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Comparing Parallel Plastic-to-X Pathways and Their Role in a Circular Economy for PET Bottles

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The United States generates the most plastic waste of any country and is a top contributor to global plastic pollution. Multiple end-of-life strategies must be implemented to minimize environmental impacts and retain valuable plastic material, but it is challenging to compare options that generate products with different lifetimes and utilities. Herein, they present a material flow model equipped with consequential life cycle assessment, cost analysis, and a plastic circularity indicator that considers product quality and lifetime. The model is used to estimate the greenhouse gas (GHG) emissions, circularity, and cost of polyethylene terephthalate (PET) bottle mechanical downcycling to lower-quality resin, closed-loop glycolysis to food-grade PET, upcycling to glass fiber-reinforced plastic, and conversion to non-plastic products (electricity, oil) on a United States economy-wide basis for the year 2020. A brute force algorithm suggests that a combination of 68% glycolysis, 11% mechanical recycling, 6% upcycling, 9% landfilling, and 5% incineration can minimize the cost and GHG emissions and maximize the circularity of the current PET economy. However, uncertainty around transportation distances, materials recovery facility efficiencies, and recycling yields can result in different "optimal" pathway mixes. This flexible framework enables informed decision-making to move toward a cost- and environment-conscious circular economy for plastic.

1. Introduction

Global momentum is building toward a circular economy capable of keeping plastics in use and out of waste streams. Given that 79% of all plastic produced since 1950 has accumulated in landfills or the natural environment,[1] rapid implementation of various end-of-life (EoL) management technologies will be needed to reach targets such as those set by the United States (U.S.) Plastics Pact and European Union to achieve 50% plastic packaging recycling or composting by 2025.[2,3] However, it can be challenging to develop an effective plastic EoL strategy when the available options—chemical or molecular recycling, energy recovery, upcycling, downcycling, closed-loop (plastic-to-plastic) or open-loop (plastic-to-*x*) recycling, among others—can generate products ranging from low-grade to virgin-quality plastic and from fuels to value-added chemicals.

Polyethylene terephthalate (PET) is a particularly representative case study for this phenomenon. PET accounted for ≈8% of plastic consumption in 2017 and 14% of plastic waste sent for disposal in 2019 in the U.S., and has many applications in single-use beverage bottles and food packaging.[4,5] Although PET is a relatively small fraction of polymer production, it has ester linkages that enable novel reclamation strategies beyond traditional mechanical recycling, such as chemical or biological depolymerization to monomers or high value chemicals.[6] Several studies have compared the environmental impacts and costs of various closed-loop[7,8] and open-loop[6,9–11] PET recycling technologies on a unit process level, while others have explored the implications of recycling on a plastic economy-wide basis.[12–14] For example, the concept paper by Shonnard et al. described a systems analysis framework for PET and polyolefins,[15] while Chaudhari et al. performed life cycle assessment (LCA) and preliminary systems analysis of PET and polyolefins in the U.S.[16,17] Gracida-Alvarez et al. also created a circular economy sustainability analysis framework combining LCA and material flow analysis (MFA) to evaluate the effect of circular economy strategies on the production of plastic packaging.[8,18] Connecting detailed process-level models with systems analysis can guide decision-making about PET waste management across multiple environmental and economic considerations. Such an approach also enables data-backed holistic insights into the overall plastics landscape.

Here, we present a flexible material flow model capable of analyzing the effects of both plastic-to-plastic and plastic-to-*x* EoL management strategies on the U.S. PET bottle economy in 2020 (**Figure 1**). This Plastic Parallel Pathways Platform (4P) assesses the environmental impacts, costs, and circularity of a PET bottle system in which waste is managed through six potential EoL

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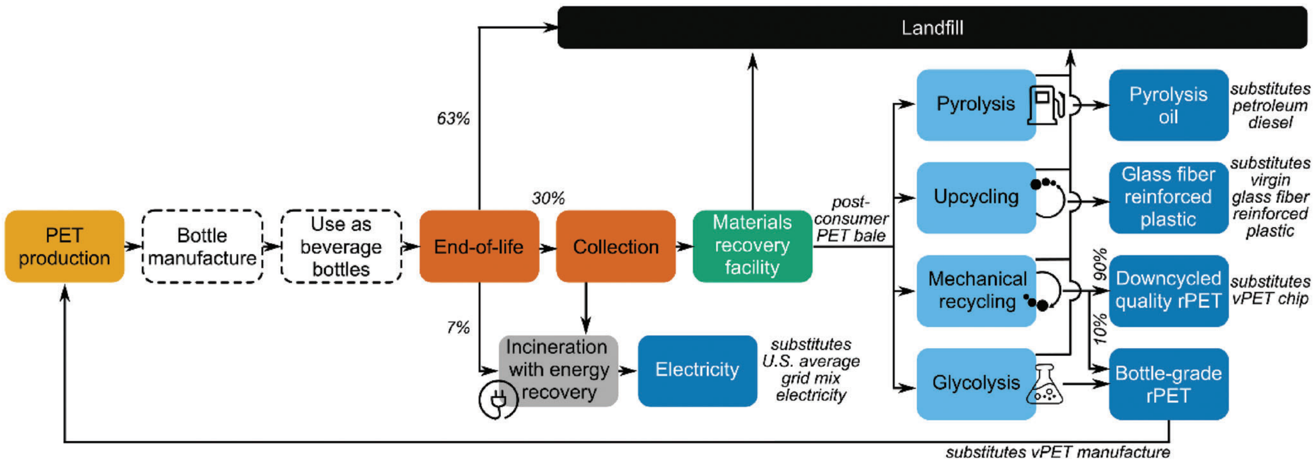

Figure 1. Schematic diagram of the analysis scope. Manufacturing PET into bottles by stretch blow molding, as well as use of the bottles, are not considered. Italicized percentages show PET bottle flows according to our 2020 baseline scenario based on data from the U.S. Environmental Protection Agency[19] and the Association of Plastic Recyclers;[20,21] these can be varied within the model.

pathways: landfill, incineration with energy recovery, pyrolysis to fuel oil, upcycling to glass fiber reinforced plastic (GFRP), mechanical recycling to low-grade PET, and chemical recycling (glycolysis) to food-grade PET. We compare the pathways across several metrics using multi-criteria decision analysis (MCDA) and then use a brute force algorithm to predict an optimal combination of EoL pathways to minimize greenhouse gas (GHG) emissions and costs and maximize circularity. Through its exploration of the PET disposal solution space, this study highlights the need to implement a diverse portfolio of EoL strategies in parallel to enable a PET economy that meets environmental, economic, and circularity requirements simultaneously.

2. Experimental Section

2.1. Previous Framework

The first version of a plastic circular economy modeling (PCEM) tool was presented in Ghosh et al.[12] The goal was to create a holistic, flexible model for analyzing the plastic circular economy that includes relevant economic, technological, and policy variables. The following sections describe the components of PCEM that were maintained in the 4P model used for this study, with further details available in Ghosh et al.[12]

2.1.1. Material Flow Model

The core of the PCEM framework was a material flow model (MFM) that tracks every mass flow in the PET bottle system in the U.S. EoL activities, such as sorting, pretreatment, incineration with energy recovery, landfilling, and recycling, were modeled as unit operations that converted material inputs into single/multiple outputs based on their underlying behavior and modeling function. The MFM connected the flow of materials between all these activities, while also tracking flows leaving the system boundary. All activities included detailed technical data from the literature[7,12,22–25] in order to simulate material conversion, recovery, and loss. The MFM also included a temporal dimension that tracks mass flows with time, enabling prospective analysis with annual resolution from 2020 to 2049. Future PET demand was estimated by linear regression based on historical data from 1991 to 2018 (Figure S1, Supporting Information).[12,26,27]

2.1.2. Life Cycle Assessment

The PyLCA module was used to perform LCA calculations. This python-based framework determined the energy and material inputs from the foreground system, attached them as final demands to the background data (sourced from the U.S. Life Cycle Inventory database)[28] and calculated emissions. The module used the TRACI 2.1 methodology[29] to convert emissions to midpoint environmental impacts. PyLCA can perform LCA of large systems in less than tenths of a second, which enables rapid computation of the thousands of calculations required for 4P.

2.1.3. TEA

The TEA module calculated the cost of processing plastics through different EoL pathways. Manufacturing costs of virgin plastic resin were included to determine the total cost of plastic production, use, and disposal. This enabled users to explore whether displacing a virgin resin with recycled resin production could provide cost benefits to the entire system. Every activity of the framework used a simple TEA model that included the capital and operating costs scaled to mass flow quantities. The operating costs included consumables, utilities, and labor requirements. The capital costs were normalized to the time resolution of the framework. Discounted rate of investment and revenues from final product sales were not considered but will be updated in future versions of the framework.

2.2. Updating The PCEM to Build 4P

There were several limitations to the previous PCEM tool, including difficulty comparing pathways that generated different products, inability to guide decision making for multiple metric considerations, a lack of uncertainty estimates, and insufficient detail around waste collection. The model was therefore modified and upgraded to develop the 4P model utilized for this study, as detailed in the following sections.

2.2.1. Agent Based Model

The original PCEM modeled collection of PET bottles with a linear regression function dependent on various socioeconomic indicators.[12] This simplistic model could not sufficiently account for consumer behavior during the waste collection stage. In contrast, agent based models (ABMs) simulate the behavior of agents (in this study: recycling, wish-cycling, and trash disposal behaviors) and their interactions such that individual decisions by agents result in emergent behaviors for the total system.[30] Agents are heterogenous, and their interactions and results might be discrete, which makes ABMs well-suited to model socio-technical systems.[31] Using an ABM provides several advantages to 4P. First, the model resolution can be preserved at both the agent (micro) level as well as the system (macro) level, conserving the heterogeneity of the population while exploring system behaviors. Second, interventions targeting individuals or the entire system can be studied and compared.[32] Third, ABM captures psychological aspects of decision-making for agents. For this study, results from social-psychology studies were used to set up the decision-making function and the ABM outputs were validated with empirical data on recycling rates. For these reasons, the linear regression model in the previous iteration of PCEM was replaced with the ABM models in 4P. Methodological details of the ABM are provided in Walzberg et al.[33]

2.2.2. EoL Pathways

The glycolysis, mechanical recycling, pyrolysis, and incineration modules already incorporated into the original PCEM were updated with the latest technical data,[7,34] and upcycling to GFRP was added as a pathway.[35] Process inventories and capital and operational costs for these EoL technologies are available in Tables S1–S5 (Supporting Information). The collection, sorting at MRFs, and landfill modules were not changed and are available in Tables S6–S8 (Supporting Information).

2.2.3. Life Cycle Assessment

Due to the nature of the multi-product system being studied with 4P, several challenges arise for determining the Goal and Scope of LCA.

Goal– The objective of this study was to understand the environmental impacts of different EoL pathways and compare them consistently to provide decision-making capabilities. However, the EoL pathways included in 4P produce different products with varied functionalities, lifetimes, and utility, making direct comparison challenging. To maintain consistency, LCA calculations were based on a functional unit of total quantity of PET bottles treated at EoL in the U.S. each year, rather than per unit of product. This enabled comparison of individual EoL pathways as well as combinations of pathways. For the current work, the assumed year is 2020.

Scope– The system boundary of 4P is shown in Figure 1. Activities included were virgin PET resin manufacture by the terephthalic acid route, waste collection, sorting, incineration, landfilling, mechanical recycling, glycolysis, upcycling to GFRP, pyrolysis, and transportation between facilities. Combustion of the pyrolysis oil or of the displaced conventional oil was not considered. 4P included internal as well as external system displacement. Internal system displacement by the recycled content approach occurs when high-quality resin is produced through recycling processes and displaces virgin resin manufacture within the system. This ensures proper accounting of recycled content in the plastic product at each time step. External displacement by the cut-off method refers to materials that leave the system boundary, such as low-grade resin, fuel oil, electricity, and GFRP. The model assumes comparable commodities are displaced by these EoL products and provides corresponding environmental credits to the respective EoL pathways (i.e., negative emissions, Table S9, Supporting Information). This consequential LCA approach enables consistent comparison of open-loop and close-loop recycling technologies regardless of their generated product. Previous studies have similarly used system expansion and consequential LCA to investigate PET down-cycling, high density polyethylene (HDPE) pyrolysis, and municipal solid waste management.[36–38]

2.2.4. Plastic Circularity Index (PCI)

The PCI for assessing circularity of the plastic system was based on the Ellen MacArthur Foundation's Material Circularity Index (MCI).[39] A Linear Flow Index (LFI) was first calculated by dividing the amount of virgin PET landfilled in a given year by the total amount of PET treated at EoL in that year. In other words, only landfilled material was considered a linear flow. A utility factor (X) was then applied to account for upcycling or down-cycling. X considers either the lifetime of the EoL product in comparison to virgin PET (electricity and oil have shorter lifetimes, whereas GFRP has a longer lifetime) or the quality of the product in comparison to virgin PET (PET recycled by glycolysis was assumed to have equal quality, while PET from mechanical recycling has lower quality). Utility factors for all scenarios and their justifications are available in Table S10 (Supporting Information). X and LFI are combined to calculate PCI using the MCI formula (Equation 1).

$$PCI = 1 - (LFI \times \left(\frac{0.9}{X} \right) \tag{1}$$

When multiple EoL pathways are switched "on" in the model simultaneously (i), PCI is calculated for each pathway, multiplied by the fraction of PET going to each individual pathway (f_i), and then summed for a final mass-weighted PCI value (Equation 2).

$$\text{weighted } PCI = \sum f_i PCI_i \tag{2}$$

2.2.5. MCDA

GHG emissions, cost, and PCI were selected as criteria for MCDA according to the Analytic Hierarchy Process.[40] GHG emissions and cost are normalized on a zero to one scale ($\bar{x}$) by dividing the value for a given scenario (x_n) by the maximum value (x_{max}) across all scenarios which are being compared, according to Equation. 3. PCI was left as is because it already utilizes a zero to one scale.

$$\bar{x} = 1 - \frac{x_n}{x_{max}} \tag{3}$$

The normalized criteria were then weighted according to four different priorities. For equal weighting, normalized GHG emissions, normalized cost, and PCI were each assigned a 33.3% weighting and summed for a final score out of one, where zero is "worst" and one is "best". For GHG prioritization, normalized GHG emissions were assigned an 80% weighting, while normalized cost and PCI were each set to 10% weighting. For cost prioritization, normalized cost was given an 80% weighting, and normalized GHG emissions and PCI were each assigned a 10% weighting. Lastly, for circularity prioritization, PCI was set at 80% weighting, while normalized GHG emissions and normalized cost were each assumed to have a 10% weighting.

2.2.6. Design of Pathways

4P can support the design of plastic recycling systems by determining the fraction of resin that should be diverted to various EoL options to meet a certain goal. The goal can be focused on a single indicator (e.g., low cost) or a combination of indicators through MCDA. 4P employed a solution space exploration methodology where the simulation was run 1000 times for different combinations of design variables. The solutions were plotted to determine the best possible design case, sketching the probable pareto front and analyzing tradeoffs between indicators as well as EoL designs. Thus, rather than taking a pure optimization route, 4P explored the design space by utilizing a rapid Monte Carlo analysis. While this approach is not guaranteed to find the globally optimal design solution as an infinite number of simulations would be required to explore the entire solution space, it nevertheless provides the user with a general idea of good system designs for meeting environmental, cost, and/or circularity targets. The benefit of using solution space exploration rather than an optimization algorithm comes from the lower computation requirement as well as facile incorporation of parameters' uncertainty.

2.2.7. Exploration of Solution Space Without Uncertainty

The 4P framework can define the solution space using the Monte Carlo scenario analysis described above without accounting for uncertainty. For these simulations, all parameters are fixed to their baseline values (Table S11, Supporting Information) and only the diversion of waste PET to the different EoL pathways is randomly varied using a uniform distribution between the upper and lower bounds (Table S11, Supporting Information). For mechanical recycling and upcycling, the upper bounds were constrained to 0.7 and 0.3, respectively, due to market demand for low-quality PET chip and GFRP being lower than production capacity (Table S12, Supporting Information). Incineration, glycolysis, and pyrolysis were all permitted to vary between zero and one.

2.2.8. Exploration of Solution Space under Uncertainty

4P can also design systems while accounting for uncertainty in the system parameters. For this analysis, upper and lower bounds were provided for the parameters from expert guidance and literature review (Table S11, Supporting Information). A triangular distribution was assumed for each of these parameters as there was insufficient data to predict a more accurate probability distribution function. For each run of 4P, a sample was randomly drawn from the distribution of the parameters as well as from the uniform distribution of design variables (i.e., EoL pathways). Repeating this procedure results in exploration of the solution space under uncertainty of system parameters.

3. Results and Discussion

3.1. Comparison of EoL Pathways

A variety of existing and emerging EoL pathways for PET bottles were selected for study. Given that an estimated 76% of PET waste is landfilled in the U.S., 9% is combusted, and 15% is recycled,[5] landfilling, incineration with energy recovery, and mechanical recycling were included as current at-scale disposal pathways. Mechanical recycling—that involves plastic shredding, washing, and extrusion into pellets—was assumed to yield lower quality PET that must be down-cycled into non-bottle products such as trays or textiles.[7,36] Emerging chemical recycling technologies were encompassed by glycolysis and pyrolysis. Glycolysis is a closed-loop recycling technique in which PET is depolymerized in the presence of ethylene glycol (EG) at elevated temperatures to the oligomer bis(2-hydroxyethyl) terephthalate (BHET), which can be subsequently repolymerized into virgin-quality PET.[6] Glycolysis was selected for inclusion here as it has been previously shown to be more economically and environmentally viable than other depolymerization strategies such as methanolysis or enzymatic hydrolysis.[7] Pyrolysis involves the heating of waste plastic in the absence of oxygen to produce fuels or platform chemicals and is currently undergoing industrial scale-up.[41] While more suitable to polyolefins (due to a lack of oxygen or other heteroatoms in their polymeric structure), pyrolysis can also convert PET into fuels, albeit at a relatively low yield and quality.[42,43] Lastly, PET upcycling to a variety of higher value products has been reported. Here, we selected upcycling to GFRP due to the availability of process inventory data and the relevance of this product to wind turbine energy targets.[35] In this chemical process, PET is first depolymerized in the presence of ethylene glycol to a lower

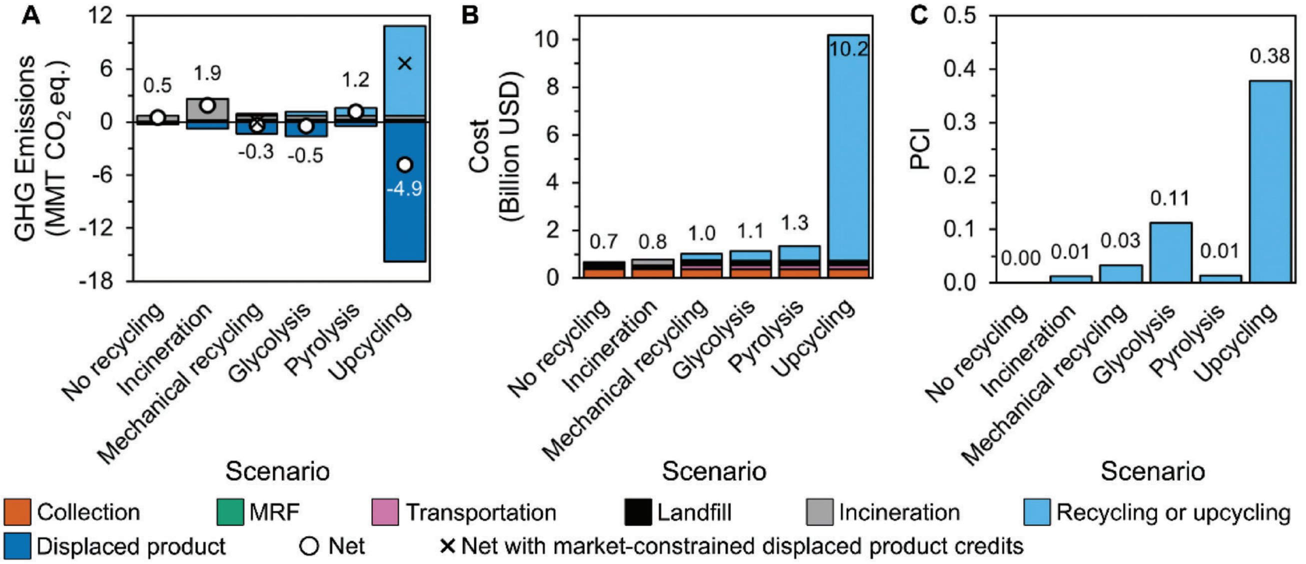

Figure 2. Comparison of the A) GHG emissions, B) cost, and C) PCI if all PET bottles collected for recycling in the United States in 2020 were used to produce lower-quality PET resin by mechanical recycling, food-grade PET resin by glycolysis, electricity from incineration, fuel oil from pyrolysis, or fiber-reinforced resin by upcycling. Circles indicate net results for the displacement approach (i.e., credits are given for avoided virgin manufacture of the respective product). The black x's indicate net results when credits are capped at the market demand for PET chip (mechanical recycling) and GFRP (upcycling) in 2020. Raw data are available in Tables S14, S16, and S10 (Supporting Information).

molecular weight unsaturated polyester, which is then blended with bio-based chemicals (malate, fumarate, and dimethyl muconate) and fiberglass to produce GFRP.[35]

The 4P model was applied to assess the cost, environmental impacts, and circularity of these EoL pathways on a U.S. economy-wide basis. We assumed that 100% of all PET bottles collected for recycling was sent to a single technology: landfill (no recycling scenario), incineration with energy recovery, or sorting at a MRF followed by mechanical recycling, glycolysis, pyrolysis, or upcycling (**Figure 2**). Of all PET bottle waste requiring disposal, only 30% was collected for these EoL pathways, while 63% was sent to landfill and 7% to incineration with energy recovery in accordance with 2016—2018 data from the U.S. Environmental Protection Agency and the Association of Plastic Recyclers.[19–21] The functional unit of our analysis is the total quantity of PET bottles treated at EoL in 2020 (3.24 MMT, Figure S1, Supporting Information). The 30% of these bottles that are assumed to be diverted from landfill (0.97 MMT) can generate 0.47 MMT low-grade PET chip if sent exclusively to the mechanical recycling pathway, 0.67 MMT food-grade PET if only glycolysis is selected, 0.63 MMT fuel oil by the pyrolysis pathway, 7.28×10^8 kWh from incineration, or 2.87 MMT GFRP via upcycling, as calculated from the life cycle inventories reported in Tables S1–S8 (Supporting Information). Material flows for these scenarios are available in Figure S2 (Supporting Information).

GHG emissions were selected to represent environmental impacts given the increasing global focus on decarbonization and climate change mitigation.[13] Additional environmental metrics such as acidification or carcinogenic toxicity are also calculated by 4P and are available in Table S13 (Supporting Information). Figure 2 shows the GHG emissions for each EoL scenario (raw data in Table S14, Supporting Information). Of the EoL scenarios, upcycling provides the lowest environmental impact at −4.9 MMT CO_2 eq per year due to its displacement of energy-intensive virgin GFRP (see virgin GFRP inventory in Table S15, Supporting Information). However, this scenario produces 3.5 times more GFRP than is currently consumed in the U.S., according to 2019 data from industry databases (Table S12, Supporting Information);[44] constraining the displacement credits to market demand would increase GHG emissions to 6.3 MMT CO_2 eq, which is at least three times higher than the impacts of the other EoL pathways. Glycolysis offers the second lowest GHG emissions (−0.46 MMT CO_2 eq), followed by mechanical recycling (−0.34 MMT CO_2 eq). Glycolysis and mechanical recycling both generate less CO_2 than the virgin PET that they displace, resulting in net negative emissions. The glycolysis process is approximately twice as impactful as mechanical recycling due to its organic solvent and energy use, but its higher recyclate yields and quality enable the displacement of more virgin PET (20% of total virgin PET bottle manufacture in the U.S. in 2020). In contrast, mechanical recycling has a slightly lower yield and generates lower quality resin that requires the manufacturer to reject a certain percentage of incoming recyclate, resulting in fewer displacement credits (see Experimental Section Section 2.2.3). Both chemical and mechanical recycling compare favorably to landfilling (0.53 MMT CO_2 eq) and incineration with energy recovery (1.9 MMT CO_2 eq). Incineration with energy recovery emits more CO_2 than conventional electricity generation (2020 U.S. grid mix assumed) because PET has a lower energy density than fossil fuels and because a portion of the U.S. grid mix is sourced from renewable energy.[45] The credits applied for electricity displacement do not counteract the EoL environmental impact; therefore, incineration exhibits net positive GHG emissions. Pyrolysis also has high GHG emissions (1.2 MMT CO_2 eq) because it is an energy-intensive process and has a low oil product yield (an optimistic 65% yield is assumed).[42] There are many uncertain parameters in 4P and so the rankings provided here—especially for scenarios with particularly close GHG emissions such as glycolysis and mechanical recycling—may not be statistically significant.

From an economic perspective (Figure 2B; Table S16, Supporting Information), landfill has the lowest estimated cost at 0.7 billion U.S. dollars (USD), followed by incineration (0.8 billion USD), mechanical recycling (1.0 billion USD), and glycolysis (1.1 billion USD). Pyrolysis—due to high energy use and capital cost—and upcycling—due to expensive fiberglass—have higher costs at 1.3 and 10.2 billion USD, respectively. Collection and transportation between EoL facilities accounts for 38–61% of costs for all pathways except for upcycling (Table S16, Supporting Information). This relatively high percentage is due to long assumed transportation distances (100 km from curbside bin to landfill, incinerator, or MRF, and 600 km from MRF to recycler)[12] and high collection costs ($100/ton).[22] Sortation at a MRF is the least economically impactful step, at 1–2% of total EoL costs for mechanical recycling, glycolysis, pyrolysis, and upcycling (Table S16, Supporting Information). The recycling or upcycling process itself accounts for 49–95% of total EoL costs (Table S16, Supporting Information). Unlike with LCA, displacement of costs (e.g., credits due to producing electricity from incineration rather than conventional generation) is not included in the current version of 4P. For example, the closed-loop nature of glycolysis could enable direct integration with PET manufacture, reducing virgin feedstock demand for bottles by 20% and thereby lowering the associated costs given that glycolysis is estimated to be less expensive than conventional PET production from virgin terephthalic acid and ethylene glycol.[7] However, these reduced costs do not necessarily correlate with decreased market prices as higher profit margins may be justified, and it is therefore challenging to apply cost credits.

Circularity (Figure 2C; Table S10, Supporting Information) is also a relevant indicator to consider given the multitude of plastic circular economy targets set by the U.S. Plastics Pact, European Union, and others.[2,3] Here, PCI is measured on a zero to one scale in which a higher value is considered “better.” Upcycling is estimated to have the best PCI of 0.36 because the expected lifetime (20 years)[46] and corresponding utility factor of GFRP are significantly higher than those of PET bottles. Glycolysis is assumed to generate virgin quality, food-grade PET and has a PCI of 0.11, while mechanical recycling has a lower PCI of 0.03 given that >90% of the produced recyclate is assumed to be of lower quality than virgin PET.[7] Pyrolysis and incineration generate short-lived energy products with lower utility factors (0.65 and 0.6, respectively, on a zero to one scale), and therefore have low PCIs that only offer an improvement over the “no recycling” scenario in which all EoL plastic is sent to landfill.

The GHG emissions, cost, and PCI results were next summed into MCDA scores ranging between zero (the “worst”) and one (the “best”). An equal weighting of the three metrics was selected for a relatively neutral perspective on PET EoL pathways (**Figure 3**; Table S17, Supporting Information, Experimental Section Section 2.2.5). MCDA conducted with prioritization of only GHG emissions, only cost, or only PCI are available in Figure S3 (Supporting Information) and exhibit trends in agreement with those observed for the single metrics in Figure 2. Under equal weighting, glycolysis and upcycling have the highest

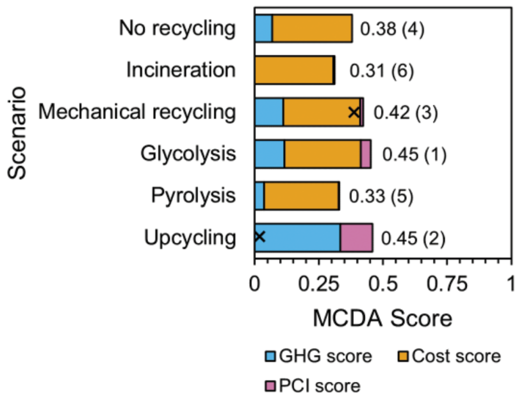

Figure 3. MCDA comparing PET EoL pathways for equal weighting of GHG emissions, cost, and PCI. The black x's indicate MCDA scores for mechanical recycling and upcycling when GHG emission displacement credits are capped at the market demand for PET chip and GFRP, respectively. Raw data are available in Table S17 (Supporting Information).

MCDA scores at 0.45, followed by mechanical recycling at 0.42, no recycling (landfilling) at 0.38, pyrolysis at 0.33, and incineration at 0.31. (Figure 3; Table S11, Supporting Information). As discussed for GHG emissions, if displacement credits for GFRP are limited to market demand, the upcycling MCDA score reduces to 0.02. All scenarios exhibit tradeoffs across metrics: mechanical recycling has low GHG emissions, but higher costs relative to no recycling and poor PCI; incineration has lower costs than recycling scenarios, but higher GHG emissions and lower PCI; and upcycling has the lowest GHG emissions and highest PCI of all scenarios, but also the highest cost. The EoL pathways with the best balance of all metrics—glycolysis and mechanical recycling—therefore emerge as the most favorable, although the close scores across most scenarios could indicate that there is no “best” technology under the assessed criteria. Furthermore, the uncertainty of such MCDA estimates is high.

3.2. Optimization of EoL Pathway Combinations

Since no single EoL technology enables simultaneous low cost, low GHG emissions, and high circularity, we developed a brute force algorithm to uniformly vary system parameters over 1000 runs and determine a local optimum that minimizes GHG emissions, cost, and maximizes PCI (**Figure 4**). This local optimum was selected by identifying the scenario with the highest MCDA score under equal weighting. Circularity is depicted in Figure 4 as 1−PCI so that the most optimal values are the closest to the graphical origin. This method determines a local minimum only and there may be other scenarios that would be more optimal but are not captured in the 1000 runs. Two experiments—one varying only the ratio of PET bottles sent to the EoL pathways (Experimental Section Section 2.2.7) and the other varying all parameters in the 4P system (Experimental Section Section 2.2.8)—were conducted.

We first varied the ratio of waste PET bottles sent to a given EoL pathway (Figure 4A-C). The ratio was permitted to vary from 0 to 1 for incineration, glycolysis, and pyrolysis, but mechanical recycling and upcycling were constrained to maxima of 0.7 and

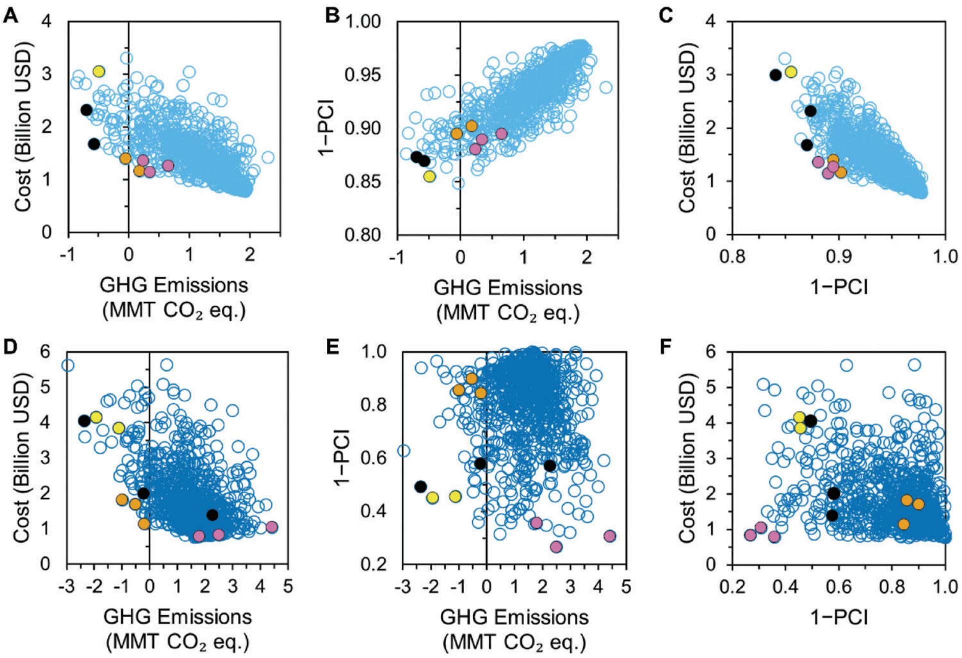

Figure 4. Brute force algorithm results for A–C) optimization of EoL pathway mix only, and D–F) optimization of EoL pathway mix and all uncertain parameters. The black markers indicate the top three scenarios optimized for low cost, low GHG emissions, and high PCI (depicted as low 1−PCI on the graphs); orange markers indicate the top three scenarios that are optimized for low cost and low GHG emissions; yellow markers indicate the top three scenarios that are optimized for low GHG emissions and high PCI; pink markers indicate the top three scenarios that are optimized for low cost and high PCI.

0.3, respectively, to account for limited market demand for their products (Table S12, Supporting Information). Of the 30% of PET bottle waste collected for EoL processing, the optimal mix of pathways is predicted to be 68.4% glycolysis, 11.4% mechanical recycling, 5.9% upcycling, and 5.1% incineration, with 8.5% sent to landfill due to process losses (**Figure 5**A; Table S18, Supporting Information). On a per total EoL PET bottle basis these ratios correspond to 65.6% landfill, 8.5% incineration, 3.4% mechanical recycling, 20.5% glycolysis, and 2.0% upcycling (Figure 5B; Table S12, Supporting Information). This scenario would reduce GHG emissions to −0.58 MMT CO_2 eq relative to no recycling (0.53 MMT), increase costs by 2.5 times (1.7 billion USD versus 0.67 billion USD), improve PCI to 0.13, and reduce virgin bottle demand by 16% (Table S18, Supporting Information). The selection of glycolysis, mechanical recycling, and upcycling is understandable since these technologies offered the top three equal-weighting MCDA scores in Figure 3. However, the higher proportion of mechanical recycling than upcycling is unexpected given the previously discussed MCDA rankings. It is important to note that when combining multiple EoL pathways, a single pathway does not necessarily need to exhibit the best performance for all metrics, so long as the *combination* of pathways facilitates a balance of GHG emissions, cost, and PCI. Mechanical recycling has significantly lower costs than upcycling, which may complement the higher circularity and lower GHG emissions of glycolysis and upcycling under the reported ratios. It is also possible that our 1000-run exploration did not capture a more optimal scenario with a higher upcycling ratio.

Given that PCI is the most uncertain metric in this work due to the subjective nature of the utility factor, the system could also be optimized exclusively for minimal GHG emissions and cost. The resulting mix of 44% glycolysis, 28% mechanical recycling, 17% incineration, 8% landfill, 2% upcycling, and 1% pyrolysis on a per collection basis would reduce GHG emissions to −0.08 MMT CO_2 eq relative to no recycling, increase costs by 93% (1.29 billion USD), improve PCI to 0.09, and decrease virgin PET bottle demand by 10% (Table S18, Supporting Information). The least optimal scenario identified by this algorithm would be a combination of 52% pyrolysis, 20% incineration, 13% mechanical recycling, 7% landfilling, 4% upcycling, and 3% glycolysis, which would increase GHG emissions by 3.7 times (1.99 MMT CO_2 eq) relative to no recycling, increase costs by 2.3 times, and provide a PCI of 0.04.

The brute force algorithm was next applied under uncertainty (Figure 4D–F). A total of 34 uncertain parameters, including transportation distances between facilities, separation efficiencies of MRF sorting equipment, yields of the mechanical recycling, glycolysis, pyrolysis, and upcycling processes, were permitted to vary within set ranges determined by expert judgement (Table S11, Supporting Information). Because many variables were changed simultaneously—not just EoL pathway mix, as in the previous experiment—the circles in Figure 4D–F cannot be compared directly. Furthermore, given that all combinations of these parameters and EoL pathways could not be captured in 1000 runs, there are likely many combinations that may prove more optimal than the mixes reported here.

Under uncertainty, the local optimum for a maximal equal-weighting MCDA score is estimated to be 66% glycolysis, 12% mechanical recycling, 8% landfilling, 5% incineration, 5% pyrolysis, and 4% upcycling (Figure 5C; Table S19, Supporting

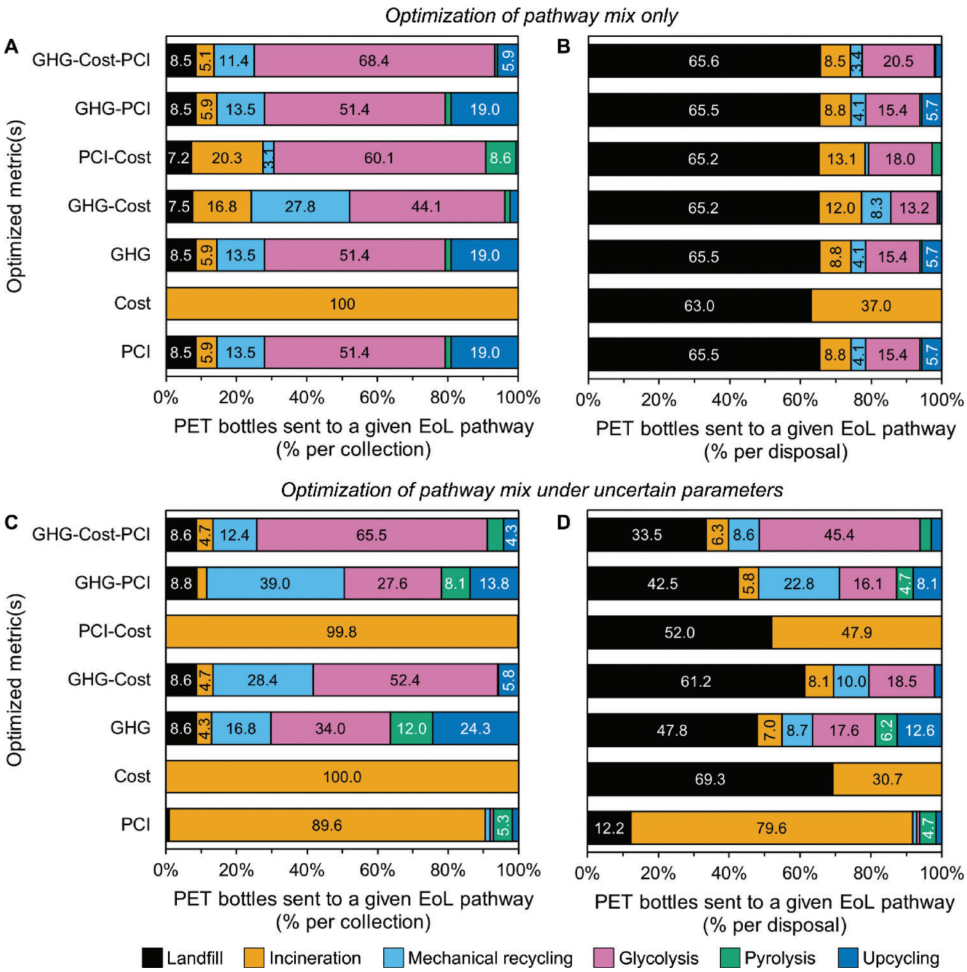

Figure 5. Predicted optimal EoL pathway mixes in 2020 when only the ratio of PET waste sent to a given EoL pathway is varied under the brute-force algorithm, shown per amount of PET bottles A) collected for recycling, or B) treated at EoL. Predicted optimal EoL pathway mixes when all model uncertain parameters are varied under the brute-force algorithm, shown per amount of PET bottles C) collected for recycling, or D) treated at EoL. Raw data are available in Tables S18 and S19 (Supporting Information). Other altered parameters for the scenarios shown in C and D are listed in Table 1 and Table S20 (Supporting Information).

Information). If these ratios are adjusted to a per total EoL PET basis, they correspond to 45% glycolysis, 9% mechanical recycling, 34% landfilling, 6% incineration, 3% pyrolysis, and 3% upcycling (Figure 5D; Table S19, Supporting Information). Additional parameter changes sampled in this scenario include increased collection rate (69% in comparison to 30% in our baseline), increased transportation distance to MRF, reduced transportation distances to incineration and landfill, improved glycolysis, upcycling, and pyrolysis yields, and both increases and decreases to certain MRF sorting equipment efficiencies (**Table 1**; Table S20, Supporting Information). This scenario would generate −1.79 MMT CO_2 eq, cost 2.65 billion USD, and have a PCI of 0.47. Previous work has similarly found that combinations of mechanical and chemical recycling can help reduce virgin plastic demand and environmental impacts, although those studies did not consider cost.[8, 17]

When neglecting PCI and optimizing exclusively for minimum GHG emissions and cost, the predicted combination of pathways is 52% glycolysis, 28% mechanical recycling, 9% landfilling, 6% upcycling, and 5% incineration. The parameters sampled in this scenario also include increased collection rate (to 35%), decreased distances to incineration and MRF, improved upcycling, pyrolysis, and glycolysis yields, as well as changes in certain MRF sorting equipment efficiencies. In this case, GHG emissions, costs, and PCI are predicted to be −1.0 MMT CO_2 eq, 1.8 billion USD, and 0.14, respectively. The least optimal scenario identified by the algorithm would be 9% landfilling, 5% incineration, 13% mechanical recycling, 13% glycolysis, 41% pyrolysis, and 20% upcycling (per collection basis) while simultaneously increasing all transportation distances, increasing collection rate, and decreasing all recovery yields. This scenario would emit 0.84 MMT CO_2 eq, cost 4.4 billion USD, and have a PCI of 0.24.

The optimal results under uncertainty are similar to those reported without uncertainty, suggesting that this optimization method is robust. Nevertheless, the ratio of mechanical recycling tends to be slightly higher in the uncertain versus non-uncertain scenarios, whereas that of glycolysis tends to be lower. Although it

Table 1. List of altered parameters for the scenarios optimized under uncertainty (shown in Figure 5C,D). Given the large number of parameters related to MRF efficiency, only those that have been varied by more than ±20% are listed here; a complete list of variables and their % variation is available in Table S20 (Supporting Information).

Optimized Scenario	Change in Collection Rate	Change in Transportation Distances	Change in MRF Efficiency	Change in Recycling Efficiency
GHG-Cost-PCI	+131%	• +34% MRF distance • −25% incineration distance	• +94% cardboard disc screen 2 • +36% film disc screen 2 • +63% paper vacuum • +42% breaker glass • +39% cardboard vacuum • −22% NIR PET-glass • −93% NIR PET-film	• +23% pyrolysis yield • +23% upcycling yield
GHG-Cost	+18%	• −23% incineration distance • −29% MRF distance	• +56% paper disc screen 1 • +25% film disc screen 2 • −23% NIR PET-cardboard • −93% NIR PET-film	• +56% upcycling yield
GHG-PCI	+95%	• −24% MRF distance	• +34% cardboard disc screen 2 • +32% paper disc screen 2 • +30% NIR PET-other • −21% NIR PET-cardboard • −91% NIR PET-film	• +25% pyrolysis yield
PCI-Cost	+41%	• +24% incineration distance	• +55% cardboard disc screen 2 • +48% paper vacuum • +27% NIR PET-aluminum • −21% paper disc screen 1 • −26% film disc screen 2 • −84% NIR PET-film	• All changes <±20%
GHG	+73%	• −50% MRF distance	• +32% cardboard vacuum • +21% NIR PET-cardboard • −21% NIR PET-aluminum • −22% paper disc screen 1 • −86% NIR PET-film	• +51% upcycling yield
Cost	−23%	• All changes <±20%	• +51% cardboard vacuum • +35% paper disc screen 1 • +29% cardboard disc screen 2 • +26% NIR PET-aluminum • +24% paper vacuum • −22% NIR PET-other • −26% NIR PET-cardboard • −89% NIR PET-film	• All changes <±20%
PCI	+191%	• +40% MRF distance • +29% incineration distance	• +69% paper disc screen 1 • +54% cardboard disc screen 2 • −28% film disc screen 2 • −84% NIR PET-film	• +33% upcycling yield

is challenging to deconvolute the compounding effects of multiple variables, one reason for this effect may be that changes in certain parameters such as MRF and reclaimer transportation distances will have a stronger effect on mechanical recycling or glycolysis, or vice versa (see sensitivity analysis in Figures S4–S10, Supporting Information). The MRF efficiency alterations listed in Table 1 are not expected to significantly affect the results as only the non-target material efficiency for each piece of equipment has changed. For example, disc screen 1 is primarily used to separate out cardboard; increasing its separation efficiency for non-target paper will therefore have a minimal impact on yields and contamination rates.[12–23] Another exception is the dramatic increase in incineration recommended for the "PCI–Cost" and "PCI" optimizations under uncertainty. This result is likely because the PCI of incineration increases with increasing collection rate at a steeper slope than other EoL pathways, eventually surpassing glycolysis at collection rates greater than 67% (Figure S11, Supporting Information). The PCI assumes that only landfilled PET is a linear flow; incineration sends virtually no material to landfill due to its high efficiency in contrast to mechanical recycling or glycolysis, where up to 40% of incoming plastic can be landfilled due to process losses at both the MRF and the reclaimer.

In a real-world situation, it is unlikely that a MRF or reclaimer would want to decrease the efficiency of its sorting equipment or recycling process, or to source postconsumer PET bottles from locations that require longer transportation distances. Certain scenarios in which efficiencies are only improved might therefore prove more insightful for future implementation. The results also showcase the high variability of the PET economy; technology mixes that are ideal for one set of parameters may no longer be optimal upon variation of those parameters. Access to less uncertain datasets around collection rates, transportation distances, and MRF and reclaimer efficiencies will be necessary to reliably guide decision-making for a PET circular economy.

While not discussed in this study, 4P calculates a full suite of life cycle indicators including acidification, ecotoxicity, eutrophication, carcinogenic and non-carcinogenic impacts on human health, ozone depletion, and smog formation. Incorporating these additional metrics into MCDA and the brute-force algorithm could significantly alter the recommended pathway mix due to disadvantages and benefits that are not currently captured. Future iterations of 4P could also incorporate additional polymers such as polyethylene (28% and 52% of U.S. plastic consumption and disposal, respectively),[4, 5] polypropylene (16% and 19%),[4,5] and biodegradable plastics like polylactic acid, as well as reuse strategies. Furthermore, a future version of 4P could consider the feasibility of deploying multiple PET bottle EoL pathways, as well as any technological, infrastructure, or policy changes that may facilitate this transition to a circular economy. For example, while mechanical recycling is already the industry standard, it would still benefit from higher yields and material quality that could be obtained through investment in improved infrastructure (e.g., for sorting).[7, 47] Glycolysis is currently being explored at pilot scale by several companies,[6] while upcycling to GFRP is at a low technology readiness level (TRL) at laboratory scale.[35] Their deployment at industrial scale will therefore occur on longer timescales than established routes like mechanical recycling, incineration, or landfill, but also offers more opportunities for innovation (e.g., novel reaction mechanisms, higher product yields).[7] EoL pathways should be complementary—with PET bottle waste of varying levels of contamination sent to different technologies—but competition may arise around limited post-consumer PET supply. Collection of PET bottle waste is a key bottleneck for any EoL strategy except incineration or landfilling,[33] and thus improvements in disposal behavior and collection infrastructure will be crucial to implement a circular plastic economy.

4. Conclusion

With a global supply chain, dozens of polymer and product formulations, and challenging EoL logistics, the plastics economy is complex. Addressing plastic waste management and promoting circularity will therefore require a correspondingly complex solution, as no single strategy will fulfill all needs. Alongside reduction and reuse mechanisms, recycling and recovery strategies are expected to play a crucial role in addressing the plastic problem. The present work aimed to compare a selection of PET bottle EoL technologies across GHG emissions, cost, and circularity metrics on a U.S. economy-wide basis. To account for the different value (plastic or energy) products generated by these EoL options and enable consistent comparison, we adapted our existing material flow model to include consequential LCA, PCI, and MCDA. The MCDA results suggested that glycolysis, mechanical recycling, and upcycling to GFRP would be the most advantageous. Furthermore, a brute force algorithm was used to identify optimal combinations of EoL pathways that minimize cost and GHG emissions and maximize circularity, both without and with consideration of the uncertainty of key parameters such as transportation distances, MRF efficiencies, and recycling yields. 4P offers a powerful platform for identifying tradeoffs and synergies across plastic-to-x technologies that are not typically directly comparable, enabling the transformation of a complex PET bottle EoL system through actionable insights for both researchers and decision-makers.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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circular economy, life cycle assessment, plastics, polyethylene terephthalate, recycling

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