A comparison of the mass, carbon and energy balance of three plastic waste treatment pathways

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Abstract

Despite plastic being a valuable material with a wide range of uses, it has proved to be a complex material to recycle. Currently, the main method of retaining value of post-consumer plastics in the EU is mechanical recycling. However, the enormous variability of polymers cause for significant challenges in producing high quality recycled materials. In an attempt to increase recycling rates, initiatives are being made to improve identification and sorting of plastics. However, despite these improvements it is unclear to which extent plastic loops may be closed in real life. As a result, chemical recycling technologies are increasingly being investigated in terms of their potential to fill the gaps where mechanical recycling is not feasible. This report provides a model that can compare the mass, carbon and energy balance of three different technology pathways of municipal plastic waste in Denmark, based on generic data that is obtained from literature. The different pathways include (a) incineration with energy recovery (with and without carbon capture), (b) mechanical recycling, and (c) chemical recycling via pyrolysis (of PE, PP, PS and a mixed 'other' fraction) and chemolysis (of PET).

Keywords

Plastic recycling — Mechanical recycling — Chemical recycling — Incineration — Pyrolysis — chemolysis — Mass balance — Carbon balance — Energy balance — Material flow analysis

1. Introduction

The production of plastic has grown exponentially over the past 70 years (European Parlament, 2018), which has caused for plastic to outgrow almost all other man-made materials (Geyer et al., 2017). Currently, the production rate exceeds over 350 Mt each year (Geyer et al., 2017). However, it is expected to exceed 500 Mt by 2050 (Sardon and Dove, 2018). Inevitably, this has also caused for an increase in the amount of plastic that ends up as waste. In fact, of the amount of plastic produced on an annual basis, approximately 80% is discarded as plastic waste (European Parlament, 2018). This is largely due to the fact that a large amount of the plastic produced is used to make single-use packaging and other short-lived consumer products, which are quickly discarded (Jeswani et al., 2021).

Globally, the majority of plastic waste is landfilled. However, in the EU the majority is incinerated with energy recovery (42.6%), about 32.5% is being recycled and 24.9% is being landfilled (European Parlament, 2018). This means that both the global and European plastic economy remains largely linear. According to the European Parlament (2018), researchers estimated that the global production and incineration of plastic in 2019, released over 850 million tonnes of greenhouse gases into the atmosphere, which they expect could rise to 2.8 billion tonnes by 2050, if no measures are taken towards improved recycling.

However, with their Circular Economy Action Plan, the EU have communicated their ambitions of moving towards a circular economy. In connection with the plan, they have renewed their recycling targets, whereby the EU hope to achieve a recycling rate of 55% for household plastic packaging waste by 2030 (European Commission, 2019), which by far makes up the largest share of all plastic waste (Ragaert et al., 2017a; Villanueva and Eder, 2011). However, according to the European Parlament (2018), recent restrictions on imports of plastic waste in China, have posed the risk of increased incineration and landfilling of plastic waste in Europe. This has resulted in increased attention being paid towards trying to find more circular and climate-friendly ways of managing plastic waste.

Nevertheless, plastic waste as a material is very complex to recycle (Ragaert et al., 2017a). This is because, plastics are made up of a very large group of individual polymers with different both chemi-

cal and technical characteristics (Hansen et al., 2013), of which the most common types are high and low density polyethylene (HDPE and LDPE), polypropylene (PP), polyvinylchloride (PVC), solid and expanded polystyrene (PS) and polyethylene terephthalate (PET) (Villanueva and Eder, 2011). These differences cause for a costly and time-intensive presorting process and an energy-intensive recycling process which often leads to low-quality polymers (Garcia and Robertson, 2017; Hansen et al., 2013). In addition, the technologies that currently exist on the market today, cannot be applied to many of the different polymeric materials (Garcia and Robertson, 2017). Thus, the end-of-life treatment options for plastic waste are in practice quite limited (Garcia and Robertson, 2017). However, recent research has caused for chemical recycling methods to attract scientific attention (Garcia and Robertson, 2017; Villanueva and Eder, 2011). Mechanical recycling has a lower energy consumption than chemical recycling, however, chemical recycling is a promising method as it avoids the need for sorting due to compatibilization of mixed plastic wastes and ability to treat traditionally non-recyclable polymers (Garcia and Robertson, 2017).

Although chemical recycling has attracted attention in terms of improving plastic circularity, some studies claim that mechanical recycling will remain the most effective method to recycle plastics in terms of time, economic cost, carbon footprint and environmental impacts (Schyns and Shaver, 2021). However, no studies provide a comparison of the mass, carbon and energy balances of mechanical and chemical recycling, and incineration. Therefore, this study seeks to provide a generic assessment, which is based on the currently existing literature, of the mass, carbon and energy balance of the following three treatment pathways:

Scenario A Incineration of all plastic fractions, with energy recovery;

Scenario B Mechanical sorting, recovery and recycling of PET, PE, PP and PS; and

Scenario C Chemical recycling and refining of the oil products, via (C1) pyrolysis of PE, PP and PS, and via (C2) chemolysis of PET.

For Scenario A, two versions of incineration is addressed: with and without carbon capture (CC) from the generated flue gas. All of the materials, which for whatever reason cannot be recycled in Scenarios B and C, are assumed sent to incineration according to Scenario A.

2. Available literature

Currently there is no available literature examining and comparing the mass, carbon and energy flows between the incineration, mechanical recycling and chemical recycling of plastic waste. However, several studies have conducted Life Cycle Assessments comparing the associated environmental impacts on one or more of the treatment pathways, and others have provided assessments of a single pathway but on the polymer level. In this section, the literature used to model the different treatment pathways in this paper will be described.

In order to model different types of treatment of post-consumer plastic, an understanding of the composition of the plastic waste stream in Denmark must be obtained. Eriksen et al. (2019) and Eriksen and Astrup (2019), has provided thorough research into the composition of Danish source-separated household plastic waste. Eriksen et al. (2019) provides insights into different sorting schemes and their efficiency, and Eriksen and Astrup (2019) provides a more detailed characterization of the rigid fraction of the waste. Both studies were used to construct the composition of the plastic waste stream being treated in by the model, and are assumed to provide a realistic overview of the composition of post-consumer plastic waste in Denmark.

As previously mentioned, the majority of post-consumer waste in the EU is incinerated with energy recovery. This is also the case for Denmark, where approximately 31% of all plastic packaging was collected for recycling in 2019, and only roughly 14% is actually recycled (The Ministry of Environment of Denmark, 2019). Although Danish incineration plants recover both electricity and heat, and thereby reduce the dependency on fossil energy, they are still net emitters of fossil CO_2, due to considerable amounts of fossil carbon present in the input waste (Bisinella et al., 2021). Consequently, within recent years waste incinerators have placed increasing interest in potential implementation of

CC as a post-treatment technology.

Bisinella et al. (2021) has carried out a study assessing the amendment of CC and storage (CCS) technology at Amager Bakke from an environmental perspective. The data obtained from the study has been used to model the results for an incineration with CC process in this report. According to Bisinella et al. (2021), Amager Bakke has one of the highest energy recovery efficiencies in Europe. This should be taken into consideration when interpreting the results, however, it should also provide a realistic overview of the mass, carbon and energy balance of state-of-the-art waste incineration in Denmark.

As for the mechanical recycling of plastic waste, no literature was found that assessed recycling of plastic waste in Denmark specifically. This is likely due to the fact that much of the plastic waste that is collected for recycling in Denmark is exported and recycled internationally in countries such as Germany (Plastic Change, 2021). However, (Eriksen et al., 2020) has provided a thorough evaluation of the potential circularity of PET, PE, and PP flows in Europe. The study considers product lifetimes, demand growth rates, and quality reductions of recycled plastic (downcycling) and represents 2016 conditions. The vast majority of the data used to model the mechanical recycling pathway in this paper was obtained from this study, and is assumed to provide a fairly realistic overview of the sorting and mechanical recycling of Danish post-consumer plastic. However, because the study is based on European plastic waste, it is assumed that some of the sorting efficiencies may be slightly lower than actual conditions.

Finally, several studies were used to model the chemical recycling pathway. However, no studies were found that assessed chemical recycling in Denmark specifically. Similar to this study, (Jeswani et al., 2021) conducted a comparison of the life cycle environmental impacts of chemical recycling via pyrolysis of mixed plastic waste, mechanical recycling and energy recovery in Germany. However, they mention that the technologies considered in their study are generic, thereby indicating that their findings may be applicable to other European countries. The majority of the data used to model the pyrolysis pathway was based on data obtained from this study, and supplemented with data from Civancik-Uslu et al. (2021), who presented a life cycle assessment of mechanical and thermochemical recycling in Belgium.

Other studies that were used include works by Solis and Silveira (2020); Barnard et al. (2021); Ragaert et al. (2017a); The European Chemicals Agency (2021) and Hann and Connock (2020). However, these were mainly used to obtain an understanding of the state-of-the-art conditions in terms of chemical recycling technologies and commercial viability, as opposed to specific recycling efficiencies and material yields. Hann and Connock (2020) did provide an insight into the yields of glycolysis, however, no data were provided in terms of mass flow and specific yields of by-products and so on. Here, Shen et al. (2010) was the only study, which was observed, that provided somewhat of an understanding of the mass flow in the glycolysis process. However, it was not possible to obtain a sufficient data for the energy requirements based on this.

According to Jeswani et al. (2021), there is a lack of literature that is based on data from operating industrial plants. This indicate that the currently available data might not necessarily represent actual or realistic conditions. Moreover, they mention that the life cycle assessments that have been conducted, do not not consider the difference in the quality of the output material. According to Jeswani et al. (2021), this could disadvantage the pathways that produce a better quality recyclate. This report seeks to present a collection of the data, which is currently available, that is necessary to model the mass, carbon and energy flows of incineration with energy recovery, and mechanical and chemical recycling of post-consumer plastic waste, whilst attempting to obtain a similar recyclate quality.

3. Methodology

3.1 Plastic waste generation

As previously mentioned, the most widely used polymers include HDPE, LDPE, PP, PVC, (E)PS and PET (Villanueva and Eder, 2011). According to Eriksen et al. (2019), the average generic composition of plastics in municipal solid waste (MSW) in Europe is LDPE (30%), PET (27%), HDPE (10%), PP (7%), PS (1%) and others (25%). Thus, this is assumed to be the composition of the initial plastic waste generation, which is inputted into the model.

In addition, the amount of bottles, rigid, soft and other plastic, which is evident for each polymer, will also be included in the model, amounting to 20 flows in total. See **Table 1** where the composition of plastic waste is presented.

Table 1. The plastic waste composition (%).

Polymer	Bottles	Rigid	Soft	Other
PET	23	4	0	0
PE	7	3	30	0
PP	0	7	0	0
PS	0	1	0	0
Other	0	5	10	10

All data is obtained from Eriksen et al. (2019).

3.2 Mass, carbon and energy balances

The efficiency of a given system can differ depending on the parameter that it is being compared by. For example, the recycling efficiency of a given polymer may be high in terms of how much of its incoming mass it is possible to recover, however, the processes required to recover that amount may prove to be very energy-intensive. This means that the same system might be very efficient in terms of recovering mass, yet not very efficient in terms of consuming energy. For this reason, this study seeks to investigate the mass, carbon and energy balance of the different treatment pathways.

Because this study seeks to provide a more generic assessment of the different types of treatment, the balances are obtained using average, existing data from literature, as opposed to data from a single plant, or from field work. However, in cases where an average is not obtainable, data from a specific plant may be used as a proxy, or from the database from the LCA software, EASETECH (Clavreul et al., 2014), which was developed at DTU Environment, for assessing environmental impacts of waste management systems. In cases where data is found for both low- and high-efficiency systems, both are included in the model in order to provide a range in the results. Thus, the model can be run using either the min and max values, in order to obtain recycling rates from both low- and high-efficiency systems. Common for all of the balances is that they are set up such that:

$$\sum Inputs = \sum Outputs \tag{1}$$

Each system is made up of a chain of unit processes. These processes are all modelled with an appropriate transfer coefficient (TC), such that they describe the appropriate partitioning of the incoming flow within the process. Thus, the specific output flow, *x*, which is being diverted from a given unit process, *i*, is found by multiplying the TC for that flow with the total input to the process, such that:

$$Output_x = \sum_i TC_{x,i} \cdot \sum Inputs \tag{2}$$

The mass balances are modelled with the intention of observing the amount (kg wet weight (ww)) of the materials (e.g. plastics) that are being circulated throughout the given treatment system. Here, the purpose is to identify which of the given pathways result in the largest amount of substituted virgin material, thereby making it the more preferable form of treatment.

The carbon balances, on the other hand, are modelled with the intention of observing the amount of carbon (kg C) that is being circulated throughout the system, whilst also accounting for any emissions such as carbon dioxide (CO_2). Here, the main purpose is to identify the amount of CO_2 which is being released into the atmosphere as a result of the given treatment pathway, thereby making it the less preferable form of treatment.

Lastly, the energy flows are modelled with the intention of assessing the amount of electricity and heat that is being consumed and produced throughout the given treatment pathway. Here, the purpose is to identify the least energy-intensive pathway, thereby making it the more preferable form of treatment from this perspective.

3.3 Recycling rate

To compare the recycling efficiency among the different recycling pathways, the recycling rate will be calculated. This will be done in accordance with the recent EU guidelines in terms of calculating recycling rates, as described in The Ministry of Environment of Denmark (2019). Here, the actual mass of the recycled material is obtained as a per-

centage of the amount of waste produced of the given waste fraction. This method differs from the methods used by The Ministry of Environment of Denmark (2019) from previous years, where the recycling rate was calculated as the amount of material that was sorted for recycling as a percentage of the total waste produced of the given fraction. Thus, the yields will be lower than those obtained by The Ministry of Environment of Denmark (2019), and reported to the EU, from previous years.

3.4 Treatment pathways

In this study, three different generic treatment pathways for treating plastic waste are modelled and compared. These pathways are divided among three different scenarios, where Scenario A represents incineration with energy recovery (with and without CC), Scenario B represents mechanical recycling and Scenario C represents chemical recycling, see the process diagram in **Figure 1** for an illustration of all three pathways. Common for all of the scenarios is that they begin with an initial source-separation process.

All of the scenarios are modelled using Excel, for which each set-up will be described in further detail in this section. In addition, the specific processes that are modelled will also be described.

3.4.1 Incineration (Scenario A)

As all of the individual pathways include incineration, the overall construction of the incineration process will mainly be described in this section. Thus, all of the TCs used to model the mass, carbon and energy balances, which take place during incineration and CC, can be found in this section.

The processes related to incineration and CC, were modelled using data obtained from Bisinella et al. (2021) and through the EASETECH database. From EASETECH, the composition of the different plastic fractions were found, see **Table 2**. These were used to model the amount of carbon and energy contained in the plastic, which passes through the system. They were also used to obtain the amount of plastic that is transformed to ash and to flue gas as a result of the incineration process. The amount of flue gas produced was assumed to be the water and VS content of the incinerated fraction combined.

The study conducted by Bisinella et al. (2021) provides an assessment of a generic energy-efficient plant, and was thus used to construct the overall processes that are included in the model. According to Bisinella et al. (2021), incineration with CC and energy recovery takes place through several different processes: First, the waste is incinerated in a furnace. The incineration process produces ashes and flue gas. The flue gas is sent to a boiler for energy recovery, with steam distributed to a turbine. In the turbine, electricity and heat are produced. The flue gas is then passed through air pollution control and condensation, before the carbon is captured.

Table 2. The composition of the plastic fractions.

Fraction	Ash	VS	Water	C	LHV
	kg	*kg*	*kg*	*kg*	*MJ*
Bottles	0.05	0.84	0.11	0.60	32.70
Rigid	0.21	0.95	0.03	0.77	36.21
Soft	0.48	0.82	0.14	0.79	34.41
Nonrec	0.51	0.88	0.07	0.65	29.69

Note: *Nonrec=non recyclable fraction, VS=volatile solids, C=carbon and LHV=lower heating value. All data is obtained from Clavreul et al. (2014)*

For simplicity, this report has divided the processes described by Bisinella et al. (2021) into two overall processes; namely (1) incineration, which includes the furnace, boiler, turbine, air pollution control and condensation; and (2) carbon capture, which merely includes the carbon capture process. This can be seen in **Figure 1**. However, because CC is not implemented in Denmark and merely at the testing stage, results are provided for incineration both with and without CC.

In addition, CCU also presents a possible future option for treating plastic waste with incineration, where the captured carbon is used to produce olefins such as ethene and propylene, which may subsequently be transformed into PE or PP polymers. However, currently this process is merely in the research stage, and it has thus been disregarded in this study. Nevertheless, it could be an interesting pathway to take into consideration in future studies, comparing the recycling efficiencies of plastic treatment scenarios.

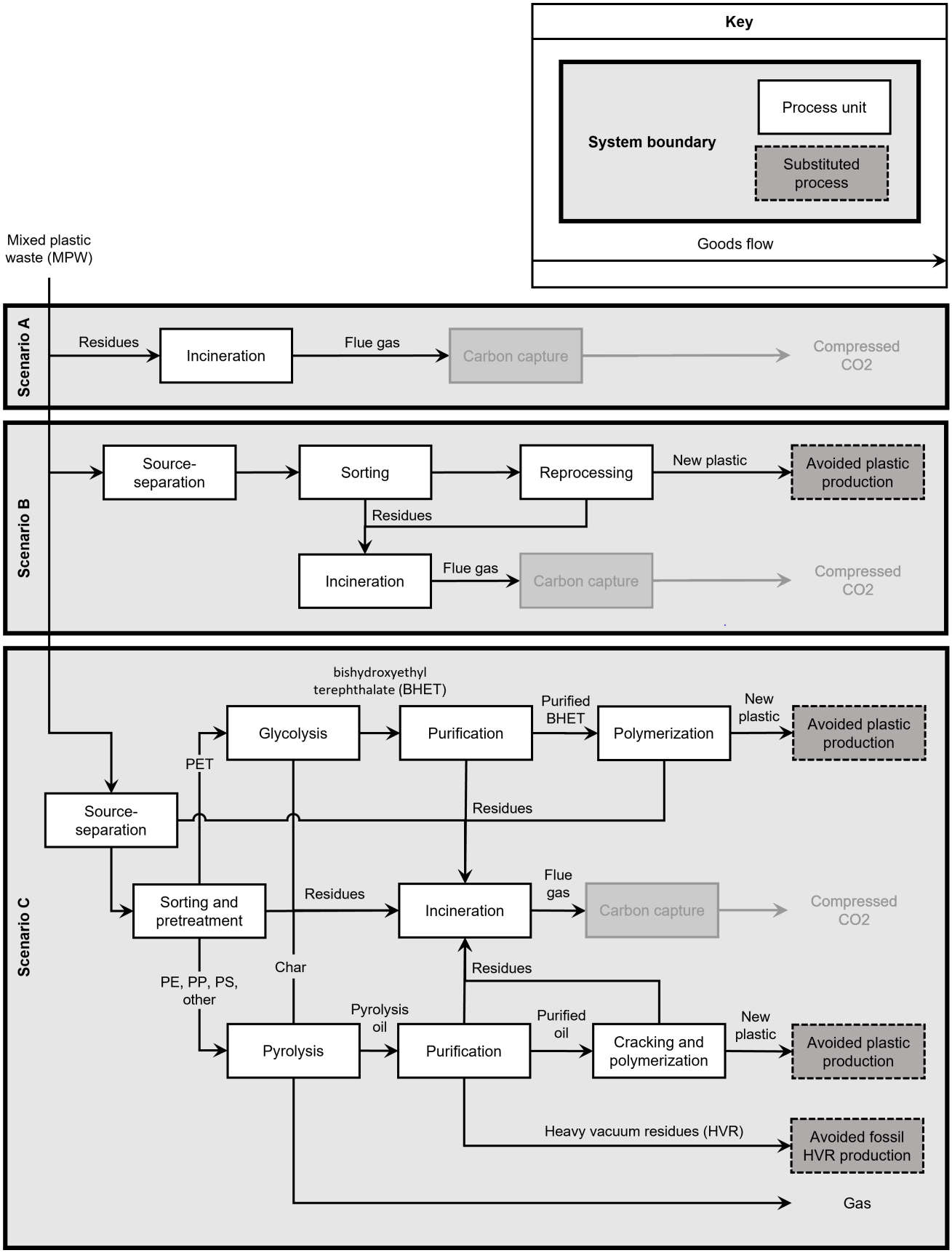

Figure 1. The process diagrams of each of the three scenarios.
Note: *The carbon capture processes are grey to indicate that they are considered a potential future option.*

Mass balance In Scenario A it is assumed that all of the plastic waste is incinerated. Thus, it is assumed that the total 1 t of plastic waste is sent directly to incineration. Here the waste is burned, which as previously mentioned, produces ashes and flue gas. Here, the ash content, and VS and water content, which are provided in **Table 2**, were used to model the amount of ash and flue gas produced, respectively.

The amount of flue gas that is produced has been assumed to amount to the VS and water content that is evident in the incinerated waste stream. However, this report does not take into consideration the chemicals or fuels that are added during the incineration process, which would add to the mass of the flue gas production. Thus, it is expected that the amount of flue gas, which would actually be produced, is higher than the one produced in the model. In addition, the weight of the oxygen that is contained in the flue gas is completely disregarded. Thus, the mass of carbon dioxide, which is compressed from the flue gas, is merely equal to the carbon content. Therefore, the mass balance and carbon balance values for the CC process are the same.

Carbon balance The carbon content in each of the different plastic fractions was also obtained through the EASETECH database, see **Table 2**. The carbon content is assumed to be directly dependent on the mass flows for each fraction. However, since ash does not contain (significant amounts of) carbon, it is assumed that all of the carbon contained in the incinerated fraction is transferred to the flue gas. The amount of carbon that is captured during the CC process is based on the findings of Bisinella et al. (2021), which suggests a CC rate of 85% to 90% (min and max).

Energy balance The energy content that is contained in each of the different fractions was also obtained through the EASETECH database, see **Table 2**. As for the carbon content, the energy content is also assumed to be directly dependent on the mass flows for each fraction. However, the electricity and heat consumption, which are required to power the different processes, are also taken into consideration. The electricity consumption that is used for the modelling of the incineration treatment pathway is listed in **Table 3**.

Table 3. The energy consumption in the incineration pathway (Scenario A) in MJ/kg plastic.

Process	INC		CC	
	min	max	min	max
Furnace	0.18	0.18	-	-
APC	0.14	0.14	-	-
Condensation	0.04	0.04	-	-
CC	-	-	0.2	0.2

Note: *INC=incineration, CC=carbon capture. All data is obtained from Bisinella et al. (2021).*

3.4.2 Mechanical recycling (Scenario B)

The mechanical recycling process typically involves sorting, washing, shredding and melting of plastic waste to produce granulate and finally new plastic products, thereby substituting the use of virgin plastic (Ragaert et al., 2017a). For simplification, the scenario is modelled such that these steps are divided among (a) a material recovery facility (MRF) for the sorting stage, where the plastic waste is sorted and prepared for reprocessing; and (b) a reprocessing facility for the mechanical recycling stage, where each of the individual polymers are recycled into granulates. All reject plastic waste, which is not considered to be mechanically recyclable, is sent to incineration.

As can be gathered from **Figure 1**, this treatment pathway is thus made up of five processes; namely (1) source-separation (SS), (2) sorting (SOR), (3) reprocessing (RE) and (4) incineration (INC) with (5) carbon capture (CC) and energy recovery.

Mass balance The mechanical recycling processes are modelled primarily using data obtained from the study conducted by Eriksen et al. (2020). According to Eriksen et al. (2020), polyethylene (PE), PP and PET are vital from a mechanical recycling perspective, and thus, these are the only polymers that the model regards as mechanically recyclable. The remaining polymers are regarded as reject and sent to incineration. The TCs that are used to model the source-separation, sorting and reprocessing processes were obtained from Eriksen et al. (2020), and can be found listed in **Table 4**. Here, the sorting and collection phases described by Eriksen et al. (2020), are comprised in the SOR process. According to Eriksen et al. (2020), some of the collected plastic is exported, and the remaining fraction is

sent to sorting. However, in this report, it is assumed that all of the exported plastic is also sent to sorting.

Table 4. The TCs used to model the mass transfer in the mechanical recycling (Scenario B).

Fraction	SS		SOR		RE	
	min	max	min	max	min	max
PET						
Bottles	0.58	0.90	0.83	0.83	0.83	0.83
Rigid	0.55	0.90	0.24	0.83	0.78	0.83
Soft	0.70	0.90	0.58	0.83	0.78	0.83
PE						
Bottles	0.45	0.90	0.76	0.88	0.93	0.93
Rigid	0.21	0.90	0.01	0.88	0.80	0.85
Soft	0.56	0.90	0.58	0.88	0.72	0.85
PP						
Bottles	0.45	0.90	0.44	0.70	0.81	0.84
Rigid	0.56[a]	0.90	0.00	0.70	0.80	0.84
Soft	0.70	0.90	0.58	0.70	0.72	0.84

Note: *SS=source-separation, SOR=sorting, RE=reprocessing. The min and max values for SS indicate the amount of waste being source-separated for sorting, the values for SOR indicate the amount of waste being sorted for reprocessing, and the values for RE indicate the amounts being reprocessed into plastic granulates. The remaining amounts of waste are sent to incineration.*

[a] Data obtained from Faraca et al. (2019).

All other data is obtained from Eriksen et al. (2020).

Carbon balance The carbon content in each of the different plastic fractions was obtained through the EASETECH database, see **Table 2**. As with the incineration pathway, the carbon content is assumed to be directly dependent on the mass flows for each fraction. See **Section 3.4.1** for a description.

Energy balance The energy content in each of the different fractions was also obtained through the EASETECH database, see **Table 2**. As for the carbon content, the energy content is also assumed to be directly dependent on the mass flows for each fraction. However, the electricity and heat consumption required to power the different processes is also taken into consideration. The total energy consumption that is used for the modelling of the mechanical recycling treatment pathway is listed in **Table 5**. The energy consumption used for modelling the incineration and CC processes are, however, found in **Table 3**.

Table 5. The energy consumption used to model the mechanical recycling (Scenario A) in MJ/kg plastic.

Polymer	SOR[a]		RE[a]	
	min	max	min	max
PET	0.096	0.42	2.80	8.65
PE	0.096	0.18	0.99	8.74
PP	0.096	0.18	0.99	8.65
PS	0.096	0.18	0.99	8.74

Note: *SOR=sorting, RE=reprocessing.*

[a] Data obtained from The Danish Environmental Protection Agency (2022).

3.4.3 Chemical recycling (Scenario C)

Chemical recycling of plastics can be performed in many different ways, with pyrolysis being among the more well-known instances (European Chemicals Agency, 2021). According to International Organization for Standardization (2013), pyrolysis is an irreversible chemical decomposition caused solely by a rise in temperature. This report, thus, assumes that the plastic fractions are treated by pyrolysis. However, not all polymers are currently suitable for pyrolysis (European Chemicals Agency, 2021). According to several different sources only PE, PP and PS are among the polymers, which this report takes into account, that are treated by pyrolysis commercially (Ragaert et al., 2017b; Solis and Silveira, 2020; Lopez et al., 2017; Datta and Kopczyńska, 2016; Vollmer et al., 2020).

According to Solis and Silveira (2020); Lopez et al. (2017), PET is only suitable for pyrolysis with in-line reforming. However, according to European Chemicals Agency (2021) this technology is only available on a pilot scale, and thus, has not been taken into account in this report. Instead, it is assumed that the PET is treated by another form of chemical recycling that is currently available at a commercial scale, namely chemolysis. According to Zhou et al. (2016), chemolysis involves treat-

ing the classified polymeric wastes with solvents and reagents (or catalysts) to depolymerize the polymer to low molecular weight chemicals and oligomers. There are various different types of chemolysis, however, glycolysis is the simplest and oldest method of PET depolymerization (Ragaert et al., 2017a; Barnard et al., 2021). According to Barnard et al. (2021) and Ragaert et al. (2017a), the technologies using glycolysis are the most advanced in terms of demonstrating commercial viability on a larger scale, in comparison to other chemical recycling technologies, and that glycolysis is currently a commercial PET recycling method that is practiced by renowned companies worldwide. It was therefore selected as the recycling pathway for PET in this report.

This scenario, thus, includes two different chemical recycling treatment pathways:

Scenario C1	Pyrolysis of PE, PP and PS
Scenario C2	Chemolysis of PET via glycolysis

This report models the pyrolysis pathway, Scenario C1, largely based on the model produced by Jeswani et al. (2021). Here, the waste is sorted once it has been collected from source-separation, where PET is removed from the plastic waste. It is assumed that the sorting process also includes a form of pretreatment. Once the waste has been sorted, the PE, PP and PS fractions are sent to pyrolysis. The pyrolysis process produces three outputs (European Chemicals Agency, 2021); namely char, which is sent to incineration with energy recovery, despite that Jeswani et al. (2021) use it as a lignite substitute (the main fuel used by the cement industry); oil, which is sent to purification; and gas, which is lost. According to Jeswani et al. (2021), only oil and char are produced, however, they describe a mass loss due to 'process inefficiencies'. This report assumes that this is lost in the form of gas. The purification process produces mainly a high-quality purified oil, along with a minor fraction of solid residues. Finally, the oil is sent to a steam cracker, which prepares it for polymerization, where plastic granulate is produced. It is assumed that all of the residues are sent to incineration.

As for the glycolysis pathway, Scenario C2, it is mainly modelled using data obtained from Shen et al. (2010). Here, they assume that the PET waste is mechanically sorted, washed and chopped into flakes prior to being sent to glycolysis. His report aggregates these steps into one sorting process. The glycolysis process yields the oligomer bis-hydroxyl ethylene terephthalate (BHET), which is sent to purification via filtration before it is repolymerized into PET and spun into fibre. For the glycolysis pathway, all residues are also sent to incineration.

As can be gathered from **Figure 1**, the chemical recycling pathway is thus made up of ten processes; namely (1) source-separation (SS), (2) sorting and pretreatment (SOR), (3) pyrolysis (PYRO), (4) glycolysis (GLYCO). (5 and 6) purification (PUR), (7 and 8) cracking and polymerization (POLY), and (9) incineration (INC) with (10) carbon capture (CC) and energy recovery.

Mass balance The chemical recycling pathway, Scenario C1, is modelled mainly using data obtained from the studies conducted by Jeswani et al. (2021) and Civancik-Uslu et al. (2021). The TCs that are used to model the pathway are summarized in **Table 6**. Because it was not possible to find precise data for bottles, rigid and soft plastic for each polymer, many of the TCs were obtained using data from mixed municipal plastic waste. For an overview of the specific assumptions made regarding each TC, see **Supplementary Materials** to this report. In addition, the TCs that were used to model the SS process in the mechanical recycling pathway, were assumed to be the same for the pyrolysis pathway, see **Table 4**.

The chemical recycling pathway, Scenario C2, is modelled similarly to Scenario C1. However, it is mainly modelled using data obtained from the study by Shen et al. (2010). As it was not possible to find data regarding different PET fractions (e.g. bottles, rigid and soft), it was assumed that all fractions were calculated using the same TCs. During the sorting and pretreatment, it is assumed that 25% of the PET fraction is removed and sent to incineration based on results from Shen et al. (2010). However, considering the date of its publication, it is assumed that this rate is farely low. Thus, a max value of 10% material removal is assumed based on the data obtained for the pyrolysis pathway. The remaining material in the PET fraction is then sent to glycolysis, purification and repolimerization, where it is assumed that 75% to 98% of

Table 6. The TCs used to model the mass transfer in the pyrolysis pathway (Scenario C1).

Fraction	PRE		PYRO				PUR				POLY	
	plastic		*oil*		*char*		*purified oil*		*residues*		*granulates*	
	min	max	min	max	min	max	min	max	min	max	min	max
PE												
Bottles	0.58[a]	0.90[b]	0.64[a]	0.80[a]	0.05[a]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
Rigid	0.58[a]	0.90[b]	0.64[a]	0.80[a]	0.05[a]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
Soft	0.80[a]	0.90[b]	0.64[a]	0.80[a]	0.05[a]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
PP												
Bottles	0.85[a]	0.90[b]	0.70[b]	0.82[a]	0.06[b]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
Rigid	0.85[a]	0.90[b]	0.70[b]	0.82[a]	0.06[b]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
Soft	0.58[a]	0.90[b]	0.70[b]	0.82[a]	0.06[b]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
PS												
Bottles	0.58[a]	0.90[b]	0.64[b]	0.70[a]	0.06[b]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
Rigid	0.58[a]	0.90[b]	0.64[b]	0.70[a]	0.06[b]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]
Soft	0.58[a]	0.90[b]	0.64[b]	0.70[a]	0.06[b]	0.07[a]	0.98[b]	0.98[b]	0.02[b]	0.02[b]	0.50[c]	0.98[b]

Note: *PRE=sorting and presorting, PYRO=pyrolysis, PUR=purification, POLY=cracking and polymerization. The min and max values for PRE indicate the min and max amount of waste being sorted for pyrolysis, where all leftover material is sent to incineration. The values for PYRO to oil and to char indicate the amount of plastic that has been transformed to oil and to char, respectively, through the pyrolysis process, and the leftover mass is assumed to be evaporated as a gas. The values for PUR to purified oil and to residues indicate the amount of oil being purified into purified oil, and the amount of residues produced in the process, respectively. The values for POLY indicate the amount of plastic granulates that have been produced from the purified oil, and all leftover mass is assumed to be considered a loss.*

[a] *Data obtained from Civancik-Uslu et al. (2021)*

[b] *Data obtained from Jeswani et al. (2021)*

[c] *Data obtained from Skraldiade (na)*

the PET is recovered (min and max) (Hann and Connock, 2020). According to Shen et al. (2010), the processes have an efficiency of about 95-96%, where roughly 2% is removed after purification and 3% is removed after repolimerization. However, because no data ranges were found for the glycolysis, purification and polymerization processes separately, they were aggregated in the model, using the range obtained from Hann and Connock (2020).

As in the mechanical recycling pathway, the processes related to incineration and CC, were modelled using data obtained from Bisinella et al. (2021) and through the EASETECH database. In **Table 2**, the composition of the different plastic fractions is summarized.

Carbon balance The carbon content in each of the different plastic fractions was obtained through the EASETECH database, see **Table 2**. As with the incineration pathway, the carbon content in the pyrolysis and glycolysis pathways are assumed to be directly dependent on the mass flows for each fraction. See **Section 3.4.1** for a description.

Energy balance The energy content in each of the different fractions was also obtained through the EASETECH database, see **Table 2**, as with the mechanical recycling pathway. Thus, it is also assumed to be directly dependent on the mass flows for each fraction. The electricity and heat consumption, which is required to power the different processes, is also taken into consideration. The energy

consumption that is used for the modelling of the pyrolysis treatment pathway is listed in **Table 7**.

As for the glycolysis treatment pathway, Shen et al. (2010) assumed that the energy consumption coupled to the sorting process was negligible. However, this study assumes the same energy consumption as seen in the pyrolysis sorting and pretreatment (PRE) process. In addition, it is not clear as to how Shen et al. (2010) have modelled the energy consumption of their glycolysis pathway. In fact, a lack of clarity was observed in connection with the overall energy inputs associated with the technologies used during the glycolysis pathway, despite it being a mature technology (Hann and Connock, 2020). According to Hann and Connock (2020), the processes associated to the pathway often require high energy inputs. Therefore, the same energy consumption is assumed as the pyrolysis pathway for the remainder of processes. For the energy consumption of the incineration process, see INC in **Table 5**.

Table 7. The energy consumption used to model the pyrolysis pathway (Scenario B1) in MJ/kg plastic.

Polymer	PRE[a]		PYRO*		POLY[a]	
	min	max	min	max	min	max
PE	0.31	0.31	2.31[a]	9.88[b]	0.33	0.33
PP	0.31	0.31	2.31[a]	9.88[b]	0.33	0.33
PS	0.31	0.31	2.31[a]	9.88[b]	0.33	0.33

Note: *PRE=sorting and presorting, PYRO=pyrolysis, POLY=cracking and polymerization.*

** The pyrolysis process (PYRO) includes the electricity and heat used to power both the pyrolysis and purification process.*

[a] *Data obtained from Jeswani et al. (2021)*

[b] *Data obtained from The Danish Environmental Protection Agency (2022).*

4. Results and discussion

4.1 Excel model

Using the data described in the previous sections, an input output model was built in Excel, which presents the mass, carbon and energy balance of the three different treatment pathways assessed in this study, see **Supplementary Materials** to this report. The model is built such that TCs are entered in ranges, ranging from a minimum value to a maximum value (i.e. from current conditions to optimal conditions). The user of the model has the full overview of all the parameters, which are necessary for the model to run, on the 'info, structure, parameters' page. These parameters require manual input by the user. This has been done such that the model can be easily updated, if more current, or accurate data is obtained. In addition, they have the ability to manually select whether they wish for the model to run using either min or max values.

Once the conditions are selected, an overview of each of the process outputs can be found for each treatment pathway on the 'incineration', 'mechanical recycling', and 'chemical recycling' pages. Here each process is presented such that the relevant TCs, which are used for the modelling of the given process, are visible at the top. These are linked to the TCs that have been manually inputted on the 'info, structure, parameters' page. Thus, no additional values should be inputted on either of the treatment pathway pages, as they merely serve to provide an overview of the processes.

At the bottom of each process, the different outputs are listed. The outputs are for the most part presented per fraction per polymer (i.e. either bottle, rigid, soft or other PET, PE, PP, PS or Other plastic) as seen in the study by Eriksen et al. (2019). For the most part each process has two outputs, one product output being transferred to the preceding process, and a residue output typically being sent to incineration. At the bottom of every process, the user can observe whether or not it contains a mistake, as it will read either 'true' or 'false' (e.g. true indicates that $input = output$). The treatment pathway pages are structured such that the mass balance can be found at the top, followed by the carbon balance, with the energy balance found at the bottom.

Finally, the overall results of the mass, carbon and energy balances can be found on the 'results' page. Here a table is provided with an overview of the mass outputs, recycling rate with and without compressed carbon, and the energy consumption and so on. These results will be discussed in the following sections

4.2 Mass outputs

When observing the mass balances for the different treatment pathways, different mass outputs from

each system are observed. In **Figures 2** and **3**, the composition of the total mass output can be seen for each treatment pathway under best and worst case conditions (using the min and max TCs), and with and without CC, respectively. The mass outputs include ash and flue gas from the incineration process without CC, ash flue gas and compressed carbon from the incineration process with CC, gas emissions from the pyrolysis process, other residues from the pyrolysis purification process (according to Jeswani et al. (2021), these consist of heavy vacuum residues) and reyclate.

As can be seen, the incineration process yields two or three different outputs depending on whether or not CC is included, namely ash and flue gas without CC, and ash, compressed carbon and flue gas with CC. The mechanical recycling process yields the same mass outputs, however, with one additional output, namely recyclate. The chemical recycling process yields an additional two outputs, including gas and other residues. In all of the treatment pathways, when the model was run on minimum values (or 'worst case' conditions), compressed carbon is the largest of the mass outputs, followed by flue gas. This is because a vast amount of the sorted plastic is removed during the sorting, and pretreatment processes. When run on maximum values (or 'best case' conditions), however, the recyclate output was the largest for the two recycling pathways. Here, the chemical recycling pathway actually produces a slightly larger amount of recyclate.

4.3 Recycling rates

As preciously described, the recycling rates are calculated using the mass of the recyclate, as opposed to the amount sorted for recycling. However, an additional recycling rate was also calculated, taking into consideration the compressed carbon, such that it encompasses both the recyclate and the compressed carbon (in this version, neglecting processing losses from further upgrading and conversion of carbon into new plastic, thereby tacitly assuming that all carbon ends up as new plastic) as a percentage of the initially produced waste. It was found that the mechanical recycling pathway had the highest recycling rate among the two when run on worst case conditions, with a recycling rate of ≈22%. The chemical recycling pathway obtained a recycling rate of ≈14%. When taking into consideration the compressed carbon, the pathways obtained recycling rates of ≈69% and ≈60%, respectively. When run on best conditions, the mechanical and chemical pathways yield recycling rates of ≈55% and ≈57%, respectively. When taking into consideration the compressed carbon, the pathways obtained recycling rates of ≈83% and ≈79%, respectively. However, it should be noted that the chemical recycling pathway also produces other outputs. These are typically made up of a co-product, heavy vacuum residue, as previously mentioned. This co-product may be used as an alternative fuel or for material production. However, this only has a minor impact on the overall recycling rate for the treatment pathway.

On a polymer level, PET obtained the highest recycling rate among all of the other polymers, with ≈36% in the mechanical recycling pathway and ≈25% during the glycolysis pathway, when run on the worst conditions. PE obtained the next highest recycling rate in both treatment pathways, with ≈23% and 12% respectively. PS obtained the lowest recycling rates in both pathways, with ≈5% and ≈6%, respectively. However, it was noticed that the TCs obtained for PS in terms of sorting and collection efficiency were quite low, as they were based on European data. It is expected that they would be higher based on current Danish conditions. When run on the best conditions, PET was found to have the highest recycling rate in the glycolysis pathway, with ≈79%, and PE in the mechanical recycling pathway, with a recycling level of ≈68%. The lowest recycling rates were seen for the other fraction, which was found to have a recycling rate of ≈26% for mechanical recycling and ≈33% for chemical recycling.

4.4 Quality of outputs

The model was set up with the basic assumption that the recyclate quality was identical in both recycling pathways. However, this is unlikely the case as the output from the mechanical recycling process is plastic granulates, while the output from the chemical recycling process is plastic fibre. Because of this, the chemical recycling pathway may be disadvantaged in comparison with the mechanical recycling pathway. However, according to Shen et al. (2010), the preferred input of plastic into the

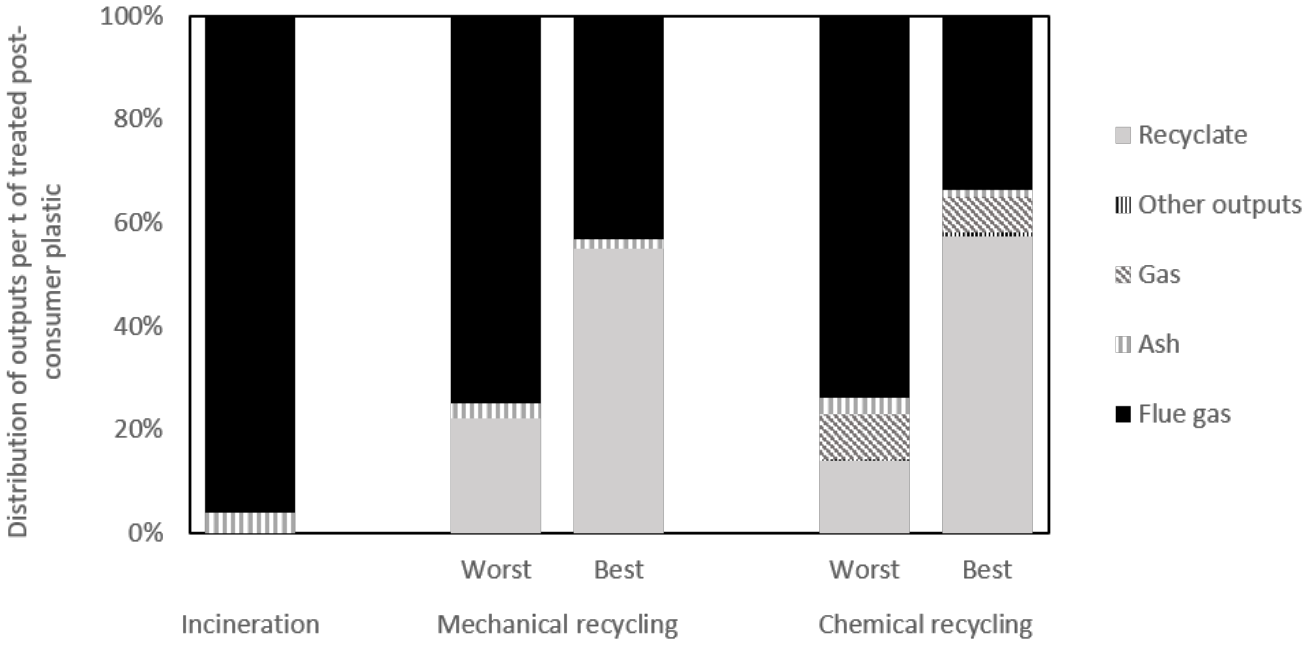

Figure 2. Mass outputs from the different treatment pathways without CC, under best and worst conditions.

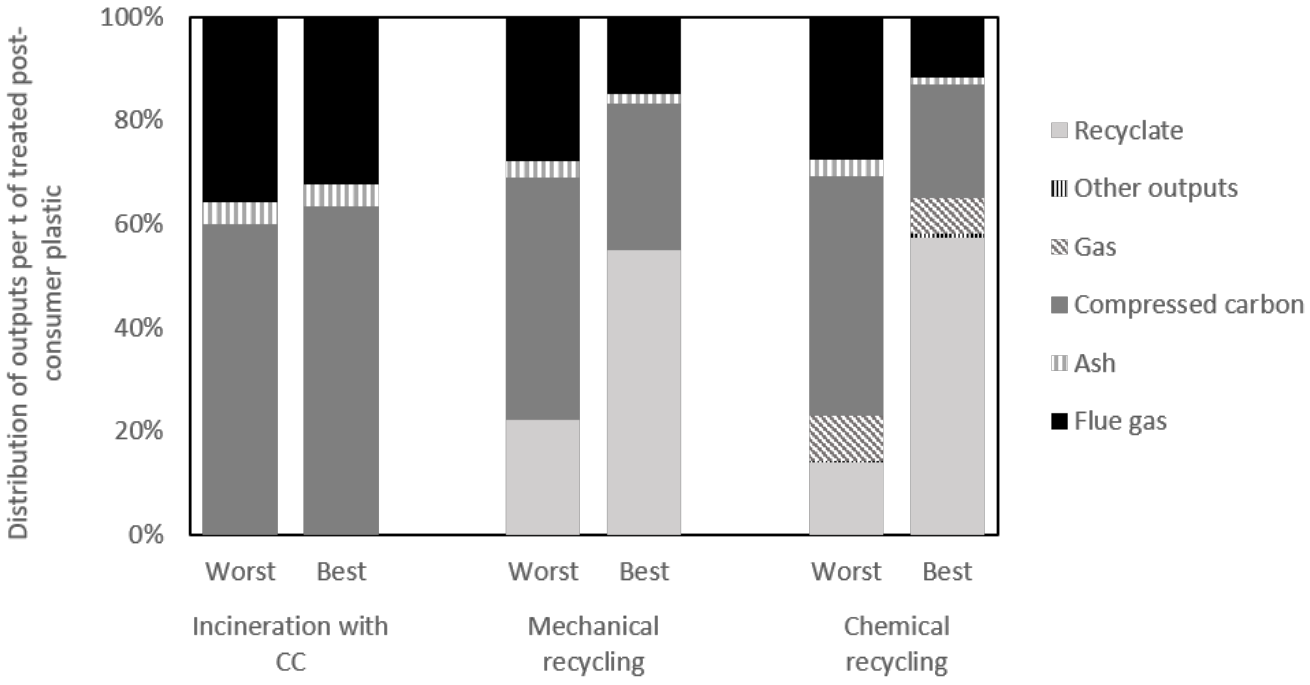

Figure 3. Mass outputs from the different treatment pathways with CC, under best and worst conditions.

chemical recycling pathway is in the form of plastic granulates. Therefore, the plastic was considered pretreated using some of the same processes as in the mechanical treatment pathway. The specific needs for (mechanical) pretreatment prior to chemical recycling is therefore unclear and may likely vary between solutions. However, it is likely that a waste management system will include both mechanical and chemical recycling, as opposed to only one of the two options, thereby the two pathways may supplement one another upon actual implementation.

4.5 Energy consumption

The energy consumption was also modelled for the different treatment pathways. However, due to lack of data, it was not possible to obtain sufficient insight into the electricity and heat required to power each process. Thus, this balance remains the most uncertain among the three balances. In **Table 8** an overview of the overall consumption and recovery of energy can be found. As can be seen, the incineration pathway has the largest energy recovery, recovering ≈35,500MJ and ≈36,400MJ with and without CC. The chemical recycling pathway is the most energy intensive pathway out of the three scenarios, as it consumes ≈976MJ and ≈1149MJ with and without CC. This does not support the statement by Garcia and Robertson (2017), in terms of chemical recycling being less energy intensive compared to mechanical recycling. In conclusion, more work needs to be done to obtain more accurate energy data, in particular regarding the chemical recycling pathway.

Table 8. The energy consumption and recovery of the three scenarios in MJ/tonne, with and without CC.

	Scenario A	**Scenario B**	**Scenario C**
	With CC		
Consumption	378	777	976
Recovery	35,500	24,500	27,300
	Without CC		
Consumption	602	959	1,150
Recovery	36,400	25,200	28,000

Note: *Scenario A=Incineration, Scenario B=Mechanical recycling, Scenario C=Chemical recycling.*

Data obtained from The Danish Environmental Protection Agency (2022) and Bisinella et al. (2021).

4.6 Data uncertainties

Overall, each of the balances for all three treatment pathways were modelled using data that was as representative of Danish conditions as possible. However, in many cases it was not possible to obtain data from Denmark or at plant level. In fact, Danish data was only obtainable for the incineration pathway, and for the composition of plastic waste. In the remainder of the pathways, the vast majority of the data obtained was based on European studies instead. It is assumed that these are fairly representative of Danish conditions, however, more work should be done to assess these values. This is especially the case for the chemical recycling pathway, where very limited data were found. To improve data availability, literature data should be supplemented by transparent and consistent data from industry.

More work could also be done to assess the data uncertainty for each parameter, to provide more clarity regarding the needs for improvement of data. Based on current data availability, definition of data uncertainty ranges for the chemical recycling pathway is particularly challenging. Updating the model to include full uncertainty analysis, rather than applying min-max values, would allow users a systematic basis for identification of parameters that may critically affect the results.

5. Conclusion

This paper has presented a model that compares the mass, carbon and energy balance of three different treatment pathways of post-consumer plastics, namely (1) incineration with energy recovery (with and without CC); (2) mechanical recycling and (3) chemical recycling via pyrolysis of PE, PP, and PS, and glycolysis of PET. The model provides a user-friendly structure, such that parameters can be easily refined once higher-quality data is obtained. In addition it allows the user to enter a data range for each parameter, such that the model can run on either current or optimal conditions (labelled 'worst case' and 'best case' conditions).

The model assessed the mechanical recycling pathway as having the highest recycling rate, compared to the chemical recycling pathway, when run on worst case conditions. Here, it was found that the mechanical recycling pathway had a recycling

rate of ≈22%, and the chemical recycling pathway had one of ≈14%. At the polymer level, PET was found to have the highest recycling rate in both treatment pathways, yielding a recycling rate of ≈36% and ≈25%, respectively. When running the model on best case conditions, the mechanical and recycling pathways were found to have recycling rates of ≈55% and ≈57%, respectively. And at the polymer level, PET was found to have the highest recycling rate in the chemical recycling pathway, with ≈79%, and PE in the mechanical recycling pathway, with a recycling level of ≈68%.

With regard to data, more information is needed regarding yields and energy consumption of chemical plastic recycling at plant level, in order to provide more realistic results. It is recommended that industry to a larger extent offers transparent mass, carbon and energy balances for chemical recycling processes, in order to supplement the relative few data available in research literature. Regardless, the model is structured such that data is easily updated if more accurate estimates are available.

In summary, this report contributes with a consistent tool to compare mass, carbon and energy flows of the three treatment pathways assessed. In addition, it provides an overview of the data that is currently available for these three pathways which should be helpful to future life cycle assessments, analysing these treatment routes.

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