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# Minimum GHG emissions and energy consumption of U.S. PET and polyolefin packaging supply chains in a circular economy†

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There is a wide agreement on the urgency of transforming linear management of plastics towards a circular economy model. However, no clear pathways exist as to required recycling technologies involved and system-wide environmental impacts. This study explores such pathways in the U.S. for the most commonly used packaging plastics through a combination of mechanical and emerging advanced recycling technologies. A system optimization model aimed at minimizing environmental impacts was developed to determine optimal end-of-life (EOL) management and locations of existing and emerging U.S. recycling infrastructures. Our study includes material flows from virgin resin production through semi-manufacturing processes to existing EOL disposal and recycling processes. An optimized circular plastics packaging system achieved greenhouse gas (GHG) emission savings of up to 28% and cumulative energy demand (CED) savings of up to 46%, compared to the linear economy. Moreover, these savings of GHG emissions and CED impacts represent a reduction of 0.16% and 0.49% compared to annual U.S. GHG emissions and energy consumption in 2022, respectively. The optimal recycling rates and systems-level circularity ranged from 78–99% and 57–75%, respectively. Increased energy savings led to increased GHG emissions showing a potential trade-off between GHG emissions and energy. Analysis of 40 scenarios showed the importance of material collection distances, blend limit of mechanically recycled resins, process yields, and mandated recycling rates for achieving a sustainable circular economy of plastics.

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## Sustainability spotlight

Existing plastics supply chains driven by fossil resources follow a linear economy model hindering the achievement of a sustainable circular economy. A linear-to-circular transition *via* integrating existing and emerging advanced/chemical recycling technologies in plastics supply chains has been recognized as one of the potential solutions to reduce consumption of fossil-based plastics. However, the sustainability impacts/trade-offs and the required changes in recycling infrastructure to achieve a sustainable circular economy are not well understood at a systems level. Our study minimizes this research gap and aligns directly and/or indirectly with following United Nations (UN) Sustainable Development Goals (SDGs): SDG 9 (industry, innovation and infrastructure), 11 (sustainable cities and communities), 12 (responsible consumption and production), 13 (climate action), and 17 (partnerships for the goals).

## 1 Introduction

The U.S. plastics industry is one of the most important industries, contributing towards the nation's economic output and was the sixth largest industry by gross output in 2020.[1] The plastics industry contributed 5.5% of the total employment and 6.3% of the total economic value added among all the U.S. manufacturing sectors in 2021.[2] However, the current plastics supply chains follow a "linear economy" ("take-make-use-dispose"[3]) model, which is associated with depletion of fossil resources and mismanagement of valuable waste plastics at their end-of-life (EOL).[4]

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The current low EOL recycling and higher disposal rates of plastics represent an economic loss of U.S. $10 billion per year[5,6] and energy loss of 1462 to 1517 Peta Joules (PJ) to landfills in the U.S.,[5] further challenging the U.S. to achieve sustainability of plastic supply chains. A transition from the existing linear to a future circular economy in plastic supply chains has been recommended by many organizations and countries, and they have recognized the need for a systems level approach to address the plastic waste problem and improve sustainability.[4]

Commercially practiced mechanical recycling processes are not able to produce virgin-quality recycled plastics that can replace fossil-derived plastics. This is due to degradation of material properties, which further affects the strength and appearance of recycled products.[7–10] Such limitations hinder the ability to close the loop on plastics supply chains.

Complementary to mechanical recycling technologies for plastics, integrating emerging recycling technologies, such as solvent-based dissolution precipitation, glycolysis, methanolysis, enzymatic hydrolysis, and pyrolysis, has been envisioned to be a potential solution to achieve a closed-loop circular economy for plastics.[11–15] These advanced recycling technologies are able to create virgin-like high quality recycled plastics that can replace fossil-derived plastics.[16–18] However, the sustainability implications of transitioning from a linear to a circular economy of the U.S. plastics supply chain are not well understood.[19]

The lack of EOL plastics supply chain infrastructure producing high quality recycled plastic resins is one of the many challenges[13,20–23] to achieve high circularity for plastics. Moreover, limited access to recycling in rural areas is another important challenge for increasing recycling rates of plastics.[24] Realizing the growing need to establish and optimize a robust EOL U.S. recycling infrastructure,[24–28] only a few studies have conducted geospatial optimization and evaluated the sustainability of EOL plastics supply chain infrastructure in a closed-loop circular economy.

Various studies are emerging in the field of optimizing and evaluating the sustainability of plastics in a circular economy.[29–36] But only a few prior studies have optimized the EOL recycling infrastructure for plastics in open-loop and closed-loop recycling in various countries including, but not limited to, the U.S.,[37–44] European countries,[45–48] China,[49] and Thailand.[50] A brief overview of this literature is provided in Table S1 of the ESI document.† The U.S. studies that focused on closed-loop recycling were limited to either a certain type of plastics, types of recycling technologies, or limited in geographical scope (state or regional level). Nearly all these studies either maximized profits or minimized total costs of the studied plastic waste management systems, while some studies included both economic and environmental objective functions. Only one U.S. national-level study[39] maximized the total profit of the closed-loop plastic waste recycling infrastructure, but their study considered only mechanical recycling and pyrolysis recycling technologies. Their study excluded the existing locations of material recovery and mechanical recycling facilities in the U.S.[39] A plastic-to-plastic circularity of 34% was determined with a combination of mechanical recycling and pyrolysis recycling technologies.[39] Their study[39] also quantified (but did not optimize) greenhouse gas (GHG) emissions and found GHG emission savings of 75%, when compared to waste-to-energy (WtE) systems.

Despite the advances described in the reviewed literature, they exhibit limitations in study scope and system understanding. None of the studies included/depicted U.S. material flows and impacts associated with production of virgin resins and their use in semi-manufacturing processes (*e.g.*, film extrusion, sheet extrusion, thermoforming, injection molding, blow molding, and stretch blow molding), which prior research finds to be the highest emitting and energy consuming processes,[51,52] and thus provided limited systems level information about the entire U.S. plastics supply chain processes. Furthermore, none of the studies reported cumulative energy demand (CED) or energy consumption of the studied systems, given the high energy consumption of plastics supply chains.[51,53] Additionally, except for a few limited studies (see ESI Table S1†), none of the studies considered the blend limit for mechanically recycled resins that accounts for the poor quality of mechanically recycled resins. Moreover, most of the studies assumed 100% of the waste plastics to be collected for sorting and recycling, *i.e.*, all of the plastic waste generated must be collected and transported to the recycling centers. This assumption does not account for appropriate EOL collection methods for waste plastics and lacks the consideration of the type of plastic waste products. In the U.S., EOL plastic waste flows are dictated by the type of plastic product and its suitability in the EOL collection stream.[23] For example, PE films, which alone account for 24% of the total U.S. plastic packaging waste,[54] are mainly collected *via* store drop-off or retail collection centers,[55] which directly go to plastic reclaimers, rather than to curbside collection programs and materials recovery facilities (MRFs). These system processes are included in the study presented here.

To fill the above-mentioned research gaps, the present study applied our previously developed systems analysis framework[56,57] that can evaluate sustainability impacts of linear as well as circular plastics supply chains. The novelty of our study is that the system model developed in our study includes material flows through upstream supply chain processes such as virgin resin production and semi-manufacturing processes, as well as downstream supply chain processes such as landfilling, incineration with energy recovery, materials recovery facility (MRF) operations, mechanical recycling, and emerging chemical/advanced recycling technologies, further providing systems-level insights. In addition, the system model presented here includes optimum locations of key recycling facilities that minimize material transportation impacts.

Our study is focused on the following important types of plastic resins: polyethylene terephthalate (PET), high density polyethylene (HDPE), low density polyethylene/linear low-density polyethylene (LDPE/LLDPE), and polypropylene (PP), that are intended for packaging applications having a short in-use average lifetime of less than one year.[58] PET and polyolefin (PO) plastic packaging waste accounts for 91% of the total plastic packaging waste and 37% of the total plastic waste

generated in the U.S.[54] Thus, this category of plastic materials is of great importance to study and optimize.

Starting with the central research hypothesis that a circular economy is more sustainable than a linear economy for plastics, the first objective of this research is to derive a "cradle-to-cradle" system optimization model for U.S. PET and PO plastic packaging supply chains in a closed-loop circular economy to minimize GHG emissions and CED and to compare against a linear economy. The second research objective is to determine the optimal EOL material management and recycling infrastructure for plastics in the U.S., including existing as well as potential locations of new facilities and lengths of transport segments to minimize the system GHG emissions and energy consumption. The third research objective is to conduct scenario analyses around different EOL supply chain configurations to understand their environmental impacts. As a part of the second and third research objectives, recycled content and circularity of environmentally optimal systems were also calculated under various scenarios.

# 2 Methods

## 2.1 Principles of systems analysis of circular supply chains

The present study applied a previously established systems analysis framework[56,57] and principles for plastics supply chains.[59] These principles are related to material flow analysis (MFA), open *versus* closed material flows, and the basis for compiling impact assessment data. Here, we repeat these principles and contribute additional ones with the purpose of providing guidance to conduct systems analysis for sustainability assessment.

• Goal and scope of the systems analysis: similar to the methods for conducting life cycle assessment (LCA), defining the goal and scope of the analysis is essential. Goal and scope definition provides the overall analysis framework and affects all aspects including data gathering, analysis methods, and result interpretation.

• Open *versus* closed systems: the analysis of a system will be different depending on whether the system is open or closed. Closed systems will normally result in fewer processes for material conversion compared to open systems, and therefore will be simpler to model with regard to impact assessments, and will achieve higher degrees of material circularity compared to open systems.

• Material flow analysis (MFA): MFA is the foundation of systems analysis because it describes not only the material flow amounts but also the processes employed in the conversion of those material types in the system.

• Transportation logistics: the transportation distances of a material between the network of processing facilities is of high importance in systems analysis, with impacts on economic performance and impacts on the environment and society. In modeling future versions of the system, optimum locations of additional facilities of varying capacities should be included.

• Impact assessment: impact assessment data, whether environmental, economic, or societal, should be expressed based on material flow, and care must be taken to avoid under or over counting of impact by judicious choice of basis and system boundary of the data.

• Optimization modeling: the optimization model should be developed to align with the goal and scope of the study. The optimization model must combine material flow data, life cycle impact assessment (LCIA) data, and geographical data (*e.g.* locations of recycling facilities, county centroids, and distances) with appropriate constraints (*e.g.* mass balance, capacity, *etc.*) to determine, for example in this study, the optimal recycling infrastructure and EOL management of waste plastics with respect to one or more sustainability metrics (*e.g.*, minimize GHG emissions).

• Scenario analysis: to better understand the effect of key parameters or assumptions on the system's performance/behavior, conducting a scenario (or sensitivity) analysis is necessary. For example, our study presented here conducted scenario analysis with respect to key system technical and operational parameters (*e.g.* blend limit for mechanical recycling, waste collection proximity, *etc.*).

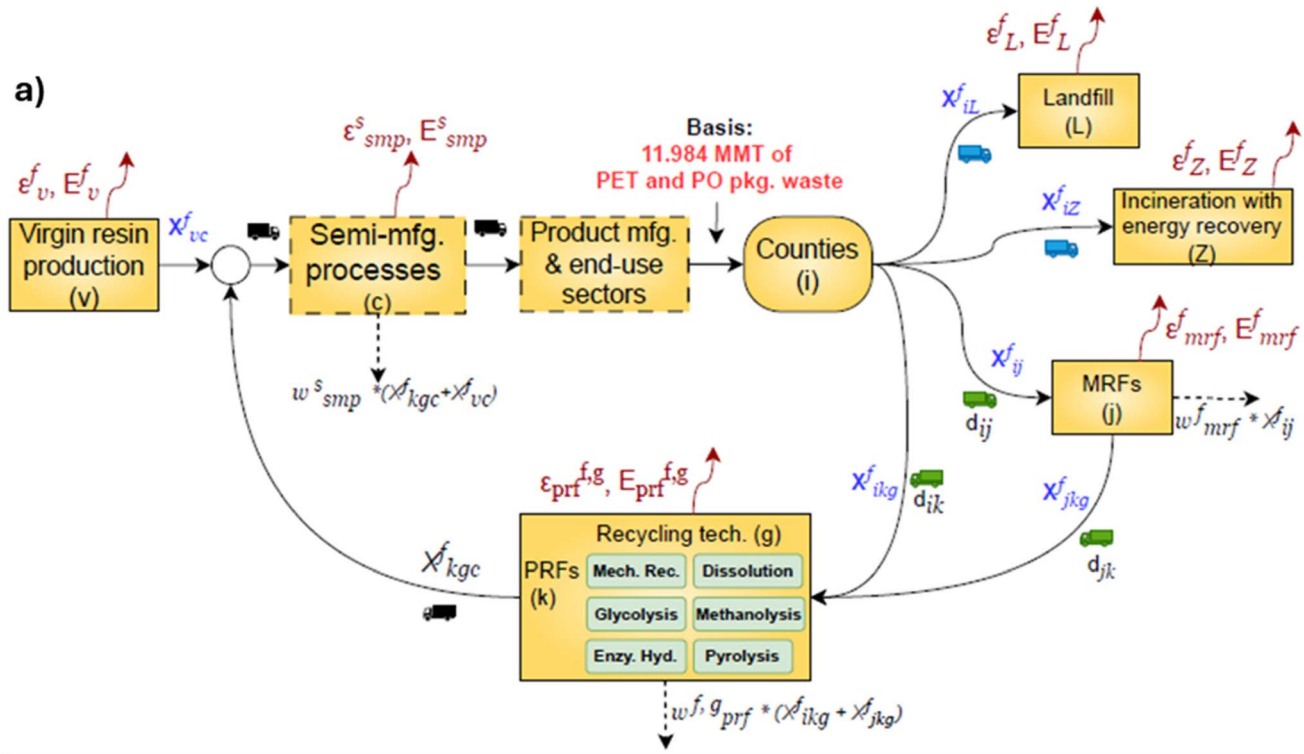
In the present study, a system optimization model was combined with MFA and LCIA factors to evaluate the environmental impacts of U.S. PET and PO plastic packaging supply chains in a closed-loop circular economy. Specifically, a mixed-integer linear programming (MILP) optimization model was developed based on our previous work[60] to minimize the GHG emissions and CED impacts of the circular PET and PO plastics packaging supply chains in the U.S. The formulation of the optimization model, including information about the objective functions and constraints, is provided in Section S2 of the ESI document.†

Briefly, the optimization model includes mass balance constraints around system processes, bottle *vs.* non-bottle deposit state constraint, collection of rigid *vs.* film packaging material constraint, MRF collection proximity constraint, minimum demand for recycled resins, blend limit constraint for mechanically recycled resins, capacity and utilization of MRFs and plastics recycling facilities (PRFs), and number of MRFs and PRFs allowed to be installed at a particular location (see ESI Section S2.1.2 for details†).

The basis for this MILP model is the total amount of EOL PET and PO plastics packaging waste generated at population-based county centroids in the U.S. Using geospatial datasets, material flow and LCA datasets, the model selects the optimal EOL material flows to landfills, incineration with energy recovery, and recycling along with locations and capacities of MRFs and PRFs to be installed across the U.S. to minimize the GHG emissions and CED impacts of the system. A brief overview of the decision variables and system being optimized is described in the next section.

## 2.2 Overview of the PET and PO plastic packaging system

The circular PET and PO plastic packaging system is comprised of the major supply chain processes (Fig. 1), based on our prior study.[57] The upstream processes (*i.e.* before the consumer's use of plastics) include production of virgin resins and different semi-manufacturing (converters) processes. The downstream



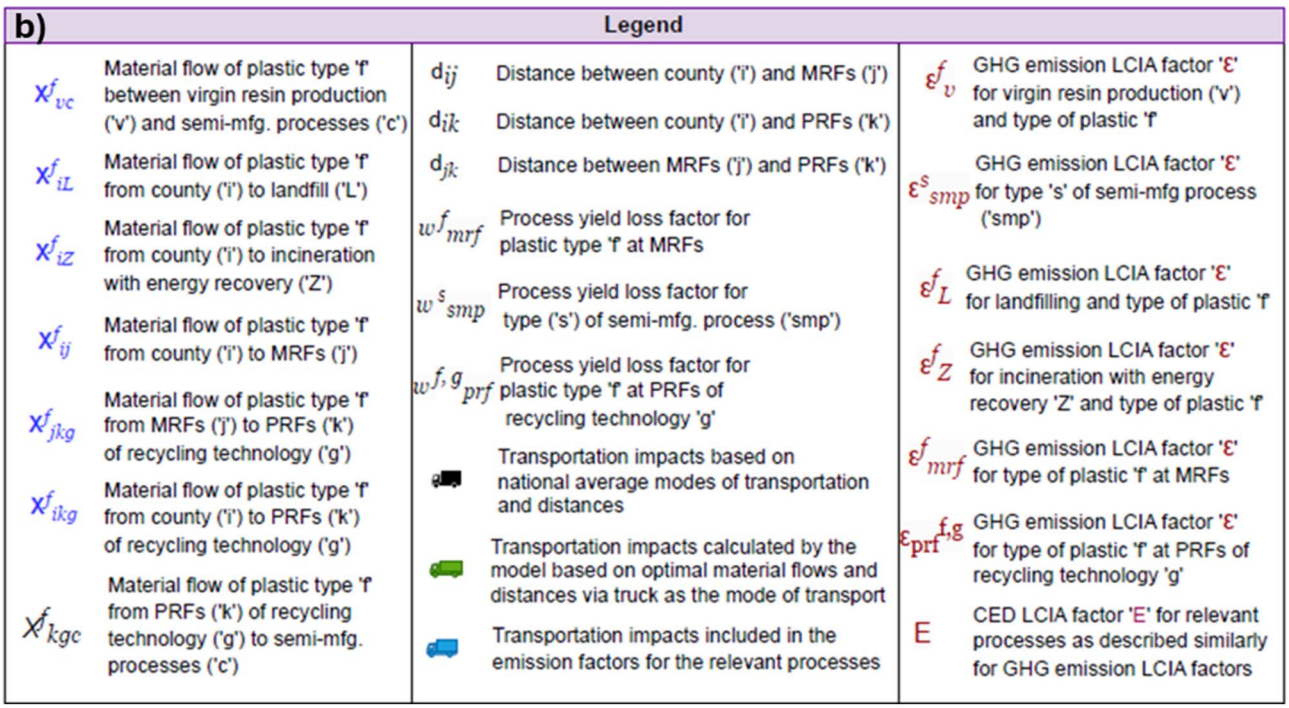




Fig. 1 Overview of the system optimization model for the U.S. PET and PO plastics packaging supply chain (in panel (a)) along with a legend (in panel (b)). Notes: (1) The decision variables are shown in blue color; (2) the dashed arrows (in panel (a)) represent the waste generated during each process, which is assumed to be sent to 80% landfill and 20% incineration with energy recovery based on conventional disposal of waste plastics in the U.S.[54] (3) The boxes with dashed lines ('semi-mfg. processes' and 'product mfg. & end-use sectors') indicate that those processes were not a part of the objective function. However, the impacts of semi-manufacturing processes were estimated and included in total GHG emissions and CED impacts. These impacts of semi-manufacturing processes were constant due to the assumed constant demand and application of PET and PO plastic packaging resins (see ESI Section S2.1.1†).

processes (*i.e.* after the consumer's use at the EOL) include collection of materials, landfilling, incineration with energy recovery (or WtE), MRFs, and PRFs. PRFs include a suite of existing and emerging recycling technologies such as mechanical recycling, solvent-based dissolution precipitation, glycolysis, methanolysis, enzymatic hydrolysis, and fast pyrolysis. All

these supply chain processes, including intermediate transportation steps, are associated with certain GHG emissions and CED impacts, as shown in Fig. 1, which contribute to the total impacts of the system. The abbreviations shown in Fig. 1 along with those used in the objective functions are provided in Table S2 of ESI Section S2.1.1.†

The starting point for this model is the total amount of PET and PO plastic packaging waste generated across all U.S. counties (11.984 million metric tons (MMT)).[54] The EOL fate of this waste could be landfilling ($X^{f}_{iL}$), incineration ($X^{f}_{iZ}$), or collection for sorting and recycling ($X^{f}_{ij}$ and $X^{f}_{ikg}$). This packaging waste, excluding PE films, could be collected *via* curbside collection programs ($X^{f}_{ij}$) and then sent to MRFs for sorting and baling. The MRFs can collect this waste within an assumed nearby collection proximity of 100 km, aligning with the value reported in the literature.[61] A total of three proximities were considered: 100 km (base proximity case), 250 km, and 500 km, to study the effects of assumed MRF collection proximity on systems performance. Alternatively, the PET and PO plastic packaging waste could also be collected *via* special collection programs ($X^{f}_{ikg}$), such as bottle deposit programs for PET bottles (only applicable for the ten bottle deposit states) and store drop-off or retail collection centers for PE films, and then sent directly to PRFs for producing recycled resins. It was assumed that the PET bottles and PE films collected *via* these special collection programs would be baled on-site at collection centers for efficient transportation of materials to PRFs.

The emerging advanced/chemical recycling technologies considered in the system model include integrated "plastics-to-plastics" processes, *i.e.*, all recycling technologies produce recycled resin as an output product in the same facility. Therefore, the material losses and environmental impacts associated with "plastic-to-monomer" conversion and "monomer-to-polymerization" steps are included for glycolysis, methanolysis, and enzymatic hydrolysis recycling technologies.[12] Similarly, in terms of fast pyrolysis of PO plastics, the environmental impacts of "plastics-to-hydrocarbon products" conversion, "hydrocarbon products-to-monomers" conversion and "monomer-to-polymerization" conversion are included in the LCIA factors. Moreover, all the recycling technologies included the impacts of the necessary pretreatment steps to remove any undesired contaminants from the baled plastics. The final produced recycled resins are then transported to plastic converters, which use various semi-manufacturing processes to produce semi-finished products. To account for the quality loss due to mechanical recycling, a maximum blend limit of 35% for mechanically recycled resins is imposed for all resins.[8,9] The recycled resins produced *via* all other emerging recycling technologies included in this study are assumed to be high quality resins directly replacing fossil derived virgin resins, and therefore they are not subject to the blend limit/quality constraint.[16–18] It is important to note the compatibility/suitability of all these recycling technologies with specific types of PET and PO packaging plastics in our optimization model (see Table S3 in ESI Section S.2.1.2†). All PET and PO packaging plastics can be recycled *via* mechanical recycling and solvent-based dissolution precipitation recycling technologies. Glycolysis, methanolysis, and enzymatic hydrolysis technologies are applicable only to PET resin. The "plastics-to-plastics" fast pyrolysis technology is applicable to all the studied PO resins only (see Table S3†).

The decision variables of this optimization model are the material flows to landfill, incineration with energy recovery, MRFs, and PRF processes, and binary decision variables are the locations and capacities of MRFs and PRFs. The potential/candidate locations of MRFs were based on our prior work.[60] The potential locations of PRFs were assumed to be the same as the existing locations of PRFs; however, installation of multiple different recycling technologies with different capacities was allowed at a given location (see ESI Section S2.1.2†). The optimization model presented in this study excluded capacity expansion of existing MRFs and PRFs, mainly due to the lack of detailed and actual facility-level operating data. Instead, the optimization model predicts the optimum capacity requirements from a given range of capacities at existing and/or new locations of MRFs or PRFs, depending on the supply and demand for packaging plastics. Such an optimization approach helps to answer the research question of what the capacities of MRFs and PRFs should be to minimize GHG emissions and energy consumption of the system considering the supply and demand of waste packaging plastics. Refer to constraints and range of capacities in ESI Section 2.1.2.†

### 2.3 System model data inputs

Briefly, the input data to this model include county-level PET and PO packaging waste generated in the U.S., locations of population-based county centroids,[62] existing and future locations of MRFs and PRFs[63,64] considering both mechanical and advanced recycling technologies, capacity ranges of MRFs and PRFs, GHG emissions and CED LCIA factors for all EOL processes, and process yield loss factors for major supply chain processes in the system.

The supply chain wide GHG emissions and CED impacts of processes were evaluated by multiplying the material flows with the respective LCIA factors, considering the input or output basis and system boundary of the LCIA factors (see Table 1 along with sources of data). The GHG emission LCIA factors were gathered from the literature, with all factors estimated using the Intergovernmental Panel on Climate Change (IPCC) 2013 and 2021 Global Warming Potential (GWP) over a 100-year timeframe LCIA method, depending on the referenced literature. Similarly, the CED LCIA factors were gathered from a literature review of studies using the Cumulative Energy Demand (CED) LCIA method. See ESI Section S2.2.1† for additional information about LCA datasets. The present study used the best available dataset for the supply chain processes at the time the research was conducted.

The county-level PET and PO plastic packaging waste generation was estimated using the population density of each county[78] with 0.036 MT of PET and PO packaging waste generated per capita annually.[54] The origin of this waste generation was assumed to be at the population-based centroid of each county.[62] The distances between population-based county centroids, MRFs, and PRFs (see Fig. 1 with green colored truck

Table 1 Summary of GHG emissions and CED LCIA factors for supply chain processes

| | GHG emissions (MT $CO_2$-eq. per MT output) | CED (MJ per MT output) | Process yield loss (%) | Reference |
|---|---|---|---|---|
| **Virgin resin production**[a] | | | | |
| PET | 2.23 | 61 400 | — | 65 |
| HDPE | 1.61 | 73 800 | — | 66 |
| LDPE/LLDPE | 1.59[d] | 74 825[d] | — | 67 and 68 |
| PP | 1.55 | 75 500 | — | 69 |
| **Semi-manufacturing processes**[b] | | | | |
| Film extrusion | 0.407 | 10 100 | 2.40% | 51 |
| Sheet extrusion | 0.331 | 7780 | 0.30% | |
| Injection molding | 1.01 | 24 700 | 0.60% | |
| Blow molding | 0.94 | 25 700 | 0.30% | |
| ISBM | 1.16 | 27 800 | 2.2% | |
| Film/sheet/others | 0.348 | 8867 | 1.6% | |
| **EOL** | | | | |
| Landfill[a] | 0.022[e] | 310[e] | — | 70 |
| **Incineration with energy recovery**[ac] | | | | |
| PET | 1.37[e] | −11 620[e] | — | 70 |
| HDPE | 1.42[e] | −21 900[e] | — | 70 |
| LDPE/LLDPE | 1.42[e] | −21 810[e] | — | 70 |
| PP | 1.42[e] | −21 850[e] | — | 70 |
| Compacting and baling only (deposit containers) | 0.036 | 541 | — | 71 and 72 |
| Compacting and baling only (PE films) | 0.025 | 160 | — | 73 |
| **MRF**[b] | | | | |
| PET | 0.06 | 864 | 11% | 74 |
| HDPE | 0.071 | 1086 | 16% | 74 |
| LDPE/LLDPE | 0.076 | 1143 | 17% | Average of HDPE and PP due to lack of data |
| PP | 0.08 | 1200 | 18% | 74 |
| **Mechanical recycling**[b] | | | | |
| PET | 0.768 | 12 740 | 20% | 74 |
| HDPE | 0.396 | 6250 | | 74 |
| LDPE/LLDPE | 0.372 | 6230 | | Average of HDPE and PP due to lack of data |
| PP | 0.347 | 6210 | | 74 |
| **Dissolution**[b] | | | | |
| PET[g] | 0.98 | 16 400 | 11% | 75 |
| HDPE[h] | 2.40 | 87 000 | 12% | 12 |
| LDPE/LLDPE[h] | 2.20 | 78 000 | 6% | 12 |
| PP[h] | 2.20 | 96 000 | 9% | 12 |
| **Glycolysis** [bf] | | | | |
| PET | 1.32 | 28 900 | 24% | 12 |
| **Methanolysis**[bf] | | | | |
| PET | 4.19 | 72 600 | 24% | 12 |
| **Enzymatic hydrolysis**[bf] | | | | |
| PET | 3.15 | 55 000 | 31% | 76 |
| **Pyrolysis**[bf] | | | | |
| HDPE | 1.40 | 17 850 | 10% | See Table S4;[65–69,77] |
| LDPE/LLDPE | 1.385[d] | 18 071[d] | | |
| PP | 1.34 | 17 250 | | |

[a] Cradle-to-gate boundary. [b] Gate-to-gate boundary. [c] Including the credits of a displaced fossil electricity grid mix. [d] Weighted average impacts based on LDPE and LLDPE mass production in the U.S.; weight for LDPE is 0.27 and weight for LLDPE is 0.73; ISBM: injection stretch blow molding. [e] LCIA factors based on input material flows. [f] LCIA factors for plastics-to-plastics recycling (*i.e.* output from the relevant recycling processes is in the form of high-quality recycled resin, assuming an integrated facility). [g] Polymer precipitation *via* the cooling method. [h] Polymer precipitation *via* the anti-solvent method.

symbols) were also determined using the law of cosines formula to estimate distances between these locations.[79] To account for the irregularities in the road network, a winding factor (or tortuosity factor) of 1.2 was assumed[80] and multiplied by the transportation distances.

The mode of transportation for shipment of EOL materials was assumed to be combination trucks (see Fig. 1 with green and blue truck symbols). For transportation of virgin resins, recycled resins, and semi-finished products, the authors used U.S. national average transportation modes from 2019 and distances specific to plastic materials (see ESI Table S5†). Additional details about the estimation of transportation impacts are described in Section S2.2.2 of the ESI document.†

### 2.4 Scenario analysis

Scenario analysis was conducted using the system optimization model (Fig. 1) to evaluate GHG emissions and CED impacts of different supply chain configurations. The studied scenarios differed in the EOL supply chain configurations, MRF collection proximities (100 km; base case, 250 km, 500 km), blend limits on mechanically recycled resins (35%; base case, 75%, no blend limit), and process yield loss factors. Some scenarios also considered the effects of increasing recycling rates and the effect of implementing the bottle bill policy across all states. A hypothetical 'zero waste' or 'ideal circular economy' scenario was also modeled representing zero material loss at each processing stage. A total of 20 scenarios were included in this study and are briefly summarized in Section S2.3 of the ESI document.† Each of these 20 scenarios (ESI Table S6†) was optimized/modeled with respect to GHG emissions and CED impacts, representing a set of 40 total system configurations. Systems-level circularity and recycled content of PET and PO packaging plastics were also evaluated (see Section S2.4 of the ESI document for the formula†).

## 3 Results and discussion

The results of MILP optimization models for minimization of GHG emissions and CED impacts of PET and PO plastic packaging supply chains include optimal EOL management of material flows (Section 3.1), network material flow supply chain maps showing optimal locations of MRFs and PRFs (Section 3.2), optimal values of GHG emissions and CED impacts and their distribution based on supply chain processes (Section 3.3). The results of scenario analysis are presented in Section 3.4.

The optimization model was solved using the CPLEX solver in Python on an Intel Core I7-9700K CPU processor at 3.60 GHz with 64 GB of RAM using two threads. The results were obtained with an optimality gap of less than 0.1%. The optimization model had a total of 23 988 990 variables and 6 697 312 constraints.

### 3.1 Optimal EOL management of plastics packaging

The state-by-state optimal EOL management decisions made by the GHG emission-based optimization model are shown in Fig. 2 for the 100 km MRF collection proximity. Out of 11.98 MMT of PET and PO plastic packaging waste, about 78% was

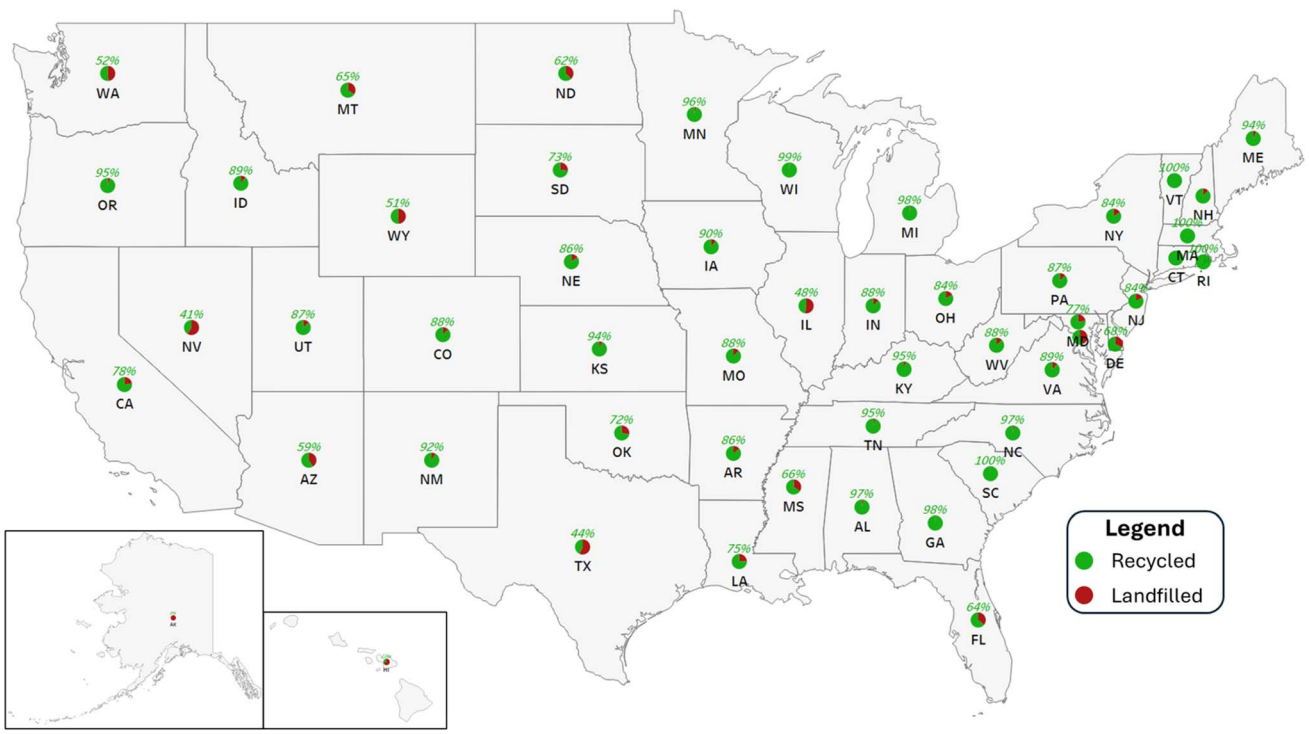



Fig. 2 Optimal EOL management of U.S. PET and PO packaging materials by state, when the system is optimized for GHG emissions with 100 km collection proximity. Notes: (1) Tabulated data by state for this figure is provided in ESI Table S7; † (2) Hawaii and Alaska have recycling rates of 27% and 0%, respectively; (3) WtE was never chosen as the EOL pathway due to its high GHG emissions and, therefore, WtE is always 0% for the GHG minimization model.

collected for sorting and recycling and 22% was landfilled across the U.S., with zero percent incineration to achieve minimum GHG emissions of the system.

Out of 51 total states (including Washington DC), 46 states had recycling rates greater than 50%, but only five states had landfilling rates greater than 50%, namely, Alaska, Hawaii, Nevada, Texas, and Illinois (see ESI Table S7†). The primary reason for high landfilling rates in certain states is the lack of MRF facilities within the 100 km county to MRF proximity limit. However, with an increased MRF collection proximity of 500 km, 49 states had recycling rates greater than 50% and only two states had landfilling rates greater than 50%, namely, Alaska and Hawaii (see ESI Table S7†). This also shows that increased MRF collection proximity distance led to increased recycling rates and lower landfilling rates for these plastics. A maximum overall optimal recycling rate of 95% and landfilling rate of 5% was achieved for the system optimized on GHG emissions with 500 km MRF proximity.

### 3.2 Material flow network supply chain maps

This section provides EOL recycling supply chain network maps for the PET and PO plastics packaging system optimized on GHG emissions with an MRF collection proximity of 100 km. These maps are shown in Fig. 3–5, for material flows between county-to-MRFs (C-2-M), county-to-PRFs (C-2-P), and MRFs-to-PRFs (M-2-P), respectively. These maps for systems optimized on GHG emissions with a MRF collection proximity of 500 km are shown in ESI Fig. S1–S3.† The C-2-M, C-2-P, and M-2-P material flows are some of the key results obtained from the integrated network optimization model. These results are presented individually in the following Section 3.2.1–3.2.3 to simplify the complexity of network supply chain maps shown in Fig. 3–5. This also enables better visualization and understanding of the recycling infrastructure as well as the origin and destination of material flows at county/state levels.

**3.2.1 County-to-MRF (C-2-M) material flows.** Based on Fig. 3, out of 3143 counties, 69% (2155 counties) are participating in 100% recycling only (no landfill or incineration), 19% (598 counties) are participating in 100% landfilling only, and 12% (390 counties) are participating in both landfilling and recycling. A total of 441 MRFs were selected by the optimization model, out of which 263 were existing MRFs and 178 were new MRFs (see ESI Table S8†). The average baled MRF output of PET and PO packaging plastics was found to be 11 800 MT per year for a 100 km MRF collection proximity. However, this average baled output increased to 14 100 MT per year with the MRF collection proximity increased to 500 km (See ESI Table S9†).

The model predicts that California (CA) had the highest number of total MRFs (36), followed by Texas (TX; 28), New York (NY; 19), and Georgia (GA; 18). Florida (FL), Michigan (MI), and North Carolina (NC) had a total of 17 MRFs each. All these seven states together accounted for about 34% of the total MRFs. Regarding states with zero existing MRFs, the optimization model determined a total of ten new MRFs to be installed in Mississippi (MS; 4), Wyoming (WY; 3), and North Dakota (ND; 3) (see ESI Fig. S4†). Apart from these states, a greater number of

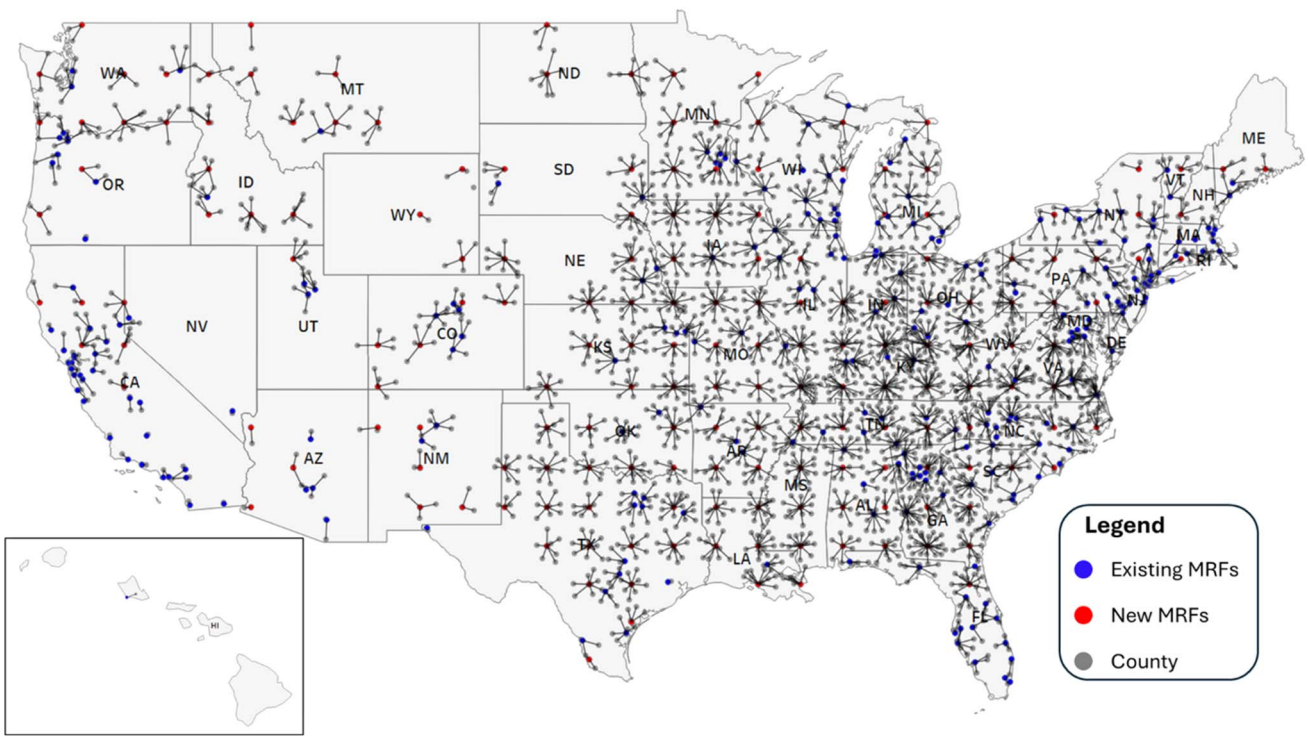



Fig. 3 County-to-MRF (C-2-M) optimal U.S. EOL PET and PO packaging recycling material flow network for systems optimized on GHG emissions with 100 km MRF collection proximity. Notes: (1) only the counties participating in recycling are shown in this figure; (2) Alaska has a 0% recycling rate, and therefore, is not shown in this figure. (3) This figure shows material flows from counties (origin) to MRFs (destination).

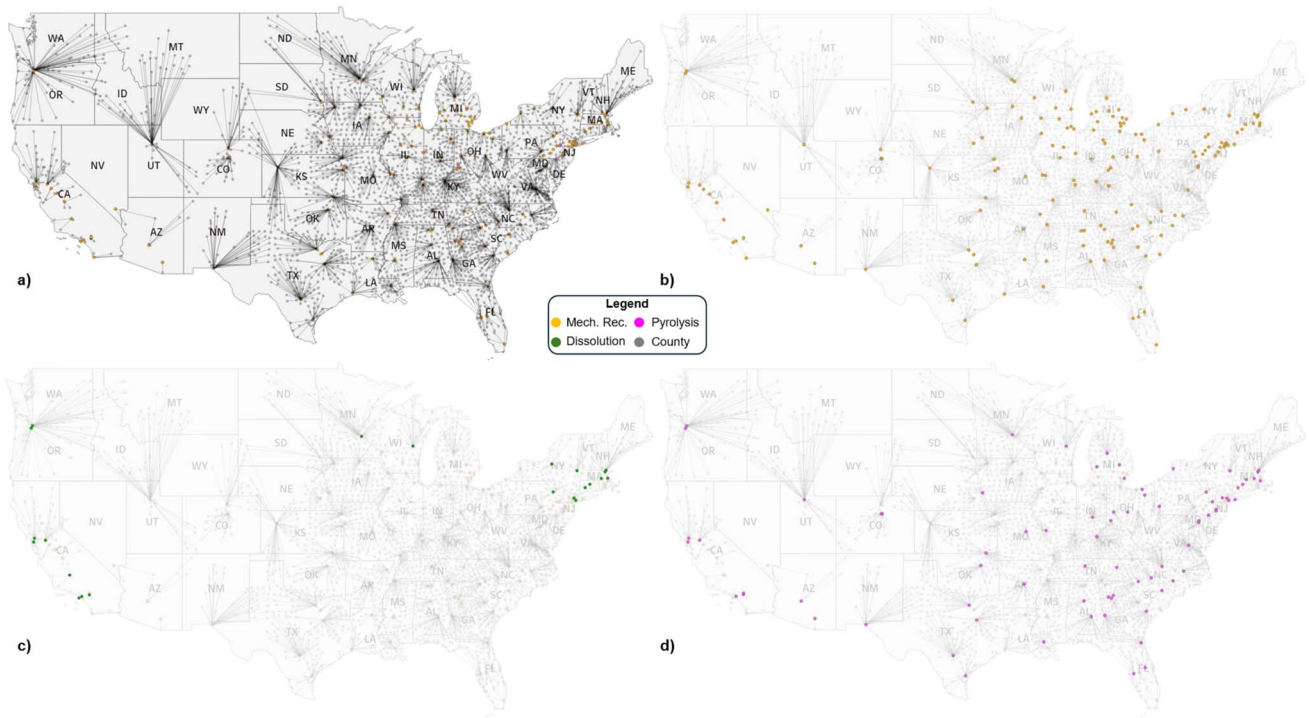



Fig. 4 County-to-PRF (C-2-P) optimal U.S. EOL PET and PO packaging recycling material flow network for the systems optimized on GHG emissions with 100 km MRF collection proximity. Note: (1) only the counties participating in recycling are shown in this figure. (2) This figure shows material flows from counties (origin) to PRFs (destination); (3) C-2-P material flows from Hawaii are shown in Fig. S13; † (4) the maps shown in (b–d) are slightly faded out to clearly highlight the locations of each recycling technology collectively shown in (a).

new MRFs would need to be established in other states, including Montana (MT), Idaho (ID), TX, Louisiana (LA), Missouri (MO), and Alabama (AL), to achieve minimum GHG emissions and high circularity of the system (see ESI Fig. S4†).

In terms of the optimal C-2-M material flows (Fig. 3), overall, a total of 6.08 MMT of PET and PO packaging material was transported with an average distance of 58 km from C-2-M across the U.S. Moreover, about 90% of the total C-2-M material flows were recycled at MRFs located in the same state as the origin county (referred to as 'in-state'), and 10% was recycled at MRFs located out-of-state (see ESI Table S10†). Only two states, New Hampshire (NH) and District of Columbia (DC), exported 100% of their total packaging plastics waste generated to their neighboring states, as both states have no in-state MRFs (see ESI Fig. S4†). NH exported all of its PET and PO packaging waste (0.03 MMT) to Maine (ME), Massachusetts (MA), and Vermont (VT), whereas DC exported all of its waste (0.01 MMT) to Maryland (MD) and Virginia (VA). However, in terms of state-wide export material flows, New Jersey (NJ), Illinois (IL), NY, and MO were the top four states that exported a total of 0.19 MMT, representing 32% of the total state-wide exports (0.58 MMT), to their neighboring states. Increasing the MRF collection proximity to 500 km led to more out-of-state recycling than in-state recycling (see ESI Table S10†). In this case, a greater number of counties (89%; 2795 counties) participated in 100% recycling, just 1% (30 counties) participated in 100% landfilling, and 10% (318 counties) participated in both landfilling and recycling (see ESI Fig. S1–S3†).

The total mass distribution of C-2-M material flows as a function of distance travelled and number of counties under the various MRF collection proximities is shown in ESI Fig. S5 and S6.† Moreover, the histograms showing the distribution of MRF output, capacity, and utilization rate are shown in ESI Fig. S7 and S8.†

**3.2.2 County-to-PRF (C-2-P) material flows.** The optimization model predicted a total of 297 plastic recycling facilities (Fig. 4a), of which 158 were mechanical recycling facilities (Fig. 4b). The total number of facilities utilizing dissolution (Fig. 4c) and pyrolysis (Fig. 4d) recycling technologies were 55 and 84, respectively (see ESI Table S8†). The state-by-state distribution of the total number of PRFs based on the type of recycling technology and the average output from technologies is shown in ESI Fig. S9–S12.†

In terms of the optimal C-2-P material flows (Fig. 4), a total of 3.29 MMT of PET and PO packaging material waste was shipped with an average distance of 158 km directly from counties to different types of recycling technologies across the U.S. The C-2-P material flows include only PET bottles collected in the ten bottle deposit states and PE films collected *via* store drop-off centers. Of the total PET and PO packaging material collected for sorting and recycling (9.37 MMT; see ESI Table S11†), 35% originated from C-2-P material flows, while 65% was from C-2-M. About 85% of the total C-2-P material flows were recycled at PRFs in-state, and 15% was recycled at out-of-state PRFs (see ESI Table S10†).

The average output, capacity, and utilization rates of PRFs by recycling technologies are shown in Table S12.† The mass

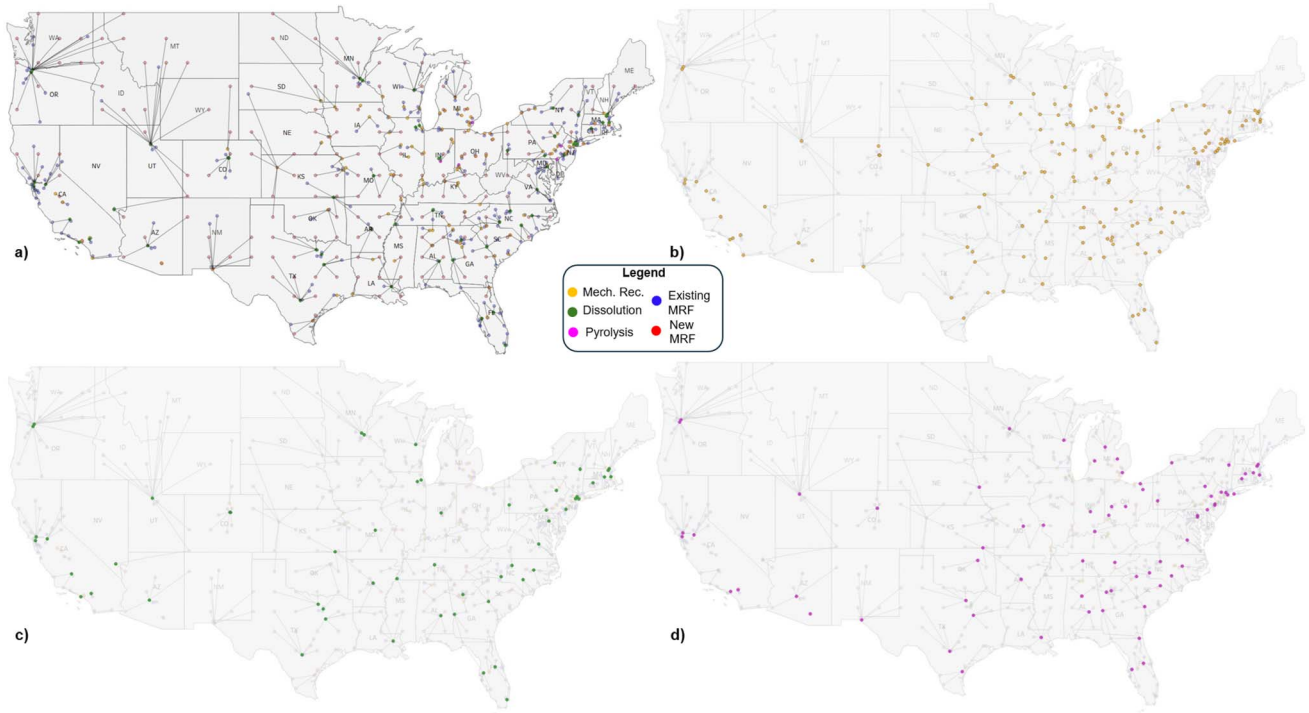



Fig. 5 MRFs-to-PRFs (M-2-P) optimal U.S. EOL PET and PO packaging recycling material flow network for systems optimized on GHG emissions with 100 km MRF collection proximity. Notes: (1) this figure shows material flows from MRFs (origin) to PRFs (destination). (2) M-2-P material flows from Hawaii are shown in Fig. S21; † (3) the maps shown in (b–d) are slightly faded out to clearly highlight the locations of each recycling technology collectively shown in (a).

distribution of C-2-P material flows as a function of distance travelled and number of counties under the various MRF collection proximities is shown in Fig. S14 and S15.† Moreover, the histograms showing the distribution of PRF output, capacity, and utilization rate are shown in Fig. S16 and S18.†

**3.2.3 MRFs-to-PRFs (M-2-P) material flows.** A total of 5.2 MMT of PET and PO plastic packaging bales were produced at MRFs, which was further shipped to PRFs by trucks (Fig. 5), as predicted by the optimization model. About 81% of these total baled plastics were shipped to in-state PRFs, and the remaining 19% were shipped to out-of-state PRFs (see ESI Table S10†). The average transportation distance between M-2-P was found to be 145 km. Moreover, the material flow distribution of M-2-P baled plastics showed that a greater number of counties shipped lower masses of baled plastics over short distances (<500 km) (see ESI Fig. S19 and S20†).

With combined C-2-P and M-2-P material flows, a total of 7.1 MMT of recycled PET and PO plastic resins were produced *via* an optimal combination of recycling technologies of mechanical recycling (for PET and PO), dissolution (only PET), and fast pyrolysis of HDPE and LDPE/LLDPE. The optimization model prescribed maximizing mechanical recycling up to the blend limit of 35%, with the remaining to be recycled *via* dissolution-precipitation and fast pyrolysis recycling technologies. This represents a recycled content of 58% for a 100 km MRF collection proximity. However, with the MRF collection proximity increased to 500 km, the recycled content increased to 71% due to increased EOL recycling.

**3.2.4 Rural *vs.* urban counties.** To further understand the role of rural and urban counties in plastics circular economy, all the counties were divided into three main categories: mostly urban, mostly rural, and completely rural.[81] These categories were based on the share of the 2020 rural population in the counties.[82] Additional descriptions and results of this analysis are presented in Section S3.2.4 of the ESI document.† ESI Fig. S22† shows the mass of PET and PO plastic packaging materials and number of counties as a function of county classification.

Overall, Fig. S22† shows that most of the materials collected for sorting and recycling originated from urban counties. With the MRF collection proximity increased to 500 km, a greater number of counties participated in 100% recycling and only a few participated in 100% landfilling. However, compared to 100 km proximity (Fig. S22-A†), the increased MRF collection proximity led to a percentage increase of up to 31% in the number of rural counties that are only recycling *versus* a percentage increase of up to 27% in the number of urban counties that are only recycling. This highlights the important role of rural counties in addition to urban counties towards achieving a circular economy of plastics.

### 3.3 Optimal GHG emissions and CED of U.S. PET and PO plastics packaging supply chains

The results of minimizing GHG emissions and CED impacts of the PET and PO packaging system model are shown in Fig. 6 for the MRF collection proximities of 100, 250, and 500 km. Fig. 6 also shows the corresponding recycling rates, optimal

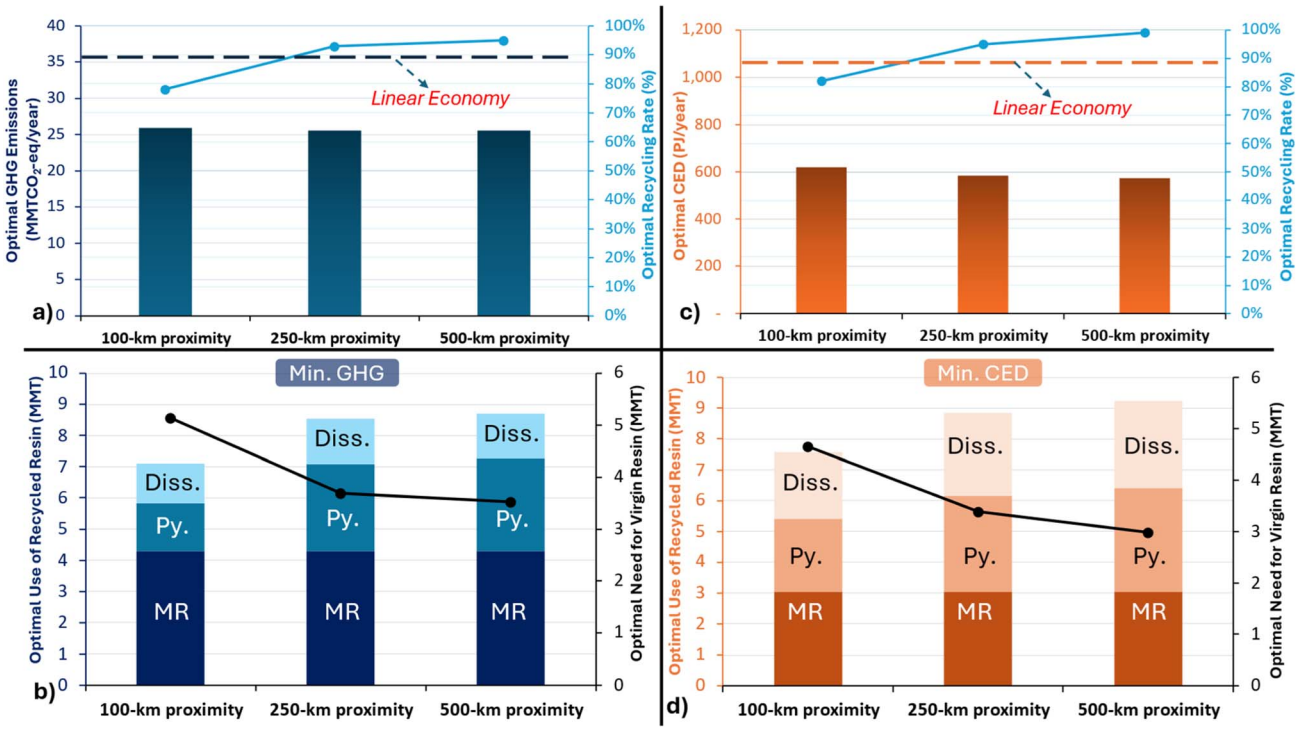



Fig. 6 Optimal GHG emissions (a and b) and optimal CED impacts (c and d) along with the respective optimal recycling rates; recycled resins produced from mechanical recycling (MR), pyrolysis (Py.) and dissolution (Diss.); and the need for virgin resins for collection proximities of 100 km, 250 km, and 500 km. Note: the dashed line represents the GHG emissions (panel a) and CED (panel c) impacts of the linear economy scenario (80% landfill; 20% WtE) that serves as a basis for comparison.

production of recycled resins based on the type of recycling technology, and the resultant need for fossil-derived virgin PET and PO plastic resins in the system. Across all MRF collection proximity cases, the optimization model prescribed reducing emissions and energy consumption with more EOL recycling *via* a combination of mechanical recycling, dissolution, and fast pyrolysis recycling technologies, rather than traditional EOL disposal methods, to reduce the need for fossil derived virgin plastics, which has the greatest emissions and energy factors in the system.

The GHG emissions and CED impacts of these supply chains in a linear economy (80% landfill and 20% WtE) were 35.60 MMT $CO_2$-equivalent (eq.) per year and 1057 Peta Joules (PJ) per year, respectively. These impacts would represent 0.56% and 1.1% of the total U.S. GHG emissions and energy consumption in 2022, respectively.[83,84] Also, the GHG emissions of linear PET and PO plastics packaging systems would represent 1.9% of the total U.S. industry-related emissions (1872.9 MMT $CO_2$-eq.; including electricity consumption emissions in the industry sector[83]) in 2022.

Overall, the optimal circular PET and PO plastic packaging systems showed lower GHG emissions and CED impacts than the fossil-dependent linear economy. Compared with the linear economy, optimized circular PET and PO packaging systems exhibited savings of GHG emissions and CED impacts by up to 28% and 46%, respectively, with increased MRF collection proximity to 500 km. In a broader context, these optimal GHG emissions and CED savings represent a reduction of 0.16% and 0.49% compared to 2022 annual U.S. GHG emissions and energy consumption,[83,84] respectively. Moreover, these optimal GHG emission savings would represent a reduction of 0.53% compared to annual U.S. industry-related GHG emissions.[83]

Considering the 2019 average market prices and higher heating values of PET and PO plastics, based on ref. 5, the linear economy of PET and PO plastics packaging would represent an economic loss of US $2.8 billion per year and energy loss of 358 PJ per year to landfills. On the other hand, GHG emissions based optimal circular PET and PO plastics packaging systems represent an economic loss of only US $0.2 to 0.8 billion per year and energy loss of 26 to 107 PJ per year to landfills, depending on the MRF collection proximity. This represents an economic gain (or decrease in economic loss) of up to 94% and decreased energy loss of up to 93% to landfills, mainly due to increased closed-loop recycling in circular PET and PO plastics packaging systems. A lower MRF collection proximity would represent higher economic and energy loss to landfills due to higher landfilling rates, compared to a higher MRF collection proximity.

Fig. 6 also shows that increased MRF collection proximity led to increased recycling rates, and thereby reduced use of virgin resins, to reach an optimal recycling rate of greater than 95% and a system circularity of up to 75% (see ESI Fig. S23† for the system's circularity). Increasing MRF collection proximity led to a small decrease in optimal GHG emissions and CED impacts;

however, it had a comparatively greater impact on recycling rates. Increasing MRF collection proximity also led to increased collection and use of recycled PO resins processed *via* pyrolysis technology. This increase in collection, transportation, and pyrolysis of GHG emissions nearly offsets the decreased GHG emissions due to avoided virgin resins, given the small savings of GHG emissions due to pyrolysis compared with virgin resins (see LCIA factors in Table 1).

The optimal PET and PO packaging systems determined by the model comprised of multiple recycling technologies, including mechanical recycling, solvent-based dissolution precipitation (for PET only), and pyrolysis (for polyolefins (PO) only). The GHG emission optimization model selected mechanical recycling (MR) up to the blend limit of 35% to recycle PET and PO packaging waste, with the remaining to be recycled *via* dissolution (PET only) and pyrolysis (PO only) processes. This selection by the optimization model was based on lower GHG emissions of recycled resins produced from these processes compared with fossil-based virgin resins. Overall, depending on the MRF collection proximity, about 50–60% of the total optimally recycled resins were produced by mechanical recycling, 17–18% by the dissolution process, and 22–34% by the plastic-to-plastics pyrolysis process (see Fig. 6b).

In terms of the system optimized on minimum CED impacts (Fig. 6d), the distribution of recycled resins produced *via* different recycling technologies was slightly different, compared to Fig. 6b. Only about 33–40% of the total optimally recycled resins were produced by mechanical recycling technology, 29–31% by the dissolution process, and 31–36% by the pyrolysis process, depending on the MRF collection proximity. This difference was mainly because PET was recycled only *via* the dissolution process rather than a combination of mechanical recycling and dissolution processes, which was in the case of systems optimized on GHG emissions. The primary reason for the avoided PET mechanical recycling in the CED optimization could be higher waste generation (or lower process yield) associated with mechanical recycling, which would bring in more embodied material feedstock energy associated with virgin PET resin. Overall, the optimal GHG emissions and CED impacts decreased slightly with increased MRF collection proximities due to increased recycling rates, consequently reducing the total amount of virgin resins required in the system.

**3.3.1 Optimal GHG emissions and CED: contributions by the supply chain process.** The distribution of optimal GHG emissions and optimal CED impacts based on PET and PO plastic packaging supply chain processes is shown in Fig. 7. The optimal GHG emissions of these plastic supply chains ranged from 25.59 to 25.88 MMT $CO_2$-eq. per year (Fig. 7a–c). These GHG emissions decreased with increased MRF collection

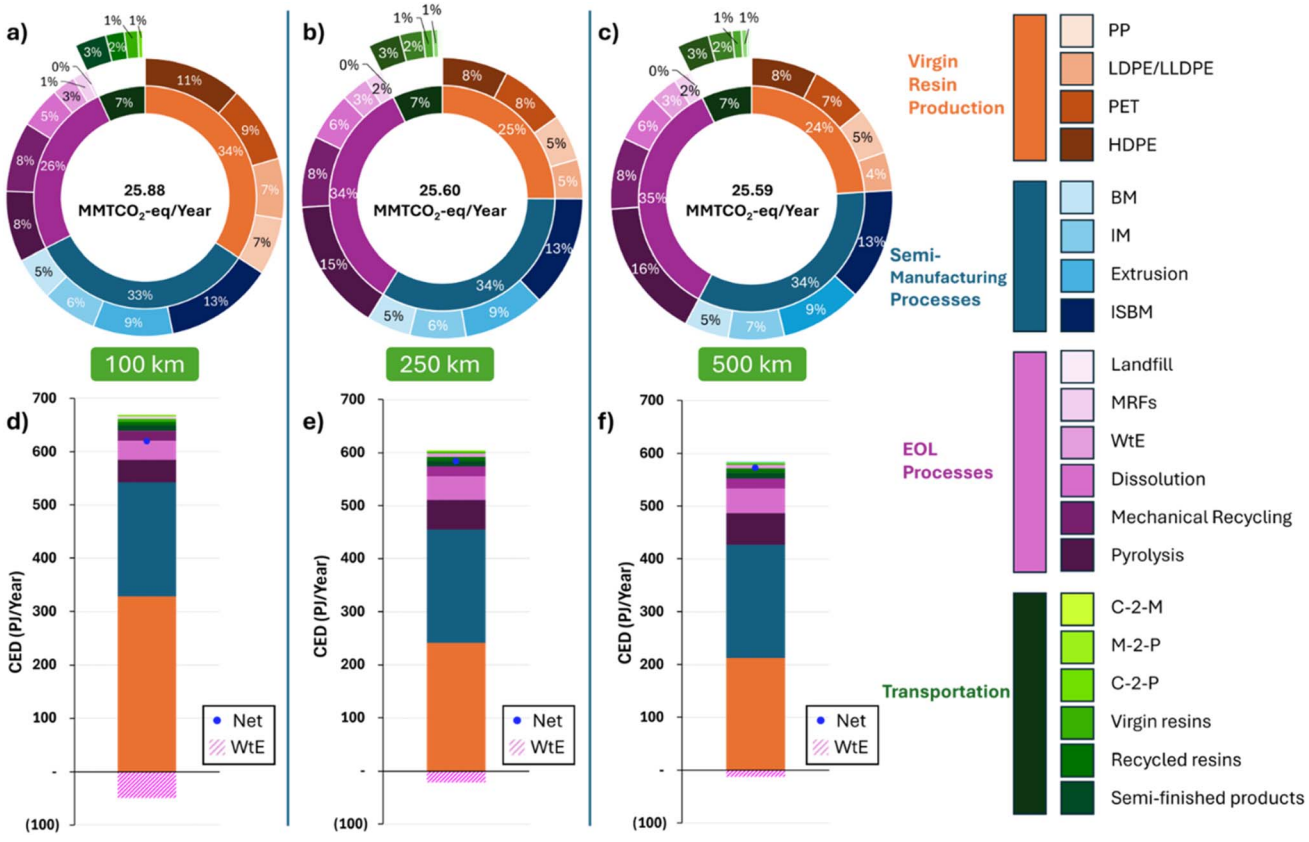



Fig. 7 Optimal GHG emissions (a–c) and optimal CED (d–f) impact distribution based on optimal PET and PO supply chain processes for three MRF collection proximities 100 km, 250 km, and 500 km. The values behind this figure are shown in ESI Tables S13–S15† for GHG emissions and ESI Tables S16–S18† for CED.

proximity. Similarly, the net optimal CED impacts decreased from 619.91 PJ per year to 572.08 PJ per year with increased MRF collection proximity (Fig. 7d–f). Overall, with increased MRF collection distance, the optimal GHG emissions of supply chains showed a shift from virgin resin production (high emissions and energy-intense) processes towards lower emissions and less energy-intense circular EOL processes (Fig. 7a–c).

These impacts could also further be compared with the national GHG emissions and energy consumption in the U.S. The optimal GHG emissions and CED impacts of circular PET and PO plastic packaging supply chain processes would represent 0.40% and 0.57% of the total U.S. GHG emissions and energy consumption, respectively, in the year 2022.[83,84] Similarly, the optimal GHG emissions would represent 1.37% of the total U.S. industry sector related GHG emissions (1872.9 MMT $CO_2$-eq.; including electricity consumption emissions in the industry sector[83]) in 2022.

In terms of the CED impacts (Fig. 7d–f), virgin resin production contributed more to the overall impacts, mainly due to the embodied material feedstock energy of virgin resins, except for the 250 and 500 km proximity cases, wherein the semi-manufacturing process contributed almost equally. The GHG emissions related to transportation were approximately 7% of the system total and were mainly dominated by the transportation of semi-finished PET and PO packaging products, which constitutes the transportation of the largest mass of materials over longer distances compared to the lower mass of materials transported between EOL processes. Therefore, as the proximity distance increased to 500 km, the emissions from transportation of materials did not change appreciably. The GHG emissions and CED impacts of C-2-M, C-2-P, and M-2-P transportation steps were less than 1% of the total optimal impacts across all collection proximities. The GHG emissions and CED impacts of semi-manufacturing processes were constant, as the demand was assumed to be constant for the production of semi-manufactured packaging products. Moreover, the highest magnitude of material flows through semi-manufacturing processes also makes it one of the major contributors to these impacts. The impacts of semi-manufacturing processes could further be reduced by using a renewable electricity grid mix. Our study also shows that additional improvements to impact savings can be achieved (see Section 3.4) by reducing waste generation in all recycling processes, increasing the blend limit on mechanically recycled resin, and reducing the impacts of emerging advanced recycling technologies through continuous research and development.

**3.3.2 Trade-offs between GHG emissions and CED impacts.** The CED impacts of the system optimized on GHG emissions (Fig. 8a) and the GHG emissions of the system optimized on CED impacts (Fig. 8b) were also evaluated, as shown in Fig. 8, for the three MRF collection proximities. Overall, a trade-off was observed between GHG emissions and CED impacts of the two optimal systems. For example, the CED of the system optimized on minimizing GHG emissions was 11% higher (Fig. 8a) than the optimal CED of the system with 100 km collection proximity. Similarly, the system optimized for minimizing CED impacts had 14% higher GHG emissions (Fig. 8b) than optimal GHG emissions with a 100 km collection proximity.

The reason for these trade-offs was mainly the selection of different EOL disposal processes when the system is optimized for minimum GHG emissions *versus* CED impacts. For example, minimizing CED impacts of the system led to selection of incineration with energy recovery as the main EOL disposal process, instead of landfilling, mainly due to the energy savings associated with incineration. Consequently, this leads to as much as 14% higher GHG emissions than optimal GHG emissions (Fig. 8b) due to higher emissions associated with incineration with energy recovery, even after accounting for the avoided emissions due to displaced fossil-based electricity.

Similarly, optimizing the system on GHG emissions led to selection of landfilling over incineration with energy recovery as the main EOL disposal process, due to lower emissions associated with landfilling. However, it leads up to 11% higher CED than the optimal CED impacts (Fig. 8a), as there are zero energy savings associated with landfilling of waste plastics. Landfilling of plastics has lower GHG emissions due to absence of biodegradable carbon in the waste plastics, and it only includes the GHG emissions and CED impacts associated with collection

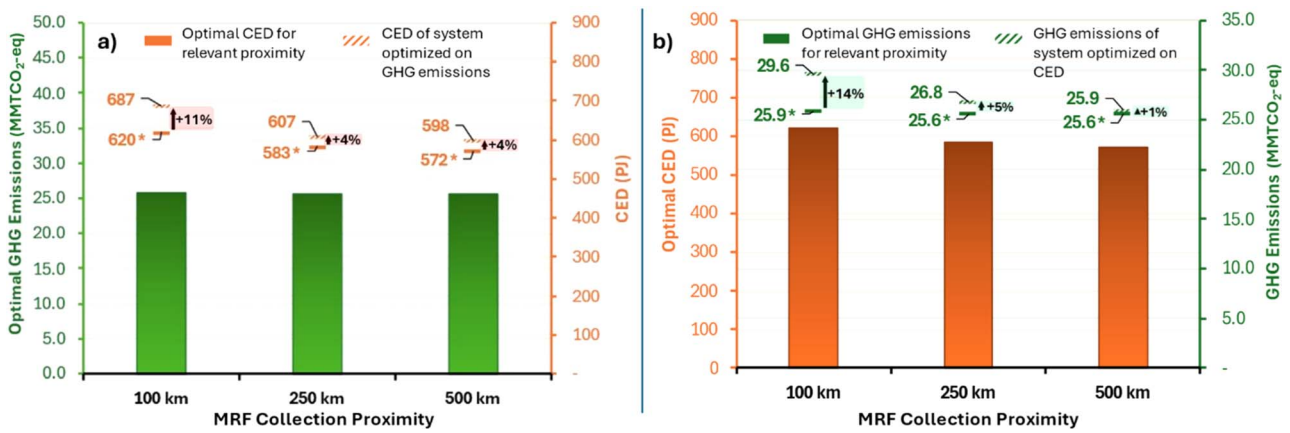



Fig. 8 Trade-offs of energy savings with higher GHG emissions in the optimal PET and PO packaging supply chain systems. Notes: panel (a): optimal GHG emissions on primary *Y*-axis; panel (b): optimal CED on primary *Y*-axis.

and transportation of waste plastics to the landfill facility, along with landfill equipment operation, which are relatively small.

Compared with 250 km and 500 km MRF collection proximities, the system optimized on 100 km proximity showed greater trade-offs between GHG emissions and CED impacts (Fig. 8), lower recycling rates (Fig. 6a), and higher need for virgin resins (Fig. 6b and d). The system optimized on 500 km proximity, however, has the highest recycling rates, lowest use of virgin resin, highest circularity, and smaller trade-offs between GHG emissions and CED impacts when compared with other collection proximities.

### 3.4 Scenario analysis

The summary of all optimal GHG emissions, CED impacts, and recycling rates for all the modeled scenarios is shown in Fig. 9. In addition to these metrics, Tables S19 and S20† summarize landfilling rates, incineration rates, recycled content, system circularity, virgin resins, and recycled resins for all modeled scenarios. The first scenario, shown in Fig. 9, represents the optimal system with base assumptions related to MRF collection proximity (100 km) and blend limit on mechanically recycled resins (35%). Overall, among all scenarios, except for linear economy and 100% WtE scenarios, the system optimization model selects the collection and recycling of PET and PO packaging waste *via* either mechanical recycling or a combination of mechanical recycling and emerging advanced recycling technologies, depending on the scenario (Fig. 9).

Among all the scenarios, there are several key points worth mentioning with regard to improving system metrics. Increasing the MR blend limit to 75% and to no limit improved GHG emission savings from 27 to 32% and CED from 41 to 42% while slightly increasing recycling rates. A 100% WtE for all collected packaging plastics increased GHG emissions by 37%

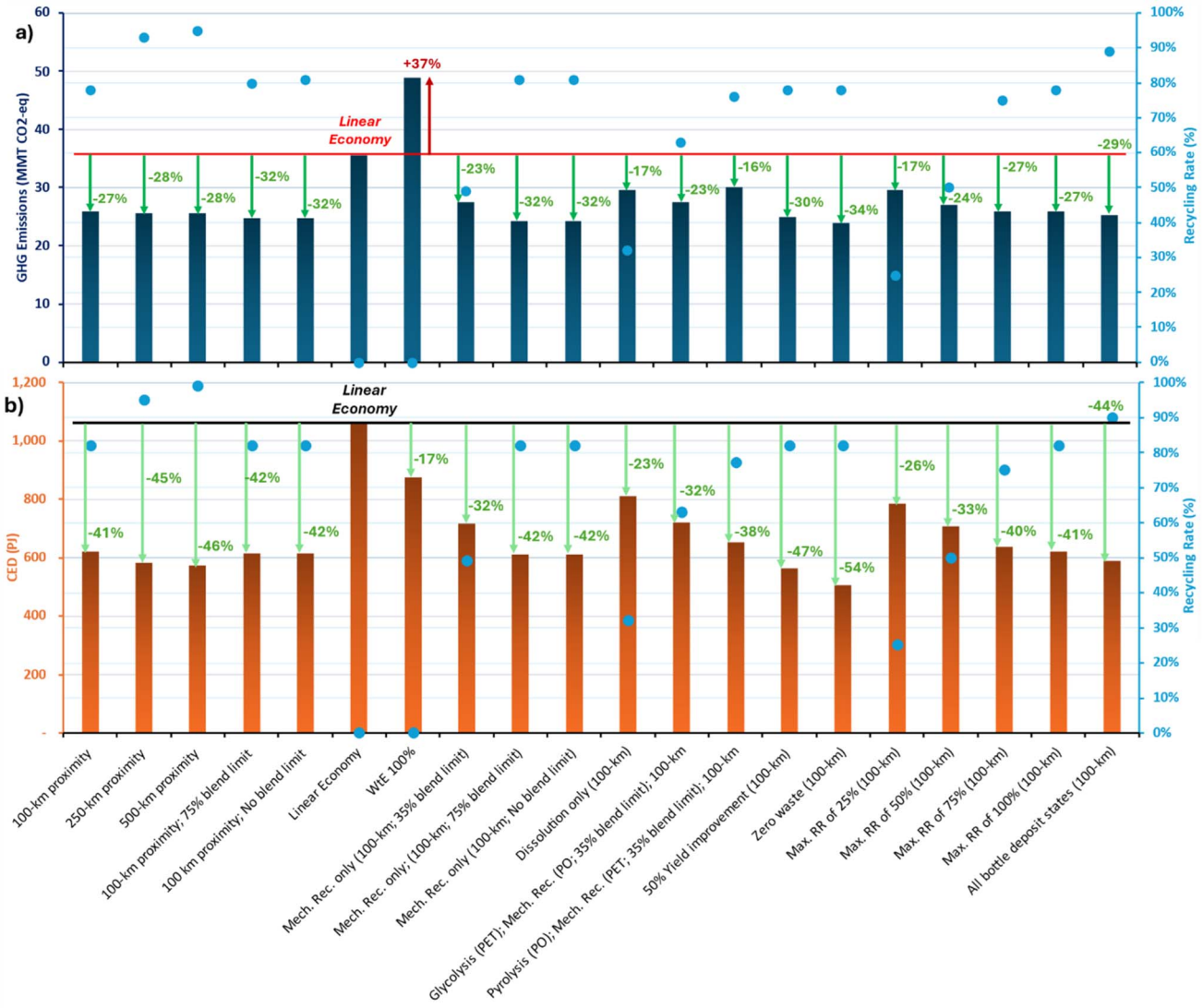



Fig. 9 Summary of optimal GHG emissions (primary *Y*-axis; in panel (a)) and CED impacts (primary *Y*-axis; in panel (b)) along with the respective recycling rates (secondary *Y*-axis) for all the scenarios modeled. Note: the dashed line represents the impacts of the linear economy scenario (80% landfill; 20% WtE) that serves as a basis for comparison. The percents of GHG emissions and CED savings relative to the linear economy scenario are shown in (a and b), respectively.

while reducing CED by 17%. The scenario with the greatest savings of GHG emissions and CED and highest circularity is "Zero waste (100 km)", although the recycling rate was unchanged from that in the "100 km proximity" scenario (see Fig. S23† for circularity). Another important set of scenarios are increasing the recycling rate (RR) from 25 to 100%, for example through mechanisms such as policy mandates, in which GHG emission and CED savings progressively improve, although the rate of improvement is greater from 25 to 50% RR. Also, implementing bottle deposit programs in all states (last scenario, no. 20) led to higher collection rates of PET *via* bottle deposit collection programs (C-2-P), compared to the optimal system with 100 km proximity (scenario no. 1). All these scenarios are further presented in Section S3.4.1 and S3.4.2 of the ESI document,† showing the effects of maximum recycling rates (Fig. S24†) and different EOL management scenarios (Fig. S25†) on system performance.

In this optimization modeling study, the results of circular systems performance depend on the availability and quality of the input data to the system model. We estimate an overall uncertainty of ±16% and ±28% in the input MFA data and LCA data, respectively, using previously developed uncertainty frameworks (see ESI Section S3.4.3†). Due to limitations in computational resources, we did not propagate these model input data errors through to the predicted GHG emissions and CED results. Future research should include such error propagation to yield a distribution of the system metrics. Moreover, the systems analysis framework applied in this study could further be expanded to other types of plastics, additional circular economy strategies, and geographical regions of interest with sufficient availability of the relevant life cycle inventory, material flow, and geographical location datasets. As research into advanced/chemical recycling technologies continues to emerge with regard to higher quality LCA and techno-economic data, the accuracy of systems analysis and optimization modeling such as that presented here will also improve, yielding benefits to stakeholders and decision-makers in circular plastics and sustainability. Further development of systems analysis and optimization should follow a path to include techno-economic results on individual processes and the incorporation of socioeconomic data such as employment changes, wages, and system revenues for these emerging closed-loop processes.

# 4 Conclusions

A comprehensive systems analysis and optimization was conducted for the U.S. PET and PO packaging plastics supply chain in a closed-loop circular economy, and the results were compared against a linear economy. The scope of our system model included all the major upstream and downstream processes, including emerging advanced/chemical recycling technologies for which LCIA emission and energy factors could be derived given the nascent status of technology development in this field. Moreover, our study evaluated systems-level circularity, recycled content, and needs for fossil-based virgin PET and PO resins in the U.S.

A transition towards a closed-loop circular economy of U.S. PET and PO plastics packaging was found to be more environmentally sustainable than the fossil-dependent linear economy. In a broader context, such a linear-to-circular economy transition would represent savings of GHG emissions and CED impacts resulting in a reduction of 0.16% and 0.49% compared to annual U.S. GHG emissions and energy consumption, respectively. The optimal circular plastics packaging systems showed increased recycling and processing of materials at EOL *via* closed-loop mechanical and emerging advanced/chemical recycling processes rather than traditional disposal processes. A system comprised of higher recycling rates showed lower environmental impacts, higher systems circularity, higher recycled content, and lower reliance on fossil-based virgin plastics. Moreover, the model showed that a linear-to-circular transition resulted in a shift of plastics material flows from high-emission and energy-intense supply chain processes towards lower-emission and less energy-intense circular supply chain processes. A trade-off between GHG emissions and CED impacts was observed due to selection of different EOL processes, further suggesting that the sustainability of circular plastics systems depends on the nature of the processes involved and will entail resolution of the inherent trade-offs among metrics.

To achieve a closed-loop circular economy of plastics and associated lower GHG emission and CED, the existing U.S. plastics recycling infrastructure will need to be expanded to process more EOL materials. The system optimization model prescribed recycling of plastics *via* mechanical recycling technology up to the blend limit of 35%, with the remaining to be recycled *via* emerging advanced/chemical recycling technologies. This shows that emerging recycling technologies such as solvent-based dissolution precipitation and fast pyrolysis processes would need to be integrated with existing mechanical recycling technology to minimize environmental impacts of the system and meet the demand for high-quality recycled plastics rather than relying on fossil-based virgin plastics.

We conclude further that additional useful information regarding the closed-loop U.S. plastics supply chain can be gleaned from the output of the optimization model. This information includes numbers, capacities and optimum locations of current and future MRFs, distances and amounts of material movement from counties to MRFs and PRFs and from MRFs to PRFs, and the relative importance of urban *versus* rural infrastructure in achieving high circularity. Furthermore, it was found that environmental impacts of material transportation between the EOL recycling facilities is small compared to the impacts of material movement upstream of EOL throughout the U.S. This knowledge encourages more recycling with the collected material being transported over long distances to achieve high circularity, and thereby reduce the negative impacts of bringing virgin plastic resins into the system.

Finally, achieving high recycling rates and a successful transition towards the circular economy would require large investments. However, high circularity would have the potential to realize positive socioeconomic outcomes such as increased employment and wages as well as reduced fossil resource

consumption. Solution strategies to achieve higher recycling rates may include, but are not limited to, careful design for recycling of plastic products (*e.g.* mono-material packaging products and appropriate consistent labelling), increased access to recycling programs, better consumer education and high participation in recycling, reduced contamination in curbside recycling, and increased process efficiency and economics of emerging advanced recycling technologies through continuous research and development.

## Data availability

The data supporting this article have been included as a part of the ESI document.†

## Author contributions

Utkarsh S. Chaudhari: writing – original draft, writing – review & editing, conceptualization, methodology, formal analysis, investigation, visualization. Abhishek Patil: methodology, software, formal analysis, data curation, investigation, visualization, writing – review & editing. Tasmin Hossain: methodology, software, data curation, investigation, writing – review & editing. David Watkins: supervision, validation, writing – review & editing. Damon Hartley: supervision, validation, writing – review & editing. Barbara Reck: writing – review & editing. Robert Handler: writing – review & editing. Anne Johnson: writing – review & editing. Vicki Thompson: writing – review & editing. David Shonnard: writing – review & editing, conceptualization, methodology, validation, resources, supervision, project administration, funding acquisition.

## Conflicts of interest

The authors declare no competing interests.

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