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Complete List of Authors:	Ma, Jiaze; University of Wisconsin, Chemical and Biological Engineering Tominac, Philip; University of Wisconsin-Madison, Chemical and Biological Engineering Olafasakin, Olumide; Iowa State University of Science and Technology, Mechanical Engineering Aguirre-Villegas, Horacio; University of Wisconsin, Biological Systems Engineering Wright, Mark; Iowa State University, Mechanical Engineering Benson, Craig; University of Wisconsin, Civil and Environmental Engineering Huber, George; University of Wisconsin, Chemical and Biological Engineering Zavala, Victor; University of Wisconsin-Madison,

Economic Evaluation of Infrastructures for Thermochemical Upcycling of Post-Consumer Plastic Waste

Jiaze Ma[1], Philip Tominac[1], Horacio Aguirre-Villegas[2], Olumide Olafasakin[3], Mark Mba-Wright[3], Craig H. Benson[4], and George W. Huber[1], Victor M. Zavala[1*]

(1) Department of Chemical and Biological Engineering, University of Wisconsin-Madison

(2) Department of Biological Systems Engineering, University of Wisconsin-Madison

(3) Department of Mechanical Engineering, Iowa State University

(4) Department of Civil and Environmental Engineering, University of Wisconsin-Madison

(*) Corresponding Author: victor.zavala@wisc.edu

Summary

Thermochemical technologies, such as pyrolysis, offer a potentially scalable pathway for upcycling diverse types of plastic waste (PW) into value-added chemicals. However, deploying these technologies in waste management infrastructures is not straightforward because such systems involve a wide range of interdependent stakeholders, processing facilities, and products. In this work, we present a holistic optimization framework that integrates value-chain analysis, techno-economic analysis, and life-cycle analysis for investigating the economic viability and environmental benefits of upcycling infrastructures that collect, sort, clean, and process post-consumer PW for producing virgin polymer resins. The framework is applied to a case study in the upper Midwest region of the US. Our analysis reveals that the infrastructures are economically viable and could activate a regional circular economy that generates over 1 billion USD in annual profit. Moreover, our analysis reveals that this economy can reduce the carbon footprint of PW incineration by half. Our framework also determines the inherent values of post-consumer PW and of derived products such as plastic bales and pyrolysis oil; we find that, in these infrastructures, PW becomes a highly valuable feedstock with a market value of 500 USD/tonne. We discuss how this market value can generate incentives that foster more effective waste pre-

sorting practices by consumers that can help bypass material recycling facilities and increase total system profit.

Introduction

The high versatility of plastics has revolutionized modern life; these materials touch nearly every aspect of our daily lives (e.g., food packaging, transportation, and construction)[1,2]. Plastics is a highly profitable and growing industrial sector; to give some perspective, US plastic production increased more than *20 times* between 1960 and 2020[3,4]. This rapid growth has outpaced the development of plastic waste (PW) disposal and recycling infrastructures. Currently, most plastic globally is either put in a managed landfill, put in an unmanaged dump, or is incinerated (less than 5% is recycled)[5]. To give some perspective on how recycling is lagging, US PW production has increased *90 times* between 1960 and 2020[6]. In 2016, it was estimated that the US per-capita PW production was 130 kg per year[7]. The wide variety of available plastic products has been a major obstacle in developing technologies that can handle ever-growing waste streams.

Landfilling and incineration are waste disposal pathways that are scalable but that also lead to a wide range of environmental and social problems[8]. For instance, PW incineration (conducted in waste-to-energy facilities) releases large amounts of carbon dioxide and of other toxic gases such as dioxins, furans, halogens (which are neurotoxic and carcinogenic)[9,10]. Existing recycling infrastructures are primarily focused on mechanical recycling technologies and target polyethylene terephthalate (PET, plastic type #1) and high-density polyethylene (HDPE, type #2). In such infrastructures, post-consumer waste is delivered to material recycling facilities (MRFs) that typically separate PW from other recyclables (e.g., glass, metal and carboard) and that sort PW into three major streams: PET bottles, HDPE bottles, and a residual (catch-all) stream that includes plastic types #3–#7 and other lower-value types such as non-bottle PET and HDPE plastics. The *catch-all stream comprises over 80% of post-consumer PW* and is typically landfilled or incinerated, due to the lack of effective processing technologies that can handle these complex waste mixtures[11,12,13]. PET and HDPE bottles obtained from MRFs, on the other hand, can be recycled mechanically and hence have an existing market and higher value[14]. This recycling pathway delivers MRF bottles to plastic reprocessing facilities (PRFs) where they are washed, ground, melted, and extruded to obtain polymer resins that can be used to produce new bottles or other products[15]. Mechanical recycling of rigid PET and HDPE is a well-established industry with profitable value-chains but only address a small portion of plastic waste[16]. The limited availability of flexible PW recycling infrastructures also exacerbates resource depletion and leads to large carbon footprints; to give some perspective, the production of 1 kg of plastic requires approximately 2 kg of crude oil[8].

Chemical recycling can overcome limitations of mechanical recycling; such technologies break down plastic waste into diverse chemical constituents which can be used in different applications such as production of virgin resins or fuels. Chemical recycling is a flexible and scalable waste processing pathway in the sense that it can handle can handle complex plastic waste mixtures generated by MRFs[17]. Thermochemical technologies are chemical recycling approaches that have been the subject of intense research. Here, we focus our attention on pyrolysis technologies, which have been the subject of extensive experimental and modeling research, including diverse aspects such as reactor design[18], process intensification[19,20,21], catalyst development[22,23,24], and techno-economic analysis[25,26].

Pyrolysis involves the breakdown of long-chain polymers at high temperatures under anoxic conditions to obtain a liquid product called *pyrolysis oil*, which is a valuable feedstock that has diverse uses. Pyrolysis oil has similar properties to petroleum products (e.g., calorific value, viscosity, density) and has value as a substitute for crude-sourced petroleum in some applications[27]. The 2020 market for pyrolysis oil was valued at 300 million USD[28] with the pyrolysis oil being valued as a feedstock to produce fuels[29,30,31]. Within a PW recycling context, it is important to note that pyrolysis has a higher conversion efficiency and a lower carbon footprint than waste incineration[32,33]. In addition, pyrolysis can accommodate mixed PW streams (including both rigid and flexible plastics) and thus has the potential to process a much greater fraction of waste than mechanical recycling[17]. However, plastic pyrolysis is a more sophisticated and cost-intensive technology than incineration and its effectiveness is highly dependent of the PW mixture composition and of the presence of impurities (e.g., chlorine).

Pyrolysis oil can be incorporated into a plastic recycling value-chain that produces virgin polymer resins. Here, the pyrolysis oil is first conditioned, processed, and broken down into monomers such as ethylene and propylene via steam cracking.[34] The monomers can then be polymerized to obtain virgin resins and thus create a circular plastic economy[35]. The products obtained from this plastic recycling strategy are chemically new are more valuable than the plastic wastes. This plastic recycling strategy turns the plastic waste into products of higher quality, it is thus called plastic *upcycling* (In opposite to plastic recycling where the quality of plastic is slightly decreased, such as mechanical recycling). To give some perspective on the potential scale that this pathway can achieve, we note that upcycling waste that would otherwise be deposited in landfills could produce 6 million tonnes of virgin PE per year, equivalent to 26% of the 2019 US market for this plastic[36]. The potential revenue of an upcycling infrastructure at this scale would be worth an estimated 13 billion USD annually and it could potentially reduce the carbon footprint of the polymer sector by roughly 5 million tonnes annually. To the best of our knowledge, there are no studies that analyze the deployment of thermochemical technologies into waste management infrastructures. These studies are necessary for understanding the economic viability, market potential, environmental benefits, and obstacles/synergies of plastic thermochemical technologies as well as understand interactions with diverse stakeholders and infrastructure elements. Moreover, it is important to understand how the efficiency of pyrolysis processes and of other elements of the value-chain can impact overall economic viability and environmental benefits.

In this work, we present an infrastructure optimization framework that incorporates value-chain analysis, techno-economic analysis (TEA), and life-cycle analysis (LCA) to investigate the economic viability and environmental benefits of upcycling infrastructures that use thermochemical technologies to generate virgin polymer resins from post-consumer PW. A high-level view of the infrastructures under study is provided in **Figure 1**. Our framework captures diverse processes, products, and stakeholders involved in these value chains (e.g., waste generators, waste collection, sorting, cleaning, grinding, thermochemical processing, and product transport). We use our framework to determine optimal infrastructure layouts that achieve maximum total economic surplus (i.e., total system profit). We also use our framework to quantify environmental benefits and to analyze the inherent value of PW as a resource, which is a complex function of costs associated with transport and processing, technology efficiencies, geographical concentration of waste, and market externalities. Moreover, we use our framework to study how the value-chain generates incentives that can impact consumer behavior and foster waste pre-sorting. For instance, in the US, post-consumer PW is typically mixed with other recyclables (e.g., cardboard and glass) and contaminants (e.g., food residues); as such, separation and cleaning in MRFs is needed. The cost of these pre-processing services is high (in the US it is estimated at 3 billion USD annually[37]) and could be mitigated via pre-sorting practices at the source (by consumers).

Results

The infrastructure optimization framework proposed is used for conducting a case study that focuses on PW generated in the upper Midwest region of the US (comprising the states of Wisconsin, Illinois, Minnesota, and Iowa). Our goal is to design infrastructures (determine optimal number, capacity, and placement of MRFs, PRFs, pyrolysis, steam cracking, and polymerization technologies) that process all the waste generated in the region and that maximize the total economic surplus of the system. The economic surplus (total profit) captures all revenue generated from the production of value-added products (e.g., virgin polymer resins) as well as all costs associated with processing and transportation. The framework interprets the upcycling infrastructure as a value-chain (a market) in which stakeholders exchange products to create economic value (i.e., generate higher-value products from lower-value products). This interpretation allows us to obtain infrastructure designs that are economically viable and that reveal the inherent value of waste, products, and byproducts generated and exchanged within the value-chain. Importantly, inherent values determined in our framework are consistent, in the sense that they cover all costs associated with processing and transportation and capture system externalities (e.g., markets outside the value-chain such as electricity markets). Details on the optimization framework are presented in the Supplementary Information. Given the large number of products and processes involved in the studied infrastructures, we use acronyms to refer to them in a compact manner and we provide an acronym list in the Appendix. All code and data needed for reproducing the results are shared as open-source software.

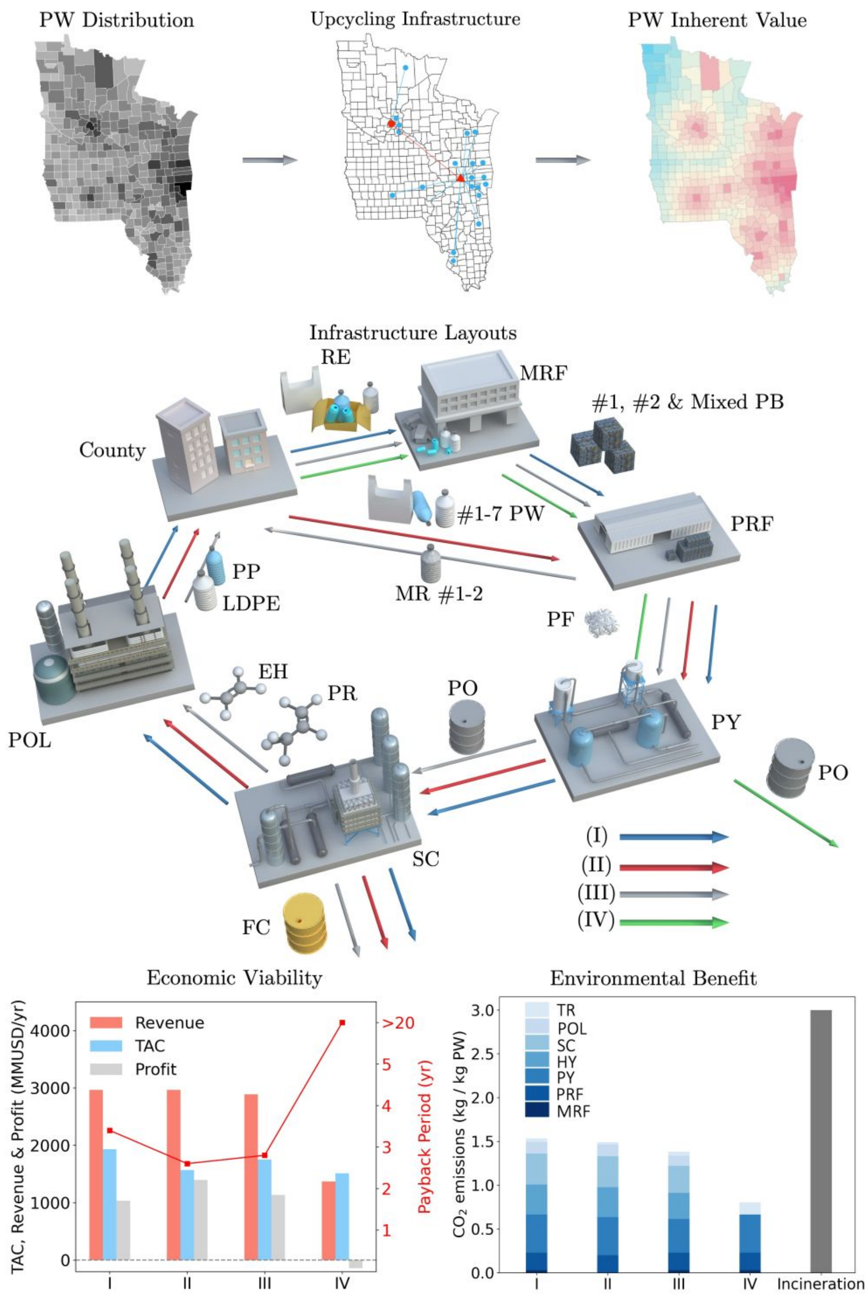

Figure 1. Schematic of computational framework for infrastructure optimization. Top: Our framework takes data on post-consumer plastic waste (PW) distribution and solves an optimization problem to identify best locations and sizes for processing technologies and the inherent values for PW and derived products. **Middle:** We explore four different infrastructure layouts, each comprising a combination of five core processing technologies. MRFs collect recyclables (RE) from consumers and separate PW. PW is then baled (PB) and shipped to PRFs where it is washed and ground into plastic flakes (PFs). The PFs are then converted into pyrolysis oil (PO) in a pyrolysis (PY) process, then into olefins (EH, PR) in a steam-cracking process (SC), and then into virgin polymers (PP and LDPE) in a polymerization (POL) process. Detailed descriptions of the technologies are included in the Supplementary Information; acronyms are summarized in the Appendix. (I) Base layout in which PW is embedded in a waste stream that is separated in the MRF; the separated PW is baled and is sent to a PRF and subsequent chemical upcycling to produce virgin polymer resins.

(II) Layout in which consumers pre-sort PW, thus bypassing the MRF and sending PW directly to the PRF. (III) Layout in which PET/HDPE bottles are separated in the MRF and PRF for mechanical recycling, and the remaining PW types (catch-all stream) are processed via PY. (IV) Layout as in (I) but PO is the final product of the value-chain. We highlight that these infrastructure layouts do not show geographical aspects (e.g., waste source, technology location, and transportation). **Bottom:** We use our framework to determine the economic viability and environmental benefits of the different infrastructure layouts.

High-Level View of Infrastructure Layouts. We first provide a high-level view of the optimal infrastructure designs associated with the four layouts analyzed (see **Figure 1**). As we see in the next section, the optimal designs involve different capacities, number, and location of technologies. In this section, we only analyze the total input-output product flows for all technologies. The product flows for the different layouts are summarized as the Sankey diagrams of **Figure 2**. These diagrams provide a high-level perspective on how waste is fed into the value-chain to obtain diverse intermediary and final products (main and byproducts). The diagrams also help illustrate interdependencies between the products and technologies that arise in these value-chains and the overall magnitude of the product flows.

Layout I is used as a base infrastructure. In this layout, MRFs process a waste stream that contains diverse recyclables to obtain PBs (containing types #1-7). The PBs are processed in a PRF to produce PFs. Pyrolysis of the plastic flakes (PF) results in pyrolysis oil (PO) with a yield factor of 77% (relative to the feed of flakes). Hydrotreatment of the PO produces naphtha and atmospheric gas oil (AGO) with yield factors of 39% and 23% (relative to the feed of PO). Naphtha and AGO undergo steam cracking (SC) to produce PP and EH (yield factors of 9.9% and 16.2%). Polymerization of these monomers produces PP and LDPE at yield factors of 9.6% and 15.7%. The yield factors for all technologies were obtained from available literature reports and from engineering insight. More details can be found in the Supplementary Information.

Layout II is used to explore the economic impact achieved by a change in consumer behavior; here, we assume that consumers pre-sort plastics from recyclables at the source (at their homes) and thus the MRFs and their associated processing costs can be bypassed. This layout is otherwise identical to layout I, producing the same products and byproducts with the same yield factors.

Layout III is used to explore whether a hybrid infrastructure consisting of mechanical recycling and chemical upcycling provides significant economic advantages over Layout I (based purely on upcycling). In this layout, the separation of PET/HDPE bottles at MRFs reduces the yield factors of plastic flakes to 88.4% and to 68% for pyrolysis oil (bottles are sent for MRF to mechanical recycling). Final product yields are reduced to 8.4% and 13.9% for PP and LDPE. This highlights a tradeoff created by diverting bottles into a different recycling process (reduced feedstock for chemical upcycling but potentially higher values can be achieved for products obtained via mechanical recycling).

Layout IV is used to explore the economic viability of producing PO without any additional upcycling. This is done to consider the possibility that oil is used for other purposes like making liquid fuels in a petroleum refinery. This also allows us to understand what prices of pyrolysis oil can make the infrastructure viable. To capture this effect, we assume that oil is transported to the Gulf region of Texas, where most petrochemical infrastructure is present.

In each layout, we account for different byproducts and waste streams and associated revenues or disposal costs. We highlight that pyrolysis produces hydrochloric acid, char, and pyrolysis gas (a mixture of light hydrocarbons such as CO and CH_4). The hydrochloric acid requires neutralization with calcium hydroxide (at an additional cost) but the char and pyrolysis gas can be recycled as fuel sources to provide heating. We also highlight that the hydrotreating process results in an oil loss of about 15% as wax. The SC leads to the generation of a mixture of C_4 chemicals (butane, butylene), fuel oil, fuel gas, and pyrolysis gasoline. Details on these additional products are provided in the Supplementary Information.

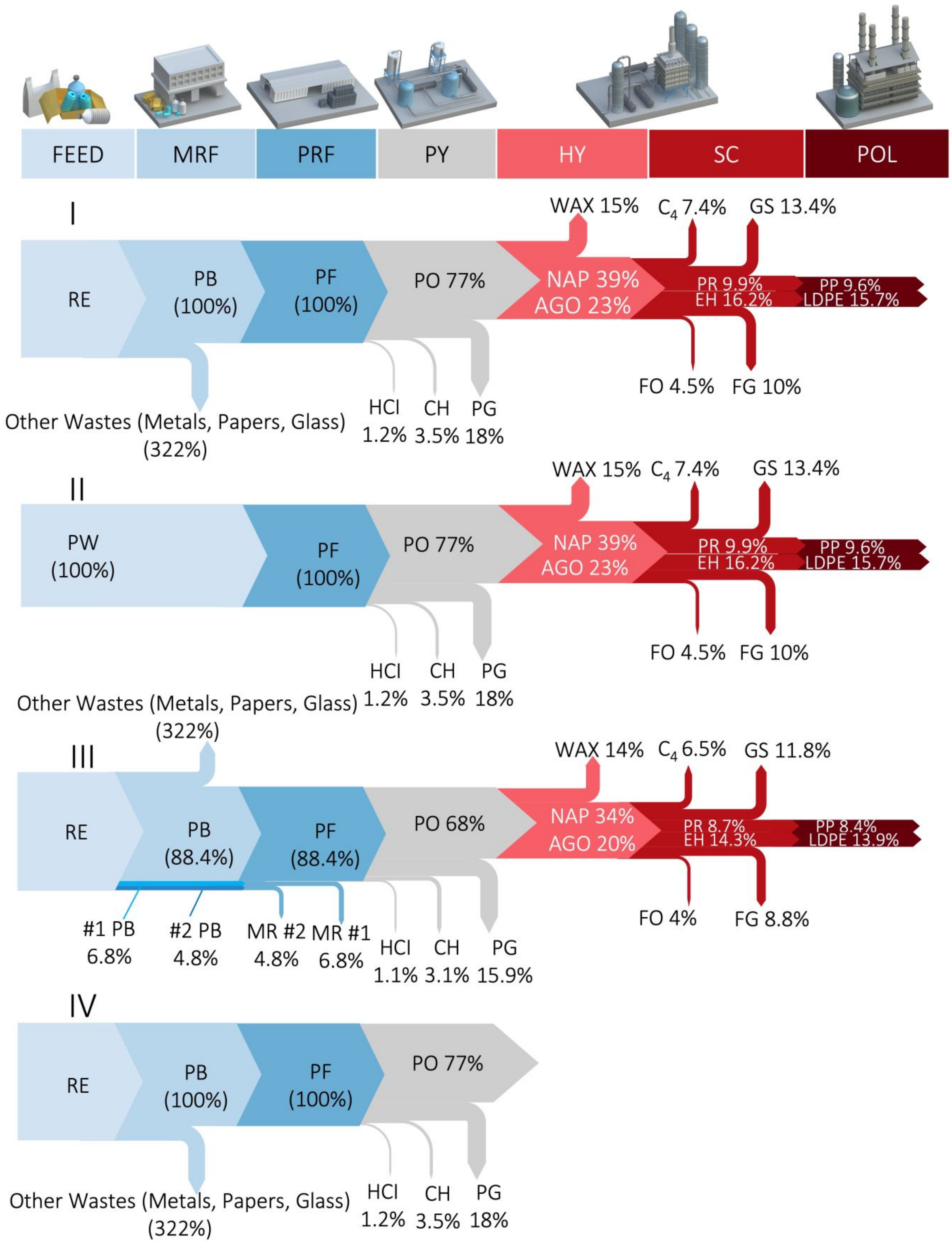

Figure 2. Sankey diagrams summarizing flows of waste and products for infrastructure layouts. In Layouts I and II, plastic waste is converted into virgin resins (25%), fuels (28%), and chemicals (22%). Layout III diverts PET

(6.8%)/HDPE (4.8%) to mechanical recycling and transforms remaining plastic waste into virgin polymer resins (22%), fuels (25%), and chemicals (21%). Layout IV transforms plastic waste into pyrolysis oil (77%) and other byproducts.

Geographical View of Infrastructure Layouts. Figure 3 presents the spatial (geographical) distribution of plastic waste in the upper Midwest region. We estimated this distribution based on population density and based on the average waste composition obtained from the US Environmental Protection Agency (EPA). As expected, waste is concentrated in dense population centers such as Milwaukee, Chicago, and Minneapolis. High waste concentration facilitates collection and processing in centralized facilities. Our aim is to locate infrastructure elements (MRFs, PRFs, PYs, SCs, and POLs) in a strategic manner to provide sufficient capacity to process all the PW produced in the region, while minimizing transportation/capital costs and maximizing the revenue of the generated products. This involves resolving a complex conflict between transportation costs (reduced by deploying more and small technologies) and capital costs (reduced by deploying few and large technologies). This conflict is resolved by solving an optimization problem that maximizes the total economic surplus of the infrastructure; this problem is a mixed-integer linear program, details are provided in the Supplementary Information.

Figure 3 also presents the optimal infrastructure designs for each layout. For layouts I, II, and III, the solutions are nearly identical. As expected, MRF locations follow population patterns, with a cluster of large units in the East to serve dense urban centers and a few units of various capacities located in the less populated areas of the Northwest. We also note that there are fewer and more centralized PRFs, with one large unit serving the entire East and a smaller unit serving the entire Northwest. This indicates that MRFs are placed to minimize transport cost and are used to extract/concentrate plastic waste from recyclable that is sent to PRFs for further processing. The optimal design also places pyrolysis units at the same locations as PRFs, thus eliminating transportation costs. A single steam cracking unit and polymerization facility are placed in the East at the same location as the pyrolysis site and serve the entire upper Midwest region. This sizing and placement strategy arises from the fact that these facilities are highly capital intensive and are benefited by economies of scale. For layout IV, we observe that that geographical distribution of MRFs is similar, but there is only a single large PRF and pyrolysis facility that produces PO; this difference is driven by transportation costs associated with shipping PO over long distances (to Texas).

The identified infrastructure designs deploy new MRFs and does not use existing MRFs, this is because we do not have enough information (e.g. capacity and types of equipment) to account for the economics of the existing MRFs. The MRF spatial distribution closely follows the yield factors and transportation costs associated with plastic waste separation. Specifically, each tonne of recyclables shipped to an MRF only yields 0.23 tonne of plastic waste (transportation costs thus need to be minimized). On the other hand, it is possible to have large, centralized PRFs because plastic bales are more economical to transport (involves smaller flows than recyclables). Centralization of the thermochemical technologies (pyrolysis and steam cracking) and of polymer synthesis is driven by capital costs (economies of scale need to be exploited). We highlight that the infrastructure layouts identified are optimal but are not necessarily unique (different designs can achieve the same optimal economic surplus), as the solutions of mixed-integer optimization problems are in general non-unique. As such, we use our solutions simply to illustrate aspects that the upcycling infrastructures aim to capture and optimize.

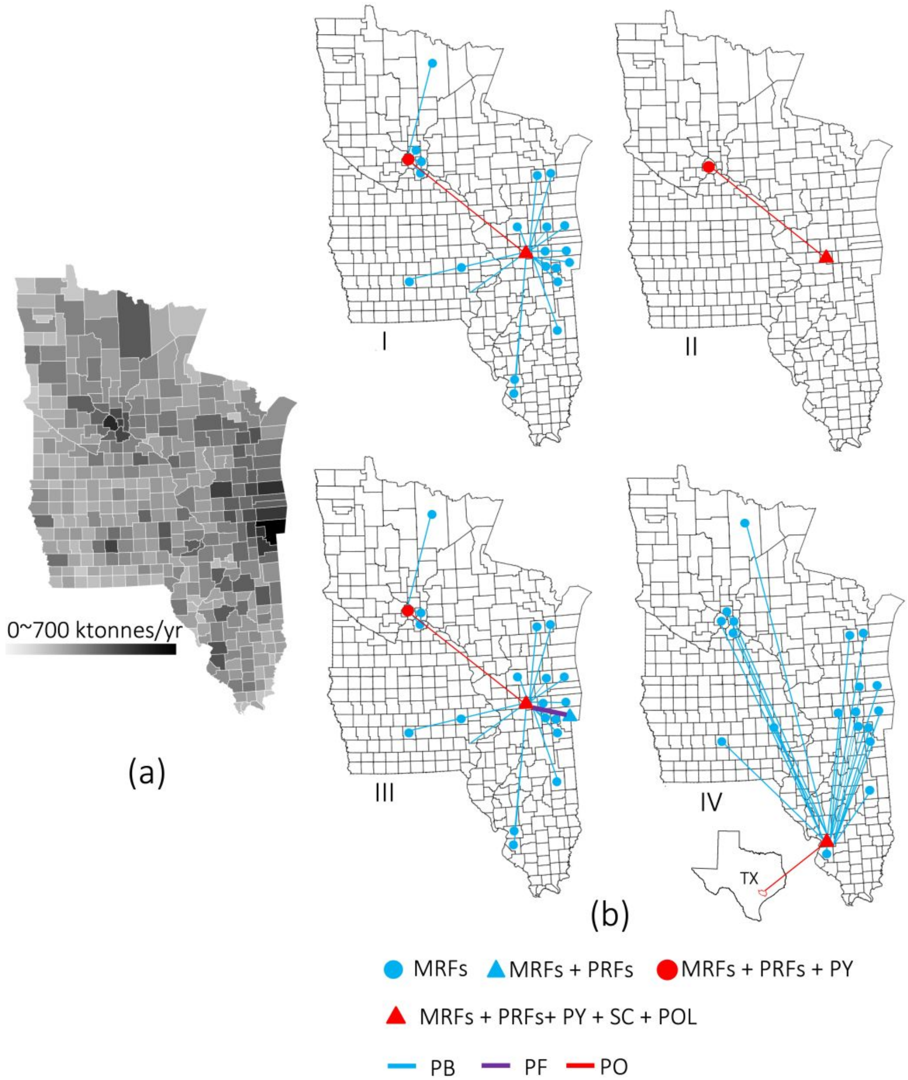

Figure 3. Geographic distribution of plastic waste and of infrastructures. (a) Post-consumer plastic waste distribution. (b) Designs of plastic upcycling infrastructures. The infrastructure designs show optimal locations for different technologies that aim to maximize the total economic surplus. In general, we see that the optimal designs aim to minimize transportation costs by deploying multiple distributed MRFs and aim to minimize capital costs by deploying a few centralized PRFs, pyrolysis, steam cracking, and polymerization facilities (centralization benefits from economies of scale).

Economic Viability of Infrastructure Layouts. We now analyze the economic viability of the different infrastructure layouts; the results are summarized in Figure 4.

In **Figure 4a** we can see that layouts I and II generate almost 3 billion USD annually in revenue. The bulk of these revenues comes from virgin resin sales (LPDE/PP). Sales of other value-added chemicals (byproducts) contribute an additional 540 million USD, and fuel sales contribute an additional 366 million USD; in percentage terms, these revenue streams correspond to 65, 16, and 11% of the total. In layout III, we observe a similar breakdown of total revenue (albeit lower), with a fraction of the LDPE/PP revenues replaced with those of mechanically recycled PET/HDPE (which together contribute 268 million USD in revenue). This illustrates that upcycling provides greater revenue than mechanical recycling. Layout IV achieves roughly half the revenue of the other three layouts. In other words, upcycling infrastructures that expand the value-chain can double the revenue that those limited to the production of PO. We note that these results assume a value for virgin resins and for pyrolysis oil that follow current markets (captured in our model as externalities). Our goal here is not to provide final recommendations but simply to illustrate how the proposed infrastructures can exploit different types of markets to generate revenue and to highlight the economic trade-offs that are involved in selecting a market over another.

In **Figures 4b** and **4c** we provide the total annual cost (TAC) associated with our optimal designs. Figure 4b compares the layouts, while **Figure 4c** groups the various cost items into capital, operating, and transportation costs. We found that layout II provides 15% savings in TAC relative to layout I; this is because layout II avoids MRF investment and operating costs (achieved via pre-sorting of waste). This highlights the potential economic impact of changing social behavior. For layout III, we observe that using mechanical recycling reduces the TAC by 176 million USD. This reduction is not as high as that achieved with layout II, as the MRFs are still the greater contributor to TAC. However, it is interesting to observe that the revenue of layout III decreases by less than the TAC, suggesting that the hybrid infrastructure approach is the most profitable. These results highlight the benefits of diversifying the portfolio of waste processing pathways. Because a fraction of plastic waste (#1-#2 rigid plastic bottles) is diverged to mechanical recycling, the sizes of capital-intensive thermochemical facilities of layout III are smaller than those of layout I. We found that the annualized capital cost of layout III (549 million USD annually) is 18% lower than that of layout I (671 million USD annually) and 11% lower than that of layout II (617 million USD annually). Steam cracking involves a substantial capital investment cost (308 million USD annualized capital cost in layout I) and contributes nearly half of the total capital cost; this indicates that any improvements in (or alternatives to) steam cracking technologies could substantially reduce the TAC. Pyrolysis is the second most expensive facility which contributes roughly 20% of total capital cost. As expected, thermochemical facilities in total account for more than 80% of total capital cost in the first three layouts.

The results of layout IV provide some interesting insights: transporting PO to Texas creates a significant cost of 491 million USD annually, representing 33% of the TAC. The annualized capital cost of layout IV is also the lowest among all the layouts we explored (245 million USD annually), with both pyrolysis and plastic pretreatment facilities attribute half of the total capital cost. Half of the TAC is associated to operating costs and the balance of 16% of TAC is attributed to capital, making this layout stand out. Notably, the TAC is almost as high as those of the other layouts, but the revenues are only half as high (illustrating how a shift to obtain value-added products creates incentives to build recycling infrastructure). Moreover, this potentially highlights the need to deploy regional plastic upcycling infrastructures, as transporting waste and intermediary products across the US can be cost-prohibitive.

Figure 4d combines the revenues and costs for the layouts and provides the resulting total economic surplus (total profit) obtained by the infrastructures. Here, we can see that layouts I, II, and III are all economic viable (obtaining positive economic surpluses between 1.03-1.39 billion USD per year). Layout II demonstrates that bypassing MRFs from the value chain represents the highest increase (35%) in surplus over the base layout I. Given the highest annual profit of layout II (1.39 billion USD per year), it has the lowest payback period as 2.6 years, followed by layout III (2.8 years) and layout I (3.4 years). We also observe that the annual profit of layout III is 10% higher than that of layout I, and layout III can take advantage of reductions in technology scale to decrease TAC, while still generating value-added products. We also observe that layout IV has a negative profit, suggesting that PO alone is not economically viable.

The large profits (over one billion USD per year) observed in our layouts suggest that incentive programs could be set up to reward consumers who pre-process plastic waste at their homes (at the source). By creating incentive programs, consumers are reimbursed for providing a service, encouraging their participation in plastic waste management, and some of the most expensive elements of collection and separation are avoided. In this layout, consumers might have an alternative to local recycling collection programs in which clean, sorted plastics are picked up separately, weighed, and reimbursed. The market values of LDPE and PP have a strong influence on the economic viability of the infrastructure; as such, we report a sensitivity analysis in the Supplementary Information. We have found that the infrastructures are robust (remain economically viable) even in the face of reductions of LDPE and PP prices of 30%).

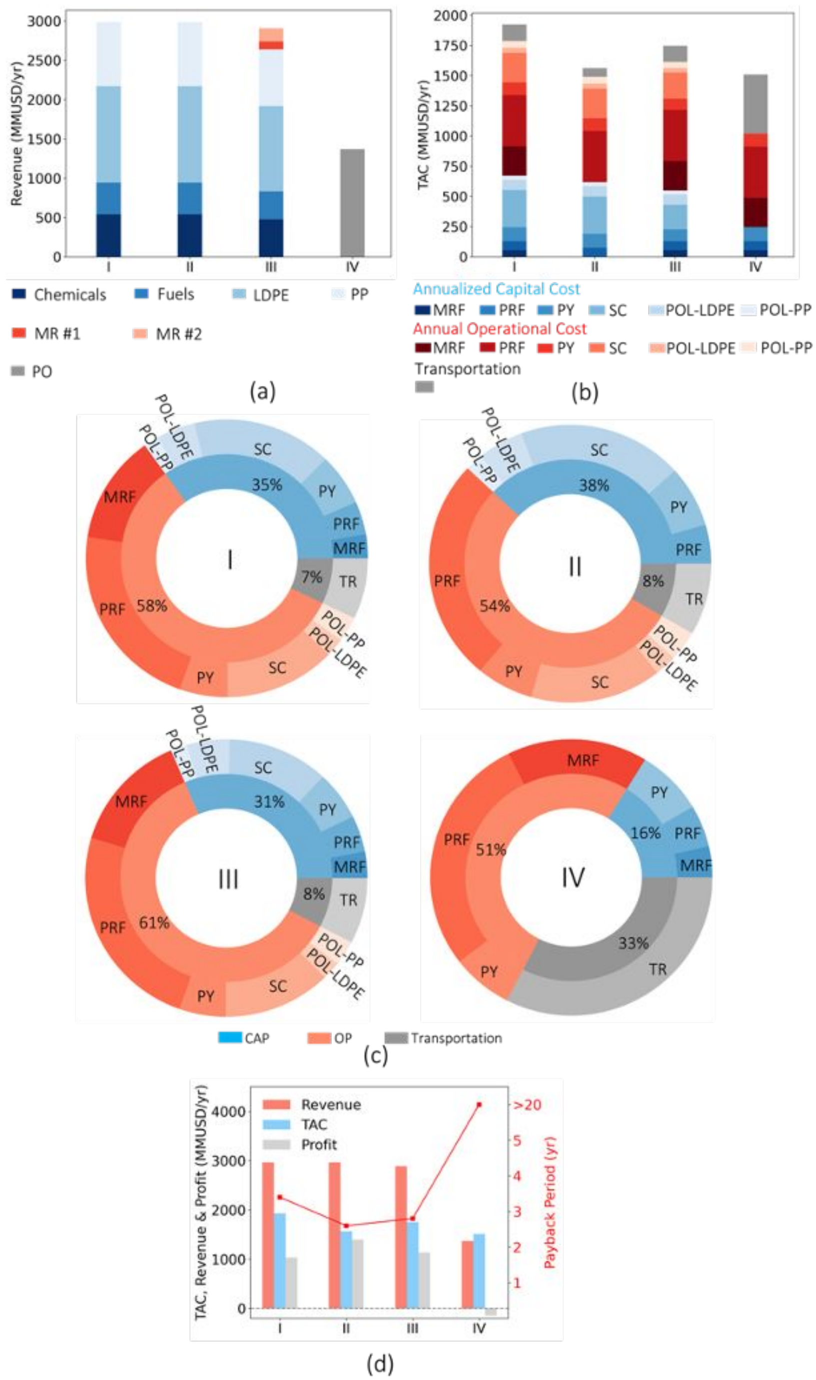

Figure 4. (a) Revenue breakdown of different infrastructure layouts. Layouts I and II generate the highest revenue; the major contributor to the revenue is the sale of polymer products. Infrastructure IV has the lowest revenue. **(b) TAC comparison.** Because MRFs are bypassed, the TAC of infrastructure II is 15% lower than that of infrastructure I. Infrastructure IV has the lowest TAC since its infrastructure is the simplest. **(c) TAC breakdown.** Inner cycles show the percentages of annualized capital cost (CAP), annual operational cost (OP) and transportation cost. Outer cycles show the detailed CAP and OP distributions of each equipment. Steam cracker has the highest investment cost and the second highest operational cost. PRF has the highest operational cost. **(d) TAC, revenue, annual profit, and the payback period (red line) of four infrastructures.** Layouts I, II, and III generate up to 1 billion USD of profit and

infrastructure II is the most profitable. The payback period of these three infrastructures are less than four years. For layout IV, the revenue cannot offset the TAC.

Environmental Benefits of Infrastructure Layouts. The environmental benefits of the proposed infrastructures are evaluated by quantifying the greenhouse gas emissions (GHGs) of the four plastic upcycling infrastructures using LCA methods (see Supplementary Information for details). GHG is an important metric that is used in analyzing the sustainability of recycling pathways, while other LCA metrics merit investigation, GHG is currently guiding industrial decision-making. Our infrastructure designs produce different end products, which makes comparing sustainability in terms of final products difficult. As such, we choose plastic waste as the basis for GHGs calculations, because it is the raw material used in all layouts.

Figure 5 presents a breakdown resulting from the processing of 1 kg of plastic waste through each layout, plus a disposal scenario representing GHGs associated with waste incineration. As a baseline comparison, all layouts outperform incineration as a means of disposal. By breaking down emissions, we find that the energy-intensive upcycling steps (pyrolysis, hydrotreating, steam cracking, and polymerization) contribute about 80% of the GHGs of the value chain, with pyrolysis contributing nearly a third of the total GHGs.

For layout III, we observe that the diversion of the 12% of PET and LDPE bottles in plastic bales into mechanical recycling lowers the total energy requirement (and GHGs) per unit of weight processed, and is our most sustainable layout, with an impact estimated at 1.38 kg CO_2 eq/kg plastic waste.

These results have a couple of important implications. First, thermochemical upcycling can reduce plastic waste accumulation by converting a substantial fraction of waste plastic into value-added new products. Second, thermochemical upcycling reduces the need for virgin plastics, demonstrating increased circularization of the plastic value chain and greater sustainability in terms of GHGs. Creating a circular economy for plastics has indirect effects such as reduction of US dependence on fossil resources.

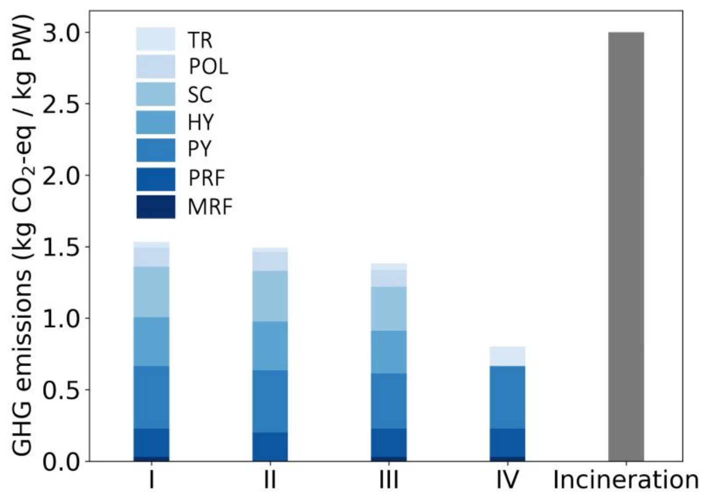

Figure 5. Breakdown of GHG emissions for upcycling infrastructures. The carbon emissions of all these infrastructures are at least 50% lower than those of incineration.

Inherent Value of Plastic Waste and Design Products. The optimization framework interprets the infrastructure as a market in which stakeholders exchange products and services to maximize the total economic surplus. This interpretation allows us to determine inherent value for different waste and product streams arising in the value-chain. The inherent values are complex functions of processing and transportation costs involved in generating a specific product, technology yields and capacities, externalities, and geographical locations. For instance, PW from a remote location is inherently less valuable than waste from a nearby location (and thus should be given less preference). Inherent values are also important to understand how value is generated and is propagated throughout the value chain.

In **Figure 6** we present inherent values for PW for the different infrastructure layouts. The optimization model is built using a county-level resolution, and we thus obtain inherent values at this level of geographic resolution. We see that the inherent values of layout I are high and in the range of 341- 498 USD/tonne. This value indicates that each tonne of PW generates roughly 500 USD of value in the value chain. Attracting plastic waste into a high-profit value chain that generates huge revenue (3 billion USD/yr) leads to high inherent values of plastic waste. The high inherent value of plastic waste which in turn implies that making virgin polymers from plastic waste via thermochemical technologies has a great economic potential. As expected, these inherent values are affected by their proximity to MRFs. This is because populated areas where MRFs are deployed tend to incur lower transportation costs and thus have higher value (these sources of waste are preferred). Transporting plastic waste to PRFs incurs a distance-based cost, reducing the value of plastic waste the farther it is from a processing center. Although there are 22 MRFs installed in the studied region, the Chicago areas (e.g., Cook County, Lake County, McHenry County, Kane County, Lake County, Winnebago County) have the highest inherent values of plastic wastes (480-498 USD/tonne). With the pyrolysis facilities located in Winnebago County, IL, short transport is required to collect plastic bale. With this geographical advantage consumers in the Chicago area enjoy higher inherent value.

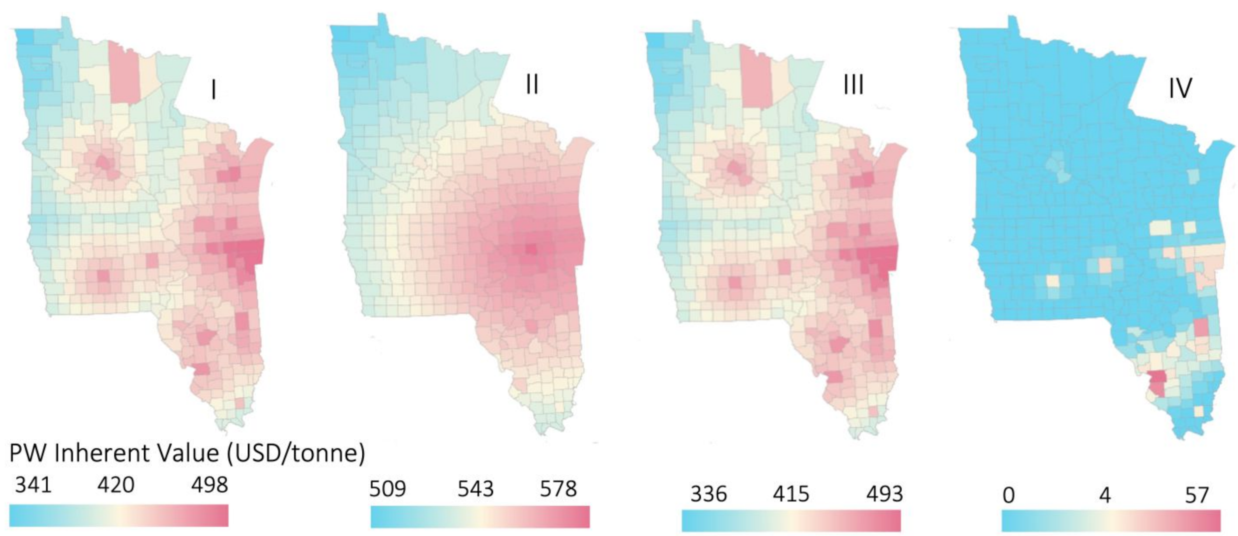

Figure 6. Inherent value of plastic waste in the study region. We see that the inherent value for PW for layouts I, II, and III is high and we see geographical variations driven by transportation costs to processing facilities. We also see that PW has a lower inherent value in layout IV, because the final product (pyrolysis oil) is less valuable than virgin resins produced in layouts I, II, and III.

Because layout II has centralized PRFs and thermochemical facilities, the distribution of inherent value is apparent. The closer to Winnebago county in Illinois, the higher the value. The value of Winnebago county reaches 578 USD/tonne and we note that the lowest value of layout II is 509 USD/tonne (Kittson County in northwest Minnesota). We thus see that, even the lowest value of layout II is still higher than the highest value of layout I. This indicates that bypassing MRFs reduces cost and ultimately increases the value of the PW. These inherent values suggest that greater incentives could be achieved by encouraging consumers to participate in a pre-sorting program. Specifically, this consumer sorting behavior increases the average value of plastic waste in the studied region by 123 USD/tonne (from 420 USD/tonne to 543 USD/tonne). The values of layout III are almost identical to those of layout I but slightly lower. These results from the fact that mechanically recycled products are not as valuable as virgin resins.

Because the economics of layout IV are less favorable, we can see that most places have an inherent value of zero, indicating that PW does not generate value in the value chain. We can see, however, that waste has a positive value in certain locations (close to PRFs and PY technologies). These results highlight how the layout of the infrastructure and the products generated have a dramatic effect on the inherent value of waste. Intuitively, attracting PW into a high-profit value chain generates more economic value than attaching waste to a low-profit value chain. For instance, attaching waste to a value chain that ends in a landfill generates no profit and thus indicates that PW has no inherent value (or even has a negative value that needs to cover costs of landfilling).

In **Figure 7,** we present the inherent values for PW and products obtained in the value chains. Because the inherent values change geographically, we present their averages. Here, we also compare the computed inherent values with current market prices. In summary, the most important observation concerning the inherent value of PW is that it is positive. This implies that PW is in fact a valuable feedstock. This contradicts the current perception of waste, which in general has a negative perceived value. For instance, consumers currently pay collectors to take away their PW. However, under an infrastructure that generates revenue from PW, consumers could potentially be incentivized to provide their PW (as this is a value resource). Our results also highlight how the inherent values of the products are in general higher than current market prices indicating that deploying plastic upcycling can open more valuable pathways. We also highlight that our framework allows us to estimate the inherent value of intermediate products such as pyrolysis oil and plastic bales. This is important because these products currently do not have well-established markets and obtaining price data is difficult. Finally, our results clearly illustrate how value is generated via different processing technologies throughout the value chain.

We highlight that the inherent value of plastic waste obtained are an implicit function of the prices of virgin resins, which are in turn a function of crude oil prices (and externality that is not captured in our model); as such, fluctuations in oil prices can affect the value of plastic waste. Our model can in principle be extended to capture market sectors that capture these dependencies and is an interesting direction of future work.

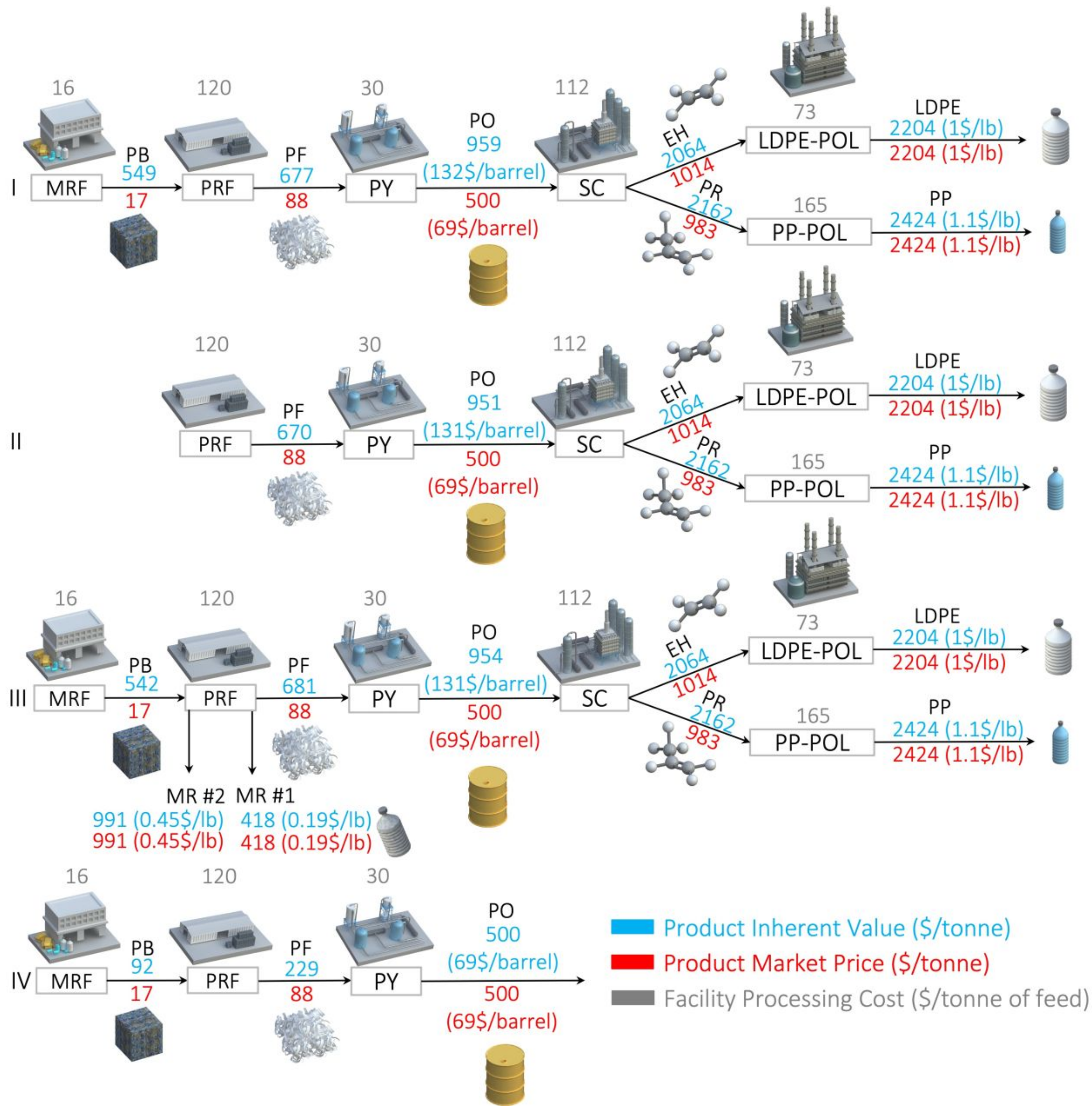

Figure 7. Average inherent values of plastic waste and derived products for the different layouts. We can see how the value of the final products (virgin resins or pyrolysis oil) propagates backwards in the value chain and makes waste valuable. The value of plastic waste is strongly affected by the final market that it is attached to; this explains why waste is more valuable in layouts I, II, and III (compared to IV).

Discussion

Driven by the ever-growing plastic waste pollution crisis, it is necessary to identify economically viable and sustainable upcycling infrastructures. Leveraging a group of well-established chemical technologies (pyrolysis and steam cracking), we investigated the viability of plastic upcycling infrastructures. To do so, we developed an infrastructure optimization framework that aims to design infrastructure layouts that maximize total economic surplus. Results show that thermochemical technologies can open new value chains that are economically viable and that generate substantial reductions in carbon emissions. We also used our framework to investigate different infrastructure layouts. Specifically, we find that pre-sorting of plastic waste by consumers and the development of hybrid infrastructures (that use mechanical recycling and chemical upcycling) can bring economic benefits. Our analysis also reveals that pre-sorting practices can be naturally incentivized in the proposed value chains because, under these settings, plastic waste becomes a valuable feedstock. The proposed framework and case study aim to highlight complex interdependencies that arise in the deployment of thermo-chemical technologies in waste management infrastructures and can be used to identify the effect of diverse factors that affect their economic viability. This information can in turn be used to guide experiments that provide more insight into the performance of pyrolysis units (the availability of such data is limited in the literature). As future work, we intend to apply the proposed model to a national level case study. Since optimizing the upcycling infrastructure at a national scale could be computationally challenging, we also aim at developing tailored algorithms to improve the solution quality and to reduce the computational time. In addition, we are currently assumed to have a common plastic composition throughout the study region (set to the national average); future work will incorporate seasonal and spatial variations of plastic composition. Geographical changes in product distribution and quality will also be considered as part of that study.

Methods

The proposed infrastructure modeling optimization framework is based on a general mixed-integer optimization formulation that aims to identify optimal selection, placement, and capacities for technologies in the geographical region that maximizes the total economic surplus. The economic surplus function simultaneously captures revenue generated from derived products and diverse costs associated with processing and transportation. This design problem can also be interpreted as a value chain (market) design problem, that aims to deploy technologies that maximize market value created. The proposed formulation is scalable, in that it can handle many candidate technologies, products, and geographical locations. The optimization formulations were all implemented in the Julia programming language and solved with a state-of-the-art solver. More details on the mathematical aspects of the formulation and of its implementation are provided in the Supplementary Information.

Our case study was built based on diverse sources of data available from the literature and from engineering judgement. The study region encompasses the upper Midwest region of the US, which in turn comprises four states (Wisconsin, Illinois, Minnesota, and Iowa) and 360 counties. Here, we treat each county in the region as a post-consumer plastic waste generator. These are consumers that generate plastic waste. In the standard value chain terminology, one would refer to such consumers as suppliers of waste. The amount of plastic (for each type) generated in each county is estimated based on population density and based on per-capita (average) plastic waste generation determined by the US EPA studies.

The infrastructures that we design must process all the plastic waste generated in the studied region. The infrastructures include combinations of six different technologies (MRF, PRF, pyrolysis, steam cracking, LDPE synthesis and PP synthesis). Each county is considered a potential location for MRFs, PRFs, pyrolysis units, steam crackers, LDPE and PP plants. Therefore, the plastic upcycling infrastructure consists of 360 potential sites for installing the MRFs, PRFs, pyrolysis units, steam crackers, and polymer synthesis. The material balances, energy demands, operating costs, and capital investment of each involved technology are obtained from literature or determined based on engineering experience (detailed data is included in the Supplementary Information).

The infrastructures also consider a wide range of primary, intermediary, and final products. The primary product (raw material) is a mixed stream of recyclables provided by each county. When consumers sort out plastics (layout II), the raw material is plastic mixed waste stream that includes all (#1-#7) types. Intermediate products are plastic bales, plastic flakes, pyrolysis oil, ethylene, and propylene. Final products are virgin polymer resins (LDPE and PP). We selected LDPE and PP as these are amongst some of the most produced plastics in industry. We also consider a set of final byproducts that include C_4, pyrolysis gasoline, fuel gas, fuel oil and wax. We place potential consumers for all products at all counties. The infrastructures include transportation services that move products across locations to satisfy requirements of technologies and consumers.

We consider that all suppliers, consumers, technology facilities and transportation providers in this infrastructure as independent stakeholders that transact products and services in a value chain to generate economic value[38,39]; this reveals helps estimate the inherent value of PW and derived products.

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Appendix

Table 1. Acronyms for Products and Technologies in Plastic Upcycling Infrastructure

Acronym	Description
AGO	Atmospheric Gasoil
C_4	Butane and Butylene
CH	Pyrolysis Char
EH	Ethylene
FC	Fuel and Chemical
FG	Fuel Gas
FO	Fuel Oil
GS	Pyrolysis Gasoline
HCL	Hydrogen Chloride
HY	Hydrotreating
HDPE	High Density Polyethylene
LDPE	Low Density Polyethylene
MR	Mechanically Recycled Plastic
MRF	Material Recycling Facility
NAP	Naphtha

PB	Plastic Bale
PF	Plastic Flake
PG	Pyrolysis Gas
PO	Pyrolysis Oil
POL	Polymerization
PP	Polypropylene
PR	Propylene
PS	Polystyrene
PRF	Plastic Reprocessing Facility
PVC	Polyvinyl Chloride
PW	Plastic Waste
PY	Pyrolysis
RE	Recyclables
SC	Steam Cracking
TR	Transportation

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