %	80 %	5 %
600°C	10 %	85 %	5 %

Table 3. Fraction yields at different temperatures.

3.4. Inventory analysis

Inventory analysis is defined in ISO 14040 (2006) standard as follows: "phase of life cycle assessment involving the compilation and quantification of inputs and outputs for a product throughout its life cycle.". In simple terms, the LCI phase involves quantifying the energy and raw material requirements, emissions to air, water, and soil, and solid wastes from the process.

Scenario 1 was calculated using literature data readily available in GaBi and the inputs and outputs related to that can be found in **Table 4**. Inputs and outputs of scenarios 2 and 3 are listed below in **Table 5** and **Table 6**, respectively.

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Incineration		Comments
		Energy recovered
Solid waste (kg)	950	waste
Residue (kg)	50	
Thermal Energy (kWh)	2635	
Electricity (kWh)	1461	

Table 4. Inputs and outputs for Scenario 1. From 1 tonne of mixed plastic waste, all of it was incinerated to recover energy in a waste-to-energy facility.

In Scenario 2, electricity is required to operate the sorting machinery, and for washing and extrusion steps. Thermal energy is needed for washing, drying, and extrusion steps. Besides the energy requirements, a NaOH solution is needed for washing, lubricant and compressed air are required for extrusion. Wastewater from the washing step is assumed to be treated in a municipal wastewater treatment plant. Outputs are resins produced after recycling and solid waste from the process. Solid waste in this table includes all that was not recycled, rejects from the sorting steps, and PVC, which is not recycled. This fraction is then assumed to be recovered in a combined heat and power (CHP) plant.

Mechanical recycling		Comments
Electricity input (kWh)	557	
Thermal Energy input (kWh)	293	
2% NaOH solution (kg)	1670	For removing contaminants
Compressed air (Nm³)	12,9	
Lubricant (kg)	0,0758	For extrusion
Outputs		
Recycled HDPE resin (kg)	94,6	
Recycled LDPE resin (kg)	203	
Recycled PS resin (kg)	40,5	
Recycled PP resin (kg)	118	
Recycled PET resin (kg)	81	
Recycled Electricity output (kWh)	685	From incineration of
Recycled Thermal energy output (kWh)	1234	rejects and PVC

Table 5. Inputs and outputs for Scenario 2. From 1 tonne of mixed plastic waste, HDPE, LDPE, PET, PP, and PS were recycled; and PVC and rejected products were incinerated with energy recovery.

In Scenario 3, the recycling of PET and the pretreatment steps have all the same requirements of energy, water, and material as scenario 2. PVC is incinerated, the residue from pretreatment steps is as well. Additionally, the pyrolysis process needs electrical and thermal energy to depolymerize plastics and water for the condensation step. The outputs from pyrolysis are mainly pyrolysis oil, CO_2 emissions from incineration of pyrolysis gas, wastewater from condensation and washing steps, a small amount of recycled aluminum and steel, PET recyclate, and energy and emissions from incineration of PVC and residue. Wastewater is assumed to be treated in a municipal wastewater treatment plant. Outputs are resins produced after recycling and solid waste from the process. More data on inputs and outputs of scenario 3 can be found in **Table 6**.

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Pyrolysis		Comments
Electricity input (kWh)	58	Sourced from the grid
Thermal Energy input (kWh)	183	Sourced internally from pyrolysis gas
Plastic waste (kg)	694	Total pyrolyzed waste input
Water	302	Used for condensation of pyrolysis vapors
Outputs		
CO2 emmissions(kg)	104	
Aluminum (kg)	9,7	Recycled
Steel (kg)	9,8	Recycled
Thermal energy (kWh)	496	Leftover energy from pyrolysis gas incineration
Pyrolysis oil (kg)	554	Output, substituted for naphtha
Disposal of PET and PVC		
Electricity input (kWh)	76	
Thermal Energy input (kWh)	44	
Recycled PET resin (kg)	81	
2%NaOH solution (kg)	252	For washing PET
Compressed air (Nm³)	1,95	For extrusion of PET
Lubricant (kg)	0,0114	
Thermal energy output (kWh)	497	
Electricity output (kWh)	263	

Table 6. Inputs and outputs for Scenario 3 at 500°C. From 1 tonne of mixed plastic waste, HDPE, LDPE, PP, and PS were pyrolyzed, PET was recycled and PVC and rejects were incinerated.

In this LCA study, several unit processes were used from pre-existing processes within the GaBi database. Electricity was modeled using "EU-28: Electricity grid mix ts". Thermal energy was modeled using "EU-28: Thermal energy from natural gas ts".

Process water used in the study for washing and condensation steps was modeled using "EU-28: Process water ts". Incineration of waste with energy recovery in Scenario was modeled using "EU-28: Plastic packaging in municipal waste incineration plant ts <p-agg>". Incineration of rejected plastics in Scenario 2 and Scenario 3 was modeled using "EU-28: Plastic packaging in municipal waste incineration plant ts <p-agg>". Steel recycling was modeled using "GLO: Credit for recycling of steel scrap <LC>". Aluminum recycling was modeled using "EU-28: Aluminum recycling (2010) EAA <p-agg>" and "EU-28: Aluminium ingot mix ts". Wastewater treatment was modeled using "EU-28: Municipal waste water treatment (sludge treatment mix) (p-agg) ts <LC>". Extrusion of HDPE was modeled using "GLO: HDPE extrusion profile ts <u-so>". Extrusion of LDPE was modeled using "GLO: LDPE extrusion profile ts <u-so>". Extrusion of PET was modeled using "GLO: PET extrusion profile ts <u-so>". Extrusion of PP was modeled using "GLO: PP extrusion profile ts <u-so>". Extrusion of PS was modeled using "GLO: PS extrusion profile ts <u-so>". Virgin HDPE production was modeled using "RER: Polyethylene high density granulate (PE-HD) ELCD/PlasticsEurope <p-agg>". Virgin LDPE production was modeled using "RER: Polyethylene low density granulate (PE-LD) ELCD/PlasticsEurope <p-agg>". Virgin PET production was modeled using "RER: Polyethylene terephthalate granulate (PET, amorph) ELCD/PlasticsEurope <p-agg>". Virgin PS production was modeled using "DE: Polystyrene granulate (PS) ts". Virgin PP production was modeled using "RER: Polypropylene granulate (PP) ELCD/PlasticsEurope <p-agg>". Compressed air and lubricant required in the extrusion step were modeled using "GLO: Compressed air 7 bar (medium power consumption) ts <u-so>" and "EU-28: Lubricants at refinery ts".

3.5. Source material

In this study, secondary data from multiple sources were used. Below are charts containing information on the composition of the feedstock studied in the LCA, mechanical recycling, and pyrolysis steps.

In a European Commission report written by Villanueva and Eder (2014), household plastic waste compositions in European Union (the United Kingdom included) were analyzed. LDPE had the biggest share with 26%, followed by PP (16%), HDPE (13%), PET (11%), PVC (8%), and PS (6%). The remaining 20% were metals, fibers, and plastics coded under "other" in industry, usually rejected from recycling (**Figure 6**).
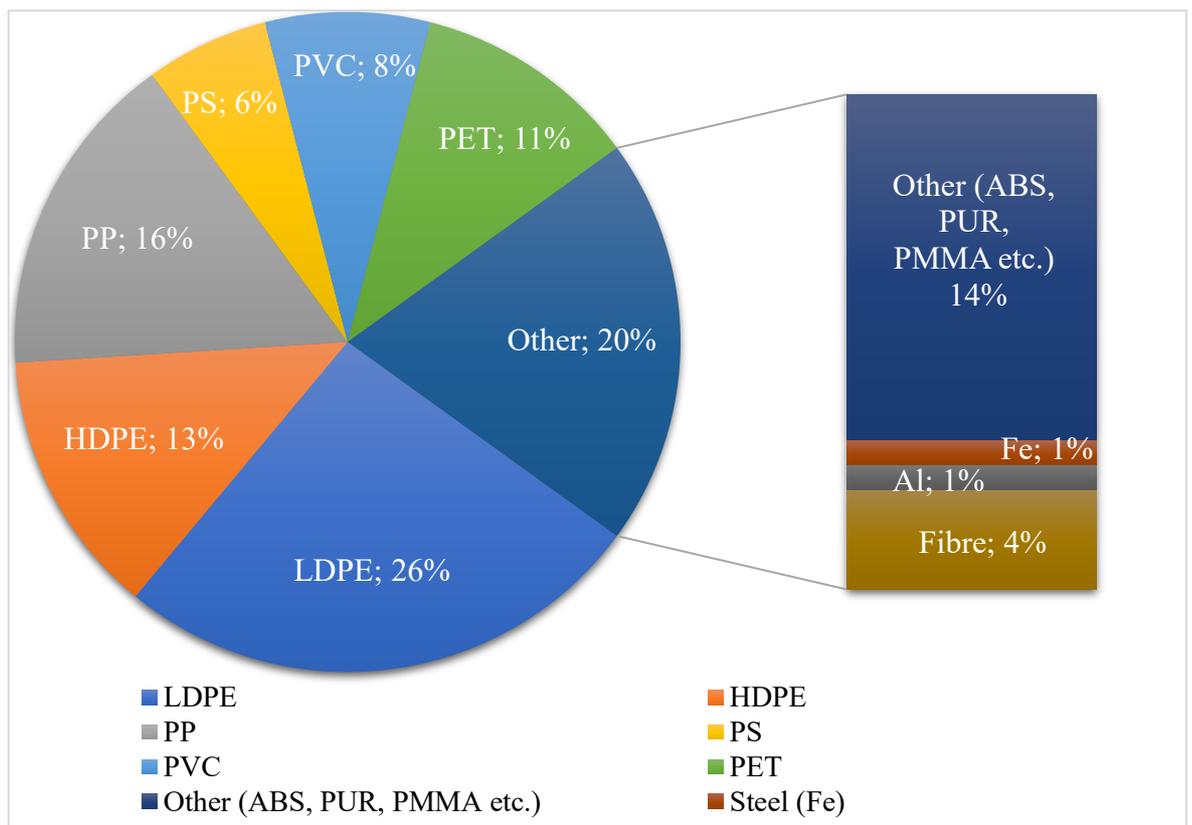
Figure 6. Composition of the feedstock in this life cycle assessment study (Villanueva and Eder, 2014).

Electricity from the grid is necessary for Scenario 2 and Scenario 3's mechanical recycling steps and for pyrolysis machinery to operate. These were calculated based on the data presented in **Table 7** and **Table 8**. **Table 7** presents the electricity demands for pretreatment steps of mechanical recycling. The system is assumed to be using a drum feeder, vacuum, magnet, ECS, NIR sensor, and shredder for the separation and sorting of plastic waste. Magnet and ECS are used for separating the ferrous and non-ferrous

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metals from the stream. They are energy-intensive machinery and although the separated waste is 1%, electricity consumed is significant. NIR Sensor is an optical separator, and it is assumed to have 70% efficiency in identifying plastics.

Sorting machinery	Drum feeder	Vacuum	ECS	Magnet	NIR Sensor	Shredder
Electricity (kWh/kg)	0,0025	0,003	0,0097	0,0118	0,012	0,024
Efficiency	100%	90%	97%	98%	70%	100%

Table 7. Electricity consumptions of sorting machinery for mechanical recycling per kg waste processed. (Schonfield, 2018; Pressley et al., 2015).

Thermal energy demands are sourced from the grid in recycling schemes in scenarios, besides the pyrolysis reaction, which is sourced internally from the byproducts of the pyrolysis reaction. The required energy is calculated according to the formula:

$$Q = m * C * \Delta T,$$

where Q is the required thermal energy for an object with a mass (of m) and specific heat capacity (of C) to increase its temperature by ΔT. Based on that, the energy required for a pyrolysis reaction to occur at 500°C was calculated. 0,264 kWh/kg energy is needed with HDPE, LDPE, PP, and PS being feedstock. For the 1 tonne of waste input, 694 kg sorted plastic waste is pyrolyzed (**Table 6**) and for the pyrolysis of that pretreated plastic waste, 183 kWh thermal energy is required.

Pyrolysis gas has a net calorific value of over 23 MJ/kg and pyrolysis char has 9,9 MJ/kg. Incineration of these covers the thermal energy demand of pyrolysis (Al-Salem et al., 2009; Jeswani et al., 2021). The energy provided by the incineration of pyrolysis gas and char is much higher than the required energy for continuing the pyrolysis reaction and surplus energy was fed into the grid. As this step is an incineration process, CO_2 is

released by burning this organic material (**Table 8**). The CO_2 emission data mentioned in Table 8 is calculated based on per kg feed pyrolyzed.

Required thermal energy for pyrolysis at 500°C temperature (kWh/kg)	0,264
Electricity demand of pyrolysis reactor (kWh/kg)	0,084
CO_2 released into the atmosphere from pyrolysis gas combustion per kg pyrolysis feed (kg)	0,15

Table 8. CO_2 emissions and energy demand for the pyrolysis process. Thermal energy demand was calculated within the model, based on the heat capacities of each plastic type (Fivga and Dimitrou, 2018; Khoo et al., 2019).

The main product of pyrolysis reaction, pyrolysis oil, has a net calorific value of 40,4 MJ/kg in this model (**Table 10**). This value was calculated within the model based on the mass-weighted average of each plastic type entering the pyrolysis reaction (**Table 9)**. This is a theoretical number as the plastics entering pyrolysis affect each other and yield fractions. Although the net calorific value of pyrolysis oil, in this case, is theoretical, it is an acceptable number within the known thresholds. This oil is used to substitute naphtha, a product of petroleum distillation, based on MJ/kg from each.

Pyrolysis gas and char are byproducts and were used to produce energy within the system. The net calorific value of pyrolysis gas is found in literature as 23 MJ/kg (Al-Salem et al., 2009). Char has a 9,9 MJ/kg net calorific value (Jeswani et al., 2021). Part of the energy produced from char and gas is used to sustain the pyrolysis reaction, the remaining energy is utilized outside of the facility.

Product	Calorific value (MJ/kg)	Calorific value (kWh/kg)
HDPE	40,5	11,3
LDPE	39,5	11,0
PET	28,2	7,8
PP	40,8	11,3
PS	43,0	11,9
PVC	21,1	5,9
Diesel	43,0	11,9

Table 9. Typical net calorific values of HDPE, LDPE, PET, PP, PS, and PVC are presented in the table (Sharuddin et al., 2018). For continuity, both MJ/kg and kWh/kg values are presented.

Pyrolysis Products	Liquid oil	Gas	Char
Net Calorific Value (MJ/kg)	40,4	23	9,9
Net Calorific Value (kWh/kg)	11,2	6,4	2,8

Table 10. Net calorific values of 3 streams of pyrolysis products are presented in the table. LHV for liquid oil was calculated within the model based on feedstock composition and LHV data for each type of plastic pyrolysis oil (Al-Salem et al., 2009; Jeswani et al., 2021).

3.6. Existing research and LCA data

The sorting step of mechanical recycling determines the economic viability of the process. If the waste stream requires many steps of sorting, energy demand, resource usage, and therefore costs would also increase. Pressley et al., (2015) investigated the electrical energy requirements and efficiency of sorting machinery used in mechanical recycling and identified which materials and machinery have a high global warming impact. HDPE was the most energy-intensive material to sort out of the mixed waste

stream. Another plastic-type, PET, required a third of the electricity compared to HDPE and had an average electricity consumption. Separation efficiency was shown to not affect energy consumption, meaning low-efficiency machinery consumes the same energy as high-efficiency machinery. In comparison to that, energy consumption changed heavily based on waste composition (more than 40% in one case). In conclusion, according to Pressley et al. (2015) sorting step of mechanical recycling is energy-intensive for plastic materials.

Gu et al. (2017) investigated the environmental impacts of mechanical recycling and compared different recyclate substitution ratios within the study. Considered substitution ratio for high-quality recyclate is 50%; whereas, for lower quality recyclate, substitution ratio can go down to 10%. The authors concluded that global warming impacts reductions of mechanical recycling depend on product (recyclate) substitution ratio, and the extrusion step has the highest greenhouse gas emissions among mechanical recycling processes.

Lazarevic et al. (2010) have findings concurring with Gu et al. (2017). Lazarevic et al. (2010) reviewed available LCA studies about end-of-life treatment options for plastic waste to identify if a specific technology was preferred as the best option available. They discovered that if the waste fraction is assumed to be a single stream with low organic contamination, resulting in recyclate substitution ratios close to 1:1, mechanical recycling is the environmentally preferred technology over chemical recycling or energy recovery. However, when these assumptions are altered, a clear plastic waste treatment hierarchy becomes difficult to define. The authors conclude by pointing out handling contamination is an important advantage of chemical recycling and the 1:1 recyclate substitution assumption lowers the impact of that.

Rigamonti et al. (2014) conduct a life cycle assessment study of plastic waste management with varying mechanical recycling percentages and energy recovery. The authors conclude that as the mechanical recycling rates increase, global warming potential steadily decreases. Although not as significant as global warming potential,

other impact categories reduce as well. Mechanical recycling lowers raw material extraction as well, as it decreases the virgin plastic used in production. The LCA study showed recycling has smaller emissions compared to incineration with energy recovery and from an environmental perspective, should be the preferred end-of-life solution over incineration with energy recovery (Rigamonti et al., 2014). However, mechanical recycling can be an energy-intensive and highly selective technology (Al-Salem et al., 2009). Al-Salem et al. (2009), analyzed the feedstock quality's effects on mechanical recycling efficiency. Their results show high-quality end-products are difficult to obtain through mechanical recycling with contaminated or mixed feedstock. Authors concur that mechanical recycling has low emissions, but a more sustainable recycling technology would be an even better option, and chemical recycling technologies are mentioned as a possible solution (Al-Salem et al., 2009).

Civancik-Uslu et al. (2021), focuses on plastic waste in Belgium and analyzes treatment options via mechanical recycling, chemical recycling via pyrolysis, and energy recovery. Pyrolysis products are used for the polymerization of new plastic products and mechanical recycling products are substituted for virgin plastic material on a 1:1 ratio. In this study, mechanical recycling has the smallest global warming impacts, followed by pyrolysis and energy recovery in such a system. Another regional article, but focused on the Singapore waste management system, Khoo (2019), argues that mechanical recycling is unable to efficiently handle the plastic waste produced in Singapore, from 822.200 tonnes of waste only 7% of the plastic waste is recycled. Alternative systems with varying process load each with a combination of mechanical recycling, pyrolysis, energy recovery, and gasification technologies were analyzed. In all scenarios, any waste that could not be treated via pyrolysis, mechanical recycling, or gasification was treated via energy recovery. Author of Khoo (2019) states they have obtained the highest reductions in climate change impact from the system with high pyrolysis and mechanical recycling ratio and no gasification technology. However, recycling technologies were utilized only in part in this article, dependent on the available infrastructure, and only 30% was recycled in the most progressive scenario. Further analysis of the potential

impacts of chemical recycling via pyrolysis is needed, to get a full picture for a sustainable system.

Jeswani et al. has conducted a similar life cycle assessment study, comparing the environmental impacts of pyrolysis with mechanical recycling and energy recovery (2021). They aimed to observe the environmental impacts of pyrolysis as a plastic waste management technology and compare it with established technologies and virgin plastic production. They have used two environmental impact assessment methods, environmental footprint, and ReCiPe, and considered waste management and production perspectives. The scenarios for pyrolysis or mechanical recycling utilized one or the other and supported each via energy recovery. After the LCA study, the authors of Jeswani et al. (2021) have observed pyrolysis has 50% less climate change impact compared to energy recovery and similar global warming potentials with mechanical recycling if the recyclate is of high quality. They have also noted that recyclate quality from mechanical recycling and pyrolysis oil conversion percentage to have significant effects on the results. Overall, mechanical recycling performs better in terms of climate change impact in Jeswani et al. (2021), by 7%. Considering the similar performances of pyrolysis and mechanical recycling and their specific advantages, the study concludes by suggesting these two technologies being supplementary to each other instead of competitors in treating municipal plastic waste. For any future research, the study recommends environmental impacts of pyrolysis be analyzed from a systems perspective, discussing the possible role of pyrolysis in such a system.

Among the studies comparing mechanical and chemical recycling methods, one of the most comprehensive is an article by Ragaert et al. (2017), in which most of the chemical recycling technologies and the machinery mechanical recycling is dependent on are discussed. Ragaert et al. (2017) introduces the main technologies under these recycling methodologies and discusses their advantages and disadvantages. After an analysis of available technologies, the authors concur with Jeswani et al. (2021) and reach a similar conclusion, that the plastic waste management problem cannot be solved by one single technology and requires an interconnected system. A system of technologies that will

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support each other and cover the shortcomings of another is required. The plastic waste disposal problem has multiple sides, and the solution should also have multiple layers to it, covering various needs. Chemical recycling is a necessary part of this system, as there are problems material recycling technology currently cannot solve.

4. Results

In this section, predicted emissions of the three scenarios will be shared and explained. This study will be focusing on global warming potential (kg CO_2 equivalent) as an impact category, and the impacts have been weighted using IPCC AR5 GWP100-excluding the biogenic carbon impact assessment method. **Figure 7** shows a comprehensive picture of the total emissions from each scenario. Scenario 3 has three different temperature points which are marked separately. These sub-scenarios have different yields depending on their heating temperature, information on these values can be found in **Table 3**.

Among the three scenarios, Scenario 1 has the highest emissions with a total of 1125 kg CO_2 eq. to air. Scenario 2 and 3 both have results below zero, meaning these scenarios lead to the reduction of some emissions in total. It can be seen from **Figure 7**, emissions released per tonne of processed waste are quite similar between the three temperature points of Scenario 3, pyrolysis at 400°C having the largest avoided emissions.

In figures from 8 to 12, scenarios are separated into steps for a clearer understanding of the source of emissions. These steps are pretreatment, washing and drying, extrusion, recyclate substitution, incineration, energy substitution, metal recycling, pyrolysis, and naphtha substitution. Besides these steps, the total amount of emissions from that scenario is also shown in the figures. The pretreatment step involves the sorting and shredding of the waste in preparation for either mechanical recycling or pyrolysis. Recyclate substitution refers to the avoided virgin plastic production and raw material extraction and the kg CO_2 eq. of those burdens. This step depends on the substitution ratio chosen, with the ratio 1:1 providing the most benefits. The incineration step, depending on the scenario, involves either the impacts from incinerating the total amount of waste, the rejects, or the rejects and pyrolysis gas. The energy substitution step corresponds to the emission reductions from the energy recovery from the incineration of either plastic waste, or plastic waste and pyrolysis gas. Metal recycling steps refer to the recycling of aluminum and steel separated from the feedstock. The pyrolysis step

includes the emissions and impacts from the pyrolysis reaction. Impact reductions of pyrolysis reaction products are included in the naphtha substitution step.
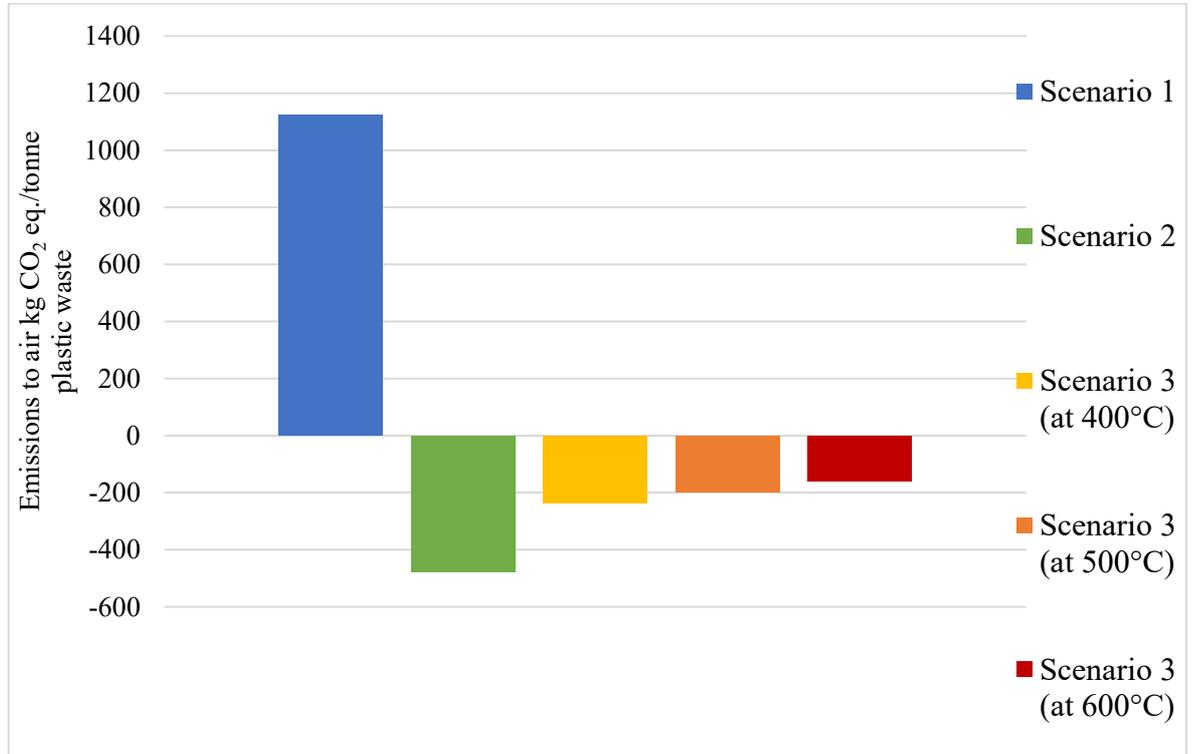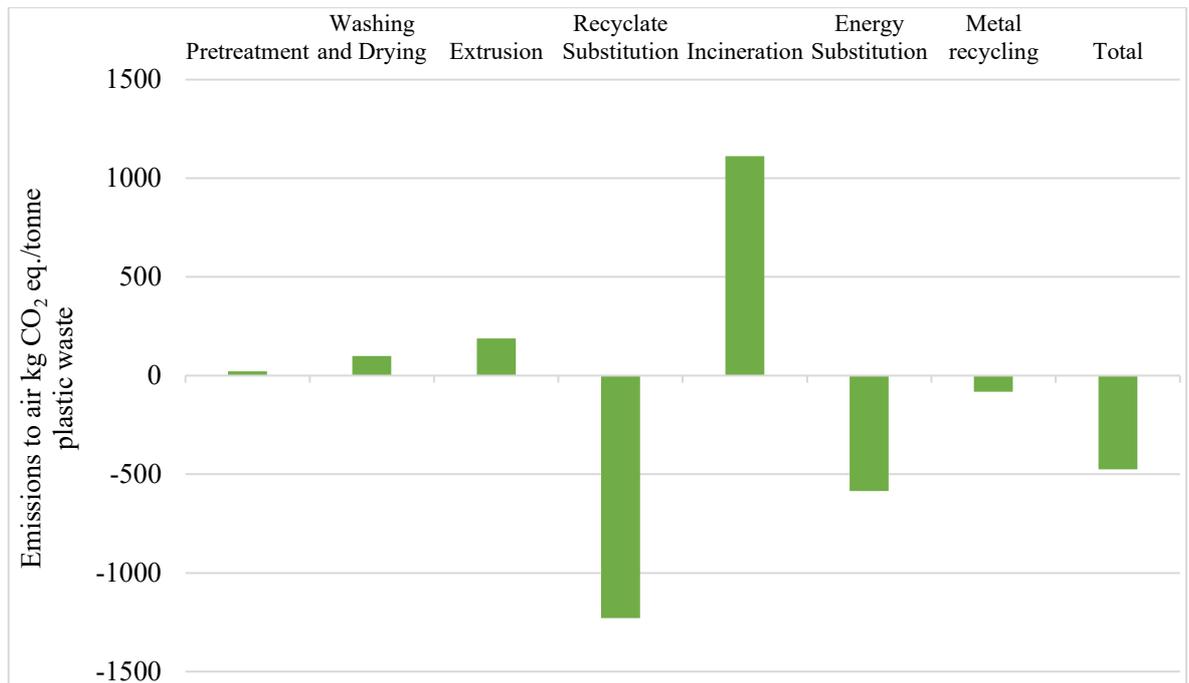

Figure 7. Emissions to air (kg CO_2 eq.) from Scenario 1 (incineration with energy recovery), Scenario 2 (mainly mechanical recycling), and Scenario 3 (mainly pyrolysis). Pyrolysis in Scenario 3 was conducted at 400°C, 500°C, and 600°C. Recycling products were assumed to be substituted 1:1 when it is included in the scenario.

In Scenario 1, all of the plastic waste is incinerated with energy recovery. Emissions from the incineration process (2373 kg CO_2 eq.) are calculated in the incineration step. Reductions from the energy recovered from the incineration process (-1247,75 kg CO_2 eq.) are shown in the energy substitution step. The total impact from incineration adds up to 1125 kg CO_2 eq., considering the emissions and reductions together.
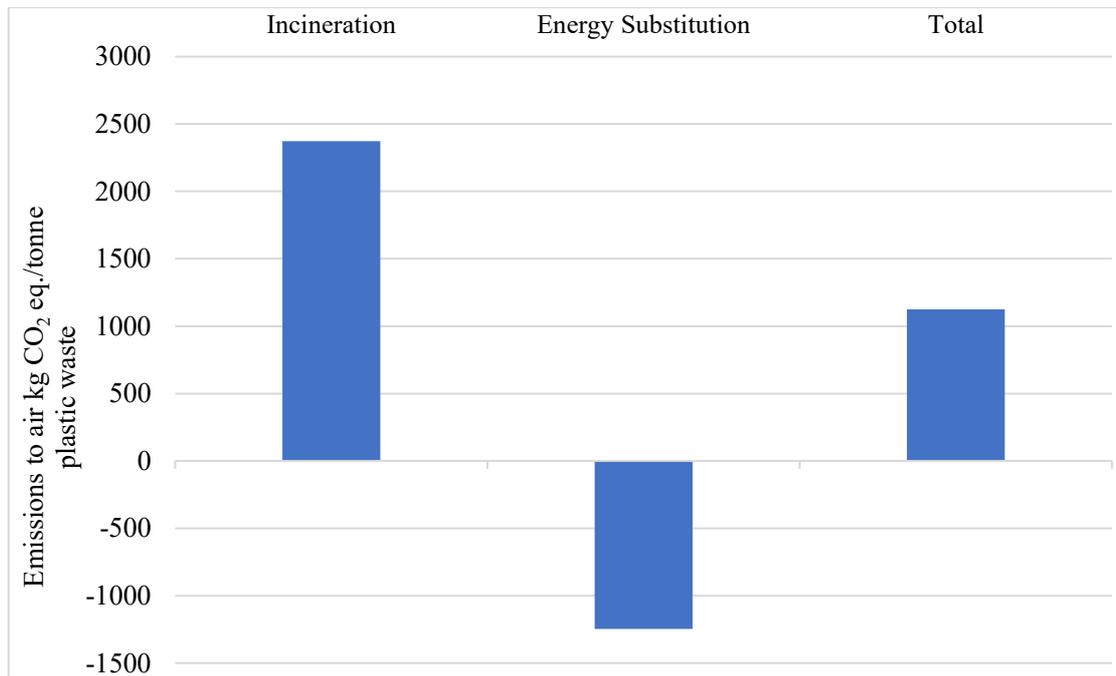
Figure 8. Emissions to air (kg CO_2 eq.) from incineration and energy recovery processes of Scenario 1.

For Scenario 2, the biggest share of emissions is from the incineration of rejected and non-recyclable plastics (**Figure 9**). Even though this is not a direct consequence of mechanical recycling, and it can even be argued mechanical recycling lowered the environmental burden from Scenario 1, it was caused because mechanical recycling was unable to handle almost 50% of feedstock (**Table 5)** and material recovery was not an option for those. Emissions avoided by recyclate substitution are substantial even with the technology constraints of mechanical recycling. It shows if 1:1 substitution of plastics was possible for all products; how much emissions could be avoided. The extrusion step has high CO_2 emissions as it is an energy-intensive process. Overall, Scenario 2 is an improvement over Scenario 1, from an environmental standpoint, as emissions were lowered.

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Figure 9. Emissions to air (kg CO_2 eq.) per tonne of post-consumer plastic waste from Scenario 2. Recycling products were substituted 1:1.

It can be seen in **Figure 10**, emissions from incineration have decreased nearly by 50% in Scenario 3 compared to Scenario 2. This is because less material is rejected from the feedstock of the pyrolysis reaction. Changes in the emissions of metal recycling and pretreatment steps are negligible. The total amount of avoided emissions in **Figure 10** compared to **Figure 9** is not because of the reduced burdens by the main technologies of these two scenarios. Reduced emissions by pyrolysis reaction itself are lower than the recyclate substitution of mechanical recycling. In **Figure 10**, reduced emissions on pyrolysis and recyclate substitution steps at 400°C amount to -435 kg CO_2 eq. whereas in Scenario 2, this number is -1230 kg CO_2 eq. for the recyclate substitution step. With the emissions from their energy substitution steps being similar, the difference between Scenario 2 and Scenario 3 is caused partly by washing and drying and extrusions steps, but mainly by the avoided emissions from incineration. These reductions are due to pyrolysis not needing washing and drying and extrusion and lowering the waste amount treated via incineration in Scenario 3.
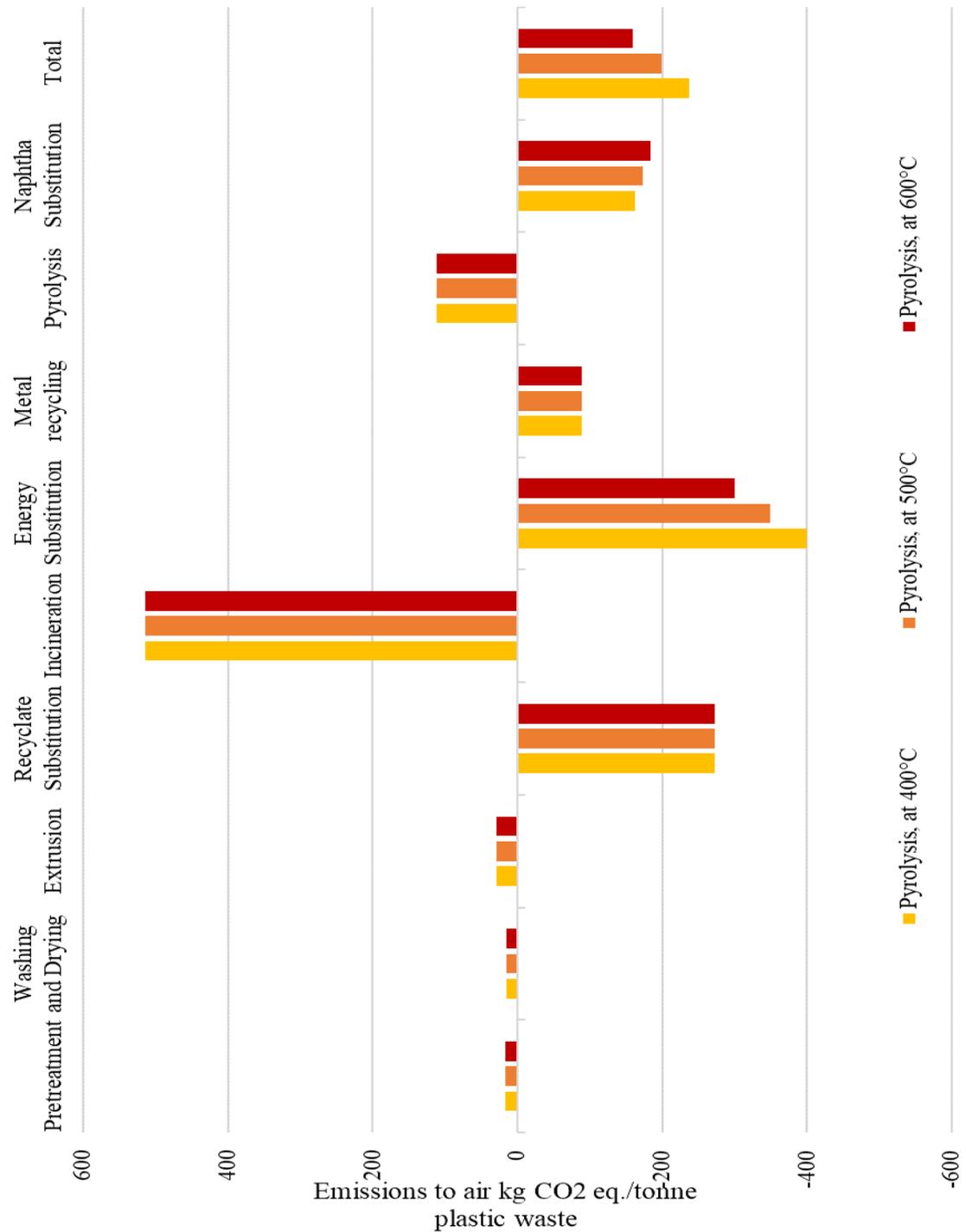
Figure 10. The figure shows kg CO_2 eq. of emissions to air per tonne of post-consumer plastic waste from Scenario 3 with pyrolysis reaction temperatures at 400°C, 500°C, and 600°C. Recycling products were substituted 1:1.

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The total amount of reduced emissions by pyrolysis is decreasing as the pyrolysis temperature increases, in **Figure 10**. With different reaction temperatures and yield fractions, only the emissions from the energy substitution step and naphtha substitution step are changing. As the temperature increases, emissions reduced by the energy substitution step decrease. This is to be expected as the amount of pyrolysis gas produced is decreasing as well. On the other hand, reduced emissions by naphtha production increased as the temperature increased. However, the increase in reduced emissions by naphtha substitution is less than the reduced emissions by energy substitution step, thus leading to the total reduction in emissions from pyrolysis to decrease. As it can be seen, Scenario 3 has higher emissions than Scenario 2 at every temperature point and these emissions change within Scenario 3 as the pyrolysis reaction temperature changes. Possible reasons for this change between pyrolysis temperatures will be discussed further in Chapter 5.

Reduced emissions from the recyclate substitution step in Scenario 2 are -1230 kg CO_2 eq. and -273 kg CO_2 eq. in Scenario 3 (the same for every temperature point). The reductions in Scenario 2 are from recycling HDPE, LDPE, PET, PP, and PS (total of 537 kg recycled waste) whereas, in Scenario 3, it is only PET (81 kg recycled waste). Despite that, Scenario 3 still has significant emission reductions from the recyclate substitution step.

5. Discussion

In this section, the results of the life cycle assessment study will be summarized and discussed to allow for conclusions to be drawn and recommendations to be given. Uncertain elements of the model will be checked by using life cycle assessment tools. Literature data will be used to judge the validity of the results where applicable.

To summarize the results from the LCA, Scenario 1 was identified as the biggest CO_2 eq. emitter, followed by Scenario 3 at 600°C, Scenario 3 at 500°C, Scenario 3 at 400°C, and mechanical recycling. In all the scenarios, the relative incineration step had the highest emissions. As the waste amount treated via incineration with energy recovery decreased, emissions from the incineration step decreased. Extrusion step is the next biggest emitter after incineration step for Scenario 2, with 188 kg CO_2 eq. but a fifth of that for Scenario 3. For Scenario 3, the pyrolysis step is the biggest CO_2 emitter after the incineration step. Extrusion in Scenario 3 releases a fifth of the emissions from Scenario 2. The pretreatment step has negligible emissions and is almost the same between Scenario 2 and Scenario 3. The washing and drying step emits 99 kg CO_2 eq. in Scenario 2, but 15 kg CO_2 eq. in Scenario 3. Recyclate substitution 1:1 leads to significant CO_2 emission reductions. Effects of recyclate substitution and the assumed 1:1 ratio of recyclate substitution on the emissions are discussed in further detail later. Emission reductions in Scenario 3 are less than Scenario 2 on all temperature points. Recyclate substitution step in mechanical recycling has significant emission reductions with a substitution ratio of 1:1. Emissions reduced from the energy substitution decrease and the naphtha substitution increase as the temperature of pyrolysis increases. The metal recycling step had a significant effect in decreasing the total emissions. The reason for these will also be discussed further in this chapter.

In Scenario 3, the biggest contributor to emissions was the incineration of PVC and rejected plastics. Energy, naphtha, and recyclate substitution steps reduce emissions and the total avoided emissions from Scenario 3 add up to be between 158-257 kg CO_2 eq. This range is due to the different pyrolysis temperatures being considered. The emissions

avoided on the recyclate substitution step are the same for all temperatures but on energy and naphtha substitution steps, there are changes. For the energy substitution step, emissions reduced at 400°C are 400 kg CO_2 eq., at 500°C they are 350 kg CO_2 eq., and at 600°C they are 300 kg CO_2 eq. The value of avoided emissions on the energy substitution step decreased as the reaction temperature increased. It was previously mentioned that gas and oil yields change as temperature changes. The yield of pyrolysis gas decreases with the increasing pyrolysis temperature, producing less energy via incineration. The opposite happens on the naphtha substitution step, reductions in emissions increase as the temperature increase. The yield of liquid oil from pyrolysis is higher at higher temperatures, increasing the amount substituted for naphtha. Among the different temperature points of Scenario 3, the scenario with the 600°C pyrolysis temperature has the highest emissions and the scenario at 400°C has the smallest emissions, indicating emissions reduced from pyrolysis gas incineration with energy recovery are higher than the pyrolysis oil substitution with naphtha.

The pretreatment step, in which the waste is separated and shredded, had a very small footprint. All the energy demands considered in this study and for the pretreatment step were sourced from works of literature. The energy demands considered could have been too low or a mistake could have been made during calculations as the emissions calculated were low. Both Scenarios 2 and 3 included some pretreatment steps. However, the steps required were much less for the pyrolysis technology, inevitably lowering the emissions from the pretreatment step for Scenario 3 even more. Pyrolysis-treated waste does need a pretreatment step but does not need washing or drying, leading to energy and water savings.

The amount of ferrous and non-ferrous metals to be recycled is only 2% of the total waste, but the metal recycling step shows a significant amount of emissions avoided. For steel scrap, reprocessing is simple and would save 15.8 GJ energy per tonne; considering the material loss, possible energy savings would be 12.9 GJ per tonne of recovered material (McDougall, pg. 442, 2001). Although the steel material present in the feedstock is low, it is significant enough. For per tonne of virgin aluminum produced, 182,8 GJ

energy is consumed, and 7640 kg of CO_2 is emitted; for recycling, energy consumption drops to 8,24 GJ/tonne aluminum produced, and 403 kg CO_2 is emitted (McDougall, pg.445, 2001). Therefore, it can be concluded that avoided emissions from the metal recycling step of this LCA study are within expectations.

Incineration was expected to have the highest emission results among the technologies included in this study. This expectation was due to the nature of combustion technology, organic materials burning in the presence of oxygen releases carbon dioxide. This step was used as a guideline scenario, to compare recycling technologies to the widely practiced energy recovery. Although Scenario 1 shows the highest CO_2 emissions among the three scenarios, it should not be written off as an end-of-life solution for plastic wastes. Each waste disposal scheme should be designed upon the needs of the community it serves and therefore has its place. Incineration with energy recovery can be preferred over landfilling as it disposes of the waste, instead of storing it somewhere, preventing chemical leaching, and if the energy provided by the grid is sourced from fossil fuels, incineration with energy recovery would be more sustainable. As a developed technology, solutions exist to cope with its problems, such as filters to prevent toxic gases from being released into the atmosphere.

Scenario 2 performs well, in the overall picture. It does not depend on landfilling, disposes of the same amount of waste as Scenario 1, but with fewer emissions. Around 50% of the material could be recovered, lowering the raw material burden of plastic production. Mechanical recycling has low emissions and recovers materials that might be otherwise incinerated during energy recovery. However, this scenario has limited reach as mechanical recycling is not a permanent solution to the waste problem for the reasons discussed in Chapter 2.2. Raw material demand can never be met entirely with the mechanical recycling technology we currently have, and even then, polymers cannot tolerate unlimited mechanical recycling cycles. Overall, Scenario 2 covers the immediate plastic waste disposal problem but does not provide a sustainable solution in the long term. If mechanical recycling cannot handle the majority of the plastic waste load single-handedly, it will always remain as support to another disposal technology.

Compared to previous scenarios, Scenario 3 emits and avoids less kg CO_2. With Scenario 2, recyclate substitution avoids a lot of CO_2 emissions by replacing raw material extraction but also releases a lot on the incineration step as well. Scenario 3 pyrolysis step has a smaller impact, but also the incineration step's emissions are lower. Washing and drying and extrusion steps have smaller emissions in Scenario 3. Pyrolysis decreases the processed waste through incineration, washing and drying, and extrusion steps and lowers emissions indirectly. At the current 1:1 recyclate substitution ratio, Scenario 2 performs better than Scenario 3 from a global warming potential impact standpoint. However, it should be mentioned that mechanical recycling was assumed to have 1:1 recyclate substitution, which is never achieved outside of a laboratory. At a lower recyclate substitution ratio, emissions from Scenario would be much higher, making Scenario 3 possibly better performing.

The difference between the recyclate substitution steps of Scenario 2 and Scenario 3 is not as high as expected. Scenario 2 treats 537 kg waste via mechanical recycling, whereas Scenario 3 treats only 81 kg via mechanical recycling. Despite the difference in the amount of treated waste, emissions reduced from the recyclate substitution step in Scenario 2 is -1230 kg CO_2 eq. and in Scenario 3 is -273 kg CO_2 eq. (the same for every temperature point). The reason for that difference is due to the types of plastics and the different emission values of their production. Emission reductions from PET recycling in Scenario 3 were the same as Scenario 2. In Scenario 2, recycling of HDPE reduced 188 kg CO_2 eq. in emissions, recycling of LDPE reduced 441 kg CO_2 eq. in emissions, recycling of PET reduced 272 kg CO_2 eq. in emissions, recycling of PP reduced 239 kg CO_2 eq. in emissions, and recycling of PS reduced 88 kg CO_2 eq. in emissions. That means recycling 1 kg of PET reduces 3,37 kg CO_2 eq., but recycling 1 kg of HDPE reduces 2 kg CO_2 eq. emissions.

Reprocessing is currently still an energy-intensive process, and extrusion is better than agglomeration for its lower emissions. The energy demand of this step cannot be lowered anymore unless a new technology is used. Incineration of waste was used for non-recyclable plastics in Scenario 2 and Scenario 3. Increasing the plastics recycled through

the main technologies in these scenarios is the only way to decrease these emissions, as that would decrease the waste processed via incineration with energy recovery. Sorting equipment with higher efficiencies for Scenario 2 could lower this incinerated waste by a small percentage. Treatment options for the pyrolysis oil could increase the amount of waste processed via pyrolysis.

Total reduced emissions kg CO_2 eq./kg plastic waste observed in this study was between 159-238 kg CO_2 eq. for pyrolysis scenarios. In a report published by CE Delft (2020) reductions in the total climate change impact of chemical recycling via pyrolysis were calculated as 200 kg CO_2 eq./tonne of plastic waste. The plastic waste composition was based on the Netherlands' waste generation statistics. The study was conducted using Dutch infrastructure data, meaning energy was sourced from the Dutch electricity mix. The estimated emissions by CE Delft (2020) are very close to the results from this study. However, CE Delft (2020) did not conduct any source separation in their study and pyrolyzed PET and PVC as well. There is no mention of any treatments considered for the pyrolysis oil. Therefore, it is reasonable for the emissions to be higher since PVC was incinerated in this study, and no emissions from the treatment of pyrolysis oil were considered in CE Delft's (2020) study. Another report by Schonfield (2008), in which pyrolysis was used to process PE, PP, and PS fractions, with PET and PVC being recycled, observed a reduction of 82 kg CO_2 eq. for global warming potential. The energy was sourced from the UK grid and collection of waste was not considered. Mechanically recycled waste was assumed to be substituted on a 1:1 ratio. Under similar conditions, both Schonfield (2008) and CE Delft (2020) reported emissions close to the results from this study.

Results in literature are affected by the differences in defined system boundaries, assumptions, and energy sources. Jeswani et al. (2021) ad Khoo (2019) are life cycle assessment studies investigating impacts of chemical recycling via pyrolysis as well, but these two articles have different system boundaries and assumptions. In Jeswani et al. (2021), collection, and transportation of waste and treatment of pyrolysis oil were all considered in system boundary, and emissions were calculated to be 238 kg CO_2 eq.

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Authors of Jeswani et al. (2021) indicate 35% of the emissions in their study are from collection and treatment steps. In addition to the changes to the system boundaries, mechanically recycled waste was assumed to be substituted on a 1:0,5 ratio in Jeswani et al. (2021). Khoo (2019) focuses on Singapore's waste management system. In that study, only 7% of the total plastic waste is treated via pyrolysis, 10% via mechanical recycling, and 83% via energy recovery, assuming a more conservative treatment capability based on infrastructure in Singapore. Emissions reported by Khoo (2019) under such conditions is 740 kg CO_2 eq. A direct comparison between Jeswani et al. (2021), Khoo (2019), and this study would not be very reliable. However, these results indicate emissions of pyrolysis can be much higher under wider system boundaries or less favorable conditions.

There were many outputs from Scenario 3 (**Table 6**), but the main product of Scenario 3 is pyrolysis oil. Net calorific values found in literature range between 40-44 MJ/kg (Fivga et al., 2018; Sharuddin et al., 2018) and higher heating value of between 41-47 MJ/kg (Miandad et al., 2017; Fivga et al.,2018). The net calorific value of the pyrolysis oil in this study was calculated as 40,4 MJ/kg, which is in line with the literature. This oil was used to substitute naphtha in this LCA study, but pyrolysis oil can also be used as diesel oil after treatment steps. Choice of utilization area depends on the economic value of diesel vs treatment of pyrolysis oil, and the quality of the produced pyrolysis oil.

5.1. Sensitivity analysis

ISO 14040 (2006) standard defines sensitivity analysis as, "systematic procedures for estimating the effects of the choices made regarding methods and data on the outcome of a study". Utilizing this technique in an LCA study allows the observation of the impacts of any uncertain elements in the study. Sensitivity analysis is conducted by assuming different values for input variables and observing the effects. This allows the predictions of the model to improve and identify the weak points when there are any.

Identified weaknesses lead to unanswered questions and can be further studied in future studies.

In plastic production, the amount of virgin material substituted by mechanically recycled material depends on the producer and the product, therefore difficult to estimate. A substitution ratio of 1:1 was assumed for mechanically recycled products for scenarios 2 and 3 in this study for ease of processing. A sensitivity analysis in which the substitution ratio of recyclate to virgin material is lowered to 1:0,5 is conducted, and results are presented in **Figure 11, Figure 12,** and **Figure 13**. A substitution ratio of 1:0,5 means recycled plastic material can replace 50% of the virgin material that would have been used in production.
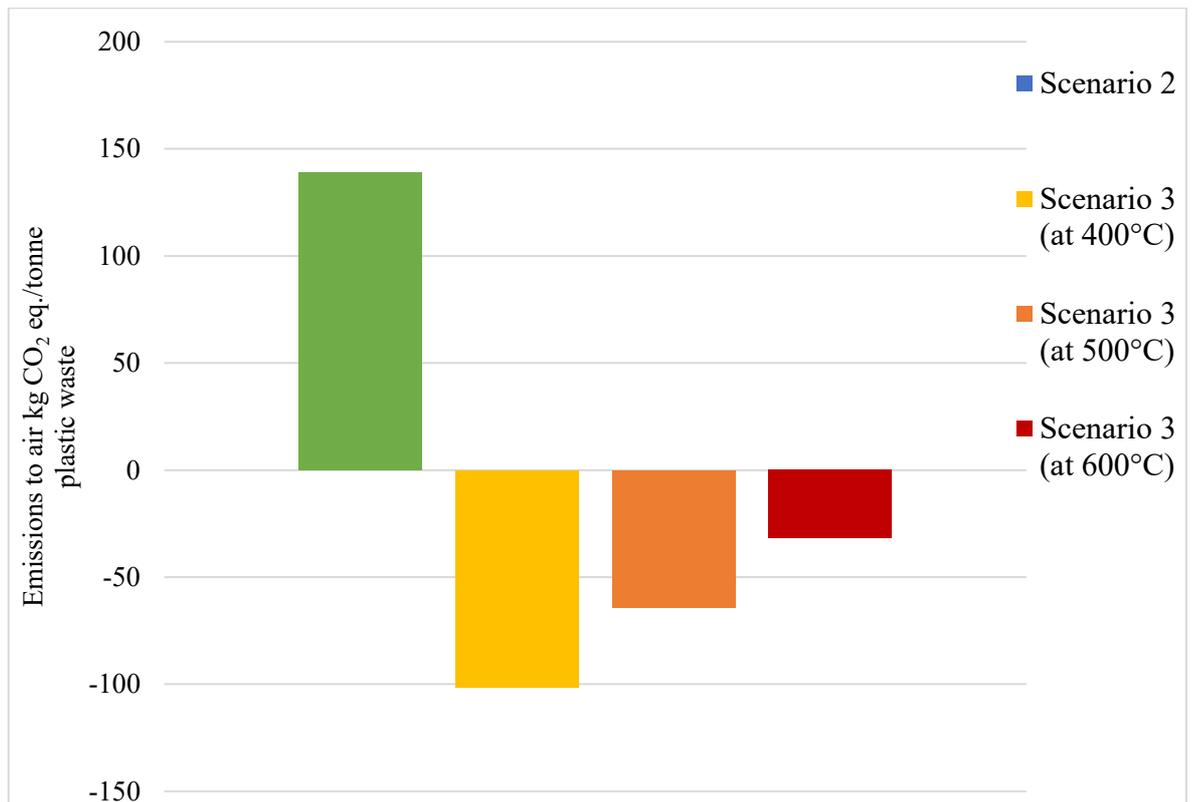
Figure 11. Emissions to air (kg CO_2 eq.) from Scenario 2 (mechanical recycling with energy recovery) and Scenario 3 (mainly pyrolysis) are presented in the figure. Pyrolysis reactions in Scenario 3 were conducted at 400°C, 500°C, and 600°C. Mechanical recycling products were assumed to be substituted 1:0,5.

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It can be seen in **Figure 12**, the avoided emissions by recyclate substitution were decreased by 50% when the substitution ratio was lowered to 1:0,5 from 1:1. None of the other steps show a change, which was expected. A substitution ratio of 1:1 in Scenario 2 allows a total of -476 kg CO_2 eq. emissions to be avoided, whereas lowering that substitution to 1:0,5 results in the total emissions of 139 kg CO_2 eq. Emissions from Scenario 2 increase when the recyclate substitution ratio is lowered (**Figure 12**). The recyclate substitution step is the only one affected by the change in assumptions as can be seen in **Figure 12**. These results indicate for mechanical recycling to be significantly effective in reducing global warming potential impacts, a high-quality product is required. A high-quality mechanical recycling product can replace virgin material close to 1:1.
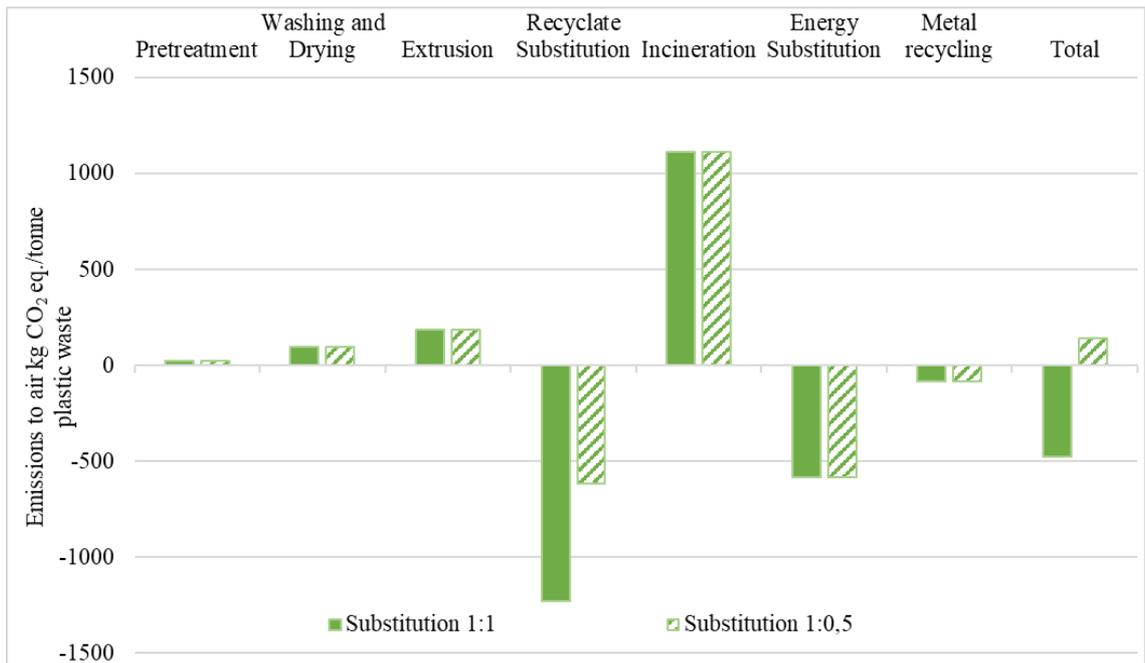

Figure 12. Emissions to air (kg CO_2 eq.) per tonne of post-consumer plastic waste from Scenario 2 are shown in the figure when recyclate substitution is 1:1 and 1:0,5 recyclate to virgin material.

Scenario 3, with a 1:0,5 recyclate substitution ratio, has a lower emission at any temperature point compared to Scenario 2 (**Figure 11**). Pyrolysis oil to naphtha

substitution is not under consideration here, only mechanical recycling product substitution is changed, and mechanical recycling is not the main process analyzed in Scenario 3. The amount of waste processed by mechanical recycling is less, only being PET and therefore, its effects are expected to be less compared to Scenario 2. Still, almost a 50% decrease in the reduced emissions can be seen in **Figure 13**. Pyrolysis temperature does not affect the avoided emissions as the substitution, in this case, is only for mechanically recycled material.
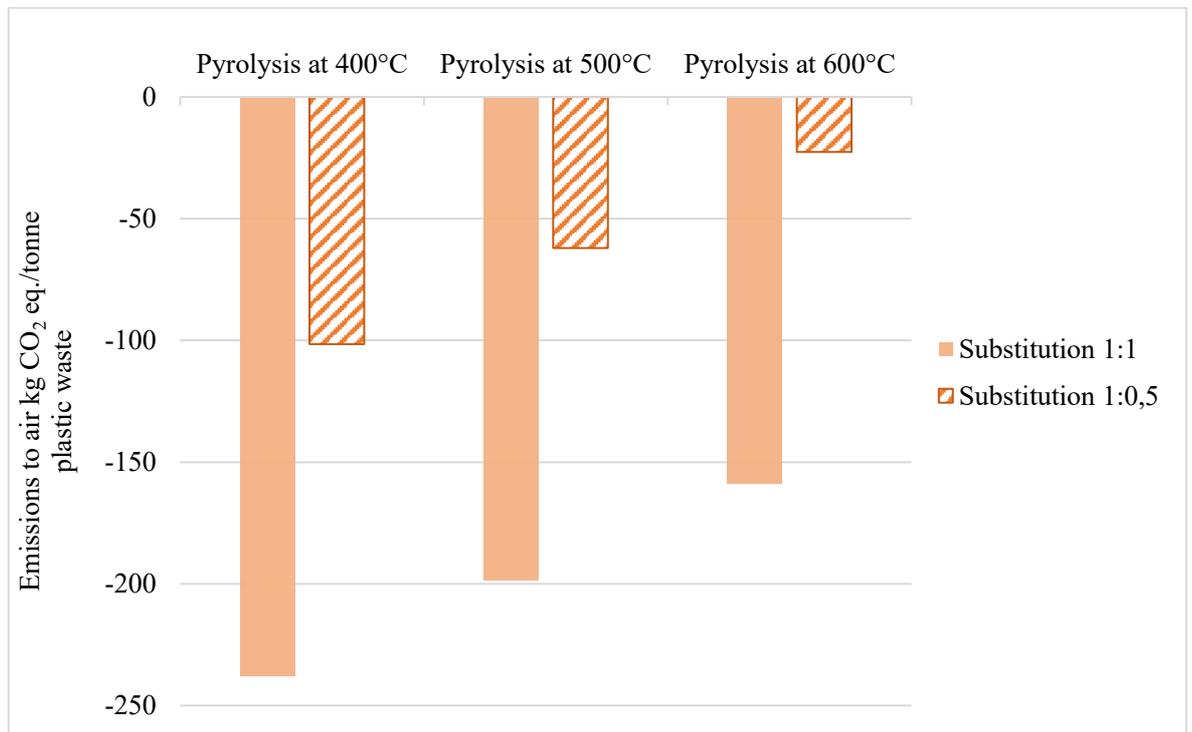
Figure 13. Emissions to air (kg CO_2 eq.) per tonne of plastic waste from Scenario 3 with pyrolysis at 400°C, 500°C, and 600° are shown in the figure when recyclate substitution is 1:1 and 1:0,5 recyclate to virgin material. The data compared in this figure is the total amount of emissions from the corresponding scenario, as that is the only variable changing in any significance between different pyrolysis temperatures.
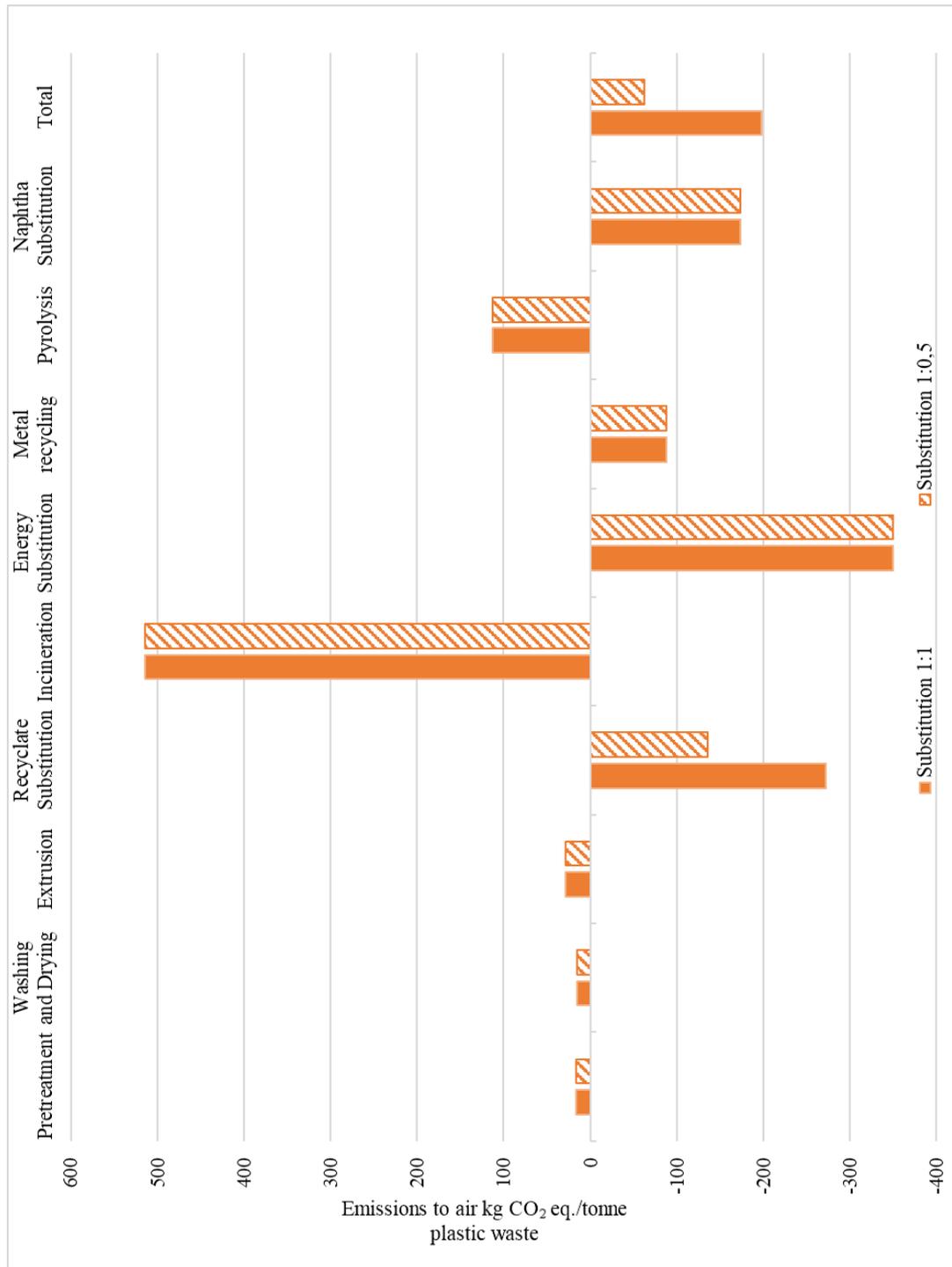
Figure 14. Emissions (kg CO_2 eq./tonne) from Scenario 3, at 500°C. Mechanical recycling product substitution ratios were investigated at 1:1 and 1:0,5 and results are presented in the figure.

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Changing the substitution ratio of recycled material has a significant effect on the emissions of both scenarios as can be observed in **Figure 12** and **Figure 13.** In comparison with that, it can be seen from **Figure 14** that besides the recyclate substitution step, no other steps show any change in Scenario 3. As the steps of all temperature points of Scenario 3 are the same, values for 500°C were used as reference. From these figures, it can be deduced that the effectiveness of mechanical recycling depends on the substitution ratio of the final product, the recyclate. Lowering the substitution ratio lowers the effectiveness of mechanical recycling.

6. Conclusions

Mechanical recycling has significant emission lowering benefits upon a scheme involving the only incineration. These benefits depend heavily on recyclate substitution ratios and the amount of rejected material from recycling. The effectiveness of mechanical recycling is limited as recycling cycles of a plastic polymer are limited, and non-recycled material can only be treated via energy recovery. With the current technology, it is not possible to achieve total efficiency in material recycling.

Pyrolysis reaction produced gas, char, and liquid oil, in which gas and char were used as an energy source for the reaction, and liquid oil was substituted for naphtha. Scenario 3 reduced more emissions at temperatures 400°C and 500°C than Scenario 2 and 600°C performed slightly below Scenario 2. When the recyclate substitution ratio was lowered, impacts from mechanical recycling increased significantly. Therefore, it can be concluded from an environmental standpoint, pyrolysis performs better compared to a scheme of mechanical recycling with incineration. More of the feedstock can be utilized through pyrolysis, resulting in higher recovery rates.

Future research can be focused on more sustainable ways of using pyrolysis oil. Using pyrolysis technology, not for fuel recovery but material recovery would be more environmentally friendly, as at the very least it would lower raw material need of plastic production, possibly remove it. However, the environmental impacts of depolymerization are not that well known yet. Impacts of pyrolysis oil as raw material for plastic products should be further investigated in an LCA study and compared to recyclate utilization in plastic production.

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