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Journal Article

Author(s):

Klotz, Magdalena; [Oberschelp, Christopher](#) ; [Salah, Cecilia](#) ; Subal, Luc; [Hellweg, Stefanie](#) 

Publication date:

2024-01-01

Permanent link:

<https://doi.org/https://doi.org/10.3929/ethz-b-000637796>

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Originally published in:

Science of The Total Environment 906, <https://doi.org/10.1016/j.scitotenv.2023.167586>

Funding acknowledgement:

- NCCR Catalysis (phase I) ()

Contents lists available at [ScienceDirect](https://www.sciencedirect.com)

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

The role of chemical and solvent-based recycling within a sustainable circular economy for plastics

Magdalena Klotz^{a,*}, Christopher Oberschelp^{a,c}, Cecilia Salah^{b,c}, Luc Subal^d, Stefanie Hellweg^a

^a Department of Civil, Environmental and Geomatic Engineering, ETH Zurich, Stefano-Franscini-Platz 5, 8093 Zurich, Switzerland

^b Department of Chemistry and Applied Biosciences, ETH Zurich, Vladimir-Prelog-Weg 1-5/10, 8093 Zurich, Switzerland

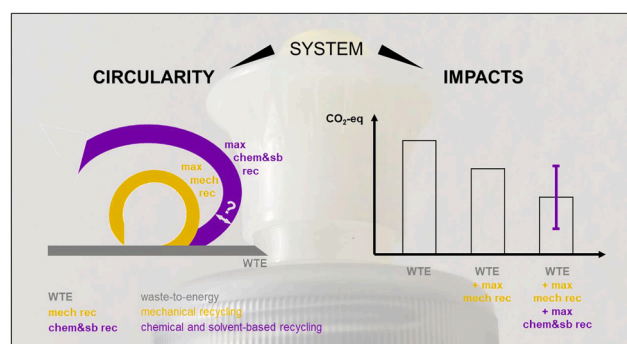
^c National Centre of Competence in Research (NCCR) Catalysis, ETH Zurich, Leopold-Ruzicka-Weg 4, 8093 Zurich, Switzerland

^d Realcycle GmbH, Hagenholzstrasse 85A, 8050 Zürich, Switzerland

HIGHLIGHTS

- Chemical and solvent-based (CSR) can complement mechanical plastic recycling.
- Gasification/pyrolysis may perform environmentally better than waste-to-energy.
- Depolymerization/dissolution achieve lowest impacts of CSR (polymer structure kept most intact).
- Impacts of CSR depend on process performance achievable on industrial scale.
- Large uncertainties related to potential benefits due to remaining technical challenges.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Daniel CW Tsang

Keywords:

Plastic recycling
Material flow analysis (MFA)
Life cycle assessment (LCA)
Environmental assessment
System assessment

ABSTRACT

Chemical and solvent-based recycling of plastic waste may help overcome some of the challenges faced by predominantly applied mechanical recycling techniques. This study quantifies the environmental impacts of chemical and solvent-based recycling as a function of varying process parameters and product composition using life cycle assessment. Furthermore, potential benefits and impacts on a system level are determined. To that end, a high-resolution material flow analysis is conducted for the reference system of Switzerland, covering all main plastic types and applications. In a scenario for the year 2040, we employ environmentally beneficial mechanical recycling where possible and convey suitable remaining waste streams to chemical or solvent-based recycling processes. Applying chemical or solvent-based recycling as a complement to maximum mechanical recycling, instead of thermal treatment with energy recovery, may achieve a reduction in the climate change impact of the system ranging from less than 10 % to almost 40 %. For achieving high environmental benefits, proper process choice and configuration are crucial. Dissolution or depolymerization provide higher benefits relative to other chemical recycling processes, but can only treat certain waste streams and require prior sorting into plastic types. Pyrolysis and gasification appeared to only have the ability to achieve substantial benefits over incineration if their output products can substitute high-impact chemicals and provided that efficient heat transfer and recovery is warranted when implemented on a large scale. As industrial-scale plants for chemical or solvent-based plastic

* Corresponding author at: Laura-Hezner-Weg 7, 8093 Zurich, Switzerland.

E-mail address: mklotz@ethz.ch (M. Klotz).

<https://doi.org/10.1016/j.scitotenv.2023.167586>

Received 13 July 2023; Received in revised form 2 October 2023; Accepted 2 October 2023

Available online 5 October 2023

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recycling are still lacking, the upscaling potential and the environmental benefits achievable in practice are highly uncertain today.

1. Introduction

Plastic recycling can reduce negative environmental impacts such as greenhouse gas emissions caused by plastic production and waste incineration. Currently, the by far most widely applied plastic recycling technology is so-called mechanical recycling, referring to reshaping processes that involve melting. However, this recycling method has a limited ability to produce useful secondary materials from mixed plastics. The reason for this is that it commonly involves an alteration of the polymer chain configuration due to a blending of different plastic wastes and thermal impact. It also leads to a transfer of (potentially hazardous) additives from the original plastics and possibly external contaminants to the recycling product. Thermosets cannot be remelted at all. Therefore, the circularity achievable via mechanical plastic recycling is limited (Klotz et al., 2023, 2022).

Alternative plastic recycling technologies that can potentially overcome the limitations of mechanical recycling mentioned are chemical or solvent-based recycling. Chemical recycling processes break down the polymer chains, to varying degrees, thermally and partly by means of reactants, potentially aided by catalysts. The polymer chains can then be rebuilt from scratch, which allows any desired chain configuration to be established, and additives or contaminants may be removed and immobilized or destroyed. Common chemical recycling technologies that are applicable for different plastic types are gasification, pyrolysis, and depolymerization (Aguado et al., 2008; Al-Salem et al., 2009; Anuar Sharuddin et al., 2016; Datta and Koczynska, 2016; Goto, 2009; Kunwar et al., 2016; Lopez et al., 2018; Miandad et al., 2016; Munir et al., 2018; Panda et al., 2010; Ragaert et al., 2017; Rahimi and García, 2017; Vollmer et al., 2020). Solvent-based (dissolution) processes do not considerably alter the polymer chain configuration and, therefore, exhibit some characteristics of mechanical recycling (Rahimi and García, 2017; Vollmer et al., 2020; Zhao et al., 2018; International Organization for Standardization, 2008). However, in contrast to mechanical recycling, they can be able to separate plastic additives from the polymer matrix or individual plastic types from multilayer structures, and hence alter the chemical composition of the input plastics mixture. Therefore, we treat solvent-based processes as a distinct recycling technology. Chemical or solvent-based recycling processes can potentially deal with a wider range of plastic waste than mechanical recycling. Moreover, the secondary feedstock materials obtained from chemical and potentially solvent-based recycling processes generally allow for a much broader application than those from mechanical recycling. It should be noted, though, that these favorable characteristics of chemical or solvent-based recycling have so far rarely been demonstrated on a full industrial scale due to, among other reasons, a current lack of profitability (Vollmer et al., 2020).

From an environmental point of view, mechanical recycling tends to outperform chemical and solvent-based recycling (Meys et al., 2020; Schwarz et al., 2021), for instance, due to a lower energy demand paired with products able to substitute for alternatives with considerable environmental impact (Meys et al., 2020). Therefore, it seems reasonable to apply mechanical recycling where possible and to complement it with chemical and solvent-based recycling of remaining waste streams. Few studies have assessed the role that chemical and solvent-based recycling could play in an integrated plastic waste management system, i.e. which waste streams would be suitable for chemical and solvent-based recycling and which environmental benefits the latter may bring. Lase et al. (2023) conducted a material flow analysis (MFA) of plastics including mechanical, chemical and solvent-based recycling, but probably overestimated the achievable mechanical recycling rate (Klotz et al., 2023) and did not couple the MFA with environmental impact data. Meys et al. (2021) optimized a plastic material flow system in order to achieve minimum climate change impacts by combining mechanical recycling and pyrolysis

with other measures such as carbon capture and utilization. Their approach, however, involves rough assumptions regarding the mechanical recyclability of different waste streams. It assumes that certain packaging plastics are completely mechanically recyclable and the remaining waste streams not at all, while all plastic waste is assumed to be chemically recyclable. In reality, not all packaging plastics offer potential for mechanical recycling, while several other plastic waste streams do (Klotz et al., 2023). At the same time, it seems unlikely that all plastics are suitable for chemical recycling since some are part of composite structures involving other materials. Another study estimated the domestic potential for chemical recycling for the Netherlands to be about 17 kg per person and year, consisting of recycling losses, mixed plastics, and polyethylene terephthalate (PET) trays (Bergsma, 2019; European Commission, 2023). Khoo (2019) determined mass-based recycling potentials based on installable capacities for mechanical and chemical recycling for Singapore (11 % and 18 % of plastic waste, respectively). Both the latter studies, however, have not or only partly analyzed plastic waste flows regarding their suitability for chemical recycling.

While a structured overview of the suitability of chemical and solvent-based recycling processes for different plastic types is lacking, life cycle assessment (LCA) data is available for several processes and plastic types, mostly for a fossil-based economy. Inventory data for different processes (energy demand, product composition, efficiency) are available from Al-Salem et al. (2014), Cardamone et al. (2022), Khoo (2019), KIT and Conversio (2018), Perugini et al. (2005), Schwarz et al. (2021), and Arena and Ardolino (2022). Climate change impacts are provided by Schwarz et al. (2021) for the vast majority of relevant processes and plastic types, as well as by Garcia-Gutierrez et al. (2023) for several different processes and plastic types. While the impacts of the former are based on theoretical models, the latter conducted an industry survey. Meys et al. (2020) quantified climate change impacts as well as fossil resource depletion for different chemical recycling processes for polyethylene (PE), polypropylene (PP), PET and polystyrene (PS). Of the different chemical and solvent-based recycling technologies, depolymerization (if applicable for a plastic type) tends to provide the highest benefits, achieving net climate change impacts of -7.2 to -0.5 kg CO₂-eq / kg of plastic input, depending on plastic type and plant size (Lindgreen and Bergsma, 2018; Meys et al., 2020; Schwarz et al., 2021; Garcia-Gutierrez et al., 2023). It is closely followed by dissolution (net climate change impacts of -6.6 to 0.3 kg CO₂-eq / kg of plastic input (Schwarz et al., 2021; Arena and Ardolino, 2022; Garcia-Gutierrez et al., 2023). Gasification tends to perform better than pyrolysis (Broeren et al., 2019; Lindgreen et al., 2017; Khoo, 2019; Schwarz et al., 2021), achieving net impacts of -1.6 to 0.4 kg CO₂-eq / kg of plastic input. Many studies have investigated pyrolysis configurations for mixed plastic waste (Al-Salem et al., 2014; Lindgreen et al., 2017; Faraca et al., 2019; Jeswani et al., 2021; Khoo, 2019; Arena and Ardolino, 2022; Garcia-Gutierrez et al., 2023), different plastic types (Civancik-Uslu et al., 2021; Volk et al., 2021), or low-density polyethylene (LDPE) (Russ et al., 2020). In most studies, pyrolysis results in a net impact of around 0.5 kg CO₂-eq per kg mixed plastic waste (Al-Salem et al., 2014; Faraca et al., 2019; Jeswani et al., 2021; Khoo, 2019; Russ et al., 2020; Arena and Ardolino, 2022; Garcia-Gutierrez et al., 2023). Exceptions with lower impacts are Lindgreen et al. (2017) (-0.8 to -0.3 kg CO₂-eq / kg of plastic input, depending on technology), Schwarz et al. (2021) (-3.5 to 0.4 kg CO₂-eq / kg of plastic input, depending on technology and plastic type) and Civancik-Uslu et al. (2021) (-1.5 to 0.5 kg CO₂-eq / kg of plastic input, depending on plastic type). The performance of pyrolysis compared to mechanical recycling depends on the assumed efficiencies of mechanical and chemical recycling, the primary material substitution ratio for mechanical recycling, and residues treatment (Al-Salem et al., 2014; Faraca et al., 2019; Jeswani et al., 2021; Volk et al., 2021). Pyrolysis

outcompetes energy recovery in waste-to-energy (WTE) plants in terms of climate change impacts according to Civancik-Uslu et al. (2021), Jeswani et al. (2021), Khoo (2019), and Russ et al. (1), the latter having impacts of 1 to 2.5 kg CO₂-eq / kg of plastic input depending on plastic type, energy efficiency and substituted energy (Civancik-Uslu et al., 2021; Jeswani et al., 2021; Khoo, 2019; Klotz et al., 2023). When the products of pyrolysis and gasification, however, are used as fuels, direct incineration of the plastic waste may perform better (Demetriou and Crossin, 2019).

The available LCA studies are almost exclusively based on theoretical models or small-scale plants. MFA studies also only approximate industrial-scale plants, assuming they can be installed. A prediction of the performance of full-scale industrial plants, therefore, involves high levels of uncertainty. Process scale-up for chemical and solvent-based recycling is technically challenging. Efficient and uniform heat transfer to the plastic waste, for instance, may be hard to implement on a large scale (Hofer and Pirker, 2020; Vollmer et al., 2020; Garcia-Gutierrez et al., 2023). Moreover, other process-related issues like corrosion or pipe clogging may arise, for example when treating PET or poly(vinyl chloride) (PVC) via pyrolysis (Al-Salem et al., 2017; Meys et al., 2020), and pilot plants usually sort out these plastic types (e.g. Quantafuel, 2023). At the same time, studies have quantified the impact of PET or PVC pyrolysis (Schwarz et al., 2021), albeit considering varied product composition and necessary removal of hydrogen chloride (HCl). Currently, numerous new plant construction activities are ongoing. Overviews of plant installations and the maturity of different process types are given by KIT and Conversio (2018), Maisels et al. (2022), Solis and Silveira (2020), and Vollmer et al. (2020). However, data on capacity and actual production are scarce. The actual practicability of chemical and solvent-based recycling is, therefore, uncertain.

In summary, up to now a comprehensive assessment with regard to which plastic wastes might be reasonably and viably treated by means of chemical or solvent-based recycling and with respect to which environmental benefits may be achieved on a system level by applying these alternative recycling processes has been lacking. In this study, we model the environmental impacts of chemical and solvent-based recycling as a function of various process parameters and product compositions. We identify plastic waste streams that are unsuitable for mechanical recycling, but appropriate for chemical or solvent-based recycling, and assess environmental impacts related to chemical or solvent-based recycling of these particular waste streams. For this, a high-resolution material flow analysis is conducted covering waste streams of the main plastic types across all relevant applications to ensure that only realistically feasible system solutions are assessed. We use Switzerland as a study region, which we consider to be representative for the whole European context since the Swiss plastics product portfolio is comparable to the one of other European countries (Klotz et al., 2023) and also the waste treatment processes modelled are pertinent for a European situation. Potentially achievable reductions in climate change impact by means of chemical and solvent-based recycling on a system level are quantified and sensitivity analyses are performed by varying process parameters based on the literature and simulation data. Finally, we assess the maturity of chemical and solvent-based recycling based on current plant operation and construction activities.

2. Methods

First, we investigated which chemical and solvent-based recycling technologies can treat which plastics (Section 2.1) and quantified related environmental impacts per input amount of plastic waste (Section 2.2). We then determined whether the composition of plastics waste streams within a material flow system that were previously identified as unsuitable for mechanical recycling (Klotz et al., 2023) allows for chemical or solvent-based recycling (Section 2.3). We determined the system impacts resulting when applying chemical or solvent-based recycling for these waste streams with LCA, performing a sensitivity analysis by varying different process parameters (Section 2.4). Finally,

to provide insight into the maturity of the application of chemical and solvent-based recycling processes in practice, we compiled data on plants in operation or under construction (Section 2.5).

2.1. Determination of suitability of chemical and solvent-based recycling processes for different plastic waste streams

We categorized chemical and solvent-based recycling processes for plastics based on process parameters (temperature level, reaction environment) and product range (type and diversity). Then we investigated, which processes are applicable for each plastic type. We also specified whether the processes are able to treat mixed waste streams. The data basis was the literature (including data given in Sections S2.1.1, S2.2.1, S2.3.1, as well as S5).

2.2. Quantification of environmental impacts related to chemical and solvent-based recycling processes

2.2.1. Goal and scope

For all chemical and solvent-based recycling processes considered (Table 1), we performed a life cycle assessment using the functional unit of 1 kg plastics input.

For impact allocation, we used system expansion with the avoided burden approach. This means, for example, that credit was given for all products of recycling as they can substitute primary materials or for energy recovered from thermal treatment.

In the impact assessment, we focused on climate change impacts as these belong to the most severe environmental impacts of plastics that can currently be assessed with LCA (see Klotz et al. (2023)). We used the IPCC 2021 GWP 100a method for assessing the climate change impacts of chemical and solvent-based recycling processes.

2.2.2. Inventory analysis with sensitivity assessment

For each chemical or solvent-based recycling technology, we investigated a range of different process configurations based on the literature and simulation data. We provide all parameters and respective values considered in Table S2.9.1. In the assessment, we used the same underlying energy scenarios for all processes and substituted chemicals. For background data, we used inventories from the Ecoinvent v3.8 and IDEA v2.3 databases.

With respect to the provision of energy for all chemical or solvent-based recycling processes and for the alternative production of substituted chemicals, we assessed two scenarios: a fossil and a renewable scenario (all details and the carbon intensities of the energy mixes are available in Section S2.5). In the fossil scenario, for the recycling processes, we assumed an energy supply via a natural gas industrial furnace as the reactors are usually located directly inside a combustion chamber. Suitable reactor types are, for instance, fluidized bed or tube reactors for gasification or pyrolysis, which provide for a uniform temperature distribution and good heat transfer on a large scale (Aguado et al., 2008; Al-Salem et al., 2010; Hofer and Pirker, 2020; Kaminsky, 2021; Kunwar et al., 2016; Lopez et al., 2017; Okuwaki, 2004; Scheirs, 2006; Solis and Silveira, 2020; Sosa Sabogal et al., 2021). We derived reactor heat transfer efficiencies from Wernet et al. (2016) and Zhang et al. (2021), and varied them in the course of the sensitivity assessment (Table S2.9.1). For chemicals production, we assumed energy supply via a natural gas combined heat and power plant.

In the renewable energy scenario, we assumed an electricity production mix of 50 % open-ground photovoltaic and 50 % offshore wind power plants as photovoltaic roof installations are assumed to mostly supply households and because many chemical companies are investing in offshore wind farms, the energy from which can partly be used directly at production sites close to the shore (BASF, 2023a). We accounted for storage losses of 25 % and assumed that storage would be required for 25 % of the electricity produced. We assumed heat provision for the recycling processes via power-to-heat. We varied the heat

transfer efficiency in the sensitivity assessment based on [Madeddu et al. \(2020\)](#) to depict different suitable reactor types (Table S2.9.1). The latter include resistance furnaces, microwave and radio frequency heaters, infrared heaters, plasma technology, or inductive heating, which can achieve sufficient temperatures ([Madeddu et al., 2020](#)) and be applied for this purpose ([Anuar Sharuddin et al., 2016](#); [Ignatyev et al., 2014](#); [Madeddu et al., 2020](#); [Rahimi and García, 2017](#); [Sosa Sabogal et al., 2021](#); [Stapf et al., 2019](#)). For chemicals production, we considered heat supply via steam, which is the most common heat source for this purpose ([Madeddu et al., 2020](#)), with average boiler efficiency of 97.5 % from [Madeddu et al. \(2020\)](#).

For crediting chemical or solvent-based recycling products, we calculated the alternative production impacts of the chemicals substituted for all processes based on cradle-to-gate heat and electricity demands as well as direct CO₂ emissions. The respective values were derived from the IDEA database v2.3. The energy mix was in line with the energy scenario. The few substances not available in the database were approximated with other similar substances (Section S2.6). Water, as part of the product, was given zero credits. CO₂ was assumed to be released as emission into the air. Since it may also be sequestered or further purified to be utilizable in industry in the future, we also did a sensitivity analysis where we did not account for these CO₂ emissions.

To assess the possible impact variations of gasification, we modelled a gasification process in Aspen Plus v11, according to [Salah et al. \(2023\)](#). We conducted process simulations for PE, PET, PP, and PS inputs, respectively, covering most of the plastic types conveyed to chemical recycling in the highest amounts (Sections S1.2.1 and S1.2.2). Steam was deployed as gasifying agent to react with the input polymers at a 1.5 mass ratio; the reactor size corresponded to a mass flow rate of 8000 t polymer input per year. This simulation allowed the effect of different parameter changes on energy demand to be quantified. The energy required for the process is provided by the partial oxidation of the input material, the low-pressure input steam, as well as external sources. The life cycle inventories were compiled based on the energy balance data obtained from the simulation, taking different reactor heat transfer efficiencies and potential catalyst use into consideration, as well as product separation efforts for the gaseous and liquid products (based on [Bisinella et al. \(2021\)](#), [RWE and Linde \(2017\)](#), [Salah et al. \(2023\)](#), [Somoza-Tornos et al. \(2020\)](#), and Section S2.1.3) in cases when product components were assumed to be individually utilized (and credited). Apart from the latter, we varied impacts and credits individually for similar reasons as outlined regarding pyrolysis. The parameter combinations considered are provided in Section S2.1.5.

To quantify the substitution benefits, we calculated which alternative production impacts can be avoided by gasification products with different compositions (Section S2.1.5). We derived a range of possible compositions from a literature review (Section S2.1.1) as well as from the simulation results (Section S2.1.2). For the gaseous product, we consider, on the one hand, the case of separation and individual substitution of all main product components and, on the other, the separation of water and CO₂ only, as well as the utilization of the remaining product as syngas. A further transformation to methanol, which can be used for plastic monomer production, was not considered. We also assessed the case of energetic utilization of the product so as to be able to compare it to direct incineration of the plastic waste. For the liquid product, we considered either separation into components or substitution of naphtha. For char, no material utilization was assumed due to potentially hazardous plastic additives contained therein, whereas energy recovery as well as landfill were assumed to be suitable alternatives. The total product amount resulted from the simulation.

For pyrolysis, we relied on energy demand data available from the literature ([Cardamone et al., 2022](#); [Khoo, 2019](#); [Perugini et al., 2005](#); [Schwarz et al., 2021](#); [Somoza-Tornos et al., 2020](#); Section S2.2.2). The range of substitution benefits were determined based on possible product compositions from the literature (Sections S2.2.1 and S2.2.3). For the target liquid product, we considered, on the one hand, a separation and individual utilization of product components and, on the other hand, a utilization of the complete product. For the gaseous

product, we considered both the case of material utilization as well as energetic utilization (as, e.g., considered in [Cardamone et al. \(2022\)](#)). For char, no material utilization was assumed, but instead either energy recovery or landfill (as in the case of gasification). As pyrolysis, similar to gasification, yields products that are typically also used as fuels, we modelled the case of energetic utilization of the complete product as a reference for comparison with direct incineration of the plastic waste. The total amount of the pyrolysis product corresponds to the plastic waste input as the reactions take place in an inert atmosphere.

When calculating the net environmental impacts of pyrolysis, we mostly varied impacts and credits independently, and did not differentiate between different individual plastic types or mixed plastic waste, for several reasons. Some parameters relevant for the impact are completely independent from product composition such as reactor efficiency and product energy recovery. For other process parameters, due to the complex behavior of waste plastics under pyrolysis conditions ([Lopez et al., 2017](#)), there is no straight-forward correlation with product composition. While a higher temperature or longer reaction time may lead to more aromatics and less aliphatic compounds being present in the product ([Aguado et al., 2008](#); [Anuar Sharuddin et al., 2016](#); [Kaminsky, 2021](#); [Scheirs, 2006](#)), the use of a catalyst may affect product composition and the temperature level of the process ([Aguado et al., 2008](#); [Anuar Sharuddin et al., 2016](#); [Cardamone et al., 2022](#); [Ignatyev et al., 2014](#); [Kaminsky, 2021](#); [Ragaert et al., 2017](#); [Arena and Ardolino, 2022](#)). Thereby, the uniformity of the temperature distribution within the reactor plays an important role as it may partly lead to undesired reaction conditions ([Kunwar et al., 2016](#); [Lopez et al., 2017](#)). The specific plastic input mix affects product composition ([Anuar Sharuddin et al., 2016](#); [Kaminsky, 2021](#); [Schwarz et al., 2021](#)), whereby secondary reactions may occur ([Kaminsky, 2021](#)) and be affected by plastic additives. From the literature considered, no direct correlation between process input and substances yielded is apparent (Section S2.2.1). For instance, high liquid yields or shares of styrene therein, providing for high substitution credits, are achieved for mixed plastic waste from electrical and electronic equipment, rather than with treatments of individual plastic types. This may be because of the different factors mentioned that affect product composition. This complex situation led to the decision not to consider potential correlations between impacts and credits or to differentiate between different plastic waste inputs, and in so doing depicting a maximum range of possible net impacts for realistic process configurations. We did, however, consider product separation efforts for the cases in which we credited individual product components separately. The parameter combinations considered are provided in Section S2.2.4.

To assess variations in depolymerization impacts, we focused on glycolysis of PET, polyamides (PA), polycarbonates (PC) and polyurethanes (PUR) since glycolysis is applied in industrial plants and due to data availability. However, depolymerization can also be achieved via other processes such as hydrolysis or alcoholysis ([Goto, 2009](#)). In the inventory, we considered the reaction enthalpy, as well as the energy required for heating both the plastic and the corresponding reactant amount up to the process temperature and for compensating heat losses. We also accounted for the production of the reactant participating in the reaction, isolation of the target product via distillation, shredding of the input material, and approximated impacts arising because of catalyst use. We calculated the required reactant and total product amounts stoichiometrically. We varied the yield of monomers and oligomers utilizable for plastic production based on the literature and considered residues treatment. The yield depends on the temperature level and reaction time ([Goto, 2009](#)), but can also be affected by catalyst use ([Datta and Kopczyńska, 2016](#); [Goto, 2009](#); [Ragaert et al., 2017](#)), and we assume it is further determined by reactants mixing and product separation efficiencies. Therefore, we varied parameters affecting impacts and credits (Table S2.9.1) independently.

For dissolution, we considered ranges for energy and solvent demand from the literature ([Cardamone et al., 2022](#); [Schwarz et al., 2021](#); [Wiprächtiger et al., 2020](#)), assuming that the reported energy demands given include efforts for shredding, solution enthalpy, heat losses, and

product separation, and fit all plastic types treatable with dissolution. We assumed that no catalyst is required (Knappich et al., 2019) and neglected the use of a drying additive (Cardamone et al., 2022). The yield depends on the reaction time and temperature level (Zhao et al., 2018; Knappich et al., 2019), but we assume factors such as homogeneity of reactant mixing and efficiency of product separation as well. Therefore, we varied impacts and credits independently, leading to a maximum range of possible process impacts.

2.3. Material flow analysis of an integrated plastic waste management system involving mechanical as well as chemical or solvent-based recycling

In Klotz et al. (2023) we concluded that the mechanical recycling rate (calculated based on utilizable secondary material) in the mid-term future is limited to a maximum of 31 % for the main plastics (Table 1) from all application segments in Switzerland (Klotz and Haupt, 2022), distinguishing 11 plastic types and 69 product groups. For the present study, we kept this mechanical recycling situation in a scenario for the year 2040 unchanged. In addition, we assumed the same reused and uncollected waste amounts (ending up in the environment e.g. due to littering or non-dismantled pipes) as in Klotz et al. (2023). For the remaining waste, which in Klotz et al. (2023) is treated mostly in waste-to-energy (WTE) plants as well as in cement kilns, we modelled the following treatment (Fig. 2):

- For the remaining post-consumer waste
 - we assumed that a share cannot be sufficiently separated from other materials so that it needs to be treated in WTE plants (15 % based on Lase et al. (2023)); this may include, for instance, plastics in mixed building waste containing large pieces of other materials, but does not comprise waste that may, despite being a material composite, be treated by chemical recycling (e.g. textiles may be made of a mixture of plastic and non-plastic fiber material, of which the plastic fiber share may still be recovered via chemical recycling);
 - for the rest (85 %) we assumed that
 - * 40 % is difficult to further separate into plastic types and can, therefore, only be treated by pyrolysis or gasification; this value is based on the share of waste not suitable for mechanical recycling in Klotz et al. (2023) because it is very inhomogeneous or composed of multi-material structures, which makes sorting very burdensome;
 - * the rest (60 %) can be separated into plastic types and therewith allows for the application of all chemical or solvent-based recycling processes.
- With regards to sorting and recycling residues, it was assumed that attachments of other materials than plastic do not inhibit chemical or solvent-based recycling as pilot plants were able to utilize these residues (Hofer and Pirker, 2020). Thereby,
 - 50 % were assumed to either arise as single-plastic-type streams or to be separable into plastic types, allowing for the application of all chemical or solvent-based recycling processes; the rationales for this choice were the following: the residues from mechanical recycling, which is done for individual plastic types, typically mainly contain one plastic type; we assumed missorted amounts Klotz et al. (2023) to be separable into plastic types as the multi-material shares in products conveyed to mechanical recycling in Klotz et al. (2023) are generally low; in contrast, the other part of sorting and recycling losses is constituted exactly by material that is not separable into plastic types (Klotz et al., 2023) and, therefore, not mechanically recyclable; as overall about half of the sorting and recycling losses in Klotz et al. (2023) are due to multi-material structures, while the other half is due to missorting and recycling process losses, we assumed 50 % of all sorting and recycling residues to be separable into plastic types;
 - the other half cannot be separated into plastic types, restricting the treatment to pyrolysis or gasification.

The treatment shares considered are meant to depict the maximum possible amounts treatable via chemical or solvent-based recycling from a technical perspective. This is consistent with the assumptions of the maximum mechanical recycling scenario of Klotz et al. (2023), which we used and expanded here with chemical and solvent-based recycling. Therefore, the results should be understood as a higher benchmark of what the combination of mechanical as well as chemical and solvent-based recycling may be able to achieve. To determine the effect of potentially lower waste shares conveyable to chemical or solvent-based recycling on the system impacts, we conducted a sensitivity assessment. In this, we considered only 50 % of remaining post-consumer plastic waste to be separable from other materials, and only 25 % of all waste recycled chemically or by means of solvents to be separable into plastic types.

Regarding the choice of specific chemical or solvent-based recycling processes applied, we depict four scenarios (Table S1). In each scenario, we conveyed all suitable waste streams to one of the four distinguished process types, respectively. Depolymerization and dissolution cannot be applied to all plastic types (Table 1). Therefore, for those plastic types for which no depolymerization process is available, we applied dissolution, and vice versa. For the waste streams restricted to pyrolysis or gasification (as they cannot be separated into individual plastic types) in the depolymerization and dissolution scenarios, we conveyed half of the concerned waste streams to gasification and half to pyrolysis, with intermediate parameter assumptions to depict an intermediate situation, whereby in the sensitivity analysis we quantified the difference between the extreme cases of applying either only worst-performing gasification or only best-performing pyrolysis.

2.4. Quantification of system impacts

Based on the MFA conducted (Section 2.3) and process impacts per kilogram for chemical and solvent-based recycling (Section 2.2), we performed a system-level LCA. The LCA compares scenarios of plastics waste management in Switzerland and, in particular, assesses the contribution that chemical and solvent-based recycling can make in reducing climate impacts in the future. The functional unit was defined as production of plastics consumed as well as treatment of plastics waste arising in Switzerland in 2040 according to the plastic material flow system described in Section 2.3.

The system boundaries included plastic production and all processes related to waste treatment. Regarding plastic waste collected for mechanical recycling, collection, sorting, and mechanical recycling was considered. For the sorting or recycling residues, additional transportation to chemical or solvent-based recycling plants is required, as well as the process of chemical or solvent-based recycling. Plastic waste to be recycled chemically or by means of solvents needs to be collected, transported to sorting, separated from other materials, transported to recycling, and recycled chemically or by means of solvents. In the case of a treatment via depolymerization or dissolution, further sorting into plastic types is considered. The assumptions regarding additional collection, sorting, and transportation efforts related to chemical or solvent-based recycling were based on Klotz et al. (2023) and Van Eygen et al. (2018), and are documented in Section S3.1. For gasification, we assumed longer transportation distances than for the other processes as the plant size must be larger (BASF, 2023b), resulting in a lower geographical density of plants.

Variations in inventory data for chemical and solvent-based recycling were considered as described in Section 2.2. For plastics production, mechanical recycling, and waste incineration, we retrieved the inventory from Klotz et al. (2023). The impacts of second-hand use and plastics reaching into the environment were not quantified as in Klotz et al. (2023).

The LCA calculations were carried out using Brightway 2, Python 3.9, and Excel 2019.

2.5. Investigation of chemical and solvent-based recycling plants in operation

To gain insight into the maturity of different chemical and solvent-based recycling technologies for different waste streams, we compiled a non-exhaustive list of chemical and solvent-based recycling plants in operation or under construction. We provide data on applied process type, treatable plastic types, plant capacity, current production amounts, locations, duration for which plants have been in operation, and other parameters in a structured way. This data was partly used as a basis for waste flow allocation to processes (Section 2.1) and for quantification of their impacts (Section 2.2.2), and can give an impression of the timeframes needed for a broad implementation of chemical or solvent-based recycling.

3. Results and discussion

3.1. Applicability of chemical and solvent-based recycling processes

We categorized available chemical or solvent-based recycling processes following common approaches. Most processes can treat nearly

all of the main plastic types (Table 1). Some polymer types pose challenges to pyrolysis and gasification, affecting process operation and product quality, however, with ways to handle undesired byproducts available (Table 1). Depolymerization is mainly used for condensation polymers (PET, PA, PC, PUR) (Goto, 2009), while high monomer yields may be achieved with pyrolysis configurations for addition polymers such as PE, PP, or PS (Goto, 2009; Meys et al., 2020). Solvent-based processes exist for all main plastic types, except for thermoset polyurethanes, which require a breaking of primary chemical bonds for recycling. This means that—if sorting into plastic types is possible—most plastic types in waste streams can be recycled via any desired chemical or solvent-based recycling process. The process choice, therefore, depends on environmental and financial considerations. Waste streams not separable into plastic types are restricted to processes that are able to recover useful products from an input of mixed plastics, while these waste streams are not suitable for processes targeting the recovery of individual plastic types.

Some chemical recycling technologies are not covered in Table 1 and this paper as they are specific to certain plastic types or less widespread. These include the production of value-added chemicals from plastic waste (Meys et al., 2020). For PVC specifically, a process exists to

Table 1

Main chemical recycling routes: characteristics and suitability for plastic types. A cross (x) means that a process type can be applied for a plastic type. A tilde (~) means that a plastic type can be treated with a certain process, but requires extra treatment steps. A slash (/) means that a plastic type cannot be treated with a certain process. LDPE comprises LLDPE, PS refers to general-purpose polystyrene (GPPS) and expanded polystyrene (EPS), and PA includes all types of polyamides, as in Klotz and Haupt (2022) and Klotz et al. (2023). ABS: acrylonitrile butadiene styrene, HIPS: high-impact polystyrene.

	Gasification	Pyrolysis	Depolymerization	Dissolution
Reaction environment ^a	Presence of sub-stoichiometric amount of oxygen	Absence of oxygen	Presence of a reactant such as ethylene glycol, methanol, or water	Presence of a solvent
Temperature level ^b	400–1500°C, typically 700–1200°C	300–800°C	50–450°C, typically around 200°C	30–200°C
Decomposition pattern ^c	Decomposition to mainly H ₂ and CO; smaller amounts of H ₂ O, CH ₄ , CO ₂ especially at lower side of temperature range; other hydrocarbons and other gaseous, solid, and liquid products depending on input composition	Decomposition to liquid and gaseous aliphatic and aromatic hydrocarbons (C ₂ -C ₁₈ +, BTX, plastic monomers, waxes), a solid product (carbon, minerals), as well as other products depending on plastic composition	Decomposition to monomers and oligomers, as well as byproducts	Rupture of secondary bonds between polymer chains, while the primary bonds within the polymer chains remain intact (as for mechanical recycling)
Suitable for recovery of	Individual and mixed plastics ^d	Individual and mixed plastics ^e	Individual plastics ^f	Individual plastics ^g
HDPE	x	x	/	x
LDPE	x	x	/	x
PET	x	–	x	x
PP	x	x	/	x
PS	x	x	/	x
PVC	–	–	/	x
ABS	x	x	/	x
HIPS	x	x	/	x
PA	–	–	x	x
PC	x	–	x	x
PUR	–	–	x	/

^a Source: Al-Salem et al. (2009), Goto (2009), Munir et al. (2018).

^b Source: Schwarz et al. (2021), Knappich et al. (2019), Solis and Silveira (1), Ragaert et al. (2017), Zhao et al. (2018), Sections S2.1.1, S2.2.1, S2.3.1.

^c Source: Baerns and Behr (2013), Arpe (2007), Ragaert et al. (2017), Goto (2009), Zhao et al. (2018), Knappich et al. (2019), La Mantia (2002), Schlummer et al. (2020), Schwarz et al. (2021), Puig-Arnavat et al. (2010), Munir et al. (2018), Meys et al. (2020), Sections S2.1.1, S2.2.1, S2.3.1; BTX: benzene, toluene, xylenes.

^d Source: Ragaert et al. (2017), Jeong et al. (2022), Borgianni et al. (2002), Sections S2.1.1, S5; suitability for technical plastics and polyurethanes assumed based on Buekens and Zhou (2014) and Guo et al. (2015); for PVC, chlorine in product gas must be eliminated, but ways to achieve this exist (Borgianni et al., 2002); polyamides and polyurethanes lead to undesired byproducts (personal communication Wolfgang Hofer, OMV, 2023-09-18).

^e Source: Ragaert et al. (2017), Meys et al. (2020), Garcia-Gutierrez et al. (2023), Sections S2.2.1, S5; suitability for technical plastics and polyurethanes assumed based on Buekens and Zhou (2014), Font et al. (2001), Zakharyan and Maksimov (2022a), and Zakharyan and Maksimov (2022b); the presence of elements other than carbon and hydrogen, as is mainly the case for PET, PVC, PA, PC, and PUR, leads to byproducts and requires different degrees of specialized treatment (personal communication Wolfgang Hofer, OMV, 2023-09-18, Zakharyan and Maksimov (2022a), Zakharyan and Maksimov (2022b)); PET and PVC have been pyrolyzed in laboratory experiments (Malik et al., 2022; Ma et al., 2002), however, in practice they can only be present in very small amounts if no additional measures are taken, because they are decomposed to corrosive products (benzoic acid, HCl) and adversely affect product quality (Aguado et al., 2008; Schwarz et al., 2021; Scheirs, 2006; Okuwaki, 2004; Hofer and Pirker, 2020; Meys et al., 2020); however, chlorine can be removed via an additional treatment step and neutralized (Ragaert et al., 2017; Aguado et al., 2008; Kunwar et al., 2016; Schwarz et al., 2021); high yield of PE, PP, and PS monomers (ethylene, propylene, styrene) can be achieved, especially for pyrolysis of these plastic types (Sections S2.2.1, S5, Meys et al., 2020; Kaminsky et al., 2004; Schwarz et al., 2021; Ragaert et al., 2017)

^f Source: Schlummer et al. (2020), Goto (2009), Lucas et al. (2018), Hong and Chen (2017), Sections S2.3.0.1, S5.

^g Source: Lucas et al. (2018), Mäurer and Schlummer (2004), Knappich et al. (2019), Zhao et al. (2018), Roelands (2020); assumed that dissolution also possible for HIPS, which usually consists to >90 % of PS (Kaiser, 2021); thermoset polyurethanes are cross-linked, therefore, primary bonds have to be broken for reshaping.

recover HCl (La Mantia, 2002). Hydrocracking is similar to pyrolysis with, however, the addition of hydrogen, causing more saturated chemicals to be produced (Munir et al., 2018; Perugini et al., 2005). Liquefaction may refer to thermal cracking of plastics in liquid form at higher pressure and lower temperature than pyrolysis (Jena and Das, 2011; Okuwaki, 2004; Scheirs, 2006). PP may be depolymerized via ionization, being subjected to an inductively coupled plasma (Rahimi and García, 2017).

3.2. Variability of chemical and solvent-based recycling impacts per kilogram of plastics input

The environmental impacts of chemical or solvent-based recycling processes may vary greatly depending on process choice and process configuration as well as input material (Fig. 1). Gasification and pyrolysis have the highest gross impacts per input amount, with gasification tending to have higher impacts. The gross impacts of depolymerization and dissolution overlap with pyrolysis, but tend to be lower. The latter two processes gain the highest credits for substituted products. Dissolution can obtain the same credits as mechanical recycling per amount of secondary plastic produced since the polymer chains are not modified to a greater extent in the course of this process than is the case with mechanical recycling. As the largest share of plastic production impacts is often caused by monomer production (Section S2.6), it is plausible that depolymerization can achieve credits that are not much below those of dissolution. The credits for gasification and pyrolysis are lower, whereby the utilization of their products may even lead to additional impacts instead of providing benefits. The net impacts of gasification and pyrolysis largely overlap and are above zero for many process configurations, while depolymerization and dissolution in all cases considered achieve net environmental benefits. The tendency is that the less the

polymer chains and monomers are decomposed, the lower the net impacts achievable.

In the renewable energy scenario, the patterns are mostly the same as for the fossil scenario, with lower absolute values for impacts, credits, and net impacts (Section S2.10). Exceptions are gasification and pyrolysis net impacts, which tend to be higher in the renewable than in the fossil scenario. The reason is that their product utilization in the renewable scenario has additional impacts for most configurations instead of receiving credits, which is due to the energetic utilization of the solid product share and the gaseous product of pyrolysis. This does not, however, affect the process performance ranking based on their intermediate net impacts.

Depending on the type of process, different factors have the biggest effect on the overall impacts (Sections S2.1.5, S2.2.4, S2.3.4, S2.4.3). For gasification, both impacts and credits may vary largely. The impacts are determined to a large extent by heat transfer efficiency and plastic type. Efforts for separating individual product components may be considerable, potentially increasing the total energy demand by more than half. The credits mainly depend on the composition of the gaseous product.

Likewise, for pyrolysis, variations in impact and credits may affect the net impact to a similar extent. The impact variation depends mostly on the energy demand (as derived from the literature); the credit variation is determined to the greatest extent by the composition of the liquid product.

When assuming zero impacts for CO₂ generated during gasification or pyrolysis (Section 2.2.2), the pyrolysis impacts are only negligibly affected (due to small CO₂ amounts produced), while the net gasification impacts decrease by 19 % for an intermediate product composition (Sections S2.1.4 and S2.2.3).

As a treatment for the solid residue of gasification and pyrolysis, landfill seems more suitable than energy recovery in terms of climate

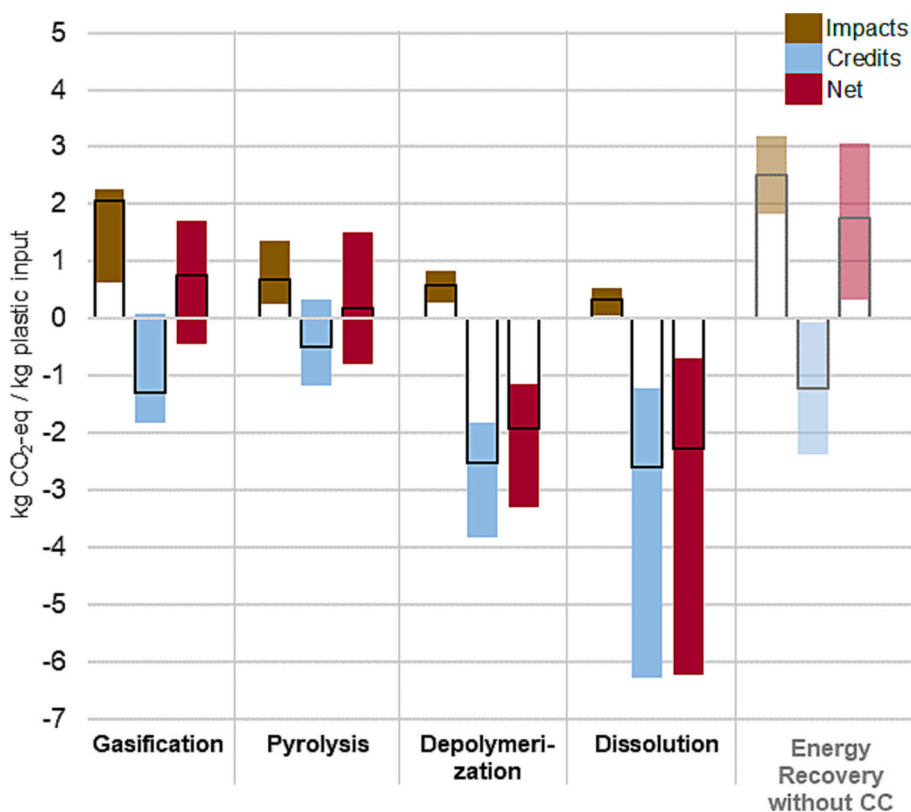


Fig. 1. Variation in gross impacts and credits, as well as net impacts for different chemical recycling processes, per amount of plastic waste treated. The framed bars show the impacts for each process based on the assumptions of medium values in the sensitivity analysis (see Table S2.9.1) and the filled bars the whole range from minimum to maximum values. For comparison, impacts of energy recovery are given. The ranges for the latter consider different fossil shares in the substituted energy mix and energy efficiencies (Section S3.2). CC: carbon capture.

change impacts. This is because the assumed alternative energy source (natural gas) has lower emissions per heating value. In the case that landfilling is chosen, preventive measures against leaching of potentially hazardous additives must be taken. However, according to Swiss legislation, the carbon content (Section S2.2.1) may be too high for landfilling (Swiss Federal Council, 2015), and from a financial point of view it may be favorable to utilize the energetic content of the char.

Energetic utilization of all gasification products may achieve a similar net impact as material utilization in the case of intermediate product composition in a fossil scenario, despite lower credits, because of the lower product component separation efforts required. In a renewable scenario, however, potential energy recovery of the complete gasification product would perform much worse than other utilization options (Section S2.1.5). For pyrolysis, energetic utilization performs worse than material utilization in both the fossil and renewable scenarios, which is due to lower estimates for product separation efforts compared to gasification.

For depolymerization, the credits for the substituted monomer, depending on plastic type input, affect the net impact the most. For a specific plastic type, the yield was the most relevant parameter.

Similarly, for dissolution, the net impacts were determined to the greatest extent by the substituted plastic type. For a specific plastic type, the yield had the greatest effect on the net impact for plastic types with high production impacts, whereas for plastic types with lower production impacts, the process energy demand was more relevant.

3.3. Impact reduction potential of chemical and solvent-based recycling on a system level

Based on the suitability of chemical and solvent-based recycling processes for different waste flows (Sections 2.1 and 2.3), in the maximum recycling scenario assessed in this paper, 55 % of the waste amount undergoes chemical or solvent-based recycling, besides 31 % of the waste amount resulting in utilizable secondary material from mechanical recycling (Fig. 2).

Chemical or solvent-based recycling of suitable non-mechanically recycled waste instead of incineration may, depending on process choice, achieve climate change impact savings of 18–34 % (referring to the system impact for the case of no recycling, 0.9–1.8 Mt. CO₂-eq; Fig. 3). The potential savings are based on a calculation with intermediate value assumptions for process parameters (Section 2.2.2) and maximum waste amounts conveyed to chemical or solvent-based recycling (Section 2.3). The impact savings result from lower impacts of chemical or solvent-based recycling per waste amount treated compared to WTE (Section 3.4). The potentially higher impact reduction achievable by chemical and solvent-based as compared to mechanical recycling on a system level is due to the fact that a larger waste volume is recycled chemically or by means of solvents than mechanically (Fig. 2). However, the specific climate change benefits per input amount into chemical or solvent-based recycling (1.7–3.2 t CO₂-eq / t on a system level, depending on process type) are smaller than those per secondary material amount from mechanical recycling (4.3 t CO₂-eq / t; Klotz et al., 2023). The main benefit of applying chemical or solvent-based recycling comes from avoiding incineration impacts (1.4 Mt. CO₂-eq), while the net impact of chemical or solvent-based recycling ranges from

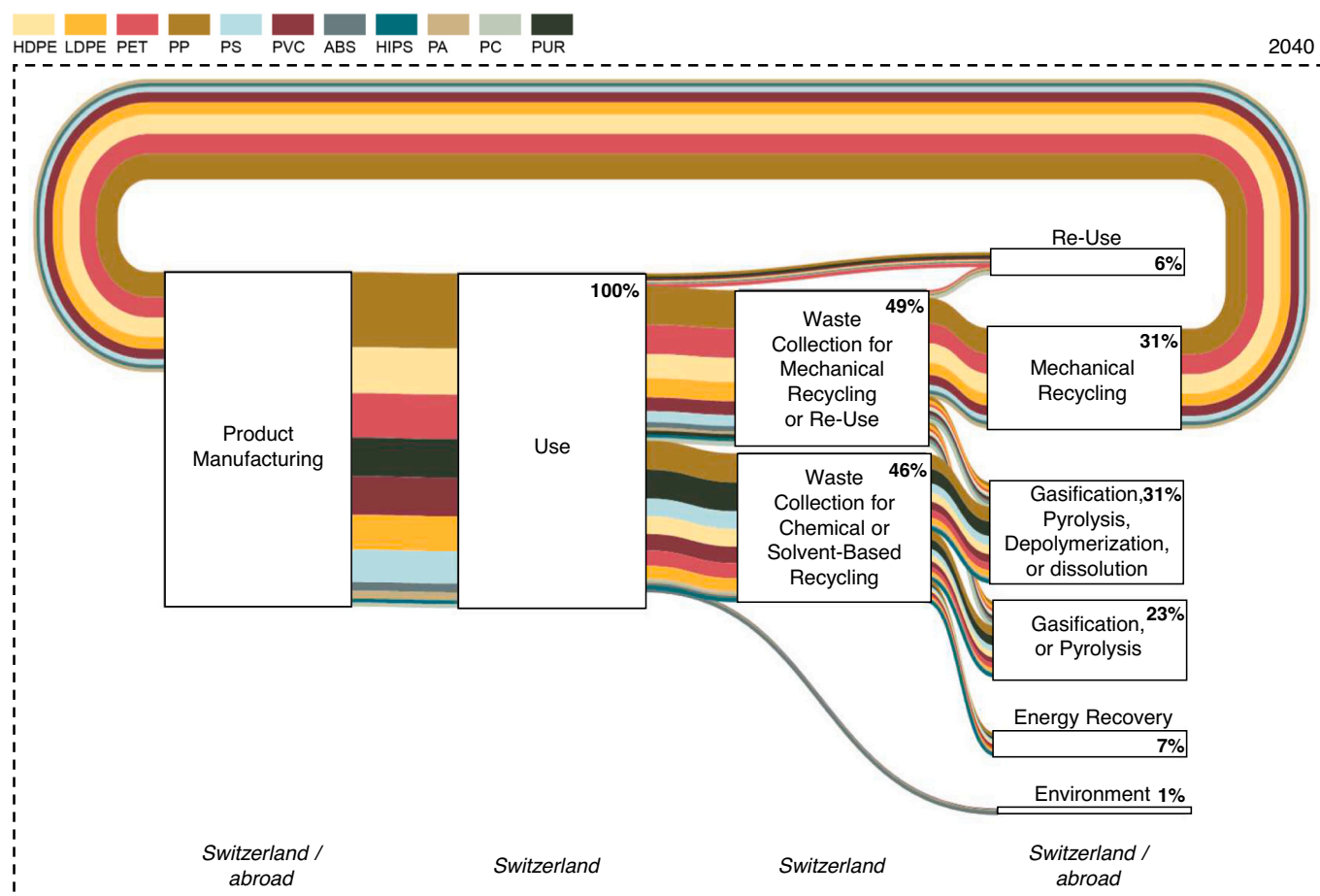


Fig. 2. Possible material flow paths for the scenarios modelled (Table S1). Material flows resulting from a sensitivity analysis assuming less waste to be suitable for chemical or solvent-based recycling are provided in Section S1.2.2. The percentages given are based on weight and refer to the total waste amount.

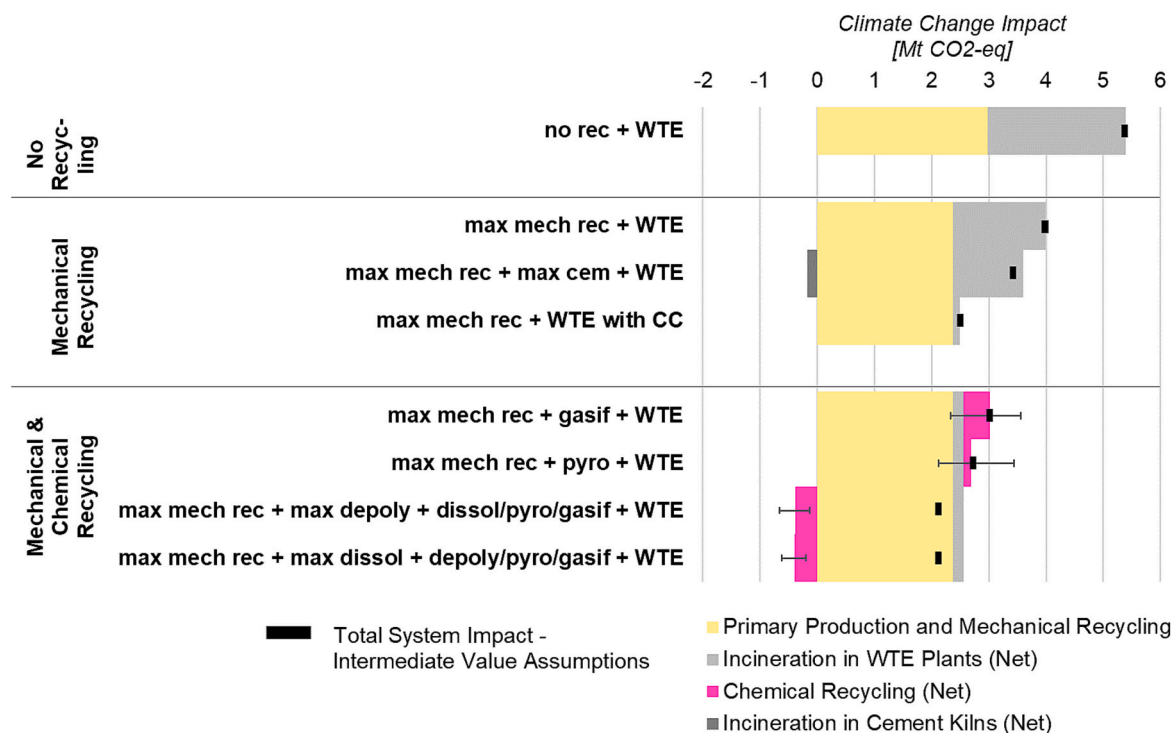


Fig. 3. System impacts of a no-recycling scenario compared to maximum mechanical recycling in combination with different incineration scenarios (Klotz et al., 2023) and, alternatively, different chemical or solvent-based recycling situations. The impacts of chemical and solvent-based recycling were calculated based on intermediate assumptions for process parameters, the uncertainty bars represent the best and worst case assumptions for each process (Section 2.2.2). The production and incineration impacts are given for a future energy and mobility situation (as in Klotz et al. (2023)). rec: recycling, WTE: energy recovery in waste-to-energy plants, max: maximal, mech: mechanical, cem: energy recovery in cement kilns, CC: carbon capture, gasif: gasification, pyro: pyrolysis, dissol: dissolution, depoly: depolymerization.

impacts of 0.5 Mt. CO₂-eq to benefits of 0.4 Mt. CO₂-eq, depending on process choice. Carbon capture and sequestration (CCS) can also diminish incineration impacts and may, therefore, achieve similar benefits as chemical or solvent-based recycling when applied in addition to maximum mechanical recycling (1.5 Mt. CO₂-eq, Fig. 3). CCS may be comparatively cheap to implement (Fellner and Brunner, 2022) and ramping up its installation in Switzerland is provided for by an agreement of the Swiss Federal Department of the Environment, Transport, Energy and Communications (DETEC) with the Association of Plant Managers of Swiss Waste Treatment Installations (VBSA) (Swiss Federal Office for the Environment (FOEN), 2023).

If more unfavorable assumptions are made with regard to the amounts of plastic waste that can be treated with chemical or solvent-based recycling (Section 2.3), only 39 % of the plastic waste is conveyed to chemical recycling (Section S1.2.2). In this scenario, the achievable benefits are lower (impact reduction of 13–19 % on a system level compared to incineration; Section S3.3.2).

From a system perspective, the great variation in chemical or solvent-based recycling impacts (Section 3.2) leads to overlapping uncertainty ranges for the different scenarios for chemical or solvent-based recycling (error bars from Fig. 3, which are based on the highest and lowest possible impact values for chemical or solvent-based recycling according to the sensitivity analysis). Dissolution and depolymerization tend to perform better than pyrolysis and gasification. The worst performance is displayed by gasification with low gaseous yield, a syngas containing large shares of components with low alternative production impacts that are separated and utilized individually, utilization of the liquid product as a naphtha substitute, and incineration of the solid product assuming a high carbon content (Section S2.1.5; Table S2.9.1). When this comes together with factors leading to a high process energy demand, such as a high reaction temperature and low heat transfer efficiency of the reactor, the impact savings of applying chemical

recycling instead of incineration in WTE plants on a system level may amount to only 8 % (Section S3.3.1). If the energy substituted by the heat and electricity generated in WTE plants is to a large extent fossil-based, energy recovery may even perform better than chemical recycling for unfavorable configurations of the latter (Section S3.3.1). This means that chemical recycling is only able to perform considerably better than incineration if the process configuration is carefully chosen.

The highest benefits can be achieved by depolymerization and dissolution. This means that sorting waste into plastic types, which is required for these processes, pays off environmentally, while from a cost perspective the situation may look different. The impact variation in the respective scenarios stems to a large extent from combining depolymerization and dissolution with either gasification or pyrolysis (see Section 2.3). Both the depolymerization and the dissolution scenarios are in any case, however, clearly beneficial compared to incineration as a complement to mechanical recycling (minimal reduction of system impact by around 30 %).

Recycling less waste mechanically, and instead doing so chemically or by means of solvents, would lead to overall higher system impacts because of the better environmental performance of mechanical recycling (Section 3.4).

3.4. Comparison to alternative circular solutions and treatments

Compared to mechanical recycling, gasification and pyrolysis tend to have higher gross impacts and receive lower substitution credits for their products. Depolymerization and dissolution, however, may perform similarly to mechanical recycling both regarding gross impacts and gross credits (see Section 3.2 and Klotz et al. (2023)). This is plausible because all of these recycling processes keep the structure of the polymers intact to a high degree.

In the present study, chemical or solvent-based recycling was

assumed to only take plastic wastes as an input that are not mechanically recyclable. Therefore, the alternatives to which chemical or solvent-based recycling processes should be compared include incineration in waste-to-energy (WTE) plants or cement kilns, utilization in steel furnaces or coke ovens, or landfilling. Pyrolysis and gasification have the potential to perform better than incineration in WTE plants, for WTE gross electric and thermal efficiencies amounting to 20.12 % and 36.21 %, respectively (Klotz et al., 2023), when the WTE energy produced substitutes a future heat and electricity mix (Klotz et al., 2023) (as considered for Fig. 3). The net WTE impacts for this case range from 1.8 to 3.1 kg CO₂-eq / kg of input, depending on plastic type (Klotz et al., 2023), and compare to pyrolysis or gasification impacts ranging from -0.8 to 1.7 kg CO₂-eq / kg (Fig. 1) of input in the fossil scenario (Section 2.2.2), as shown in Fig. 3, and -0.1 to 1.8 kg CO₂-eq / kg of input in the renewable scenario. However, when WTE energy substitutes heat from natural gas and electricity from the European mix (representing today's energy situation in Klotz et al. (2023)), the impact ranges of pyrolysis and gasification overlap with the WTE impacts (net WTE impacts of 1.0–2.1 kg CO₂-eq / kg of input; Klotz et al., 2023). This is because of the high credits received by WTE for CO₂-intensive energy substituted in such a scenario. Consequently, if we fail in reducing fossil-based heat and electricity supply, WTE may perform similarly or even better than chemical recycling (Section S3.3.1). In such a case of largely fossil energy substitution, achieving high WTE efficiencies and concentrating plants in locations allowing for a high heat utilization rate may provide for WTE net impacts as low as 0.3–1.6 kg CO₂-eq / kg of input (Section S3.2). For a future (low-impact) energy mix substituted, in contrast, WTE efficiency has a minor effect on net impacts. An advantage that pyrolysis and gasification could in any case offer compared to WTE is that their output products can be transported and stored. Thus, they allow for flexible energy generation or other utilization in terms of site and time.

Incineration of plastics in cement kilns for substituting coal may perform better than gasification and pyrolysis per amount of waste treated (net impacts of -1.7 to -0.2 kg CO₂-eq / kg of waste treated for cement kilns (Klotz et al., 2023); 0.2 and 0.8 kg CO₂-eq / kg of waste treated for pyrolysis and gasification, respectively, in the intermediate case). From a system perspective, however, gasification and pyrolysis are able to achieve higher benefits by avoiding WTE impacts (Fig. 3) because the amount treatable with these technologies is not limited, while for cement kilns it depends on the cement production volumes (Klotz et al., 2023). In addition, coal has already partly been substituted by alternative fuels in cement kilns, which additionally limits the substitution potential (Klotz et al., 2023).

Landfilling of plastics has low climate change impacts when considering a time frame of 100 years (0.1 kg CO₂-eq / kg of landfilled plastics; Wernet et al., 2016), lying within the range of gasification and pyrolysis impacts. However, landfills occupy land and hazardous additives included in waste plastics may leach to the groundwater.

In a future circular plastics economy, additional circularity strategies to those prevalently applied today, for instance leading to reduced consumption amounts, may assume a more prominent role (Lisiecki et al., 2023). If such measures environmentally outperform chemical and solvent-based, or also mechanical plastic recycling, they should be prioritized from an ecological perspective, while they may partly be complementary to recycling.

3.5. Relation to the literature and limitations of the study

The climate change impacts of dissolution as well as depolymerization of our study are similar to those of Schwarz et al. (2021) (except for PA), while for pyrolysis and gasification they are higher. The latter corresponds to the findings of other studies (Al-Salem et al., 2014; Faraca et al., 2019; Jeswani et al., 2021; Khoo, 2019; Russ et al., 2020; Arena and Ardolino, 2022; Garcia-Gutierrez et al., 2023). The climate change impact of pyrolysis for intermediate value assumptions from this

study is similar to other studies (0.2 compared to 0.5 kg CO₂-eq/kg, see Section 1). Possible reasons for the lower impacts in Schwarz et al. (2021) may be potentially assumed higher yields of high-impact substances and higher alternative production impacts, as well as lower energy demands or energy production impacts. Another reason may be the different Ecoinvent versions used for inventory data. Since not all assumptions regarding these issues were openly reported in Schwarz et al. (2021), it is not possible to identify the reasons for the differences with certainty. The overlap of net impacts for different gasification and pyrolysis configurations from this study is in line with Schwarz et al. (2021). However, our finding that pyrolysis tends to perform better than gasification contradicts the findings of Schwarz et al. (2021) and other studies (Broeren et al., 2019; Lindgreen et al., 2017; Khoo, 2019). The reason for the good performance of gasification in the literature may be high assumed yields of aliphatic as well as aromatic compounds, which amount to up to 80 % in Schwarz et al. (2021), excluding methane, compared to less than 10w% of aliphatic and aromatic compounds (without methane) in the gasification product in our study. These aliphatic and aromatic components may arise when reaction time is short according to Schwarz et al. (2021) and are assumed to be due to an incomplete reaction, which may be difficult to control in practice. The impacts of the substituted chemicals in the fossil scenario of this study are generally lower than those from the Ecoinvent database. This may be because natural gas was assumed as energy source, while the Ecoinvent datasets may include energy from oil or coal. This leads to lower credits, while at the same time using natural gas as energy source for the processes lowers the gross impacts.

The present study, while trying to depict a range of process variations, is subject to uncertainties that were not quantified. These largely concern parameters affecting the gross impact of the processes. Regarding gasification and pyrolysis, efforts related to product component separation were only very roughly estimated, and catalyst use was approximated. Catalyst use may be required to achieve the desired product composition and be financially relevant, while with the chosen model parameters, it is not very relevant in terms of climate change impacts. However, the specific material used as a catalyst may drastically affect the environmental impact. Gasification impacts were determined for all high-volume plastic types except for polyurethanes (Sections 2.2.2, S1.2.1), which constitute the second biggest plastics fraction available, by mass, for chemical recycling. PVC was neither considered for determining gasification impacts. Although PVC is conveyed to chemical recycling in smaller amounts, it may be relevant in terms of process impacts due to required HCl removal (Schwarz et al., 2021). Regarding the impacts of depolymerization, reaction enthalpy was considered, while potentially required activation energy and solution enthalpy were neglected. Depolymerization was approximated with glycolysis, while it may also be achieved by other processes such as methanolysis, which can yield other products (e.g. dimethyl terephthalate (DMT) instead of bis(2-hydroxyethyl terephthalate) (BHET) for PET; Goto, 2009) and consume different amounts of reactant. Methanolysis or hydrolysis may achieve a slightly better performance than glycolysis for the example of PET (Garcia-Gutierrez et al., 2023).

Substitution credits for individual components of chemical recycling products may vary considerably. Many of the substituted substances may be produced from different raw materials via different production routes and typically stem from multi-product systems, which makes the impacts dependent on allocation methods. We only considered one production option based on the approach used in the IDEA database. We did, however, align the energy source for production with the one supplying the recycling processes.

The impact ranges are rather overestimated by this study as we varied many parameters independently, partly combining worst- and best-case parameter settings (Section 2.2.2). Possibly, further correlations between process parameters and product composition could be identified. One example is PET gasification, which needs much less energy than required for PE, PP, or PS, while at the same time the product

composition is different because of the oxygen contained in PET (Section S2.1.2). Considering this dependency would lead to lower impacts along with lower credits, which would narrow the net impact range. This may also be relevant for other plastic types than PET, which may as well lead to different products than gained for PE, PP, and PS (Jeong et al., 2022). The process input composition, in general, affects the product output composition to a certain extent (Sections 2.2.2 and 3.1). For the plastic material flow system assessed in this study, the composition of plastic waste input to chemical recycling is given, resulting from the MFA, while for calculating gasification and pyrolysis impacts, we varied the product output composition over a range of potential plastic waste inputs. This likely resulted in an overestimated impact variation for the system assessed. Besides, while gasification and pyrolysis are theoretically able to treat all plastic types, certain polymers may be more suitable for one process or the other (Table 1). It may, for instance, be convenient to treat polymers containing oxygen by gasification rather than by pyrolysis (personal communication Wolfgang Hofer, OMV, 2023-09-18). An appropriate allocation of plastic types to gasification and pyrolysis may lead to lower impacts, which we did not consider. We do, however, provide the impacts for different process configurations and product compositions separately so that they may be combined in different ways and matched with specific input compositions.

Impact categories other than climate change may also be important when evaluating chemical and solvent-based recycling. Future research should, for instance, assess potential toxic impacts of process emissions. Process design of future plants should safeguard that potentially arising undesired byproducts such as dioxins or persistent organic pollutants (POPs) (Paladino and Moranda, 2021; Weiland et al., 2021; Joung et al., 2007), possibly formed from substances present as plastic additives, are removed or destroyed (Chang et al., 2009; Yamawaki, 2003; Maric et al., 2020).

The saving of fossil resources by means of chemical recycling was not assessed in this study. It seems to be of secondary importance given the climate emergency. Whatever the case may be, chemical recycling may make regions like Europe more independent of fossil resource imports.

The estimation of waste amounts recyclable via chemical or solvent-based recycling processes may be too high. This is, on the one hand, because sufficient information to assess the suitability for chemical or solvent-based recycling was not available for all waste plastic streams. The lack of large-scale plants in operation (Section 3.6), on the other hand, makes it very uncertain to which degree the application of chemical or solvent-based recycling may be expanded. Despite possibly overly large waste amounts conveyed to chemical recycling in the model, the resulting environmental benefits may anyhow to be low.

An aspect that was not regarded in this study, but is of utmost relevance, is the behavior of plastic additives in chemical recycling processes, which may impair performance. For instance, metals ending up in pyrolysis oil may lead to higher coke formation when the latter is used in steam crackers compared to naphtha (Kusenberget al., 2022). Plastic additives may, therefore, affect the practical feasibility and product usability of chemical recycling, and must be further investigated.

3.6. Current situation of chemical and solvent-based recycling implementation in practice

There are many ongoing activities worldwide devoted to the upscaling of chemical or solvent-based recycling processes with the aim to install industrial-scale plants for a range of plastic types (Table 2). While plants operating on an industrial scale exist for each process type, actual production data was found for only a few gasification and pyrolysis plants (Table 2, Section S5). The maximum installed capacities of individual plants for different process types range from 8,000 to 70,000 t/a (Table 2), with the median size of installed plants decreasing in accordance with the sequence gasification, pyrolysis, depolymerization, dissolution (Section S5).

The process choice in practice may be driven by financial aspects. These are determined, among others, by the demand for energy, catalysts, solvent-reactants, solvents, and product revenue. The financial expenses are related to specific process configurations and, therewith, environmental performance. For instance, regarding energy demand, the degree of separation of individual product components is very relevant (Section 3.2), and concrete catalyst selection is likely to determine product composition to a significant extent as well. Another factor that can affect process choice is plant size: gasification tends to require larger plant sizes (BASF, 2023b), which means that a large waste amount needs to be available per area, or transport distances increase. The share of transportation, as well as collection and sorting processes related to chemical recycling, however, is only responsible for 1 % of the total system impact (Sections S3.1 and S3.3.1), while transportation efforts may financially play a role.

Achieving process design so as to gain products that are able to substitute chemicals with high climate change impacts in favor of low-impact ones on a large scale is crucial for an environmental point of view (Section 3.2). Some of the industrial-scale plants in operation may currently only produce fuels (Section S5). As the utilization of chemical recycling products as fuels may not create substantial environmental benefits (Sections S2.1.5 and S2.2.4), process improvements are potentially required. Activities related to upgrading pyrolysis oil, for instance, are ongoing (Section S5). Important technical challenges include the implementation of uniform heat distribution and efficient heat transfer, and avoiding coke formation on a large scale (Hofer and Pirker, 2020; Scheirs, 2006; Vollmer et al., 2020; Qureshi et al., 2020; Garcia-Gutierrez et al., 2023). Uncertainties regarding whether chemical and solvent-based recycling processes can be implemented as modelled are, therefore, still considerable.

4. Conclusions

The climate change impacts of chemical or solvent-based recycling processes for plastics can range from values similar to those resulting from direct energy recovery from plastic waste via incineration to benefits approximating those achievable for mechanical recycling. To achieve low environmental impacts, plastic waste should be sorted into plastic types, where possible, to allow for dissolution or depolymerization. If pyrolysis or gasification are applied, efficient heat transfer and recovery as well as a

Table 2
Summary of data on existing and planned chemical recycling plants worldwide in 2022 (Section S5).

	Gasification	Pyrolysis	Depolymerization	Dissolution
Maximum actual production per plant	50,000 t/a	20,000 t/a	no data	no data
Maximum installed capacity per plant	70,000 t/a	20,000 t/a	50,000 t/a	8,000 t/a
Maximum planned capacity per plant	360,000 t/a	100,000 t/a	160,000 t/a	48,000 t/a
Treated plastic types	Mixed plastic waste	HDPE, LDPE, PP, PS, PA, rubber, and others; lower shares of PET, PVC	PET, PA	HDPE, LDPE, PET, PP, PS, PVC, PA

composition of the main product enabling a substitution of chemicals with high alternative production impacts should be aspired to. The strong dependence of the environmental impact of chemical or solvent-based processes on process type as well as specific parameters suggests that the specifics matter and no general conclusions about the performance of these alternative recycling processes can be drawn. By providing chemical and solvent-based recycling impacts in dependence of process parameters, the present study enables process configurations that are environmentally competitive to be identified and, hence, can inform decisions about technology use and implementation.

In plastic waste management systems, chemical and solvent-based recycling seem suitable for application as a complement to mechanical recycling due to the tendency towards better environmental performance of the latter. Recycling mixed or contaminated waste streams that are unsuitable for mechanical recycling chemically or by means of solvents instead of recovering their energy appears to be potentially environmentally beneficial in most cases. The reduction in system impact achievable may amount up to 40 %. However, impact savings over energy recovery may only be minor for certain process configurations, even when making generous assumptions regarding the applicability of chemical recycling given its current marginal utilization. Chemical recycling may for certain configurations even perform worse than energy recovery if WTE energy is able to achieve high benefits by substituting for fossil heat and electricity and WTE efficiencies are high. This broad range of possible outcomes demonstrates the necessity of careful process selection and optimization. Whether chemical or solvent-based recycling will in fact be able to provide considerable environmental benefits depends on process performance achieved on a large scale. Several challenges for implementation remain, such as designing reactors that achieve a specific target product composition, understanding the behavior of the diverse plastic additives in chemical recycling processes, and minimizing process energy demand.

CRediT authorship contribution statement

Magdalena Klotz: Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Visualization, Writing – original draft. **Christopher Oberschelp:** Conceptualization, Data curation, Investigation, Methodology, Writing – review & editing. **Cecilia Salah:** Data curation, Investigation, Methodology, Writing – review & editing. **Luc Subal:** Data curation, Investigation, Methodology, Writing – review & editing. **Stefanie Hellweg:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

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

The data that support the findings of this study are available in the supporting information of this article.

Acknowledgements

This work was supported by the Swiss Federal Office for the Environment (grant numbers 8T20/17.0103.PJ and 8T20/18.0118.PJ); the Swiss Federal Office of Public Health (grant number 18.000809); and the Canton of Zurich's Office for Waste, Water, Energy and Air (grant number 85P-1454 and within the framework of the project 'Clean Cycle 2.0'). This publication was created as part of NCCR Catalysis, a National Centre of Competence in Research funded by the Swiss National Science Foundation (grant number 180544).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.167586>.

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