Master Thesis

Life Cycle Assessment of Pyrolysis Process of Plastic Waste at the Chemelot Site

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Summary

Plastic causes numerous issues due to its accumulation in the environment. In the Netherlands, 60% of all plastic is incinerated, which has a great impact on carbon dioxide emission and consequently on climate change. Chemical recycling represents a solution to seal the gap between incineration and mechanical recycling. A proposed chemical recycling option is pyrolysis of plastic waste. By adding pre and post treatment steps, pyrolysis oil with similar characteristics as naphtha can be derived from the process. This can be used in a steam cracker to produce polyolefins. Chemelot's vision is to become carbon neutral by 2050. Thus, in the Brightsite research programme, a collaboration between TNO and Chemelot, the carbon footprint of the planned pyrolysis process can be investigated. This study conducts a Life Cycle Assessment (LCA) to determine the global warming potential of 1 ton of mix plastic waste stream, classified as DKR 350, treated in the pyrolysis process. Based on the resulted carbon footprint it was concluded that the whole pyrolysis of plastic waste emits 866 CO_2 eq. by pyrolyzing only 28% of the waste stream. 75% of all the emissions are assigned to incineration of leftover plastic. Scenario analysis has been performed, which showed that additional mechanical recycling of PET and using DKR 310 film waste stream significantly reduces the current emissions of the base case scenario.

Contents

1 Introduction

1.1 Background

The large plastic production dates back to the 1950s and has since grown exponentially. 70 years later it is estimated that more than 6,300 million tons of global plastic waste has accumulated in landfills and the environment[1]. In 2017 almost 350 million tons were produced and if the worldwide production continues at the same pace, these numbers could quadruple by 2050[2] [3]. The widespread use of plastic is due to its low costs, versatile application and durability. It is made of petrochemical hydrocarbons with additives such as flame-retardants, stabilizers, and oxidants that make it difficult to bio-degrade and they stay in the environment for decades[4].There are more than 30 types of plastic used in a vast number of applications in various sectors. In the EU, one person on average uses 100 kg of plastic per year[5]. Such extensive use of this material causes enormous strains on the environment.

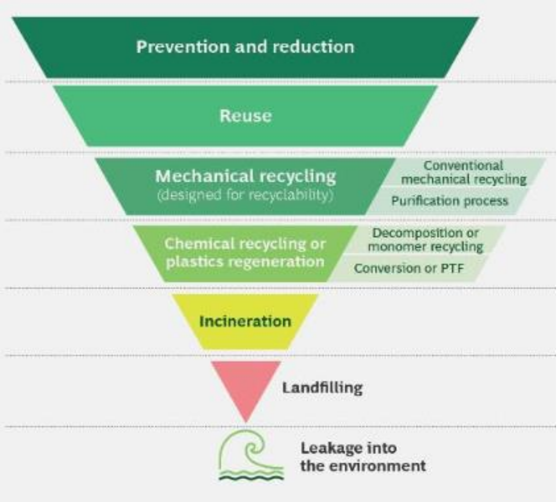
In order to solve all these major environmental issues, alternatives such as recycling and energy recovery methods have been developed. In Europe alone, 60 million tons of plastic waste were generated in 2016. Less than half of that has been collected, from which only 31,1% was recycled and 41,6% was incinerated, the rest was landfilled.[2] This shows that the percentage of plastic waste which ends up in landfills globally is still very high. In developing countries, the recycling rate is close to 0%, since the open or landfill disposal is a common practice for plastic waste management and it is also the perfect habitat for insects and rodents, which may cause diseases. In addition, due to rapid urbanization, the land available for landfills, especially in cities, is reducing[3]. A densely populated country such as the Netherlands has little space intended for landfilling, therefore the Netherlands waste management consisted of 2% landfilling, 40% recycling and 58% incineration in 2017[6]. Even though incineration recovers energy, it still heavily contributes to the climate change and air pollution. Consequently, it lies at the bottom of the waste hierarchy[7].

The Dutch government took an extra incentive within the goals of the European Union to achieve a 50% reduction in the use of primary raw materials by 2030 and become completely circular by the end of 2050[8]. In order to reach this target, the recycling processes have to be optimized and strive to achieve the highest percentage possible in the waste treatment options, as well as keep the environmental impact at the minimum.

Recycling still has a low share in plastic waste management due to its highly complex collection process, separation of several different flows, and cleaning prior to the recycling process, which today is very labour intensive[9]. Mechanical recycling operates optimally when plastic is collected separately; such is the case with Polyethylene terephthalate (PET) bottles. Other waste streams such as mixed waste is very difficult to manage with mechanical recycling due to the variety of additives to adapt their characteristics. It usually results in down cycling into lower quality products which do not satisfy markets demand of high value plastic[10]. Overall, companies in recycling and waste management field are struggling with two main problems. Firstly, most of the high value recycled plastic waste is already being recycled and secondly, the prices (quality) of secondary raw materials are low, compared to those of the primary raw materials. This stimulates the development of new technological solutions to increase the overall usage of recycled plastic in new products[11].

Currently in Europe, over 5 million tons of plastic waste is mechanically recycled and only around 50,000 tons of plastic waste is recycled chemically[12]. Chemical recycling is able to seal the gap between mechanical recycling and incineration as shown in Figure 1[13]. It provides potential for handling the complexity of plastic products such as multi-layer materials or plastic that include numerous harmful substances as additives. In this way, the recycled waste streams can be expanded to more complex and contaminated ones. In addition to that, it ensures a truly closed loop recycling of materials and provide raw materials such as hydrocarbon[11].

Figure 1 The pyramid of plastic waste management.



Solvolysis, dissolution and pyrolysis are some of the prime technologies in chemical recycling. A combination of these processes will likely ensure the improvement in current recycling processes. In terms of LCA, the best way to achieve that would be by keeping the same polymer structure throughout dissolution. This is a process which separates one polymer from a mixture of other polymers in multilayers films. The downside of this process is the use of hazardous organic solvents. However, the polymers remain intact, which leads to lower environmental impact. The second best option is to break down the polymers into monomers and repolymerize them.

Solvolysis attains the highest monomer recovery rate; however, the feedstock is limited to polyesters and polyamides. Pyrolysis is the least preferred option regarding the CO_2 avoidance. Nevertheless, it is a very valuable process when included in a refinery infrastructure to produce feedstock for a steam cracker. This is also the focus of this study[14].

1.2 Pyrolysis

Pyrolysis is a process by which a solid (or a liquid) undergoes thermal degradation to generate a mixture of hydrocarbons without interaction with oxygen[15]. In the case of plastic, the polymeric materials are heated to high temperatures in order to break down their macromolecular structure into a wide spectre of hydrocarbons, which are found in the form of synthetic oil, gas and char. The composition of these outputs may vary widely depending on the variety of chemical compounds. This process allows recovery of reaction products with added value, which can be used later on as raw materials for petroleum derived products. Both liquid and gaseous fractions of such products compose of complex combination of hydrocarbons, which depends solely on the composition of the plastic or biomass feed stream[16].

Pyrolysis is a mature technology. There have been years of research behind optimizing the process to the level where the feedstock challenges and end product needs are satisfied. A big number of commercial plants currently operate with biomass feedstock, whereas plastic pyrolysis is gaining momentum at the moment[11].

Plastic feedstock delivered to the plant site might vary in size and shape, which could cause problems while feeding it to the pyrolysis reactor. Therefore, it is necessary to shred and ground it to the uniform size for easier feeding process. Another optional step in preparing the feedstock is its densification. This process uses blending to form high mass and energy dense feedstock. Following the pre-treatment, the feedstock enters the pyrolysis chamber where thermal or catalytic decomposition takes place in an oxygen-free environment. Usually, the chamber is flooded with steam or nitrogen gas. The choice of reactor is essential when defining the desired product spectrum. There is a distinguished number of reactors when doing a slow and fast pyrolysis. On the one hand, slow pyrolysis operates under long retention time of several minutes and low temperatures, with the main product being charcoal. On the other hand, fast pyrolysis occurs in a span of a few seconds in a high-temperature settings to produce oil and lighter fractions of liquids and vapour[17]. Heat and mass transfer play a crucial role in this step. In the last step, pyrolysis product is treated with hydrogen to remove all the impurities and heteroatoms such as sulphur and oxygen to improve the quality of the oil.

2 Conceptual design

2.1 Research aim and relevance

This study has been conducted within the frames of the Brightsite programme created by TNO and industrial area Chemelot, in order to support Chemelot's vision to become carbon neutral by 2050. Chemelot is a cluster of chemical companies known for their strong inclusion of utilities and services, which groups together more than 50 factories and R&D facilities[18]. Half of the infrastructure at Chemelot belongs to SABIC, which is the fourth largest chemical producer in the world[19]. They own three Low-density polyethylene (LDPE) production lines, two high-density polyethylene (HDPE) production lines, and two polypropylene (PP) reactors, as well as two naphtha crackers (NAK3 and NAK4)[20]. Their main interest is to produce high quality oil through pyrolysis of plastic waste. Oil will be used as feed for their cracker to produce polyolefins in a circular and CO_2 neutral way in the long run. The goal of this research is to investigate the global warming potential of implementing a pyrolysis plant on the site.

2.2 Research questions

In line with the research aim, this main research question is presented:

What is the carbon footprint of the possible pyrolysis plant at the Chemelot site?

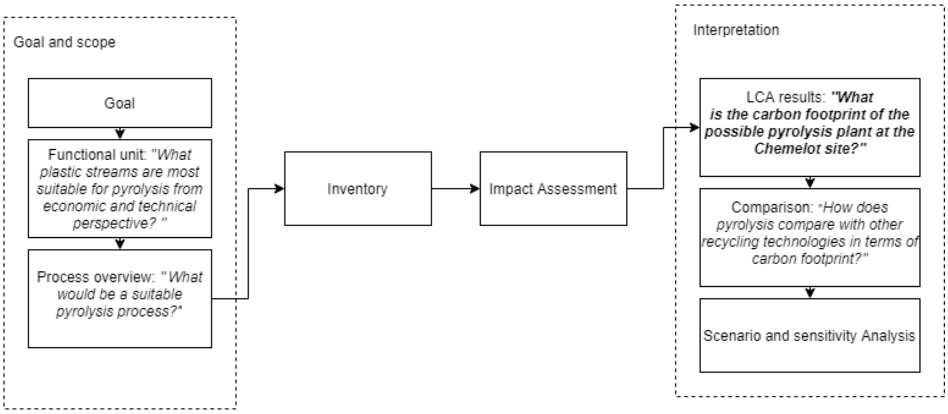
The following sub-questions are defined:

1. **What plastic streams are most suitable for pyrolysis from economic and technical perspective?**
2. **What would be a suitable pyrolysis process?**
3. **How does pyrolysis compare to other recycling technologies in terms of carbon footprint?**

2.3 Research framework

From the research question shown, the research framework can be constructed.

Figure 2 The research framework



The research framework of this Life Cycle Assessment consists of four parts as seen in Figure 2: Goal and scope, Inventory, Impact Assessment and Interpretation. The research questions are set to fill in the most crucial knowledge gaps in this study.

The goal and scope of LCA sets the context of the study. In this section it is important to state the goal of the research, its functional unit and the process overview with the system boundaries. Since the goal has been already stated in the research aim of this report, the first sub-question helps to define the functional unit, which in this case is the amount of certain stream of plastic treated through pyrolysis. The second sub-question focuses on the best suitable overall process which helps to construct the system boundaries and with that also the inventory.

From this point on, a thorough inventory of the energy and materials can be conducted and the impact assessment method can be defined.

In the study, the plastic waste input is considered as burden-free. However, the environmental benefits of the products are included, by subtracting the impacts of the conventional production of these products from the impacts of the recycling technology itself. The LCA follows guidelines as laid down in the ISO standards[21] and handbooks[22]. The inventory database ecoinvent 3.5 present in the LCA software SimaPro 9.0 serves as a basis for the analysis. The ReCiPe 2016 method[23] is chosen as an impact assessment method for this study. The method in this research takes the midpoint approach, where the characterization factors are located at the point where the environmental mechanism is identical for all the environmental flows assigned to one impact category. There are 18 different impact categories and this study focuses on the impact category of climate change, as this is the most significant category in terms of the end life of plastic.

The results from the impact assessment are summarized during the interpretation phase. The main results are the most important ones and answer the main research question. To put said results into perspective, the last sub-question focuses on the comparison of pyrolysis process to other technologies recycling the same waste stream. The last section focuses on extensive scenario and sensitivity analysis in order to test all the assumptions taken in this study.

3 Life Cycle Assessment

3.1 Goal

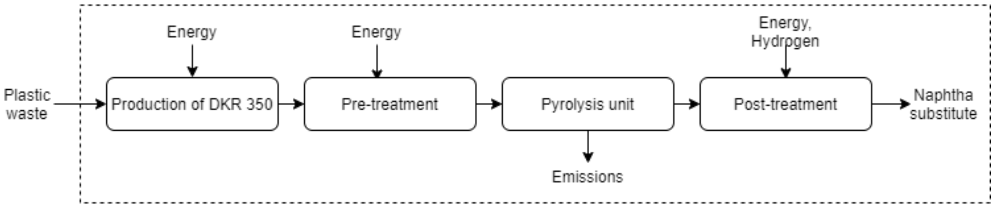
As already stated above, the goal of this LCA is to assess the global warming potential of the pyrolysis process of plastic waste at Chemelot site.

3.2 Functional unit

CE Delft report on “Exploration of Chemical Recycling” provides an overview of the amount of available plastic waste streams that could be used as inputs for chemical recycling in the Netherlands through different processes such as pyrolysis, gasification, solvolysis and depolymerization[24]. Potential feedstock for chemical recycling has no profitable financial flows in mechanical recycling and a big part of the waste stream cannot be mechanically recycled. Mixed plastic waste stream, also classified as DKR 350 fits this criterion as well. This is the largest stream delivered by households and it the most suitable one for pyrolysis. Therefore, the functional unit of this study is *1 ton of mixed plastic waste stream – DKR 350*. In addition, the results are also expressed in 100 ktons of DKR 350, to grasp the potential of overall use of annual supply of the waste stream for chemical recycling[24].

3.3 Process overview and system boundaries

Figure 3 Visual representation of the studied system.



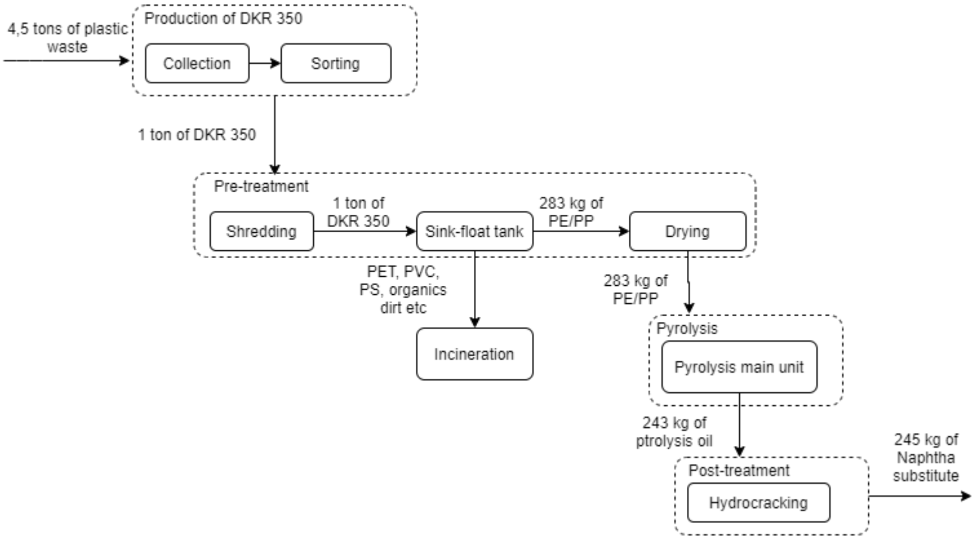
The system boundaries visualized in Figure 3 are divided into four stages: production of DKR 350, pre-treatment, pyrolysis process itself and the post-treatment of products. In the first stage, waste is collected and sorted into different plastic waste stream which comply with the DKR standards at the sorting centre. Mixed plastic – DKR 350 waste stream is therefore transported to the Chemelot site, where the pyrolysis plant is located. Before entering the pyrolysis reactor, the plastic needs to undergo pre-treatment. During pre-treatment, the waste stream is firstly shredded and put into the sink-and-float tank, where all the plastic is cleaned and the impurities as well as the undesirable plastic types such as PS, PVC and PET are removed. After the sink-and-float method, the plastic going into the reactor is dried, while the leftover plastic is incinerated. Once the plastic waste is turned into the pyrolysis oil, it is hydrocracked into the oil similar to that of naphtha composition.

4 System description and Inventory

4.1 Process overview with mass balance

The following Figure 4 depicts the process overview with the mass balance.

Figure 4 Process overview with mass balance.



4.2 Inventory

4.2.1 *Production of DKR 350*

Plastic waste has different origins and can be collected in various ways. In 2017 it was reported that 47,5% of all the collected plastic waste packaging was recycled[24]. Once the collected plastic enters the sorting facilities, different residual components are removed, such as organic material, glass and metals. Later on, the plastic films are taken out with ballistic separators or wind-sifters. At the end of the process, all the rigid plastic object are put through a near infrared (NIR) sorting machine, which separates the four major plastic products: PP, PE, PET and MIX plastic. These streams must comply with DKR standards (Deutsche Gesellschaft für Kreislaufwirtschaft und Rohstoffe mbH)[25]. These specifications allow the legal transport of these waste streams to recycling facilities.

The following base assumptions are made for this step:

- 45% of the recycled plastic is sorted into mono-stream plastic and the remaining 55% into mixed plastic (DKR350). During the production of DKR 350, another 15% of the mixed plastic is lost in the process of sorting. Consequently, 22% of all the collected plastic ends up as mixed plastic waste (DKR350), which can be used in this study[24]. It was calculated that 4,5 tons of plastic have to be collected in order to obtain 1 ton of DKR 350 waste stream.

- CE Delft[6] mentions three major ways to collect plastic waste packaging: collection via deposit, collecting plastic separately and collecting plastic in residual waste. It was assumed that collection via deposit has a negligible impact, because the trips to the supermarket to hand in the deposit is not the main reason to undertake the trip and in the Netherlands, many trips to the supermarket are by bike, which has no direct CO2 emissions.
- A transport distance of 100 km by a collection truck for both separately collected plastic and residual waste was assumed. After collection, the different plastic streams have to be sorted into the DKR 350 specifications. For this, an energy use of 4 kwh/ton input was assumed. Since the emission were mass allocated to 1 ton, the energy use for sorting the 3,5 tons of the collected plastic is not accounted in this study.

4.2.2 DKR 350 composition

The data for the mix plastic DKR 350 composition was taken from Brouwer et al. (2018)[26], where the compositional data of the MIX sorted product made from recovered plastic packaging waste offered to mechanical recovery facilities was analysed. The data was taken from 21 samples between the years of 2009 and 2013. The exact composition is depicted in the following Table 1.

Table 1: Composition of the mixed plastic DKR 350 waste stream based on Brouwer et al. (2018)[26].

Mixed Plastic DKR 350	**%**
PET	18,5
PE	19,4
PP	8,9
PVC	0,2
PS	0,4
Not identifiable	3,2
Misc plastic	0,1
Laminated packages	1,6
EPS (expanded ps) blocks	0,1
Silicone tubes	0,1
Non-packaging plastics	3,0
Organics	5,2
Paper/cardboard	4,4
Metal	0,6
Moisture and dirt	34,5
Total	100

4.2.3 *Pre-treatment*

4.2.3.1 *Transport*

It is expected that the waste stream is transported from the sorting centre of Renewi to the Chemelot site[20]. The average transport distance between the sorting centre of Renewi and Chemelot site is estimated to be 100 km. There are DKR 350 bails coming from the ports of Rotterdam and Antwerp; however, only occasionally, thus they are not taken into account. It is assumed that one truck can transport one ton of DKR 350 to the site. The type of truck is assumed to have EURO 6 norm, as this covered 60% of the rides in 2019[27].

4.2.3.2 *Shredding*

In order to facilitate homogenization the incoming waste stream is shredded with the estimated electricity use of 4 kWh per ton of DKR 350[28]. The waste stream is shredded to an average size of a few hundred millimetres and it is assumed that there are no material losses. The losses would appear in the form of dust or micro particles, which cannot be recovered. However, no data on that was found.

4.2.3.3 *Float-and-sink method*

In the next step, the shredded waste material goes into the float-and-sink tank, which is filled with water and it is used to separate different types of plastics based on their specific gravity in the mixed plastic streams. In that way, rigid PP/PE or film have a lower density, which causes the plastic to float, while the contaminant particles and the types of plastic such as PET, PVC and PS sinks to the bottom to be discarded [29]. For this process, a float-and-sink tank was chosen with the capacity of 1000 kg of input mixed plastic stream. The estimated energy use amounts to 2,82 kWh per 1 ton of input, which is taken at the 75% of the stated full capacity of the machine[29].

The water consumption can be given as mass percentage from the stream throughput (rule of thumb)[30].In this case 15 wt% of PE/PP mass throughput is taken for water consumption which amounts to 43,4 kg of water usage per 1 ton of input.

4.2.3.4 *Drying*

The incoming mix of plastic undergoing the drying comprises of 283 kg of wet PE and PP with the assumption that 10% of solid content is moisture. The dryer is assumed to be a rotary type[31] with the reported efficiency in literature around 83-95%[31], therefore the estimated efficiency in this case is 90%. The energy demand is calculated following Equation 1 below. The explanation of the calculation is given in Table 2.

Equation 1 : Equation to calculate heat requirements[40].

$$E_{drying} = m_{w-eva} * \left(C_{p,water} * \Delta T + \Delta H_{v,water}\right) + \frac{(m_{w-re} * C_{p,water} + m_s * C_{p,s}) * \Delta T}{\eta_{drying}}$$

Table 2 Explanation of the calculation.

Item	Description	Amount	Unit
E drying	*energy demand for drying*	31,69	kWh
m (w-eva)	*mass of evaporated water*	25,47	kg
m (w-re)	*mass of remaining water*	2,83	kg
m (s)	*mass of solid substance in*	283,00	kg
C (p, water)	*specific heat capacity water*	4,20	kJ/(kg˚C)
C (p, s)	*specific heat capacity of PP*	1,74	kJ/(kg˚C)
ΔT	*temperature difference (20 °C) and drying (105 °C)*	85,00	˚C
ΔH (v, water)	*evaporation heat of water*	2257,20	kJ/kg
η (drying)	*efficiency of drying*	0,90	

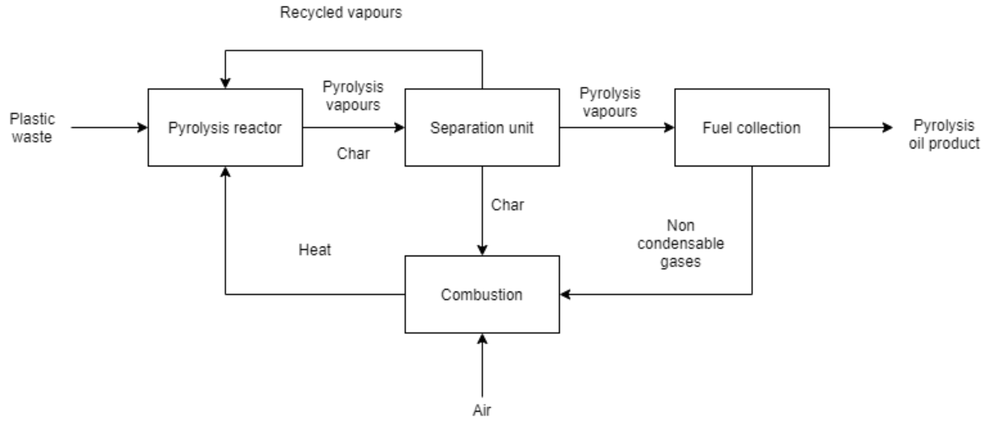
4.2.3.5 Incineration of left over plastic

All the left over plastic that is removed by the float-and-sink method and does not contain PE or PP is assumed to be incinerated. This also includes the dirt, organics, metal, etc. In this case, 545 kg of plastic, including 10% of moisture content, is incinerated with energy recovery. Efficiency of the heat and electricity recovery is estimated to be 20,2% and 16,9% respectively. These are the paramters in Simapro software. To correctly model the incineration process in Simapro, the energy and carbon content of the left over plastic has been calculated.

4.2.4 Pyrolysis process

The pyrolysis system investigated in this study was developed by a recycling company based in the UK[32] and its data has been entirely attained from Fivga et al. (2018)[32]. It is shown in Figure 5 that it consists of four functional units: pyrolysis of plastic waste, char separation, collection of the pyrolysis fuel oil and combustion unit. Combustion unit generates the thermal energy by combusting char in a secondary fluidized bed reactor. This thermal energy is delivered to the pyrolysis reactor, therefore no external energy sources are needed to operate the pyrolysis unit. The process capacity in the article is 100 kg/h.

Figure 5 An overview of the pyrolysis system for the production of fuel.



The pyrolysis process occurs in a fluidized bed reactor, in an inert atmosphere, at atmospheric pressure and at a fixed reaction temperature of 530 °C. Nitrogen is used as a fluidizing gas only at the start-up of the plant and it is later replaced with the recycled pyrolysis vapours from the separation unit. The product vapours enter the solid separation section at the temperature of 530 °C, where char is separated by using a ceramic hot vapour filter unit (HVF). The before-mentioned vapours are recycled back whereas the non-recycled pyrolysis vapours are kept above their dew point until they reach the fuel collection unit[32].

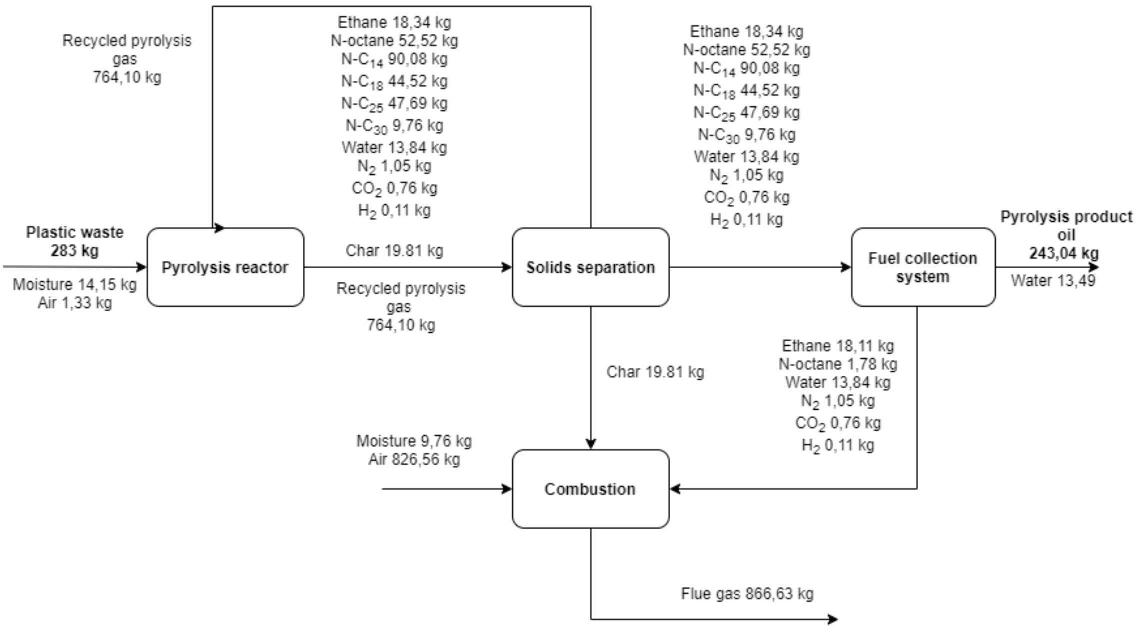
There are four heat exchangers in the collection unit. The first one collects heavier pyrolysis vapours and it is set at 400 °C. The next series of heat exchangers and separators are set at 200 °C, 80 °C and 25 °C respectively. Water is provided in a closed loop at 40°C to the first three exchangers and at 10 °C to the fourth exchanger. Non-condensable gases separated in the fuel collection system are combusted together with char in secondary fluidized bed reactor, which provides thermal energy to the whole system. A fan supplies the air at a 10% excess to the fluidized bed combustor and provides the required pressure drop across the fluidized bed[32].

4.2.4.1 Flows composition

In Fivga et al. (2018)[32] article, the process flowsheet of the plastic waste pyrolysis process was modelled by the Aspen HYSYS process simulation software. In addition, all the material balances, energy and utility requirements were estimated by using the Peng-Robinson thermodynamic property method, which is usually recommended for modelling refinery process unit operations. It provides accurate results for hydrocarbon and light gases systems, such as H_2 and CO_2. Because only the product yield data of pyrolysis reactor was provided, the Aspen HYSYS yield shift reactor is used to simulate the pyrolysis reactor. This is considered in the case when stoichiometry and the kinetics are unknown, but the product yield is available. Since the process operating conditions have a large impact on the different primary and secondary products, the modelled hydrocarbons in the products fuel are not straightforward.

For this LCA, the inputs based on the 100 kg of plastic waste are scaled up to 283 kg, which is the fraction of waste stream that it is put into the pyrolysis reactor. Table 3 shows the process scheme and the mass balance.

Table 3 Mass-based process scheme of the pyrolysis unit.



4.2.4.2 Base assumptions, mass, energy and carbon balance

Feedstock:

- The plastic waste feedstock contains 283 kg of PE and PP and its elemental composition amounts to 86 wt% of carbon and 14 wt% of hydrogen, which correspond to the one stated in Fivga et al. (2018)[32] with 85 wt% carbon and 15 wt% of hydrogen with a slight deviation.

- The higher heating value (HHV) is calculated by the following equation:

$$HHVdry\left(\frac{MJ}{kg}\right) = 0{,}3491 * C + 1{,}1783 * H + 0{,}1005 * S - 0{,}1034 * O - 0{,}014 * N - 0{,}0211 * A$$

  which in this case amounts to 48,00 MJ/kg.
- And calculated lower heating value (LHV) of feedstock follows this equation: $LHVdry\left(\frac{MJ}{kg}\right) = HHVdry - 2{,}442 * 8{,}936 * H/100$, which results in 44,51 MJ/kg.

Pyrolysis reactor unit:

- For carbon balance calculation, composition of char of 100 wt% carbon and composition of air of 22 wt% oxygen and 78 wt% nitrogen is taken.
- The recycled pyrolysis gas was not included in the carbon balance calculation.
- Carbon balances have been calculated for each unit separately; however, in the report it is presented only for the overall process.

Table 4 Mass balance of the pyrolysis reactor.

Pyrolysis reactor- Mass balance					
Input			Output		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
Plastic waste	283,00	kg	*Ethane*	18,34	kg
Moisture	14,15	kg	*N-Octane*	52,52	kg
Air	1,33	kg	n-C_{14}	90,08	kg
Recycled pyrolysis gas	764,10	kg	n-C_{18}	44,52	kg
		kg	n-C_{25}	47,69	kg
			n-C_{30}	9,76	kg
			Water	13,84	kg
			N2	1,05	kg
			CO2	0,76	kg
			H2	0,11	kg
			Char	19,81	kg
			Recycled pyrolysis gas	764,10	kg
Total	**1062,58**	**kg**		**1062,58**	**kg**
Total – recycled gas	**298,48**	**kg**		**298,48**	**kg**

Separation unit

In the separation unit, the mass and carbon balance stays the same, as no chemical reactions take place. The only difference is that char is separated and directed into the combustion unit, while the pyrolysis vapours continue to the fuel collection system. The recycled pyrolysis gas is recycled back into the pyrolysis reactor; however, it is not included.

Fuel collection unit

In Table 5, the mass balance of fuel collection unit is presented.

Table 5 Mass balance of the fuel collection unit.

Fuel collection unit- Mass balance					
Input			Output		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**

			<u>Pyrolysis product oil</u>		
Ethane	18,34	kg	*Ethane*	0,23	kg
N-Octane	52,52	kg	*N-Octane*	50,74	kg
n-C_{14}	90,08	kg	*n*-C_{14}	90,11	kg
n-C_{18}	44,52	kg	*n*-C_{18}	44,52	kg
n-C_{25}	47,69	kg	*n*-C_{25}	47,69	kg
n-C_{30}	9,76	kg	*n*-C_{30}	9,76	kg
Water	13,84	kg	<u>*Non-condensable gases*</u>		kg
N2	1,05	kg	*Ethane*	18,11	kg
CO2	0,76	kg	*N-Octane*	1,78	kg
H2	0,11	kg	*Water*	13,84	kg
			N2	1,05	**kg**
			CO2	0,76	**kg**
			H2	0,11	**kg**
Total	**278,67**	**kg**		**278,67**	**kg**

Combustion unit

In Table 6 and

Table 7, the flue gas composition and mass balance of combustion unit are given.

The amount of emitted CO_2 was calculated by the amount of (fossil) carbon combusted. The following flue gas composition aligns with the elemental composition of the inputs and it also correspond with the literature, where it says that the typical flue gases from coal-fired boilers may contain 12-14% of CO_2, 8-10% of H_2O, and 3-5% of O_2 and 72-77% N_2[33].

Table 6 Flue gas composition.

Flue gas composition			
Item	**Amount**	**Unit**	**wt%**
N_2	635,05	kg	***73%***
CO_2	131,77	kg	***15%***
O_2	54,53	kg	***6%***
H_20	45,28	kg	***5%***
Total	866,63	kg	***100***

Table 7 Mass balance for the combustion unit.

Combustion Unit- Mass balance					
Input			Output		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
			<u>Flue gas composition</u>		
Char	19,81	kg	*N_2*	635,05	kg
Moisture	9,62	kg	*CO_2*	131,77	kg

Air	815,04	kg	*O_2*	54,53	kg
Ethane	18,11	kg	*H_20*	45,28	kg
N-Octane	1,70	kg			
Water	0,42	kg			
N_2	1,05	kg			
CO_2	0,76	kg			
H_2	0,11	kg			
Total	**866,63**	**kg**		**866,63**	**kg**

Overall process of pyrolysis unit

The mass and carbon balance of the whole process are presented in Table 8 and Table 9.

Table 8 Mass balance of the overall process.

Overall process- Mass balance					
Input			Output		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
Plastic waste	283,00	kg	Pyrolysis fuel oil		
Moisture	23,77	kg	*Ethane*	0,23	kg
Air	816,37	kg	*N-Octane*	50,74	kg
			n-C14	90,11	kg
			n-C18	44,52	kg
			n-C25	47,69	kg
			n-C30	9,76	kg
			Water	13,49	kg
			Flue gas		kg
			N_2	635,05	kg
			CO_2	131,77	kg
			O_2	54,53	kg
			H_20	58,58	kg
Total	**1123,14**			**1122,97**	

Table 9 Carbon balance of the overall process.

Overall process– Carbon balance		
	Input	**Output**
C	0,22	0,22
H	0,04	0,04
O	0,18	0,18
N	0,57	0,57
Sum	1,00	1,00

4.2.5 Hydrocracking unit

According to the product composition distribution in the Fivga et al. (2018)[32], the product oil does not contain any heteroatoms such as oxygen and sulphur, and it only contains paraffins. Therefore, technically, the hydro treatment is not needed in this case. For the base case scenario, only the hydrocracking unit is used, where the pyrolysis oil is hydrocracked into the composition similar to that of naphtha. Hydrocracking of the oil breaks carbon-carbon bonds and converts them into shorter-chain hydrocarbons, which are more suitable as a feedstock for the steam cracker. The hydrogen needed to hydrocracked pyrolysis oil to naphtha composition is calculated using Excel.

Base assumptions:

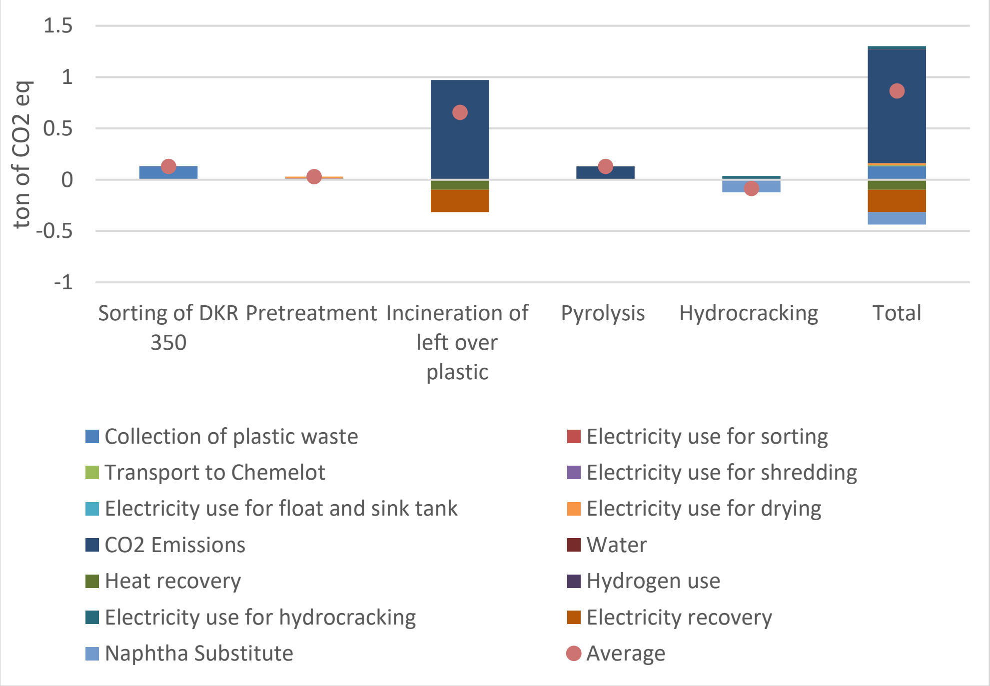
- 243 kg of pyrolysis oil with a product distribution of large hydrocarbons chains.
- 2,31 kg of hydrogen is needed to hydrocrack the pyrolysis oil to the naphtha (C_7H_{16}) substitute composition.
- Electricity consumption for a hydrocracking unit is estimated to be 0,22 kWh/kg of produced oil[34].

5 Environmental Assessment Results

5.1 Main results

Figure 6 details a CO_2 contribution of a pyrolysis treatment of 1 ton of DKR 350 waste stream to global warming potential. Total GWP is measured in terms of CO_2 as the equivalent substance. The results show that the total of processes of the pyrolysis life cycle generates 0,866 ton of CO_2 eq. The greenhouse gas emissions mainly come from the incineration of the leftover plastic, which amounts to 76% of the overall emissions. The second biggest emitter is the sorting process for production of DKR 350, as well as the pyrolysis unit. Their net values both amount to 0.131 ton of CO_2 eq. The hydrocracking unit emits in total negative 85 kg of CO_2 eq., because the product that comes out of the hydrocracking step is assumed to have the same composition as naphtha. Therefore, naphtha is modelled as an avoided product in this step, which results in negative emissions.

Figure 6 GWP of treatment of 1 ton of DKR 350 waste stream by life cycle process step.



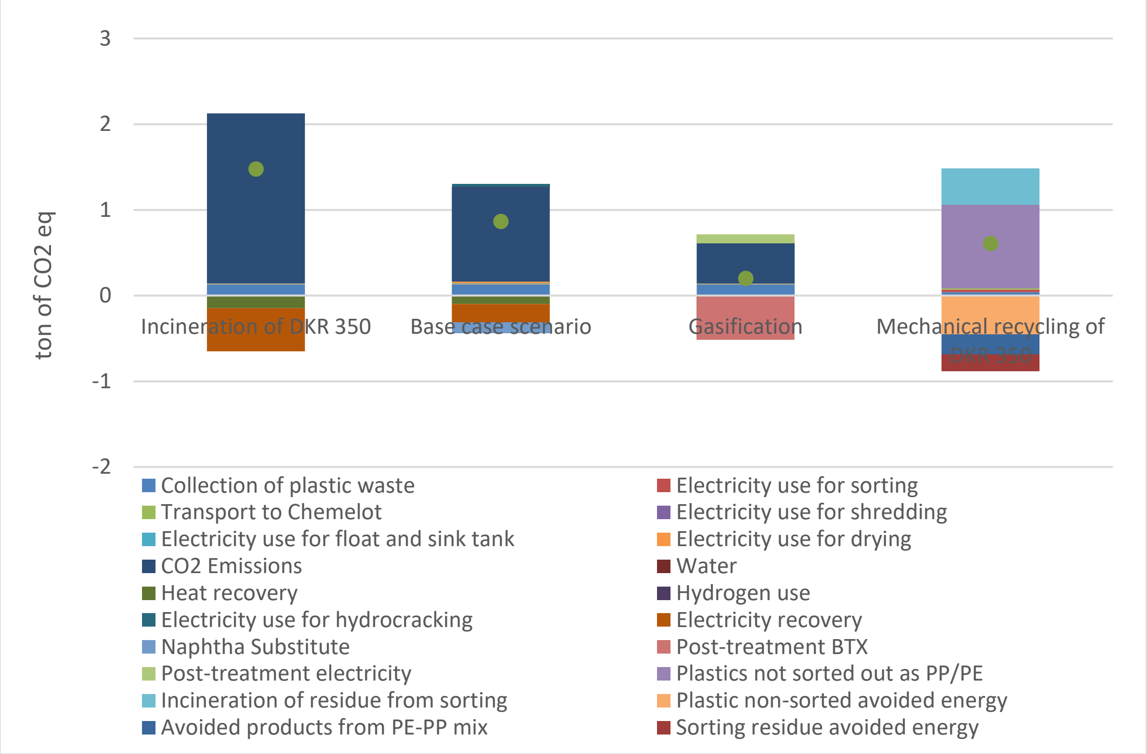
5.2 Comparison with other recycling technologies

Disclaimer: The recycling technologies cannot be entirely compared to each other because of the different final products. The only way a fair comparison can be made in this case is by comparing the treatment of DKR 350 through different recycling technologies, such as gasification, incineration and mechanical recycling.

DKR 350 can be treated with different recycling technologies. In order to really assess the advantages and disadvantages of pyrolysis of DKR 350, it needs to be placed along with other recycling alternatives such as incineration, gasification and mechanical recycling. In parallel to this research, a LCA of gasification process has been conducted with equal system boundaries and assumptions[35]. The LCA of gasification process of plastic waste is based on experimental data on MILENA - OLGA gasifier at the pilot scale, with three sets of products investigated: BTX + electricity, BTX + methanol and BTX + substitute natural gas (SNG) production. It has the same functional unit and the production step of DKR 350. The data on the incineration process as well as pyrolysis and gasification has been extracted entirely from the ecoinvent database. The production of DKR350 is also included for incineration to make a fair comparison.

The mechanical recycling of DKR 350 is also added to the comparison. DKR 350 is downcycled into a material that is used to make benches in parks. The LCA of mechanical recycling of DRK 350 is beyond the scope of this study, therefore the data has been taken from Ligthart et al. (2019)[36]. In this case, the DKR 350 composition has been assumed to consist of 40,4% of PP/PE, 35,2% of unsorted plastic going into incineration for energy recovery and 24,5% of non-plastic material that it is removed during the processing. Other assumptions regarding the collection, sorting and processes in Simapro have also been taken into account.

Figure 7 Comparison between the GWP of different recycling technologies in treatment of DKR 350.



In total, the GWP for base case gasification and pyrolysis is 202 kg and 866 kg CO_2-eq. respectively. The major difference between the gasification and pyrolysis is that gasification can process the whole ton of DKR 350 waste, whereas the pyrolysis can process only 28% of the waste stream. With that assumption, gasification avoids the additional steps of cleaning and drying in the pre-treatment and it potentially converts more plastic into valuable products. The major contribution of CO_2 emission in pyrolysis is the incineration of the leftover plastic.

The total emissions of incineration amount to 1,475 ton of CO_2 eq. That means that with pyrolysis, a 40% reduction of emissions can be achieved by only pyrolyzing 28% of the waste stream.

Mechanical recycling of DKR 350 amounts to 606 kg of CO_2 eq., which is 30% lower than the carbon footprint of pyrolysis. However, the PE/PP share in a study by Ligthart et al. (2019) is 12% higher than in the base case scenario, therefore more plastic in the base case scenario is incinerated. The differences in the CO_2 contribution also change due to the different choice of processes in the Simapro software between the studies. Here it is assumed that this could affect the results by up to -/+ 100 kg of CO_2 eq. The mechanical downcycling of DKR 350 would still result in lower emissions than pyrolysis; however, from the economical perspective, it is not profitable. The roadmap to circular plastic[37] states that mechanical recycling of DKR 350 showcases negative business case of -110 €/ton of DKR 350.

Additionally, the following Table 10 demonstrates the useful products that were created from 1 ton of DKR 350 through different recycling technologies.

Table 10 Comparison between technologies on useful product created out of one ton of DKR 350

Technology	Useful product/ ton DKR 350 used
Pyrolysis	245 kg of naphtha
Gasification	1.51 MW_eh of electricity
Mechanical recycling	404 kg of bulk of plastic for benches
Incineration	4025 MJ electricity recovery + 3188 MJ of thermal energy

5.3 Conclusion

To conclude, pyrolysis in the base case scenario reduces CO_2 emissions by 40% compared to incineration, but reaches similar CO_2 performance as mechanical recycling. Furthermore, it has higher CO_2 emissions than gasification due to a 60% lower input into the pyrolysis unit compared to the gasifier.

6 Scenario and Sensitivity Analysis

6.1 Introduction

In the previous chapters, an environmental assessment of a base case scenario with specific assumptions has been displayed. A number of assumptions were made regarding the yields and efficiencies of the processes, composition, energy mix etc. Changing these assumptions can potentially have large effect on the results. Therefore, a scenario and sensitivity analysis, in which some of the values of selected assumptions are varied, has been made. Said scenario analysis investigates alternative pathways whereas sensitivity analysis looks into the uncertainties in key parameters in the present and future.

The following topics for the scenario analysis have been selected based upon the possibility of improving the waste treatment stream and as well as having more options regarding feedstock for chemical recycling:

- **Mechanical recovery of PET**. In the base case scenario, it was assumed that the leftover waste stream of plastic is going to be incinerated. The waste stream included a significant portion of PET, which could potentially be mechanically recovered.
- **Changing the composition from DKR 350 to DKR 310.** There are other waste stream specifications that are suitable for chemical recycling besides the mix plastic waste stream of DKR 350. DKR 310 contains large amount of plastic films in the form of bags, shrink-wrapping film and labels. This is an interesting waste stream to consider, because it requires a second sorting step to increase its quality.
- **Theoretical full potential of pyrolysis.** Currently only 28,3% of mix waste stream is considered as an input to pyrolysis reactor. Hence, it would be interesting to consider what the potential of the pyrolysis process where 100% of the waste stream is converted into pyrolysis oil. The composition of DKR 350 is replaced with the 100% of PE/ PP, eligible for chemical recycling.

The following topics for the sensitivity analysis have been selected:

- **Sensitivity analysis of DKR 350.** In the base case scenario, the DKR 350 composition is based on the real samples taken throughout the years at the sorting facilities. However, the official requirements of the DKR stream vastly differ from the samples, especially in the reduced moisture content. In this analysis, the moisture content is removed to align with the composition of the stream that is required for further processing.

- **Post-treatment alternatives.** Regarding the post-treatment, the base case scenario is considered as an optimistic option, since it takes into account only the hydrocracking unit and the impurities and heteroatoms in the pyrolysis oil. In this sensitivity analysis, the environmental impacts are investigated by adding extra units of hydrotreatment by the company called CLARITER. Since the final composition of their product is not known, the impacts of different final product options are also considered.
- **Future electricity mix.** The environmental impacts of pyrolysis process in the future are modelled for the years 2030, 2040 and 2050. The data is taken from the integrated assessment model (IAM) E3ME-FTT-GENIE[38], which assumes that the climate targets are based on the maximum global warming of 2 ˚C scenario. The model was recommended by an TNO expert.

6.2 Scenario analysis

6.2.1 Mechanical recovery of PET

PET can be mechanically recovered with a profit. Since the DKR 350 stream includes almost 20% of PET plastic, it can be suggested that this portion is recycled mechanically, instead of being incinerated. Table 11 shows the base assumptions taken in this case.

Table 11 Assumptions on mechanical recycling of PET.

Assumptions on mechanical recycling of PET		
Item	**Amount**	**Unit**
Efficiency	81	%
Electricity use	0,855	kWh/kg of PET
Water	2,91	l/kg of PET

Figure 8 depicts the changes between the incineration step in the base case scenario and in this one. From 1 ton of mix plastic waste, 185 kg of PET is assumed to be mechanically recovered. Therefore, only 359,5 kg of plastic is incinerated compared to 545,5 kg in the base case. This is translated into lower CO_2 emissions. Virgin PET is modelled as avoided product, therefore it results in large CO_2 avoidance comprising of 464 kg of CO_2 benefit. There are 87 kg of CO_2 emissions from the electricity use for mechanical recycling of PET, which is quite small compared to the environmental benefit. The last bar depicts the joint effect of incineration of left over plastic and mechanical recycling of PET in this scenario. It can be seen that due to PET recovery, 666 kg of CO_2 emissions can be saved compared to the incineration step in the base case scenario.

An important remark has to be made here. DKR 350 stream generally contains very low quality plastic. Therefore, fully replacing virgin PET with the mechanical recycling PET trays from the DKR 350 waste stream is currently unlikely and it could be considered for a future scenario.[1]

Figure 8: Comparison between the incineration of the leftover plastic in the base case scenario and incineration with additional PET recovery.

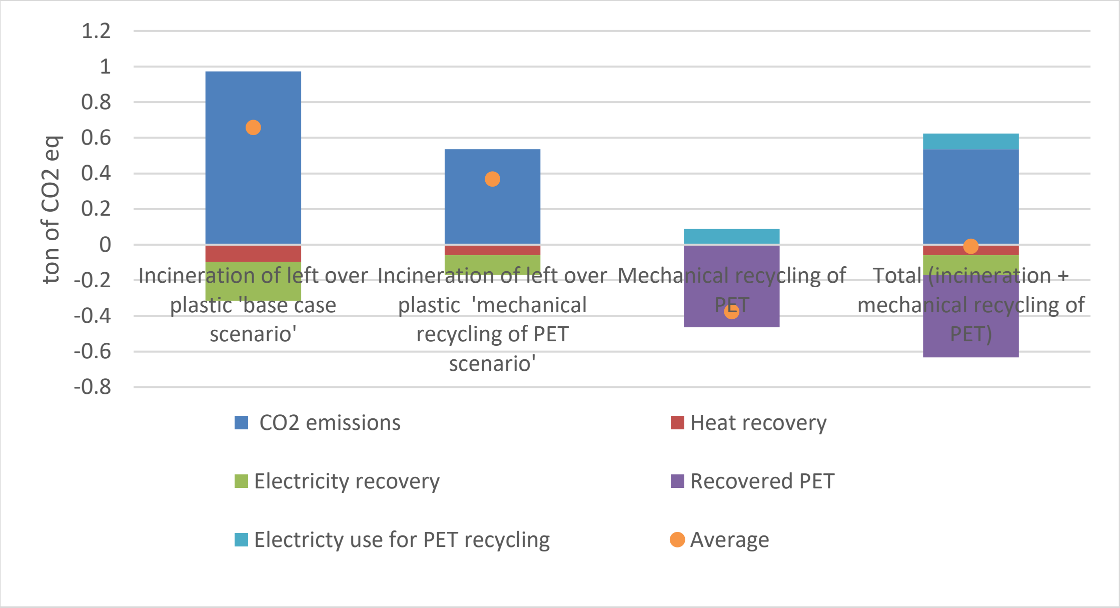


Figure 9 shows the detailed environmental profile of the base case scenario when PET is mechanically recovered. The graph demonstrate that the overall process causes 0,377 ton of CO_2 eq. The major part of greenhouse gas emissions originates from the incineration of the leftover plastic. Nevertheless, in total this could be reduced by mechanically recycling PET. All the emissions of the other process units remain the same.

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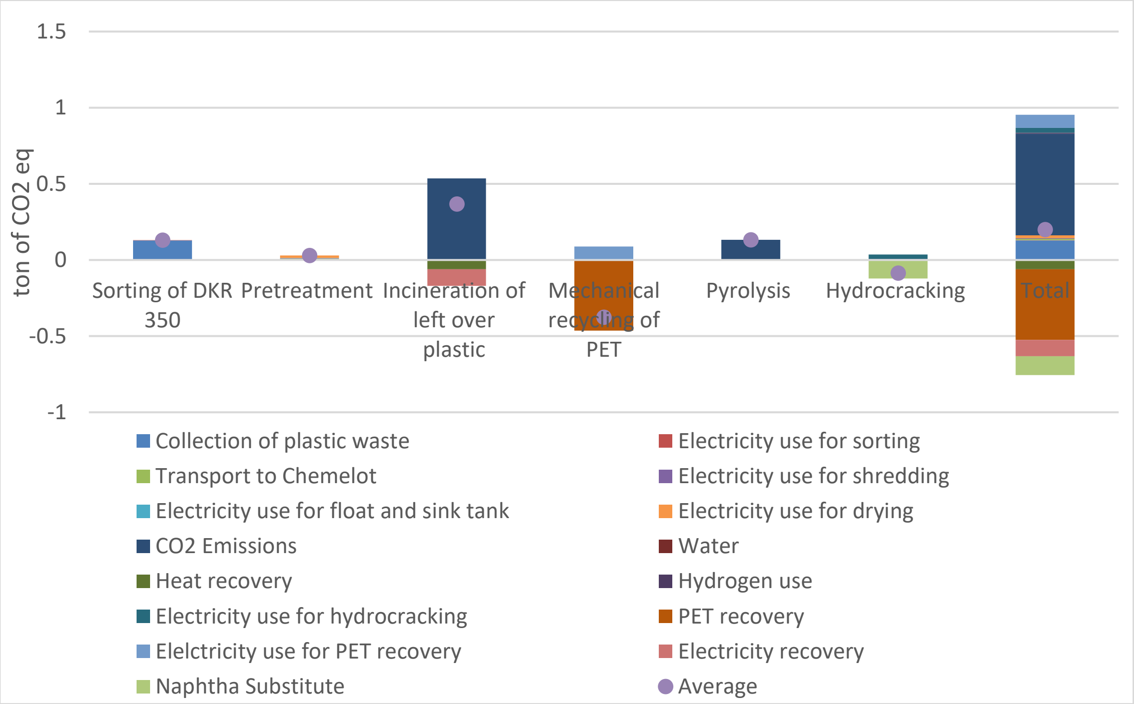
[1] At the Chemelot site, a 10 ktons/a demonstration depolymerisation plant was built in 2018. The plant depolymerizes PET trays into bi-hydroxyethylene terephthalate (BHET), which can be used to produce virgin-PET. This can be a very interesting option for Chemelot to consider, however it is outside of this research.

Figure 9: GWP of DKR 350 with a PET mechanically recovered scenario.



6.2.2 *DKR 310*

It has been estimated that a large portion of waste flows coming from households. such as foil (DKR 310), PE (DKR 329), PP (DKR 324), PET (DKR 328), mix (DKR 350) cannot be mechanically recycled in a profitable manner[37]. The DKR 310 and DKR 350 both showcase a negative business case in mechanical recycling of - 50€ and -110€/ton, respectively. This is due to the very low quality of PE in DKR 310 stream, which requires additional steps in mechanical recycling to increase the quality. That implies that DKR 310 can add more value when undergoing chemical recycling option as well. Hence, a scenario was made where 1 ton of DKR 350 was replaced with 1 ton of DKR 310.

The following Table 12 displays the comparison between the compositions of both waste streams. The most significant differences between the streams are the increase of the PE share and reduction of PET. The portion of PE is more than double in DKR 310, whereas PET percentage decreases from 18,5% to 2,6%.

Table 12: Comparison of the composition of mix plastic (DKR 350) and film plastic (DKR 310) waste streams.

Composition	DKR 350 (%)	DKR 310 (%)
PET	18,5	2,6
PE	19,4	54,3

PP	8,9	5,5
PVC	0,2	0,1
PS	0,4	0,7
Not identifiable	3,2	4
Misc plastic	0,1	0,3
Laminated packages	1,6	2,9
EPS blocks	0,1	0
Silicone tubes	0,1	0
Non-packaging plastics	3,0	7,7
Organics	5,2	0,2
Paper/cardboard	4,4	0,4
Metal	0,6	0
Moisture and dirt	34,5	21,3
Total	100	100

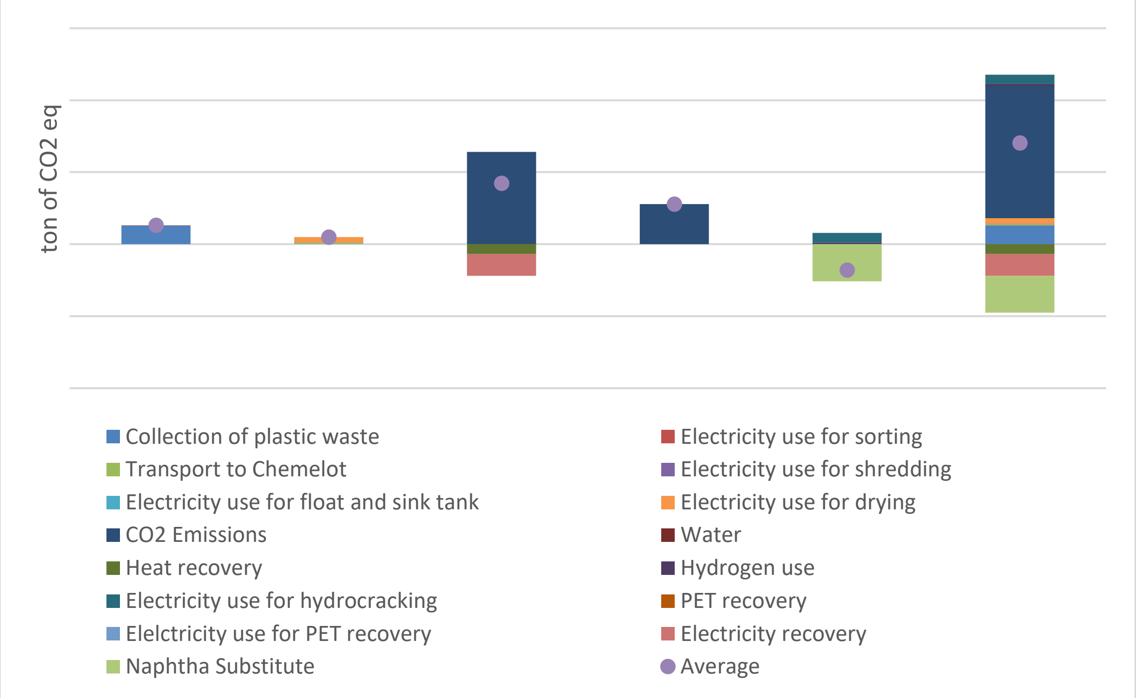
By taking the DKR 310 portion of PE and PP as a pyrolysis reactor input, the yield of product oil increases for a factor of 2,1. In the pre-treatment and in the hydro processing unit more energy for the drying and hydrogen, respectively, is required due to a larger amount of plastic input. The exact changes in numbers are shown in Table 13.

Table 13: Comparison in the exact changes in numbers between the waste streams of DKR 350 and DKR 310.

	DKR 350		**DKR 310**	
Item	**Amount**	**Unit**	**Amount**	**Unit**
Plastic available for pyrolysis (PE/PP)	283,00	kg	598,00	kg
Plastic waste for incineration	545,00	kg	296,00	kg
Energy use for thermal drying	32,00	kWh	67,00	kWh
Hydrogen consumption	2,31	kg	5,13	kg
Pyrolysis oil yield	245,31	kg	519,00	kg

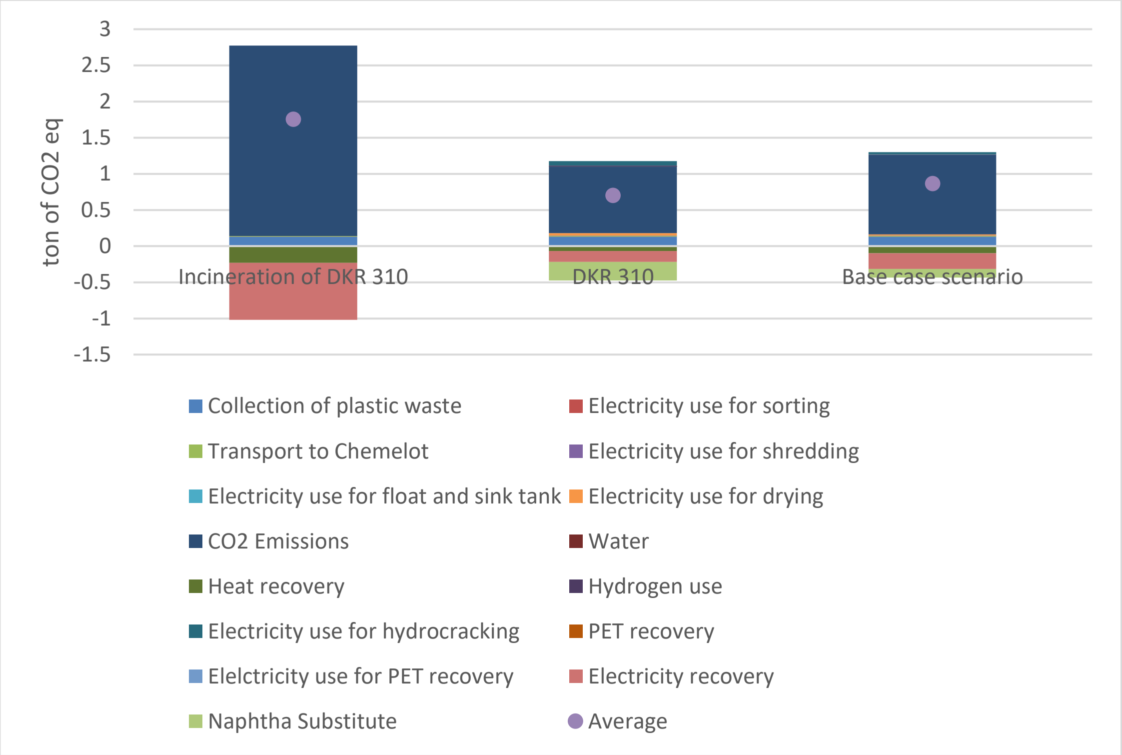
Figure 10 details the CO_2 contribution of a pyrolysis treatment of 1 ton of DKR 310 waste stream to the global warming potential. In this case it is evident that the overall process generates 0,702 ton of CO_2 eq. The incineration of the leftover plastic still largely contributes to the overall process emissions; however. only with 60 %. The second biggest emitter with the 0,278 ton of CO_2 eq is the pyrolysis unit itself without the sorting part as in the base case scenario. This is due to a higher amount of plastic going into the pyrolysis process. The emissions for the pre-treatment and pyrolysis unit are more than doubled, whereas the environmental benefits at the hydrocracking step increased by 135 kg of CO_2 eq.

Figure 10: Environmental profile of treatment of 1 ton of DKR 310.



In Figure 11, the comparison between the base case, DKR 310 scenario and the incineration of DKR 310 is shown. The net value of the CO_2 emissions contribution in the base case scenario and in the DKR 310 is set at 0,866 and 0,702 kg of CO_2 eq. It can be seen that the environmental benefits are larger in DKR 310 than in DKR 350.Due to the halved amount of plastic going into the incineration process, the CO_2 emissions and the energy recovery of the DKR 310 profile are lower, which results in an environmental benefit.

Figure 11: Comparison between the DKR 350 and DKR 310.



6.2.3 *Theoretical potential of pyrolysis*

In the base case scenario only 28,3 % of mix plastic waste stream is currently suitable to enter the pyrolysis reactor. What would be the case if 100% of waste stream were treated through pyrolysis? An interesting scenario to investigate would be the full potential of the pyrolysis process. In this case, the composition of DKR 350 is theoretically replaced with 100% of PE/PP eligible for chemical recycling. It is estimated that the same amount of energy goes into the collection and sorting of the new theoretical waste stream and that there is no plastic left to be put in the incineration.

Figure 12 depicts the environmental profile of the full potential scenario. The total CO_2 emissions result in 0,377 ton of CO_2 eq. The pyrolysis unit is the biggest emitter, comprising 57% of the overall emissions. The environmental benefits of naphtha substitute reduces these emissions by almost one half.

Figure 12: GWP profile of a full potential scenario.

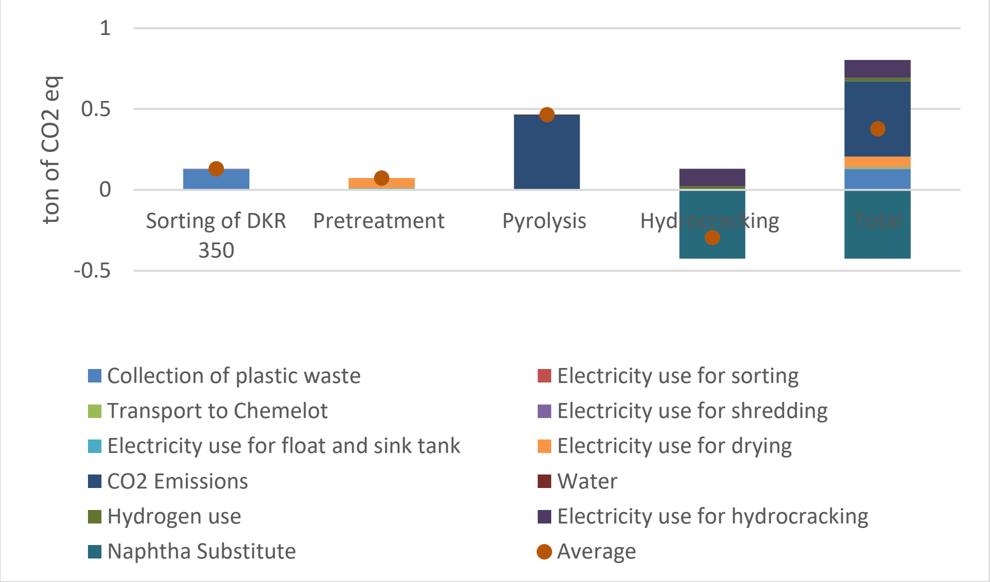
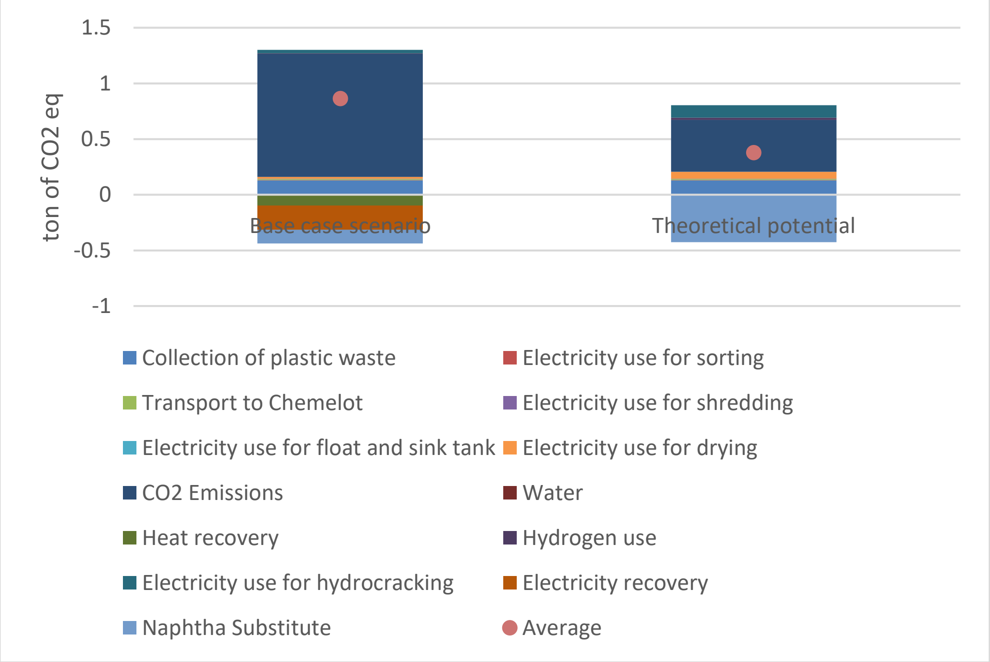


Figure 13 shows the comparison between the overall emissions of the full potential scenario and the basecase. The base case scenario emits 0,866 ton of CO_2 eq., whereas the full potential scenario results in only 0,377 tons of CO_2 eq. The reduction in emissions comprises of 0,487 ton of CO_2 eq. Due to the incineration exclusion, the impact in CO_2 emissions are much lower, but the environmental benefits of naphtha substitute are almost 3,5 times higher.

Figure 13: Comparison between the full potential and the base case scenario.



6.2.4 Conclusion and discussion

There have been several additional scenarios carried out to potentially optimize the CO_2 reduction by assuming a different feedstock (DKR 310 scenario), as well as different processing of the leftover waste (mechanical recovery of PET). Additionally, a separate scenario was conducted in order to assess the highest theoretical potential of pyrolysis, where 100% of feedstock is used in pyrolysis. All three additional scenarios show immense improvements in CO_2 avoidance compared to the base case scenario. Therefore, it is recommended to further look into the possibility of recovering PET and to consider other waste streams, such as DKR 310 as a feedstock.

6.3 Sensitivity analyses

6.3.1 DKR 350

Der Grüne Punktons company established a dual disposal system of waste and provides specifications on all recyclable fractions[25]. Table 14 states the requirements for mix plastic waste fraction.

Table 14 DKR 350 specifications.

	DKR 350 - MIXED PLASTIC
Specifications	Packaging (PE, PP, PS, PET)
Purity	90%<
Impurities	Paper (5%), metal (2%), PET(4%), PVC (0.5%), Other (3%)
Delivery	Transportable bales, dry stored

There is no detailed information on separate plastic fractions; however, it is evident that there is no moisture and dirt present in the fractions. To get closer to the composition in requirements, moisture and dirt content from the DKR 350 composition in the base case is removed. This is shown in Table 15.

Table 15 Changes between the composition of DKR 350 before and after the removal of moisture.

Composition	DKR 350 (%)	NEW DKR 350 (%)
PET	18,5	28,2
PE	19,4	29,5
PP	8,9	13,5
PVC	0,2	0,3
PS	0,4	0,6
Not identifiable	3,2	4,9
Misc plastic	0,1	0,2
Laminated packages	1,6	2,4
EPS blocks	0,1	0,2
Silicone tubes	0,1	0,2

Non-packaging plastics	3,0	4,6
Organics	5,2	7,9
Paper/cardboard	4,4	6,7
Metal	0,6	0,9
Moisture and dirt	34,5	-
Total	100	100

Figure 13 Total GWP of the new DKR 350 shows the total GWP of the new DKR 350 composition. The emissions amount collectively to 1090 kg od CO_2 eq. This is due to the higher carbon content in the waste stream. 43,1% of the waste stream goes into the pyrolysis unit, which consequently emits more CO_2. In addition, the leftover plastic without dirt has a higher carbon content.

Figure 13 Total GWP of the new DKR 350 composition.

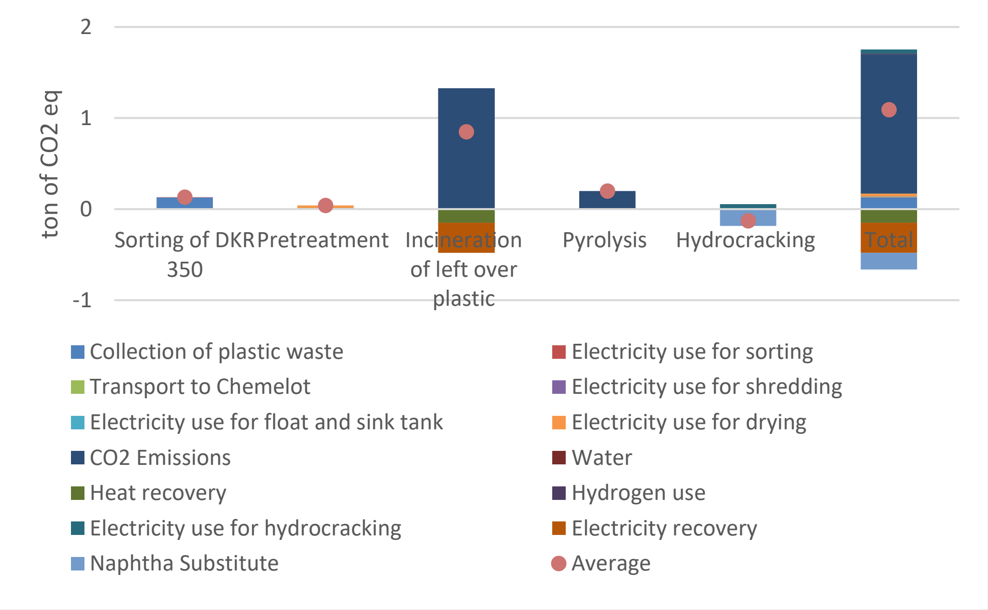


Figure 14 depicts the CO_2 emissions of the pyrolysis and incineration processes. It is seen that the incineration CO_2 contribution amounts to 1559 kg of CO_2 eq. and with that, pyrolysis archives a 30% emissions reduction compared to the 40% reduction with the base case DKR 350 composition.

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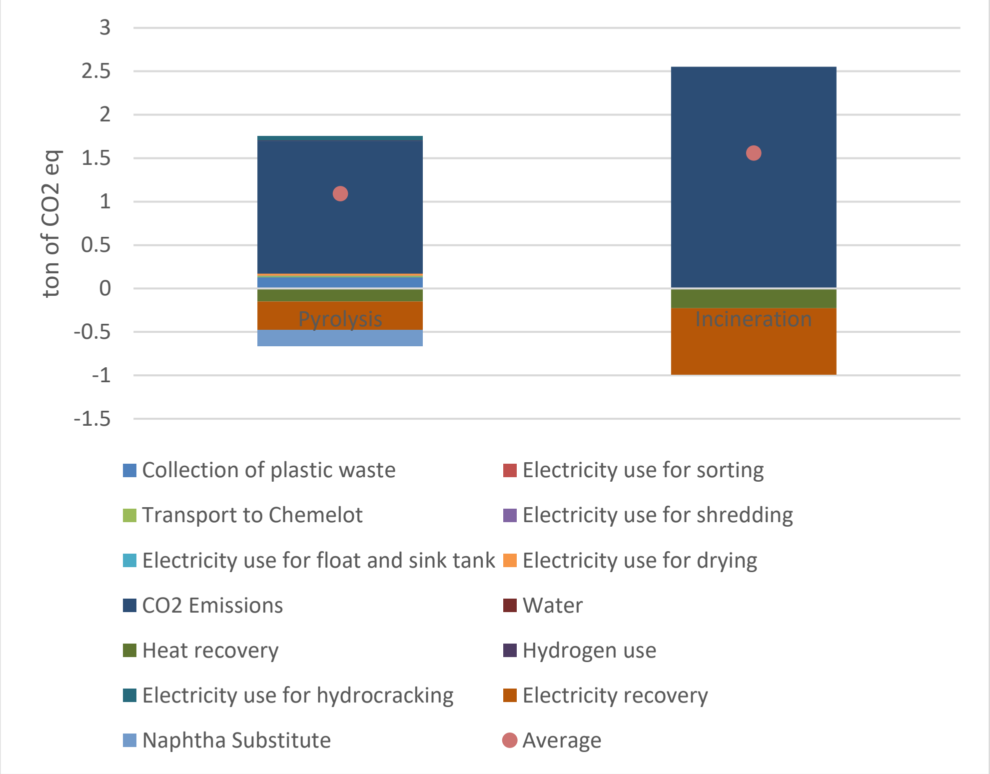
Figure 14 Comparison of GWP of pyrolysis and incineration of DKR 350.



6.3.2 *Post treatment alternatives*

In the base case scenario, the pyrolysis oil does not contain any impurities such as heteroatoms and aromatic compounds. Therefore, the oil is upgraded only via hydrocracking in order to reach the composition similar to that of naphtha. However, this scenario is very optimistic. During the process, there will occur some level of contamination, which will result in the product oil being oxygenated and unstable. In order to improve the quality of the oil, it needs to undergo the hydrotreatment step, where the use of hydrogen and catalysts reduces levels of sulphur, nitrogen and oxygen.

Due to the lack of data regarding the contamination levels and the hydrogen needed to remove the impurities, the hydrorefining technology and its hydrogen consumption are taken from the company called CLARITER. CLARITER is a company which produces oil and wax from plastic waste through pyrolysis and then hydrorefines it. The input of the process and the process itself are very similar to the one studied. Therefore, it is taken as a reference for this sensitivity analysis. The data on production rates and their catalyst and hydrogen consumption for the hydrorefining step are publicly available and therefore used in this study[39]. The description of the pyrolysis process is taken from their publication[39] as well.

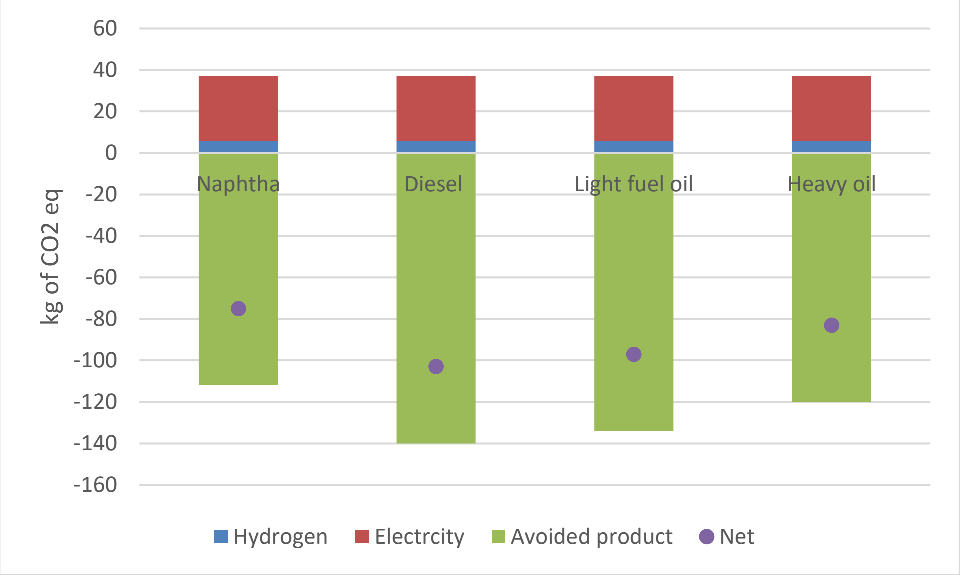
''Plastic feed, a combination of soft and hard plastics is before coming to the plant already separated cleaned, washed and dried therefore it is no longer a waste stream but mix of plastics. The conversion of plastics into hydrocarbons is based on relatively mild conditions thermal cracking producing a so called "Cracked Oil". Cracked Oil is a semi solid in ambient conditions mixture of various hydrocarbons that later on and hydro-refined. The Cracked Oil is catalytically hydrogenated (HDT) and hydro-desulfurized (HDS – basically hydro-treated i.e. refined in the presence of hydrogen) at high pressure/high temperature conditions in order to saturate all double bonds (olefins) and great majority of the aromatic bonds as well as to eliminate all heteroatoms like sulfur. nitrogen. oxygen and metals impurities to a single ppm level. This processes result in extremely clean products being used by cosmetic and food industry for example. Following this HDT/HDS step the obtained hydro-treated paraffin mass ("paramass") is separated in a two stage distillation process into light (solvents). middle (oil) and heavy (wax) fractions. The oil fraction is further dewaxed (HDX). at high pressure medium temp. in presence of H2. to improve its cold flow properties and viscosity. Dewaxing is followed by a hydro-finishing (HDF) step to eliminate any potential colour species produced during dewaxing.''[39]

From the description above it is evident that the oil is hydrotreated as well as hydrocracked in the dewaxing step of the process. From the publications it is not clear until what composition the final product is cracked. Hence, the uncertainty lies in the hydrogen consumption and the final product composition.

6.3.2.1 Sensitivity regarding the final product

The CLARITER company provides hydrotreatment and hydrocracking unit where the oil is treated to a certain composition. The base assumptions of the hydrogen and electricity consumption are the same as in the base case. However, the final composition of their product is unknown. In order to discover the deviation of the environmental impact of different final products, four different products were modelled in this analysis. In Figure 15, the environmental impacts when pyrolysis oil is hydro processed to naphtha, diesel, light oil and heavy oil are presented.

Figure 15 Comparison between the different final product options, modelled as the avoided product in the post treatment step.



As it is seen from figure 15, there are no large deviations in terms of the environmental impact of different products. Naphtha was modelled in the base case scenario as an avoided product. Diesel and light fuel oil represent the largest anomaly from naphtha with 28 and 20 kg of CO_2 eq., respectively. Heavy fuel oil deviates only with 8 kg of CO_2 eq. Therefore, it can be concluded that the final product composition does not play a significant role in the environmental impact assessment. And if the final product differentiates from the one in the base case scenario, it will only have a positive impact on the GWP. It should be emphasized that it plays a role as an input into the steam cracker. The final product is determined by the flexibility of the steam cracker.

6.3.2.2 Sensitivity analysis in terms of hydrogen consumption

Hydrogen consumption is another topic that is uncertain at this step. As mentioned before, the pyrolysis oil should also be hydrotreated and not only hydrocracked due to a highly possible contamination of the pyrolysis oil. Because the impurities content in the oil is unknown, and with that also the hydrogen consumption, the data is taken form the CLARITER company.

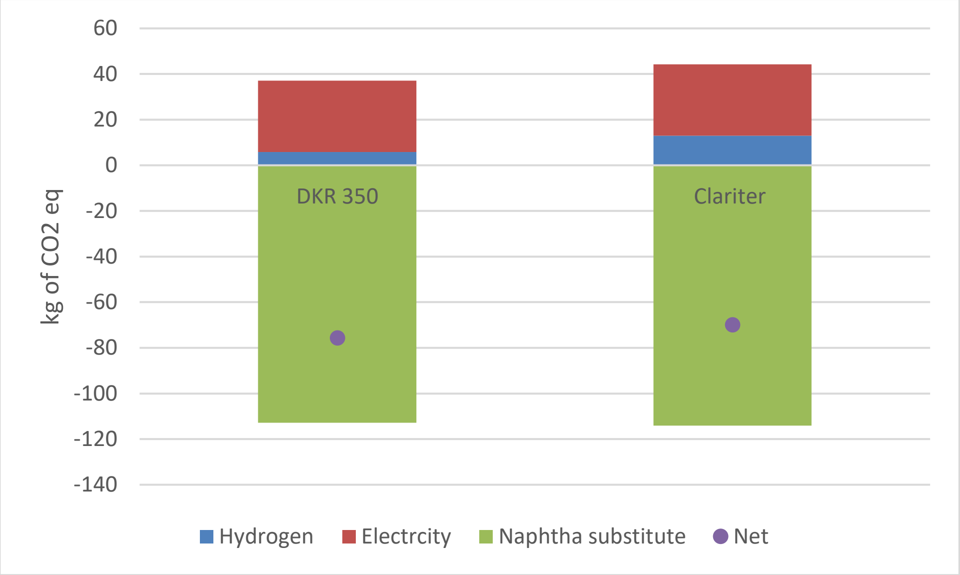
Base assumptions:

- Yearly hydrogen consumption of CLARITER amounts to 17,52 Mg/y of hydrogen per 960 Mg/y of PE and PP input. This translates to 5,16 kg/h of hydrogen per 283 kg of PE and PP input (or per functional unit – 1 ton of DKR 350).
- Electricity consumption was not provided by the company, therefore it was taken from the Vinescu et al (2017)[34], where different upgrading scenarios are

investigated. Electricity consumption for a hydrotreatment unit is 0,23 kWh/kg of produced oil.

Figure 16 shows the increase in GWP in the CLARITER hydrogen consumptions by 7,7 kg of CO_2.

Figure 16 Comparison of hydrogen consumption in the base case and CLARITER scenarios.



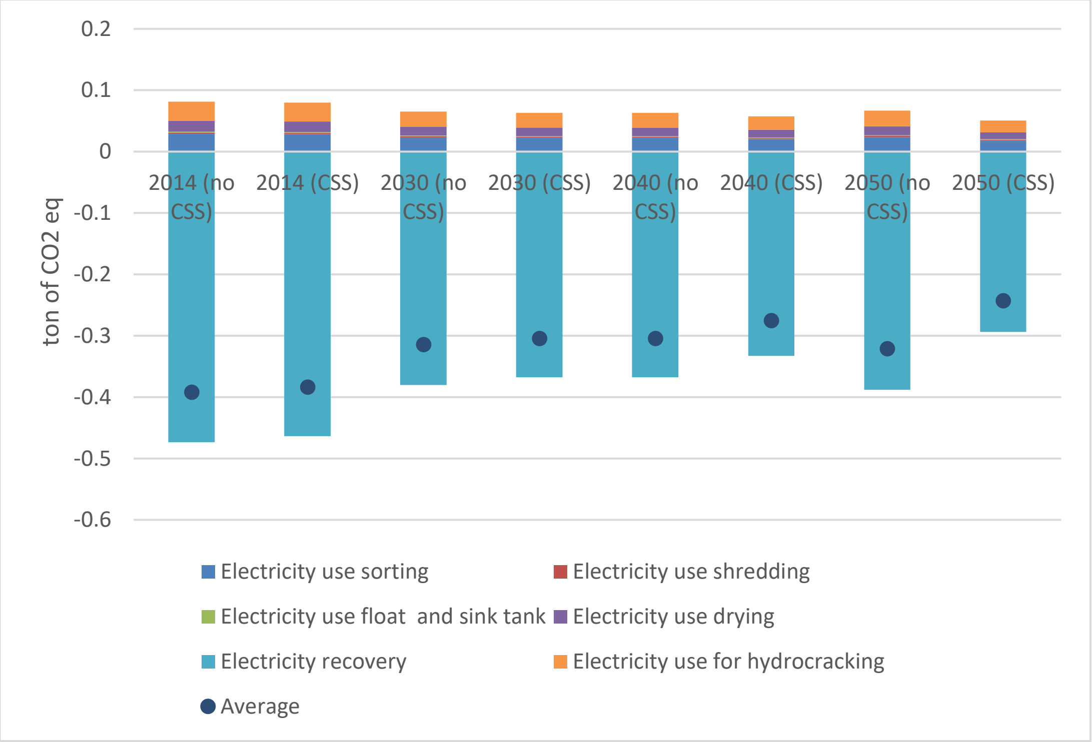
6.3.3 *Future electricity mix*

In order to predict the environmental impacts of the pyrolysis process in the future, the future possible energy mixes were included in the analysis. The future scenarios for the years 2030, 2040 and 2050 were taken from the integrated assessment model (IAM) E3ME-FTT-GENIE[38]. This model assumes that the climate targets are based on the maximum global warming of 2 ˚C scenario. The future scenarios were implemented in the Simapro software using the Dutch electricity production mix from the ecoinvent dataset. The model was recommended by an expert at TNO.

However, there are significant changes visible between the ecoinvent datasets and the IAM. The major one is the inclusion of carbon capture and storage (CCS) in the IAM. In order to convert all the categories in the IAM to the ecoinvent model, two scenarios were constructed. The first scenario assumes no CCS technology in the future. This means that electricity production of all technologies with or without CCS are summed together and this leads to an overestimation of the CO_2 emissions. The second scenario includes CCS by assuming that all the CO_2 emissions can be captured. In the process, this is calculated by subtracting all the CO2 emissions from the particulate technology including CCS. As the first scenario depicts the overestimation in CO2 emission, the second one tends to underestimate them and consequently their impact. Thus, it can be concluded that the middle ground between the both scenarios shows a realistic image of the future impacts.

Figure 17 depicts the comparison between the environmental impacts of various electricity mixes used in the whole pyrolysis process with or without CCS for the years 2014, 2030, 2040 and 2050. It is evident that throughout the years, the CO_2 emissions are reduced due to a larger share of renewable technologies in the energy mix. However, more renewables also cause a reduction in the environmental benefits since less and less CO_2 is saved compared to the current electricity mix. Comparing electricity mixes also with or without the CCS technology shows that CCS will be gaining more significance in the future decades. In 2014 the differences between the scenarios is negligible, whereas in 2050 there is a difference in emissions equal to 32 kg of CO_2 eq.

Figure 17 Future electricity mix.



6.3.4 *Conclusion and discussion*

Sensitivity analysis regarding the adjusted DKR 350 compositions, the future electricity mix, the post treatment options have been performed. The future electricity mix has demonstrated that the higher inclusion of renewables in the electricity mix could potentially cause lower savings in terms of CO_2. By including two scenarios: one with the CCS technology and one without it showcase the significance of CCS in the future, especially by 2050. Third analysis showed that a lot of uncertainties are present in the post treatment unit, where the hydrogen consumption and the final composition of a product vastly depend on each other. It was established that choosing between different products such as diesel, light oil, or heavy oil brings only positive environmental impact and therefore changes in the final composition do not have a negative impact on the results. However, the steam cracker might impact the products choice. On the other hand, an increased hydrogen consumption for the hydrotreatment unit results in higher emissions, which are, however, not significant. Thus, adding additional hydrogen to eliminate all the potential impurities does not significantly change the CO_2 contribution of the whole process.

6.4 Chemelot

As mentioned before, there are 100 ktons of mixed plastic waste stream available for chemical recycling in the Netherlands. This amount of feedstock could potentially produce 24.5 ktons of pyrolysis oil serving as naphtha substitute for the stream crackers. Annual naphtha consumptions in the two steam cracker at Chemelot amounts to 4000 ktons, which means that 24,5 % of pyrolysis oil could only replace 0,6% of the whole annual consumption[20]. An option would be to process additional feedstock such as DKR 310. This would add another 30,8 ktons of pyrolysis oil per year. Even combining both of the waste streams to produce naphtha substitute could only replace 1,4% of the annual naphtha usage at the site. It can be concluded that there is not enough plastic in the Netherlands to feed two steam crackers at Chemelot. Alternatives in the upper part of the waste management hierarchy have to be found in order to keep the plastic circular with the minimum downgrading.

As stated in the previous chapters, pyrolysis is also in need of a very clean stream of PP and PE in order to be efficient. The input of pyrolysis is very low and the majority of the left over material has to be incinerated. Therefore, it can be concluded that some other recycling technologies, such as gasification, currently perform better environmentally. By analysing different potential scenarios, it is highly recommended in the future to optimize the current waste treatment by recovering PET, and to consider other waste streams such as DKR 310 as a feedstock.

Due to the lack of plastic waste in the Netherlands and the current poor environmental performance of the pyrolysis process compared to other chemical recycling technologies, it can be summed up that pyrolysis is not the best alternative for the Chemelot site.

Conclusion and recommendations

This study investigated the climate change impacts of the pyrolysis treatment of 1 ton of mix plastic waste stream. The processes used in this study can be implemented right now and therefore reflect the current state of the environmental impacts. In the LCA, four major steps were included: production of DKR350, pre-treatment, pyrolysis and post-treatment. The pyrolysis oil is in the post-treatment unit hydrocracked to the oil with naphtha composition. This could be easily put into the steam cracker to produce ethylene and propylene. The results show that the total of processes of the pyrolysis life cycle generates 0,866 ton of CO_2 eq In the base case scenario, incineration of leftover plastic has the biggest contribution of the CO_2 emissions to the overall process emissions, amounting to 76% of the emissions. The pyrolysis unit and the production of DKR 350 follow with both 15% and 14%, respectively, whereas the pre-treatment only amounts to 3% of all CO_2 and hydrocracking has a negative 8% due to naphtha considered as an avoided product.

Pyrolysis oil annual production of DKR 350 collected in The Netherlands at the pyrolysis plant at Chemelot could potentially replace 0,5% of the yearly naphtha consumption of the steam crackers at Chemelot. This is far too low to achieve carbon neutrality at Chemelot. The plastic needs to be additionally imported from the neighbouring countries or other alternatives need to be found. Currently, pyrolysis has a lower environmental performance than gasification. However, this can be improved in the future by PET recovery or improving the feedstock composition.

The future research can be dedicated to expanding the boundaries to the steam cracker. This would give a fairer comparison to other recycling technologies, as well as enable us to look into the future of sorting, which could affect the waste stream composition and with that improve or reduce the options for chemical recycling. Since there is a clear lack of plastic in the Netherlands to satisfy the steam cracker current input, it is recommended to investigate the environmental aspects and efficiency of pyrolysis with biomass feedstock. Evaluating different practical aspects of the waste treatment on the site is also something that has not been investigated in depth in this research and could add value in the future.

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8 Appendix 1: Inventory table

Table 16 Inventory table for the production of DKR 350.

Input			**Output**		
Input			**Output**		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
Plastic packaging waste	4500	kg	*DKR350*	1000	kg
Municipal waste collection service	100	tkm			
Dutch electricity sorting	4	kWh			

Table 17 Inventory table for the pre-treatment unit.

Input			**Output**		
Input			**Output**		
Item	Amount	Unit	Item	Amount	Unit
DKR350	1000	kg	*Shredded PE/PP*	283	kg
Transport lorry (>32 metric ton). EURO6	100	tkm			
Dutch electricity mix for shredding	4	kWh			
Dutch electricity mix for float and sink tank	2.81	kWh			
Water consumption for float and sink	43.05	kg			
Dutch electricity mix for drying	31.69	kWh			

Table 18 Inventory table for the pyrolysis reactor.

Pyrolysis reactor- Mass balance					
Input			Output		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
Plastic waste	283,00	kg	*Ethane*	18,34	kg
Moisture	14,15	kg	*N-Octane*	52,52	kg
Air	1,33	kg	$n\text{-}C_{14}$	90,08	kg
Recycled pyrolysis gas	764,10	kg	$n\text{-}C_{18}$	44,52	kg
		kg	$n\text{-}C_{25}$	47,69	kg
			$n\text{-}C_{30}$	9,76	kg
			Water	13,84	kg
			N_2	1,05	kg
			CO_2	0,76	kg
			H_2	0,11	kg
			Char	19,81	kg
			Recycled pyrolysis gas	764,10	kg
Total	**1062,58**	**kg**		**1062,58**	**kg**
Total – recycled gas	**298,48**	**kg**		**298,48**	**kg**

Table 19 Inventory table for the fuel collection unit.

Fuel collection unit- Mass balance					
Input			Output		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
			Pyrolysis product oil		
Ethane	18,34	kg	*Ethane*	0,23	kg
N-Octane	52,52	kg	*N-Octane*	50,74	kg
n-C_{14}	90,08	kg	n-C_{14}	90,11	kg
n-C_{18}	44,52	kg	n-C_{18}	44,52	kg
n-C_{25}	47,69	kg	n-C_{25}	47,69	kg
n-C_{30}	9,76	kg	n-C_{30}	9,76	kg
Water	13,84	kg	*Non-condensable gases*		kg
N_2	1,05	kg	*Ethane*	18,11	kg
CO_2	0,76	kg	*N-Octane*	1,78	kg
H_2	0,11	kg	*Water*	13,84	kg
			N_2	1,05	**kg**
			CO_2	0,76	**kg**
			H_2	0,11	**kg**
Total	**278,67**	**kg**		**278,67**	**kg**

Table 20 Flue gas composition.

Flue gas composition			
Item	**Amount**	**Unit**	**wt%**
N2	635,05	kg	***73***
CO2	131,77	kg	***15***
O2	54,53	kg	***6***
H20	45,28	kg	***5***
Total	866,63	kg	***100***

Table 21 Mass balance for the combustion unit.

Combustion Unit- Mass balance					
Input			**Output**		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
			Flue gas composition		
Char	19,81	kg	N_2	635,05	kg
Moisture	9,62	kg	CO_2	131,77	kg
Air	815,04	kg	O_2	54,53	kg
Ethane	18,11	kg	H_20	45,28	kg
N-Octane	1,70	kg			
Water	0,42	kg			
N_2	1,05	kg			
CO_2	0,76	kg			
H_2	0,11	kg			
Total	**866,63**	**kg**		**866,63**	**kg**

Table 22 Mass balance of the overall process.

Overall process- Mass balance					
Input			**Output**		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
Plastic waste	283,00	kg	Pyrolysis fuel oil		
Moisture	23,77	kg	*Ethane*	0,23	kg
Air	816,37	kg	*N-Octane*	50,74	kg
			n-C_{14}	90,11	kg
			n-C_{18}	44,52	kg
			n-C_{25}	47,69	kg
			n-C_{30}	9,76	kg
			Water	13,49	kg
			Flue gas		kg
			N_2	635,05	kg
			CO_2	131,77	kg
			O_2	54,53	kg
			H20	58,58	kg
Total	**1123,14**			**1122,97**	

Table 23 Carbon balance of the overall process.

Overall process– Carbon balance		
	Input	**Output**
C	0,22	0,22
H	0,04	0,04
O	0,18	0,18
N	0,57	0,57
Sum	1,00	1,00

Table 24 Inventory table for the post-treatment.

Post-treatment					
Input			**Output**		
Item	**Amount**	**Unit**	**Item**	**Amount**	**Unit**
Pyrolysis Oil	243,04	kg	*Naphtha Substitute*	243,35	kg
Hydrogen consumption	2,31	kg			
Electricity use for hydroporcessing	56,14	kWh			

9 Appendix 2: Future energy mix

In order to predict the environmental impacts of the pyrolysis process in the future, the future possible energy mixes were included in the analysis. The future scenarios for the years 2030, 2040 and 2050 were taken from the integrated assessment model (IAM) E3ME-FTT-GENIE[38]. This model assumes that the climate targets are based on the maximum global warming of 2˚C scenario. The future scenarios were implemented in the Simapro software using the Dutch electricity production mix from the ecoinvent dataset.

However, there are significant changes visible between the ecoinvent datasets and the IAM. The major one is the inclusion of carbon capture and storage (CCS) in the IAM. In order to convert all the categories in the IAM to the ecoinvent model, two scenarios were constructed. The first scenario assumes no CCS technology in the future. This means that electricity production of all technologies with or without CCS are summed together and this leads to an overestimation of the CO_2 emissions. The second scenario includes CCS by assuming that all the CO_2 emissions can be captured. In the process, this is calculated by subtracting all the CO2 emissions from the particulate technology including CCS. As the first scenario depicts the overestimation in CO2 emission, the second one tends to underestimate them and consequently their impact. Thus, it can be concluded that the middle ground between the both scenarios shows a realistic image of the future impacts.

Since the ecoinvent and IAM datasets are so different, the electricity mix of 2014 in IAM was also not the same to the one in ecoinvent dataset. Therefore, an electricity mix of IAM was also used in the base case scenario. There are numerous other assumptions made in this scenario analysis.

- Firstly, the technologies fuel cells and the combined heat and power were omitted from the IAM, as these technologies were not in the ecoinvent dataset. Another reason behind the exclusion is their low contribution in the future. In 2014, their input comprised only 0,08% and 0,001%, respectively. That number is estimated to become even lower in the upcoming decades.
- Secondly, the ecoinvent dataset provides more detailed technologies than IAM. This gap was breached by assigning a certain value to the more specific technology than the main one in the IAM. These values were split up according to the ratios between the specific technologies in ecoinvent Dutch production mix. It was assumed that these ratios would remain equal in the future scenarios. For

example, the value for the combined cycle gas turbine (CCGT) was split up into electricity production and heat & power co-generation using natural gas (for both the combined cycle power plant and the conventional power plant). The onshore wind electricity production was split up according to the power of the turbine, i.e. <1MW. 1-3MW and >3MW. Lastly, the solar PV for the production of low voltage was split up into multi-Si and single-Si panels. The integrated gasification combined cycle (IGCC) was assumed to be heat and power co-generation of hard coal in SimaPro. Finally, the solid biomass and biomass-based integrated gasification combined cycle (BIGCC) was assumed to be the heat and power co-generation using wood chips.

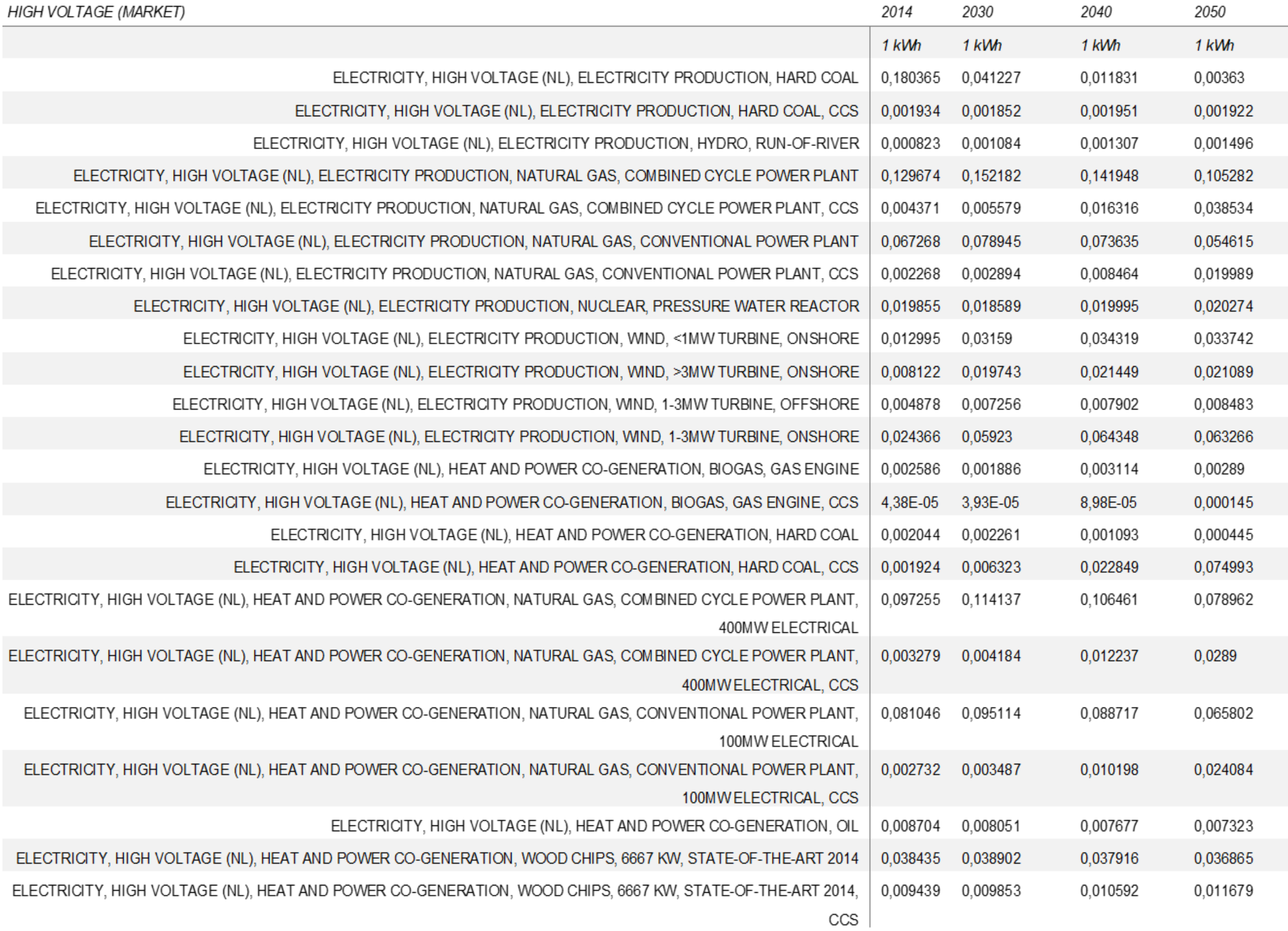
- The solar PV was also not included in the future electricity mix. Solar electricity is usually connected to the low voltage grid and the electricity mix used in the base case scenario was modelled for the medium voltage. The only exception is incineration, which was modelled for the high voltage production mix.
- The import of electricity from Belgium, Germany, Great Britain and Norway was included because it was already incorporated in the ecoinvent database. It was assumed that the amount of imported electricity of each country remained equal in the future scenarios of the years 2030, 2040 and 2050.

The following TablesTable 25Table 26 represent the contribution of different technologies to the electricity generation in the Netherlands for 2014, 2030, 2040 and 2050 including and excluding the CCS, divided into ecoinvent categories.

Table 25 Contribution of different technologies to the electricity generation in the Netherlands for 2014, 2030, 2040 and 2050 excluding CCS, divided into ecoinvent categories.

HIGH VOLTAGE (MARKET)	**2014**	**2030**	**2040**	**2050**
	1 kWh	*1 kWh*	*1 kWh*	*1 kWh*
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, HARD COAL	0,1823	0,043079	0,013782	0,005552
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, HYDRO, RUN-OF-RIVER	0,000823	0,001084	0,001307	0,001496
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NATURAL GAS, COMBINED CYCLE POWER PLANT	0,134045	0,157761	0,158264	0,143816
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NATURAL GAS, CONVENTIONAL POWER PLANT	0,069536	0,081839	0,082099	0,074605
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NUCLEAR, PRESSURE WATER REACTOR	0,019855	0,018589	0,019995	0,020274
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, <1MW TURBINE, ONSHORE	0,012995	0,03159	0,034319	0,033742
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, >3MW TURBINE, ONSHORE	0,008122	0,019743	0,021449	0,021089
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, 1-3MW TURBINE, OFFSHORE	0,004878	0,007256	0,007902	0,008483
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, 1-3MW TURBINE, ONSHORE	0,024366	0,05923	0,064348	0,063266
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, BIOGAS, GAS ENGINE	0,00263	0,001925	0,003204	0,003034
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, HARD COAL	0,003968	0,008584	0,023942	0,075438
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, NATURAL GAS, COMBINED CYCLE POWER PLANT, 400MW ELECTRICAL	0,100534	0,118321	0,118698	0,107862
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, NATURAL GAS, CONVENTIONAL POWER PLANT, 100MW ELECTRICAL	0,083778	0,098601	0,098915	0,089885
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, OIL	0,008704	0,008051	0,007677	0,007323
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, WOOD CHIPS, 6667 KW, STATE-OF-THE-ART 2014	0,047874	0,048754	0,048508	0,048544

Table 26 Contribution of different technologies to the electricity generation in the Netherlands for 2014, 2030, 2040 and 2050 including CCS, divided into ecoinvent categories.



HIGH VOLTAGE (MARKET)	*2014*	*2030*	*2040*	*2050*
	1 kWh	*1 kWh*	*1 kWh*	*1 kWh*
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, HARD COAL	0,180365	0,041227	0,011831	0,00363
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, HARD COAL, CCS	0,001934	0,001852	0,001951	0,001922
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, HYDRO, RUN-OF-RIVER	0,000823	0,001084	0,001307	0,001496
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NATURAL GAS, COMBINED CYCLE POWER PLANT	0,129674	0,152182	0,141948	0,105282
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NATURAL GAS, COMBINED CYCLE POWER PLANT, CCS	0,004371	0,005579	0,016316	0,038534
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NATURAL GAS, CONVENTIONAL POWER PLANT	0,067268	0,078945	0,073635	0,054615
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NATURAL GAS, CONVENTIONAL POWER PLANT, CCS	0,002268	0,002894	0,008464	0,019989
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, NUCLEAR, PRESSURE WATER REACTOR	0,019855	0,018589	0,019995	0,020274
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, <1MW TURBINE, ONSHORE	0,012995	0,03159	0,034319	0,033742
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, >3MW TURBINE, ONSHORE	0,008122	0,019743	0,021449	0,021089
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, 1-3MW TURBINE, OFFSHORE	0,004878	0,007256	0,007902	0,008483
ELECTRICITY, HIGH VOLTAGE (NL), ELECTRICITY PRODUCTION, WIND, 1-3MW TURBINE, ONSHORE	0,024366	0,05923	0,064348	0,063266
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, BIOGAS, GAS ENGINE	0,002586	0,001886	0,003114	0,00289
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, BIOGAS, GAS ENGINE, CCS	4,38E-05	3,93E-05	8,98E-05	0,000145
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, HARD COAL	0,002044	0,002261	0,001093	0,000445
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, HARD COAL, CCS	0,001924	0,006323	0,022849	0,074993
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, NATURAL GAS, COMBINED CYCLE POWER PLANT, 400MW ELECTRICAL	0,097255	0,114137	0,106461	0,078962
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, NATURAL GAS, COMBINED CYCLE POWER PLANT, 400MW ELECTRICAL, CCS	0,003279	0,004184	0,012237	0,0289
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, NATURAL GAS, CONVENTIONAL POWER PLANT, 100MW ELECTRICAL	0,081046	0,095114	0,088717	0,065802
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, NATURAL GAS, CONVENTIONAL POWER PLANT, 100MW ELECTRICAL, CCS	0,002732	0,003487	0,010198	0,024084
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, OIL	0,008704	0,008051	0,007677	0,007323
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, WOOD CHIPS, 6667 KW, STATE-OF-THE-ART 2014	0,038435	0,038902	0,037916	0,036865
ELECTRICITY, HIGH VOLTAGE (NL), HEAT AND POWER CO-GENERATION, WOOD CHIPS, 6667 KW, STATE-OF-THE-ART 2014, CCS	0,009439	0,009853	0,010592	0,011679