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From sustainable macro debris chemical recycling to microplastic reclamation: Overview, research challenges, and outlook

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ABSTRACT

The current surge in plastic waste generation from human activities has led to environmental hazards associated with Sustainable Development Goal (SDG) 6 (Clean Water and Sanitation), from mismanaged polymeric waste and its derived free plastic debris. Chemical recycling enables the effective end-of-life (EoL) of plastic wastes and debris by transforming them into value-added chemicals, reducing their offsite manufacturing's environmental and social risks related to SDG 13 (Climate Action) while mitigating plastic pollution associated with SDG 6, SDG 14 (Life Below Water), and SDG 15 (Life on Land). Herein, this work examines the recent research progress, technological advancements, and policy implications on the chemical recycling of plastic waste and derived debris from macro to micro levels and implications for achieving SDGs. We identify three primary research challenges related to plastic pollution mitigation: 1. Understanding the extent of plastic pollution and consequences by sources originating from human activities; 2. Exploring the sustainable material life cycle of plastics with minimum waste generation, free debris exposure and pollution related to SDGs 14 and 15, and carbon emissions aligned with SDG 13; 3. Indicating the necessary local and global efforts to deploy plastic production, use, and waste management as per sustainable plastic life cycle concepts associated with SDG 12 (Responsible Production and Consumption). Future perspective directions lie in mitigating plastic pollution on both the sources identified by plastic waste and derived debris generation from individual human activity and the sinks associated with free debris exposure to natural environments. Reducing plastic use, waste reuse, repurposing, and recycling can fully mitigate these negative externalities but also generate greenhouse gas emissions, which requires decarbonization under climate commitments.

1. Introduction

Since its first invention in 1950, plastics have benefited our lives for decades through their high durability with the virtue of hydrophobicity, good physical properties, and low manufacturing costs (Ügdüler et al., 2020). In 2020, over 390 million tons of plastics were produced for industrial and residential use (PlasticEurope, 2023), posing fresh strains to the global waste management capacities, particularly in developing economies, and resulting in 367 million tons of discarded materials globally (Co-operation and Development, 2022). While plastics' good chemical inertness has benefited many Anthropocene activities, it also contributed to the long-term retention of mismanaged wastes in the natural environment for millennia (Fojt et al., 2022). Untreated plastic waste spreads through air, water, and land, releasing debris in macro (>5 mm) and micro sizes (<5 mm), depicted by macro- and microplastics, that burden air, water, soil, and ecosystems (Cappello et al., 2022). Chemical additives in plastic wastes, including Bisphenol A, acidify the aquatic environments, and plastic debris, especially microplastics, can pollute the air and harm human health through ingestion and respiration (Sheldon and Norton, 2020; Zhao and You, 2024). Although technological advancements in non-toxic, recyclable, and biodegradable materials can mitigate the environmental risks of single-use plastics, replacing all conventionally used polymers with advanced materials is currently cost-prohibitive (Sheldon and Norton, 2020). Therefore, greening the plastic end-of-life (EoL) by reducing solid waste generation and material losses through repurposing and economically feasible reuse of discarded materials is essential for mitigating the plastic's life cycle environmental burdens aligned with the United Nations' (UN's) Sustainable Development Goals (SDGs).

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Abbreviations

A&HCI	Arts & Humanities Citation Index
BAU	Business-as-usual
BECCS	Bioenergy with carbon capture and storage
CCS	Carbon capture and sequestration
CCU	Carbon capture and utilization
EoL	End-of-life
ESCI	Emerging Sources Citation Index
GHG	Greenhouse Gas
LCA	Life cycle assessment
MFA	Material flow analysis
PC	Polycarbonate
PE	Polyethylene
PET	Polyethylene terephthalate
PLA	Polylactic acid
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride
SCIE	Science Citation Index Expanded
SDG	Sustainable Development Goal
SSCI	Social Sciences Citation Index
UN	United Nations
UNEP	United Nations Environmental Programme
VOC	Volatile organic chemicals
3R	Reuse, repurposing, and recycling

Chemical recycling effectively converts environmentally harmful plastic wastes into alternative chemicals that can substitute their offsite production (Rahimi and García, 2017). Currently, lab-scale chemical reclamation studies on downgrading (Lim et al., 2023), recycling (Zhao and You, 2023), or upgrading (Yuan et al., 2022) discarded plastics were surging, with some pilot projects backed by governmental incentives, such as 18 new U.S. chemical recycling plants endorsed by policies from the American Chemistry Council (Hogue, 2022). While most plastic waste conversion practices focus on macro debris treatment, incineration remains the primary solution for reducing plastic losses, generating tons of greenhouse gas (GHG) emissions if all these discarded materials are treated (Zhao and You, 2022). With chemical properties similar to bulk plastics, polymeric debris can also be treated in existing solid waste chemical reclamation processes (Chen et al., 2022). Onsite tandem conversion of multiple types of plastic wastes, either alone or with their derived debris, can produce crucial input chemicals for onsite conversion of other wastes and improve environmental sustainability by reducing offsite chemical manufacturing (Pichler et al., 2021). When adopting new technologies, it is crucial to weigh the technology readiness level (Garcia and Robertson, 2017), yields of alternative chemicals (Vollmer et al., 2020), and their cost effectiveness (Ma et al., 2023), as well as incurred environmental emissions and social risks, such as worker safety concerns from onsite and offsite energy generation and raw chemical production (Zhao et al., 2022a). Balancing and enhancing these economic, environmental, and social factors are essential for sustaining the application of the plastic chemical recycling process to mitigate pollution in practice. A holistic sustainability analysis, encompassing economic, environmental, and social aspects, can help identify the pros and cons of adopting technologically viable plastic chemical recycling processes for pollution mitigation whilst maintaining sustainability (Fan et al., 2022). Assessing the sustainability of chemical recycling of plastic wastes should consider the entire life cycle, including those of bulk plastics and the formation, transmission, and EoL fate of derived particles (Hu et al., 2022b), to inform technological innovations that address both macro- and micro-wastes simultaneously. An ideal case is to minimize plastic waste generation to achieve zero waste, as the advanced EoL management technology can handle all accumulative untreated wastes in the long term (Zimmerman et al., 2020). However, this study did not consider the potential environmental impacts of material losses, such as microplastic emissions, from plastic production and waste management, which remains a knowledge gap in most existing studies.

Examining the effectiveness of adopting these chemical recycling technologies for plastic pollution mitigation, as the United Nations Environmental Programme (UNEP) recommended, requires understanding the level of accumulative plastic pollution globally and its derived environmental consequences, both of which remain knowledge gaps in current studies (Lau et al., 2020). Specifically, investigating the level of plastic debris generation from human activities and exposure to natural environments, which also remains unclear in existing studies, is crucial to indicate the environmental aftermaths of global plastic pollution and inform effective mitigation measures (Lau et al., 2020). One direct consequence of plastic debris exposure is uptake by humans in the form of microplastics from diet and respiration, which are identified as potential public health risks (Yan et al., 2021). Relevant studies on plastic debris human uptake focused on the microplastic uptake source and origin without explicitly investigating the effectiveness of plastic pollution reduction measures (Yan et al., 2021). Under the global plastic treaty, mitigation measures of plastic pollution posed by macro- and microplastic debris demand global and local actions on minimizing plastic waste generation, mismanagement, and free debris exposure within the material life cycle. Efforts for mitigating local plastic pollution, such as chemical recycling, may vary with the plastic waste types, amounts, and technological readiness for advanced plastic pollution mitigation technologies across different regions with various social-economic contexts (Kubiczek et al., 2023). Current plastic waste and debris mitigation measures mostly remain localized (Lau et al., 2020), and understanding what, when, and how to adopt these sustainable measures across regions is critical and an unmet research need (Zhao and You, 2023).

This paper summarizes the current research progress in plastic waste chemical recycling from the technology, environmental, economic, and social sustainability perspectives, highlights the existing research challenges and unmet research needs, and suggests future research directions to fill these knowledge gaps under the green chemistry context. Section 2 overviews the current plastic waste and debris chemical recycling and emerging microplastic processing technologies to draw key policy and technological insights listed in Section 3, which also identifies research challenges and knowledge gaps associated with understanding the extent of plastic pollution from human activities and the efforts needed for complete pollution mitigation. Addressing these research challenges involves assessing the amount of plastic waste and microplastics generated by each human activity and exploring the adoption of plastic chemical recycling measures to handle the current levels of plastic waste generated domestically and globally. Sections 4–6 present detailed background information, research questions, and future research directions to support further research in addressing these knowledge gaps. All sections are concluded in Section 7.

2. Sustainability analysis of plastic waste and macro- and microplastic debris chemical recycling

We review the relevant literature from 2002 to 2023 gathered from Science Citation Index Expanded (SCIE), Social Sciences Citation Index (SSCI), Arts & Humanities Citation Index (A&HCI), and Emerging Sources Citation Index (ESCI) in the Web of Science Core Collection. As shown in Fig. 1, the four typical chemical recycling processes classified by downstream product use are open-loop recycling, closed-loop

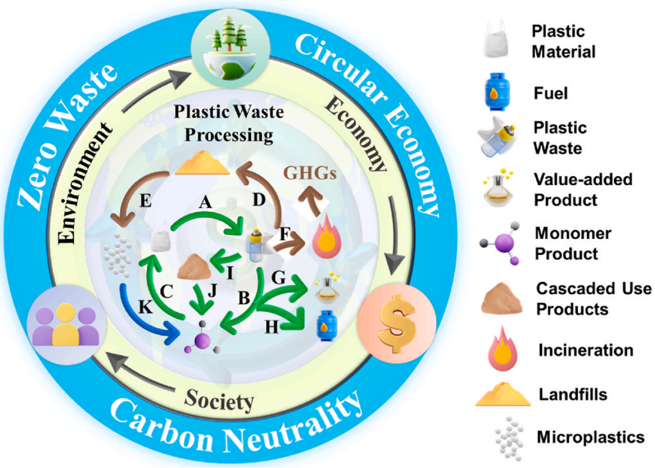

Fig. 1. Overview of the sustainable chemical recycling process and its sustainability assessment. Routes A–K are included in the life cycles of plastics treated by different EoL management practices. Closed-loop recycling converts plastic wastes from route A to basic chemicals by conversion route B, used for remanufacturing virgin materials indicated by route C. Plastic waste open-loop recycling yields basic chemicals for plastic production or other products by conversion route H, such as fossil fuels. The plastic cascade use utilizes wastes for secondary use in route I, and discarded materials are treated in tandem with bulk wastes in route J. Plastic upcycling adds values to the wastes from route A by producing hydrocarbons via route G. The combination of pathways D and E represents the plastic waste landfill and degradation, yielding microplastics that pollute the environment. Pathway F indicates the incineration process. Route F represents the incineration of plastic wastes generating greenhouse gases (GHGs), while route K denotes the chemical upcycling process converting microplastics into monomeric products.

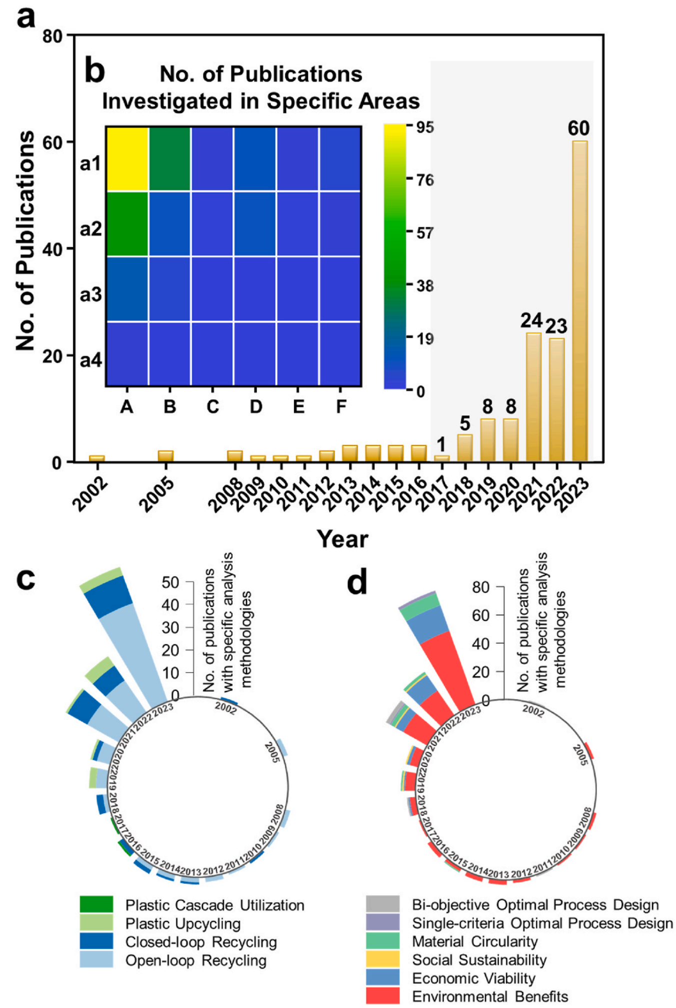

Fig. 2. The number of recent publications focusing on plastic waste chemical recycling by referring to the SCIE, SSCI, A&HCI, and ESCI in the Web of Science Core Collection. **a**, Temporal variations of the number of recent publications from 2002 to 2023. **b**, The distribution of the number of recent publications by recycling processes and methodology used. **c**, The breakdown of the number of publications from 2002 to 2023 by the target recycling processes. **d**, The breakdown of the number of publications from 2002 to 2023 by the sustainability analysis methodology used. The notations a1–a4 in all graphs represent the recycling processes classified by the use of downstream products: a1: Closed-loop recycling that converts wastes into monomers used for manufacturing original plastics; a2: Open-loop recycling that produces chemicals from wastes for alternative use, not limited to producing original plastics; a3, Upcycling that manufactures chemicals with added values over producing the monomers of virgin plastics; a4, Cascade use that reuses the waste multiply in alternative applications. A–F denote the assessment goal: A, Environmental benefits; B, Economic viability; C, Social sustainability; D, Material circularity; E, Single-criteria optimal process design; F, Bi-objective optimal process design.

recycling, upcycling, and cascade use (Davidson et al., 2021). Each subsequent subsection summarizes the current research progress for one type of chemical recycling process and its sustainability performance from environmental, economic, and social aspects.

2.1. Three sustainability pillars and assessments on macro- and microplastic chemical recycling

Fig. 2a shows the growing trend of chemical recycling sustainability analysis studies since the first publication on open-loop plastic chemical recycling's environmental analyses in 2002. Four studies were investigated in 2005 and 2008, all targeting the environmental performances of the open-loop plastic waste chemical recycling systems. Twenty-four studies on all three pillars of sustainability and optimization-based process design and synthesis were investigated for plastic close- and open-loop recycling and upcycling processes in 2021. Many research dedications emerged in 2022, with 23 publications in plastic chemical recycling associated with a similar research aim in 2021, while more studies on plastic waste chemical recycling sustainability analyses are provided in 2023. We found 60 relevant studies in 2023 (Ajaj et al., 2023; Alqahtani et al., 2023; Arena et al., 2023; Arifuzzaman et al., 2023; Azam et al., 2023; Berger et al., 2023; Boone et al., 2023; Chairat and Gheewala, 2023; Chari et al., 2023; Choi et al., 2023; Corella-Puertas et al., 2023; Cristóbal et al., 2023; Cudjoe et al., 2023; Dai et al., 2023; Diez-Cañamero and Mendoza, 2023; Dou et al., 2023; Gabisa et al., 2023; Gadaleta et al., 2023; Ghosh et al., 2023; Gracida-Alvarez et al., 2023; Hernández et al., 2023; Huang et al., 2023; Hubble et al., 2023; Ibrahim et al., 2023; Istrate et al., 2023; Iturrondobeitia et al., 2023; Jayawardane et al., 2023; Jin et al., 2023; Kuroda et al., 2023; Lang et al., 2023; Liang et al., 2023a, 2023b; Luo et al., 2023; Mumtaz et al., 2023; Muniyappan et al., 2023; Olafasakin et al., 2023a; Olafasakin et al., 2023b; Ozoemena and Coles, 2023a; Ragab et al., 2023; Ravina et al., 2023; Rebolledo-Leiva et al., 2023; Salah et al., 2023; Sangtani et al., 2023; Schmidt and Laner, 2023; Schneider et al., 2023; Schulte et al., 2023; Schwarz et al., 2023; Shan et al., 2023; Stallkamp et al., 2023; Stegmann et al., 2023; Tan et al., 2023; Tomatis et al., 2023; Verbeek et al., 2023; Vlasopoulos et al., 2023; Williams and Bourtsalas, 2023; Wyss et al., 2023; Yadav et al., 2023; Yan et al., 2023; Zhang et al., 2023a; Zhao et al., 2023), with 45.9%, 13.11%, 14.75%, 3.28%, and 22.95% of them focusing on "cradle-to-grave", "cradle-to--gate", "cradle-to-cradle", "gate-to-gate", and "gate-to-grave" analyses, respectively, as shown in Fig. 3a. 73.02% of the 2023 investigations studied open-loop recycling of plastic wastes, while 67.07% targeted environmental sustainability analyses. Detailed time trend of relevant studies can be found in the Supporting Information.

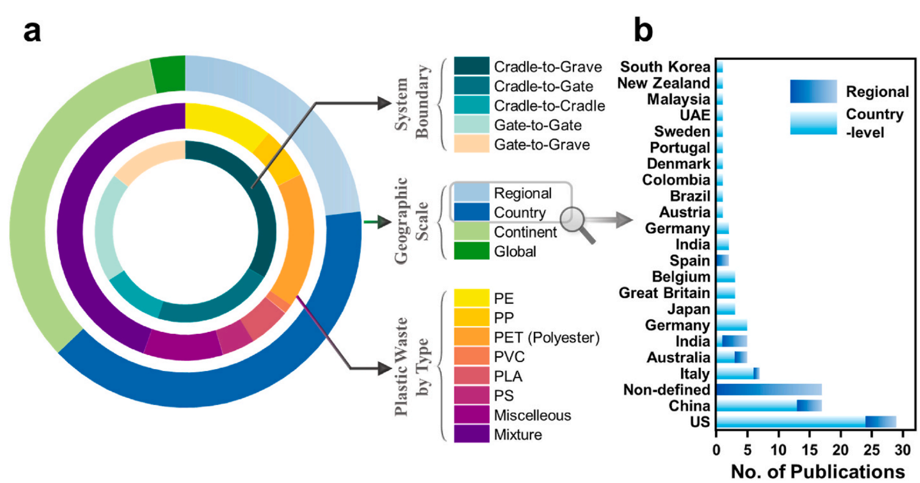

Fig. 3. The breakdown of the number of recent plastic waste chemical recycling publications. **a**, Total publications from 2002 to 2023 classified by the geographic scale depicted in the outer circle, plastic waste types depicted in the mid-circle, and system boundary denoted by the inner circle. **b**, Breakdowns of the regional and country-level analyses publications by countries. The non-defined indicates that the publication did not contain a region-specific investigation. The values to the right of the bar chart in Panel **b** show the number of publications in the regions associated with the y-axis values.

For the sustainability analysis methodology, footprint analyses focus on the impacts posed by a specific material/element over the complete life cycles of plastics, while life cycle assessment (LCA) can help evaluate based on full-spectrum impact categories, enabling a holistic understanding of the social and environmental impacts posed by plastic chemical recycling processes. The material flow analysis (MFA) focuses on certain materials and evaluates their metabolisms in the Anthroposphere based on mass and energy balance. The cash flow analysis aided by the techno-economic assessment can reflect the economic performance, interpreted by the net present value, levelized costs, and unit profits or revenue of the plastic chemical recycling system. Using all these studied environmental, economic, and social impact analysis methodologies can help inform future technological innovations for improving plastic waste chemical conversion and its effectiveness whilst minimizing sustainability risks. Based on the sustainability analysis goals of existing plastic chemical recycling processes, Fig. 3a shows five system boundary configurations classified by the goal and downstream product use: Cradle-to-grave, cradle-to-gate, cradle-to-cradle, gate-to-gate, and gate-to-grave.

Specifically, focusing on the waste mixture, relevant research performs sustainability investigation from the "factory" gate that generates waste plastics. Gate-to-gate studies focus on the chemical recycling process and evaluate its sustainability performance compiled by the mass and energy input/output. When expanding into the "gate-to-grave" system boundary, relevant sustainability analyses accounting for 20% of existing studies can help inform the sustainable chemical recycling practice from the product ends especially the EoL use of downstream products. The type of plastic waste can also influence the sustainability performance of waste chemical recycling processes. By considering the upstream processes within the plastic waste life cycles, including raw material extraction as cradle and polymer production, relevant studies can help inform the sustainable life cycle design of plastics. This can be achieved by comparing the target plastic materials and their solid waste disposal with conventional plastic processed by widely used EoL waste management, including incineration, open burning, and sanitary landfills. Due to the data unavailability in the downstream product EoL use, 27% of these studies only evaluate the sustainability performances from a "cradle-to-gate" system boundary that truncated the waste life cycle stages in the "factory" gate. 25% of the existing study defines the downstream product EoL uses, including fossil fuels, basic chemicals, and monomers for plastic remanufacturing, and evaluates the entire life cycle performances of plastic waste treated by chemical recycling processes. Cradle-to-cradle analysis, which aims to identify the pros and cons of chemically repurposing plastic wastes back to monomers, contributes to 8.7% of the reviewed literature.

2.2. Substitute upstream plastic use by alternative materials to mitigate pollution

Minimizing plastic pollution over the entire material life cycle demands mitigation measures from both material sources and sinks. Substituting plastic use with alternative materials combined with effective waste management enables minimum mismanaged plastic waste and derived debris release to the natural environment (Lazarevic et al., 2010). By durability, the type of plastic materials includes rigid and flexible plastics, which involve single or multiple chemical compositions of plastics (represented by mono- and multimaterial, respectively) (de Mello Soares et al., 2022). Flexible monomaterial plastics widely used for carrier bags, films, and household goods are recyclable with easy substitution, given their simple chemical composition (Guerritore et al., 2022). Biodegradable plastics, such as polylactic acid (PLA), as well as other cellulosic products involving fabrics, paper board cartons, coated papers, and wood, are typical alternative materials to replace flexible mono-material plastic use, especially single-use packaging materials (UNCTAD, 2021). These alternative materials showed similar elasticity, durability, and reusability compared to original plastic materials (UNCTAD, 2021). Rigid plastic materials, mostly polyolefins, such as polyethylene and polypropylene, are typically used for plastic bottles and business-to-business packaging, demanding high material durability, usability, and chemical inertness (Di et al., 2021). Current investigations on substitute materials of rigid plastics mainly focus on rigid inorganic materials, such as tinplate, aluminum, stainless steel, and glass (UNCTAD, 2021). However, higher manufacturing costs of these materials than the global average rigid plastic price hindered their substitution without effective government support (Reijnders, 2000). Moreover, the substitution of multilayered or multi-material plastics, both of which contribute to more than 15% U.S.'s domestic plastic wastes by mass (EPA, 2020), faces technological difficulties due to complex chemical compositions and remain inadequate in existing practices (de Mello Soares et al., 2022). For instance, Dow's "Recycle-Ready" has initiated replacing these materials with multiple mono-material plastic components sharing similar properties, but this shift may decrease the product quality and require more effective waste recycling, such as chemical recycling processes, to tackle impurities (de Mello Soares et al., 2022). Chemical recycling, especially thermal processing has been proven as effective technology to treat the wastes of these alternative materials and further mitigate the plastic pollution (Meys et al., 2020).

2.3. Upstream plastic waste processing and debris collection of macro- and microplastics before chemical recycling

Collection, sorting, and pretreatment of plastic waste and its derived material debris are crucial for the high yield of downstream products and for maintaining the practicability and sustainability of chemical recycling (Mangold and von Vacano, 2022). Most macro debris is easy to collect through ocean clean-ups and land-based debris collection, including curb-side waste collection (Groot et al., 2014), vehicular collection (Bing et al., 2014), and drop-off recycling (Larsen et al., 2010). However, the miniature sizes of microplastics, smaller than the pore size of the filter used in traditional manual sorting-based practices (Tirkey and Upadhyay, 2021), hamper their effective collection. Microplastics' high surface areas enhance the surface charge aided by the good particle hydrophilicity owing to natural weathering (Mukherjee et al., 2023), which inspires electrostatic interaction-based microplastic capture methods by using autonomous nano/microrobots to remove suspended plastic particles from the natural environment (Urso and Pumera, 2022). Good surface properties of microplastics also make surface-sensitive capturing methods (Bang et al.), such as using liquid crystals and bioreactors (Bang et al.; Mukherjee et al., 2023), effective in plastic particle removal at the lab scale. All collected macro plastic debris is then sorted by plastic type using float-sink (Pongstabodee et al., 2008), near intra-red (Ragossnig and Agamuthu, 2021), thermal adhesion (Lim et al., 2022a), dry zig-zag (Lee et al., 2022a), or electrostatic sorting methods (Tilmatine et al., 2009), or directly sent to chemical recycling facilities. For the treatment of collected wastes, the type of plastics can impact the composition and yield of downstream products, thus altering the three pillars of chemical recycling's sustainability performance (Ma et al., 2023). Plastic compounds can be classified into the following categories: polyethylene terephthalate (PET), polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polystyrene (PS), and miscellaneous plastics such as polycarbonate (PC), PLA, acrylic, acrylonitrile butadiene, styrene, fiberglass, and nylon. Polyolefin plastics, involving PE and PP, are widely investigated in the chemical recycling of single plastic compounds given their fossil fuel savings (Kumar et al., 2011), lower GHG emissions (Volk et al., 2021), and lighter plastic pollution (Lee et al., 2021a) compared to conventional EoL waste management, such as incineration and sanitary landfills. Chemical recycling studies of polyester and PLA plastics with high biodegradability mainly focus on the sustainability of the non-thermal technologies, including solvolysis (Aryan et al., 2021; Fehér et al., 2022), enzymatic recycling (Bendourou et al., 2021; Singh et al., 2021), and hydrolysis (Carta et al., 2003; Iñiguez-Franco et al., 2018), and provide technical and policy insights towards plastic pollution mitigation. For the sustainable conversion of other plastics, including PS (Achilias et al., 2007), PVC (Yu et al., 2016), and miscellaneous plastic wastes (Putra et al., 2022), thermal processing technologies, especially pyrolysis, is the primary research focus. These plastics can undergo thermal cracking and produce basic chemicals for polymer remanufacturing (Achilias et al., 2007), gas fuels (Yu et al., 2016), and liquid fuels onsite (Putra et al., 2022), yielding environmental, economic, and social benefits from substituting offsite production.

2.4. Closed-loop recycling of plastic waste and macro- and microplastic debris

Closed-loop recycling, which aims to convert collected plastic wastes back into its virgin form, namely monomers, used to remanufacture polymeric materials, in a closed recycling loop (Xu and Wang, 2022), has gained growing attention on a lab scale to enhance plastic waste conversion rate (Liu and Lu, 2023), monomer yields (Guillaume, 2023), and production efficiency of monomers back to polymeric materials (Uekert et al., 2023). This recycling process can typically treat polyester plastics involving PET or PLA, which can decompose under mild conditions (Aryan et al., 2021; Bendourou et al., 2021; Fehér et al., 2022; Singh et al., 2021). Recent research has also identified novel polymers, like poly(diketoenamine), that can self-recover from waste back to virgin materials under solvolysis (Christensen et al., 2019). Despite these technological advances, closed-loop recycling faces challenges when processing thermal-static plastics, such as polyolefins, which are difficult to crack into homogeneous monomers (Vogt et al., 2021). Although research efforts have been paid to improve operating conditions and versatility in handling waste mixtures, such as wise solvent use (Uekert et al., 2023), ambient reaction (Zhang et al., 2023b), and catalyst design for monomer recovery (Liu and Lu, 2023), recycling scale-ups are hindered by high capital costs (Larrain et al., 2020) and significant energy consumption from chemical input manufacturing (Liu et al., 2022d). These challenges, combined with the high chemical composition of polyolefin plastics in solid waste and debris (Zhao and You, 2023), including microplastics, make closed-loop recycling less effective in managing plastic waste. Chemical impurities, such as plasticizers and other additives (Dai et al., 2022), and residues from plastic degradation (Hu et al., 2022a), can also deactivate the catalysts used in closed-loop recycling, thus decreasing plastic waste's conversion rates and monomer yields. Meticulous waste separation processes, such as solvent-based selective separation (Walker et al., 2020), are required to split the polymeric mixture into virgin materials before closed-loop recycling for material remanufacturing. However, these processes can introduce incurred sustainability risks posed by additional infrastructure and input chemicals requirements for offsite production, which may offset the economic, environmental, and social benefits of reusing virgin plastics.

2.5. Open-loop recycling of plastic waste and macro- and microplastic debris

Reallocating the carbon that will form carbon dioxide in plastic waste incineration to alternative uses can extensively reduce GHG emissions and fossil fuel use (Williams et al., 2010). One sustainable approach for plastic waste conversion is to produce alternate hydrocarbon chemicals used as raw materials or renewable fuels, offsetting their external production and associated environmental and social risks (Lim et al., 2022b). Different from the closed-loop recycling that produces monomers for plastic remanufacturing in a closed recycling loop, open-loop recycling aims to convert solid wastes or mixtures from the collection into chemicals other than monomers for primary plastic remanufacturing (Sheldon, 2023). In contrast with closed-loop recycling, open-loop processes are less hampered by the chemical purity of plastic waste (Larrain et al., 2020). Current research has evidenced an effective conversion of polyolefin materials (Chu et al., 2022b), highly contaminated plastics (Huysveld et al., 2019), plastic material mixtures like personal protective equipment (Zhao et al., 2022b), and plastic debris such as waste residues (Larrain et al., 2020) to high yields of renewable fuels and monomers. This chemical recycling process can also co-convert plastics with other wastes like biomass to produce hydrocarbon fuels (Beydoun and Klankermayer, 2020). The most common plastic open-loop recycling method involves thermal cracking processes, such as pyrolysis (Jahirul et al., 2022), gasification (Dai et al., 2022), and hydrothermal processes (Dogu et al., 2021), which help break the carbon bonds within polymer molecules. Currently, pyrolysis operated under anaerobic conditions is the most commercially and technologically feasible method, with low GHG emissions and high yields of alternative fuels and monomers (Alston and Arnold, 2011). Other thermal-cracking processes, such as hydrothermal liquefication and carbonization, are now only operated at a lab scale and require intensive water use, high capital costs, and labor intensity for maintaining high-pressure operating conditions (Dogu et al., 2021). A recent LCA study on hydrothermal treatments (HydroPRS™ technology) located in the UK has illustrated technological feasibility in piloting these hydrothermal processes to maintain environmental sustainability (Ozoemena and Coles, 2023b). Besides macro plastic debris, open-loop recycling, of which conversion rate is not hampered by chemical inpurities (Nabgan et al., 2022), can also convert microplastics into renewable fuels. However, the offsite input chemical production and energy generation required to maintain effective waste conversion and downstream product separation can pose full-spectrum environmental burdens and offset the sustainability benefits of onsite downstream product manufacturing. Hence, weighing the impact of chemical recycling across three sustainability pillars is critical.

2.6. Upcycling of plastic waste and macro- and microplastic debris

Both open-loop and closed-loop recycling enable the effective conversion of plastic waste to various downstream products but also require intensive input chemical and energy use. The low selectivity, yield, and conversion rate of value-added downstream products limit their economic profitability and may not offset the high capital expenditures and operating expenses (Solis and Silveira, 2020). Chemical upcycling technologies, which enhance economic profitability by giving solid waste a second life with additional value, have attracted wide research attention since 1999 (Jehanno et al., 2022). Research efforts involve enhancing the conversion and selectivity of value-added products by advancing catalyst design (Zhang et al., 2022a), upcycling methods, and operating conditions (Rorrer et al., 2020; Tennakoon et al., 2020; Zhuo and Levendis, 2014), and manipulating the polymer molecules to retain effective conversion by design (Abel et al., 2021; Westhues et al., 2018). For polyolefin plastics, thermal conversion-based upcycling rearranges the carbon bonds under high-temperature conditions to produce value-added hydrocarbons with high selectivity aided by advanced catalysts (Liu et al., 2021a). This plastic chemical recycling process also provides opportunities to implement green chemistry methodology, which aims to manufacture products from solid waste, such as manufacturing basic value-added chemicals via electrocatalysis (Zhou et al., 2021), bioconversion (Kulas et al., 2021), and solvolysis (Singh et al., 2021). Current investigations on microplastic collection lie in advancing microplastic capture processes from air, water, and soil, as well as waste- and drinking water treatment that homogeneously removes particles from water. Theoretically, as plastic particulates share similar chemical properties with macro wastes, upcycling is also technologically feasible to create values from microplastics via electrocatalysis (Liu et al., 2022a; Ma et al., 2022), photocatalysis (Qin et al., 2022), and steam reforming to produce renewable fuels (Nabgan et al., 2022) under mild operating conditions, which can aid in future scale-ups. Recycling of these collected microplastics is still in the initial stage (Pan et al., 2023). These research efforts help reduce the sustainability risks from offsite chemical and energy generation whilst enhancing economic benefits from downstream products. As plastic waste upcycling is economically attractive for scale-ups (Hou et al., 2021), evaluating the three pillars of sustainability of this process is critical to help inform the technological innovations needed to maintain good practicability with enhanced economic benefits.

2.7. Cascaded use of plastic waste and macro- and microplastic debris

Besides alternate fuels or energy production, repurposing plastic wastes for multiple uses can also extend resource availability while minimizing waste generation and environmental exposure. The “plastic cascade use”, where plastic wastes are reused by many times in various alternative applications, such as concretes (Almohana et al., 2022), is growing for plastic recycling purposes (Zhao and You, 2023). This process also enables the co-treatment of solid wastes of other kinds, such as biomass, to reduce economic costs and enhance waste conversion per treatment cycle while mitigating plastic pollution (Beydoun and Klankermayer, 2020). Existing plastic cascade use studies also investigate the tandem conversion of bulk wastes alone (Wang et al., 2022) or along with intermediate products to enhance the economic values of downstream products and usability of plastic wastes over multiple cycles (Xu et al., 2022), which help curb plastic waste generation and material loss in a single-life cycle. Since the chemical impurities in the plastic waste mixture do not impact its alternative use other than plastics, cascading use of the microplastic alone or along with macro polymeric debris can be technologically viable in mitigating sustainability risks from mismanaged solid wastes. Identifying the pros and cons of these tandem processes requires cascading the life cycles of micro- and macroplastics, but the relevant investigation remains a knowledge gap due to the few understandings of material losses and their subsequent chemical emission estimates along the entire plastic life cycle.

2.8. Other emerging microplastic recycling solutions

The material losses from untreated wastes can generate microplastics in all earth compartments and pose full-spectrum environmental burdens (Luan et al., 2022). Unlike macro debris with similar chemical properties, microplastics are hard to collect manually due to their miniature size, thus resulting in tons of suspended particles exposed to the earth (Hong et al., 2017). Removing these microplastics is critical as they are being detected in drinking water and are illustrated as a threat to public health and ecosystems (Yan et al., 2021). Most aqueous microplastics come from freshwater sources and are treated in waste/-drinking water treatment plants with easy operations. These plastic particulates are often separated and processed with wastewater treatment sludge in incineration or open burning processes, posing GHG emissions and air pollution that worsen public health and social risks (Zhao and You, 2021c). Plastic waste chemical recycling, which can effectively repurpose solid wastes into value-added chemical products, can also sustainably treat complex plastic compounds within microplastics and reduce their environmental exposure (Bacha et al., 2021). Previous lab-scale studies proved the technical feasibility of the chemical conversion of microplastics to renewable fuels to power plastic debris collection processes (Nabgan et al., 2022), reducing the environmental pollution posed by microplastics and saving fossil fuels. The good chemical properties, including reactivity induced by the high surface area (Chen et al., 2022), can promote microplastic conversion via biodegradation and mitigate pollution. In practice, the ideal scenario for minimizing environmental exposure is capturing microplastics and converting them into essential basic chemicals for plastic remanufacturing (Othman et al., 2021). However, the economic and environmental benefits of this onsite chemical production may not offset the energy and cost of collecting sufficient microplastics for conversion at a pilot scale (Gallo et al., 2018). A cascade life cycle, including a tandem of macroplastic recycling and microplastic removal processes, can help assess the pros and cons of microplastic removal processes to help inform their future technical innovations toward maintaining environmental, economic, and social sustainability. Recent studies have illustrated the environmental benefits of waste PP cascaded use in reducing fossil fuel use and plastic losses, but this process also requires scale-ups to reduce the capital cost per mass plastic waste treated (Zhao and You, 2023).

2.9. Decarbonization in plastic waste and debris chemical recycling

Over 3.4% of global GHG emissions come from plastic material production and conversion and are set to double by 2060 given the expanding use of plastics (OECD, 2024). For the plastic EoL treatment, chemical recycling processes are also energy and carbon-intensive because of the high-temperature conditions maintained for cracking plastic wastes with complex chemical compositions and offsite production of input chemicals (Khoo, 2019). Therefore, the plastic material life cycle requires implementing decarbonization technologies, including carbon capture and utilization (CCU) and sequestration (CCS), under the climate commitment (Bauer et al., 2022). Current investigations focused on converting the captured CO_2 from direct air capture to basic chemicals for plastic production, including methanol (M and E, 2016), ethanol (Harris et al., 2021), syngas (Bachmann et al., 2023), and formic acid (M and E, 2016). Biogenic carbon emissions could be reduced by cultivating and producing biomass, including wood (Saraeian et al., 2020), bark (Hu, 2015), miscanthus (Götz et al., 2022), and sugar and oil crops (Ramirez and Rainey, 2019), which are used for producing ethylene and propylene as bio-based plastic monomers by thermal processing, such as fast pyrolysis (Hu, 2015) and gasification (Ahlström et al., 2019). Utilizing biomass for basic chemical production can facilitate the carbon capture by plants and replace the fossil counterparts, both of which can reduce carbon emissions by 20% to negative, as given in Table 1. Direct onsite conversion of carbon dioxide to basic chemicals via the aforementioned CCU processes can also reduce up to 56% of plastics' life cycle carbon emissions (Table 1). Direct air capture enables a 25% reduction in life cycle carbon emissions, which are largely driven by the local energy source mix and carbon capture efficiency. However, a trade-off exists between the carbon emissions and total economic costs, given the relatively high capital investments of thermal processing equipment of biomass conversion and product separation.

3. Sustainable plastic waste and macro- and microplastic debris chemical recycling: insights and challenges

3.1. Policy and technological insights

Technological and policy insights are informed in existing plastic waste chemical recycling studies to secure effective conversion of wastes and their derived debris from macro- to microscale to maintain environmental, economic, and social sustainability (Villarrubia-Gómez et al., 2022). The technological insights are categorized by the feedstock development, product exploration, process innovation, sustainability analysis methodology improvement, and product value chain enhancement listed as follows (Zhang et al., 2022b):

- **Feedstock development:** Advance the input material wastes by design based on the lab-scale investigation to pursue effective conversion with sustainability benefits.
- **Product exploration:** Manufacture new products from plastic wastes guided by theoretical design for more applicable and versatile use with low sustainability burdens.
- **Process innovation:** Implement a novel or improve process design to enhance the purity and yield of downstream products with better overall sustainability performances.
- **Sustainability analysis methodology advancement:** Advance the sustainability assessment methodology for a comprehensive, precise, and holistic analysis of chemical recycling processes.
- **Product value chain enhancement:** Promote the supply and use of downstream products to pursue sustainability throughout supply chains.

These technological insights towards three pillars of sustainability cover the life cycle stages of wastes encompassing: 1) Feedstock design and production; 2) Upstream processing: Collection and sorting; 3) Downstream processing: Conversion and product separation; 4) Product design, trade, and use; 5) Product EoL fate (Mitrano et al., 2021). Wise feedstock design and selection can help avoid tackling complex mixtures, reallocate environmental pollutants to upstream conversion, and reduce sustainability burdens in downstream processes compared to conventional plastic EoL management. By manufacturing more practical, value-added, and recyclable products from waste, the sustainability performance of chemical recycling processes can be further improved according to product development investigations. Tuning the EoL fate of plastic wastes/material losses referring to the associated technological innovations can also help relieve the sustainability burdens from untreated or mismanaged plastics within their product life cycle. Detailed technological challenges by categories are listed in Table 2.

Implementing these technology innovations in chemical recycling processes should receive government support concerning technology, economic, environment, and social (public) oriented policies (Gontard et al., 2022). Technology-oriented policies aim to incentivize the existing or technologically promising chemical recycling processes to convert macro and micro debris effectively into value-added products with high yields. Reinforcing the sell, supply, distribution, and effective use of those downstream products under existing environmental interventions can facilitate plastic waste chemical recycling under economic support by referencing relative economic-oriented policies. Relevant environmental policies aim to balance the technological performances of chemical recycling, including conversion rates and product yield, with potential environmental hazards/benefits of implementation under existing sustainability interventions. Timely planning and execution of these interventions, in line with social-oriented policies, is urgently needed to maintain the three pillars of sustainability of chemical recycling. In these contexts, stakeholders within the value chains of plastic waste and its downstream products can benefit from these policy implications associated with 1) Industrial entities that cover waste recyclers and product manufacturers, suppliers, and sellers; 2) Governmental and non-governmental organizations; 3) Product customers. Table S1 in the Supporting Information shows the detailed classifications of these policies.

3.2. Research challenges and knowledge gaps

Efforts to mitigate plastic pollution through chemical recycling, shown in Table 2, focus on reducing plastic waste generation from Anthropocene activities and reallocating wastes away from natural environments to downstream value-added products manufacturing that

Table 1
Decarbonization practices utilized in plastic material life cycle and their relative reduction on carbon footprints.

References	Decarbonization Practices	Relative Carbon Emissions Reduction vs. Conventional Pathways
Kaiser and Bringezu (2020)	Solvent-based (Monoethanolamine) absorption	−25%
Thonemann et al. (2022)	Carbonation and carbon mineralization	[−74%, −15%]
Mufarrij et al. (2023)	Tri-reforming process	[−94%, −62%]
Walker and Rothman (2020)	Biomass production for plastic manufacturing	[−100%, −20%]
Benavides et al. (2020)	Biomass production and thermal processing for plastic manufacturing	[−140%, −20%]
Van Roijen and Miller (2024)	Biomass production for plastic manufacturing: Fermentation coupled with thermal processing	[−140%, −20%]
Tsiropoulos et al. (2015)	Biomass production and thermal processing for plastic manufacturing	[−140%, −20%]
Chu et al. (2022a)	Decarbonizing plastic production (no specific technology indicated)	Up to Reduce 74.9%
M and E (2016)	Carbon dioxide conversion to basic chemicals, including syngas, formic acid, ethylene, and methanol	[−55%, −34%]
Harris et al. (2021)	Direct air capture coupled with basic chemical production from biomass	[−56%, −31%]
Bachmann et al. (2023)	Reverse water-gas shift process	[−49%, −5%]
Saraeian et al. (2020)	Biomass production for plastic manufacturing	[−140%, −20%]
Götz et al. (2022)	Thermal valorization of biomass for plastic manufacturing	[−100%, −20%]

Table 2
Technical challenges associated with product and feedstock development, process innovations, and evaluation methods to pinpoint effective plastic pollution mitigation practices, including chemical recycling and upcycling processes.

Categories	Technological Insights	References
1. Product exploration	1.1. Develop products guided by holistic lab investigation.	(Simões et al., 2013; Zamani et al., 2015)
	1.2. Replace natural materials with recycled ones with similar or improved properties.	(Bensadoun et al., 2016; Bortolini et al., 2018; Civancik-Uslu et al., 2021; Haylock and Rosentrater, 2018; Ingrao et al., 2014; Kayili and Çelebi, 2021; Singh et al., 2021; Yao et al., 2022)
	1.3. Minimize the product life cycle environmental impacts and production costs.	(Muiruri et al., 2022; Özdemir and Önder, 2020; Tamburini et al., 2021; Yuan et al., 2022)
	1.4. Produce renewable fuels replacing fossil alternatives.	(Pacheco-López et al., 2021; Tomic et al., 2022)
	1.5. Substitute original materials with re-granulates and flakes.	Civancik-Uslu et al. (2021)
	1.6. Use biomass or industrial wastes in waste processing.	(Manjunatha et al., 2022; Zibunas et al., 2022)
	1.7. Design sustainable products specific to regions.	Venkatachalam et al. (2022)
2. Feedstock development	2.1. Use bio-based plastics with valid EoL treatment.	(Karayılan et al., 2021; Zheng and Suh, 2019)
	2.2. Innovate plastic fillers for effective recycling.	Al-Majali et al. (2019)
	2.3. Pyrolyze discarded plastics with other wastes.	Lai et al. (2021)
	2.4. Reduce the complexity of plastic packaging.	(Civancik-Uslu et al., 2021; Pauer et al., 2020)
	2.5. Secure a consistent feedstock, e.g. plastic waste supply.	Singh et al. (2021)
	2.6. Use sustainable energy/feedstocks specific to regions.	Zibunas et al. (2022)
	2.7. Use industrial by-products for public goods production.	Zibunas et al. (2022)
3. Process innovation	3.1. Improve sustainability and process performances	
	3.1.1. Reduce carbon emissions with energy savings.	(Dastjerdi et al., 2021; Demarteau et al., 2022; Jeswani et al., 2021; Karayılan et al., 2021; Lawler et al., 2012; Meys et al., 2020, 2021; Park and Gupta, 2015; Uekert et al., 2022; Volk et al., 2021; Wang et al., 2019; Zheng and Suh, 2019)
	3.1.2. Enhance the resource or material circularity (cascade use).	(Hongshen and Ming, 2013; Huysman et al., 2015; Karayılan et al., 2021; Klemeš et al., 2021; Passarini et al., 2012; Singh et al., 2021; Tamburini et al., 2021; Thakker and Bakshi, 2021)
	3.1.3. Mitigate (aqueous) plastic pollution.	(Bora et al., 2020; Cappello et al., 2022)
	3.1.4. Improve socio-economic performances.	(D'Adamo et al., 2020; Demarteau et al., 2022; Klemeš et al., 2021; Meys et al., 2020; Meys et al., 2021; Park and Gupta, 2015; Vadoudi et al., 2022; Wang et al., 2019; Zhao et al., 2022b)
	3.1.5. Enable treating complex/contaminated plastics.	(Carvalho et al., 2018; Guran, 2019; Singh et al., 2021; Teuber et al., 2016; Uekert et al., 2022; Vadoudi et al., 2022; Venkatachalam et al., 2022)
	3.2. Process operation	
	3.2.1. Adjust the feedstock composition.	(Al-Salem et al., 2014; Bhogayata and Arora, 2019)
	3.2.2. Increase the recycling capacity.	Al-Salem et al. (2014)
	3.2.3. Enhance the labor quality and ergonomics.	(Bortolini et al., 2018; Cudjoe and Wang, 2022; Huysveld et al., 2022; Vitale et al., 2018)
	3.2.4. Improve the schedule and worker safety.	Yao et al. (2022)
	3.3. General technology innovations	
	3.3.1. Adopt proper technology and equipment selection.	(del Oso et al., 2021; Khalil, 2019; Simões et al., 2013; Veksha et al., 2022; Zhao and You, 2021b, c)
	3.3.2. Enhance the material recovery facility capacity for handling hard plastics.	Faraca et al. (2019)
	3.3.3. Replace incineration and landfills with chemical recycling.	(Cardamone et al., 2022; Civancik-Uslu et al., 2021; Mastellone, 2020; Zhao et al., 2022b)
	3.3.4. Specify the treatment technology by waste.	Schwarz et al. (2021)
	3.3.5. Advance the feedstock or wax recovery.	(Cappello et al., 2022; Schwarz et al., 2021)
	3.3.6. Implement multiple recycling in practice.	Singh et al. (2021)
	3.3.7. Adopt chemical recycling technology globally.	Guran (2019)
	3.4. Plastic conversion innovations	
	3.4.1. Advance the plastic sorting system nationwide.	(Horodytska et al., 2020; Pauer et al., 2020; Sommerhuber et al., 2017; Teuber et al., 2016; Uekert et al., 2022)
	3.4.2. Enhance the efficiency via optimization.	Pillain et al. (2019)
	3.4.3. Promote cross-sectoral waste valorization.	Karayılan et al. (2021)
	3.4.4. Treat thermoset polymers via dissolution or depolymerization.	Schwarz et al. (2021)
	3.4.5. Treat polyolefins or debris via thermal cracking.	(Al-Majali et al., 2019; Tomic et al., 2022; Volk et al., 2021)
	3.4.6. Treat low-grade plastics via pyrolysis or co-incineration.	(Hossain et al., 2021; Joshi et al., 2021; Neha et al., 2022; Tomic et al., 2022; Veksha et al., 2022; Yousef et al., 2022)
	3.4.7. Use surfactant treatment for processing low-grade polyhydroxyalkanoates.	del Oso et al. (2021)
	3.4.8. Improve pyrolysis efficiency via catalyst design.	Yousef et al. (2021)
	3.4.9. Adopt solvolysis in treating complex plastic mixtures.	Vadoudi et al. (2022)
	3.4.10. Treat complex plastics by tandem vapor-phase hydrotreatment process.	Wang et al. (2022)
	3.5. Downstream processes innovations	

(continued on next page)

Table 2 (*continued*)

Categories	Technological Insights	References
	3.5.1. Use solvents in solvolysis for effective separation.	(del Oso et al., 2021; Khalil, 2019; Liu et al., 2022b,c)
	3.5.2. Include CCU/CCS for reducing carbon emissions.	Meys et al. (2021)
	3.5.3. Use biomass technologies considering local renewable electricity mixes and biomass availabilities for carbon reduction.	Bientinesi and Petarca (2009)
	3.5.4. Advance or use alternative separation processes.	Singh et al. (2021)
	3.5.5. Reduce energy use of solvent regeneration.	Liu et al. (2022b,c)
	3.5.6. Pyrolysis crude requires further processing.	Reznichenko and Harlin (2022)
4. Sustainability analysis methodology advancement	4.1. Evaluate the life cycle sustainability for decision-making based on transparent information.	(Demarteau et al., 2022; Guran, 2019; Lazarevic et al., 2010; Passarini et al., 2012; Stegmann et al., 2022; Vitale et al., 2018)
	4.2. Optimize environmental hotspots of specific process designs.	(Gear et al., 2018; Rochat et al., 2013)
	4.3. Evaluate the circular economy benefits.	(Guran, 2019; Klemeš et al., 2021; Muiruri et al., 2022; Pillain et al., 2019; Schwarz et al., 2021; Simões et al., 2014; Thakker and Bakshi, 2021)
	4.4. Evaluate the sustainability of waste trade.	Ma et al. (2020)
	4.5. Systematically overview and determine the technologies.	Jeswani et al. (2021)
	4.6. Extend the system boundary by including product use, trade, and market.	(Pacheco-López et al., 2021; Schwarz et al., 2021; Thakker and Bakshi, 2021; Yao et al., 2022; Zhao and You, 2021a)
	4.7. Incorporate biomass pathways and ecosystem management in evaluation.	Pacheco-López et al. (2021)
	4.8. Use the case/region-specific data and uncertainties.	(Hossain et al., 2021; Singh et al., 2021; Stegmann et al., 2022)
	4.9. Consider technology performances and waste purity in LCA models.	Schwarz et al. (2021)
	4.10. Use prospective LCA for future technology evaluation.	Schwarz et al. (2021)
	4.11. Evaluate the entire plastic life cycle, including microplastics.	(La Rosa et al., 2021; Liu et al., 2022b,c; Meys et al., 2021)
	4.12. Use multi-life cycle assessment for products with multiple EoL use.	Liu et al. (2022b,c)
	4.13. Propose and use a comprehensive plastic material selection framework.	Venkatachalam et al. (2022)
	4.14. Develop and use the social life cycle assessment framework.	Venkatachalam et al. (2022)
5. Product value chain enhancement	5.1. Optimize the packaging value chain with a circular economy approach.	(Ardolino et al., 2021; Karayılan et al., 2021; Thakker and Bakshi, 2021)
	5.2. Reduce waste exportation with improper EoL treatment.	Ardolino et al. (2021)
	5.3. Reduce capital cost or enhance income from products.	Zhao et al. (2022b)

replaces offsite manufacturing and maintains three pillars of sustainability. Measures such as redesigning the plastic materials for easy 3Rs (reuse, repurposing, and recycling), promoting the downstream product market and trade, secondary product use for facilitating plastic production, and reducing carbon emissions by using renewables can improve the sustainability of chemical recycling endorsed by necessary policies associated with technology, economic, and environmental policies in Table S1 in the Supporting Information. Effectively adopting these measures requires inter- and multisectoral cooperation among technology, environmental, economic, and social practitioners, facilitated by information exchange based on an improved public understanding of the extent of plastic pollution and its environmental aftermaths within short and long terms, as shown in Table S1 in the Supporting Information.

Understanding the extent of plastic pollution, the consequences from human activities, and the efforts needed for full mitigation is an existing research challenge with growing research and policy attention (Karbalaei et al., 2018). The total amount of untreated plastics and free material debris in the natural environments, their specific environmental impacts, and the social and economic efforts required to alleviate these environmental consequences govern the extent and consequences of plastic pollution (Borrelle et al., 2020). Thorough removal of these plastic wastes from natural environments by technological measures is the ultimate solution for pollution mitigation. However, exploring the sustainable material life cycle of plastics with minimum waste generation, free debris exposure, and carbon emissions remains an unmet research need. Since plastic pollution is a global environmental issue, adopting technological measures to combat it necessitates intersectoral information exchange (Lee et al., 2021b), collaboration, and coordination among environmental, economic, and social decision-makers under the global treaty (Johansen et al., 2022). Assessing the sustainability performance of chemical recycling in managing the cumulative domestic plastic waste can indicate the inter- and multisectoral efforts needed among environmental, economic, and social practitioners. These implications can guide the achievement of a global zero-plastic pollution treaty by promoting economic performance and positive societal impacts through enhancing plastic circularity (Zhao and You, 2023). Explicit investigations are still lacking in the necessary local and global efforts to deploy plastic production, use, and waste management as per relevant studies on sustainable plastic life cycle concepts. This knowledge gap originates from the absence of understanding of the total amount of plastic waste and microplastics released to natural environments, technology transitions from conventional EoL to zero-waste measures, and global practices on removing accumulative untreated plastics on earth. The following three sections present detailed background information, research questions, and research directions to initiate future thinking on filling in these three knowledge gaps.

4. Green solutions for reducing plastic pollution and exposure by human uptake

Plastics, due to their diverse properties and applications, are integral to various human activities, including food packaging, building materials, and industrial manufacturing (Agarwal and Gupta, 2018; Psychology). If not managed properly at the EoL (Suzuki et al., 2022), these polymers can generate microplastics within 5 mm in size. Microplastics can easily enter the food chain through ingestion by organisms and contaminate foodstuffs or drinking water (Galloway et al., 2017). Once ingested by humans, these particles in vivo may cause organ inflammation, carcinogenic effects, and digestive problems due to toxic plasticizers absorbed on microplastics (Amato-Lourenço et al., 2020). The problem could worsen with the growing consumption of plastic materials and inadequate waste management. Targeting the source of plastic waste generation can help inform effective measures to curb microplastic generation, but identifying the exact origins of these micro particulates remains an unmet research need (see Fig. 4).

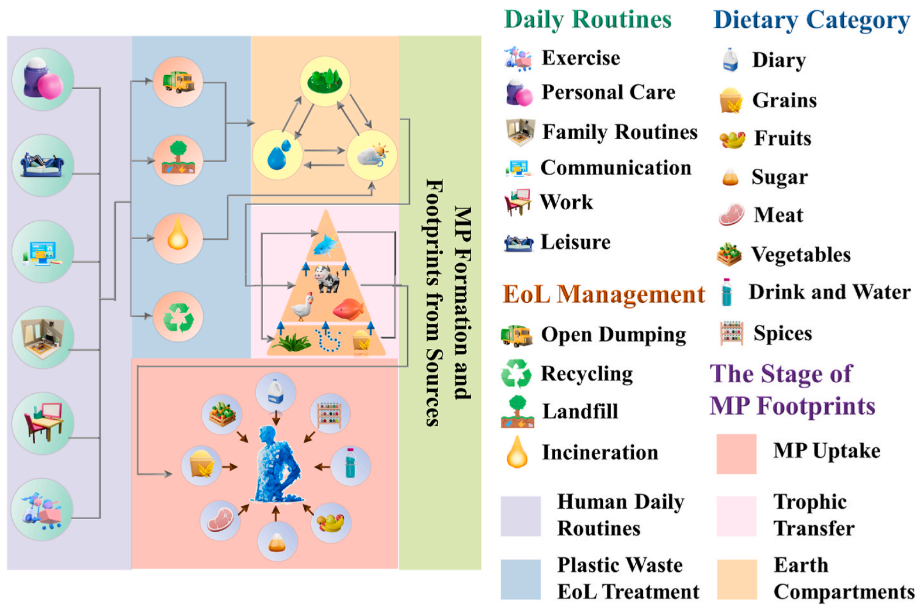

Fig. 4. Overview of the sources, distribution, and pathways of microplastic ingestion by humans. Plastic waste from daily human routines can generate microplastics from solid waste EoL management processes. Those microplastics then undergo compartment transmission and trophic transfer to be exposed to foodstuffs ingested by humans.

First, how do human activities generate macro- and microplastic debris? Daily routines consuming plastics, the type of plastic consumed, and the consumption amount can influence plastic debris generation. Daily routines reflect the general human activities occurring within a specific socio-economic context (Miró, 2014). Identifying how plastic serves different functions in daily routines, such as PET for food packaging or nylons for fabricates, can help assess overall plastic consumption (Periyasamy and Tehrani-Bagha, 2022). Plastic material production and use can generate material losses from abrasion and natural weathering, including the microfibers originating from fabrication and laundry wastewater effluents (Lahiri et al., 2023). All used plastic materials are ultimately treated by the EoL practices, generating microplastics from mismanaged wastes or being recycled for the second use (De Almeida and Borsato, 2019). In this context, the overall waste and material debris generated from human activities is assessed by aggregating the material losses information by plastic type at the domestic production, use, and EoL stages (Luan et al., 2022).

Second, how does free plastic debris generate particles? Plastic particles originate from material losses from production, use, and uncontrolled waste release from landfills and open dumping (Wen et al., 2021a), or fly ash from incineration (Shen et al., 2021). Compartment transport and degradation govern plastic particle distribution, release into natural environments, and human exposure (Li et al., 2020), as indicated by multimedia transport models, including the Simplebox4nano model and NanoDUFLOW model (Besseling et al., 2017; Meesters et al., 2014). Shapes and sizes of plastic debris alter the transmission rates and compartment distributions (Wang et al., 2021), of which variations can be quantified based on the existing multimedia transport model supported by real-time monitoring data (Nizzetto et al., 2022). Natural degradation aided by photo- and biodegradation, including natural and hydraulic weathering, can generate volatile organic chemicals (VOCs) along with particle transport and pollute the air (Chamas et al., 2020). Real-time simulation of the natural degradation of plastic waste (Chamas et al., 2020) can provide accurate degradation profiles. Coupled with the microplastic exposure by organisms along the food chains (Athey et al., 2020), this degradation profile information enables a holistic and precise estimation of the impact of plastic debris on natural environments and public health.

Third, how much plastic particulates can be ingested by people, and what is the removal cost? Microplastic ingestion by organisms and subsequent human consumption is influenced by factors such as microplastic concentration in prey, predatory behavior, and excretion rate (Athey et al., 2020; Sun et al., 2022). Measuring the microplastic accumulation in organisms along the food chain and their specific nutrient transfer rates helps assess the microplastic concentration in foodstuffs belonging to each trophic hierarchy (Sun et al., 2020). The microplastic exposure level in these foodstuffs and intake based on domestic dietary habits (Parfitt et al., 2010) dictate the plastic particulate uptake. Tracing microplastics from food intake back to human activities can help identify their exact origins and inform how and where to take effective mitigation measures. Assessing the economic, environmental, and social costs of innovation and adoption of plastic debris clean-up and repurposing processes, such as water quality control measures for industrial and residential effluents, informs the necessary government incentives and public support to reduce the impact of ingested microplastics.

A clear and concise visualization of microplastic exposure and its origins helps identify the human activity with the most prevalent plastic release and inform region-specific policies to reduce particle ingestion and potential health risks (Mofijur et al., 2021) aligned with SDG 3 (Good Health and Well-being). Particular attention should be paid to monitoring the quality of foods with high microplastic contents as they escalate with age before effective waste management measures are taken for pollution mitigation. A real-time dashboard showing region-level microplastic footprints and derived disease risks globally enables advancing public awareness of the health burdens caused by plastic pollution. A holistic understanding of mitigation measures' timing, implementation, and costs indicates the efforts required to minimize plastic footprints and negative impacts on public health, but the relevant investigation remains a knowledge gap.

5. Sustainably addressing macro- and microplastic debt

Minimizing plastic footprints requires adjusting consumption habits and curbing waste generation from material production, use, and EoL waste management. Striving for a "waste-free" plastic life cycle through the 3Rs—reuse, repurpose, and recycle—can maintain and enhance plastic material circularity (Marino and Pariso, 2020). Chemical recycling and upcycling processes enable efficient plastic waste conversion to downstream products with versatile applications, facilitating cost-effective and low-emission material circularity (Pichler et al., 2021). While some thermal-based processes are scaling up (Hogue, 2022), most chemical recycling processes remain at the lab- or pilot scales due to insufficient government incentives (Garcia and Robertson, 2017). Before these processes are fully adopted, portions of plastic waste still end up in landfills and incineration plants and pollute the planet. The term "accumulative plastic debt", which imitates the "plastic toxicity debt" defined by Rillig et al. (2021), reflects the quantity of untreated harmful plastic waste accumulating with age. Additionally, plastic waste treatment contributes to climate change by sharing 3.4% of total GHG emissions from Anthropocene activities (OECD, 2023). Decarbonizing all plastic life cycle stages is crucial to curb the rise in global temperature under the climate commitment. CCU and CCS processes, which aim to capture the carbon dioxide for storage or conversion to applicable chemicals, show potential for reducing carbon emissions in chemical recycling (Meys et al., 2021) aligned with SDG 13. Adopting this zero-waste, carbon-neutral strategies is essential to mitigating plastic pollution and climate change, which requires answering the following three research questions.

First, what are the zero-waste and zero-carbon technologies? Zero-waste pathways aim to minimize waste accumulation by reducing waste generation and enhancing treatment capacity. Implementing the 3Rs alongside chemical recycling and upcycling can reallocate waste to secondary uses and cut generation (Jehanno et al., 2022). An ideal approach involves 3R various types of wastes in tandem to reduce their generation, inspired by the cascaded conversion of plastics with solid biomass wastes (Beydoun and Klankermayer, 2020), which further minimizes material use, losses, and derived environmental and social burdens. Using alternative materials like biodegradable plastics to substitute fossil-based counterparts can further reduce material losses over usage and alleviate environmental and social burdens from EoL waste management (Sangroniz et al., 2019). However, these eco-friendly materials still cause pollution if mismanaged in open composting or casual discarding. Cascaded processing of plastic waste and alternative materials enables the manufacturing of useable products and reduces waste generation per material usage (see Fig. 5).

Extracting resources from waste, such as capturing plastic debris from natural environments and converting it into useful materials, can also impede generation and pollution. Advanced plastic particle capture approaches, which leverage particles' surface properties, can assist in microplastic removal by using liquid crystals and bioreactors (Bang et al.; Mukherjee et al., 2023). Synthesis of these materials is energy intensive and premature in scale-ups (Wang et al., 2014). Recent LCA studies have explored these processes' energy and material consumption and predicted future scenarios (Wang et al., 2014) based on the foreground data (Arvidsson et al., 2018). Thermal degradation processes like pyrolysis and gasification can effectively treat captured plastic particles (Nabgan et al., 2022). Cascading macro- and micro-debris processing minimizes plastic particle release, although current technologies may require high energy and input chemicals for effective conversion (Zhao and You, 2023).

High GHG emissions from intensive energy and input chemical consumption can be reduced by incorporating the CCU or CCS processes, which repurpose the carbon emissions from plastic processing into useable chemicals. Producing monomers or plastic materials from carbon dioxide through CCU or biomass from bioenergy with carbon capture and storage (BECCS) simultaneously mitigates GHG emissions and waste pollution (de Oliveira et al., 2021). Co- or cascaded conversion of plastic wastes with carbon dioxide or biomass from BECCS to produce monomers is technologically feasible and can further reduce environmental burdens (Esso et al., 2022; Lee et al., 2022b). However, scaling up these carbon-neutral, zero-waste practices to fully address plastic pollution and climate change still requires time.

Second, when and how should zero-waste combined with

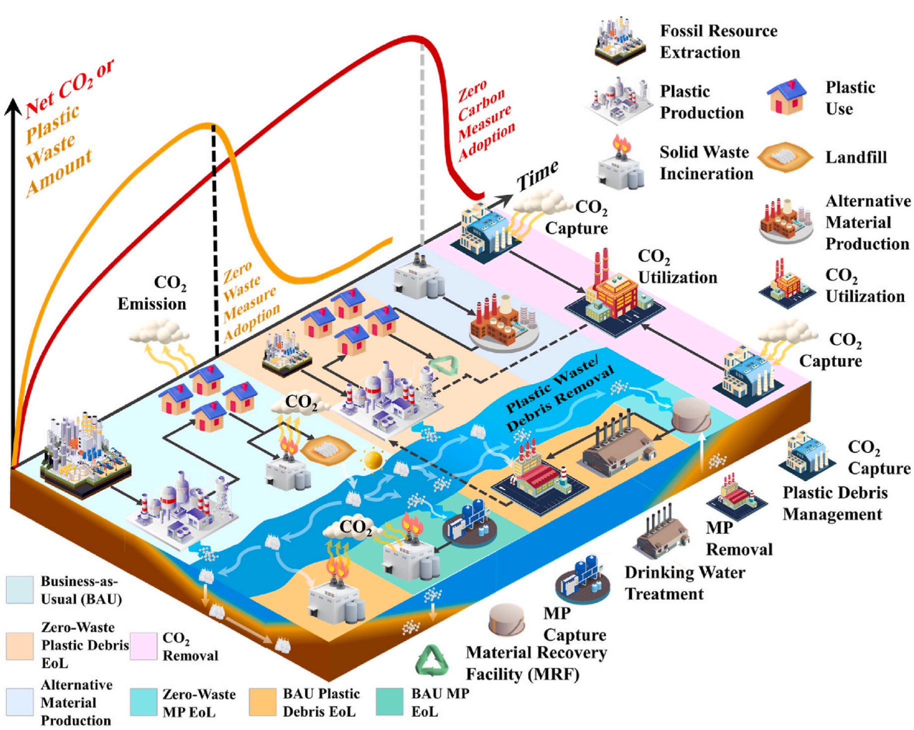

Fig. 5. Overview of the plastic life cycle in business-as-usual (BAU) and zero-waste scenarios under climate commitment. In the BAU scenario, plastic waste is treated in chemical recycling, incineration, and landfills. The polymeric debris generated from mismanaged wastes can undergo photodegradation to form macro- and microplastic debris, both of which are treated by incineration. Plastic wastes in the zero-waste scenario are fully recycled, while the accumulated waste debris in the natural environment is collected and treated by chemical recycling processes for material recovery. The carbon dioxide capture and utilization processes are also incorporated in the zero-waste plastic treatment practices to meet climate commitment by cutting carbon emissions.

carbon-neutral technologies start full adoption? Even through the zero-waste movement began 20 years ago (Seldman, 2016), a large portion of U.S. plastics still end up in landfills (Wen et al., 2021a), posing pollution from mismanaged wastes. Technological, economic, and societal hurdles challenge the adoption of zero-waste practices (Pietzsch et al., 2017). With sufficient government incentives, many lab-scale plastic waste upcycling processes can progress alongside technology improvement and integration of carbon-neutral practices in EoL waste management. The time required for these technological transitions to practical adoption is estimated by associated technology learning curves (Hundertmark et al., 2018). While adopting CCS (excluding BECCS) technology faces controversy in the site selection (Stephens and Jiusto, 2010), relevant policies (Management, U.S.D.o.t.I.B.o.L., 2022) are under development to guide CCS project operation on public lands. By contrast, CCU technology adoption is not site-selective and can be retrofitted from existing chemical plants, such as incineration facilities (Christensen and Bisinella, 2021), to capture GHGs from the flue gas.

Technology transitions from conventional plastic waste management to advanced processing practices necessitate transparent and efficient information sharing among scientific, environmental, public health, and technology stakeholders (Tanveer et al., 2022). This collaboration can hasten the development of supportive initiatives for technology diffusion before full adoption (Simeone, 2018). The longevity of supportive policies on technological innovations determines the right time to initiate technology adoption, with the balance of economic incentives and costs dictating the duration. Furthermore, intersectoral education and training, supported by the United Nations Environmental Programme, help expedite the diffusion of technology (Programme, 2023).

Third, when and how to reach a plastic debt-free future under the climate commitment? The reduction of accumulated plastic waste, or “plastic debt”, occurs when plastic debris removal processes adequately manage waste generation. The technology transition pace and the types and amounts of plastic waste treated drive the plastic pollution reduction rate. Socio-economic contexts, which impact people’s daily routines and industrial manufacturing, can affect plastic material consumption (such as packaging) and waste generation (Ribeiro et al., 2019). The pace of technology transition also changes in different socio-economic contexts (Chen et al., 2023), with different government incentives provided. Therefore, efforts to mitigate the cumulative plastic debt vary and require explicit investigation under diverse socio-economic contexts.

High GHG emissions from waste incineration and energy-intensive plastic chemical recycling make the technology transition to zero-waste measures carbon-intensive, offsetting the sustainability benefits of waste mitigation. Decarbonization strategies include repurposing carbon emissions into long-term sinks, such as renewable energy, virgin chemicals, and reusable plastic materials (Langie et al., 2022), which can combat these environmental burdens. The socio-economic contexts drive the adoption and innovation pace of climate-neutral technology. Once carbon-neutral technologies are fully adopted in large-scale zero-waste plastic processing, a decarbonized, plastic debt-free waste management sector can start adoption under climate commitments.

Achieving regional plastic debt-free is a crucial initial step towards global zero waste. Expanding the proposed zero-waste and carbon-neutral strategies to a global scale can help curb worldwide plastic pollution under the climate commitment. However, strategies for eliminating “global plastic debt” remain contentious, particularly as many developing countries weigh appropriate measures’ economic, environmental, and social benefits.

6. Solving global macro- and microplastic debt under circular plastic economy

Mitigating global plastic pollution necessitates regional waste reduction efforts (Cowan and Tiller, 2021). Across different regional economies, the pace of technology innovation and adoption to eliminate local plastic debt varies due to differences in government incentives (Uyarra, 2010). Rapid industrialization in developing countries increases plastic consumption, but the adoption of chemical recycling processes still lags due to insufficient research and development funding in these regions (Walther et al., 2020). Industrialized countries, capable of implementing and innovating zero-waste strategies sooner, often export wastes to less developed economies owing to low economic benefits from processing discarded materials (Liu et al., 2021b). This scenario exacerbates the waste generation in developing countries, exceeding the local EoL treatment capacities and contributing to global plastic pollution. Supportive measures to suppress this incapability of local zero-waste measures and reallocation of untreated waste from technologically immature to advanced regions can address the growing plastic waste generation. Relevant investigations remain a knowledge gap corresponding to the following three key questions:

First, what should be considered to apply effective zero-waste measures under different socio-economic contexts? Plastic debt varies across regions with different quantities of plastic waste generation, EoL management practices employed (incineration, 3R, and landfills), and the duration of technology adoption. Factors like geographical restrictions, which challenge the adoption of CCS, potentially impact the technical feasibility of implementing zero-waste measures. Governmental incentives necessitate the technology transition to zero-waste and carbon-neutral mitigation practices. The duration before full adoption is driven by regional research and development funding for scale-ups and the efficiency of multisectoral information sharing, which helps understand and reconcile the benefits of plastic pollution mitigation. Adequate government incentives throughout technology development and transition can facilitate the adoption of zero-waste measures across diverse socio-economic contexts.

Second, how to reallocate the untreated plastic debt across regions? Ideally, countries executing advanced plastic waste recycling practices should be capable of absorbing excess waste from major plastic consumers. The global plastic waste trade, recommended by the United Nations (UN), could redistribute plastic waste for treatment in other economies to generate economic benefits (Liu et al., 2022b,c). Due to the technical immaturity of creating economic values from waste, most developed countries often export wastes to industrializing countries for cost-effective EoL treatments. This practice can overwhelm the waste management capacities of developing countries and exacerbate global plastic pollution. Reversing this process could lessen the environmental burden but is still hampered by factors including 1. Small-scale plastic waste chemical processing. 2. Low values waste conversion products. 3. Slow progress in zero-waste technology transitions. Government incentives remain crucial for technology scale-up, complemented by economic values generated from upcycling plastic into useable chemicals. Enhanced collaboration and information sharing among stakeholders can expedite transitions to waste-free technology and requires reconciling economic and environmental performances of waste trade, reallocation, and processing (see Fig. 6).

Third, how to reinforce and support **the global treaty in plastic pollution mitigation?** The UN encourages collaboration among social, environmental, and economic sectors to achieve waste-free plastic processing through technology scale-ups, transitions, and adoption (UN, 2022). Necessary steps for maximizing all these sectors’ joint benefits can be identified by assessing the sustainability performances of zero-waste technology innovation, transitions to replace unsustainable practices, and full adoption across the plastic waste value chains. An optimal global plastic waste trade should bolster waste-free environments while maximizing the co-benefits. Redistributing waste from regions with nascent processing technology to those with advanced practices and subsequent trading of downstream products for plastic remanufacturing can help curb plastic pollution while fulfilling the plastic demand in developing economies. Encouraging 3R practices and shifting technology transitions to chemical upcycling processes can enhance the economic profits from downstream products and sustain the

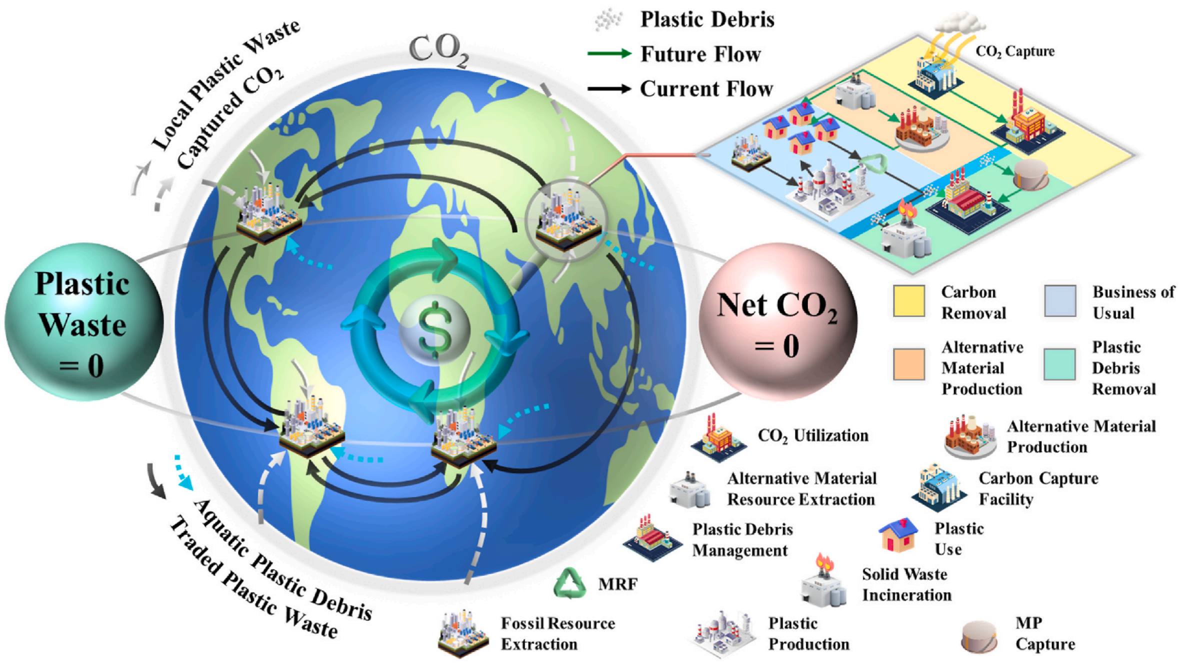

Fig. 6. Overview of the global plastic value chain to free of plastic debt. Innovation in local plastic chemical recycling combined with carbon-neutral technologies is the first crucial step to mitigate regional plastic debt under the climate commitment and circular economy context. Promoting the trade of accumulative plastic wastes from less developed countries to regions with advanced plastic recycling practices can further keep zero waste generation globally.

waste trade. While centralizing zero-waste measures in scale-ups is cost-effective, most plastic waste upcycling technologies remain decentralized due to their low technology readiness in handling diverse real-world plastic waste. As effective downstream product use enhances upstream waste conversion and technology transitions, recovered plastic or other upcycled product applications should be integral to the sustainable plastic waste value chain. Major plastic producers often rely on imports of basic chemicals for plastic manufacturing and alternative material production to relieve plastic waste burdens. Multiregional and multisectoral cooperation aligned with SDG 17 (Partnerships for the Goals) can keep global treaties to curb plastic pollution. Cross-border economic and technological support endorse sustainable waste trade and technology adoption in developing economies, yielding long-term environmental and social benefits through reducing waste and derived particle release.

Negative externalities to the plastic waste trade, such as the domestic waste trade bans in developing countries, can cease excessive imported plastic waste, ending in EoL management (Wen et al., 2021b). However, such bans may not help achieve zero plastic pollution globally if domestic waste generation outstrips management capacity over time. Waste import bans can also impede export to other regions and accumulate untreated plastic waste onsite. A sustainable solution should consider treating waste directly rather than avoiding it, harmonizing zero-waste management practices with trade bans for maximum sustainability. Facilitating smooth technological adoption and transitions to absorb domestic discarded plastics is a prerequisite before importing foreign wastes for onsite management. Retrofitting existing plastic (waste) supply chains with maximum sustainability benefits and introducing versatile alternative materials similar to plastics can ease the EoL waste management burden. Examining these strategies' sustainability performances can guide pragmatic policy-making on plastic waste redistribution practices, aiming to maximize co-benefits while minimizing environmental pollution.

Transparent and effective information sharing across social, economic, and environmental sectors is vital for timely decision-making on pollution mitigation based on comprehensive technology and sustainability information. Real-time updates to an open-source information pool featuring the latest innovative zero-waste measures can advance public understanding and help more accurate decision-making for the public. Tools like ChatGPT can then help deliver succinct information across the life cycle of product manufacturing, use, and waste treatments, assisting in prompt responses to varying plastic waste treatment needs (Zhu et al., 2023).

7. Discussion

Plastics, while beneficial to human activities, also contribute to long-lasting pollution from mismanaged wastes and debris, including macro- and microplastics, posing environmental burdens to water and ecosystem counteracting with SDGs 6, 14, and 15. Existing plastic waste EoL practices utilize incineration and landfills as two conventional and convenient approaches but come with climate burdens and plastic pollution. Replacing these EoLs with effective waste recycling processes can help minimize waste generation and pollution, of which adoption rates vary across countries as they are driven by research and development support tailored to socio-economic contexts. However, the effective adoption of all these processes to sustainably address global plastic pollution lacks the answers to how and when these measures should be taken into practice. Our work addressed these knowledge gaps based on a systematic review of the sustainability performances of existing plastic waste EoLs by technologies and proposing perspective research directions on addressing pollution from source to sink by innovating existing waste management technologies and integrating with decarbonization practices, including CCU and CCS. Adopting effective chemical recycling, which transforms discarded waste and plastic particles into virgin materials to gain both environmental and economic benefits, also yields environmental and social risks from intensive energy and chemical use. Reduction of these embodied sustainability risks demands avoiding excessive plastic waste generation over treatment capacity, technological innovations in recycling to improve plastic recyclability, and integration of decarbonization practices within the material value chain. Plastic manufacturers and consumers should prioritize and be responsible for improving material circularity (SDG 12) to facilitate effective downstream waste recycling while benefiting SDGs 6, 14, and 15 by avoiding excessive waste generation polluting water, land, and their associated ecosystems. Meeting climate commitments (SDG 13) necessitates coupling plastic waste chemical recycling practices with CCU or CCS technologies in waste processing, which also enables onsite production of plastic and alternative materials, including ephemeral packaging, and minimizes waste generation from production (SDG 12). On the global front, addressing these issues requires future research on exploring adopting and redistributing plastic waste across countries, reinforcing global treaties, and advancing public understanding of the underlying environmental and social drivers of plastic pollution mitigation (He et al., 2023). Future work will also provide a more detailed review of the criteria and methodologies for EoL technology selection to improve its sustainability performance.

8. Conclusion

This study reviewed the recent progress and sustainability analyses of chemical recycling processes of plastics waste and its derived debris from macro- to micro scales. Relevant investigations were growing with age to pinpoint the environmentally and economically sustainable technology solutions in upcycling both plastic wastes and microplastics. Understanding these processes' plastic pollution mitigation performances demanded a holistic understanding of the extent of plastic pollution originating from human activities and the local and global mitigation efforts needed for technology adoption, which remains uninvestigated in existing studies. We further proposed three future research directions associated with the extent of plastic pollution and exposure to humans, plastic pollution mitigation reduction within climate commitments, and addressing global plastic pollution. Understanding how human activities generate plastic wastes and their derived particles (footprints) could guide effective plastic waste reduction strategies from sources. Reducing plastic material consumption and employing 3R approaches (reuse, repurpose, recycle) aided by chemical processes in EoL management demanded investigating local and international collaborative efforts across global developing and industrialized regions to minimize microparticle uptake.

CRediT authorship contribution statement

Xiang Zhao: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Fengqi You:** Writing – review & editing, Visualization, Supervision, Software, Resources, Funding acquisition, Formal analysis, Data curation, Conceptualization, Investigation, Methodology, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2024.142281.

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