Plastics to fuel or plastics: life cycle assessment-based evaluation of different options for pyrolysis at end-of-life

Sabyasachi Das,[1] Chao Liang,[2] Jennifer B. Dunn, *[1,3,4]

1. Chemical and Biological Engineering, Northwestern University, Evanston, IL, USA
2. Institute of Sustainability and Energy at Northwestern, Northwestern University, Evanston, IL, USA
3. Center for Engineering Sustainability and Resilience, Northwestern University, Evanston, IL, USA
4. Northwestern-Argonne Institute of Science and Engineering, Northwestern University, Evanston, IL, USA

*jennifer.dunn1@northwestern.edu

Abstract

Pyrolysis is a leading technology to convert non-recyclable plastic waste to fuels or chemicals. As interest in the circular economy grows, the latter option has seemingly become more attractive. Once waste plastic is pyrolyzed to, for example, naphtha, however, additional steps are required to produce a polymer product. These steps consume additional energy and water and emit greenhouse gases (GHG). It is unclear whether this more circular option of non-recyclable plastics to virgin plastics offers environmental benefits, compared to their conversion to fuels. We therefore examine whether it is possible to determine the best use of pyrolyzing non-recyclable plastic – fuels or chemicals (low-density polyethylene (LDPE) as product)– from a life cycle perspective. We use recently published life cycle assessments of non-recycled plastics pyrolysis and consider two functional units: per unit mass of non-recyclable plastics and per unit product - MJ of naphtha or kg of LDPE. In the U.S., on a cradle-to-gate, per unit mass waste basis, producing fuel is lower-emitting than producing LDPE from pyrolysis . The opposite is true in the EU. But expanding the system boundary to the grave results in LDPE as the lower-emitting product in both regions. Naphtha and LDPE produced from non-recyclable plastics are less GHG-intensive than conventional routes to these products. Fossil fuel and water

consumption and waste generation are all lower in the P2F case. Our results highlight that prioritization of P2P and P2F may depend on regional characteristics such as conventional waste management techniques and water scarcity.

Keywords

Waste plastic, pyrolysis, life cycle assessment

1. Introduction

Addressing ever-growing volumes of waste plastic that make their way to landfills, incinerators, and the environment has reached a critical stage with societal, industrial,(Alliance to End Plastic Waste, 2021) investor (Crowley, 2021) and international and federal agency interest (United Nations Environment Programme, 2022) in reducing waste generation stronger than ever. While some types of polymers (e.g., polyethylene terephthalate and high-density polyethylene) are amenable to recycling, others are not because their properties deteriorate upon recycling, technology to recycle them does not exist, or their recycling is not economically viable. These factors underlie the very low plastics recycling rate (e.g., ~8% in the United States(US EPA, 2017a)).

There are numerous options for managing plastics at the end of life. The most common option, landfilling, wastes the inherent energy in plastics and takes up land. Another option is incinerating waste plastic to recover energy, which can result in toxic air emissions (e.g., dioxins) (Hou et al., 2018) depending upon the composition of plastic waste and air pollution control measures. Increasingly, mechanical and chemical (Chen et al., 2021) recycling methods are receiving attention as paving the way towards a circular economy. It should be noted that various recycling methods may be more or less beneficial for different plastic types as compared to more conventional waste handling methods (landfill, energy recovery) depending on a number of factors including energy content.(Meys et al., 2020)

Pyrolysis is one of the most prominent chemical recycling technologies that is in the early stages of commercialization.(Luu, 2021; Solis and Silveira, 2020) Examples of pyrolysis entering commercial-scale operations include Brightmark's 100,000 ton plastic waste per year facility under construction in Indiana, United States and the partnership between Fuenix Ecogy and Dow in the Netherlands.(Chemical and Engineering News, 2020; Luu, 2021) It can be used with mixed plastic wastes that are not economically viable for recycling. Pyrolysis products include a mix of energy products (e.g., naphtha, diesel, char, and fuel gas).(Benavides et al., 2017; Jeswani et al., 2021) Naphtha can be used as a fuel or a raw material for chemicals production. The latter option has been a focus of recent chemical recycling efforts that target circularity as a primary objective. It is unclear, however, whether expending energy to convert naphtha to a polymer such as low-density polyethylene (LDPE) to achieve circularity is better than stopping at the production of naphtha. Combusting a fuel made from naphtha releases CO_2 to the air and breaks any link to circularity for the plastic, but uses the energy the waste plastic contains – potentially in place of energy in virgin fossil fuels - and does not contribute to solid waste generation. On the other hand, converting the naphtha to a plastic consumes energy and emits GHGs. The end-of-life fate of that plastic is most likely (in the U.S. and many countries that contribute the most to plastic waste)(Law et al., 2020) either landfilling or release to the environment. As a result, it is not immediately clear which option offers comparably more environmental benefits. Importantly, regional differences may affect the relative merits of the

P2P and P2F pathways. Regions differ in their waste management methods, the energy- and GHG-intensity of the production of virgin chemicals and fuels which may use different feedstocks, technologies, energy sources, water availability, and the electricity grid mix. To evaluate whether circularity is the better objective for end-of-life plastics that undergo pyrolysis – and what data gaps and regional differences might exist that influence the relative merits of fuels compared to plastics in chemical recycling - we carried out a simple life cycle assessment (LCA) using existing data and high-level assumptions to compare plastics-to-plastics (P2P) and plastics-to-fuels (P2F) routes for waste plastics with two different functional units (mass of waste plastic managed and per unit product). Given the above-mentioned regional differences, we carried out this evaluation in the context of the United States (U.S.) and the European Union (EU).

2. Methods

Both P2P and P2F pathways comprise a pyrolysis step to convert waste plastic feedstock into naphtha. In the P2F process, naphtha is the main product and is used as a fuel. In the P2P process, the naphtha is converted to ethylene, which is then polymerized to low-density polyethylene (LDPE). The parameters used in our analysis are documented in Table 1 including feedstock composition, product yields, electricity grid, and conventional waste management parameters. We determined the GHG emissions and fossil energy consumption for both pathways. We considered two functional units: mass of waste plastic and unit product (MJ naphtha in the case of P2F and kg LDPE in the case of P2P). In the former case, we consider how results are affected by recycling, landfilling, and incineration rate differences between the U.S. and the EU. When adopting a product perspective, we considered that the plastic produced via chemical recycling could be recycled at the rate specific to the region of study. This may be a best case assumption in terms of retaining the inherent value (e.g., avoiding downcycling) of the end-of-life plastic because mechanical recycling, which may entail downcycling, is the dominant recycling method operating commercially.(U. S. Government Accountability Office, 2021) We note that the methodology of evaluating plastics circularity in LCA remains under debate and that other analysis approaches to addressing circularity or that use other recycling methods for the waste-plastic derived polymers would be possible.(Huysveld et al., 2019) It is beyond the scope of this analysis to explore the sensitivity of results to the method of evaluating circularity's effects.

INSERT TABLE 1

One previous study(Benavides et al., 2017) that supports our analysis considered conversion of non-recycled plastic (NRP) waste (25 wt% HDPE, 33 wt% LDPE and 42 wt% PP) to diesel fuel. No chlorine or oxygen entered the pyrolysis reactor based on the feedstock composition; accordingly, no post-pyrolysis purification was required. This is consistent with another analysis of plastics pyrolysis(Jeswani et al., 2021) which also relied on industry data and observed that less than 0.5% of the pyrolysis reactor feed contained chlorine. Jeswani et al. assumed a feedstock composition of 90 wt% PE, PP and PS and 10 wt% impurities. Based on surveys of P2F companies, Benavides et al. determined that the liquid pyrolysis product was 80% (by vol.) diesel and 20% naphtha. In our analysis, however, we changed the liquid product composition to 100% naphtha and 0% diesel. While energy consumption of the process would likely change with this shift of product slate, we envision the pyrolysis process much like a petroleum refinery that can shift its product slate through changes in temperature, pressure, and other parameters without extraordinary differences in carbon intensity that would greatly change LCA results. It has been shown(Elgowainy et al., 2014) that a range of a few g CO_2e/MJ for refinery products exists over 43 different refineries and among different refinery products. We take this assumption in this perspectives piece as sufficient to explore the big-picture question we are asking regarding best use of pyrolysis for end-of-life plastics. A detailed analysis, however, would require a close look at the implications of changing the product slate and, depending on the feed composition, consider post-pyrolysis refining steps. Benavides et al. explored different uses of the fuel gas and char co-products. In our calculations, we assumed that fuel gas would be used for internal heat. Excess fuel gas would be sold as an energy product. We used the system expansion co-product handling approach. In this approach, the main product (naphtha or LDPE) is assigned all emissions from the process. Subsequently, emissions associated with conventional manufacturing of the amount of co-products produced along with the main product (e.g., from virgin fossil fuels) are subtracted from this GHG intensity. System expansion is also applied to the electricity produced from waste incineration in scenarios that incorporate conventional waste management. Section 6 of the Supplementary Information provides the equations and parameters we used in these calculations. Benavides et al. observed that LCA results for the P2F pathway were relatively insensitive to co-product handling choice. The Supplementary Information contains a life cycle inventory data table and schematic depicting data sources.

For the U.S. context, we modified the existing P2F pathway in the Greenhouse gases, Regulated Emissions and Energy use in Technologies (GREET) model (Argonne National Laboratory, 2020) to characterize the P2P pathway. The produced naphtha was assumed to undergo cracking to produce ethylene(Yang and You, 2017). The ethylene needs to be separated from other cracking products like propylene, hydrogen, and C_4 and C_5 hydrocarbons. We used mass allocation to spread the energy and emissions burdens and credits for this among cracking process co-products. Subsequently, ethylene undergoes polymerization to produce polyethylene. Material and energy consumption data from the GREET Bioproducts module (Dunn et al., 2015) were used to characterize the polymerization process.

For the EU context, data for the production of ethylene from waste plastic feed were extracted for European conditions.(Jeswani et al., 2021) Data for polymerization of ethylene to LDPE was also derived for European conditions.(Vanderreydt et al., 2021)

We adopted two functional units in this analysis. Figure 1 depicts the system boundary when the functional unit is 1 kg of waste plastic. Figure 2 reflects a product-based functional unit: 1 MJ of naphtha fuel (P2F) or 1 kg LDPE (P2P).

The baselines depend on the choice of functional unit. For the waste-based perspective (Figure 1), the baseline for non-recycled plastic is the current conventional waste-handling in the U.S. (20% incineration, 80% landfill). It is important to note that in the EU, on average, 63% and 37% of waste is incinerated and landfilled, respectively. (Benavides et al., 2017) 33% of plastic waste (Centro De Documentacion Europea, 2021) is recycled in the EU whereas in the US only 8% is. We assume that emissions from landfilling operations are negligible in this analysis, which aligns with previously published results(Demetrious and Crossin, 2019) that report negligible landfilling emissions associated with mixed plastic waste. In the U.S. context, for the product perspective (Figure 2) the baseline for the P2F pathway is the production of naphtha in the GREET Petroleum module. For the P2P pathway, the baseline is the conventional LDPE production as modeled in

GREET. For the EU context, we also use similar values for conventional production of naphtha(Boustead, 2005) and LDPE(Vanderreydt et al., 2021).

INSERT FIGURES 1 and 2

3. Results

Figure 3 illustrates life-cycle GHG emissions results for the P2P and P2F cases when the functional unit is 1 kg NRP. The results in Figure 3 and all subsequent figures are tabulated in the supplementary information.

INSERT FIGURE 3

In Figure 3, contributors to emissions with solid outlines are activities that occur when the system boundary ends at the (solid lines in Figure 1) pyrolysis factory gate. Contributors with dashed lines reflect emissions from activities between the gate and the grave. In the P2P case, we assume that the regionally-specific plastic recycling, incineration, and landfilling rates apply to the kg of waste. That is, (in the US) 1 kg of NRP that undergoes pyrolysis and subsequent processing to produce LDPE yields 0.25 kg of LDPE. When that mass of plastic reaches its end of life, 18% of it is incinerated, 74% of it is landfilled, and 8% of it is recycled (we assume chemically to produce LDPE). In our high-level analysis, we assume that the pyrolysis process has the same yield when the feed is only LDPE as opposed to having the pyrolysis feed composition in Table 1.

In Figure 3, P2F *cradle-to-gate* emissions are lower than in the P2P case for US context. Compared to the P2F pathway, P2P requires additional processing that raises emissions over those of the P2F pathway when the system boundary ends at the gate. This is not true for the EU context because of the much higher LDPE yield (~2X), which results in greater credits for system expansion for the P2P case. When the system boundary extends to the grave, however, GHG emissions from naphtha combustion drive up P2F emissions and they exceed those of the P2P case for both U.S. and EU contexts. As illustrated in Figure 1, the P2P pathway in the U.S. includes processing the 0.25 kg LDPE produced from the original 1 kg of NRP through the "average" system for dealing with plastic waste. In the US, this means that only 8% (or 0.02 kg) of it is recycled. Accordingly, the second cycle emissions in Figure 3 are effectively negligible. We therefore only account for one recycling process in the U.S. context as subsequent cycles deal with vanishingly small masses. When the system boundary extends to the end-of-life of the fuel or plastic, the P2F cases have higher emissions than conventional waste handling. We note that if waste were only landfilled, (and none were incinerated) emissions from conventional waste handling would be effectively zero, but there would be no circularity at all in the management of waste. In the P2F case, there is no waste at the end-of-life. Assuming the 0.25 kg of LDPE produced in the U.S. P2P case experiences average end-of-life conditions, 0.74 kg of waste will be generated (Figure 4). No waste is generated in the P2F scenario (in the US or the EU).

INSERT Figure 4

In the EU, conventional waste management of non-recycled plastics entails 37% landfilling and 63% incineration. As a result, baseline waste management emissions are higher in the EU than in the US. In the EU, P2P GHG emissions are lower than P2F. In the P2P pathway, the effects of recycling are more evident than in the U.S. because 33% versus 8% of plastic waste is recycled. Second cycle emissions (e.g., chemical recycling of the LDPE (0.54 kg from 1 kg of NRP)) are higher because the higher recycling rate translates to an appreciable amount of mass that is chemically recycled. Whereas in the US analysis we could only achieve one pass of the pyrolysis-derived LDPE through chemical recycling, we can evaluate two such passes in the EU context – extending to three cycles.

When interpreting Figure 3, it is also important to note the differences in the electricity grid mix between the U.S. (33% renewable) and the EU (48% renewable) based on our underlying data sources (Table 1). The greater share of renewable electricity in the European context lessens the systems expansion-based emissions benefit from accounting for using electricity from waste incineration in place of conventional grid electricity. But in the EU context the LDPE yield is higher so, overall, P2P emissions are lower than in the U.S. context. P2F GHG emissions in the EU-based analysis are lower than in the U.S. context because Benavides et al. reported a higher yield than Jeswani et al. (Table 1). With more naphtha produced, naphtha combustion emissions are higher per kg of waste processed.

INSERT Figure 5

From a product perspective, both the P2P and P2F routes from waste plastic are environmentally more favorable than their baseline scenarios (Figure 5). This result holds in both the U.S. and EU contexts. The baseline for the P2F route is conventional naphtha production from petroleum; the baseline for the P2P route is conventional LDPE production. In the U.S., the life-cycle GHG emissions are 12% lower for naphtha when it is produced via NRP pyrolysis compared to conventional routes. This GHG emissions reduction is driven mostly by the lower GHG emissions within the cradle-to-gate system boundary because combustion emissions per unit mass of naphtha are the same for pyrolysis- and conventionally-derived naphtha. On the other hand, in the U.S. context, the process to produce LDPE from NRP is lower-emitting than the baseline LDPE process by 18%. From a product perspective, the P2P route offers a greater reduction in GHG emissions than the P2F route when compared to their respective baselines. The same overall conclusion holds true in the EU, with a 7% reduction for the P2F route compared to conventional naphtha production and a 37% reduction when LDPE is prepared via chemical recycling instead of from virgin fossil fuels (Boustead, 2005; Vanderreydt et al., 2021).

4. Discussion

The results of our analysis indicate that, when the system boundary extends beyond the pyrolysis plant gate, the P2P route consistently offers GHG emissions benefits regardless of region or functional unit, although the extent of these reductions does depend on the viewpoint that the functional unit defines. The benefit is greater from the perspective of unit of mass waste managed compared to per mass of plastic product produced from pyrolysis. Notably, emissions are higher in the EU context because of the higher share of incineration in waste management.

Beyond GHG emissions reductions, a circular plastics economy has the potential to reduce fossil fuel consumption. Figures S4 and S5 in the supplementary information contain life-cycle fossil fuel energy consumption with a per mass or energy functional unit (product perspective). For both products (naphtha and LDPE), fossil fuel consumption is significantly lower for the products derived from waste plastics rather than virgin fossil fuels. It is also lower for the P2F route than for the P2P route (99% v 56%). We also evaluated life-cycle water consumption from the product perspective. Figure S3 illustrates that this metric is favorable in the P2F route but not in the P2P case because of water consumed in converting naphtha to LDPE. This may be a notable distinction with a strong regional influence given projected water scarcity in the future(He et al., 2021). Solid waste and associated terrestrial ecotoxicity are other important metrics that would favor the P2P and P2F pathways over conventional waste management, especially in the U.S given its high rate of landfilling(Saling et al., 2020). The P2F route holds an advantage in this regard because it will not generate any solid waste. These results show the importance of considering metrics beyond GHG emissions reductions alone.

It is worth considering the total emissions reductions that could result if waste plastics were used at a large scale as a source of fuels or chemicals. Annual production of naphtha between 2018 and 2020 in the United States ranged from 320-400 billion MJ.(Energy Information Administration (EIA), 2021) We could conceivably produce about 825 billion MJ(US EPA, 2017b) naphtha from waste plastics, exceeding this demand. If we entirely replaced naphtha production from petroleum with naphtha production from waste plastic, emissions reductions would be 3.9 million tonnes of CO_2e. On the other hand, approximately 3.5 million metric tons of LDPE are produced annually in the U.S.(American Chemistry Council, 2020) From waste plastic, it would be possible to produce 1.8 times this amount. If all LDPE were produced from pyrolysis of NRP, emissions reductions would be 1,940 tonnes of CO_2e. This reduction is orders of magnitude lower than the reduction associated with displacing conventional naphtha with fuels from waste plastic. Clearly, viewing plastic at its end-of-life as a resource to be converted to either fuels or plastics could generate significant GHG savings although P2F routes may offer greater savings.

5. Conclusion

Based on this analysis, we can be fairly certain that P2P pathways enable plastic waste management that is lower-emitting than current waste management practices and would produce plastics that are lower-emitting than conventional production routes. However, when taking into account a larger environmental picture that includes solid waste generation and the largely uncertain fate of plastic waste (even the U.S. with its strong waste management system is the third largest contributor of mismanaged plastic waste to the coastal environment world's plastic waste globally), (Law et al., 2020) it is important to keep P2F on the table as a waste management option, especially in regions that incinerate a large share of plastic waste and/or have very low recycling rates. Furthermore, P2F pathways are less water-intensive than P2P pathways, which may be a critical factor in water-scarce regions.

To improve decision making regarding the best direction for pyrolysis of waste plastic, we need to first evaluate the different data sources for the pyrolysis process. We need to account for the use of early TRL data in this analysis and perhaps set targets for yields and other parameters in the pyrolysis and subsequent processing steps to help us design and implement recycling processes that offer environmental advantages across multiple categories (e.g, GHG emissions, solid waste generation, water consumption). Moreover, in this study, we used literature-based data for the cracking step, which would benefit from the use of real-world data. We also need to better understand how regional differences might influence the "best" options for managing plastic wastes, by performing such analyses for different geographic regions and varying starting waste plastics. Three salient examples of regional differences could include varying leakage rates in upstream production of methane (would influence baseline LCA results for plastics), electricity grid mix differences, and baseline waste management practices. It also would be helpful to have sound regional projections of anticipated changes in waste management practices. As the United Nations negotiate the Treaty on Plastic Pollution (United Nations Environment Programme, 2022) policies that will lend themselves to such projections may become clearer. Furthermore, if baseline products are produced differently in different parts of the world, we may need to adjust approaches to dealing with waste plastics accordingly so using them to produce new fuels and chemicals, at a minimum, does no harm compared to what we do today.

We also note that the approach we took in evaluating recycling in the P2P pathway is just one of many methods that are possible (Demets et al., 2021; Schaubroeck et al., 2021; Tian et al., 2022). Results will differ depending on the methodology employed. Furthermore, in many regions, recycling is not common at all. In this case, it is best to assume the waste is either landfilled or released to the environment. In the latter case, it is important to ask whether the reduction of waste through combusting pyrolysis-derived fuel where fuel might already be used is a better outcome than further pollution of waterways and oceans with plastic waste. If so, P2F may be the better option. In addition, this study only considers pyrolysis as a recycling technology. Other recycling technologies could potentially make the P2P process less energy-, GHG-, and water-intensive. Similarly, the P2F pathway assumes naphtha as the only fuel produced. Scenarios could be explored to produce other fuels like diesel to see the effect of this choice on the GHG intensity of the P2F process. Finally, environmental impact categories other than GHG emissions that encapsulate the toxicity and land use aspects of waste plastics should also be explored to provide a more well-rounded analysis of the environmental benefits of plastics recycling.

As society grapples with how best to manage plastic waste, addressing data gaps, and continuing to discuss how best to manage circularity in LCA will be important ways LCA can contribute to decision making in communities, at companies, and within policy making. Furthermore, incorporating LCA metrics that account for plastics effects on the environment beyond GHG emissions is essential. For example, incorporating emissions of air pollutants, including air toxics, is a necessary step given the ongoing policy debate regarding whether pyrolysis-based plastics recycling constitutes incineration and should be regulated accordingly (Hogue, 2022). Finally, LCAs should robustly account for the effect of recycling technology to reduce the effects of plastic waste on aquatic and other ecosystems (Maga et al., 2022; Saling et al., 2020)

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Tables and Figures

Table 1: Key parameters in Benavides et al. and Jeswani et al.

Parameter	**Benavides et al.**(Benavides et al., 2017)	**Jeswani et al.**(Jeswani et al., 2021)
Geographic Region	United States of America	Europe
Waste plastic feed	(25 wt % HDPE, 33 wt% LDPE, 42 wt% PP)	(Mix of PE, PP and PS (~90% by wt.) and 10% impurities)
Conventional Waste Handling method	80% landfill, 20% incineration	37% landfill, 63% incineration
Yield of naphtha from pyrolysis	75%	63%
Co-products	Fuel gas (16%), Char (9%)	Fuel gas (19%), Char (7%), Heavy vacuum residue (1%)
Electricity Grid	U.S. Electricity Mix in 2016 (33% renewable)	EU Electricity Mix in 2013 (48% renewable)
Plastic Recycling Rate	8%	33%
Yield of LDPE	25%	54%

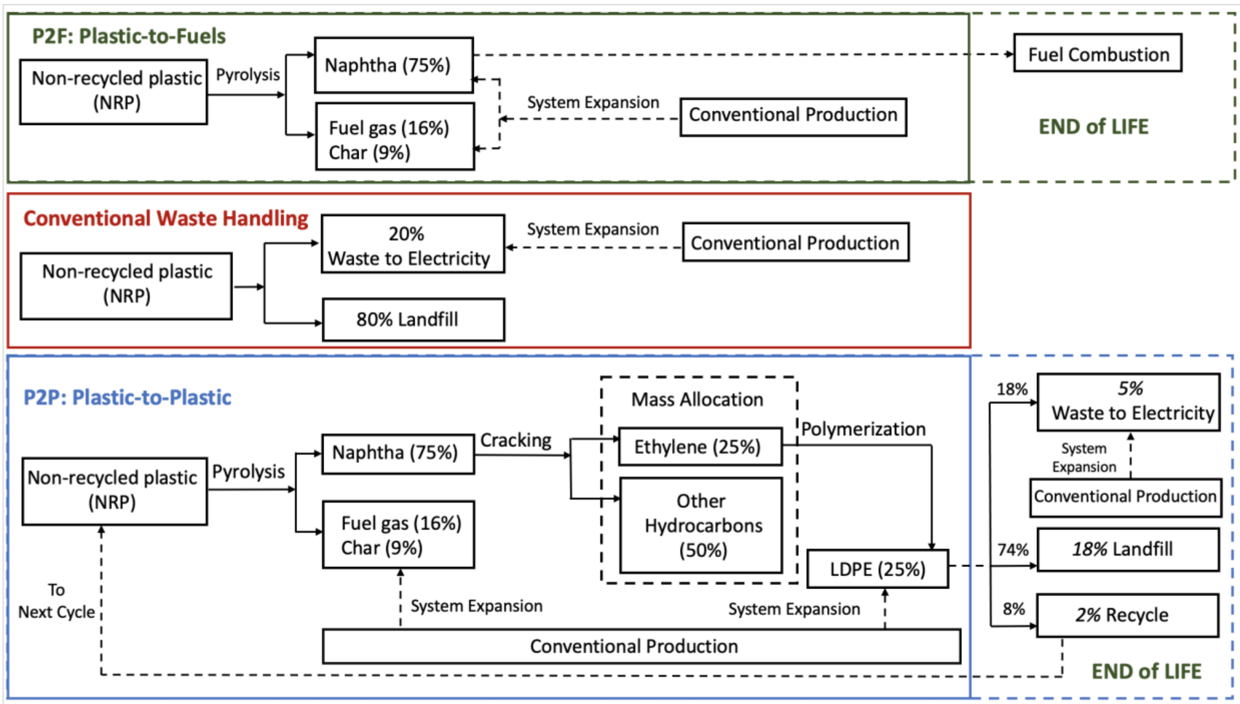

Figure 1: System boundary diagram for a functional unit of mass of waste plastic. The mass shares reported in this figure reflect the US context. Percentages in italics within the end of life portion of the system boundary reflect the percent of mass of the original kg of NRP that advances to this stage. Shares relevant to the EU context are reported in Table 1. The feedstock to the pyrolysis process in the US context is 25 wt% HDPE, 33 wt% LDPE and 42 wt% PP. The feedstock to the pyrolysis process in the EU context is 90 wt% PE, PP and PS and 10 wt% impurities.

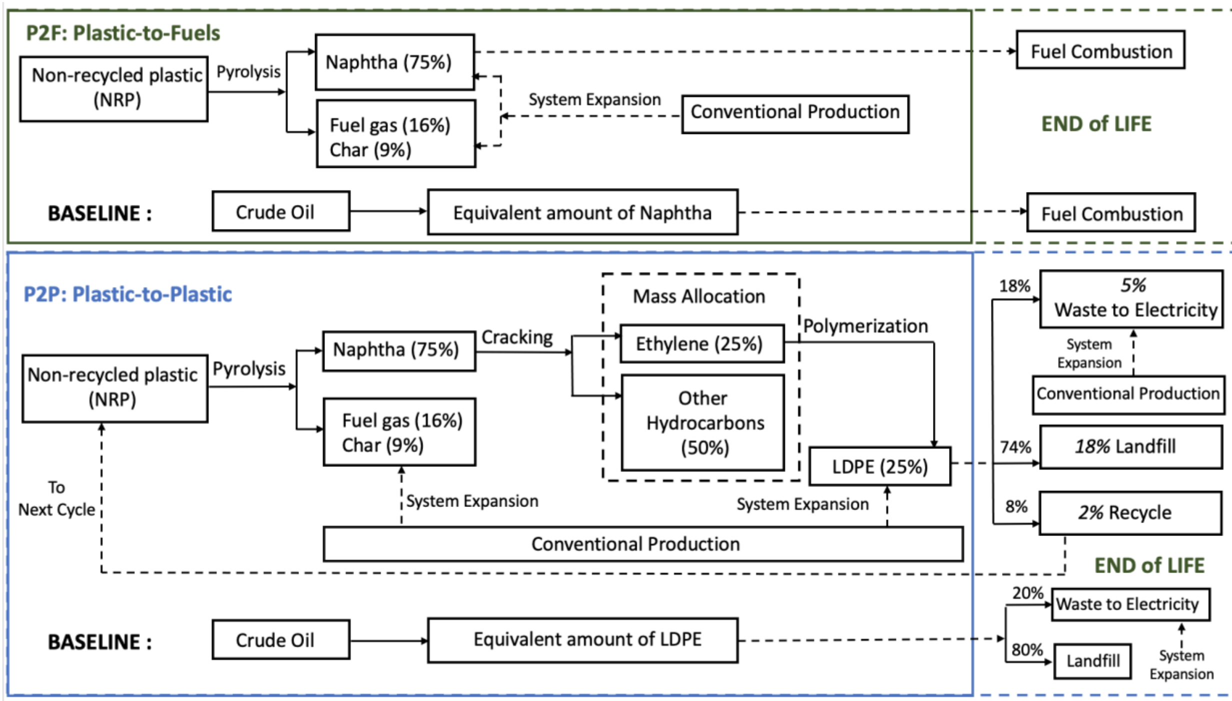

Figure 2. System boundary diagram for a functional unit of per product (MJ naphtha or kg LDPE). The mass shares reported in this figure reflect the US context. Percentages in italics within the end of life portion of the system boundary reflect the percent of mass of the original kg of NRP that advances to this stage. Shares relevant to the EU context are reported in Table 1. The feedstock to the pyrolysis process in the US context is 25 wt% HDPE, 33 wt% LDPE and 42 wt% PP. The feedstock to the pyrolysis process in the EU context is 90 wt% PE, PP and PS and 10 wt% impurities

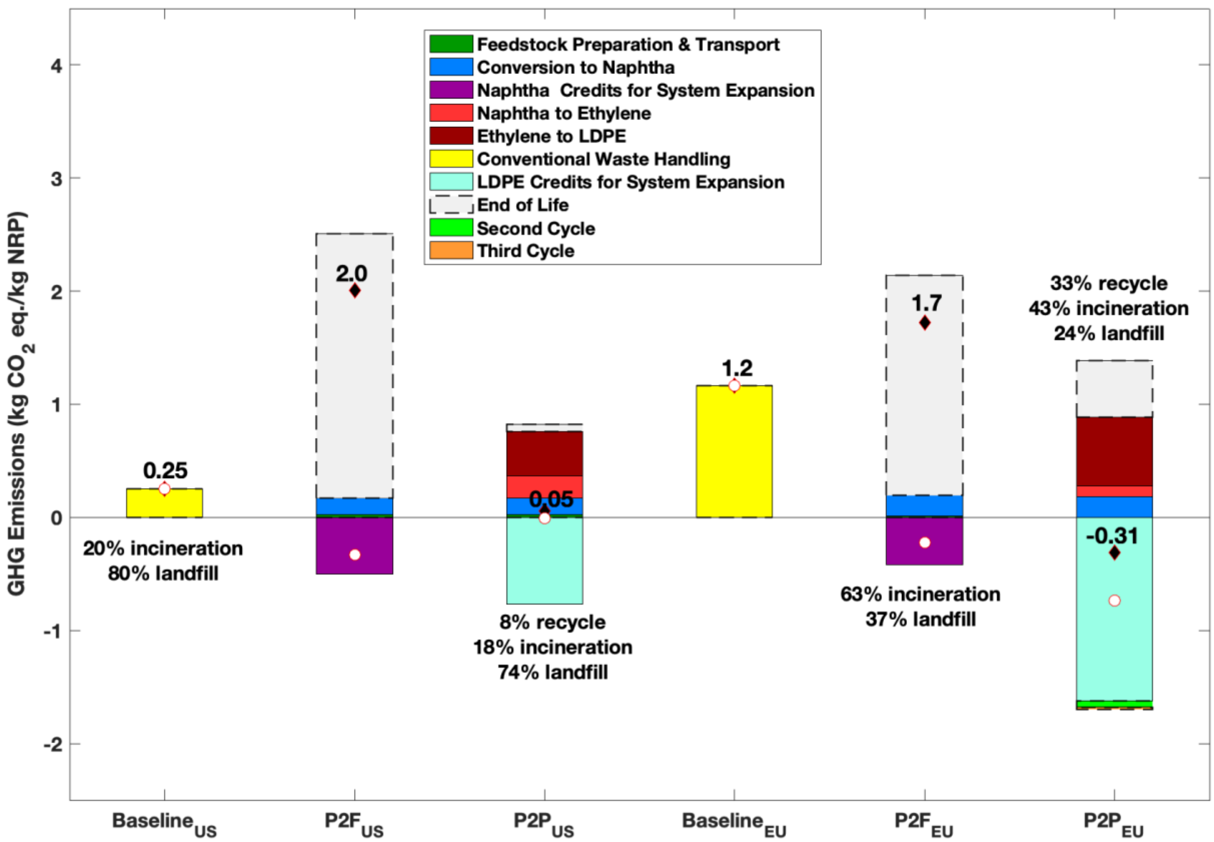

Figure 3. Life-cycle GHG emissions per kg NRP. Diamonds and circles reflect the sum of positive and negative GHG emissions in the system boundary in cradle-to-grave and -gate system boundaries, respectively. The feedstock to the pyrolysis process in the US context is 25 wt% HDPE, 33 wt% LDPE and 42 wt% PP. The feedstock to the pyrolysis process in the EU context is 90 wt% PE, PP and PS and 10 wt% impurities.

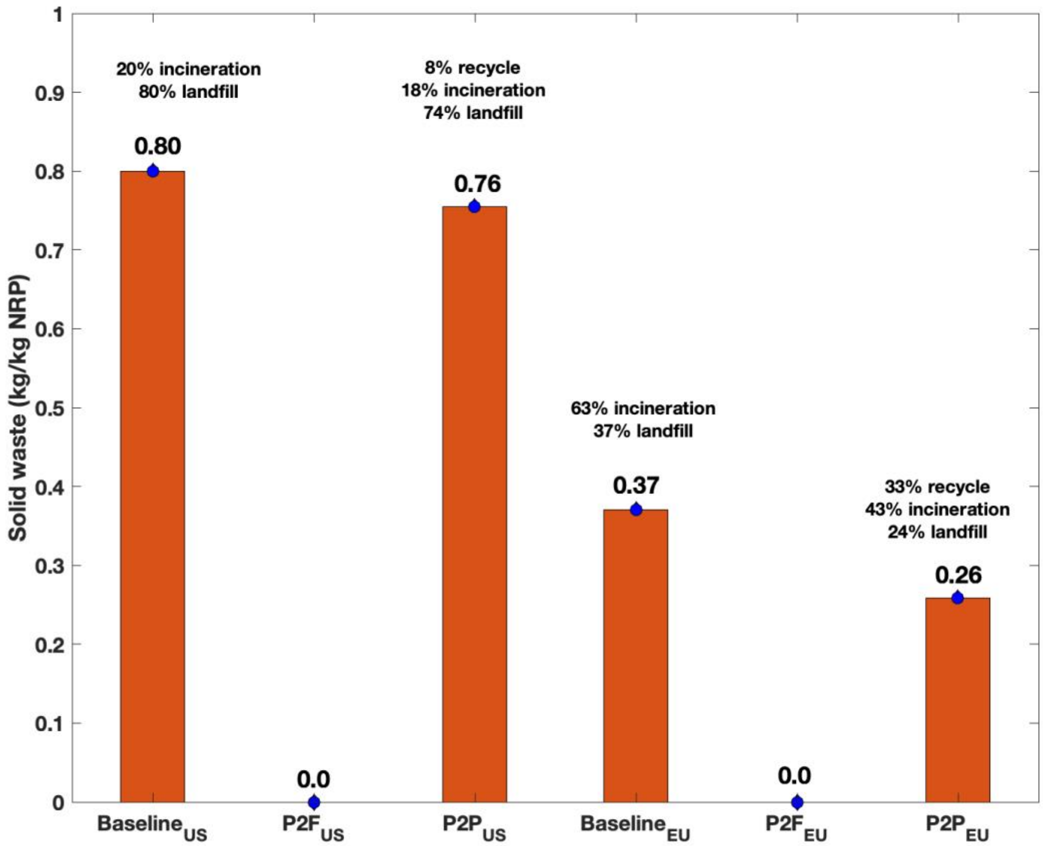

Figure 4: Solid waste generation per kg NRP. There is no solid waste generation for the P2F case.

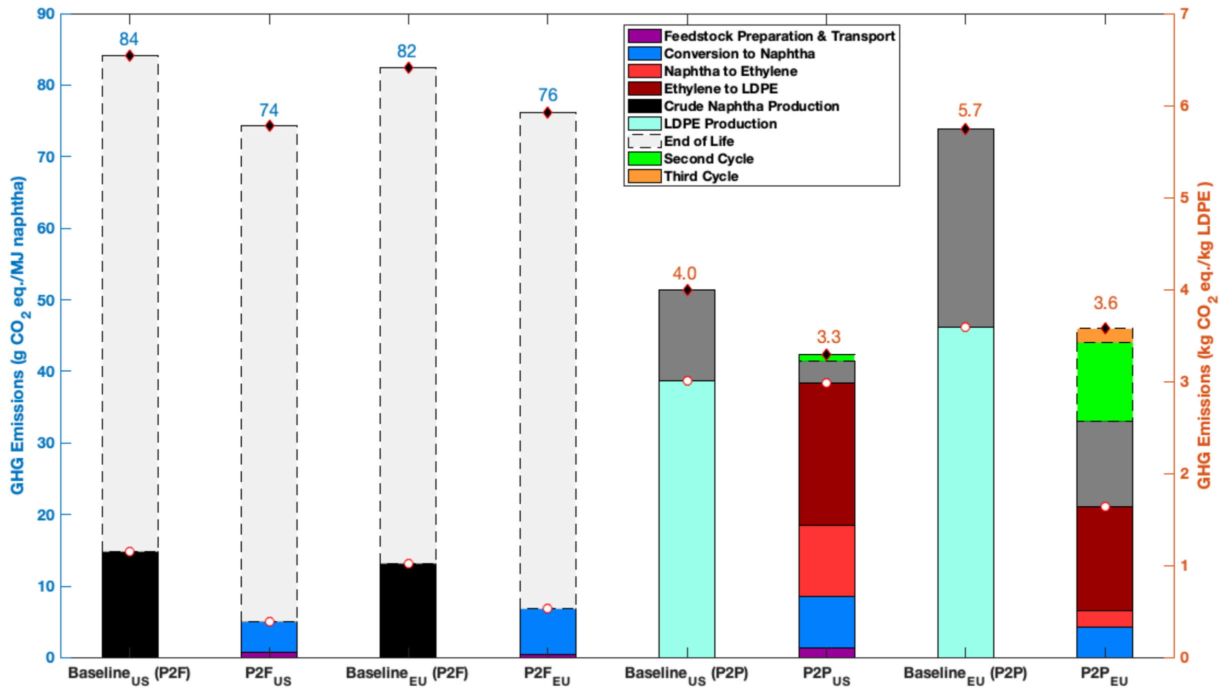

Figure 5. Life-cycle GHG emissions using a product-based functional unit (per MJ for naphtha, per kg for LDPE). Diamonds and circles reflect the sum of positive and negative GHG emissions in the system boundary in cradle-to-grave and -gate system boundaries, respectively. The feedstock to the pyrolysis process in the US context is 25 wt% HDPE, 33 wt% LDPE and 42 wt% PP. The feedstock to the pyrolysis process in the EU context is 90 wt% PE, PP and PS and 10 wt% impurities