Cascading Polymer Macro-Debris Upcycling and Microparticle Removal As An Effective Life Cycle Plastic Pollution Mitigation Strategy

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Abstract

Plastic pollution caused by material losses and their subsequent chemical emissions is pervasive in the natural environment and varies with age. Cascading the life cycles of plastic losses with solid waste reclamation via re-manufacturing virgin polymers or producing fuels and energy, may extend resource availability while minimizing waste generation and environmental exposure. Here we systematically investigate this cascaded plastic waste processing over other waste end-of-life management pathways by analyzing the environmental consequences of plastic losses across the entire life cycle. Plastic losses can form volatile organic chemicals via photo-degradation and pose non-negligible global warming, ecotoxicity, and air pollution effects that worsen by at least 189% in the long run. These environmental burdens increase by above 9.96% under high ultraviolet radiation levels and participation rates, which facilitate plastic particulate compartment transport and degradation. Cascaded plastic waste processing aided by fast pyrolysis upcycling technologies can effectively cut environmental losses and outperform landfills and incineration in reducing 23.35% ozone formation and 19.91% air pollution by offsetting the external monomer manufacturing and fuels and energy production while saving at least 25.75% fossil fuels.

Keywords: plastic losses, cascaded plastic waste processing, microplastic, life cycle assessment, end-of-life management.

Synopsis

Cascading macroplastic upcycling and microplastic removal reduce material losses and air pollution compared to landfills and incineration and save fossil fuels.

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Introduction

Plastic, a crucial building block in Anthropocene activities, has soared exponentially in its production since it was invented in 1950[1]. High chemical resistance, good physical elasticity, and low production costs make this material versatile in industrial and residential sectors[2]. However, limited reuse before disposal has driven tonnages of polymer wastes to their end-of-life (EoL) treatment sites daily[3]. Effective waste-recovery technologies, such as incineration and recycling[4], can provide discarded plastic alternative uses, including fuels and/or energy production, to extend resource availability while minimizing waste generation and environmental exposure. Since waste-recovery processes are not dominant in EoL waste management, most polymer wastes still end in landfills or casual discarding[5]. Once exposed to the natural environment, these solid wastes lose their mass denoted by plastic losses[6] from hydraulic weathering or photo-degradation and form macro-particles and microplastics (MPs) less than five millimeters in size[7]. These material losses are transported globally and yield wide dispersal into the air, water, and land-based natural systems[8]. Unlike soluble chemicals or sediments, polymer particles cannot run directly into the mineral cycle aided by biogenic degradation, and their degree of intermingling with the natural environment remains an unmet research need[9].

Complex earth compartment transport and degradation phenomena can affect particle mobility and exposure to the natural system[10]. Lighter than soil substances, plastic particles are easily blown off from water or ground and suspended in the air as pollutants[11]. These airborne particulates can also undergo slow degradation to form volatile organic chemicals (VOCs) or fall off to land-based systems[12]. Aquatic plastics mostly sourced from the land-based runoffs[13] can be cracked into micro- or nano-particles under photodegradation[12]. Their minute nature allows easy toxic substance adsorption that poses harm to the aqueous organisms by ingestion[14]. Therefore, polymer degradation kinetics in earth compartments determine the extent of MP formation across the entire life cycle and chemical releases, which contribute to environmental pollution. These environmental burdens vary with age and require a holistic time-dynamic analysis to reflect the pervasive environmental pollution posed by post-disposal plastic wastes. However, the explicit evaluation of plastic pollution requires assessing the extent of plastic losses measured by their mass flow rates in earth compartments, which are still lacking in existing studies[15].

Cutting plastic waste generation and its environmental exposure can reduce their associated environmental burdens. Repurposing these solid wastes for other uses, such as concrete

production[16] or waste-to-energy[17], enables economical resource and energy use and limits direct waste release to the ecosystem. In practice, an ideal case to minimize this environmental exposure is to capture and convert the material losses into basic chemicals for plastic remanufacturing[18]. Environmental sustainability evaluation of this process requires cascading the macro- and microplastics' life cycles, including macro-plastic reclamation and microplastic removal processes[19]. Effective repurposing of discarded polyolefin plastics[20], such as polypropylene (PP), can be achieved via pyrolysis-based upcycling[21] that yields monomeric products under high temperatures[22]. Pyrolysis is the most commercially viable and technically promising plastic waste upcycling technology that has been implemented in 18 U.S. states[23], while the hydrothermal carbonization process is still operated on a lab scale and embodies a relatively lower technology readiness level[24]. With more than 20% (wt.%) in total yield[25], these monomeric products, including ethylene and propylene, will then be effectively separated and sold as value-added products to enhance economic profitability. Fast pyrolysis operated under anaerobic conditions hinders plastic oxidation and can cut greenhouse gas and toxic chemical emissions compared to incineration and gasification[26]. Adoption of hydrothermal liquefaction in polyolefin plastic waste conversion requires intensive water use and a higher labor and equipment procurement cost due to the high-pressure operating condition compared to fast-pyrolysis processes[27]. Relevant plastic waste fast pyrolysis studies identified the environmental advantages of onsite renewable fuel manufacturing from plastic cradle to the factory gate due to undetermined product EoL use[28]. A more complete "cradle-to-grave" life cycle was recently investigated for plastic mixture fast pyrolysis and downstream fuel and monomeric product use[29], but this study did not consider the potential environmental impacts of material losses from the entire life cycle. Existing MP ecotoxicity and human toxicity investigations have proved the potential environmental hazards of plastic particulates from diet ingestion[30], dermal contact[31], and air inhalation[32] without providing a basic understanding of the effects and extent of plastic losses, as well as effective removal technologies. All these knowledge gaps can be filled by precise estimates of the plastic losses and their subsequent chemical emissions occurring across the cascaded life cycle involving macroplastic upcycling and microplastic removal. Relevant holistic analysis of cascading macroplastic upcycling and microplastic removal remains a knowledge gap.

Previous studies only assessed microplastic's full-spectrum life cycle environmental impacts without evaluating how many microplastics are formed from the whole plastic life cycle[33]. This

work analyzes the extent of material losses released to the natural environment from the plastic entire life cycle and their derived environmental impacts to help estimate the environmental impacts from the cascaded plastic life cycle. We cascade the life cycles of plastic waste and their material losses treated by pyrolysis upcycling to assess its environmental performance over other EoL waste management pathways in plastic pollution alleviation. We consider cascading the multi-stage monomer recovery and fuel and energy production from plastic wastes and collected marine debris. This cascaded plastic waste processing after EoL waste disposal may benefit from onsite monomer, fuels, energy production, and debris mitigation, but it lacks quantitative understanding in the existing literature. We study the thermoplastic that is most widely produced (57% of total plastic manufactured) in the U.S.[34] and specifically focuses on PP in plastic bags because of its similar chemical property to other polyolefins widely used in packagings[35] as the major plastic pollution sources[36], and identical EoL waste treatment methods to other polymeric materials. The compartment transport and degradation kinetics are accounted for to determine the extent of plastic pollution[37], which denotes the full-spectrum environmental burdens of material losses and their derived chemical releases in a certain time frame. Given the long degradation time of plastics in the natural environment, a holistic understanding of their environmental consequences across the entire life cycle requires both short- and long-term quantitative analyses. Through systematic comparisons of these environmental consequences concerning different plastic EoL waste treatment pathways shown in Figure 1, we identify the pros and cons of the cascaded plastic waste processing over other solid waste management processes, shedding light on the technical innovations and relevant policy implications in mitigating pollution.

Key novelties of this study are:

- A novel mass balance-based plastic post-disposal fate model concerning earth compartment transport phenomena and degradation profiles was developed to investigate the life cycle material losses and their derived chemical emissions that help assess the extent of plastic pollution.
- A systematic evaluation of the time-dynamic environmental performances of cascading (macro)plastic waste upcycling and microplastic removal processes was performed to show their pros and cons over other EoL waste management pathways in plastic pollution mitigation.

We summarized the key findings as follows:

- Over 20 to 500 years, plastic losses posed by MPs and their derived chemical releases from photodegradation can increase 422% in climate change, 2,061% in ecotoxicity, 189% in air pollution, and 2,105% in ozone formation.
- Water hydraulics and meteorology can affect the long-term environmental consequences of plastic losses, as illustrated by an above 20% increment under a higher precipitation rate, wind speed, or ultraviolet (UV) radiation levels that enhance the surface degradation rate to facilitate polymer particulate generation and their derived chemical emissions.
- In the long run, cascaded plastic waste processing can be environmentally advantageous compared to other EoL waste management pathways composed of sanitary landfills and incineration in declining material losses and reducing 6.74% photochemical ozone formation, 4.57% global warming potential (GWP), and 25.75% fossil fuel consumption by offsetting external virgin chemical production.

This work deciphers the time-dependent environmental consequences of unavoidable plastic losses from existing EoL waste management pathways and demonstrates the benefits of employing cascaded plastic waste processing to achieve the circular economy. Relevant policy implications that help reach this environmental sustainability goal are:

- Zero waste policy: Cascaded plastic waste processing aided by fast pyrolysis technologies should be incentivized to reduce material losses by over 99.99% and recover these solid wastes onsite into monomeric products and fuels to offset their external manufacturing and avoid its environmental burdens by at least 4.35%.
- Environmental policy: Landfills should be replaced with effective plastic EoL waste management technologies that facilitate cascaded plastic waste processing, such as chemical reclamation, with governmental incentives to promote pollution mitigation and reduce over 23.35% life cycle ozone formation and 19.91% air pollution.
- Plastic pollution act: Minimizing plastic environmental exposure via galvanizing solid waste collection and meticulous release control or elimination can mitigate short-term environmental consequences and impede over a 422% increment in long-term effects.

We describe the plastic cascaded use processes and the life cycle assessment methodology in the **Materials and Methods** section by defining the research scope, introducing the key data

needed and their derivation, and outlining the impact assessment method applied. The following **Results and Discussion** section examines plastic waste's short and long-term environmental impacts, those from current plastic EoL management, and the pros and cons of cascading plastic use in three subsections. In the **Discussion** subsection, we inform the technological and policy insights associated with cascaded plastic use over other solid waste management processes. Conclusions are drawn in the last section.

Materials and Methods

Cascaded Plastic Use Overview

Repurposing PP plastic waste for a tandem of multiple reuses can extend resource availability while minimizing waste generation and environmental exposure[38]. Fast pyrolysis, which enables the efficient conversion of polyolefin plastics to value-added products, can effectively treat complex or contaminated plastic mixtures[19]. Utilizing the fast pyrolysis process to break down PP into renewable fuels and monomers for plastic re-manufacturing assists in cascading EoL of plastic wastes and debris. Other EoL options, such as incineration and landfills, are also considered; however, these technologies can result in the formation and transportation of MPs in the air, soil, and water due to natural weathering. All of the aqueous plastic debris collected is then converted into fuels and monomers through fast pyrolysis, enabling material recovery and cascading of plastic use. The drinking water treatment plant and incineration are included in the system to remove the retained MPs from water effectively[39].

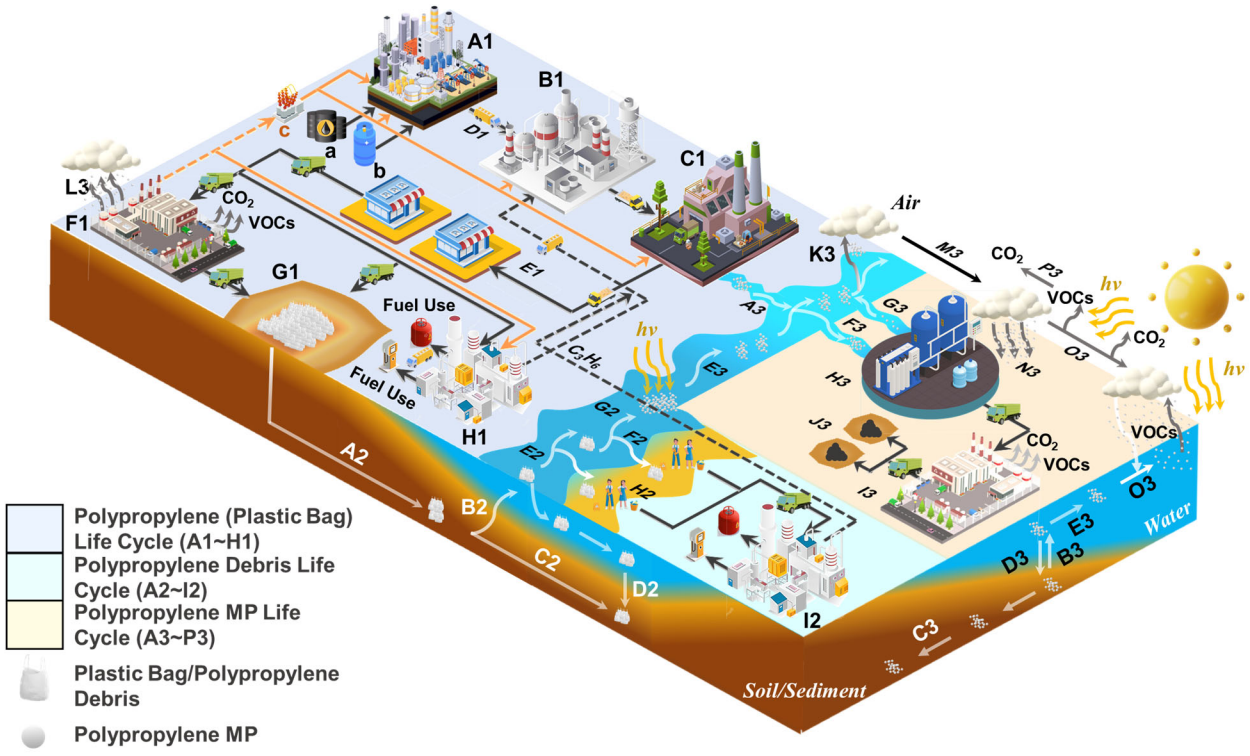

Figure 1. The "cradle-to-grave" system boundary of the PP plastic accounting for the material losses (macro- and MPs) formation from the entire life cycle and cascaded waste processing via waste-recovery to monomers and polymer re-manufacturing.

Life Cycle Assessment Methodology Overview

The LCA approach is employed to assess the environmental impacts of a product's entire life cycle, from raw material acquisition to EoL management[40]. This holistic approach provides an understanding of the associated air, water, soil, and ecosystem pollution, helping to inform decision-making that supports environmental sustainability[41]. We adopted this holistic LCA approach to identify the pros and cons of cascaded PP plastic waste processing by assessing the plastic losses and environmental impacts across the entire life cycle shown in Figure 1. Four typical phases are accounted for in the LCA: Goal and scope definition, inventory analysis, impact assessment, and interpretation. The assessment goal and its associated system boundary for investigation are defined in the stage of the Goal and Scope Definition. The Inventory Analysis stage assesses the life cycle inventories (LCIs), which describe the material and energy flow within the system boundary. Specifically, the upstream LCIs are built by the mass and energy flows of all PP pre-disposal life cycle processes, while the downstream data represent the missing mass

balances of the plastic losses and their derived chemical emissions. By compiling all collected LCIs with the characterization factors in the Impact Assessment phase, the full-spectrum environmental impacts of cascaded plastic use could be assessed and compared with other EoL waste management options. Finally, the Interpretation phase elucidates the assessment results to meet the LCA goal for decision-making, ultimately leading to effective plastic EoL waste management choices that reduce pollution and promote long-term environmental sustainability.

Goal and Scope Definition

We focused on evaluating the life cycle environmental impacts of PP plastics because they are widely used in packaging materials, of which solid waste releases are well-known as the major (micro)plastic pollution sources. The system boundary covers the upstream and downstream processes corresponding to the life cycle of PP wastes and their environmental losses, respectively. The upstream processes encompass raw material and resource extraction for propylene production, PP manufacturing, use, and EoL waste management pathways composed of incineration, landfill, and PP chemical recycling. The downstream processes involve plastic losses formation, degradation, aquatic waste collection and reclamation by fast pyrolysis-based chemical recycling, MP removals via drinking water treatment (DWTP), and incineration. The functional unit was chosen as one ton waste PP plastic bag treated to align with the mass and energy balance across the entire life cycle. Detailed explanation of the life cycle stages is given in the Supplementary Information.

Inventory Analysis

A. Upstream LCIs

Mass- and energy balance set the basis of LCI data collection. We extracted process-based LCIs from the Ecoinvent V3.8 database and related literature for all PP pre-disposal life cycle stages corresponding to all upstream processes. LCIs of onsite energy and chemical production, including the mid-voltage electricity and process heat co-generation from PP waste incineration and virgin chemicals produced from PP chemical recycling, were modelled as their market processes avoided by their internal production, referring to the avoided burden approach[42]. Chemical yields from PP waste chemical reclamation can be found in relevant plastic fast-pyrolysis LCA studies[43].

B. Problem Statement of Downstream LCI Analysis

Degradation and earth compartment transport govern plastic losses' natural exposure, mobility, and EoL chemical emissions that can interweave with earth compartments or biota and pose environmental effects with age. We developed a mass balance-based EoL fate model to generate the LCIs corresponding to plastic losses and their derived chemical emissions each year to air, water, and land, based on the random sampling methodology, which has shown its more than 95% accuracy represented by less than 5% relative errors[44] in multimedia transport and exposure dose model.

This mass balance-based EoL fate model can calculate the mass flow rate of plastic losses in particles and derived chemical emissions by accounting for various degradation and transport pathways in air, water and land compartments indexed by set *K*. Set *I* represents the chemicals yielded from plastic losses by photodegradation, including methane, ethane, peroxide, and acetone. All these mass flow rates are determined by the degradation mechanisms varying across compartments. Compiling these mass flow rates with their specific environmental impact estimates within the time horizon *t* can assess the overall environmental burdens posed by plastic losses. The general model formulation and equations are outlined as follows, with further details provided in the **Supplementary Information**.

- Perform random sampling to show a degradation and transport pathway given in Equations (1)–(3).
- Evaluate the amount of airborne and aquatic plastic particles (losses) generated from the sampled degradation and transport pathway by Equations (4)–(8).
- Evaluate the mass flow rates of the plastic losses and their derived chemical emissions by Equations (S21)–(S40).

C. Mass Balance-based EoL Fate Model Formulation

Specifically, we discretized each degradation and transport pathway ($\mathbf{T}_t$) by earth compartments and time with the timestep of one year. The plastic loss $\mathbf{T}_t$ comprises a set of binary parameters $TT_{k,n}$ generated from random sampling and shows the microplastic degradation and transport node. $TT_{k,n}$ equals to one, which indicates the microplastic transport to compartment *k* over time period *n* in time horizon *t*, as given in Equation (1). By compiling all these nodes ($TT_{k,n}$)

within the time horizon *t*, as shown in Equation (2)[45], the plastic loss $\mathbf{T}_t$ can represent a certain transport trajectory of microplastics,

$$\mathbf{T}_t = \left[TT_{k,n} \right], \qquad \forall n \in \{1,2,...,t\} \tag{1}$$

$$\sum_k TT_{k,n} = 1, \qquad \forall n \in \{1,2,...,t\} \tag{2}$$

where $\mathbf{T}_t$ represents the trajectory of plastic losses determined by time *n* and compartment *k*, and the binary parameter $TT_{k,n}$ equals one when they are transported to compartment *k* in time *n*.

Photo-oxidizing the airborne (k = 1) polymer particles can yield two sizes of particles[46] represented by binary variables TS_n and TSS_n as shown in Equation (3)[47]. The sizes of particles from losses can be reduced under photodegradation, and Equation (4) calculates the size of polymer particulates ($D(\mathbf{T}_t)$) yielded from degradation and earth compartment transport pathway based on the initial size of particulates (D_0, 1000 μm) given in Equation (5)[48]. We compiled the overall degradation rate calculations in a function *L* and its detailed mathematical formulation can be found in Equations (S6)–(S8).

$$TS_n + TSS_n = TT_{1,n}, \qquad \forall n \in \{1,2,...,t\} \tag{3}$$

$$D(\mathbf{T}_t) = (L_t \circ L_{t-1} \circ \cdots \circ L_1)(D_0) \tag{4}$$

$$D(\mathbf{T}_0) = D_0 \tag{5}$$

The number of plastic particulates ($N(\mathbf{T}_t)$) yielded from the degradation and transport pathway $\mathbf{T}_t$ are assessed in Equation (6) based on the initial number of particles in material losses (N_0), transfer rate $E_{k,n}$ within compartment *k* in time *n*, and transport rate $aa_{k,k'}$ from compartment *k* to *k'*[49]. Detailed mathematical formulation of the transfer rates can be found in Equations (S10)–(S13)[47].

$$N(\mathbf{T}_t) = N_0 \prod_{n=1}^{t-1} \left(\sum_{k'} \sum_{k} TT_{k,n} \cdot aa_{k,k'} \cdot TT_{k',n+1} \cdot E_{k',n+1} \right) \qquad \forall n \le t-1 \tag{6}$$

The number of MPs formed by marine plastic degradation ($N_2(\mathbf{T}_t)$) is evaluated in Equation (8) by the MP yield (α) from material losses and their number of particles ($N(\widetilde{\mathbf{T}}_t)$) in degradation and transport pathway $\widetilde{\mathbf{T}}_t$ [47], of which formulation is shown in Equations (7) and (S15)[49-50].

Detailed mathematical formulation of the transfer rates and the transport ratios of MPs (A_n , B_n , and C_n) calculated by Equations (S10)–(S13) and (S18)–(S20)[47], respectively.

$$\widetilde{\mathbf{T}}_t = \left[\mathbf{TT}_{1,t} \mid \mathbf{0} \mid \mathbf{TT}_{3,t} \right] \tag{7}$$

$$N_2\left(\mathbf{T}_t\right) = \sum_{n=1}^{t} \left[N\left(\widetilde{\mathbf{T}_{t-n}}\right) \cdot \alpha\left(D\left(\widetilde{\mathbf{T}_{t-n}}\right)\right) \cdot A_n \cdot B_n \cdot C_n \right], \qquad \forall n \le t-1 \tag{8}$$

Besides mass flow rates of plastic losses and chemical emissions evaluated in Equations (S39)–(S40)[51-53], process-based LCIs of current polymer material debris removal practices, including coastal solid waste collection and MP removals via drinking water treatment (DWTP), were also collected from Ecoinvent V3.8 Database and relevant literature. The collected coastal PP wastes and MPs were treated effectively in the fast pyrolysis-based chemical recycling plant and incineration sites [54]. The incineration ashes are then sent to sanitary landfills.

Impact Assessment

MPs and VOCs formed in 100 years do not pose environmental impacts to the same extent as releasing the same amounts in 500 years[55]. Therefore, environmental assessment of plastic losses should not be time-static. This work only considered the time dynamic GWP calculation because other environmental indicators' characterization factors are proven not to change with analysis time horizons[56]. Detailed dynamic GWP calculation can be found in the Supplementary Information.

The cascaded waste processing aided by fast pyrolysis processes can reduce the raw material use and its environmental impacts, and these environmental benefits were reflected as the "reduction" of offsite raw materials (natural gas and crude oil) and recovered propylene production[57]. We then evaluated the functional unit-based environmental impacts by the overall environmental effects based on GWP, EF3.0, ReCiPe 2016, and USEtox indicators, which were typically used in plastic processing LCA studies, divided by the total PP manufactured from the cascaded life cycle. Specifically, the proposed fate modelling methodology given the previous work was applied to evaluate the ecotoxicity characterization factors, where the effect factors were assumed to be proportional to the Hazardous Concentration above 20% species (HC20) data and calculated by the dose-response result of polyethylene[33]. Since the exposure factor of microplastics was postulated to be one, the ecotoxicity characterization factors were equal to the product of the

evaluated fate factor and effect factors. By collating these characterization factors with the investigated LCI data on PP manufacturing, use, EoL waste disposal, and material losses, we can then evaluate the full-spectrum environmental impacts across the complete life cycle. Detailed calculations can be found in (S53).

Impact Assessment Elucidation

The elucidation of the environmental assessment results should help reinforce the effective plastic EoL waste management choices for pollution reduction. We first compiled and displayed material losses' temporally dynamic environmental consequences to reflect their time-dependent effects over the material lifetime. Effect factors and their influences on these environmental burdens are unveiled in a comprehensive sensitivity analysis by varying the key parameters corresponding to photo-degradation and earth-compartment transport. We then evaluated and visualized the time-dependent environmental impacts of plastic losses and the results of other life cycle stages to identify the environmental pros and cons of the current EoL waste management pathway. This holistic approach is then applied to evaluate the overall environmental effects and breakdowns of various EoL waste management pathways to show their pros and cons.

Techno-economic Assessment

We further analyzed the total cost of treating waste PP by waste management pathway composed of landfill, incineration, and recycling receiving various percentages of domestic PP wastes by referencing the relevant literature[25]. The total cost (*TC*) was calculated in Equation (9) by the difference between the total annual expense (*TAE*) and income (*INC*) from downstream products. The total annual expense evaluated in Equation (10) equaled the summation of the operating expenses (*OPEX*) and the discounted value of total capital costs (*CAPEX*), which comprised the direct equipment installation (*DIC*), indirect (*IC*), and working capital (*WC*), equipment procurement (*PEC*), and land costs (*LC*) given in Equations (11) and (14)−(16). The direct and indirect capital costs of the processing facilities were scaled by their treatment capacities (*CAP*) and scaling factors (*SFF*) given in Equations (12)−(13). Equation (17) calculates the OPEX, which denotes the necessary cost for maintaining operations and stable processing, accounting for the operation and maintenance (O&M) costs (*OMCC*), transportation costs (*TRAN*), feedstock

(*FEC*) and utility costs (*UC*), and property and insurance (PT&I, represented by *PTIC*). The income from chemical recycling was generated by the onsite production of monomers and fuels, while the income from incineration came from heat and electricity generated onsite. Parameters *DICC* and *PECC* denote the base-case direct equipment installation and procurement costs, respectively. *CAPC*, *CEPCI*, and *CEPCIB* represent the base-case processing capacity and chemical engineering indices of the current year and base-case year, respectively. The coefficients *ICC*, *WCC*, and *LCC* indicate those for calculating indirect capital, working capital, and land costs based on the direct equipment installation and procurement costs.

$$TC = TAE - INC \tag{9}$$

$$TAE = CAPEX + OPEX \tag{10}$$

$$CAPEX = DIC + IC + WC + LC \tag{11}$$

$$DIC = DICC \cdot \left(\frac{CAP}{CAPC}\right)^{SFF} \cdot \left(\frac{CEPCI}{CEPCIB}\right) \tag{12}$$

$$PEC = PECC \cdot \left(\frac{CAP}{CAPC}\right)^{SFF} \cdot \left(\frac{CEPCI}{CEPCIB}\right) \tag{13}$$

$$IC = ICC \cdot PEC \tag{14}$$

$$WC = WCC \cdot DIC \tag{15}$$

$$LC = LCC \cdot PEC \tag{16}$$

$$OPEX = OMCC + TRAN + FEC + UC + PTIC \tag{17}$$

Results and Discussion

Short- and Long-term Plastic Waste Environmental Consequences

Previously thought to be chemically inert, plastic waste runoffs from landfills have now proved to be the main source of ecotoxic material losses, which can slowly yield VOCs by photo-oxidation over centuries[58]. The summation of the environmental consequences of these polymer and chemical releases over a certain time scale denotes the extent of plastic pollution during this period. We evaluated these environmental consequences over 20 (short-term), 100, and 500 years (long-term), of which time spans are typically used in assessing the climate change impacts as the environmental hotspot of polymeric material processing across the entire life cycle. The mass flow rates of plastic losses and derived chemical releases are evaluated based on fate modelling and

reaction kinetics given in **Materials and Methods**.

Overall, global warming, ecotoxicity, air pollution corresponding to particulate matter (PM) formation, and ozone formation corresponding to photochemical ozone formation and its specific effects on human health and terrestrial ecosystems are four major environmental consequences of the chemical and MP releases. In the short run, plastic debris from runoffs is mostly retained on the land and transmitted to water and air with slow transport rates of 0.05 and 0.1 for air and water, respectively. These aquatic and airborne particulates then undergo photo-oxidization to a small extent. Few airborne polymer particulates are downgraded into VOCs, and aquatic polymer debris is mostly converted into large-size MPs (>100 μm) with less unit ecotoxicity effect of 1.323×10^4 CTU_e per ton PP bag waste compared to small-size polymer particles with 9.2×10^5 CTU_e per ton PP bag waste, resulting in relatively unpronounced climate change, ecotoxicity, and ozone formation consequences as observed in Figure 2a. At length, these particulates break up and reduce their sizes in air and water as the earth compartment transport and photo-degradation proceed. Airborne MPs smaller than 10 and 2.5 μm in diameter can pollute the air analogous to PM 10 and PM 2.5, respectively, and worsen the PM formation and major air pollution effects by 15 times from short- to long terms. VOC emissions from photo-oxidizing these particulates increase as more MPs form with age, shifting the ozone formation impacts that harm human health and the terrestrial ecosystem by 2,105%. On the other hand, respiratory effects posed by PM 2.5 do not monotonically improve with time, as shown in Figure S1. This environmental impact increases as the airborne MP formation predominates over degradation in the first 100 years, and these particles will then undergo degradation over the next 50 years and reduce the PM 2.5 formulation. This environmental burden will continue to grow as the MP particulates accumulate in the air in the long run. Besides air pollution effects, the long-term impact of the ecotoxicity is also aggravating at least four times from incremental aquatic MPs formation and organic chemical emissions, including ketones, peresters, and peroxides formed by photo-degradation. With greater land-based polymer debris input and transmission to air and water, environmental consequences of plastic losses, in the long run, can be foreseen as a monotonic increment observed in Figure 2a.

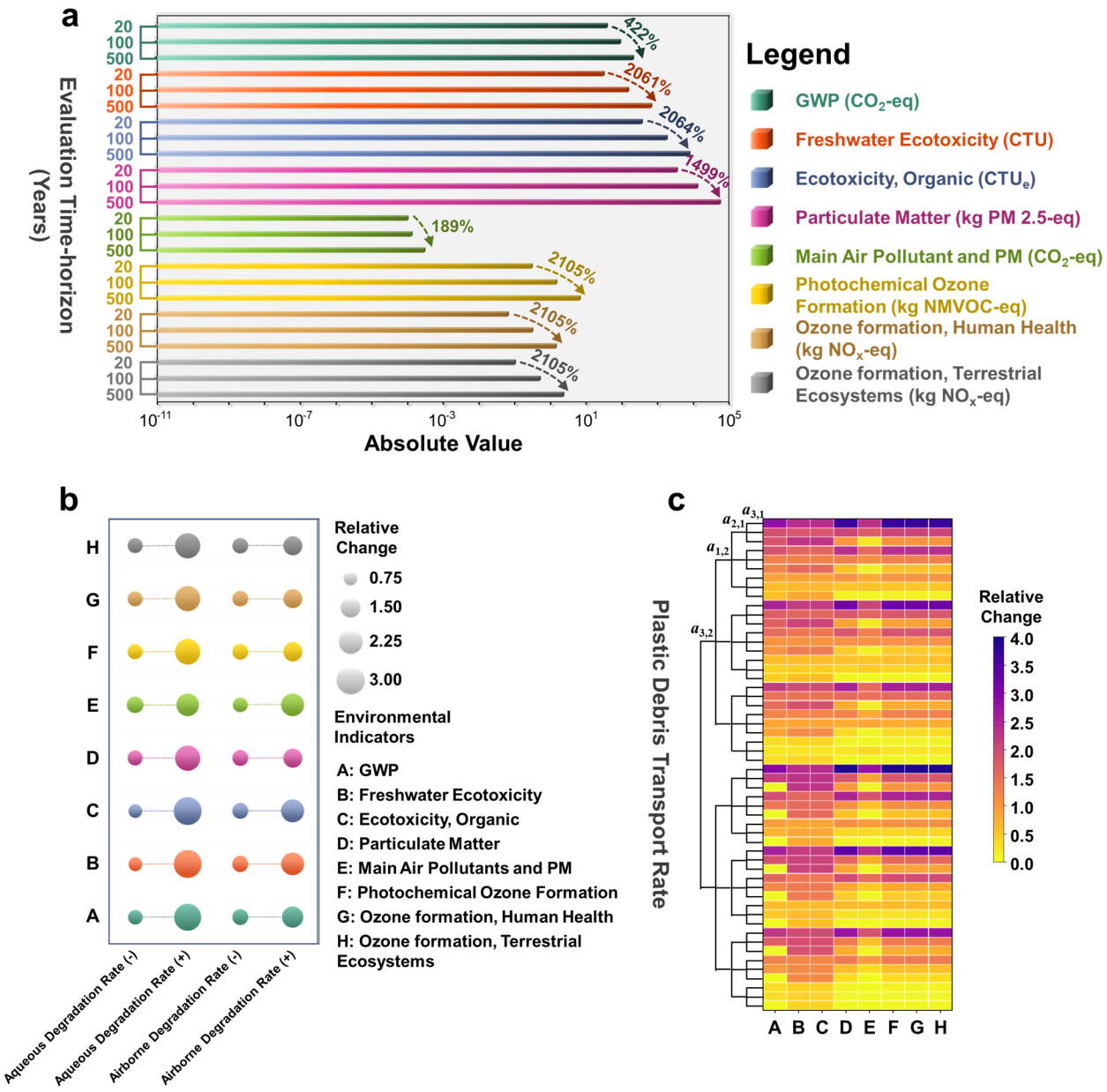

Figure 2. Short- to long-term plastic losses-related environmental impacts and their impact factors. (a) Absolute environmental impacts over 20, 100, and 500 years. (b) Effects of degradation rates on plastic losses' environmental consequence over 500 years. (c) Effects of multimedia transport rates on plastic losses' environmental consequence over 500 years. The environmental indicators used include GWP (CO_2-eq: CO_2 equivalent); Freshwater Ecotoxicity (PAF: Potentially Affected Fraction); Freshwater Ecotoxicity, Organics (CTU_e: The comparative toxic unit for aquatic ecotoxicity impacts); Major Air Pollutants and PM (UBP: Eco-points); PM (PM2.5-eq: Equivalent air pollution effect caused by PM with sizes smaller than 2.5μm); Photochemical Ozone Creation (NMVOC-eq: Non-methane volatile organic chemical equivalent); Ozone Formation,

Human Health (NO_x-eq: Nitrogen-oxides equivalent); Ozone Formation, Terrestrial Ecosystems (NO_x-eq: Nitrogen-oxides equivalent). The values near the ticks in Figure 2a represent the relative increment of environmental impacts over 500 years compared to those over 20 years. The bubble diameters given in Figure 2c are proportional to the relative change in environmental impacts under parameter variations. Parameters $a_{i,j}$ represent the plastic transport rate from compartments i to j: 1, 2, and 3 denotes air, water, and land-based compartments, respectively.

Key photodegradation parameters, including temperature and UV radiation dose[50], can determine plastic losses and their subsequent chemical releases. The plastic debris airborne degradation rate constant is calculated by the product of the ultraviolet irradiation level (*lv*), quantum yield (*yi*), and efficiency (*ef*), as given in Equation (18)[50], while the water temperature (T) can change both the kinetic rate constant of plastic degradation in water (Equation (19)) and the retention of microplastics, as illustrated by the relevant microplastic multimedia transport studies[59]. Parameters k_{d0}, E_a, T_0 represent the referenced kinetic rate constant, activation energy, and referenced temperature, respectively. Results show that airborne polymer particulates can degrade faster under a two-fold higher UV radiation level in summer, resulting in at least 41.48% increments in full-spectrum environmental impacts, as shown in Figure 2b. These environmental impacts are deterred by over 10.90% when the water temperature drops to near 0 °C in winter and slows down the aquatic polymer photodegradation. Season change can affect plastic degradation rates and the subsequent long-term environmental burdens as the temperature varies."

$$kk = yi \cdot ef \cdot lv \tag{18}$$

$$k_d = k_{d0} \cdot e^{E_a\left(\frac{1}{T_0}-\frac{1}{T}\right)} \tag{19}$$

Besides temperature variations, earth compartment transport phenomena can also govern MP formation kinetics and photo-degradation product profiles. Multimedia transport of polymer particles in the air, water, and land driven by water hydraulics and meteorology conditions can redistribute MPs and chemicals to different compartments with specific degradation characteristics and thus influence long-term environmental impacts shown in Figure 2c[60]. High wind speeds that increase transport rates from water and soil to air by 2.90% and 150% can facilitate more plastic particulates blown off land and water. Airborne suspension of these solid particles acts as PM and can form VOCs from photo-degradation, which improves human health and terrestrial ecosystem

hazards by at least 60.03%. High precipitation rates, which can promote airborne particle deposition to land and water by a 36.84% enhancement in transportation rates, can enhance aquatic exposure to ecosystems and ecotoxicities by at least 9.96%. Earth compartment transport between land and water driven by water flow rates can be influenced differently by environmental impact indicators. As more plastic particles are transmitted from soil and held within water compartments, which is represented by the $a_{3,2}$ increment to 0.1, the global warming effect decreases by at least 20.3% because fewer particulates will be instantly transported to the air and generate VOCs. However, the more polymer debris stored in water or land facilitated by compartment transports, the higher amount of airborne plastic particles will yield from wind-blown-offs, which can worsen the air pollution corresponding to PM and ozone formation by at least 10.5% and 7.88%, respectively.

Our results suggested that the environmental consequences of plastic losses are detectable and can worsen in the long run. Due to the data gap, this conclusion was drawn based on the assumption that the meteorological and hydrological conditions remain consistent across different years by referencing the assumption of existing microplastic fate investigation[61]. The temporal variations of these external conditions could tune the multimedia transport of plastic losses and their derived chemical emissions, thus affecting the environmental assessment results. To better account for these impacts, a more comprehensive framework for evaluating plastic material losses should be developed in future studies. This framework should combine real-time meteorological and hydrological monitoring with long-term extrapolated data and analyze these elements using dynamic environmental assessment methodologies.[62]

Current Plastic End-of-Life Option's Environmental Cost

Currently, the U.S. is one of the world's top plastic polluters, and 78.31% of the domestic PP waste ends in landfills, and only 2.73% is effectively recycled[63]. As landfilled plastics can runoff into the natural environment and generate material losses with the time-dependent environmental hazards illustrated by Figure 1, the EoL fates and removal of these polymeric materials should also be accounted for within the complete life cycle. Therefore, we evaluated and outlined these environmental impacts across the entire plastic life cycle to stress the environmental consequences of the current EoL waste management pathway.

In the short run, the plastic losses mainly from the landfills can pose 0.05%, 0.49%, and 3.00% of life cycle ecotoxicity, PM formation, and ozone formation effects from MPs and subsequent

chemical emissions, respectively, as shown in Figure 3. These environmental impacts posed by low flow rates of pollutants shown in the right-end ticks in Figure 4b are relatively unpronounced in the short run but can be more prominent with age, as illustrated by Figure 4a. In the long run, the incremental yields of airborne MPs and VOCs from photo-degradation, including methane and ethylene in the air, of which flow rates are shown in Figure 4b, can exacerbate 94.25% of total ecotoxicity posed by organic chemical emissions, 41.65% photochemical ozone formation, and 0.0174% PM air pollution. Therefore, these trivial plastic losses from the current EoL waste management pathway, which were not explicitly reported in existing LCA studies, can pose conspicuous environmental burdens across the entire life cycle.

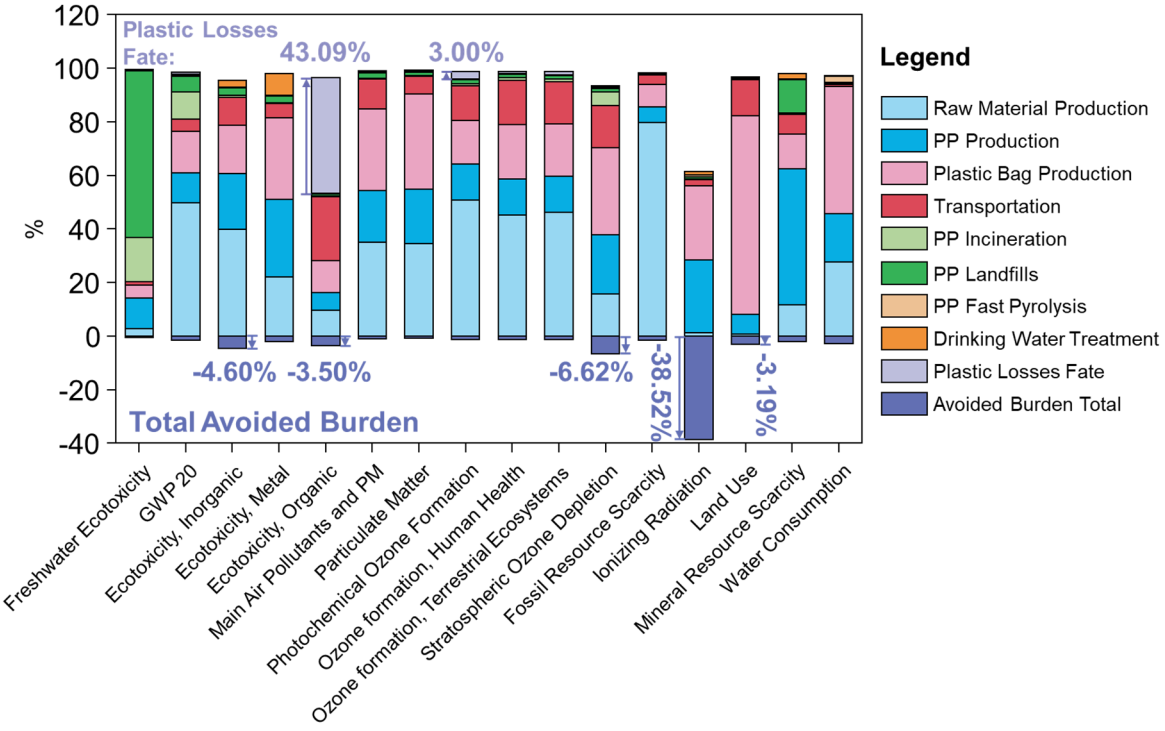

Figure 3. Full-spectrum environmental results and breakdowns of plastic waste treated by the current U.S. EoL waste management scenario that accounts for treating 2.73% of wastes (wt.%) in recycling, 19.96% in incineration, and 78.31% in sanitary landfills.

Effective plastic waste reclamation technologies can replace landfills and gain environmental benefits from secondary use and solid waste runoff reduction. We considered fast pyrolysis technology in reclamation because it effectively breaks PP carbon backbones under high temperatures and reverses the waste plastics back into high-yield monomers used for secondary production[64]. Figure 3 shows the environmental advantages (shown as Avoided Burden Total) of

deploying the fast pyrolysis in treating 2.73% of domestic PP wastes by reducing 1.00% main air pollutant and PM, 1.89% photochemical ozone formation, and 1.50% climate change. The environmental benefits of plastic reclamation can be further enhanced by improving its use in EoL waste management. This technology is not environmentally benign because of the intensive fossil resource use for process heat, water use, and direct greenhouse gas (GHG) emissions in the flue gas from the incorporated incineration process[65]. Therefore, a systematic comparison between different EoL waste management pathways is required to determine the plastic EoL waste treatment method with minimum environmental pollution.

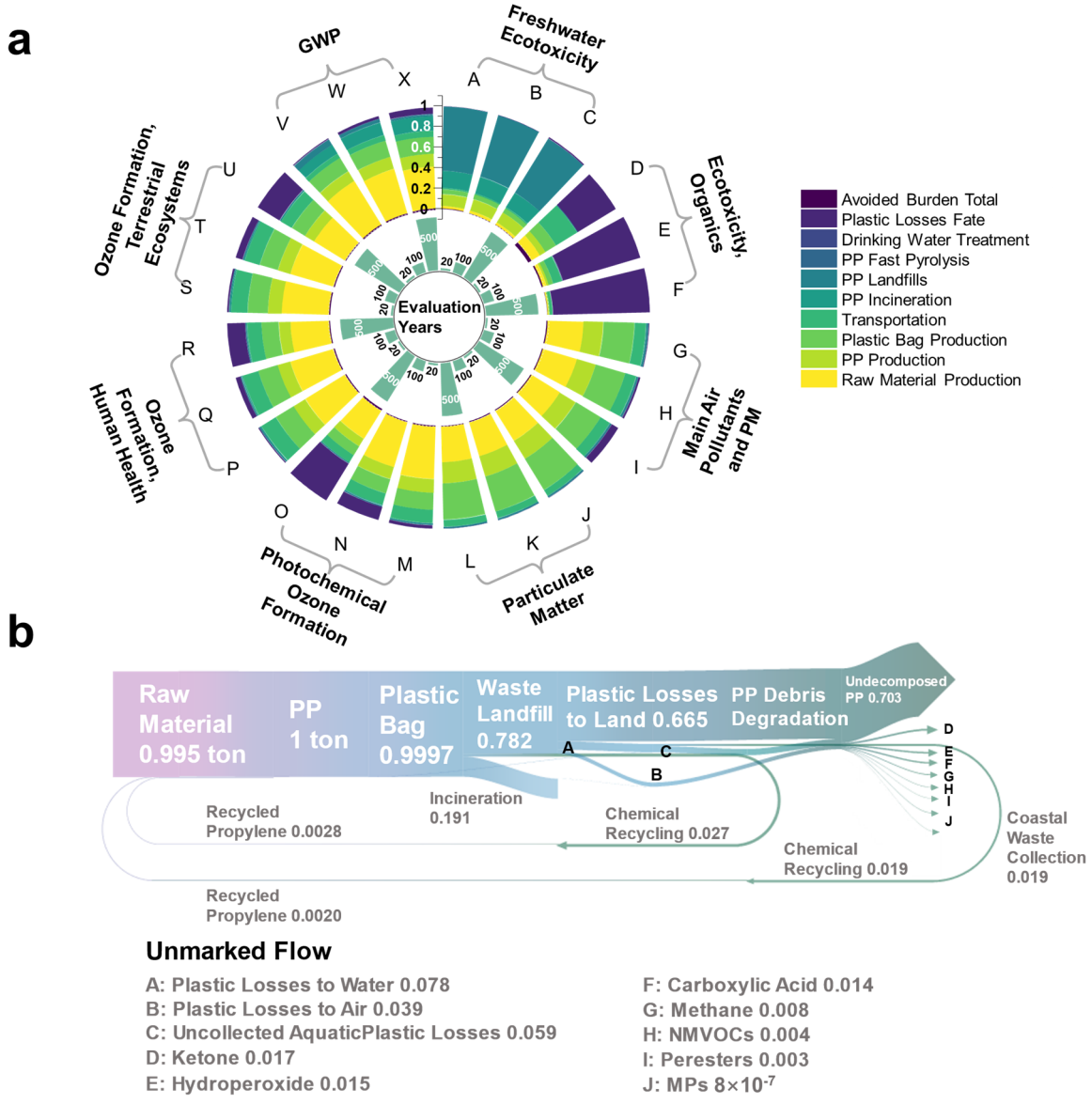

Figure 4. Environmental and flow profiles of PP waste treated by the current U.S. EoL waste management pathway (78.31% landfilled, 19.96% incinerated, and 2.73% recycled[63]) evaluated in

different time horizons. (a) Environmental profile based on various environmental indicators in 20, 100, and 500 years. (b) Mass flow distribution across the entire life cycle in 500 years shows the mass flow rates in tons from raw material and resource extraction to the photo-degradation products as graves.

Reducing Environmental Burdens through Cascaded Plastic Waste Processing

We evaluated the overall environmental effects of the plastic EoL waste management pathway composed of major disposal options, including landfill, incineration, and recycling, which receive a specific percentage of domestic PP wastes. The summation of the percentages of all these three EoL waste technology options equaled one. The maximum percentage of total plastic waste received for landfilling (wt.%) was set as 78.3%. We compared the life cycle environmental impacts of three typical plastic EoL waste treatment scenarios named with the **U.S. 2018**, **50/50/0**, and **100/0/0**. The **U.S. 2018** scenario refers to the latest U.S. domestic plastic EoL waste management pathway proposed by the U.S. Environmental Protection Agency[63], which denotes that 2.73%, 19.96%, and 78.31% of total PP waste are received at recycling, incineration, and landfill sites, respectively. The "**50/50/0**" represents the future EoL waste management pathway in 2030, an extrapolation based on the current European Union (EU)[66] case, that 50% of total solid wastes are recycled while another 50% end in incineration. The "**100/0/0**" denotes an ideal case that plastic waste is fully recycled.

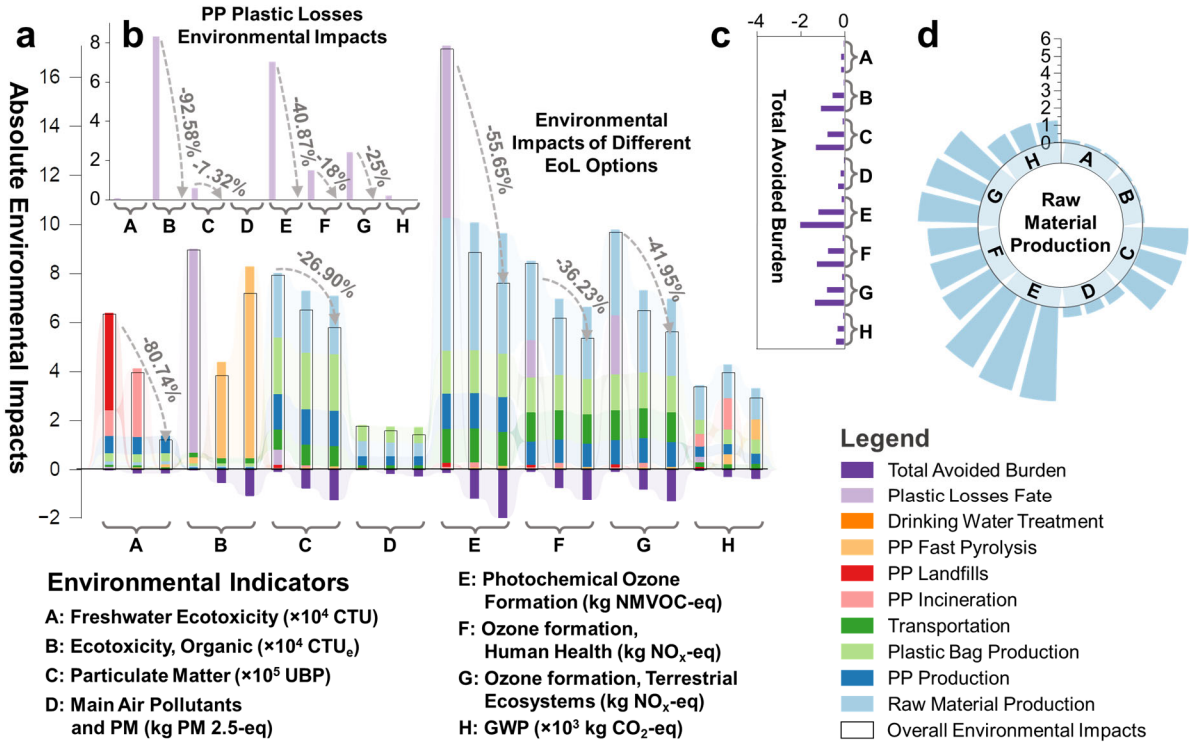

Figure 5. Absolute life cycle environmental impacts (over 500 years) and impact assessment results for specified life cycle stages of plastic waste treated by three different EoL scenarios. (a) The entire life cycle stages. (b) The fate of PP plastic material losses. (c) Total avoided burden from onsite monomer and fuels and energy production. (d) Raw material and resource extraction. The *y*-axis values for all subfigures represent the absolute environmental impact results. The three bar charts in subfigures for each impact category indicate the current U.S. EoL waste management pathway, 50% incineration with 50% recycling, and 100% recycling cases from left to right (Figure 5a and Figure 5b), top to bottom (Figure 5c), and clockwise (Figure 5d).

Overall, Figure 5a and Figure 6 show that the plastic losses and their environmental impacts can be significantly reduced if landfilling is replaced by other EoL waste management technology options, including incineration or recycling. When 50% of solid wastes are treated by incineration and another half by reclamation, plastic losses are decreased by a minimum of 99.99% and become environmentally benign. However, PP waste incineration can generate CO_2 intensively and contributes 32.15% to the life cycle GHG emissions. Compared to onsite incineration, PP fast pyrolysis-based reclamation processes can allocate the carbons to fuels, such as gasoline and diesel, and monomer products, including ethylene and propylene, and reduce direct GHG emissions.

Figure 6a-6h show that the freshwater ecotoxicity and air pollution impacts associated with PM formation and ozone formation can reach their minimum when treating the PP waste completely by chemical recycling. Plastic losses and their environmental impacts are also heavily reduced, as illustrated in Figure 5b, which shows 92.58%, 40.87%, and 25% decrements in total ecotoxicity posed by life cycle organic chemical emissions, photochemical ozone formation, and ozone formation that harms terrestrial ecosystems, respectively. Although high organic solvent use in the aromatic extraction process within PP chemically recovery processes can increase ecotoxicity from life cycle organic emissions[67], this EoL waste management process still outperforms other EoL options, including incineration and landfills, in terms of the full-spectrum environmental impacts.

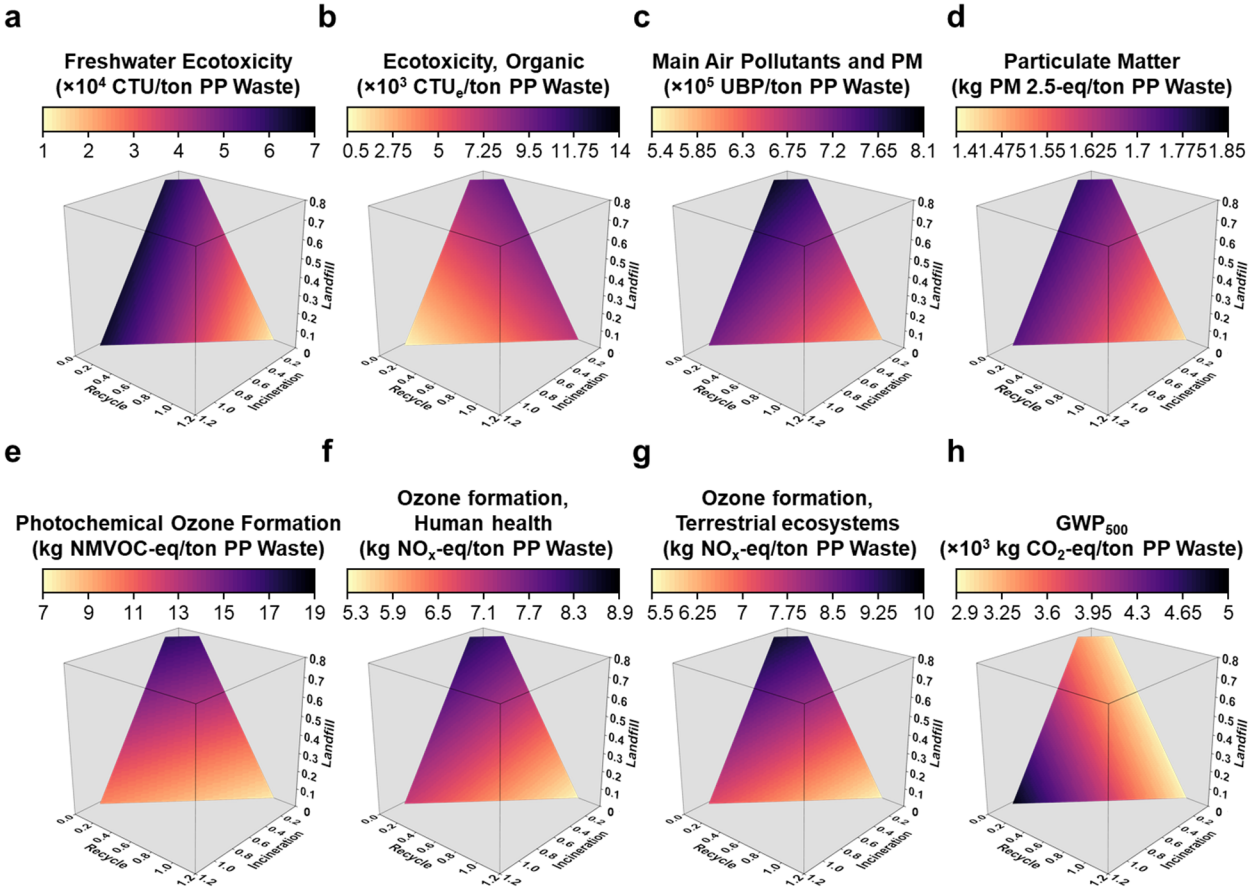

Figure 6. Absolute life cycle environmental impacts (in 500 years) of PP wastes treated by various PP waste EoL options. (a) GWP (CO_2-eq: CO_2 equivalent); (b) Freshwater Ecotoxicity (PAF: Potentially Affected Fraction); (c) Freshwater Ecotoxicity, Organics (CTU_e: The comparative toxic unit for aquatic ecotoxicity impacts); (d) Major Air Pollutants and PM (UBP: Eco-points). (e) Particulate Matter (PM2.5-eq: Equivalent air pollution effect caused by particulate matter with sizes smaller than 2.5μm); (f) Photochemical Ozone Creation (NMVOC-eq: Non-

methane volatile organic chemical equivalent); (g) Ozone Formation, Human Health (NO_x-eq: Nitrogen-oxides equivalent); (h) Ozone Formation, Terrestrial Ecosystems (NO_x-eq: Nitrogen-oxides equivalent).

Onsite monomer and fuels and energy production in PP recovery processes can decrease environmental burdens from offsite manufacturing. Figure 5c illustrates this environmental benefit by reducing 19.91% PM formation, 26.49% photochemical ozone formation, and 23.35% ozone formation in the **100/0/0** scenario. The current global plastic waste management practice in 2023 is to process plastic wastes by 30%, 24%, and 46% in incineration, chemical recycling, and landfills, respectively. Figure 6h shows that this process can reduce more than 20% of full-spectrum environmental impacts when processing PP wastes compared to the existing U.S. practices. The climate impacts, which are the environmental hotspot in maintaining plastic life cycle sustainability, can reach their minimum when no PP wastes are received and treated by incineration. The propylene produced from PP waste recovery can be re-polymerized into plastics, enabling cascaded waste processing to reduce virgin chemical consumption. Figure 5d shows the full-spectrum environmental advantages of this fast pyrolysis-based cascaded PP waste management on declining 6.74% photochemical ozone formation, 5.70% ozone formation that harms human health, 5.86% ozone formation harming the terrestrial ecosystem, and 4.57% GWP. Applying the cascaded plastic waste processing can mitigate these life cycle environmental impacts via declining environmental losses and reducing 11.25% raw material and 25.75% fossil use via onsite monomer and fuels and energy production, as observed in Figure 5d and Figure S3. However, implementing chemical recycling does not necessarily indicate a full-spectrum environmental impact reduction. Figure 6b indicates that the high energy and solvent consumption over this process can enhance the ecotoxicity impacts associated with organic chemical emissions. Further improving the monomer recovery rate[68], limiting energy use, and specialized air pollution controls on the exhaust gas can better hinder environmental consequences from plastic losses and reduce fossil resource consumption.

We also assessed the total cost of treating waste PP by waste management pathway composed of landfills, incineration, and recycling receiving various percentages of domestic PP wastes. The **50/50/0** scenario presented in Figure 7 shows the highest total costs of the three investigated scenarios owing to the higher capital costs of the incineration plant than the sanitary landfills. Due

to the economy of scale, the total equipment installation costs of two processing plants (chemical recycling and incineration) in the **50/50/0** scenario can be higher than the **100/0/0** scenario with the same total treatment capacity by cascaded plastic use. Despite having lower costs due to simpler operations and processes, sanitary landfills cannot offset the high environmental burdens caused by plastic losses. Therefore, government incentives should be provided for cascaded plastic use to effectively reduce plastic losses and their derived environmental consequences.

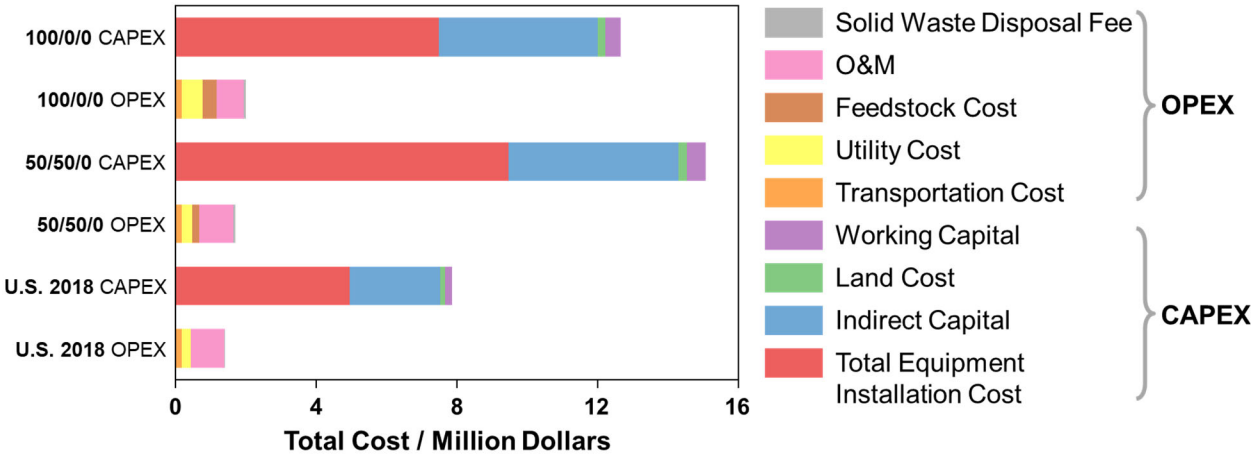

Figure 7. Breakdowns of the total costs of the **100/0/0**, **50/50/0**, and **U.S. 2018** scenarios. The total cost comprises CAPEX and OPEX. CAPEX equals the summation of the total equipment installation costs, indirect capital, land cost, and working capital, while the OPEX is comprised of solid waste disposal fee, O&M, and costs of feedstocks, utilities, and transportation.

Discussion

Growing plastic production has benefited our Anthropocene activities, but the prices of widespread use should be reconciled with its environmental burdens across the entire life cycle[69]. These environmental consequences from material losses can be evaluated via understanding their environmental fate, including degradation and transport phenomena and exposure to different earth compartments (i.e., air, water, and land). Plastic losses can undergo natural photo-degradation and pose ecotoxicity, climate change, and air pollution that can worsen by at least four times as the decomposition continues with age. Meteorology and river hydraulics are two key effect factors of these environmental consequences. A two-fold higher UV radiation levels on sunny days could increase 41.48% of these environmental consequences, and intensive wind can also drive these environmental effects by at least 20%. Water temperature dropping to near 0 ℃ in winter slows

595 the aquatic degradation and reduces by above 10.90% of full-spectrum environmental impacts.
596 Environmental burdens from plastic losses are not negligible and can increase with age.
597 Therefore, previous studies underestimated the environmental burdens of plastics due to the
598 lacking investigation of their environmental losses across the whole life cycle[70], though they were
599 detected in tonnages from virgin material production, use, and EoL waste disposal[71]. For instance,
600 the current domestic PP EoL waste management pathway of the U.S., which is one of the world's
601 top plastic polluters, causes growing environmental pollution with age and can pose a minimum
602 of 41.65% full-spectrum environmental impacts on ecotoxicity (organics) and ozone formation in
603 the long run. Implementing solid waste reclamation technologies can effectively limit waste runoff
604 and alleviate environmental losses to reduce these long-term environmental legacies. When
605 substituting plastic landfills with 50% chemical recycling and 50% incineration, the plastic release
606 to the environment can be reduced by over 99.99%. These environmental benefits can be more
607 pronounced if applying cascaded waste processing to reclaim solid wastes back to virgin materials
608 multiple times and cut raw chemical consumption, polymeric material release, and GHG emissions
609 compared to the widely used incineration process. Environmental benefits of the cascaded PP
610 waste processing can be outlined as reductions in the 19.91% life cycle PM formation, 26.49%
611 photochemical ozone formation, and 23.35% ozone formation compared to the current U.S. EoL
612 waste management pathway. Overall, cascaded waste processing is effective in chronic polymeric
613 material losses and pollution mitigation, which fits and sheds light on the EoL waste management
614 policy implications corresponding to the zero-waste policy, environmentally-specific policies, and
615 the plastic pollution act.
616 Practices in the circular plastic economy[35] aided by cascaded waste processing can minimize
617 environmental burdens from waste losses and virgin chemical production via closing the cycle [72].
618 Specifically, cascaded waste processing can cut the environmental release and exposure to reduce
619 pollution and help achieve zero-waste by reducing over 99.99% material losses. Incentivizing
620 cascade plastic waste processing also supports recovering the solid wastes onsite into monomeric
621 products and fuels to offset their external manufacturing and avoid its environmental consequences
622 by at least 4.35%. Enhancing the monomer recovery rate, which determines plastic reuse time, by
623 advancing the fast pyrolysis catalyst design[73], reaction condition optimization[74], and meticulous
624 control of yield separation[17] enable cascaded waste processing to reduce environmental losses. In
625 practice, these technology innovations should combine with effective solid waste sorting

technologies to handle polymer wastes with complex chemical compositions.

Plastic wastes can limit their pollution if treated by environmentally sustainable EoL management with minimum toxic release and gas emissions. Landfills should be replaced with other effective EoL waste management technologies that facilitate cascaded plastic waste management, such as chemical reclamation, with governmental incentives to promote pollution mitigation and reduce 23.35% life cycle ozone formation and 19.91% air pollution posed by PM. The environmental benefits of cascade plastic waste processing can be further improved by reducing energy use and effective off-gas emission control and mitigation by integrating carbon capture sequestration or capture technologies.

Adopting advanced and sustainable materials like cellulose products can replace fossil alternatives and generate fewer material losses[75-76]. Existing studies have also supported informing policies for using these materials to substitute plastic with less pollution[77]. Future studies can investigate the pros and cons of displacing polyolefin plastics with advanced or sustainable materials that can be reused multiple times, specifically focusing on the effects of limiting material losses and their derived environmental impacts. However, these technological innovations for achieving high natural compatibility and degradability by referencing the current Green Chemistry framework are not a permanent solution to reducing pollution or other environmental drawbacks[78], given our findings on high environmental exposure and hazards of plastic losses from improper EoL waste management. Minimizing environmental exposure via integrating effective solid waste collection with meticulous material release control or elimination can mitigate short-term environmental consequences of plastic losses and impede their four-fold increment in long-term effects. Direct MP reclamation methods proposed by existing experimental studies can also cease these environmental exposures and consequences[79]. Improving the technology maturity of these MP recycling processes and incorporating them with cascaded waste processing still requires scale-ups and government incentives for future development.

Testing and galvanizing the cascaded plastic use supported by the policy implications is critical, not only for gaining environmental benefits but also for maintaining the potential economic growth and positive societal effects of enhancing plastic circularities. However, practical implementation of this process is currently hindered by technology limitations for treating plastic waste mixtures and contaminated plastics with minimal energy usage and material loss. To address these limitations, government incentives for identifying the best cascaded use method for each

type of plastic or mixture are necessary for maximizing reuse and sustainable EoL treatment. On the other hand, future societal impact analyses on the proper solid waste collections and effective conversions, especially cascaded use, can advance our understanding of how and where this technology should go to gain its maximum full-spectrum sustainability benefits. All these technological implications can aid in reaching a global treaty on plastic pollution mitigation by fostering transparent information sharing between scientific and negotiating communities.

Supporting Information

Input data and model formulation for the mass-balance based PP post-disposal fate model, time dynamic environmental assessment methodology and results of plastic losses, absolute LCA results of PP plastics treated by waste EoL options, and environmental breakdowns of PP plastics treated by waste EoL options.

Acknowledgments

This material is based upon work supported by the National Science Foundation (NSF) under Grant No. 1643244 and 2029327.

References

(1) Horton, A. A., Plastic pollution: When do we know enough? *Journal of Hazardous Materials* **2022,** *422*, 126885.
(2) Cywar, R. M.; Rorrer, N. A.; Hoyt, C. B.; Beckham, G. T.; Chen, E. Y.-X., Bio-based polymers with performance-advantaged properties. *Nature Reviews Materials* **2022,** *7* (2), 83-103.
(3) Thomas, K. V., Understanding the plastics cycle to minimize exposure. *Nature Sustainability* **2022,** *5* (4), 282-284.
(4) Rahimi, A.; García, J. M., Chemical recycling of waste plastics for new materials production. *Nature Reviews Chemistry* **2017,** *1* (6), 1-11.
(5) Wen, Z.; Xie, Y.; Chen, M.; Dinga, C. D., China's plastic import ban increases prospects of environmental impact mitigation of plastic waste trade flow worldwide. *Nature communications* **2021,** *12* (1), 1-9.
(6) Min, K.; Cuiffi, J. D.; Mathers, R. T., Ranking environmental degradation trends of plastic marine debris based on physical properties and molecular structure. *Nature communications* **2020,** *11* (1), 1-11.
(7) Fojt, J.; Denková, P.; Brtnický, M.; Holátko, J.; Řezáčová, V.; Pecina, V.; Kučerík, J., Influence of Poly-3-hydroxybutyrate Micro-Bioplastics and Polyethylene Terephthalate Microplastics on the

691 Soil Organic Matter Structure and Soil Water Properties. *Environmental Science & Technology*
692 **2022,** *56* (15), 10732-10742, DOI: 10.1021/acs.est.2c01970.
693 (8) Galloway, T. S.; Cole, M.; Lewis, C., Interactions of microplastic debris throughout the marine
694 ecosystem. *Nature ecology & evolution* **2017,** *1* (5), 1-8.
695 (9) Allen, D.; Allen, S.; Abbasi, S.; Baker, A.; Bergmann, M.; Brahney, J.; Butler, T.; Duce, R. A.;
696 Eckhardt, S.; Evangeliou, N., Microplastics and nanoplastics in the marine-atmosphere
697 environment. *Nature Reviews Earth & Environment* **2022**, 1-13.
698 (10) Li, L.; Luo, Y.; Li, R.; Zhou, Q.; Peijnenburg, W. J.; Yin, N.; Yang, J.; Tu, C.; Zhang, Y.,
699 Effective uptake of submicrometre plastics by crop plants via a crack-entry mode. *Nature*
700 *Sustainability* **2020,** *3* (11), 929-937.
701 (11) Revell, L. E.; Kuma, P.; Le Ru, E. C.; Somerville, W. R.; Gaw, S., Direct radiative effects of
702 airborne microplastics. *Nature* **2021,** *598* (7881), 462-467.
703 (12) Wu, X.; Chen, X.; Jiang, R.; You, J.; Ouyang, G., New insights into the photo-degraded
704 polystyrene microplastic: Effect on the release of volatile organic compounds. *Journal of*
705 *Hazardous Materials* **2022,** *431*, 128523.
706 (13) Morgado, V.; Palma, C.; Bettencourt da Silva, R. J., Bottom-Up Evaluation of the Uncertainty
707 of the Quantification of Microplastics Contamination in Sediment Samples. *Environmental*
708 *Science & Technology* **2022,** *56* (15), 11080-11090.
709 (14) Pfohl, P.; Wagner, M.; Meyer, L.; Domercq, P.; Praetorius, A.; Hüffer, T.; Hofmann, T.;
710 Wohlleben, W., Environmental degradation of microplastics: how to measure fragmentation rates
711 to secondary micro-and nanoplastic fragments and dissociation into dissolved organics.
712 *Environmental Science & Technology* **2022,** *56* (16), 11323-11334.
713 (15) Schwarz, A. E.; Ligthart, T. N.; Boukris, E.; Van Harmelen, T., Sources, transport, and
714 accumulation of different types of plastic litter in aquatic environments: a review study. *Marine*
715 *Pollution Bulletin* **2019,** *143*, 92-100.
716 (16) Sharma, R.; Bansal, P. P., Use of different forms of waste plastic in concrete–a review.
717 *Journal of Cleaner Production* **2016,** *112*, 473-482.
718 (17) Bora, R. R.; Wang, R.; You, F., Waste polypropylene plastic recycling toward climate change
719 mitigation and circular economy: energy, environmental, and technoeconomic perspectives. *ACS*
720 *Sustainable Chemistry & Engineering* **2020,** *8* (43), 16350-16363.
721 (18) Hahladakis, J. N.; Iacovidou, E., An overview of the challenges and trade-offs in closing the
722 loop of post-consumer plastic waste (PCPW): Focus on recycling. *Journal of Hazardous Materials*
723 **2019,** *380*, 120887.
724 (19) Hu, Q.; Ok, Y. S.; Wang, C.-H., Sustainable and highly efficient recycling of plastic waste
725 into syngas via a chemical looping scheme. *Environmental Science & Technology* **2022,** *56* (12),
726 8953-8963.
727 (20) Ni, B.-J.; Zhu, Z.-R.; Li, W.-H.; Yan, X.; Wei, W.; Xu, Q.; Xia, Z.; Dai, X.; Sun, J.,
728 Microplastics mitigation in sewage sludge through pyrolysis: The role of pyrolysis temperature.
729 *Environmental Science & Technology Letters* **2020,** *7* (12), 961-967.
730 (21) Roosen, M.; Mys, N.; Kusenberg, M.; Billen, P.; Dumoulin, A.; Dewulf, J.; Van Geem, K.
731 M.; Ragaert, K.; De Meester, S., Detailed analysis of the composition of selected plastic packaging
732 waste products and its implications for mechanical and thermochemical recycling. *Environmental*
733 *science & technology* **2020,** *54* (20), 13282-13293.
734 (22) Alston, S. M.; Arnold, J. C., Environmental impact of pyrolysis of mixed WEEE plastics part
735 2: life cycle assessment. *Environmental science & technology* **2011,** *45* (21), 9386-9392.
736 (23) Hogue, C. Chemical recycling of plastic gets a boost in 18 US states—but environmentalists

question whether it really is recycling. https://cen.acs.org/environment/recycling/plastic-recycling-chemical-advanced-fuel-pyrolysis-state-laws/100/i17 (accessed May 15, 2022).
(24) Munir, M. T.; Mansouri, S. S.; Udugama, I. A.; Baroutian, S.; Gernaey, K. V.; Young, B. R., Resource recovery from organic solid waste using hydrothermal processing: Opportunities and challenges. *Renewable and Sustainable Energy Reviews* **2018,** *96*, 64-75.
(25) Gracida-Alvarez, U. R.; Winjobi, O.; Sacramento-Rivero, J. C.; Shonnard, D. R., System analyses of high-value chemicals and fuels from a waste high-density polyethylene refinery. Part 1: conceptual design and techno-economic assessment. *ACS Sustainable Chemistry & Engineering* **2019,** *7* (22), 18254-18266.
(26) Dai, L.; Zhou, N.; Lv, Y.; Cheng, Y.; Wang, Y.; Liu, Y.; Cobb, K.; Chen, P.; Lei, H.; Ruan, R., Pyrolysis technology for plastic waste recycling: A state-of-the-art review. *Progress in Energy and Combustion Science* **2022,** *93*, 101021.
(27) Dogu, O.; Pelucchi, M.; Van de Vijver, R.; Van Steenberge, P. H.; D'hooge, D. R.; Cuoci, A.; Mehl, M.; Frassoldati, A.; Faravelli, T.; Van Geem, K. M., The chemistry of chemical recycling of solid plastic waste via pyrolysis and gasification: State-of-the-art, challenges, and future directions. *Progress in Energy and Combustion Science* **2021,** *84*, 100901.
(28) Jeswani, H.; Krüger, C.; Russ, M.; Horlacher, M.; Antony, F.; Hann, S.; Azapagic, A., Life cycle environmental impacts of chemical recycling via pyrolysis of mixed plastic waste in comparison with mechanical recycling and energy recovery. *Science of the Total Environment* **2021,** *769*, 144483.
(29) Klemeš, J. J.; Fan, Y. V.; Jiang, P., Plastics: friends or foes? The circularity and plastic waste footprint. *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects* **2021,** *43* (13), 1549-1565.
(30) Cox, K. D.; Covernton, G. A.; Davies, H. L.; Dower, J. F.; Juanes, F.; Dudas, S. E., Human consumption of microplastics. *Environmental science & technology* **2019,** *53* (12), 7068-7074.
(31) Hale, R. C.; King, A. E.; Ramirez, J. M.; La Guardia, M.; Nidel, C., Durable Plastic Goods: A Source of Microplastics and Chemical Additives in the Built and Natural Environments. *Environmental Science & Technology Letters* **2022,** *9* (10), 798-807.
(32) Zhang, Q.; Xu, E. G.; Li, J.; Chen, Q.; Ma, L.; Zeng, E. Y.; Shi, H., A review of microplastics in table salt, drinking water, and air: direct human exposure. *Environmental science & technology* **2020,** *54* (7), 3740-3751.
(33) Zhao, X.; You, F., Life Cycle Assessment of Microplastics Reveals Their Greater Environmental Hazards than Mismanaged Polymer Waste Losses. *Environmental Science & Technology* **2022,** *56* (16), 11780-11797, DOI: 10.1021/acs.est.2c01549.
(34) Rorrer, J. E.; Troyano-Valls, C.; Beckham, G. T.; Román-Leshkov, Y., Hydrogenolysis of polypropylene and mixed polyolefin plastic waste over Ru/C to produce liquid alkanes. *ACS Sustainable Chemistry & Engineering* **2021,** *9* (35), 11661-11666.
(35) Rosenboom, J.-G.; Langer, R.; Traverso, G., Bioplastics for a circular economy. *Nature Reviews Materials* **2022,** *7* (2), 117-137.
(36) Zhang, J.; Zou, G.; Wang, X.; Ding, W.; Xu, L.; Liu, B.; Mu, Y.; Zhu, X.; Song, L.; Chen, Y., Exploring the occurrence characteristics of microplastics in typical maize farmland soils with long-term plastic film mulching in northern China. *Frontiers in Marine Science* **2021,** *8*, 800087.
(37) Wang, Y.; Levis, J. W.; Barlaz, M. A., An assessment of the dynamic global warming impact associated with long-term emissions from landfills. *Environmental Science & Technology* **2019,** *54* (3), 1304-1313.
(38) Gast, L.; Cabrera Serrenho, A.; Allwood, J. M., What contribution could industrial symbiosis

783 make to mitigating industrial greenhouse gas (GHG) emissions in bulk material production?
784 *Environmental science & technology* **2022,** *56* (14), 10269-10278.
785 (39) Faragò, M.; Damgaard, A.; Logar, I.; Rygaard, M., Life Cycle Assessment and Cost-Benefit
786 Analysis of Technologies in Water Resource Recovery Facilities: The Case of Sludge Pyrolysis.
787 *Environmental science & technology* **2022,** *56* (24), 17988-17997.
788 (40) Ji, L.; Wang, Y.; Xie, Y.; Xu, M.; Cai, Y.; Fu, S.; Ma, L.; Su, X., Potential Life-Cycle
789 Environmental Impacts of the COVID-19 Nucleic Acid Test. *Environmental science & technology*
790 **2022,** *56* (18), 13398-13407.
791 (41) Littlefield, J.; Rai, S.; Skone, T. J., Life Cycle GHG Perspective on US Natural Gas Delivery
792 Pathways. *Environmental science & technology* **2022,** *56* (22), 16033-16042.
793 (42) Zhao, X.; You, F., Consequential Life Cycle Assessment and Optimization of High-Density
794 Polyethylene Plastic Waste Chemical Recycling. *ACS Sustainable Chemistry & Engineering* **2021,**
795 *9* (36), 12167-12184, DOI: 10.1021/acssuschemeng.1c03587.
796 (43) Zhao, X.; You, F., Waste high‐density polyethylene recycling process systems for mitigating
797 plastic pollution through a sustainable design and synthesis paradigm. *AIChE Journal* **2021,** *67*
798 (4), e17127, DOI: 10.1002/aic.17127.
799 (44) Wang, S.-W.; Georgopoulos, P. G.; Li, G.; Rabitz, H., Random sampling− high dimensional
800 model representation (RS− HDMR) with nonuniformly distributed variables: application to an
801 integrated multimedia/multipathway exposure and dose model for trichloroethylene. *The Journal*
802 *of Physical Chemistry A* **2003,** *107* (23), 4707-4716.
803 (45) Bronstert, A.; de Araújo, J.-C.; Batalla, R. J.; Costa, A. C.; Delgado, J. M.; Francke, T.;
804 Foerster, S.; Guentner, A.; López-Tarazón, J. A.; Mamede, G. L., Process-based modelling of
805 erosion, sediment transport and reservoir siltation in mesoscale semi-arid catchments. *Journal of*
806 *Soils and Sediments* **2014,** *14* (12), 2001-2018.
807 (46) Lu, T.; Solis-Ramos, E.; Yi, Y.; Kumosa, M., UV degradation model for polymers and
808 polymer matrix composites. *Polymer degradation and stability* **2018,** *154*, 203-210.
809 (47) Maga, D.; Galafton, C.; Blömer, J.; Thonemann, N.; Özdamar, A.; Bertling, J., Methodology
810 to address potential impacts of plastic emissions in life cycle assessment. *The International*
811 *Journal of Life Cycle Assessment* **2022,** *27* (3), 469-491.
812 (48) Li, D.; Shi, Y.; Yang, L.; Xiao, L.; Kehoe, D. K.; Gun'ko, Y. K.; Boland, J. J.; Wang, J. J.,
813 Microplastic release from the degradation of polypropylene feeding bottles during infant formula
814 preparation. *Nature Food* **2020,** *1* (11), 746-754.
815 (49) Chamas, A.; Moon, H.; Zheng, J.; Qiu, Y.; Tabassum, T.; Jang, J. H.; Abu-Omar, M.; Scott,
816 S. L.; Suh, S., Degradation rates of plastics in the environment. *ACS Sustainable Chemistry &*
817 *Engineering* **2020,** *8* (9), 3494-3511.
818 (50) Vega, G. C.; Gross, A.; Birkved, M., The impacts of plastic products on air pollution-A
819 simulation study for advanced life cycle inventories of plastics covering secondary microplastic
820 production. *Sustainable Production and Consumption* **2021,** *28*, 848-865.
821 (51) Niki, H.; Maker, P.; Savage, C.; Breitenbach, L., A Fourier transform infrared study of the
822 kinetics and mechanism for the reaction hydroxyl+ methyl hydroperoxide. *The Journal of Physical*
823 *Chemistry* **1983,** *87* (12), 2190-2193.
824 (52) Atkinson, R., Kinetics and mechanisms of the gas-phase reactions of the hydroxyl radical
825 with organic compounds under atmospheric conditions. *Chemical Reviews* **1986,** *86* (1), 69-201.
826 (53) Altshuller, A., The production of carbon monoxide by the homogeneous NO x-induced
827 photooxidation of volatile organic compounds in the troposphere. *Journal of atmospheric*
828 *chemistry* **1991,** *13* (2), 155-182.

(54) Fan, Y. V.; Jiang, P.; Tan, R. R.; Aviso, K. B.; You, F.; Zhao, X.; Lee, C. T.; Klemes, J. J., Forecasting plastic waste generation and interventions for environmental hazard mitigation. *Journal of Hazardous Materials* **2022,** *424*, 127330, DOI: 10.1016/j.jhazmat.2021.127330.
(55) Wang, Y.; Levis, J. W.; Barlaz, M. A., Life-Cycle Assessment of a Regulatory Compliant US Municipal Solid Waste Landfill. *Environmental Science & Technology* **2021,** *55* (20), 13583-13592.
(56) Huijbregts, M. A.; Steinmann, Z. J.; Elshout, P. M.; Stam, G.; Verones, F.; Vieira, M.; Hollander, A.; Zijp, M.; van Zelm, R., ReCiPe 2016: a harmonized life cycle impact assessment method at midpoint and endpoint level report I: characterization. **2016**.
(57) Höglmeier, K.; Weber-Blaschke, G.; Richter, K., Utilization of recovered wood in cascades versus utilization of primary wood—a comparison with life cycle assessment using system expansion. *The International Journal of Life Cycle Assessment* **2014,** *19* (10), 1755-1766.
(58) Masry, M.; Rossignol, S.; Gardette, J.-L.; Therias, S.; Bussière, P.-O.; Wong-Wah-Chung, P., Characteristics, fate, and impact of marine plastic debris exposed to sunlight: A review. *Marine Pollution Bulletin* **2021,** *171*, 112701.
(59) Kooi, M.; Besseling, E.; Kroeze, C.; Van Wezel, A. P.; Koelmans, A. A., Modeling the fate and transport of plastic debris in freshwaters: review and guidance. *Freshwater microplastics: Emerging environmental contaminants?* **2018**, 125-152.
(60) Ali, M. U.; Lin, S.; Yousaf, B.; Abbas, Q.; Munir, M. A. M.; Ali, M. U.; Rasihd, A.; Zheng, C.; Kuang, X.; Wong, M. H., Environmental emission, fate and transformation of microplastics in biotic and abiotic compartments: Global status, recent advances and future perspectives. *Science of The Total Environment* **2021,** *791*, 148422.
(61) Besseling, E.; Quik, J. T.; Sun, M.; Koelmans, A. A., Fate of nano-and microplastic in freshwater systems: A modeling study. *Environmental pollution* **2017,** *220*, 540-548.
(62) Brendel, C. E.; Dymond, R. L.; Aguilar, M. F., Integration of quantitative precipitation forecasts with real-time hydrology and hydraulics modeling towards probabilistic forecasting of urban flooding. *Environmental Modelling & Software* **2020,** *134*, 104864.
(63) Plastics: Material-Specific Data. https://www.epa.gov/facts-and-figures-about-materials-waste-and-recycling/plastics-material-specific-data (accessed Sep. 10).
(64) Ragaert, K.; Delva, L.; Van Geem, K., Mechanical and chemical recycling of solid plastic waste. *Waste management* **2017,** *69*, 24-58.
(65) Zhao, X.; You, F., Waste respirator processing system for public health protection and climate change mitigation under COVID-19 pandemic: Novel process design and energy, environmental, and techno-economic perspectives. *Applied Energy* **2021,** *283*, 116129, DOI: 10.1016/j.apenergy.2020.116129.
(66) Geyer, R.; Jambeck, J. R.; Law, K. L., Production, use, and fate of all plastics ever made. *Science advances* **2017,** *3* (7), e1700782.
(67) Zhao, X.; Klemeš, J. J.; You, F., Energy and environmental sustainability of waste personal protective equipment (PPE) treatment under COVID-19. *Renewable and Sustainable Energy Reviews* **2022,** *153*, 111786.
(68) Vollmer, I.; Jenks, M. J.; Roelands, M. C.; White, R. J.; van Harmelen, T.; de Wild, P.; van Der Laan, G. P.; Meirer, F.; Keurentjes, J. T.; Weckhuysen, B. M., Beyond mechanical recycling: Giving new life to plastic waste. *Angewandte Chemie International Edition* **2020,** *59* (36), 15402-15423.
(69) Mitrano, D. M.; Wagner, M., A sustainable future for plastics considering material safety and preserved value. *Nature Reviews Materials* **2022,** *7* (2), 71-73.

(70) Gontard, N.; David, G.; Guilbert, A.; Sohn, J., Recognizing the long-term impacts of plastic particles for preventing distortion in decision-making. *Nature Sustainability* **2022**, 1-7.
(71) Peng, Y.; Wu, P.; Schartup, A. T.; Zhang, Y., Plastic waste release caused by COVID-19 and its fate in the global ocean. *Proceedings of the National Academy of Sciences* **2021,** *118* (47), e2111530118.
(72) Chin, H. H.; Varbanov, P. S.; You, F.; Sher, F.; Klemes, J. J., Plastic Circular Economy Framework using Hybrid Machine Learning and Pinch Analysis. *Resources, Conservation and Recycling* **2022,** *184*, 106387, DOI: 10.1016/j.resconrec.2022.106387.
(73) Huang, J.; Veksha, A.; Chan, W. P.; Giannis, A.; Lisak, G., Chemical recycling of plastic waste for sustainable material management: A prospective review on catalysts and processes. *Renewable and Sustainable Energy Reviews* **2022,** *154*, 111866.
(74) Queiroz, A.; Pedroso, G. B.; Kuriyama, S. N.; Fidalgo-Neto, A. A., Subcritical and supercritical water for chemical recycling of plastic waste. *Current Opinion in Green and Sustainable Chemistry* **2020,** *25*, 100364.
(75) Krupp, L. R.; Jewell, W. J., Biodegradability of modified plastic films in controlled biological environments. *Environmental science & technology* **1992,** *26* (1), 193-198.
(76) Zhao, X.; Klemeš, J. J.; Saxon, M.; You, F., How sustainable are the biodegradable medical gowns via environmental and social life cycle assessment? *Journal of Cleaner Production* **2022,** *380*, 135153, DOI: 10.1016/j.jclepro.2022.135153.
(77) McDevitt, J. P.; Criddle, C. S.; Morse, M.; Hale, R. C.; Bott, C. B.; Rochman, C. M., Addressing the Issue of Microplastics in the Wake of the Microbead-Free Waters Act A New Standard Can Facilitate Improved Policy. *Environmental science & technology* **2017,** *51* (12), 6611-6617.
(78) Mitrano, D. M.; Wohlleben, W., Microplastic regulation should be more precise to incentivize both innovation and environmental safety. *Nature communications* **2020,** *11* (1), 1-12.
(79) Arpia, A. A.; Chen, W.-H.; Ubando, A. T.; Naqvi, S. R.; Culaba, A. B., Microplastic degradation as a sustainable concurrent approach for producing biofuel and obliterating hazardous environmental effects: a state-of-the-art review. *Journal of Hazardous Materials* **2021,** *418*, 126381.

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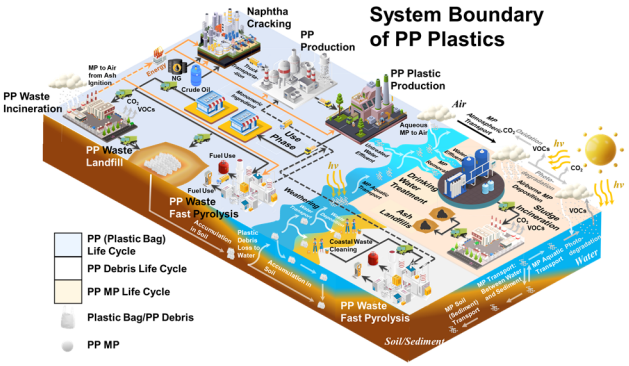

Description: Cascading plastic macro debris chemical upcycling and microparticle removal processes to reduce material losses and their derived pollution from incineration and landfills.