

# **Life-cycle Analysis of Conversion of Post-Use Plastic via Pyrolysis with the GREET Model**

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**Energy Systems and Infrastructure Analysis Division**

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# Life cycle Analysis of Conversion of Post-Use Plastic via Pyrolysis with the GREET Model

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prepared by

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## ABBREVIATIONS AND ACRONYMS

ACC	American Chemistry Council
Argonne	Argonne National Laboratory
C	carbon
CA	California
CAP	criteria air pollutant
CH <sub>4</sub>	methane
CO	carbon monoxide
CO <sub>2</sub>	carbon dioxide
$E_{gp}$	average energy use per tonne of product $p$ in group $g$
$E_i$	energy input in plant $i$
EPA	U.S. Environmental Protection Agency
E.U.	European Union
FRCC	Florida Reliability Coordinating Council
$g$	group
gal	gallon(s)
GHG	greenhouse gas
REET	Greenhouse gases, Regulated Emissions, and Energy use in Technologies
GWP	global warming potential
H	hydrogen
HDPE	high-density polyethene
HTL	hydrothermal liquefaction
IPCC	Intergovernmental Panel on Climate Change

ISO	International Organization for Standardization
kg	kilogram(s)
LCA	life-cycle analysis
LCFS	Low Carbon Fuel Standard
LDPE	low-density polyethylene
LHV	lower heating value
LLDPE	linear low-density polyethylene
$m_i$	mass of feedstock used in plant $i$
MJ	megajoule(s)
MRF	material recovery facility
MRO	Midwest Reliability Organization
MSW	municipal solid waste
$n$	total number of plants in group $g$
NDA	nondisclosure agreement
NO <sub>x</sub>	nitrogen oxides
N <sub>2</sub> O	nitrous dioxide
O	oxygen
$p$	product
PCP	post-consumer plastic
PET	polyethylene terephthalate
PIP	post-industrial plastic
PM <sub>2.5</sub>	particulate matter with a diameter $\leq 2.5 \mu\text{m}$

PM <sub>10</sub>	particulate matter with a diameter $\leq 10 \mu\text{m}$
PP	polypropylene
$P_{pi}$	mass of product $p$ generated in plant $i$
PS	polystyrene
PTF	plastic to fuel
PUP	post-use plastic
PVC	polyvinyl chloride
RFS	Renewable Fuels Standard
S	sulfur
SO <sub>x</sub>	sulfur oxide
ton	short ton
tonne	metric ton
ULS	ultra-low sulfur
U.S.	United States
USD	U.S. dollars
VOC	volatile organic compound

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## EXECUTIVE SUMMARY

Recent initiatives and policies have focused on creating a circular economy by developing and promoting pathways to convert plastics into valuable products, such as molecules suitable for remaking new, high-quality plastics and fuels with low sulfur content. Therefore, in recent years, processes and technologies for converting post-use plastic (PUP) into chemicals and fuels have been gaining traction.

Building on a previous collaboration between the American Chemistry Council (ACC) and Argonne National Laboratory (Argonne), in this report we present an updated version of the life-cycle analysis (LCA) of PUP conversion via pyrolysis to produce either intermediate products such as pyrolysis oil or fuels such as ultra-low sulfur (ULS) diesel. We used the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET®) model developed at Argonne to calculate the greenhouse gas (GHG) emissions, fossil energy, and water consumption of PUP intermediate product pyrolysis oil and final product ULS diesel. We then compared the results to those metrics for their conventional counterparts such as petroleum naphtha and petroleum ULS diesel fuel, respectively. To collect the data, we conducted a survey of eight pyrolysis companies, which we classified based on their annual capacities, and allocated the results accordingly. Three groups were established for the analysis: (1) pioneer, (2)  $N^{\text{th}}$ -plant, and (3) highest capacity. Pioneer plants were grouped in the lowest capacity range: less than 50,000 tonnes of PUP per year. Plants with higher capacity than 50,000 tonnes are referred to as  $N^{\text{th}}$ -plants. We examined one company with a processing capacity of 291,800 tonnes of PUP per year, which we refer to as the highest capacity. **Based on the survey, we observed that pyrolysis companies focus their production on intermediate chemicals, like pyrolysis oil, rather than fuels. This shift in production practices is in line with the establishment of a circular economy for plastics.**

As shown in Table ES-1 and ES-2, the results varied significantly depending on the coproduct treatment method (displacement, energy, and market allocation) and the plant capacity group.  $N^{\text{th}}$ -plants and the highest capacity plant exhibited lower GHG emissions, fossil energy use, and water consumption compared to their petroleum-based counterparts, crude naphtha and petroleum ULS diesel. For instance, using energy allocation method as the default case to allocate the emissions and energy burdens,  $N^{\text{th}}$ -plants showed 29%, 73%, and 83% lower GHG emissions, fossil energy use, and water consumption per megajoule of pyrolysis oil, respectively, compared to crude naphtha. Similarly, PUP-derived ULS diesel presented a reduction of 3%, 96%, and 80% in the GHG emissions, fossil energy use, and water consumption, respectively, compared to petroleum-based ULS diesel. In contrast, pyrolysis oil and ULS diesel produced from pioneer plants showed higher GHG emissions and water consumption compared to their fossil-based counterparts.

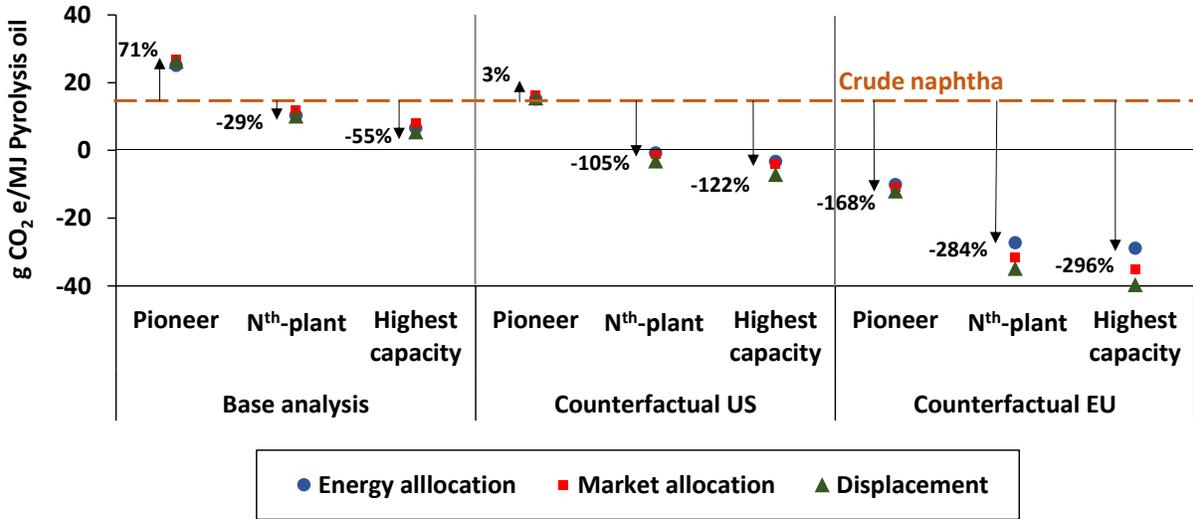
**Table ES-1. Change in Impact (%) from Producing 1 MJ of Pyrolysis Oil via Pyrolysis vs. Producing 1 MJ of Crude Naphtha (Base Analysis)**

Impact evaluated	Allocation method	Plant capacity group		
		Pioneer	N <sup>th</sup> -plant	Highest capacity
GHG emissions	Displacement	79%	-31%	-63%
	Energy-based allocation	71%	-29%	-55%
	Market-based allocation	83%	-19%	-45%
Fossil energy consumption	Displacement	-66%	-70%	-77%
	Energy-based allocation	-68%	-73%	-80%
	Market-based allocation	-66%	-69%	-75%
Water consumption	Displacement	13%	-83%	-84%
	Energy-based allocation	6%	-83%	-84%
	Market-based allocation	13%	-81%	-80%

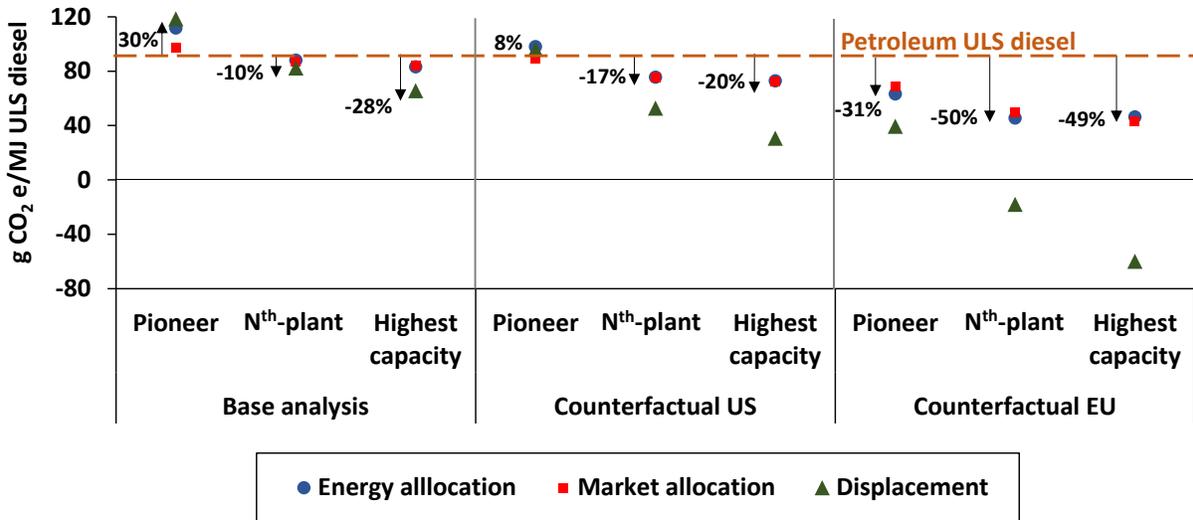
**Table ES-2. Change in Impact (%) from Producing 1 MJ of ULS Diesel via Pyrolysis vs. Producing 1 MJ of Petroleum ULS Diesel (Base Analysis)**

Impact evaluated	Allocation method	Plant capacity group		
		Pioneer	N <sup>th</sup> -plant	Highest capacity
GHG emissions	Displacement	30%	-10%	-28%
	Energy-based allocation	23%	-3%	-9%
	Market-based allocation	7%	-5%	-8%
Fossil energy consumption	Displacement	-185%	-249%	-339%
	Energy-based allocation	-81%	-96%	-98%
	Market-based allocation	-89%	-97%	-98%
Water consumption	Displacement	51%	-173%	-218%
	Energy-based allocation	54%	-80%	-83%
	Market-based allocation	-8%	-82%	-81%

The analysis also included a counterfactual scenario that subtracted the credits from avoiding traditional waste management in two geographical regions: United States and the European Union (E.U.). In the United States, the majority of PUP is disposed in landfills while in the E.U. it is incinerated with energy recovery. The results are presented in Figure ES-1 and Figure ES-2 and indicate that the GHG emissions per megajoule of pyrolysis oil with energy allocation were 105% lower than those of crude naphtha when the avoided emissions of traditional waste management in the United States are subtracted. The comparison of the U.S. counterfactual scenario also showed that emissions from ULS diesel produced in N<sup>th</sup>-plants are 17% lower compared to petroleum ULS diesel using energy allocation. Pyrolysis oil and ULS diesel obtained from pioneer plants showed similar GHG emissions compared to those of fossil counterparts, even after including avoided emissions from the traditional waste management in the United States. Greater GHG emission reductions were obtained when the results were compared with the E.U. counterfactual scenarios, because a higher percentage of PUP is sent for incineration in the E.U. **The results of the analysis presented here indicate that economies of scale for pyrolysis conversion reduce the environmental impacts and use of resources.**



**Figure ES-1. GHG Emissions to Produce 1 MJ of Pyrolysis Oil by Converting PUP via Pyrolysis.** Percentages shown in the Figure refer to the energy allocation method.



**Figure ES-2. GHG Emissions to Produce 1 MJ of ULS Diesel by Converting PUP via Pyrolysis.** Percentages shown in the Figure refer to the energy allocation method.

## 1 INTRODUCTION

The unique properties of plastics mean they have innumerable applications. They can be designed to be durable, so they can resist rough and harsh conditions; or they can be lightweight, which produces benefits in logistics and transportation. They are easy to make, because they can be molded according to the need, shape, and size of the desired product. Items made of plastic can reduce product costs, which means they are often available at reasonable prices. However, these characteristics have also driven the significant increase of plastic consumption worldwide, and plastics' end-of-use fate is creating considerable disposal problems.

Global plastics manufacturing has grown from 1.50 million tonnes per year in 1950 to more than 322 million tonnes per year in 2015 (Plastics Europe, 2017); this correlates with an increase in plastic waste disposal that affects human life, wildlife, and habitats (Law et al., 2010; Jambeck et al., 2015; Lebreton et al., 2018). The growth of the plastics industry is expected to continue into the near future, with an estimated global production of about 1,633 million tonnes by 2050 (Ryan, 2015). According to an American Chemistry Council report (ACC, 2020) the production of North American plastic resin grew 1.7%, to 55 million tonnes in 2019, up from 54 million tonnes in 2018.

In addition, the U.S Environmental Protection Agency (EPA) estimated that in the United States the total amount of plastic found in municipal solid waste (MSW) in 1960 was around 390,000 tonnes. This number has increased significantly, to 35 million tonnes in 2017 (EPA, 2019). Of the waste generated in 2017, only 8.4% of plastic in MSW was recycled; 16% was combusted, and the remaining 76% was landfilled. The accumulation of plastic waste in landfills and oceans has motivated researchers to develop new plastics and technologies to enhance the use phase and end-of-life treatment of plastic to improve sustainability.

Plastic that has served its designed purpose or that is generated as scrap is known as post-use plastic (PUP). When PUP is diverted from conventional waste-management treatment, such as a landfill, it can be transformed into a valuable product because it has a high energy content (46.4–49 MJ/kg, depending on the resin; Kunwar et al., 2016) and high carbon content (84–93% carbon, depending on the resin; Kunwar et al., 2016). This means PUP can be used to make new plastics, make precursors for chemicals, or produce energy.

High-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP) are common plastics used in industry. These polymers contain only carbon and hydrogen, which can be beneficial for pyrolysis. The products (chemical intermediates and fuels) generated from PUP can have high heating values, and they can be non-acidic and non-corrosive because they do not contain water or oxygen (Kunwar et al., 2016). Therefore, bringing plastic waste back into the supply chain can help drive circular economy. In addition, pyrolysis companies are changing their target products to intermediate chemicals, like pyrolysis oil, to promote circular economy strategies for plastics.

As PUP turns into a new source of feedstock, emerging conversion technologies are becoming popular and relevant to tackle the plastic waste problem. Conversion technologies like pyrolysis, gasification, hydrocracking, catalytic conversion, hydrothermal liquefaction (HTL), and

enzymatic depolymerization are providing alternatives to these fossil-derived products. They can potentially lower the environmental impact of plastics and are encouraging companies around the world to produce products derived from PUP. In this report, we analyze the process of converting PUP into an intermediate product or transportation fuels via pyrolysis. In pyrolysis the macromolecule structures of polymers are broken down into smaller molecules and monomer units under high temperatures and in the absence of oxygen (Khot and Basavarajappa, 2017). In gasification, high temperatures and oxidizing agents decompose the plastic into syngas, which is used to produce chemicals (Kunwar et al., 2016). In hydrocracking, polymers are cracked into fuel-range hydrocarbons using hydrogen and high temperatures. Catalytic conversion involves adding a special catalyst to the pyrolysis reaction to improve conversion, fuel quality, and selectivity. Catalysts are also added to improve operating conditions; they can, for example, reduce the temperature and the residence time (Kunwar et al., 2016). Finally, HTL uses supercritical water to liquefy PUP into oil that includes olefins, kinds of paraffin, and aromatics, which can be then used to produce gasoline-like blendstocks or other chemicals such as naphtha (Chen et al., 2019).

Life-cycle analysis (LCA) can be used to evaluate the environmental impacts of these different conversion technologies. LCA takes a holistic approach to evaluate each stage of the process: converting plastic waste into intermediate feedstocks or fuels, identifying the key drivers that influence GHG emissions and other sustainability metrics, and pursuing opportunities to mitigate adverse environmental impacts.

In this report, we present an updated LCA of the plastic-to-fuel (PTF) pathway developed at Argonne National Laboratory (Argonne). In previous research, Benavides et al. (2017) analyzed the environmental impact of producing ultra-low sulfur (ULS) diesel fuel from PUP via pyrolysis (the most common PTF technology). In this study, we used the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET<sup>®</sup>) model and collected data from eight pyrolysis companies to assess the potential energy and environmental benefits associated with this technology (Argonne, 2020). We conducted a survey, and from the different responses we aggregated a dataset that includes plant capacity, process yields, feedstock composition, material and energy inputs, and outputs, and other parameters. We calculated the energy, greenhouse gas (GHG) emissions, and water consumption of intermediate product, pyrolysis oil, and final product, PUP-derived ULS diesel, and then we compared the results to those metrics for their conventional counterparts (crude naphtha and petroleum ULS diesel fuel).

For this new analysis, we followed the same methods previously used by Benavides et al. (2017): we collected industry data from eight companies (some of which participated in the first study), analyzed the survey results, and aggregated the data in three different categories based on plant capacity—pioneer plant, *N*<sup>th</sup>-plant, and highest-capacity plant. This study focused on the United States, because the majority of the pyrolysis companies surveyed are located in the United States. The metrics evaluated included the energy consumption, water consumption, and GHG emissions per 1 MJ of pyrolysis oil and per 1 MJ of PUP-derived ULS diesel. We compared these data to comparable data for conventional crude naphtha and petroleum ULS diesel fuel, respectively.

This report is organized as follows: Section 2 presents the data and methodology used to collect and process the information. Section 3 describes the different environmental metrics used during the LCA of converting plastic waste into pyrolysis oil and ULS diesel fuel. This section also focuses on the key drivers that affect GHG emissions, fossil energy consumption, and water consumption. We also present sensitivity scenarios and the effect of varying key parameters on the GHG emissions. Finally, conclusions are presented in Section 4.

## 2 DATA AND METHODOLOGY

### 2.1 DATA COLLECTION AND SURVEY DEVELOPMENT

This section discusses data collection and key process parameters that were used to conduct the LCA of PUP-derived pyrolysis oil and PUP-derived ULS diesel fuel. To collect the data, we adapted the survey that was developed for the previous study (Benavides et al., 2017) to include detailed questions related to the type of PUP feedstock used, its pretreatment, conversion to oil, and upgrading or separation step (if available). Eight companies involved in pyrolysis technology provided information for the survey. During collection, the study team exchanged emails and phone calls with each company to discuss their participation and to explain the data input required. The team reviewed the survey responses and communicated with the companies to obtain clarification and explanations of missing data. All information was provided under a nondisclosure agreement (NDA). In combination with the survey results, we also used publicly available data for verification.

The survey was divided into four main sections, as shown in Table 1. Section 1 of the survey, “About the Company,” collected general information about each company, such as design capacity, technology stage, years of operation, and location. Survey section 2, “Feedstock,” asked questions related to the type, source, quality, quantity, and properties of feedstock used. It also discussed whether or not the company performs *any* pretreatment and the amount of energy (or material) used before the feedstock goes to conversion. Survey section 3, “Conversion and Separation,” discussed the conversion of the feedstock to oil (i.e., PUP-derived pyrolysis oil) or final product (i.e., PUP-derived ULS diesel-range fuel). This section was divided into two subsections based on the product obtained by the companies. For example, if the company produces an intermediate oil or pyrolysis oil that is upgraded offsite, the company selected option 1; however, if the conversion of plastic and intermediate oil upgrading process both occur onsite, then the company selected option 2. The last part of the survey was section 4, “Emissions.” This section gathered information about emissions associated with non-combustion activities and criteria air pollutants (CAPs). Additional details about the survey appear in Section A.1 of the appendix.

**Table 1. Key Parameters of the Survey**

Section of Survey	Sections
1. About the Company	<ul style="list-style-type: none"><li>• Capacity</li><li>• Stage of technology</li><li>• General</li></ul>
2. Feedstock	<ul style="list-style-type: none"><li>• Logistics</li><li>• Market</li><li>• Feedstock type</li><li>• Quality</li><li>• Properties and composition of feedstock</li><li>• Consumption</li><li>• Pretreatment</li></ul>

**Table 1. (Cont.)**

Section of Survey	Sections
3. Conversion and Separation	<b>Option 1</b> <ul style="list-style-type: none"> <li>• Properties and composition at the reactor throat</li> <li>• Conversion from plastic to intermediate oil</li> <li>• Conversion inputs/outputs</li> <li>• Intermedia oil production rates</li> <li>• Residues and coproducts production rates</li> <li>• Properties of products</li> <li>• Transportation distances and methods</li> </ul>
	<b>Option 2</b> <ul style="list-style-type: none"> <li>• Properties and composition at the reactor throat</li> <li>• Conversion from plastic to intermediate oil</li> <li>• Conversion inputs/outputs</li> <li>• Separation/Upgrading</li> <li>• Residues and coproducts after separation/upgrading</li> <li>• Final output after separation/upgrading</li> <li>• Properties of products</li> <li>• Transportation distances and methods</li> </ul>
4. Emissions	<ul style="list-style-type: none"> <li>• Non-combustion emissions</li> <li>• Other emission considered hazardous</li> </ul>

## 2.2 DATA PROCESSING

After collecting the data, we classified the companies into three groups based on their plant capacity: (1) pioneer plant, (2)  $N^{\text{th}}$ -plant, and (3) highest capacity plant. Details about this classification system are described in Section 3.1. The results were aggregated for each group. For instance, weighed average values were used to represent values of the composition, source, and type of feedstock, while other process parameters such as transportation distance for feedstock and products, process yields, non-combustion emissions, and product properties were aggregated using arithmetic mean calculations. For qualitative parameters, such as transportation type or fate of coproduct, we reported the more predominant answer as the representative for each plant capacity group. There were two main products for the surveyed companies: pyrolysis oil, which can be used as a chemical intermediate, and ULS diesel fuel. The material and energy inputs considered the standardization of the data into a per product basis using equation (1):

$$E_{gp} = \frac{\left[\left(\frac{1}{n}\right) \cdot \sum \frac{E_i}{m_i}\right]}{\left[\left(\frac{1}{n}\right) \cdot \sum \frac{P_{pi}}{m_i}\right]} \quad (1)$$

where  $E_{gp}$  is the average energy use per tonne of product  $p$  in group  $g$ ,  $E_i$  is the energy input in plant  $i$ ,  $m_i$  is the mass of feedstock used in plant  $i$ ,  $P_{pi}$  is the mass of product  $p$  generated in plant  $i$ , and  $n$  is the total number of plants in group  $g$ . After estimating representative values for each

plant capacity group, we used these values as inputs to perform the LCA. The considerations and assumptions of the LCA are described in the following sections.

### 2.3 SYSTEM BOUNDARY

Figure 1 shows the system boundary for the analysis presented here. The process starts with pretreatment. This step reduces contaminants and adjusts physical properties such as particle size, moisture content, and other properties needed to meet the specifications for conversion. The feedstock that is ready for conversion is sent to the pyrolysis reactor, from which the solid (char), gaseous (non-condensable gases or fuel gas), and liquid (pyrolysis oil) fractions are obtained. As described above, because there are two reported products, we defined two system boundaries for (1) companies producing pyrolysis oil, and (2) companies producing ULS diesel. The feedstock used for the conversion process is PUP that can be classified as post-consumer plastic (PCP) and post-industrial plastic (PIP).

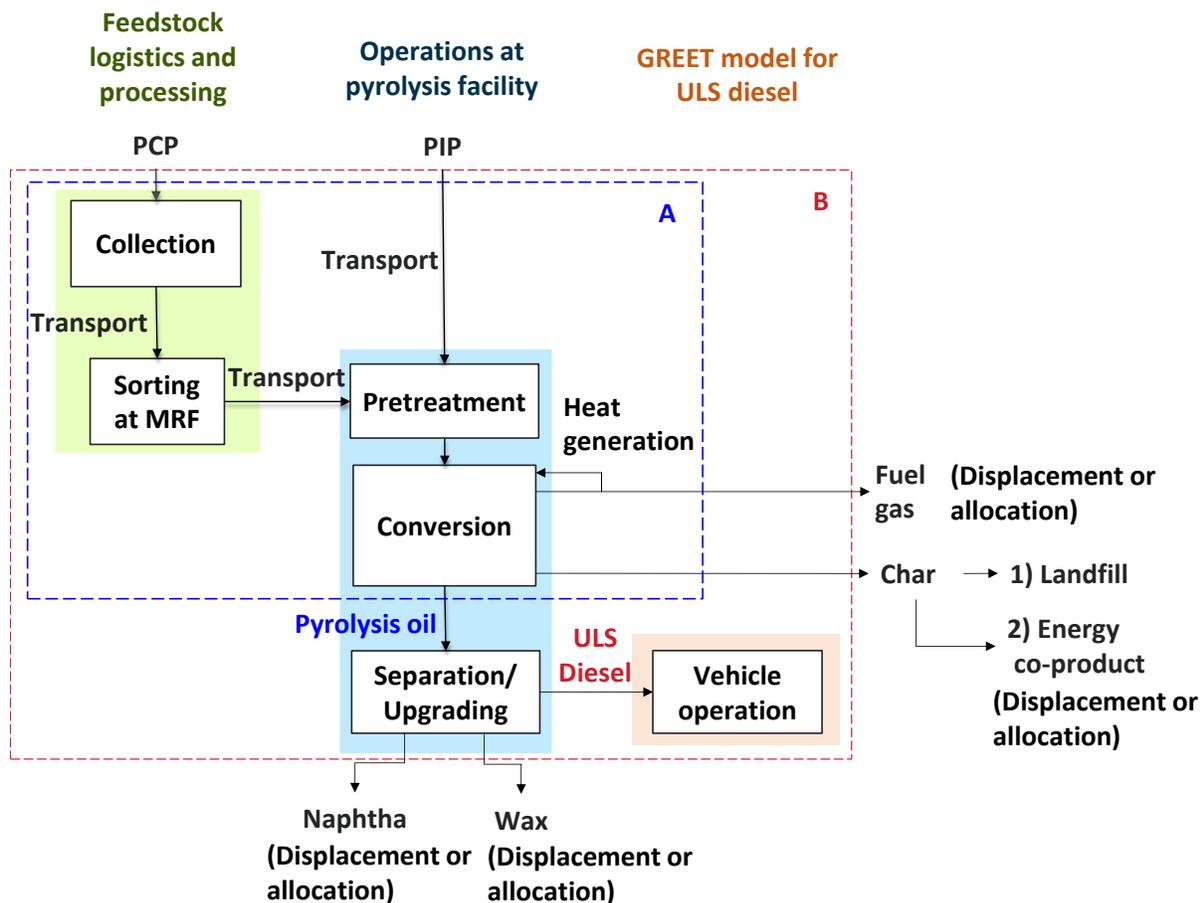
In this report we define PCP according to the International Organization for Standardization (ISO, 2008) and EPA (2020), which define PCP as a plastic material that is diverted from waste disposal after it is no longer useful. PIP is also known as pre-consumer plastic. PIP is a byproduct of industrial or manufacturing processes (Ocean Recovery Alliance, 2015). Regardless of the final product obtained, the process starts with the feedstock logistics and processing stage (green section in Figure 1).

Following the same assumptions presented in the previous study by Benavides et al. (2017), the PCP is collected using a refuse truck. The transportation distance was assumed to be 50 miles from the source of collection to the material recovery facility (MRF) where PCP is separated from MSW (Franklin Associates, 2018). The modeling of the MRF is based on information provided by Franklin Associates (2018), which used data from various MRF surveys. Details about the collection and sorting are presented in Section A.2 of the appendix.

According to the data provided by the surveyed companies, bales of PCP from the MRF were transported to the pyrolysis facility using a diesel truck for pioneer and  $N^{\text{th}}$ -plant facilities. The highest capacity plant used rail transport. Unlike the plastic in PCP, PIP is largely homogeneous and has little contamination; therefore, pretreatment processes such as sorting or washing might not be necessary for plastic conversion technologies like pyrolysis, so it could avoid going through the municipal recycling program. PIP is typically sent directly to recycling, energy recovery, or other end uses (Ocean Recovery Alliance, 2015). Where that was the case, no burdens from processing PIP were assigned and only transportation burdens were considered.

When companies produce ULS diesel, two additional stages were considered: (1) upgrading and/or separation, where the diesel fraction is separated from the rest of the liquid fraction, and (2) vehicle operation, where the ULS diesel is combusted in the vehicle. Vehicle operation emissions were leveraged from the information available in the GREET model for ULS diesel fuels (Argonne, 2020). Based on the information provided by the companies about the composition of the liquid fraction, naphtha and waxes were obtained as coproducts after the separation of ULS diesel. Only three of the surveyed companies provided data about upgrading and separation. Fuel gas and char are also two important coproducts of this process.

The next section discusses the different methods used to treat and allocate all the coproducts.



**Figure 1. System Boundary of PUP Conversion via Pyrolysis for: (A) Pyrolysis Oil Production, and (B) ULS Diesel Production.** We assume that upgrading/separation occurs at the conversion facility because most surveyed companies employ onsite upgrading (PCP: post-consumer plastic; PIP: post-industrial plastic; ULS: ultra-low sulfur; MRF: material recovery facility).

## 2.4 COPRODUCT TREATMENT AND USE

Figure 1 shows alternative scenarios for the use of fuel gas and char. Fuel gas can be combusted to generate electricity or heat. Depending on the amount of fuel gas generated, it can be used at the facility or exported, and char can be sent to landfill or considered as an energy coproduct. In Section A.3 of the appendix describes all possible alternative scenarios for treating these coproducts.

In this report, we focus our analysis on what the companies described as their current practice regarding the use of fuel gas and char. For instance, we assumed that fuel gas was combusted to produce heat used for internal operations. For companies with heating demands lower than the heat generated by the fuel gas, the leftover energy was considered an energy product. Electricity generation from char combustion was not considered a source of internal energy because most

companies mentioned that char is sent to landfill. The carbon content of this char was taken as a credit for GHG emissions because it is sequestered in the landfill.

As shown in Table 2, three methods were used to allocate the emissions and energy burdens of the main products (pyrolysis oil or ULS diesel, depending on the system boundary chosen for analysis):

- **Displacement:** The main product receives a credit for the energy, water consumption and emissions associated with the conventional products displaced by the coproducts. When no direct conventional product could be used to displace the alternative product, we approximated based on functionality and applications. That is the case for waxes, excess fuel gas, and char that are assumed to displace residual oil from petroleum refineries, natural gas, and petroleum coke, respectively. Details of these assumptions can be found in Benavides et al. (2017).
- **Energy allocation:** Energy, water consumption, and emissions burdens are allocated based on the energy output share of each coproduct.
- **Market allocation:** Energy, water consumption, and emissions burdens are allocated based on the revenue share of each coproduct.

In this report we present the results for the three allocation methods. We focus the discussion on our major findings using the energy and market allocation methods because these are conservative and will avoid the large coproduct effects from displacement methods. Market and energy allocation do not include the influence of char in the estimation of allocation factors because we assumed that the end use of char is for it to be landfilled (not used for energy recovery or sold as a product). Waxes are treated only as material feedstock because some facilities mentioned using them to produce lubricants or as a replacement for paraffin waxes; therefore, we did not include the energy from waxes in the energy allocation factors calculation. These allocation methods are consistent with those we previously presented in Benavides et al. (2017).

**Table 2. Approaches for Coproduct Treatment**

Product	Displacement	Energy- or Market-Based Allocation
Naphtha	Displaces conventional low-sulfur naphtha	Sold as energy product
Wax	Displaces petroleum wax (residual oil as approximation)	Sold as energy product
Fuel gas	Displaces natural gas	Excess fuel gas sold as energy product
Char	Displaces petroleum coke	Landfilled

Table 3 shows the values used in our estimates of the allocation factors for each product. The lower heating values (LHVs) for pyrolysis oil, fuel gas, and char were taken from the survey. Because product properties vary between the capacity groups, LHVs range between the maximum and minimum values of the groups. The LHV of waxes was calculated based on the average composition of the pyrolysis oil.

Because there was a lack of publicly available market values for fuel gas and char produced from pyrolysis, we used the market values of natural gas and petroleum coke, respectively, as

approximations. For the market values of diesel and fuel gas, we used the average price over the last 5 years, as reported by the U.S. Energy Information Administration (EIA, 2021a, 2021b). For naphtha, we considered the average of three values reported for the Gulf Coast, and for char we used the average of the prices of two types of pet coke.

**Table 3. Values Used to Estimate Allocation Factors**

<b>Product or Coproduct</b>	<b>For Energy Allocation (LHV [MJ/kg])</b>	<b>For Market Allocation (Market Value)</b>
Pyrolysis oil	42.7–43.4 <sup>a</sup>	3.2 USD/gal <sup>c</sup>
ULS diesel	43.1 <sup>b</sup>	1.9 USD/gal <sup>d</sup>
Naphtha	44.9 <sup>b</sup>	1.4 USD/gal <sup>e</sup>
Waxes	25.2–42.6 <sup>a</sup>	0.6 USD/lb <sup>f</sup>
Fuel gas	43.5–41.6 <sup>a</sup>	75 USD/ton <sup>g</sup>
Char	17.8–11.0 <sup>a</sup>	156 USD/ton <sup>h</sup>

<sup>a</sup> Data obtained from the surveyed companies.

<sup>b</sup> Source: Argonne (2020).

<sup>c</sup> Source: Raczkowski and Willard (2019).

<sup>d</sup> Source: EIA (2021a).

<sup>e</sup> Source: OPIS (2020).

<sup>f</sup> Source: ChemAnalyst (2021).

<sup>h</sup> Source: EIA (2021b)

<sup>g</sup> Source: Argus Media (2020).

## 2.5 GREET UPGRADE AND IMPLEMENTATION OF NEW INFORMATION

Although a PTF module had already been implemented in the GREET model, we adapted a new version of this module to incorporate the results of our new survey. **From the survey, we learned that most of the participating companies produce a pyrolysis oil product that can be converted into new chemicals or plastic, and very few companies produce fuel-type products.** Therefore, the updated PTF module has two sets of results, which represent the pyrolysis oil and fuel product pathways. The PTF module shows all stages of the system boundary (see Section 2.3 for details) from feedstock collection, sorting at MRF, pretreatment, conversion, and separation to final product (for the ULS diesel analysis).

## 3 RESULTS AND DISCUSSION

### 3.1 SURVEY RESULTS

We surveyed eight companies: seven operating in the United States and one in Europe. Because the survey obtained a variety of results, we decided to classify the surveyed companies based on their annual capacities, and the results were allocated accordingly (see Section 2.2). The annual capacity of the surveyed companies ranged from 270 to 291,800 tonnes of PUP per year, as shown in Table 4. Three cases were chosen for the analysis:

- **Pioneer plant:** Pyrolysis plants with a processing capacity of less than 50,000 tonnes of PUP per year. This type of plant reflects the current state of technology for the pyrolysis plants surveyed in this analysis. Their annual capacity ranges from 270 to 18,800 tonnes of PUP processed per year. Most of the surveyed companies (six) were in this category.
- **$N^{\text{th}}$ -plant:** Pyrolysis plants with a processing capacity of more than 50,000 tonnes per year. This type of plant represents the expected capacity of future commercial-scale pyrolysis facilities, likely between 50,000 and 100,000 tonnes per year (Goyal, 2021; Bettenhausen, 2020).
- **Highest-capacity plant:** Modeled pyrolysis facility with a processing capacity of approximately 291,800 tonnes per year.

Two of the eight plants were classified in the  $N^{\text{th}}$ -plant category, with one of them reaching 291,800 tonnes per year of plastic processing capacity. This latter plant was also classified as the highest-capacity plant. The data reported for this plant was based on simulated data that represents an increased capacity with optimized operations. The classification of the three categories is designed to help examine the effects of increasing capacity and the economy of scale. The results of this study focused on the  $N^{\text{th}}$ -plants because they reflect the projected capacity of the pyrolysis technology. Most of the data represent operations for the year 2020. However, data from 2021 and 2017 were provided by two different companies.

For the pioneer plants, five companies provided data from their operations, while one shared data based on process simulation. The data used to describe the highest-capacity plant is also based on process simulation. Table 4 summarizes other key characteristics of the surveyed companies. For identification, a code was assigned to each company. In addition to plant capacity based on feedstock processed, Table 4 presents the stage of development and annual production capacity based on the amount of pyrolysis oil each company produced.

Pioneer companies produced between 230 and 13,100 tonnes of pyrolysis oil per year. Pioneer plants are more established in the market, with longer operation times compared to an  $N^{\text{th}}$ -plant that has been in the market for less than a year. Pioneer plants that have been functioning for over a year are in the demonstration/commercial stage, while most plants that recently started operation are in the pilot and simulation stages. The definitions of each stage of development appear in Table A.1-2 of the appendix.

Table 4 also mentions the type of conversion being used at each facility: thermal or catalytic. Thermal conversion decomposes the structure of PUP into shorter hydrocarbon molecules using high temperatures (450–700°C). Catalytic conversion uses catalysts to increase the rate of

decomposition, which requires lower temperatures than thermal conversion. One of the surveyed plants performs catalytic pyrolysis using gamma alumina as the catalyst, but the great majority of plants use thermal conversion. Only two of the companies reported further processing of pyrolysis oil to ULS diesel; one of these companies is in the pioneer capacity group, and the other is in the  $N^{\text{th}}$ -plant category.

**Table 4. Key Characteristics of the Surveyed Companies**

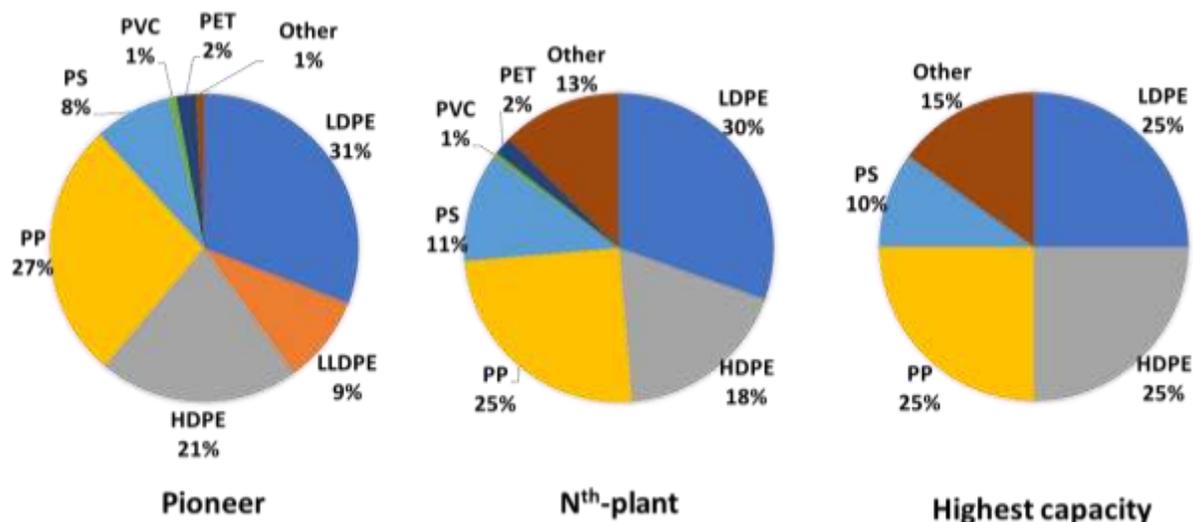
Type of Plant	Company Code <sup>a</sup>	Processing Capacity (thousand tonnes PUP/year) <sup>b</sup>	Production Capacity (thousand tonnes pyrolysis oil/year)	Type of Conversion	Stage of Development
Pioneer	1	0.3	0.2	Catalytic	Pilot scale
	3	5.1	2.7	Thermal	Commercial
	4	6.0	5.1	Thermal	Demonstration/ commercial
	6	15.0	12.3	Thermal	Demonstration/ commercial
	7	18.8	13.1	Thermal	Demonstration/ commercial
	8	7.0	5.3	Thermal	Simulation/ commercial
	<b>Average</b>	<b>8.7</b>	<b>6.6</b>		
$N^{\text{th}}$ -plant	2	113.5	66.1	Thermal	Demonstration/ commercial
	5	291.8	178.6	Thermal	Simulation/ commercial
	<b>Average</b>	<b>197.4</b>	<b>126.3</b>		
<b>Highest Capacity</b>	5	291.8	178.6	Thermal	Simulation/ commercial

<sup>a</sup> Company code is an identification number.

<sup>b</sup> Feedstock entering pretreatment (when applicable).

Figure 2 shows the aggregated shares of plastic types reported in the survey. The surveyed companies process a variety of polyolefins including HDPE, LDPE, linear low-density polyethylene (LLDPE), and PP as the major resins to produce pyrolysis oil or diesel. These shares ranged from 73% to 88% of the feedstock mixture. The two main polyolefins being processed were PP and LDPE for pioneer and  $N^{\text{th}}$ -plants. For the highest-capacity case, however, LDPE, PP, and HDPE were equally considered for conversion.

These results are consistent with current trends in plastic recycling in the United States, according to the EPA (2020). In the United States, polyethylene terephthalate (PET) and HDPE resins are not commonly processed by pyrolysis; instead, they are recycled at a high rate (19% and 9% of total waste in MSW, respectively) compared to other resins like PP (0.6%) and LDPE (4.3%). Therefore, the companies limited the share of PET in the feedstock mixture to a maximum value of 5%. PET can shift the pyrolysis to combustion because PET contains oxygen and generates low yields of pyrolysis oil.

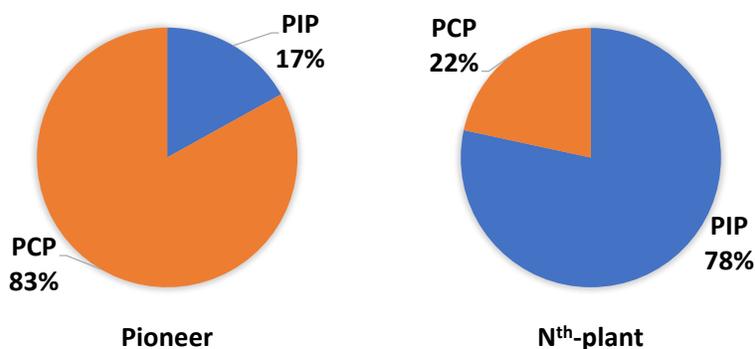


**Figure 2. Composition of Feedstock Mixture Entering the Pyrolysis Facility for Pioneer Plants, N<sup>th</sup>-plants, and Highest-capacity Plants** (PET: polyethylene terephthalate; PVC: polyvinyl chloride; PS: polystyrene; PP: polypropylene; HDPE: high-density polyethylene; LLDPE: linear low-density polyethylene; LDPE: low-density polyethylene).

Companies also restrict the moisture content in feedstock to less than 5%, and the percentage of polyvinyl chloride (PVC) resins to 3% or less of total feedstock mass. This is mainly because during pyrolysis the chlorine contained in PVC produces hydrochloric acid, which is corrosive to equipment. These chemicals could deteriorate equipment and increase production costs. However, chlorine is present in some additives used in the production of other resins, so it is impossible to completely remove it from the feedstock.

As already mentioned, PCP is shipped to the companies from MRFs, because it must be sorted after being collected. According to ISO (2008) and EPA (2020), this category includes furniture and furnishings, packaging waste, transportation plastic, and agricultural plastic. In contrast to PCP, the pyrolysis facilities receive PIP directly from industries or commerce, because this source is comprised of non-compliant products and scrap material and therefore is less contaminated than PCP.

The contributions of PCP and PIP as feedstocks varied considerably among the three plant capacity groups, as shown in Figure 3. For example, pioneer plants rely more on PCP (83%), while N<sup>th</sup>-plants and the highest-capacity plants use more PIP (78% and 100%, respectively). Pioneer plants depend more on PCR, because this is less expensive for their operations and MRF plants are located to closer to the facilities. The surveyed plants with higher capacities rely more on PIP because it is necessary to process a more homogenous feedstock and secure a consistent supply.



**Figure 3. Contribution of Different Sources to Feedstock Mixture for Pioneer and N<sup>th</sup>-plants** (PCP: post-consumer plastic; PIP: post-industrial plastic. The highest-capacity plant uses 100% PIP as feedstock).

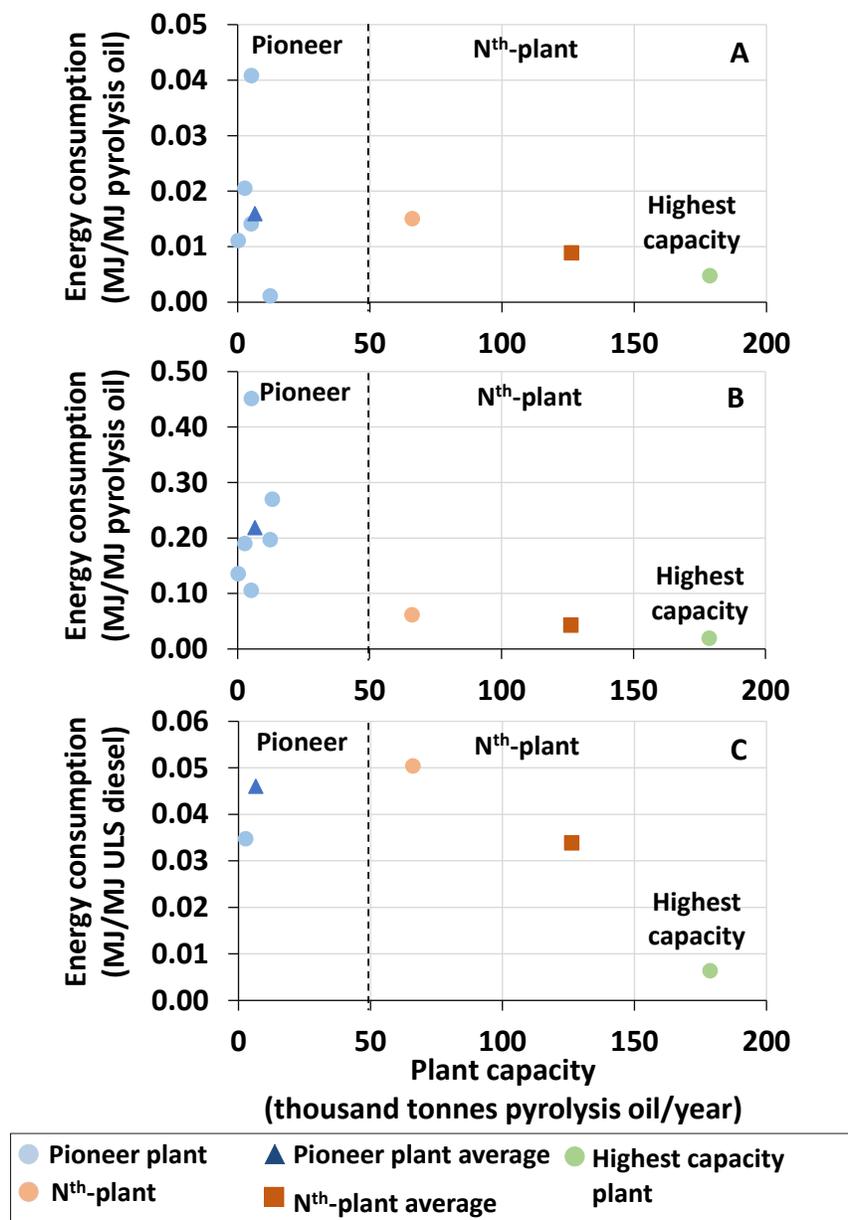
The transportation data is presented in Table 5. This table includes information about the transportation of feedstocks and products, and the types of transportation and fuel used. As shown in this table, pioneer plants have the shortest distances for collection and distribution compared to higher capacity plants, which implies that pioneer plants rely on local markets. As their capacity increases, the market will need to expand in order to supply more material. The highest-capacity case must travel a greater distance; therefore, the material is transported by rail, which makes the process more economical.

**Table 5. Transportation Data for Each Capacity Group**

Parameter	Type of Plant		
	Pioneer	N <sup>th</sup> -plant	Highest Capacity
Distance from feedstock supplier to pyrolysis facility (mi)	82	207	300
Type of transportation	Truck	Truck	Rail
Type of fuel	Diesel	Diesel	Diesel
Distance from pyrolysis facility to customer <sup>a</sup>	182	225	300
Type of transportation	Truck	Truck	Rail
Type of fuel	Diesel	Diesel	Diesel

<sup>a</sup> Customer to the end product. This refers to a refinery or chemical producer, depending on what the final product is.

The gross energy requirements per unit of energy (MJ) of product for the different process stages in the pyrolysis facilities (i.e., pretreatment, conversion, and upgrading/separation) are shown in Figure 4. In general, the gross energy requirements of the pioneer plants are higher than those from the N<sup>th</sup>-plants and highest-capacity plants, because pioneer plants have lower energy efficiency and lack economies of scale. In Figure 4A, only five companies in the pioneer group reported energy data for pretreatment; one company receives the feedstock ready for conversion.



**Figure 4. Gross Energy Requirements per Stage at Pyrolysis Facilities: (A) Pretreatment, (B) Conversion, and (C) Upgrading/separation.** For pretreatment and conversion cases, data are presented per megajoule of pyrolysis oil. Upgrading and separation data are presented in megajoules of ULS diesel product. The dotted line separates the data from pioneer and  $N^{\text{th}}$ -plant groups.

The lowest energy requirements for pretreatment were in the highest capacity facility; requirements are 70% and 46% lower compared to the average values of the pioneer and  $N^{\text{th}}$ -plant facilities, respectively. All facilities used 100% electricity as their energy source, and none reported using water during pretreatment. In the conversion stage, the energy requirements of the highest capacity facility were 91% and 55% lower than the average value of the pioneer and  $N^{\text{th}}$ -plants, respectively.

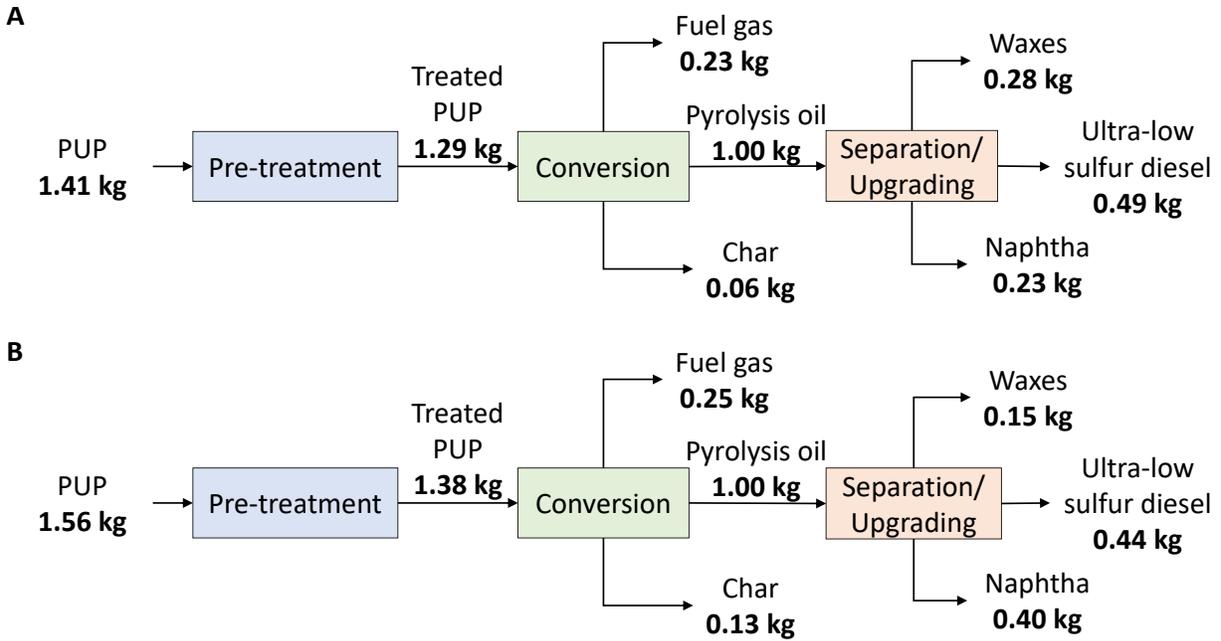
Table 6 presents the shares of energy sources in the conversion stage. Pioneer plants used a larger share of electricity for conversion (34%) compared to  $N^{\text{th}}$ -plants and the highest-capacity facility (3% and 17%, respectively). Pioneer plants used between 13% and 48% of electricity as an additional energy source to support the conversion process, and one pioneer facility operates the conversion reactor using 100% electricity. In the case of  $N^{\text{th}}$ -plants, one facility used 100% natural gas for conversion energy, while the highest-capacity facility used 83%. Due to a lack of information about the upgrading/separation stage in the pioneer group, we estimated the average energy consumption by adjusting the data of the company that produced diesel fuel as its final product using the average yield of ULS diesel estimated for the pioneer plants. The energy requirements of upgrading/separation at the highest-capacity operation were 86% and 81% lower compared to the pioneer and  $N^{\text{th}}$ -facilities, respectively.

**Table 6. Share of Energy Sources for Conversion and Upgrading/Separation**

Stage	Energy Source	Type of Plant		
		Pioneer	$N^{\text{th}}$ -plant	Highest Capacity
Conversion	Natural gas	66%	97%	83%
	Electricity	34%	3%	17%
Upgrading/Separation	Natural gas	97%	94%	19%
	Electricity	3%	6%	80%

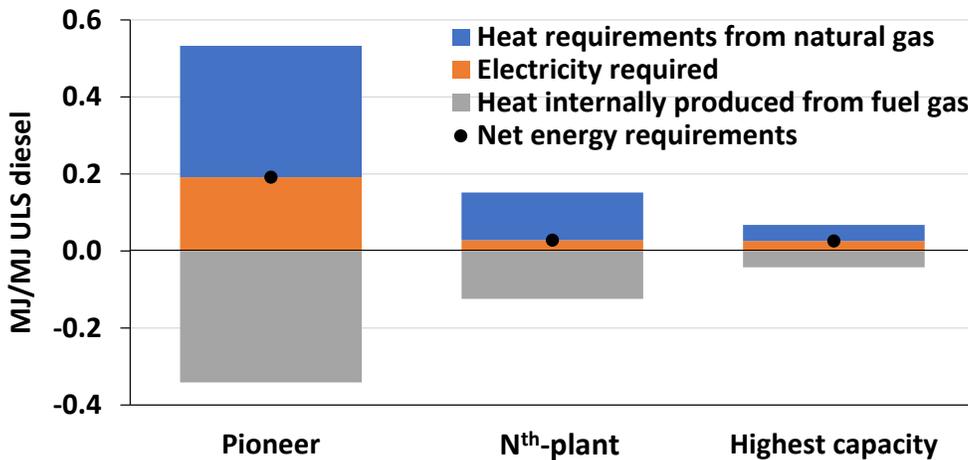
The mass balances of the pioneer and  $N^{\text{th}}$ -plant pyrolysis facilities are shown in Figure 5A and 5B, respectively. This figure only provides data about the three steps the pyrolysis facilities reported in the surveys. Results for the highest capacity plant are not presented, in order to protect its confidentiality; however, its information is aggregated in the flow diagram of Figure 5B.

Interestingly, the yield of the pioneer facilities was slightly higher than  $N^{\text{th}}$ -plants, because 7% less PUP feedstock is required to produce 1 kg of pyrolysis oil in pioneer plants. The amount of fuel gas was very similar between the two plant capacity groups (0.23 kg in pioneer versus 0.25 kg in  $N^{\text{th}}$ -plants); however, char production was 2.1 times higher in  $N^{\text{th}}$ -plants than in pioneer plants. The yields of the solid, gaseous, and liquid fractions were estimated, respectively, in 4%, 18%, and 78% (pioneer plants) and 10%, 18%, and 72% ( $N^{\text{th}}$ -plants). Another interesting result was that more waxes and less naphtha were produced in the pioneer facilities compared to  $N^{\text{th}}$ -plants. This selective production of hydrocarbon fractions can be derived from the operation conditions and current target products of the companies in each category.



**Figure 5. Average Mass Balance for (A) Pioneer Plants and (B) N<sup>th</sup>-plants**

After calculating the gross energy requirements of the pyrolysis facilities—which do not incorporate primary energy estimates—and the yields of the different products and coproducts, we estimated the energy that can be internally produced from fuel gas. The net energy requirements were obtained by subtracting this internally produced energy from the gross energy requirements. The results of these estimates are shown in Figure 6.



**Figure 6. Energy Requirements and Internal Energy Production per Plant Capacity Group**  
The net energy requirements are indicated by black dots. Fuel gas is a coproduct from the conversion process.

Two of the companies provided information about their CAP emissions. These CAPs are regulated by the EPA and include emissions of volatile organic compounds (VOCs), nitrogen

oxides (NO<sub>x</sub>), sulfur oxides (SO<sub>x</sub>), particulate matter (PM<sub>10</sub> and PM<sub>2.5</sub>), and carbon monoxide (CO) (Sun et al., 2019). The EPA establishes a threshold of 100 tonnes of CAP emissions per year to require a Title V permit (Good Company, 2021). The EPA regulations are established annually and are not evaluated per megajoule of product. Some companies mentioned that they are categorized as low-volume emitters according to these regulations from EPA.

Good Company (2021) also confirmed that plastic pyrolysis facilities have annual CAP emissions that are below the limits established by the EPA. Plastic pyrolysis facilities emit 15 tonnes of VOCs per year, an amount approximately 85% lower than that from food production facilities (101 tonnes of VOCs per assembly plant per year). In addition, the annual PM<sub>10</sub> emissions of plastic pyrolysis facilities (11 tonnes per year) are less than half of those from food-processing companies (28 tonnes per facility per year) (Good Company, 2021).

VOC and PM emissions from one pioneer company were less than 6 tonnes per year, which confirms that pyrolysis facilities generate emissions below the EPA threshold mentioned above. However, the inputs of the highest-capacity plant showed that the emissions of CO, PM<sub>10</sub>, and PM<sub>2.5</sub> exceeded the limits established by EPA (90 tonnes per year). Table 7 shows the aggregated data for non-combustion emission per megajoule of pyrolysis oil for the few pyrolysis facilities that provided that information. Because the density and the fraction of diesel in the pyrolysis oil vary in each plant capacity group (Figure 5), the non-combustion emissions are presented as ranges in Table 7.

**Table 7. Non-combustion Emissions from Pyrolysis Facilities (mg/MJ of product)**

Product	VOCs	CO	NO <sub>x</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	SO <sub>x</sub>
Pyrolysis oil	6.8–6.9	37.7–38.3	10.3–10.5	21.2–21.6	18.2–18.4	0.01
Diesel	14.0–19.2	77.8–106.7	21.3–29.3	43.8–60	37.5–51.4	0.03–0.04

Material inputs were another outcome obtained from the survey. However, within each plant capacity group not all facilities need to use the same materials to operate. Therefore, we obtained the mass of material inputs or chemicals (energy in the case of hydrogen) per megajoule of product in each group based on the average of only those facilities that reported it. Because these quantities do not represent all facilities, their influence was evaluated in a sensitivity analysis (Section 3.3.3). The materials and their corresponding amount per megajoule of product are presented in Table 8.

These materials are only used either in the conversion or in the separation/upgrading stages for both pioneer and *N*<sup>th</sup>-plants. For example, only pioneer plants report the application of calcium oxide, hydrochloric acid, sodium hydroxide, and nitrogen in the conversion process, while only *N*<sup>th</sup>-plants mentioned the use of solvents (methyl ethyl ketone and toluene) and hydrogen in the separation/upgrading process. These chemicals were used to improve the quality of liquid products (calcium oxide, sodium hydroxide and hydrochloric acid), recover oil from coproducts (methyl ethyl ketone and toluene), provide inert conditions in the reactor (nitrogen), and production of fuels (hydrogen and Ni/Mo/gamma alumina). As shown in Table 8, the highest-capacity plant did not report the use of chemicals or catalysts.

**Table 8. Material Inputs in Each Plant Capacity Group (g/MJ of product)<sup>a</sup>**

Material Input	Stage of Use	Pioneer		N <sup>th</sup> -plant	
		Pyrolysis Oil	ULS Diesel	Pyrolysis Oil	ULS Diesel
ZSM-5 catalyst <sup>b</sup>	Conversion	0.0002	0.0004	—	—
Ni/Mo/gamma alumina catalyst	Separation/upgrading	—	—	—	0.002
MEK	Separation/upgrading	—	—	—	0.021
Toluene	Separation/upgrading	—	—	—	0.023
Hydrogen (MJ/MJ of product)	Separation/upgrading	—	—	—	0.07
Calcium oxide	Conversion	4.2	8.7	—	—
Hydrochloric acid	Conversion	0.3	0.5	—	—
Sodium hydroxide	Conversion	0.6	1.2	—	—
Nitrogen	Conversion	0.1	0.3	—	—

<sup>a</sup> The highest-capacity plant does not require any material inputs. MEK: methyl ethyl ketone.

<sup>b</sup> ZSM-5: Zeolite Socony Mobil-5.

### 3.2 LCA RESULTS

In this section, we assess the environmental impacts of converting PUP to pyrolysis oil and ULS diesel. Because of these two potential products, two functional units were chosen for comparison: per 1 MJ of pyrolysis oil or per 1 MJ of ULS diesel. In the pyrolysis oil case, we report results on cradle-to-gate basis. For the ULS diesel case, we present the results on a cradle-to-grave basis.

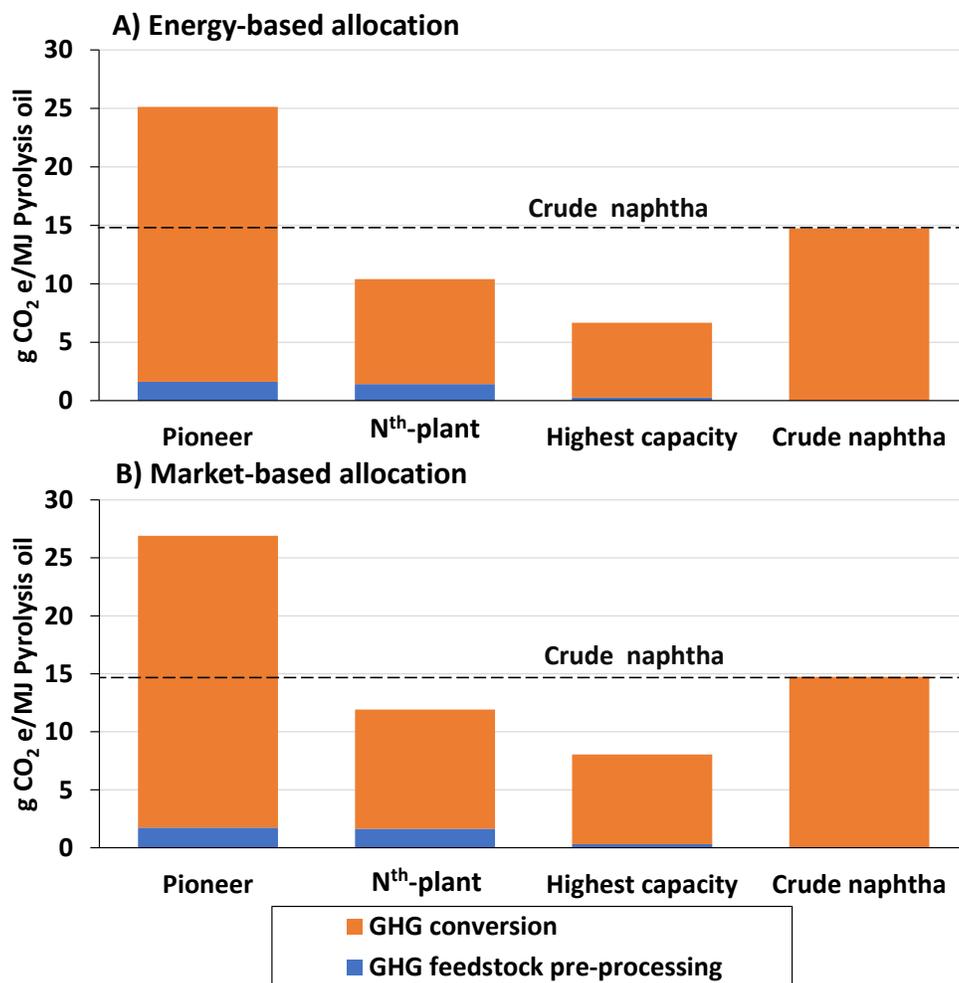
The GREET model was used to calculate the lifecycle GHG emissions (grams of CO<sub>2</sub>-equivalent), fossil energy consumption (MJ), water consumption (liters, L), and criterial air pollutants (grams). These results were obtained by modeling environmental metrics in the updated PTF tab in GREET using the new survey results. The GHG emissions include carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O), calculated based on the 100-year global warming potentials for CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O emissions, which are 1, 30, and 265, respectively (IPCC, 2014). For this analysis, we used the U.S. electricity generation mix and natural gas produced in the United States (Argonne, 2020).

Section A.4 of the appendix presents the LCA results for all types of plants, differentiating between simulated and non-simulated data.

#### 3.2.1 GHG Emissions

The results for GHG emissions breakdown of the pyrolysis oil case are presented in Figure 7. To compare the results, we chose crude naphtha as the baseline because of the potential to use pyrolysis oil as a replacement of crude naphtha (Jeswani et al., 2021). In the United States crude naphtha is a major feedstock for steam cracking (Kootungal, 2015) which can be converted to other chemicals (i.e., ethylene). The GHG emissions of crude naphtha are leveraged from the GREET model (Argonne, 2020).

Figure 7 shows results for two allocation methods (energy- and market-based). Details on GHG emissions for the displacement method are shown in Figure A-4 of the appendix. Although both allocation methods report similar emission reduction trends, in the energy allocation case the GHG emissions are slightly lower than those from the market allocation case, because the allocation factors in the energy-based method are lower than those in the market case (see Table A.3-2 in the appendix). Figure 7A shows that the  $N^{\text{th}}$ -plant and highest capacity facilities produce 29% and 55% lower emissions, respectively, than the baseline. Pioneer plants do the opposite: they produce 71% more GHG emissions than the baseline.



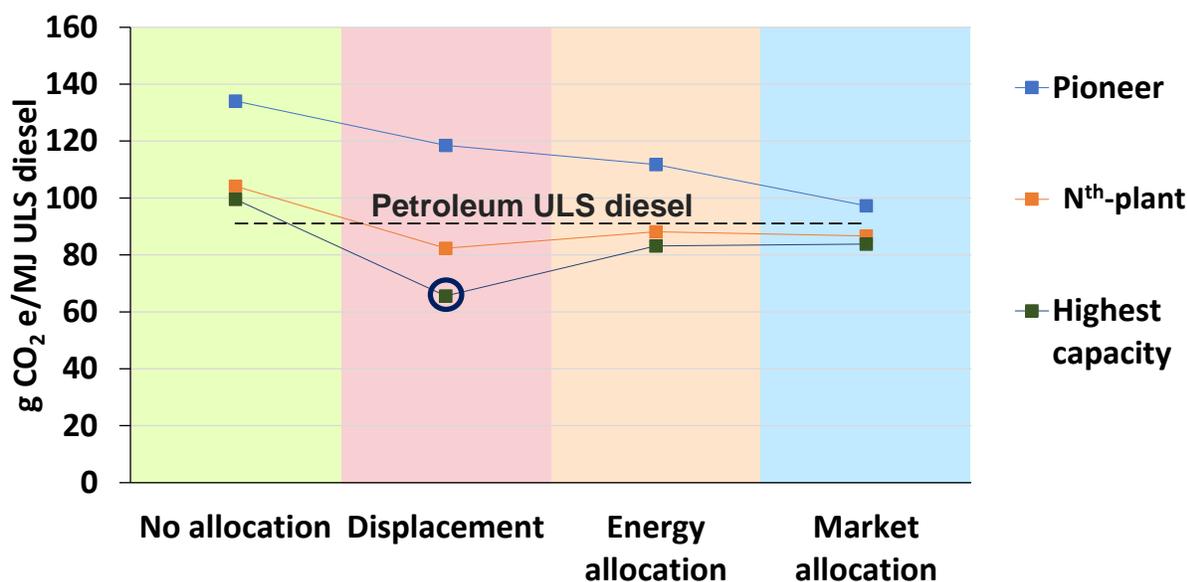
**Figure 7. GHG Emissions by Stage for Pyrolysis Oil Production.** (A) energy-based allocation, (B) market-based allocation. For pyrolysis oil, GHG emissions of feedstock preprocessing consider feedstock logistics and processing and GHG emissions of conversion include the stages of pretreatment and conversion at the pyrolysis facility. For crude naphtha (baseline), the GHG emissions of conversion include cradle-to-gate emissions of its production from crude oil. The GHG emission of crude naphtha are 14.7 gCO<sub>2</sub>e/MJ (Argonne, 2020).

Figure 7B (Market allocation) shows emission reductions of 19% and 45% for the  $N^{\text{th}}$ -plant and highest-capacity facilities, respectively, compared to the baseline and an increase of 83% for pioneer plants compared to the baseline. Conversion emissions are the major contributors to total GHG emissions in all facility groups because the energy requirements in pioneer plants are

higher than those of the  $N^{\text{th}}$ -plants and the highest-capacity facility. This is particularly important because it highlights the impact that economies of scale have in reducing the GHG emissions for the production of pyrolysis oil.

Figure 8 presents the lifecycle GHG emission results for PUP-derived ULS diesel for all of the coproduct allocation methods described in Section 2.4. The petroleum-based ULS diesel GHG emissions reported in GREET were used for comparison (Argonne, 2020). The lowest GHG emissions were achieved for the highest-capacity case with the displacement method. These emissions were 28% lower than petroleum-based ULS diesel. For the energy- and market-allocation case, the highest-capacity plant only had GHG emission reductions of 9% and 8%, respectively, compared to petroleum-based ULS diesel.

The displacement method represents a more optimistic emission-reduction scenario because of the credits taken from the coproducts (see Table A.3-3 of the appendix). However, these credits depend upon the assumptions used for displacing the coproducts (see Table 2).

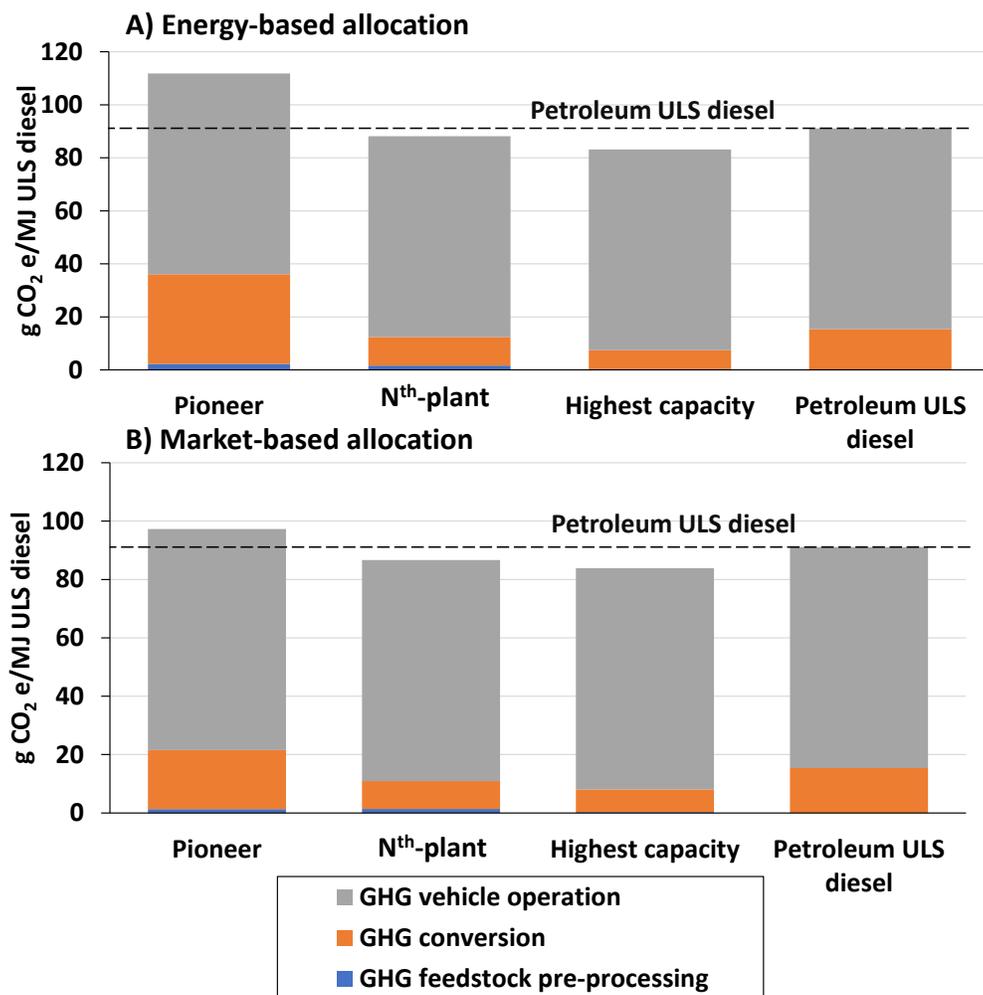


**Figure 8. Comparison of Lifecycle GHG Emissions under Different Coproduct Allocation**

**Methods.** These emissions include the emission from feedstock processing, conversion, upgrading/separation, and vehicle use. GHG emissions of petroleum-based ULS diesel are 91.1 g CO<sub>2</sub>e/MJ (Argonne, 2020).

In the pioneer plants, an increase of up to 47% in the GHG emissions was observed compared to petroleum-derived ULS diesel (baseline) when no allocation is taken into account. Regardless of the allocation method chosen for the pioneer plants, the GHG emissions were higher than those of petroleum-based ULS diesel; values ranged from 30% (displacement) to 7% (market allocation). In contrast, the GHG emission of the  $N^{\text{th}}$ -plant and highest-capacity facilities were slightly lower than those of petroleum ULS diesel, with the emission reduction slightly improving as the plant capacity increased. However, there is not a major difference between energy and market allocation results.

To better understand the contributors to the GHG emission of PUP-derived ULS diesel production via pyrolysis, Figure 9 shows a breakdown of the GHG emissions presented in Figure 8. Regardless of the allocation method used, the GHG emissions of vehicle operation were the major contributors to the lifecycle GHG emissions, representing 68%, 86%, and 91% for the pioneer,  $N^{\text{th}}$ , and highest-capacity facilities, respectively. The GHG emissions for conversion and upgrading/separation were significantly higher in the pioneer facility (30% of lifecycle GHG emissions) compared to the other facility groups and ULS petroleum diesel, because the pioneer plants have higher energy demands. Similar to the analysis of pyrolysis oil, the economies of scale, represented by the  $N^{\text{th}}$ -plants and the highest-capacity facility, lead to GHG emissions reductions of PUP-derived ULS diesel that are even lower than those of fossil-based counterparts.



**Figure 9. GHG Emissions to Produce ULS Diesel via Pyrolysis.** For PUP-derived ULS diesel, GHG emissions of feedstock preprocessing consider feedstock logistics and processing; GHG emissions of conversion refer to the stages of pretreatment, conversion, and separation/upgrading at pyrolysis facility; and GHG vehicle operation refers to the emissions produced during fuel combustion. For petroleum-derived diesel, GHG emissions of conversion refer to the cradle-to-gate emissions from producing ULS diesel. GHG emissions of petroleum-based ULS diesel are 91.1 g CO<sub>2</sub>e/MJ (Argonne, 2020).

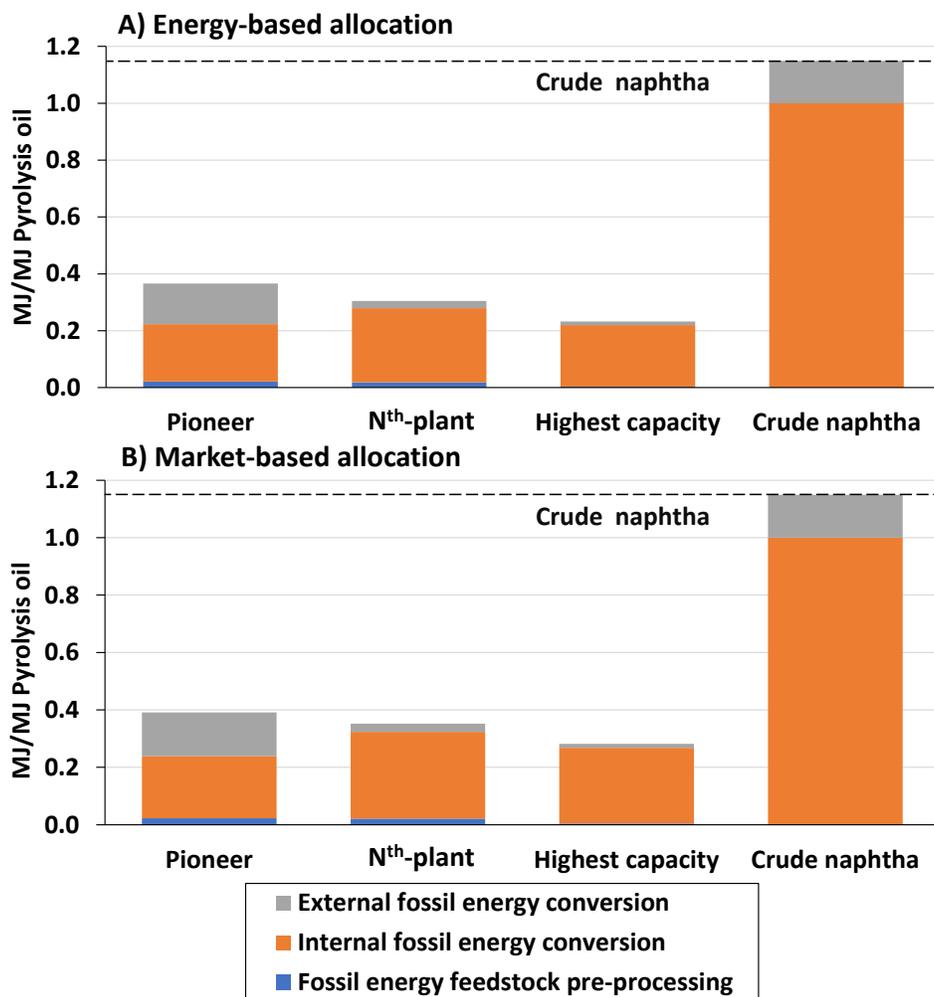
### 3.2.2 Fossil Energy Consumption

Estimates of the fossil energy consumption to produce PUP-derived pyrolysis oil are presented in Figure 10 for both energy- and market-allocation methods. In all capacity groups there was a considerable reduction in fossil fuel consumption. Figure 10 shows that, using energy allocation, the fossil fuel consumption decreased by 68%, 73%, and 80% in pioneer plants,  $N^{\text{th}}$ -plants, and the highest-capacity facility, respectively, compared to crude naphtha. This shows how much this metric improves as plant capacity increases.

Most of the fossil energy came from the internal energy used during the conversion process, which is the energy embedded in the PUP used as feedstock. For the pioneer plants, this internal energy represented 55% of the total fossil energy. For the  $N^{\text{th}}$ -plants and highest-capacity facilities it represented 85% and 93%, respectively, of the total fossil energy.

The external energy, referring to the natural gas and electricity consumption during conversion, represented 39% of the total fossil energy in the pioneer case, more than the amounts for the  $N^{\text{th}}$ -plants and highest-capacity facilities. This is consistent with the results presented in Figure 6. Pioneer plants have lower internal fossil energy consumption for conversion (0.2 MJ/MJ pyrolysis oil) compared to  $N^{\text{th}}$ -plants and highest-capacity facilities.

When the external fossil energy for conversion was added, the total fossil energy consumption was higher for pioneer plants compared to  $N^{\text{th}}$ -plants and highest-capacity facilities. Therefore, the increased requirements of natural gas and electricity offset the benefits of higher conversion yields in the pioneer plants. Overall, no major differences were observed when the results from energy and market value allocation were compared.



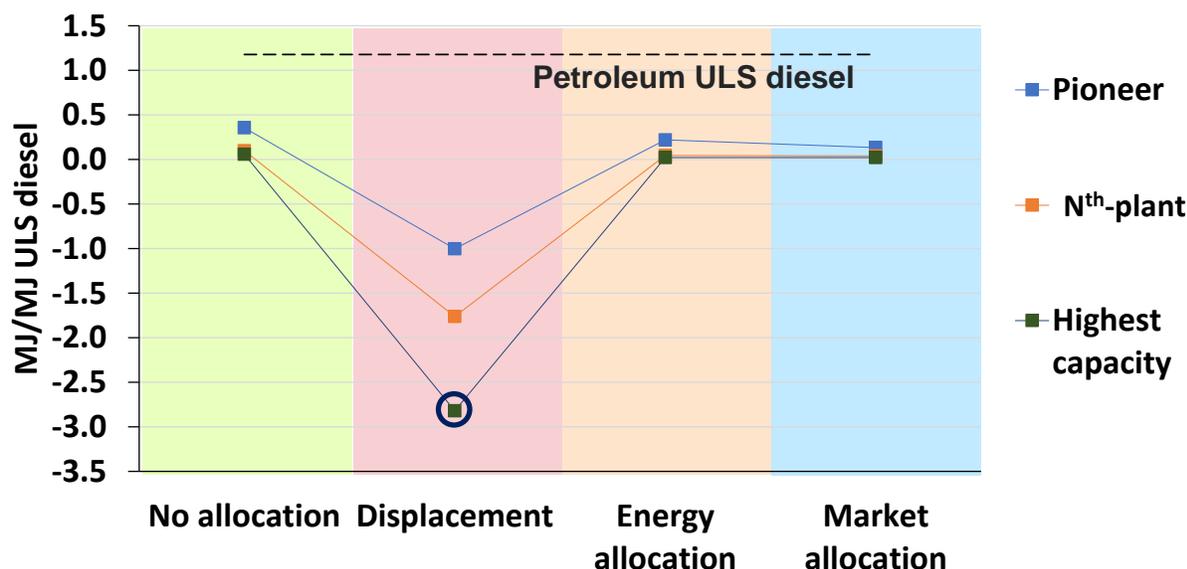
**Figure 10. Fossil Energy Consumption by Production Stage for Pyrolysis Oil.** For pyrolysis oil, fossil energy for feedstock preprocessing includes feedstock logistics and processing; internal fossil energy for conversion refers to the energy embedded in the PUP used in the conversion process; and external fossil energy for conversion refers to the use of natural gas and electricity to provide energy to the process. For crude naphtha, internal fossil energy for conversion considers the energy embedded in the product and external fossil energy from conversion refers to the energy from fuels use to produce crude naphtha. Fossil energy consumption for crude naphtha is 1.1 MJ/MJ (Argonne, 2020).

The fossil energy consumption of the different allocation methods for ULS diesel (Figure 11) shows trends similar to the results presented for GHG emissions (see Figure 8). Using the displacement method, the highest-capacity plant provided the lowest fossil energy consumption (negative 2.8 MJ/MJ ULS diesel) among all scenarios evaluated due to coproduct credits (0.8, 1.7, and 0.1 MJ/MJ ULS diesel due to fuel gas, naphtha, and waxes, respectively), as shown in Figure A-4 of the appendix.

For all allocation methods, there were significant reductions in fossil energy consumption. For example, using energy allocation, these reductions were estimated to be 81%, 96%, and 98% for pioneer plants, N<sup>th</sup>-plants, and the highest capacity facility, respectively, compared to petroleum-based ULS diesel. For the displacement treatment, all plant capacity groups show negative fossil

energy consumption, because the credits from coproducts are higher than the energy demand of the process.

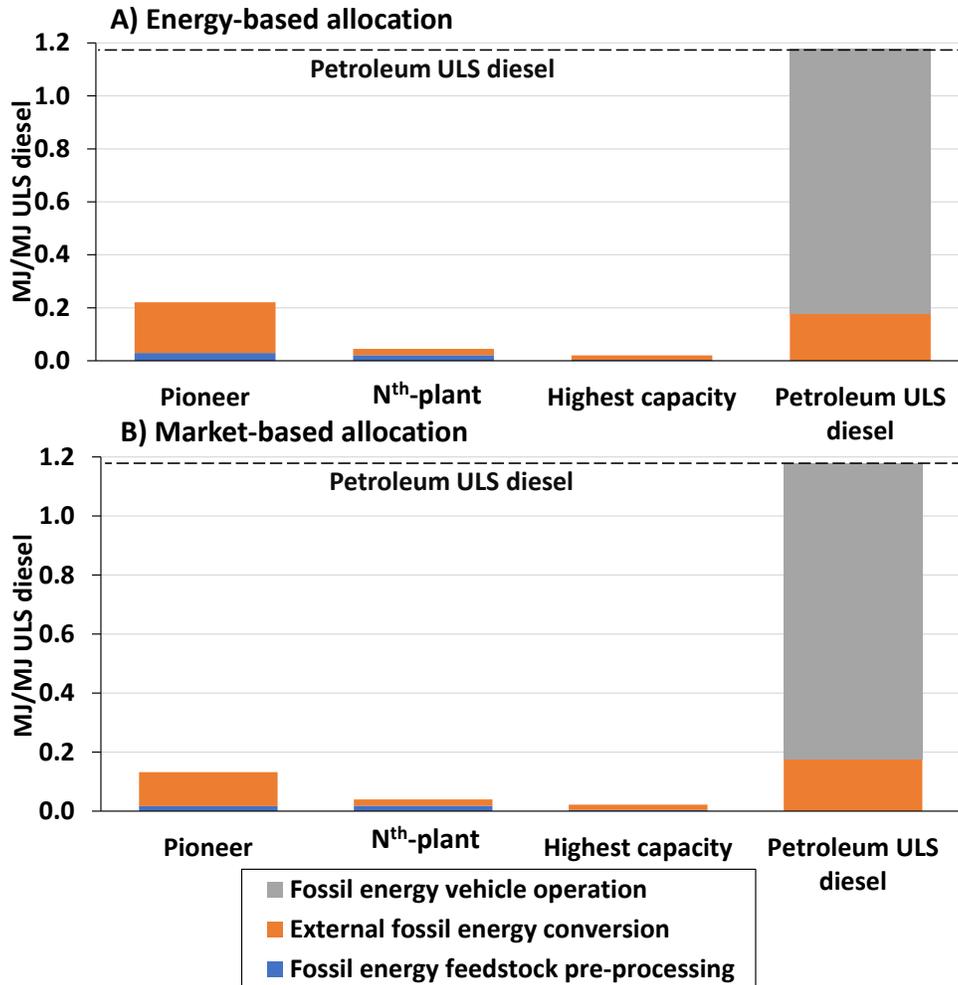
Because we included the use phase in the ULS diesel comparison, an important assumption for the PUP-derived ULS diesel analysis is that the internal energy from the feedstock in conversion and vehicle operation is not included in the estimates of fossil energy consumption, in order to make a fair comparison with petroleum-based ULS diesel. The major reason for this assumption is that the feedstock used to produce PUP-derived ULS diesel comes from second-generation feedstock and its internal energy has already been used to produce plastic in its first life. This distinction is shown in Figure 12, which presents the fossil energy consumption breakdown for energy and market allocations.



**Figure 11. Fossil Energy Consumption to Produce PUP-derived ULS Diesel for Different Coproduct Treatment Methods.** The fossil energy consumption of PUP-derived ULS diesel includes the energy of feedstock processing and external energy for conversion. Fossil energy consumption for petroleum-derived ULS diesel includes the energy of feedstock processing, internal and external energy for conversion, and energy for vehicle operation. Fossil energy consumption of fossil-based ULS diesel is 1.2 MJ/MJ (Argonne, 2020).

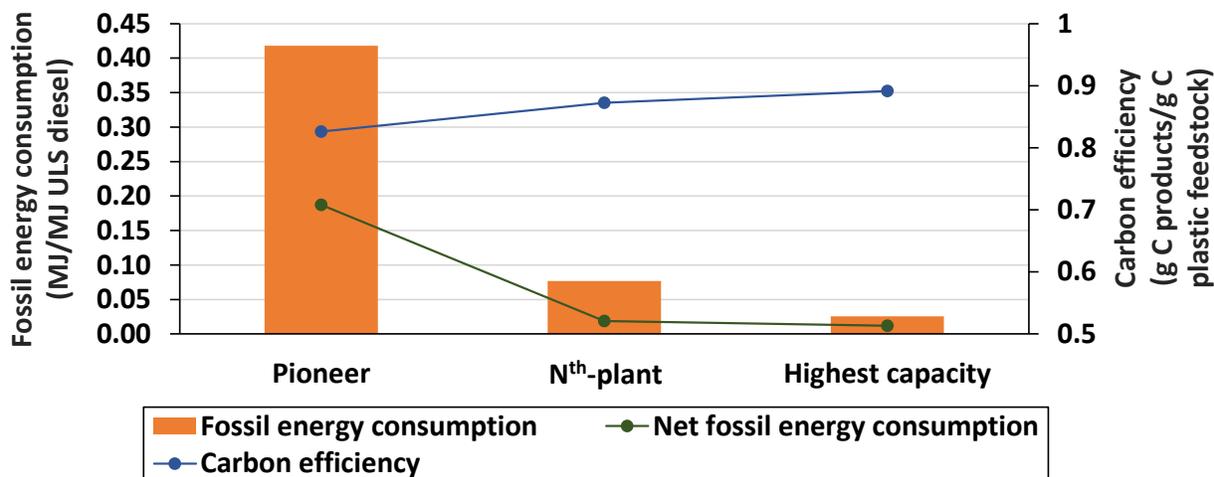
Figure 12 clearly shows how fossil energy consumption is reduced as a result of producing PUP-derived ULS diesel via pyrolysis, as compared to petroleum ULS diesel; none of the internal energy was considered during the conversion separation/upgrading and vehicle operation. Only the energy from feedstock logistics and processing and the external energy for pretreatment, conversion, and separation/upgrading were included to estimate total fossil energy consumption.

Pioneer plants have the largest fossil energy consumption (0.2 MJ/MJ ULS diesel) among all capacities, because they have a high demand for energy (external energy in the figure) to convert and upgrade the PUP into ULS diesel fuel. This external energy comprises 87%, 53%, and 83% of the total fossil energy consumption of the pioneer plants, N<sup>th</sup>-plants, and highest-capacity plant, respectively.



**Figure 12. Fossil Energy Consumption to Produce ULS Diesel via Pyrolysis.** For PUP-derived diesel, fossil energy for feedstock preprocessing includes feedstock logistics and processing, and external fossil energy for conversion refers to the use of natural gas and electricity to provide energy to the process. For petroleum-derived ULS diesel, fossil energy for conversion represents the energy from fuels required in the cradle-to-gate production and fossil energy for vehicle operation refers to the energy consumed during fuel combustion. Fossil energy consumption for petroleum-derived ULS diesel is 1.2 MJ/MJ (Argonne, 2020).

The GHG emissions and fossil energy results imply the effect from the economies of scale and the benefits of transitioning to more advanced and consolidated technology. The lower energy requirements and reduced GHG emissions of a more efficient use of energy per amount of feedstock processed reflect the improvements of a continuous operation at a larger scale. Figure 13 shows that not only do *N<sup>th</sup>*-plant and highest capacity facilities have lower net fossil energy requirements compared to the pioneer plants, but also their carbon efficiency is higher—meaning that most of the carbon in the feedstock ends up as part of their products and coproducts. Overall, we can see an advantage when transitioning from the current technologies to those that are optimized and using economies of scale.

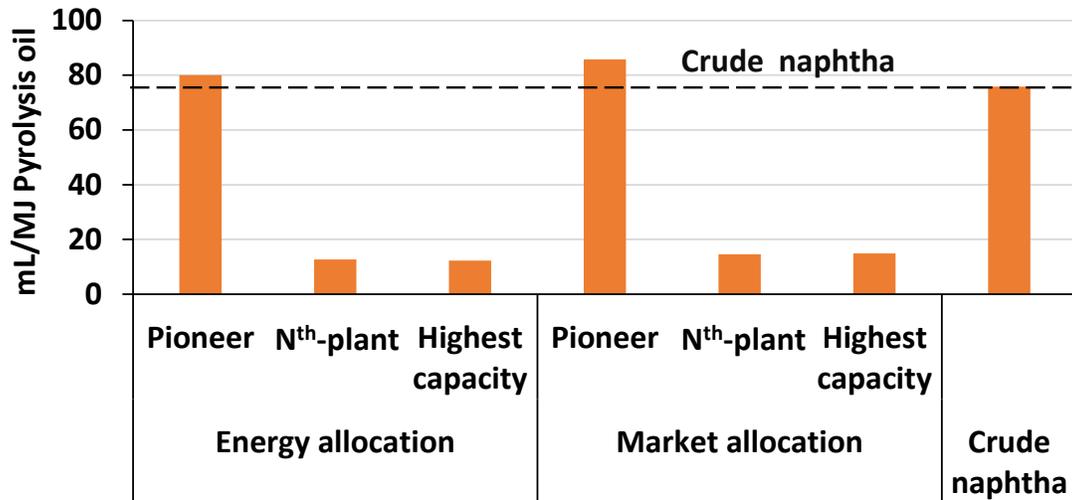


**Figure 13. Net Energy Requirements and Carbon Efficiency of the Plant Capacity Groups.**

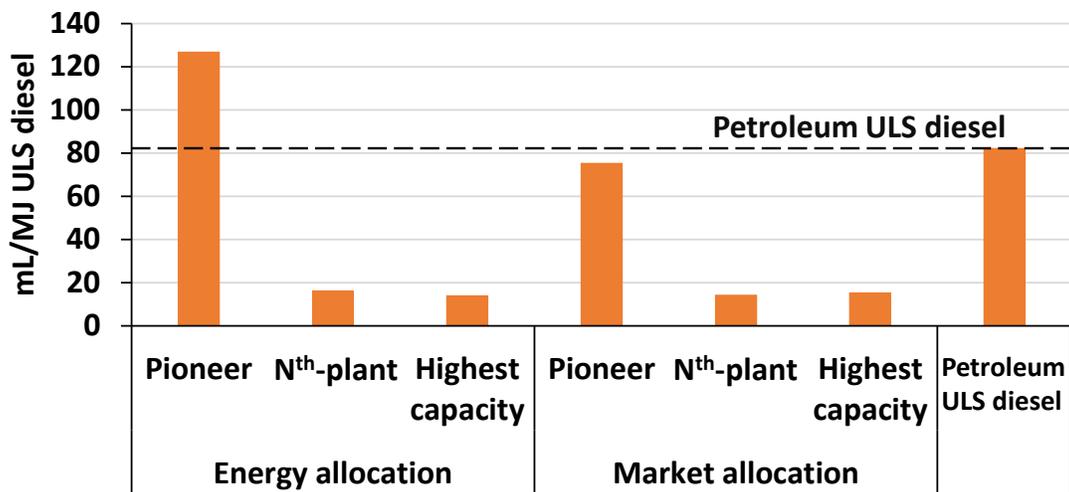
This figure represents the results for the energy allocation case. Carbon efficiency is the percentage of carbon from PUP incorporated in products and coproducts. Fossil energy is the total energy consumption from fossil-based fuels in the pretreatment, conversion, and separation/upgrading processes. Net fossil energy requirements are the fossil energy consumption after using heat from internally produced fuel gas.

### 3.2.3 Water Consumption

Water consumption is the volume of water used in the process per megajoule of product that cannot be recycled. Figure 14 and Figure 15 compare water consumption for pyrolysis oil and ULS diesel, respectively, between the plant capacity groups. Pioneer plants have the highest water consumption of all the facilities, an estimated 85.8 and 126.9 mL/MJ for pyrolysis oil and ULS diesel, respectively. This difference occurs because pioneer plants use more electricity per megajoule of product compared to  $N^{\text{th}}$ -plants and the highest-capacity plant. The amount of make-up process water in pioneer plants is 5 and 3 times larger compared to the  $N^{\text{th}}$ -plant and highest-capacity facilities, respectively, which suggests that conversion at pyrolysis facilities with higher capacity reduces water consumption. The water consumption of pioneer plants is 6% higher than the crude naphtha and 54% higher than petroleum-based ULS diesel assuming the energy allocation.



**Figure 14. Water Consumption to Produce Pyrolysis Oil Using Different Allocation Methods.** Water consumption of crude naphtha is 75.8 mL/MJ (Argonne, 2020).



**Figure 15. Water Consumption to Produce ULS Diesel Using Different Allocation Methods** Water consumption of petroleum-based ULS diesel is 82.3 mL/MJ. (Argonne, 2020).

The water consumption for PUP-derived ULS diesel production shows a higher value than water consumption for pyrolysis oil production because the yield of ULS diesel is between 35% and 48% lower than that of pyrolysis oil, which increases the water consumption per megajoule. Additional water is also required in the separation/upgrading process, which is only included in ULS diesel production. This additional volume of water is estimated to be between 4 and 26 mL/MJ ULS diesel for *N<sup>th</sup>*-plants and pioneer facilities, respectively. The water consumption per megajoule of ULS diesel is 80% and 83% lower than petroleum-based ULS diesel for *N<sup>th</sup>*-plants and the highest-capacity plant, respectively, and 87% and 89% lower when compared to the water consumption in the pioneer plants for the energy allocation.

For market allocation, this reduction of water consumption per megajoule of ULS diesel in  $N^{\text{th}}$ -plants and highest capacity plants, respectively, is 82% and 81% compared to petroleum-based ULS diesel and 81% and 79% compared to pioneer plants. This higher water consumption in pioneer plants is related to the reported consumption of process water from the survey (see Table A.3-4 in the appendix). Pioneer plants consume almost 5 times more process water than  $N^{\text{th}}$ -plants and twice the amount of the highest capacity plant. Comparing water consumptions from energy and market allocation showed no more than a 10% difference (for energy allocation in pioneer plants).

### 3.3 SENSITIVITY ANALYSIS

The sensitivity analysis evaluates the effects of certain process parameters in the LCA results compared to a base case. In this report, the base case takes into account the use of energy allocation for coproduct treatment, the production of heat from fuel gas generated from conversion, and the supply of electricity from of the average U.S. grid mix. Because some companies reported using chemicals during certain stages of the processes (see Table 8), we analyzed how including these materials would influence the fossil energy consumption and GHG emissions of pyrolysis oil and ULS diesel produced from PUP. Because these materials are specific to certain capacity groups and process stages, the burdens of materials were allocated only to those products and capacity groups that incorporate them. For example, chemicals that are only used for separation/upgrading (i.e., hydrogen) were only counted in the analysis of ULS diesel.

The companies reported that electricity was one of the energy sources they used to provide power to operate equipment. Because the electricity grid mix varies throughout the country, the location of a pyrolysis facility can determine the environmental performance of its conversion process if the facility has a high electricity demand. Therefore, a second sensitivity case analyzed the effects of three electricity grid mixes: one with a high contribution of renewable energy (California [CA]), one with a reduced supply of renewable energy sources (Florida [FRCC]), and one with high GHG emissions (Midwest [MRO]). Details about these electricity mixes appear in Argonne (2020).

A third sensitivity case analyzed the use of renewable materials to generate heat by substituting renewable natural gas for natural gas. However, because the heat requirements in all plant capacity groups are fulfilled using fuel gas (see Figure 6), changes in the external supply of natural gas did not produce effects in the sensitivity analysis.

The results of the sensitivity analysis for pyrolysis oil and ULS diesel are presented in Figure 16 and Figure 17, respectively. Results from the third sensitivity cases are not shown in these figures because they did not show differences compared to the base case. The use of material inputs, as shown in Table 8, increased the fossil energy consumption of pyrolysis oil and ULS diesel compared to the base case by 11% and 26%, respectively, in the pioneer plants (see Figure 18A).

Sodium hydroxide and calcium oxide were major contributors to the amount of fossil energy consumed by the pioneer plants. Sodium hydroxide is very energy-intensive to produce (energy consumption of 28 MJ/kg). Calcium oxide, although less energy intensive than sodium

hydroxide (5 MJ/kg), still requires more energy in the conversion process (4.2 g/MJ of pyrolysis oil) compared to the other material inputs.

In the  $N^{\text{th}}$ -plants, fossil energy consumption is 3% higher than in the base case only for the ULS diesel (see Figure 18C), because the material inputs are only used in separation and/or upgrading. Most of this energy comes from the use of methyl ethyl ketone and toluene. The analysis of GHG emissions shows values 26% and 8% higher for pyrolysis oil and diesel, respectively, compared to the base case in the pioneer plants. This increase in GHG emissions is mostly induced by the use of calcium oxide.

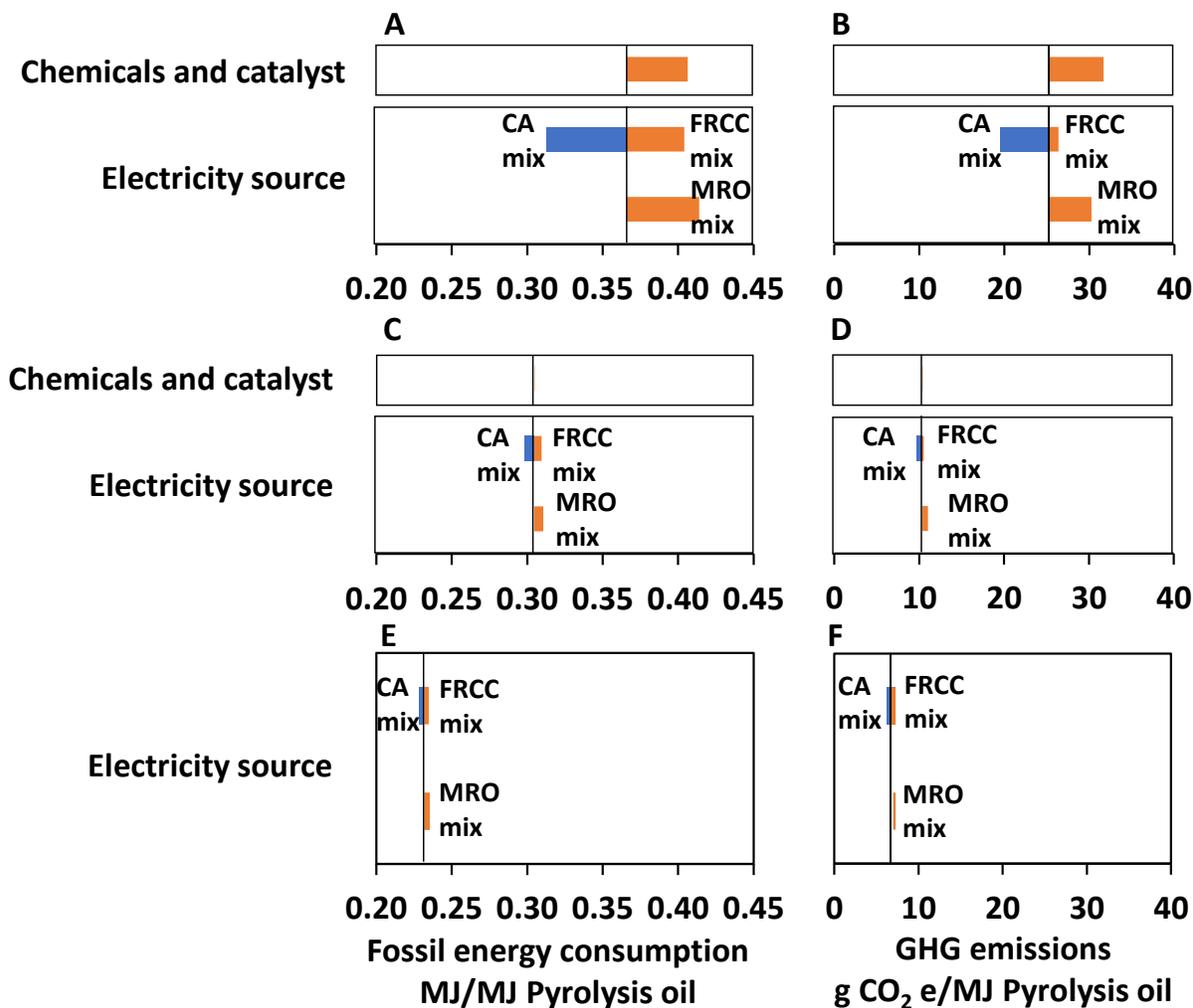
Using a more renewable-dependent electricity grid, such as the CA grid mix, reduced the fossil energy consumption for pyrolysis oil and ULS diesel, respectively, by 15% and 34% for pioneer plants (Figures 18A and 19A), 2% and 18% in  $N^{\text{th}}$ -plants (Figures 18C and 19C), and 2% and 25% in the highest-capacity plant (Figures 18E and 19E), all compared to the base case. We expect that changes to the grid mix affect the performance of the pioneer plants more because they consume more fossil energy per megajoule of product (see Figure 10 and Figure 12) and a higher share of electricity demand (Table 6) than  $N^{\text{th}}$ -plants and the highest-capacity plant.

The GHG emissions decrease by 22% and 7% for pyrolysis oil and ULS diesel, respectively (Figures 18B and 19B) for pioneer plants, 7% and 1% for  $N^{\text{th}}$ -plants (Figures 18D and 19D), and 6% and 1% for the highest-capacity facilities (Figures 18F and 19F). The reduction of GHG emissions in pyrolysis oil was more prominent than in ULS diesel, because in the latter more than 68% of the emissions are derived from vehicle operation. The CA grid mix generates 40% less GHG emissions per kilowatt-hour of electricity compared to the U.S. average grid mix, because 47% of its energy sources are renewable (Table A.5-1 in the appendix).

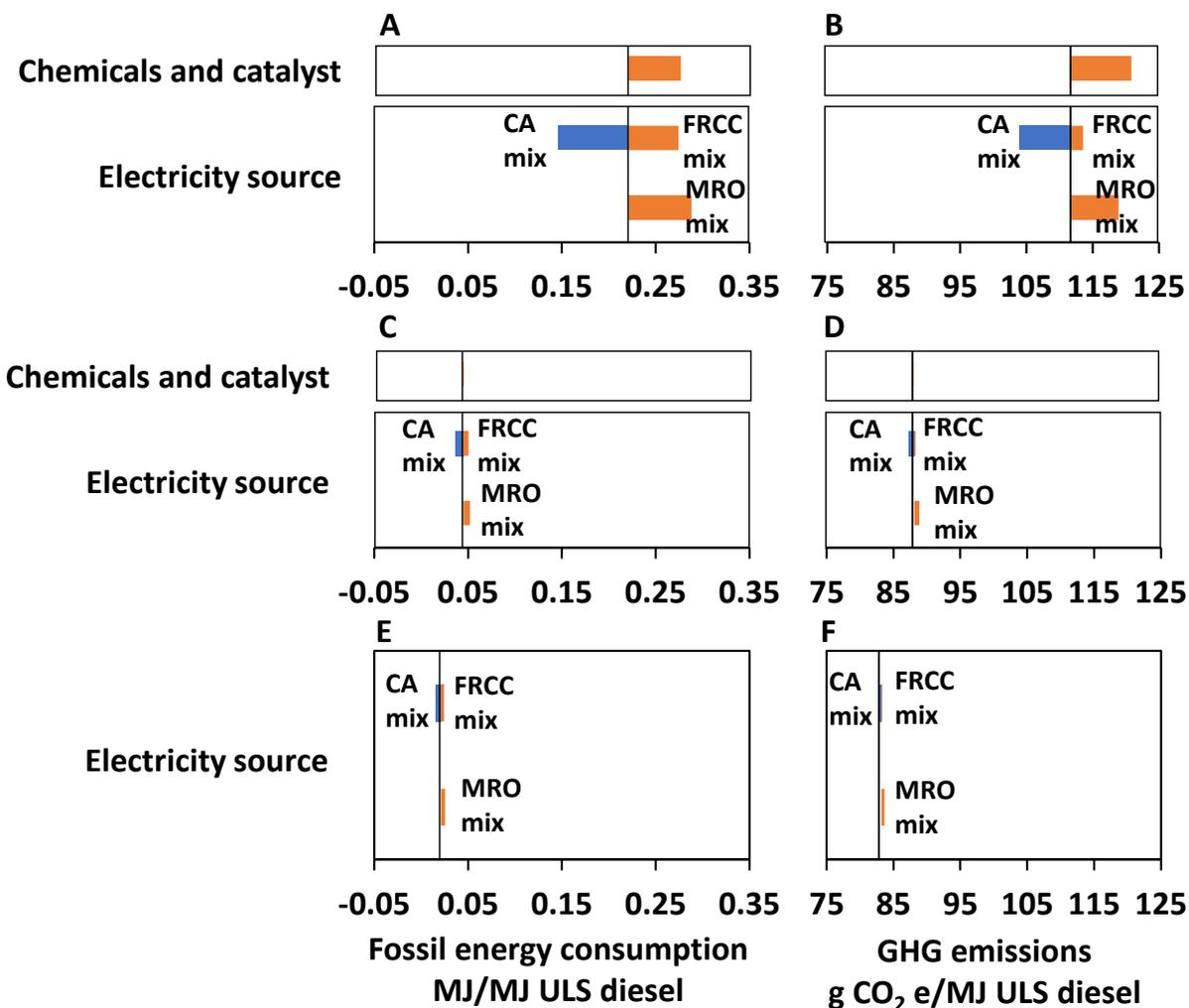
Shifting the electricity grid mix to FRCC increases the fossil energy consumption of pioneer plants by 11% and 25% for pyrolysis oil and ULS diesel, respectively, compared to the base case. GHG emissions of pioneer plants increased by 5% and 2% for pyrolysis oil and ULS diesel, respectively, also compared to the base case. The change is small because the GHG emissions from the U.S. grid mix and the FRCC grid mix are very similar (449 versus 501 g CO<sub>2e</sub>/kWh, respectively).

In pioneer plants, fossil energy consumption using the MRO grid mix increases by up to 13% and 30% for pyrolysis oil and ULS diesel, respectively, while GHG emissions increase by up to 20% and 6% in pyrolysis oil and ULS diesel, respectively. A high percentage of coal (43%) in the energy mix is responsible for the overall increase of fossil energy consumption and GHG emissions derived from the use of the MRO grid mix.

Comparing the sensitivity analysis with the GHG emissions of crude naphtha and petroleum-derived ULS diesel (14.8 and 91.1 g CO<sub>2e</sub>/MJ, respectively) reveals that using the electricity grid mix with a higher percentage of renewable sources does not reduce the GHG emissions of pioneer plants below the values for petroleum-based products. However, the use of a grid mix with almost 50% more GHG emissions than the U.S. grid did not increase the GHG emissions of  $N^{\text{th}}$ -plants and the highest-capacity plant over the threshold of petroleum-based products.



**Figure 16. Sensitivity Analysis of the Production of Pyrolysis Oil.** (A) Fossil energy consumption of pioneer plants; (B) GHG emissions of pioneer plants; (C) fossil energy consumption of  $N^{\text{th}}$ -plants; (D) GHG emissions of  $N^{\text{th}}$ -plants; (E) fossil energy consumption of the highest-capacity plant; and (F) GHG emissions of the highest-capacity plant (energy allocation is the coproduct treatment method).

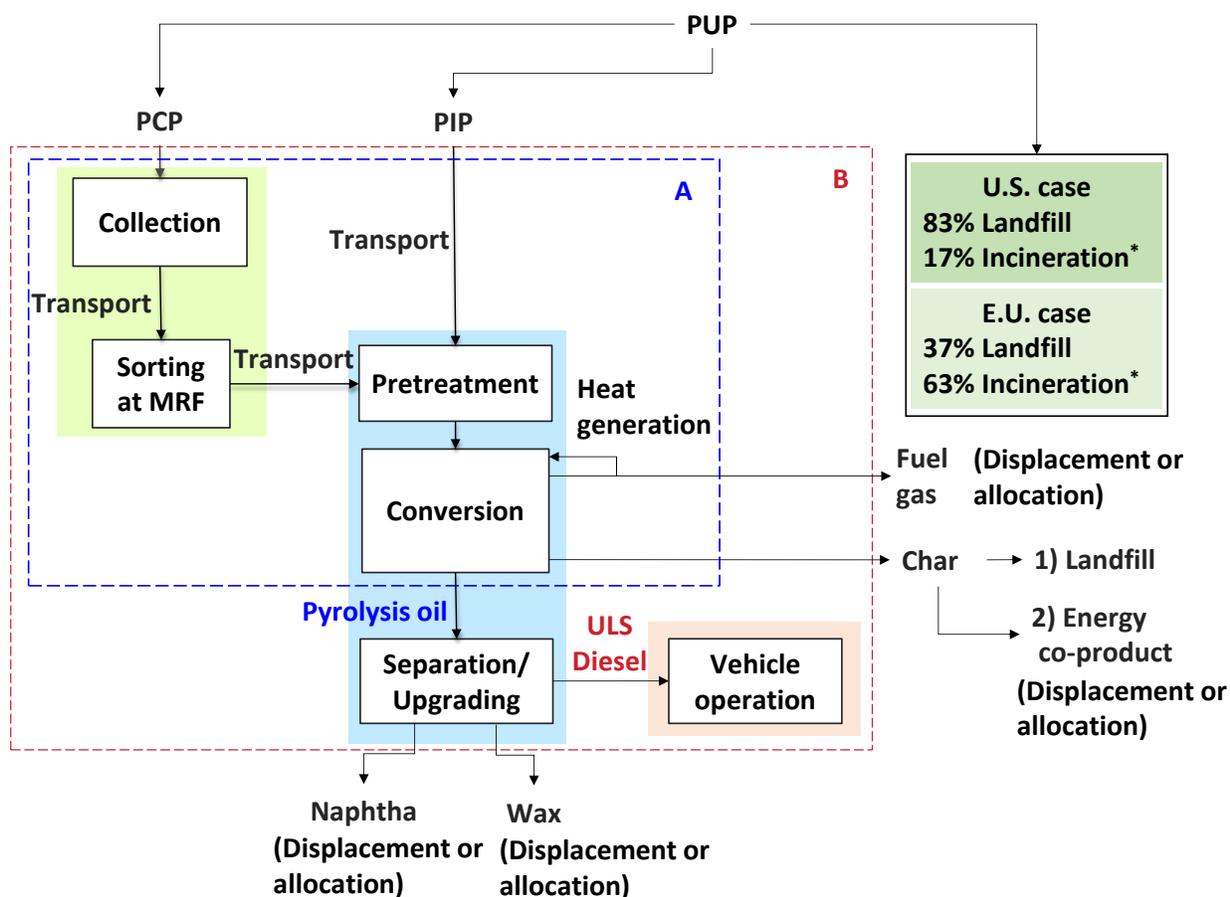


**Figure 17. Sensitivity Analysis of the Production of ULS Diesel.** (A) Fossil energy consumption of pioneer plants; (B) GHG emissions of pioneer plants; (C) fossil energy consumption of N<sup>th</sup>-plants; (D) GHG emissions of N<sup>th</sup>-plants; (E) fossil energy consumption of the highest-capacity plant; and (F) GHG emissions of the highest-capacity plant. Energy allocation is the coproduct treatment method.

### 3.4 COUNTERFACTUAL SCENARIO

So far, in the base analysis we discussed the case where PUP is used to produce two main products: one that can be used as a chemical precursor such as pyrolysis oil, and another that can be used as transportation fuel such as ULS diesel. However, we did not include the impact of diverting the PUP from traditional waste management treatment. Traditional waste management generates emissions, mainly from the CO<sub>2</sub> formed during combustion of PUP used for electricity generation. These emissions are avoided when PUP is diverted to produce chemicals and fuels. Hence, a credit based on these avoided emissions could be included in the estimation of GHG emissions to fully capture the impacts brought by pyrolysis conversion.

According to the EPA (2020), 76% of the plastic waste in the United States in 2018 was landfilled, while 16% was incinerated with energy recovery. Considering only the major resins that were part of feedstock mix of the pyrolysis facilities (i.e., HDPE, LDPE/LLDE, PS and PP, [Figure 2]) and normalizing for the fraction (91%) that was not recycled, the amount of PUP in waste plastic that we assumed would otherwise be landfilled corresponded to about 83%. We assumed that the remaining 17% was incinerated with energy recovery. In other geographical regions like the E.U. the situation is different; only 25% of plastic waste is landfilled, and 43% is incinerated for energy recovery (European Parliament, 2018). After normalization, in the European case 63% of PUP is incinerated while 37% is landfilled. Therefore, in this section we discuss the impact of the avoided emissions from the traditional waste management treatment of PUP. Figure 18 includes traditional waste management of PUP in the United States and the E.U. in the system boundary presented in Figure 1.



**Figure 18. System Boundary of PUP Conversion, Including the Counterfactual Scenarios for PUP Processing in the United States and the E.U.: (A) Pyrolysis Oil Production, and (B) ULS Diesel Production.** Incineration with energy recovery. PCP: post-consumer plastic; PIP: post-industrial plastic; ULS: ultra-low sulfur; MRF: material recovery facility; U.S.: United States; E.U.: European Union.

Electricity can be produced from incineration, which could displace grid electricity. Based on this, we estimated the amount of carbon produced from incineration and subtracted the emissions from the electricity generated, assuming it replaces U.S. average grid electricity. To estimate the

carbon that was combusted to produce energy and the CO<sub>2</sub> emitted, we relied on data provided by the companies, such as the average carbon content of PUP (86%) and the LHV of PUP (40 MJ/kg). We assumed that the efficiency of electricity production was 25% (Argonne, 2020), and used this number to estimate the amount of energy generated in incineration. GHG credits were calculated based on the U.S. grid mix, as shown in Table A.5-1 of the appendix. Transportation of the PUP to landfill provided by the companies was also included in the analysis (Table 5). We subtracted the electricity generation credits from the incineration emissions to calculate the emissions avoided by diverting PUP from end-of-life management to the production of pyrolysis oil and ULS diesel.

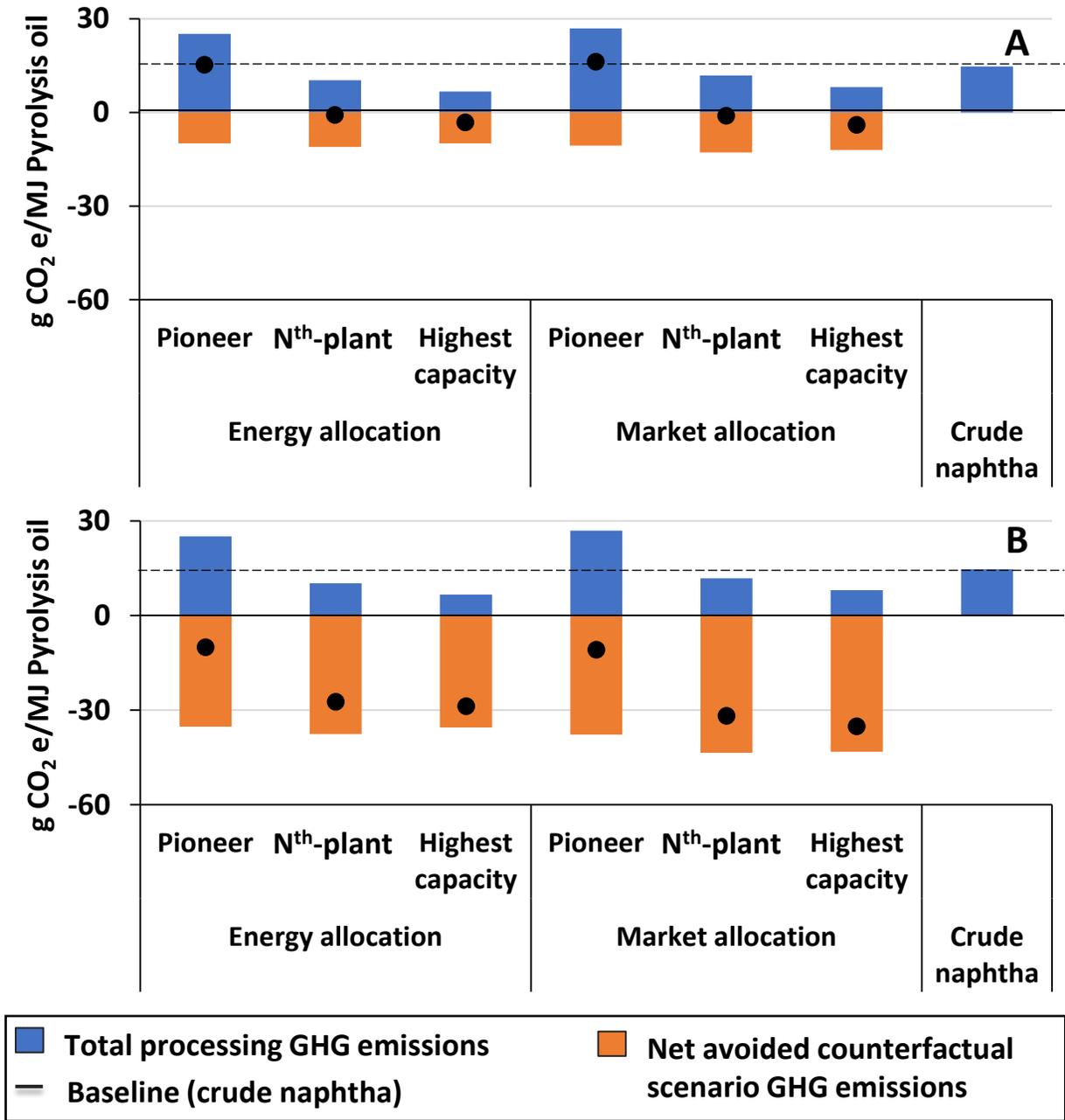
The GHG emission results including the counterfactual scenario are shown in Figure 19 and Figure 20 for the energy and market allocations. Figure 19 compares the results to naphtha GHG emissions as the baseline, while Figure 20 compares them to petroleum ULS diesel.

No major differences were found between using energy and market allocation. The U.S. counterfactual scenario for pyrolysis oil showed that pioneer plants can achieve the same GHG emissions as naphtha, while the highest-capacity plant had the lowest GHG emissions reduction compared to other facilities, including the naphtha baseline. For the E.U. case, the GHG emissions were negative for all facilities due to an increase in incineration credits.

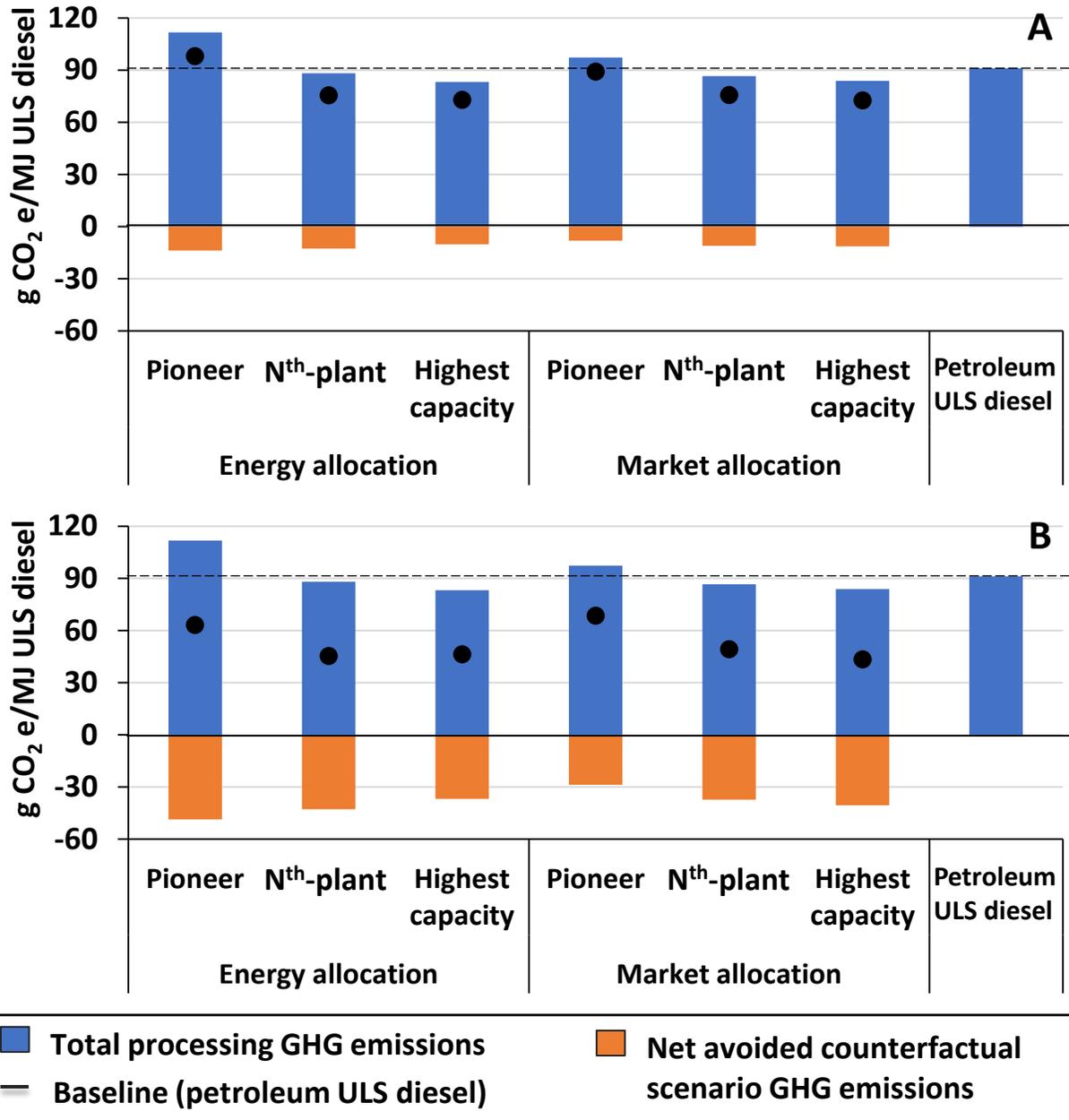
The GHG emissions of pyrolysis oil from  $N^{\text{th}}$ -plants and highest-capacity plants are already lower compared to crude naphtha (see Figure 7). Therefore, subtracting the credits of the counterfactual cases generated negative emissions in the energy and market allocations. In the analysis of ULS diesel, the highest-capacity plant offered the greatest reduction benefits: 20% and 49% less GHG emissions compared to petroleum-derived ULS diesel for the U.S. case and the E.U. case, respectively, when energy allocation is used.

The counterfactual scenario results for pioneer plants showed that GHG emissions of PUP-derived ULS diesel were similar to the petroleum ULS diesel in the U.S. case and 31% lower compared to petroleum ULS diesel in the E.U. case, also for the analysis using energy allocation. The results from including the counterfactual scenario showed that the GHG emissions of PUP-derived pyrolysis oil and ULS diesel in all the plant capacity groups are lower than or very close to the baseline.

The counterfactual scenario results are driven only by the fraction of PUP incinerated for power generation. The GHG emissions for PUP landfilled were negligible because we assumed that carbon in PUP does not degrade. The scenarios in the E.U. counterfactual case generated less GHG emissions than the U.S. counterfactual case because there are more credits (due to increased incineration emissions) in the E.U. counterfactual scenario compared to the U.S. counterfactual case. However, the percentage of PUP sent to incineration in the United States is not expected to be similar to the E.U. percentage in the future. This is because the EPA's goal is to increase the recycling rate of materials and reduce traditional waste management (EPA, 2020). Therefore, the use of pyrolysis technology can shift the fate of PUP from incineration or disposed in landfills to be used as a feedstock for intermediate chemicals and fuels production.



**Figure 19. Counterfactual Analysis for Pyrolysis Oil.** (A) GHG emissions for the U.S. case and (B) GHG emissions for the E.U. case. The points on the graph indicate net GHG emissions after subtracting the avoided emissions from traditional waste management.



## 4 CONCLUSIONS

In this study, we quantified the life-cycle environmental benefits of pyrolysis-based products (pyrolysis oil and ULS fuel) from PUP plastic in terms of GHG emissions, fossil energy consumption, water consumption, and tradeoffs. This analysis was conducted for three types of facility: pioneer plants,  $N^{\text{th}}$ -plants, and the highest-capacity plant. The results were compared to their conventional petroleum-based counterparts, that is, crude naphtha and ULS diesel fuel. Multiple allocation methods were included in the analysis (displacement, energy, and market).

Table 9 and 10 summarize the fossil energy consumption and GHG emissions results for PUP-derived pyrolysis oil and PUP-derived ULS diesel, respectively. Greater reductions were achieved in the displacement method due to the credit received when allocating coproducts (e.g., fuel gas, char, naphtha, and wax); however, because the potential of large coproduct effects can create a risk of generating distorted results, we focused the discussion of our results on the energy- and market-allocation methods.

Pioneer plants showed higher GHG emissions, fossil energy, and water consumption compared to the baselines (naphtha and petroleum ULD diesel), and the additional facility types ( $N^{\text{th}}$ -plants and the highest capacity).  $N^{\text{th}}$ -plants and the highest-capacity facility, on the other hand, showed greater benefits (e.g., lower GHG, fossil energy and water consumption) compared to the baselines. For instance, pyrolysis oil obtained from  $N^{\text{th}}$ -plants could reduce GHG emissions, fossil energy, and water consumption by 19–29%, 69–73%, and 81–83% when market or energy allocation was used, respectively. Reduction benefits still remained when the system boundary was expanded to PUP-derived ULS diesel. In this case, GHG emissions, fossil energy, and water use of  $N^{\text{th}}$ -plants decreased by 3–5%, 96–97%, and 80–82%, respectively, with energy or market allocation compared to their fossil base counterpart.

**Table 9. GHG Emissions and Fossil Energy and Water Consumption to Produce 1 MJ of Pyrolysis Oil, via Pyrolysis of PUP, and Production of 1 MJ of Crude Naphtha (percentage of difference from baseline)**

Impact evaluated	Allocation method	Pyrolysis oil			Crude naphtha
		Pioneer	$N^{\text{th}}$ -plant	Highest capacity	
GHG emissions (g CO <sub>2</sub> e/MJ)	Displacement	26.3(79%)	10.2 (-31%)	5.5 (-63%)	—
	Energy-based	25.1 (71%)	10.4 (-29%)	6.7 (-55%)	14.7
	Market-based	26.9 (83%)	11.9 (-19%)	8.0 (-45%)	—
Fossil energy consumption (MJ/MJ)	Displacement	0.39 (-66%)	0.34 (-70%)	0.27 (-77%)	—
	Energy-based	0.37 (-68%)	0.30 (-73%)	0.23 (-80%)	1.15
	Market-based	0.39 (-66%)	0.35 (-69%)	0.28 (-75%)	—
Water consumption (mL/MJ)	Displacement	86.0 (13%)	12.8 (-83%)	12.5 (-84%)	—
	Energy-based	80.1 (6%)	12.7 (-83%)	12.3 (-84%)	75.8
	Market-based	85.8 (13%)	14.6 (-81%)	14.9 (-80%)	—

**Table 10. GHG Emissions and Fossil Energy and Water Consumption to Produce 1 MJ of ULS Diesel, via Pyrolysis of PUP, and Production of 1 MJ of Petroleum ULS Diesel (percentage of difference from baseline)**

Impact evaluated	Allocation method	PUP-derived ULS diesel			Petroleum ULS diesel
		Pioneer	N <sup>th</sup> -plant	Highest capacity	
GHG emissions (g CO <sub>2e</sub> /MJ)	Displacement	118.5 (30%)	82.4 (-10%)	65.6 (-28%)	—
	Energy-based	111.8 (23%)	88.1 (-3%)	83.2 (-9%)	91.1
	Market-based	97.3 (7%)	86.6 (-5%)	83.8 (-8%)	—
Fossil energy consumption (MJ/MJ)	Displacement	-1.00 (-185%)	-1.76 (-249%)	-2.82 (-339%)	—
	Energy-based	0.22 (-81%)	0.04 (-96%)	0.02 (-98%)	1.18
	Market-based	0.13 (-89%)	0.04 (-97%)	0.02 (-98%)	—
Water consumption (mL/MJ)	Displacement	123.9 (51%)	-60.4 (-173%)	-97.0 (-218%)	—
	Energy-based	126.9 (54%)	16.5 (-80%)	14.2 (-83%)	82.3
	Market-based	75.4 (-8%)	14.5 (-82%)	15.6 (-81%)	—

For all scenarios studied in this analysis, electricity was the major driver of the fossil energy and GHG emissions results. Sensitivity analyses of the electricity grid showed that GHG emissions per megajoule of pyrolysis oil can be reduced by 22% and 7%, for pioneer and N<sup>th</sup>-plants, respectively, when the electricity mix changed from a fossil-based mix to a more renewable mix, like the grid of the state of California. In the case of GHG emissions per megajoule of ULS diesel, we estimated that the reductions would be 8% and 1% for pioneer and N<sup>th</sup>-plants, respectively. When additional material and catalyst were incorporated in the analyses, pioneer plants showed an increase of 8% in the GHG emissions per megajoule of ULS diesel compared to the base analysis when these materials and catalysts are not required. No significant effects on the GHG emission were observed in the analysis of N<sup>th</sup>-plants and the highest-capacity facility.

The higher GHG emissions and fossil energy consumption of pioneer plants was mostly due to their higher net energy requirements on a per unit of product basis, and an increased share of electricity compared to N<sup>th</sup>-plants and the highest-capacity facility. Currently, pyrolysis facilities do not use coproducts to produce electricity, therefore it is obtained from the grid. Most of the water consumed during PUP conversion to either pyrolysis oil or ULS diesel occurs in the conversion stage. The amount of water used was considerably higher in pioneer plants compared to N<sup>th</sup>-plants and the highest-capacity facility. In addition, out of the three plant capacity groups pioneer plants presented the lowest carbon efficiency, because they needed to use more internally-produced fuel gas to supply energy for the conversion process. The results obtained here suggest that the pyrolysis technology can obtain environmental benefits from economies of scale; the increased processing capacity of the pyrolysis plants reduced the environmental impacts associated with the production of pyrolysis products (pyrolysis oil and ULS diesel) compared to petroleum-based counterparts (crude naphtha and petroleum ULS diesel).

**Table 11. GHG Emissions to Produce 1 MJ of Pyrolysis Oil via PUP Pyrolysis (sensitivity cases)**

Plant capacity group	CO <sub>2</sub> e/MJ pyrolysis oil				
	Base analysis	Materials inputs	CA grid	FFRC grid	MRO grid
Pioneer	25.1	31.7	19.5	26.4	30.2
<i>N</i> <sup>th</sup> -plant	10.4	10.4	9.7	10.6	11.0
Highest capacity	6.7	6.7	6.3	6.7	7.0

**Table 12. GHG Emissions to Produce 1 MJ of ULS Diesel via PUP Pyrolysis (sensitivity cases)**

Plant capacity group	g CO <sub>2</sub> e/MJ ULS diesel				
	Base analysis	Materials inputs	CA grid	FFRC grid	MRO grid
Pioneer	111.8	120.8	104.0	113.5	118.9
<i>N</i> <sup>th</sup> -plant	88.1	88.1	87.3	88.3	88.9
Highest capacity	83.2	83.2	82.6	83.3	83.7

When we included the credits from the avoided emissions of traditional waste management, GHG emissions of PUP-derived pyrolysis oil and PUP-derived ULS diesel from *N*<sup>th</sup>-plants were reduced by 105% and 17% compared to the crude oil-based naphtha and ULS diesel, respectively, in the U.S. case. The GHG emissions of pioneer plants also showed values close to petroleum-based counterparts in the U.S. case and lower than the petroleum-based counterparts in the E.U. case. Although the results for the E.U. case further reduce the GHG emissions, the percentage of PUP managed through incineration is not expected to increase in the United States.

**Table 13. GHG Emissions to Produce 1 MJ of Pyrolysis Oil by Converting PUP Pyrolysis in the Counterfactual Scenarios (percentage of difference from crude naphtha)**

Plant capacity group	Counterfactual United States			Counterfactual European Union		
	Displacement	Energy allocation	Market allocation	Displacement	Energy allocation	Market allocation
Pioneer	15.5 (5%)	15.2 (3%)	16.2 (10%)	-11.9 (-181%)	-10.1 (-168%)	-10.9 (-174%)
N <sup>th</sup> -plant	-3.1 (-121%)	-0.7 (-105%)	-0.9 (-106%)	-34.8 (-336%)	-27.2 (-284%)	-31.7 (-315%)
Highest capacity	-7.1 (-148%)	-3.2 (-122%)	-4.0 (-127%)	-39.6 (-369%)	-28.8 (-296%)	-35.1 (-338%)

**Table 14. GHG Emissions to Produce 1 MJ of ULS Diesel by Converting PUP Pyrolysis in the Counterfactual Scenarios (percentage of difference from petroleum ULS diesel)**

Plant capacity group	Counterfactual United States			Counterfactual European Union		
	Displacement	Energy allocation	Market allocation	Displacement	Energy allocation	Market allocation
Pioneer	96.2 (6%)	98.1 (8%)	89.2 (-2%)	39.5 (-57%)	63.3 (-31%)	68.6 (-25%)
N <sup>th</sup> -plant	52.7 (-42%)	75.5 (-17%)	75.6 (-17%)	-18.1 (-120%)	45.4 (-50%)	49.3 (-46%)
Highest capacity	30.5 (-66%)	72.9 (-20%)	72.5 (-20%)	-60.0 (-166%)	46.3 (-49%)	43.4 (-52%)

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## APPENDIX

### A.1 SURVEY DEVELOPMENT

Table A.1-1 presents the list of questions that were incorporated in the survey to collect data for the analysis presented in this report. As previously described, the survey was divided into four main sections: (1) about the company and plant, (2) feedstock, (3) conversion and separation, and (4) emissions. Each company had the opportunity to go through all the questions presented here and provide details about their technology. The data from this survey was aggregated as explained in the report. No individual data was presented in this report.

**Table A.1-1. Survey Development for Collecting Data of Conversion of Waste Plastic Technologies**

Section of the Survey	Category	Question
<b>1. About the company and plant</b>	<b>1.1 Capacity</b>	1.1a What is the design capacity of the process? (Please provide the amount in metric tons of plastic per month and per year or specify if other units are used.)
		1.1b Please provide the number of hours or days of operation per year.
	<b>1.2 Stage of technology</b>	1.2 What is the technology stage of development for the data shared in this survey (e.g., bench, pilot, demonstration, commercial)? Note: for clarification, please look at <b>Definitions</b> tabs: State of technology.
	<b>1.3 General</b>	1.3a Years of operation.
		1.3b Location of the facility where the data come from.
		1.3.c Please provide some information about your electricity grid supply. For example, do you rely on renewable energy suppliers, or do you rely on your regional electricity grid? We will take this information into account for sensitivity analysis; however, we will use a U.S. electricity grid mix for the analysis. Note: for clarification, please look <b>Definition</b> tab: U.S. Electricity Mix.
<b>2. Feedstock</b>	<b>2.1 Logistics</b>	2.1a Does the feedstock come from a local source?
		2.1b Please provide the transportation method(s) (e.g., truck, rail) and distance from the feedstock source to the conversion facility. Please provide the information on an average per year.
		2.1c What is the fuel type (e.g., diesel) and capacity of transportation mode?
		2.1d What is the moisture content (%) of the aggregated feedstock that enters the collection site?
	<b>2.2 Market</b>	2.2 Do market conditions significantly influence the feedstock type you use? Please explain if possible.
	<b>2.3. Feedstock Type</b>	2.3a What is the source of the waste plastic? Please provide the relative mass share of each waste material (example: industrial plastic waste 20%, packaging 80%).
		2.3b What are the types of plastic/resin waste used in the process? Please provide the relative mass share of each waste material on an annual average basis (example: LLDPE 20%, PP 80%).
		2.3c Does the variability in feedstock composition affect the process conditions? (For example, having more PP vs. PE vs. PS will change reaction temperatures or increase the aromatic content in the product).

**Table A.1-1. (Cont.)**

Section of the Survey	Category	Question
	2.4. Quality	2.4a What kind of plastic waste is not appropriate/acceptable for the process (example: PVC, chlorinated polyethylene, or CPE)? What other resins are removed?
		2.4b Is there a maximum contamination level? If yes, please explain and list contaminants of concern.
		2.4c What do you do with the discarded plastic from your feedstock? For example: sell, send back to reclaimers, send to landfill.
	2.5. Properties and composition ( <u>before entering pretreatment</u> )	2.5a What is the density of the aggregated feedstock? Please provide in units of mass per volume.
		2.5b What is the moisture content (%) of the feed stream into the pretreatment process?
		2.5c What is the C, H, and O content of the aggregated feedstock (in % by dry weight)?
		2.5d What is the aggregated feedstock's lower heating value (e.g., energy per mass)?
	2.6. Consumption	2.6 What is the aggregated feedstock feed rate to the pretreatment process? Year average data is preferred.
	2.7. Pretreatment	2.7a What type of pretreatment is required (e.g., additional sorting, separation, size reduction)?
		2.7b Does the pretreatment use any chemicals? If so, please specify the type and consumption rate.
		2.7c What type of fuel is used in the pretreatment process? Please indicate the consumption rate in units per day.
	3. conversion and separation	3.1 Properties and composition ( <u>before entering conversion stage</u> )
3.1b What is the moisture content (%) of the feed stream into the conversion process?		
3.1c What is the C, H, and O content of the aggregated feedstock (in % by dry weight)?		
3.1d What is the aggregated feedstock's lower heating value (e.g., energy per mass)?		
3.2. Conversion		3.2a What is the technology used for conversion (e.g., pyrolysis, gasification, depolymerization)? If you use pyrolysis, please provide information about type of pyrolysis reaction (thermal, catalytic, microwave, plasma, hydrothermal, etc.).
		3.2b What is the feed rate of plastic into the conversion process? Please provide it in metric tons.
3.3. Conversion inputs		3.3a What amount of each type of process fuel is consumed? In what type of equipment is it consumed? <b>This question refers to the conversion process only.</b>
		3.3b Is any fuel produced within the conversion process and used to supply internal energy? If yes, please specify the fuel type and amount.
		3.3c If a catalyst is used in the process to convert the feedstock to an oil, please provide the catalyst type and net consumption rate.
		3.3d Does the conversion process use any other chemicals (i.e., hydrogen)? If so, specify the chemicals and their consumption rates.
		3.3e Does the conversion process consume water? If yes, what is the net consumed amount per one gallon of oil produced? Note: This question refers to the amount of makeup water.
		3.3f Is any portion of the process conducted in a vacuum?

**Table A.1-1. (Cont.)**

Section of the Survey	Category	Question
		3.3g If the answer to 3.2f is yes, how much energy is involved in the process? Please provide units.
		3.3h Do you upgrade the intermediate oil onsite? If yes, please move to section 3.4. If no, continue with question 3.3i.
		3.3i What is the production rate of the intermediate oil? Please provide it in metric tons.
		3.3j What are the properties of the intermediate oil?
		3.3k What is the composition of the intermediate oil? Please provide the results in weight percentage.
		3.3l Please provide the transportation method(s) and distance from your facility to the site where the intermediate oil is used. Please provide the information on an average per year.
		3.3m What is the fuel type (e.g., diesel) and capacity of transportation mode for the intermediate oil?
		3.3n What are other products that you obtain from the conversion and their applications?
		After completing this section, please move to section 4.
		3.4 Separation/Upgrading
	3.4b Is any fuel produced internally and used to supply internal energy?	
	3.4c If a catalyst is used in the upgrading process, please provide the catalyst type and net consumption rate.	
	3.4d Does the separation/upgrading process consume water? If yes, what is the net consumed amount per one gallon of oil produced? This question refers to the amount of makeup water.	
	3.4e Do you use H <sub>2</sub> to upgrade the intermediate product? How much?	
	3.4f If the answer to 3.4e is yes, describe where the H <sub>2</sub> comes from (e.g., purchased directly from a supplier, steam reforming of methane).	
	3.4g If H <sub>2</sub> is purchased, please provide the method of transportation and distance to your facility.	
	3.4h Does the process consume any other chemicals? If so, specify which chemicals and at what rates they are consumed.	
	3.5 Residues and coproducts after separation/upgrading	3.5a What are the residues and coproducts? What is the fate of each residue or coproduct (e.g., disposal, further treatment by third party, sold directly to consumer)?
	3.5b If residues/coproducts are managed outside the facility, please provide the transportation method and distances to facilities. Please specify for which residue the data is provided.	
	3.6 Final output after separation/upgrading	3.6 What is the <b>final output product or products</b> after upgrading? Please provide amounts, and final application. For clarification, please look at <b>Definitions</b> tab: petroleum products.
	3.7 Properties of products	3.7 Please describe the properties of the final product(s) ( <b>this question includes main products (diesel gasoline, naphtha) and coproducts such as char, wax, syngas, fuel gas, etc.</b> ): density, C-H-O S content (wt.%), lower heating value (e.g., energy per mass). If several intermediate products are produced, please provide properties for each of them. Please add more rows if needed.

**Table A.1-1. (Cont.)**

Section of the Survey	Category	Question
4. Process air emissions	4.1 Emissions	4.1a Please provide the type and number of emissions (fugitive or non-combustion). For example, a pyrolysis process could emit volatile organic compounds (VOCs) and pre-treatment processes could emit particulate matter (PM). We will use our model to calculate air emissions from combustion of process fuels.
		4.1b Other emissions considered as hazardous air pollutants.
		4.1c If the company has a facility permitting (e.g., EPA title V), would you share that with us?

We defined four stages of technology according to (Ocean Recovery Alliance, 2015; Closed Loop Partners, 2019). These definitions use the scale of development that the company has proven to be feasible for its conversion process.

**Table A.1-2. Stage of Development Definitions**

Stage of Technology	Definition
Bench scale	Laboratory scale: basic infrastructure to test proof of concept.
Pilot scale	Small-scale model of commercial offering with the goal of demonstrating proof of concept, testing different feedstocks, and evaluating oils yields and product quality. Pilot-scale systems can be used to test design modifications for other preprocessing and back-end processing configurations.
Demonstration scale	Full commercial-scale system with the goal of demonstrating proof of concept, testing different feedstocks, evaluating oil, yields, and product quality at commercial scale. Demonstration scale plastic-to-fuel systems can be converted into commercial systems followed a period of testing and optimization.
Commercial scale	Full-scale infrastructure built at the design capacity of company’s commercial offering. The goal of a commercial scale system is to produce a liquid petroleum product to be marketed of off takers for the purposes of achieving profitability.

**A.2 COLLECTION AND SORTING OF INVENTORY DATA**

Collection, transportation, and sorting at the material recovery facility (MRF) comprised the stages in the feedstock logistics and processing stage. The assumptions and inventory data used for the calculations of fossil energy consumption, greenhouse gas (GHG) emissions, water consumption, and criteria air pollutant (CAP) emissions of this stages are shown in Tables A.2-1 and A.2-2.

**Table A.2-1. Assumptions Used for Collection (Argonne, 2020)**

Parameter	Value
Fuel economy (km/L)	2
Distance (mi)	80.5
Carried payload (wet tons)	12.4

**Table A.2-2. Inventory of Post-use Plastic (PUP) Sorting at MRF**

Energy Inputs	Amount (MJ/kg)
Natural gas	0.0001
Electricity	0.05
Diesel	0.07
Liquified petroleum gas	0.08

**A.3 ADDITIONAL INFORMATION USED IN THE LCA**

Apart from the base case for analysis defined in Figure 1 (energy and market allocation, fuel gas only used to produce heat, and char sent to landfill) there are other possible cases that can be explored in the analysis. These other cases consider other coproduct treatment methods and uses for coproducts. Tables A.3-1 to A.3-3 show the other possible cases that can included and the parameters that can be used in the analysis.

**Table A.3-1. Extended Scenarios in the LCA**

Product	All Fuel Gas Combusted (Heat and Electricity)		Fuel Gas for Heat Only	
	Displacement	Energy or Market Value Allocation	Displacement	Energy or Market Value Allocation
Naphtha	Displaces conventional low-sulfur naphtha	Sold as energy product	Displaces conventional low-sulfur naphtha	Sold as energy product
Wax	Displaces residual oil (approximation)	Sold as energy product	Displaces residual oil (approximation)	Sold as energy product
Fuel gas	Excess electricity displaces grid electricity	Excess electricity sold to electricity grid	Displaces natural gas	Excess fuel gas sold as energy product
Char	Landfilled or displaces petroleum coke	Landfilled or sold as energy product	Landfilled or displaces petroleum coke	Landfilled or sold as energy product

**Table A.3-2. Allocation Factors in Each Plant Capacity Group <sup>a</sup>**

Plant Capacity Group	Energy Allocation		Market Allocation	
	Pyrolysis Oil	ULS <sup>b</sup> Diesel	Pyrolysis Oil	ULS <sup>b</sup> Diesel
Pioneer	92%	61%	99%	36%
<i>N</i> <sup>th</sup> -plant	84%	43%	97%	37%
Highest capacity	79%	-29%	96%	32%

<sup>a</sup> Fuel gas is used internally only to produce heat for the pyrolysis plant. Char is sent to landfill.

<sup>b</sup> ULS: ultra-low sulfur.

**Table A.3-3. Energy Displaced by Coproducts in Each Plant Capacity Group  
(MJ/MJ of product)**

Plant Capacity Group	Pyrolysis Oil		ULS Diesel			
	Fuel Gas	Char	Fuel Gas	Char	Naphtha	Wax
Pioneer	0.09	0.00	0.14	0.00	0.49	0.57
<i>N</i> <sup>th</sup> -plant	0.20	0.00	0.41	0.00	0.94	0.29
Highest capacity	0.27	0.00	0.75	0.00	1.65	0.13

The companies provided information about the consumption of process water in the different stages. The water consumption is defined as the volume of makeup water per megajoule of product supplied to the process because it cannot be recovered. Data about water consumption, as reported by the companies, is shown in Table A.3-4.

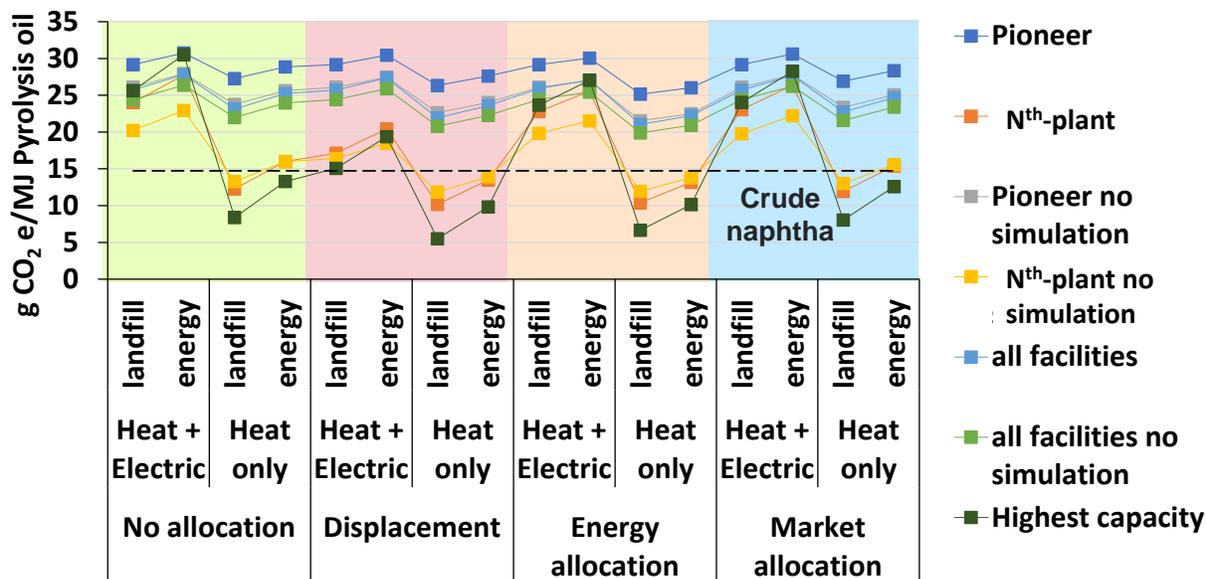
**Table A.3-4. Water Consumption from Survey (mL/MJ of product)**

Plant Capacity Group	Pyrolysis Oil	ULS Diesel	
	Conversion	Conversion	Separation/Upgrading
Pioneer	22.6	46.6	26.1
<i>N</i> <sup>th</sup> -plant	4.7	10.6	4.0
Highest capacity	10.1	28.2	0.0

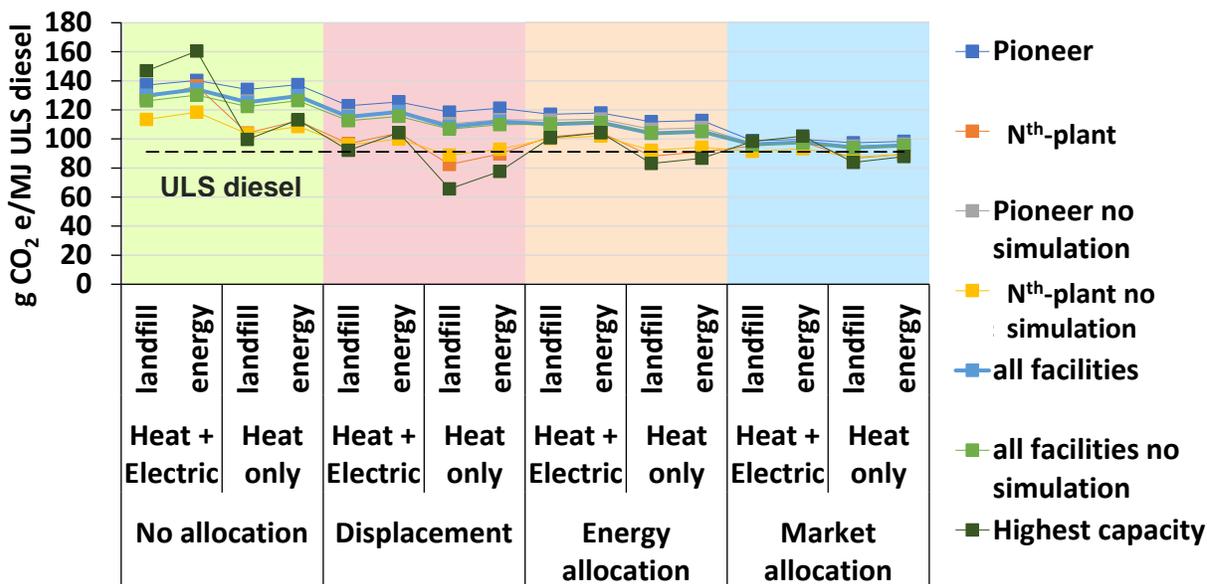
#### **A.4 LCA RESULTS FOR ALL TYPES OF PLANTS**

The results from the different cases presented in Table A.3-1 are shown in Figures A.4-1 through A.4-4. As shown in the figures, the analysis incorporated additional plant capacity groups that compare simulated and non-simulated data and a plant capacity group that includes all the facilities. Overall, there were no major differences between simulated and non-simulated data for all the facility groups.

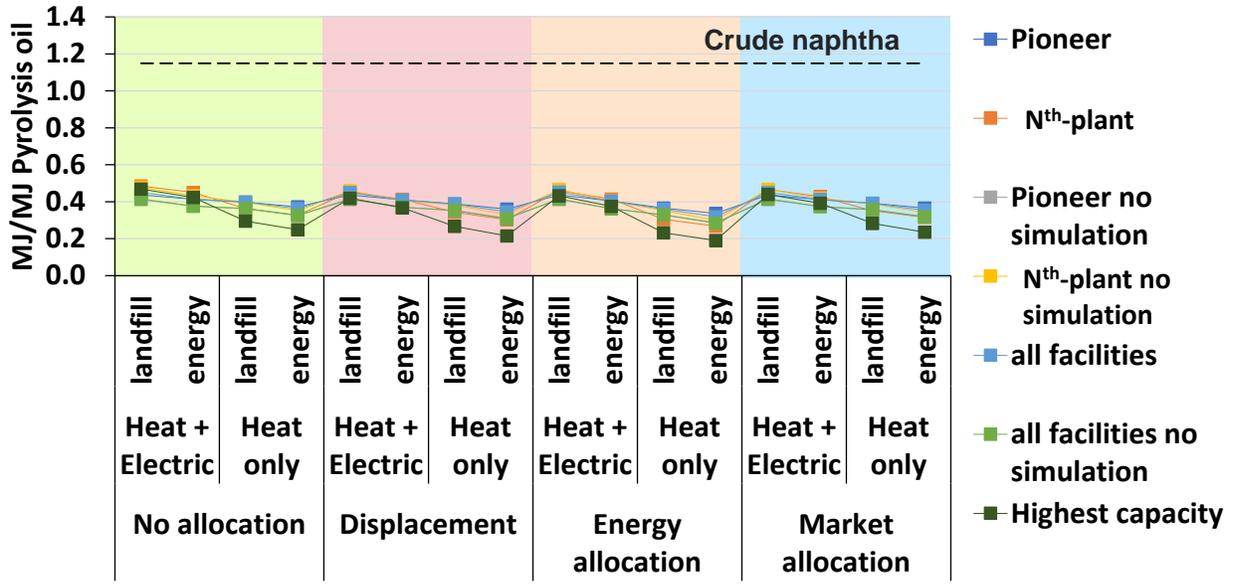
In addition, the displacement methods resulted in the lowest values of fossil energy consumption and GHG emissions for PUP-derived ULS diesel and pyrolysis oil. Similarly, to the results of the base case, *N*<sup>th</sup>-plants and highest capacity showed the lowest values of fossil energy consumption and GHG emissions from all the groups analyzed. In addition, the scenarios where the fuel gas is only used for the heat generation (“heat only”) showed lower GHG emissions compared to those where fuel gas is also used to produce electricity (“Heat + Electric”). This is because the GHG emissions to produce electricity from fuel gas (generation efficiency of 25%) are higher than the credits from avoiding the use of external energy.



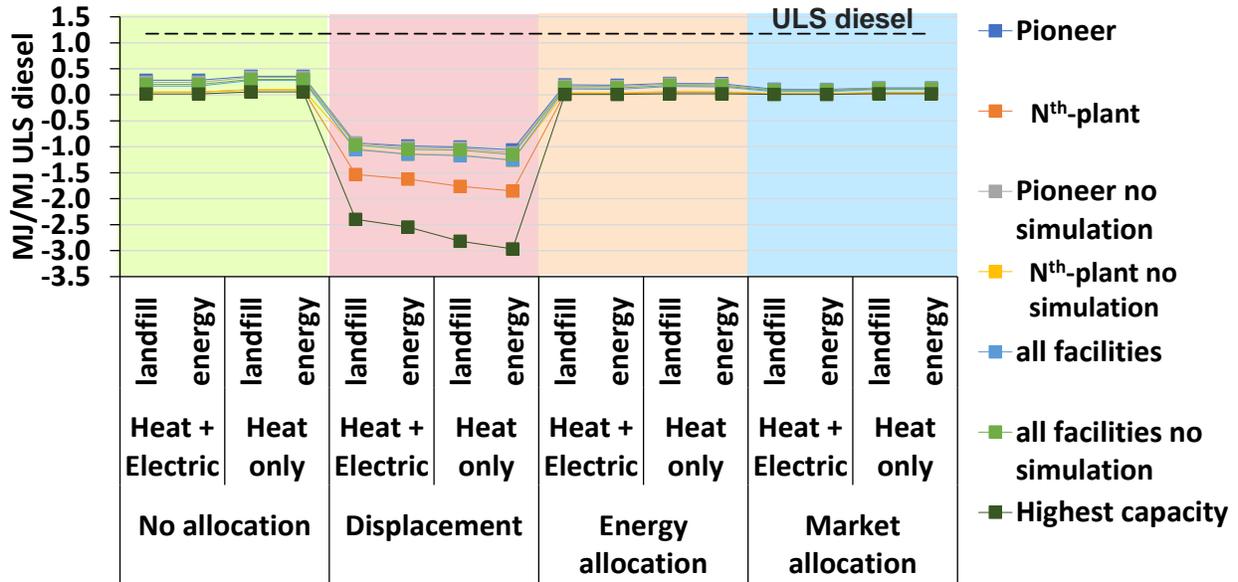
**Figure A.4-1. Lifecycle GHG Emissions Using Different Coproduct Allocation Methods for PUP-derived Pyrolysis Oil.** This figure includes emissions from feedstock processing and conversion; GHG emissions of crude naphtha: 14.7 g CO<sub>2</sub>e/MJ pyrolysis oil (Argonne, 2020).



**Figure A.4-2. Lifecycle GHG Emissions Using different Coproduct Allocation Methods for PUP-derived ULS Diesel.** This figure includes emissions from feedstock processing, conversion, upgrading/separation, and vehicle use; GHG emissions of petroleum-based ULS diesel: 91.1 g CO<sub>2</sub>e/MJ pyrolysis oil (Argonne, 2020).



**Figure A.4-3. Fossil Energy Consumption to Produce PUP-derived Pyrolysis Oil for Different Coproduct Treatment Methods.** Fossil energy consumption estimates include the energy of feedstock processing and internal and external energy for conversion; fossil energy consumption for crude naphtha: 1.1 MJ/MJ pyrolysis oil (Argonne, 2020).



**Figure A.4-4. Fossil Energy Consumption to Produce PUP-derived ULS Diesel for Different Coproduct Treatment Methods.** Fossil energy consumption of PUP-derived ULS diesel include the energy of feedstock processing and external energy for conversion; fossil energy consumption for petroleum-derived ULS diesel includes energy of feedstock processing, internal and external energy for conversion, and energy for vehicle operation; fossil energy consumption of fossil-based ULS diesel: 1.2 MJ/MJ pyrolysis oil (Argonne, 2020).

## A.5 ELECTRICITY GRIDS USED IN THE SENSITIVITY ANALYSIS

The sensitivity analysis considered variations in the sources of the electricity grid mix. Three possibilities were explored: an electricity grid with a high percentage of renewable feedstocks (California [CA] mix), an electricity grid with a low percentage of renewable feedstocks (Florida Reliability Coordinating Council [FRCC] mix) and an electricity grid with high GHG emissions, based on a high use of coal (Midwest Reliability Organization [MRO] mix). The contribution of the different energy sources along with the GHG emissions per kilowatt-hour of electricity of each of these grid mixes is presented in Table A.5-1.

**Table A.5-1. Electricity Mixes for the Grids Used in the Sensitivity Analysis (Argonne, 2020)**

Energy Source	U.S. Mix	CA Mix	MRO Mix	FRCC Mix
Residual oil	0.4%	0.0%	0.8%	0.2%
Natural gas	36.7%	42.5%	28.5%	69.9%
Coal	24.6%	0.0%	43.3%	13.2%
Nuclear power	20.4%	10.3%	15.4%	13.4%
Biomass	0.3%	1.2%	0.2%	0.2%
Hydroelectric	7.3%	14.1%	1.4%	0.8%
Geothermal	0.4%	4.5%	0.0%	0.0%
Wind	7.5%	8.1%	9.8%	0.0%
Solar PV	1.8%	18.3%	0.3%	1.3%
Others	0.5%	1.1%	0.4%	1.1%
GHG (g CO <sub>2</sub> e/kWh)	449	253	661	501

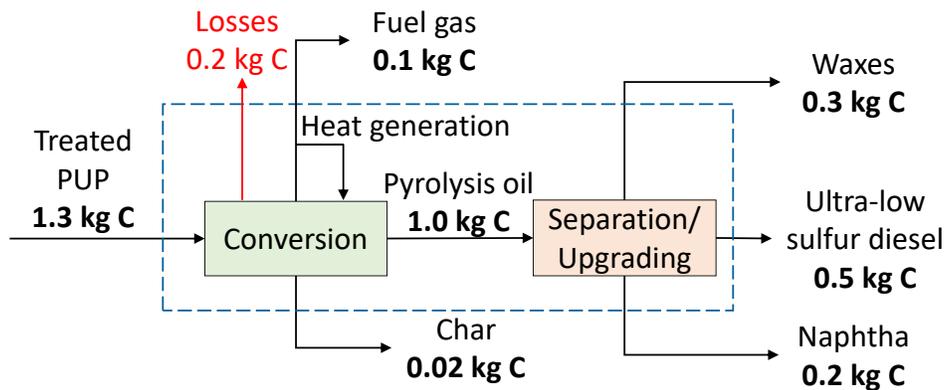
## A.6 CARBON EFFICIENCY AND BALANCE

As described in Section 3.2, carbon efficiency represents the amount of carbon from the PUP incorporated in the products. Because there are differences in the carbon content (expressed as percentage of carbon) of the pyrolysis products, we performed a carbon balance to estimate the distribution of carbon between fuel gas, char, diesel, naphtha, and waxes. Data on the carbon content of pyrolysis oil, fuel gas, and char were obtained from the survey, and the carbon content of naphtha and diesel was obtained from GREET (2020). The carbon content of waxes considered the mass composition of the fractions of pyrolysis oil in its estimation. The carbon content of each product was multiplied by its corresponding mass as presented in Figure 5 to obtain the amount of carbon in each product stream. After completing of the carbon balance, the carbon efficiency was estimated using equation (A.1):

$$C_{\text{eff}} = \frac{C_{\text{FG}} + C_{\text{C}} + C_{\text{D}} + C_{\text{N}} + C_{\text{W}}}{C_{\text{PUP}}} \quad (\text{A.1})$$

where  $C_{\text{eff}}$  is the carbon efficiency and  $C_{\text{FG}}$ ,  $C_{\text{C}}$ ,  $C_{\text{D}}$ ,  $C_{\text{N}}$ , and  $C_{\text{W}}$  are the carbon amount in fuel gas, char, diesel, naphtha, and waxes, respectively.

Figure A.6-1 shows the results of the carbon balance for pioneer plants, normalized to 1 kg of carbon in the pyrolysis oil. As observed, there are carbon losses occurring during conversion; these are a consequence of the combustion of fuel gas to produce heat and the emissions of VOCs present in some plastic additives (Noguchi & Yamasaki, 2020; Miandad et al., 2019). These carbon losses are converted to carbon dioxide (CO<sub>2</sub>) and included in the GHG emissions calculations. According to Figure 13, the carbon efficiency increases with the transition from pioneer to the highest capacity facility, which is a consequence of the reduced amount of combusted fuel gas for facility operations. As shown, this presents an advantage due to economies of scale. The carbon losses (indicated by a red arrow) are due to the combustion of fuel gas for heat generation and emissions of volatile organic compounds (VOCs) in PUP during thermal degradation.



**Figure A.6-1. Carbon Balance for Pioneer Plants in the Conversion and Separation/Upgrading Process**



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