Resources, Conservation & Recycling 162 (2020) 105010

Contents lists available at ScienceDirect

Resources, Conservation & Recycling

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

Full length article

Towards a circular economy for plastic packaging wastes – the environmental potential of chemical recycling

Raoul Meys[a], Felicitas Frick[a,c], Stefan Westhues[b], André Sternberg[a], Jürgen Klankermayer[b], André Bardow[a,d,e,*]

[a] Institute for Technical Thermodynamics, RWTH Aachen University, Schinkelstr. 8, 52062 Aachen, Germany
[b] Institute of Technical and Macromolecular Chemistry, RWTH Aachen University, Worringerweg 1, 52074 Aachen, Germany
[c] HTP GmbH & Co. KG, Maria-Theresia Allee 35, 52064 Aachen, Germany
[d] Institute of Energy and Climate Research - Energy Systems Engineering (IEK-10), Forschungszentrum Jülich GmbH, Jülich, Germany
[e] Energy & Process Systems Engineering, ETH Zürich, 8092 Zürich, Switzerland

ARTICLE INFO

Keywords:
Chemical recycling
Life cycle assessment
Circular economy
Plastic waste
Plastic waste treatment

ABSTRACT

Plastic packaging waste faces increasingly stringent sustainability targets such as recycling rates of 55% imposed by the European Commission. To realize the vision of a circular economy, chemical recycling is advocated as a large-scale avenue to decrease fossil resource depletion and greenhouse gas (GHG) emissions. In this work, we develop a theoretical model for chemical recycling technologies assuming ideal performance. The theoretical model allows us to compute the minimal environmental impacts for chemical recycling technologies and compare them to real-case benchmark waste treatments. Thereby, we robustly identify chemical recycling technologies that will not result in environmental benefits, since their minimal environmental impacts are already higher than those of current benchmark waste treatments. In this way, we show that PET, HDPE, LDPE, PP and PS should not be recycled chemically to refinery feedstock or fuel products and rather be treated by mechanical recycling and energy recovery in cement kilns in order to reduce global warming impacts. In contrast, chemical recycling to monomers or value-added products could potentially reduce global warming impacts compared to all benchmark waste treatments by up to 4.3 kg CO_2-eq per kg treated PET packaging waste. By analyzing 75 waste treatment scenarios for 5 environmental impacts, our analysis offers guidance to stakeholders involved in chemical recycling to identify the most promising as well as the least promising chemical recycling technologies.

1. Introduction

Plastics have experienced unique market growth of 23 % between 2008 and 2015, because of their versatile properties and cheap production (European Comission, 2013). However, about 91 % of all produced plastic has not been recycled. If the current trend of plastic production and waste management continues, a cumulative 12,000 Mt of plastic waste would end in landfills or the natural environment until 2050 (Geyer et al., 2017). Furthermore, plastics production has been shown to emit 1.78 Gt of CO_2-eq in 2015. These emissions will increase to 6.5 Gt. CO_2-eq in 2050 (Zheng and Suh, 2019). Consequently, the increasing amount of virgin plastic production and waste threatens our natural environment by both plastic pollution and greenhouse gas emissions.

Almost half of the globally produced plastic waste consists of 5 types of plastic packaging: (1) polyethylene terephthalate (PET), (2) low-density polyethylene (LDPE), (3) high-density polyethylene (HDPE), (4) polypropylene (PP) and (5) polystyrene (PS). In 2015, these types of plastic packaging wastes amounted to 141 Mt and are expected to further increase. (Geyer et al., 2017) Thus, proper handling of plastic packaging waste is a key challenge to evolve into an environment-friendly future (World Economic Forum and Ellen MacArthur Foundation, 2017).

The challenges are addressed by governmental institutions and scientists through shifting towards a circular economy. The circular economy redesigns man-made systems to align economic and environmental well-being by recycling and thus, circulating material flows (Reichel et al., 2016). For plastic packaging waste, the European Commission's circular economy package imposed a recycling rate of 55 % for 2030 (EU Commission, 2018). Similarly, the United States

---

[*] Corresponding author at: Energy & Process Systems Engineering, ETH Zürich, CLA F 19.1, Tannenstrasse 3, 8092 Zürich, Switzerland.
E-mail address: abardow@ethz.ch (A. Bardow).

https://doi.org/10.1016/j.resconrec.2020.105010
Received 5 February 2020; Received in revised form 13 May 2020; Accepted 10 June 2020

(The REMADE Institute, 2019), as well as China (McDowall et al., 2017), boost the circular economy towards higher recycling rates.

To efficiently recycle large amounts of plastic packaging wastes, a circular economy requires suitable technologies, such as chemical recycling (Nat Sustain, 2018; Rahimi and García, 2017). Chemical recycling turns plastic packaging waste into chemical products, avoiding their production from fossil feedstock in the first place. Therefore, chemical recycling is expected to decrease the demand for the planets' finite fossil resources as well as the emissions of greenhouse gases (World Economic Forum and Ellen MacArthur Foundation, 2017). At the same time, chemical recycling provides chemical products that are chemically identical to the replaced products. Thereby, chemical recycling avoids performance losses currently observed for mechanical recycling of plastic packaging, so-called "downcycling" (Hong and Chen, 2017). Due to downcycling, products ultimately have to be incinerated or end up in landfills after shorter use cycles.

However, environmental benefits by chemical recycling are intensely debated: Geyer et al. (2016) show that closed-loop recycling systems have no intrinsic environmental benefit over open-loop recycling systems. In fact, Shen et al. (2010) found that linear recycling pathways for PET by mechanical recycling is environmentally superior to circular pathways by chemical recycling to feedstock monomers, even if the mechanically recycled PET is ultimately incinerated. Additionally, studies indicate that large amounts of collected and sorted plastic packaging waste can be efficiently recycled mechanically with material properties sufficient to substitute virgin polymer (Michaud et al., 2010; Prognos AG, 2008).

In conclusion, there is a mismatch between expected environmental benefits and results from prospective environmental evaluations for chemical recycling. At the same time, chemical recycling is still in early development (Rahimi and García, 2017) and has not been fully assessed environmentally. It is therefore timely to assess if and to which extent chemical recycling of plastic packaging waste can achieve environmental benefits before research funds and time are invested.

The most established method for environmental assessments of products and technologies is a standardized Life Cycle Assessment (LCA) (ISO 14040, 2006; ISO 14044, 2006). LCA requires full energy- and mass-balances of the full-scale processes. Consequently, LCA is complex and time-consuming for novel technologies such as chemical recycling, where only limited data is available. Furthermore, reviews of LCA studies showed that methodological variations hinder comparisons of individual case studies (Laurent et al., 2014). Thus, methods to assess environmental benefits of chemical recycling have to overcome two challenges: (1) the assessment of many chemical recycling technologies despite low data availability and (2) a consistent LCA methodology to ensure comparability.

In this work, we overcome both challenges by determining the maximum environmental benefits of 26 chemical recycling technologies compared to 18 benchmark waste treatment technologies, based on a consistent LCA-based methodology. Maximal possible environmental benefits are denoted as the *environmental potential of chemical recycling.*

For this purpose, we derive an LCA-based method that is based on reaction chemistry and basic thermodynamic data. The LCA-based method is used to assess the environmental potential of chemical recycling for the major plastic packaging wastes: PET, HDPE, LDPE, PP, and PS. We compare the environmental impacts of (1) ideal chemical recycling to (2) the benchmark waste treatment. The ideal thermodynamic assessment results in minimal environmental impacts for chemical recycling. Thus, we robustly identify chemical recycling without any promise of environmental benefits since their minimal environmental impacts are already higher than those of current benchmark waste treatments. By analyzing 26 chemical recycling technologies compared to 18 benchmark waste treatment technologies, our analysis identifies the chemical recycling technologies with the highest potential to reduce global warming, fossil resource depletion, terrestrial acidification as well as freshwater and marine eutrophication.

2. Material and Methods

The maximal environmental benefits of a chemical recycling technology are denoted as the *environmental potential of chemical recycling.* The environmental potential is based on a comparative LCA between chemical recycling and its benchmark waste treatment. In this section, we present the general methodology to calculate the environmental potential: the system boundaries and the functional unit, the calculation of ideal chemical recycling inventories, the relevant impact categories, the uncertainty and robustness of results, and the scope of the analysis. Values for all inventories are given in the Supplementary Material.

2.1. System boundaries and functional unit

LCA evaluates the environmental impacts of products and processes from cradle-to-grave, e.g. from the extraction of raw materials over the use-phase to the disposal or recycling (Baumann and Tillman, 2004). In comparative LCAs, only changing activities need to be considered, because identical activities cancel each other out in all scenarios (Eriksson et al., 2005). For instance, waste generation can be neglected when comparing different energy recovery options for the same plastic waste. Here, it is assumed that waste is collected and sorted before entering waste treatment. Thus, collection and sorting can be omitted in our comparative LCA study.

Chemical recycling has two major effects: the benchmark waste treatment (e.g., landfill) is substituted and primary chemical production is avoided. However, the benchmark waste treatment might produce a valuable product, which now needs to be compensated by primary materials. Thus, system boundaries need to include the environmental impacts of (1) the benchmark waste treatment (WT) or chemical recycling (CR) and (2) the avoided benchmark product (avP) or the avoided chemical production (avC) (see Fig. 1). The avoided production leads to a credit in LCA, denoted as "avoided burden", and is commonly used in LCAs for waste treatment.

In LCA, the functional unit ensures a consistent comparison between technology options (Baumann and Tillman, 2004). Previous LCA studies highlighted that the composition of plastic waste affects LCA results (Lazarevic et al., 2010). Thus, we define the functional unit as the treatment of 1 kg of plastic packaging waste of defined composition, including organic/inorganic contamination and moisture.

2.2. Computation of the environmental potential

The environmental potential (E_{pot}) is the difference between the net

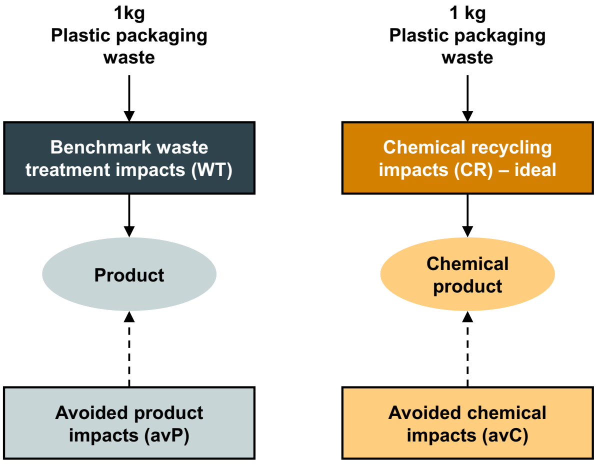


Fig. 1. Comparison between the benchmark waste treatment (WT) and chemical recycling (CR) including the avoided conventional products (avP) and chemicals (avC). The input stream is 1 kg collected and sorted plastic packaging wastes.

environmental impacts of the benchmark waste treatment ($EI_{\mathrm{WT,net}}$) and ideal chemical recycling ($EI_{\mathrm{CR,\ ideal,\ net}}$), equation (1). Net environmental impacts consist of direct environmental impacts of the benchmark waste treatment ($EI_{\mathrm{WT,direct}}$) and chemical recycling ($EI_{\mathrm{CR,\ ideal,\ direct}}$) minus the credit for avoided products (EI_{avP}) and chemicals (EI_{avC}):

$$E_{\mathrm{pot}} = \overbrace{(EI_{\mathrm{WT,\ direct}} - EI_{\mathrm{avP}})}^{EI_{\mathrm{WT,net}}} - \overbrace{(EI_{\mathrm{CR,ideal,direct}} - EI_{\mathrm{avC}})}^{EI_{\mathrm{CR,ideal,net}}} \tag{1}$$

The direct environmental impact of the benchmark ($EI_{\mathrm{WT,\ direct}}$) includes all environmental impacts required to treat 1 kg of plastic packaging waste. For example, energy recovery from plastic packaging waste leads to direct environmental impacts from flue gas emissions, the supply of auxiliary materials and energy for flue gas cleaning, as well as residue treatment (fly ash, sludge, wastewater, and slag treatment) (Doka, 2003, 2013). Avoided environmental impacts (EI_{avP}) of energy recovery include a credit for the products heat and electricity (Eriksson and Finnveden, 2017). For all benchmarks, inventories are taken from industrial datasets (see section 2.5.3).

Chemical recycling converts 1 kg of treated plastic packaging waste into m_{j} kg of one or more chemicals. These chemicals substitute their conventional production and the corresponding environmental impacts (EI_{j}), leading to a credit (EI_{avC}):

$$EI_{\mathrm{avC}} = \sum_{1}^{j} EI_{\mathrm{j}} m_{\mathrm{j}} \tag{2}$$

Environmental impacts (EI_{j}) are based on data from LCA databases and represent the production of the identical chemical from cradle-to-gate. For chemically identical products, the gate-to-grave phase is the same and does not lead to further credits if market-mediated effects can be excluded.

The remaining challenge is to calculate the direct environmental impacts of the ideal chemical recycling ($EI_{\mathrm{CR,\ ideal,\ direct}}$), because only limited data is available, e.g. only the reaction equations. However, this limited data is sufficient for our analysis since our goal is to compute the minimal environmental impacts of chemical recycling. For this purpose, we propose an approach based on stoichiometry and thermodynamic data. The approach includes (1) the reactants ($\sum_1^i EI_{\mathrm{i}} m_{\mathrm{i}}$), (2) residual wastes ($\sum_1^k EI_{\mathrm{k}} m_{\mathrm{k}}$), and (3) thermal energy ($EI_{\mathrm{H}} Q_{\mathrm{H}}$):

$$EI_{\mathrm{CR,ideal,direct}} = \sum_{1}^{i} EI_{\mathrm{i}} m_{\mathrm{i}} + \sum_{1}^{k} EI_{\mathrm{k}} m_{\mathrm{k}} + EI_{\mathrm{H}} Q_{\mathrm{H}} \tag{3}$$

Here, m is the respective mass of reactants i and residual k. $EI_{\mathrm{i/k}}$ represents the environmental impacts per kg to produce reactant i or treat residual k.

Our goal is to calculate the minimal environmental impacts for chemical recycling. In line with this goal, Q_{H} is the minimal energy demand for the complete chemical recycling process per 1 kg of plastic packaging waste. It is assumed that energy is provided by heat, because the energy demand is largely driven by the chemical decomposition of polymers that takes place above 300°C (Lopez et al., 2017; Rahimi and García, 2017). Using electric heating above these temperatures is possible, however, would lead to higher global warming impacts under current emission intensities of electricity supply. Thus, EI_{H} denotes the environmental impact of providing 1 MJ of thermal energy by industrial furnaces using natural gas as fuel. All masses and the required thermal energy are calculated assuming ideal thermodynamics:

(I) 100 % conversion of polymer waste based on the stoichiometric reaction

$$\underbrace{\sum_{1}^{\mathrm{i}} \nu_{\mathrm{i}} M_{\mathrm{i}}}_{\text{reactants}} + \underbrace{\nu_{\mathrm{p}} M_{\mathrm{p}}}_{\text{polymer}} = \underbrace{\sum_{1}^{\mathrm{j}} \nu_{\mathrm{j}} M_{\mathrm{j}}}_{\text{products}} \tag{4}$$

(II) The minimal energy requirement from an energy balance of the process assuming 100% conversion:

$$Q_{\mathrm{H}} = \Delta H_{\mathrm{R}}^{0} \tag{5}$$

where the reaction enthalpy $\Delta H_{\mathrm{R}}^{0}$ at standard conditions (25°C, 1 atm) is the difference of the standard enthalpies of formation of the pure in- and outputs (see Supplementary Material section S8).

2.3. Environmental impacts

In the main text, we focus on global warming and fossil resource depletion, because these are stated as the major targets of a circular economy for plastic waste (World Economic Forum and Ellen MacArthur Foundation, 2017). Additionally, we study terrestrial acidification and marine/freshwater eutrophication (sections S15 and S16 of the Supplementary Material). All impact categories are modeled according to Recipe midpoint indicators implemented in Ecoinvent (Ecoinvent, 2019). It is important to keep in mind that the physical lower bound of the environmental impacts depends on the method and version used for impact assessment.

2.4. Robustness and uncertainty of the environmental potential

Full LCAs require, at best, industry-based inventories. Based on these inventories, the environmental benefits (E_{ben}) by chemical recycling could be calculated as the difference between the benchmark ($EI_{\mathrm{WT,\ net}}$) and the industrialized chemical recycling ($EI_{\mathrm{CR,\ net}}$):

$$E_{\mathrm{ben}} = EI_{\mathrm{WT,net}} - EI_{\mathrm{CR,net}} \tag{6}$$

However, many chemical recycling technologies are in early development and sufficient data is missing. To still analyze chemical recycling, we assume thermodynamically ideal chemical recycling (cf. section 2.2). The ideal assumptions lead to an optimal chemical recycling process with minimal environmental impacts ($EI_{\mathrm{CR,\ ideal,\ net}}$). Real-case chemical recycling ($EI_{\mathrm{CR,\ net}}$) will always lead to increased environmental impacts such that $EI_{\mathrm{CR,\ net}} > EI_{\mathrm{CR,\ ideal,\ net}}$. For instance, ideal thermodynamic models result in minimal energy demands. Real chemical recycling would increase energy demands and environmental impacts.

As a result, the environmental potential ($E_{\mathrm{pot}} = EI_{\mathrm{WT,net}} - EI_{\mathrm{CR,ideal,net}}$, cf. section 2.2, eq. (1)) will always be higher than the environmental benefits of chemical recycling, i.e. the following equation is always true: $E_{\mathrm{pot}} > E_{\mathrm{ben}}$.

Thereby, our analysis allows identifying the most promising chemical recycling pathways. However, a positive environmental potential does not imply that a chemical recycling pathway *is* beneficial but that it *could be* better than the benchmark. A full LCA is warranted to ensure benefits of the real chemical recycling technology. We assess the uncertainty of the obtained results in a sensitivity analysis (see section 3.4). This sensitivity analysis varies the energy demand and the conversion rate of chemical recycling.

In contrast, our method is robust for negative environmental potentials: a negative value indicates that chemical recycling is inferior to the benchmark even for ideal conditions. Thus, for real conditions no benefits can be expected, because environmental impacts can only increase ($E_{\mathrm{pot}} > E_{\mathrm{ben}}$).

2.5. Scope

Packaging represents approx. 50 % (Geyer et al., 2017) of global plastic waste. Our study covers 5 major plastic packaging wastes: PET, HDPE, LDPE, PP, and PS. Our literature review identified 26 chemical recycling technologies and 18 benchmark waste treatments, e.g. waste incineration, energy recovery in cement kilns, and mechanical recycling for each plastic packaging waste. All packaging wastes are assumed to be collected and sorted before the waste treatment. Furthermore, the environmental potential is calculated in the European context, where the circular economy is gaining increased attention (European Comission, 2015) and sufficient data is available. In the main text, the technologies are only briefly described. Details can be found in the Supplementary Material.

2.5.1. *Benchmark waste treatments*

Waste treatment of plastic packaging waste is dominated by three technologies: landfilling, energy recovery, and mechanical recycling (Geyer et al., 2017).

In 2015, *landfilling* represented the dominating technology and accounted for 59 % of the treatment of plastic waste (Geyer et al., 2017). However, landfill bans are discussed in several world regions and were proven highly effective. In Europe, landfill rates dropped by 38 % and recycling rates increased by 64 % from 2006 to 2014. Based on the success, several European countries are planning to implement landfill bans (PlasticsEurope, 2018). Thus, landfilling is expected to play a minor role in Europe and thus is not considered as benchmark waste treatment.

If landfilling is banned, *energy recovery* currently is the only option for plastic packaging waste that cannot be recycled mechanically. In principle, energy recovery can be performed by (1) incineration in municipal solid waste incinerators or (2) co-combustion with other fuels (Lazarevic et al., 2010). When assessing the environmental impacts of energy recovery, the energy production that is substituted has to be carefully assessed (Eriksson et al., 2007). In regions with cement industries, plastic packaging waste is often co-combusted in cement kilns to reduce lignite or hard coal utilization (Dehoust and Christiani, 2012). However, in countries were no cement industry exists, plastic packaging waste is incinerated in municipal solid waste incinerators for combined heat and power production (Eriksson and Finnveden, 2009). Here, we include both scenarios for energy recovery.

The first scenario substitutes coal, e.g. lignite, as fuel in cement kilns. This assumption is based on the large share of coal in cement plants' fuel supply of 70% in 2017 (IEA and CSI, 2018). For the future, even the most sustainable scenario of the International Energy Agency (IEA) predicts that in 2030, 55% of the fuel used in the cement industry is coal. Despite the important role of coal as fuel in the cement industry, alternative fuels are expected to increase: For 2030, the IEA predicts a share of 18% of biomass, waste and other renewable fuels as well as 12% of natural gas in the fuel mix of cement plants. To account for future fuels shifts in the cement industry, we consider two alternative scenario in which either natural gas or biomass, e.g. waste wood pellets are substituted as fuel in cement plants. In the second scenario, we assume that plastic packaging waste is incinerated in a municipal solid waste incinerator with an energy efficiency of 41 % and a power to heat ratio of 0.35. These values represent the average efficiencies for municipal solid waste incinerators in Europe (Eriksson and Finnveden, 2009). The products heat and electricity are assumed to substitute average European grid electricity and district heating (Ecoinvent, 2019).

Mechanical recycling currently plays a minor role in global waste treatment: today, only 16 % of plastic packing waste is recycled mechanically (Geyer et al., 2017). In contrast, a recent survey for Germany in 2015 showed that up to 73 % of collected and sorted plastic packaging waste was recycled mechanically (consultic, 2015). Thus, if plastic packaging waste is collected and sorted, it can be efficiently recycled. However, the recycled granulates cannot fully substitute virgin polymer granulates due to losses of product properties. To account for this downcycling, a so-called substitution factor is used frequently (BIO Intelligence Service, 2013; Michaud et al., 2010; Prognos AG, 2008). The substitution factor represents the amount of virgin plastic that can be substituted by 1 kg recycled plastic (e.g. technical quality measures). The substitution factor typically ranges from 0.7 for HDPE, LDPE, and PP to 1 for PET (BIO Intelligence Service, 2013; Michaud et al., 2010; Prognos AG, 2008).

2.5.2. *Chemical recycling*

Chemical recycling was recently discussed in excellent reviews (Clark et al., 2016; Hong and Chen, 2017; Lopez et al., 2017; Ragaert et al., 2017; Rahimi and García, 2017) from which we derived 4 categories for chemical recycling: (1) use as refinery feedstock, (2) fuel production, (3) monomer production and (4) chemical upcycling (cf. Table 1)[i]:

(1) **Refinery feedstock:** Plastic packaging wastes can be liquefied and substitute crude petroleum oil and intermediates, such as naphtha, in refinery and steam cracking processes. More precisely, polyolefins and PS can be utilized (Lopez et al., 2017), whereas PET results in large amounts of corrosive benzoic acid (up to 0.5 kg per kg PET). Benzoic acid can block pipes and heat exchangers (Hong and Chen, 2017) . Hence, we exclude PET as a refinery feedstock. We assume substitution on an equivalent mass basis. This assumption is justified by the small difference of carbon and hydrogen contents[ii] of polyolefins (C: 85.63 wt. %, H: 14.37 wt. %) and polystyrene (C: 92.26 wt. %, H: 7.74 wt. %) compared to petroleum oil (C: 85 wt. %, H: 12 wt. %)[12]. Due to the small difference, no substantial change of products from refinery units are expected by utilizing either polyolefins, PS or petroleum oil/naphtha.[13]

(2) **Fuel production:** Even though fuel production might not be considered a "full recycling", it is considered in the subsequent analysis, because at least the plastics energy content is re-used, the production of fossil fuels is avoided and many reviews of chemical recycling include fuel production. Fuel production decomposes the molecular structure of the polymer by gasification or pyrolysis and results in mixtures of gaseous fuels (C_1-$C_{5/6}$) (Honus et al., 2018a; Honus et al., 2018b) or liquid fuels (C_5-C_{20}) (Lopez et al., 2017). For gaseous fuels, yields of up to 99 % (Li et al., 1999) are reported for polyolefins, while PET and PS result in lower yields of 76.9 % and 9.9 %, respectively (Anuar Sharuddin et al., 2016). The low yields for PS are due to the aromatic fraction in PS. This aromatic fraction produces solids and liquids during gasification and thus, decreases gaseous fuel yields (Honus et al., 2018a; Honus et al., 2018b). For PET-derived gaseous fuels, gasification leads to high CO_2-concentrations which reduce net calorific values to only up to 9.7 MJ/kg (Honus et al., 2018a; Honus et al., 2018b) compared to natural gas with 50.4 MJ/kg (Ecoinvent, 2019). Thus, PET-derived gaseous fuels are excluded from our study. The considered gaseous fuels are expected to substitute natural gas based on an equal net calorific value. Liquid fuels can be produced from polyolefins with yields of up to 95.7 % (Zeaiter, 2014). For PET and PS, yields of 38.89 % and 96.73 % have been reported (Anuar Sharuddin et al., 2016). Liquid fuels are expected to substitute diesel and gasoline based on the net calorific value (Kalargaris et al., 2017).

(3) **Monomer production:** Polyolefins can be recycled back to monomers by thermal or catalytic pyrolysis. High yields of up to 75 % (Milne et al., 1999) can be achieved for conversion to ethylene and propylene by utilizing high temperatures above 800°C and short residence times or highly selective catalysts such as HZSM-5, HY and Hβ zeolites at 500°C (Elordi et al., 2009). For PET, many pathways have been proposed to produce feedstock monomers: e.g. hydrolysis, alcoholysis, acidolysis, or aminolysis (Carta et al., 2003; Paszun and Spychaj, 1997). Most pathways apply trans-

---

[i] Note that LDPE, HDPE and PP are collectively denoted polyolefins in the following.

[ii] Calculated based on chemical composition of raw materials ethylene, propylene and styrene.

esterification reactions with nucleophiles such as ethylene glycol or methanol, to produce terephthalic acid or dimethyl terephthalate together with ethylene glycol (Rahimi and García, 2017). Conversion rates of 90 % were reported for the production of both terephthalic acid and dimethyl terephthalate (Paszun and Spychaj, 1997). For PS, the most efficient chemical recycling technologies to monomers employ metal oxides such as MgO, BaO, CaO, or K_2O. The resulting yield to styrene is approx. 70 % (Zhang et al., 1995). The production of monomers is assumed to substitute an equal mass of monomers because chemically identical molecules are produced.

(4) **Chemical upcycling:** Plastic packaging waste can be converted to value-added chemicals by so-called chemical upcycling. Recent advances show that molecular ruthenium catalysts enable selective chemical upcycling of mixed plastic wastes (e.g. PET/PLA) to value-added chemicals, such as 1,4-benzenedimethanol and ethylene glycol (Westhues et al., 2018). Furthermore, aminolysis or glycolysis of PET can produce bis(2-hydroxyethyl) terephthalate (Paszun and Spychaj, 1997). Bis(2-hydroxyethyl) terephthalate and 1,4-benzenedimethanol are regarded as promising monomers for cyclohexane di-methanol (Guo et al., 2015; Westhues et al., 2018). Cyclohexane di-methanol is used in the polymer industry for value-added polyester fibers (Turner, 2004) and as polycarbonate substitute (Ritter, 2011). For polyolefins and PS, no chemical upcycling technologies have been so far proposed to the best of the authors' knowledge.

Table. 1
Overview of substituted chemicals, produced by chemical recycling of plastic packaging waste PET, PS, and polyolefins.

category	PET	PS	polyolefins (LD/HDPE + PP)
(1) refinery feedstock	excluded from study	crude petroleum oil	
(2) fuel production	gasoline and diesel	natural gas, gasoline, and diesel	
(3) monomer production	terephthalic acid or dimethanol terephthalate; both with ethylene glycol	Styrene	ethylene or propylene
(4) chemical upcycling	cyclohexane di-methanol and ethylene glycol	not available	

2.5.3. Data requirements and used datasets

All details about datasets can be found in the Supplementary Material. Waste compositions were obtained from industrial sorting facilities (DSD, 2018). For mechanical recycling, environmental impacts are based on current industry datasets from recycling practitioners from the late-year 2017 (BIO Intelligence Service, 2013; HTP GmbH & Co. KG, 2017; Prognos AG, 2008). For energy recovery, we calculated the environmental impacts according to Doka (2003, 2013). Modeling principles and assumptions, as well as thermodynamic properties required to calculate the environmental impacts for chemical recycling, are summarized in Sections S7 to S9 of the Supplementary Material. All background datasets are obtained from LCA databases for supplied materials (e.g. reactants) and energy carriers (e.g. heat and electricity) and avoided conventional products, except for lignite, diesel, and gasoline (Ecoinvent, 2019; thinkstep AG, 2019).

3. Results and discussion

3.1. Global warming impacts and fossil resource depletion

Results are discussed for each benchmark waste treatment separately. For fossil resource depletion, the environmental potential is almost always positive. Thus, most chemical recycling technologies have the potential to reduce the use of fossil resources. Therefore, the following discussion focuses on global warming impacts if not mentioned otherwise.

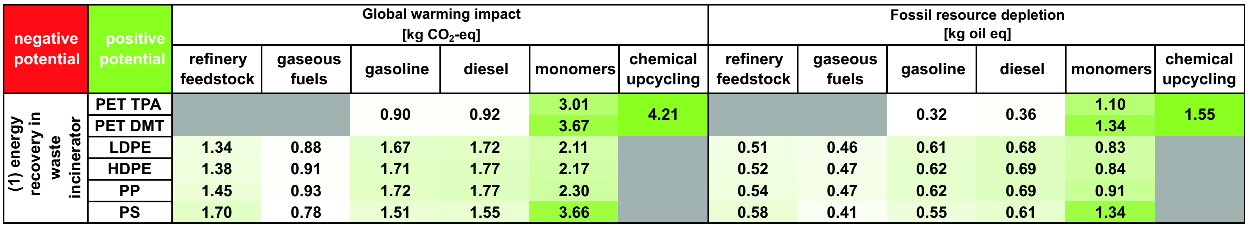
3.1.1. Chemical recycling vs. energy recovery in municipal solid waste incinerators

All chemical recycling technologies have a positive environmental potential compared to energy recovery in waste incinerators (see Fig. 2). The environmental potential for global warming ranges from 0.78 kg CO_2-eq for gaseous fuels from PS to 4.21 kg CO_2-eq for chemical upcycling of PET to cyclohexane di-methanol. Chemical recycling could potentially reduce global warming, because municipal solid waste incinerators receive low credits for electricity and heat production. Credits for global warming impacts are low since large amounts of electricity in Europe are already produced from renewables. Furthermore, district heating is mainly based on natural gas which represents the least harmful fossil source of heat (Connolly et al., 2014). In the near-term future, electricity production and district heating will be further decarbonized and thus the environmental impacts avoided by municipal solid waste incinerators will further decrease. This trend will further increase the environmental potential for all chemical recycling technologies compared to municipal waste incinerators.

3.1.2. Chemical recycling vs. energy recovery in cement kilns

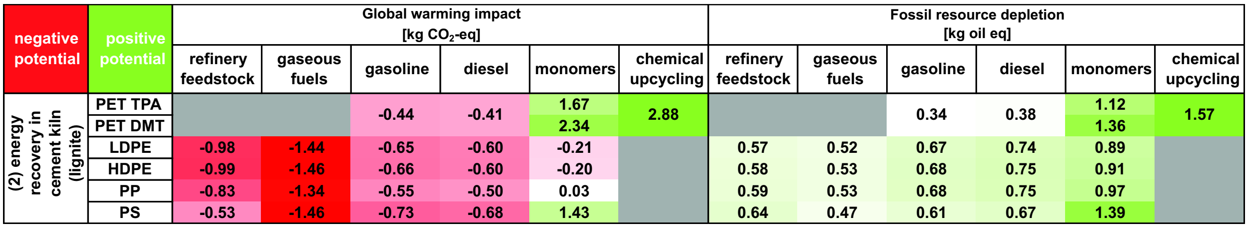
Environmental potentials for global warming are less homogeneous for the comparison with energy recovery in cement kilns (see Fig. 3). Note that, in cement kilns, plastic packaging waste is assumed to substitute lignite, if not stated otherwise. The results differ for two groups of chemical recycling: (1) *refinery feedstock* and *fuel* production and (2) *monomer* production and *chemical upcycling*.

Recycling plastic packaging waste to *refinery feedstock* and *fuels* has negative environmental potentials ranging from -1.46 kg CO_2-eq for gaseous fuels from HDPE to -0.44 kg CO_2-eq for liquid fuel production



negative potential	positive potential	Global warming impact [kg CO_2-eq]						Fossil resource depletion [kg oil eq]					
		refinery feedstock	gaseous fuels	gasoline	diesel	monomers	chemical upcycling	refinery feedstock	gaseous fuels	gasoline	diesel	monomers	chemical upcycling
(1) energy recovery in waste incinerator	PET TPA			0.90	0.92	3.01	4.21			0.32	0.36	1.10	1.55
	PET DMT					3.67						1.34	
	LDPE	1.34	0.88	1.67	1.72	2.11		0.51	0.46	0.61	0.68	0.83	
	HDPE	1.38	0.91	1.71	1.77	2.17		0.52	0.47	0.62	0.69	0.84	
	PP	1.45	0.93	1.72	1.77	2.30		0.54	0.47	0.62	0.69	0.91	
	PS	1.70	0.78	1.51	1.55	3.66		0.58	0.41	0.55	0.61	1.34	

Fig. 2. Environmental potential for global warming impacts and fossil resource depletion of chemical recycling compared to energy recovery in municipal solid waste incinerators. Red indicates negative environmental potentials. Green indicates positive environmental potentials. White indicates values equal to zero. Grey indicates that chemical recycling does not exist or has been omitted. PET can be used to produce ethylene glycol and two types of monomers: terephthalic acid (PET TPA) and dimethyl terephthalate (PET DMT).



negative potential	positive potential	Global warming impact [kg CO_2-eq]						Fossil resource depletion [kg oil eq]					
		refinery feedstock	gaseous fuels	gasoline	diesel	monomers	chemical upcycling	refinery feedstock	gaseous fuels	gasoline	diesel	monomers	chemical upcycling
(2) energy recovery in cement kiln (lignite)	PET TPA			-0.44	-0.41	1.67	2.88			0.34	0.38	1.12	1.57
	PET DMT					2.34						1.36	
	LDPE	-0.98	-1.44	-0.65	-0.60	-0.21		0.57	0.52	0.67	0.74	0.89	
	HDPE	-0.99	-1.46	-0.66	-0.60	-0.20		0.58	0.53	0.68	0.75	0.91	
	PP	-0.83	-1.34	-0.55	-0.50	0.03		0.59	0.53	0.68	0.75	0.97	
	PS	-0.53	-1.46	-0.73	-0.68	1.43		0.64	0.47	0.61	0.67	1.39	

Fig. 3. Environmental potential for global warming impacts and fossil resource depletion of chemical recycling compared to energy recovery in cement kilns. Red indicates negative environmental potentials. Green indicates positive environmental potentials. White indicates values equal to zero. Grey indicates that chemical recycling does not exist or has been omitted. PET can be used to produce ethylene glycol and two types of monomers: terephthalic acid (PET TPA) and dimethyl terephthalate (PET DMT).

from PET. Negative environmental potentials result from two effects: (1) using plastic packaging waste as fuel in cement kiln results in high credits from substituting lignite and (2) chemical recycling of plastic packaging waste to refinery feedstock or fuels results in low credits from substituting fossil products. Thus, the substitution of lignite is favorable over the production of refinery feedstock or liquid and gaseous fuels.

Monomer production results in negative environmental potentials for HDPE and LDPE, a small positive potential for PP and positive environmental potentials for PET and PS. The results are based on two opposite effects: (1) the higher net calorific values and thus higher credit for lignite substitution of HDPE, LDPE, PP and PS compared to PET and (2) the lower credits for monomer production from HDPE, LDPE and PP compared to PET and PS.

Chemical recycling of HDPE and LDPE achieves a low credit for the global warming impacts avoided from the conventional production of ethylene. At the same time, high calorific HDPE and LDPE must be compensated by lignite in cement kilns resulting in higher emissions. As a result, environmental potentials are negative. For PP, the increase in avoided global warming impacts in cement kilns is slightly less than for HDPE and LDPE. Thus, chemical recycling to propylene has a small positive environmental potential. For PS, the global warming impacts avoided from conventional styrene production are higher than for HDPE, LDPE, and PP and thus, outweigh the increase of cement kiln impacts due to increased lignite utilization. Thus, monomer production from PS has a positive environmental potential.

For PET, low net calorific values lead to small avoided global warming impacts from lignite utilization. At the same time, credits are high for replacing conventional terephthalic acid/dimethyl terephthalate production. Thus, the environmental potential of monomer production from PET is positive. Furthermore, the environmental potential for PET reaches up to 2.88 kg CO_2-eq if cyclohexane di-methanol is produced, due to very high avoided global warming impacts of conventional cyclohexane di-methanol production.

From the perspective of global warming, energy recovery in cement kilns is superior to chemical recycling to refinery feedstock and fuels for all polymer types if lignite can be substituted in cement kilns. Thus, sorted plastic packaging waste should rather be used as a substitute for lignite in cement plants than chemically recycled to refinery feedstock or fuel products. For LDPE and HDPE, energy recovery in cement kilns is even preferable to ideal chemical recycling to monomers. In contrast, PET and PS otherwise being used in cement kilns seem promising candidates for monomer production and chemical upcycling.

In contrast to lignite usage, chemical recycling seems more promising if biomass or natural gas would be replacing plastic packaging as fuel in cement kilns (cf. Figure S7 in the Supplementary Material). The only chemical recycling technology still resulting in negative environmental potentials for both global warming impacts and fossil resource depletion is recycling to gaseous fuels due to the low credit for substituting natural gas.

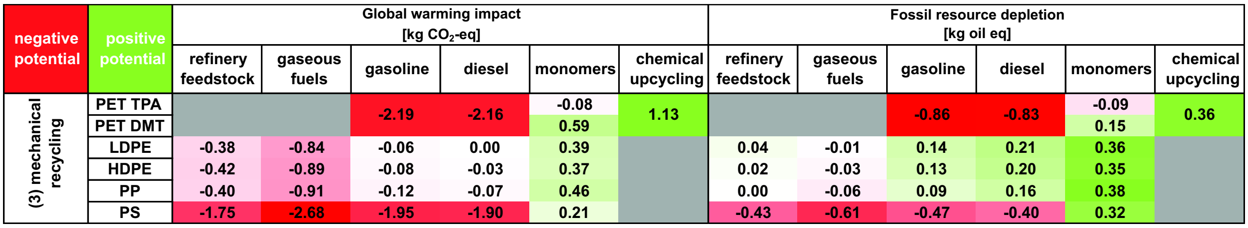
For all other chemical recycling technologies except the production of gaseous fuels, the environmental potentials range from 0.35 kg CO_2-eq for diesel from PET to 3.77 kg CO_2-eq for chemical upcycling of PET to cyclohexane di-methanol if plastic packaging waste replaces natural gas as fuel. If plastic packaging waste would replace biomass, positive environmental potentials reach up to 5.06 kg CO_2-eq for chemical upcycling of PET to cyclohexane di-methanol. The positive environmental potentials are based on the fact that natural gas and biomass are less emission-intensive fuels than plastic packaging waste. Thus, plastic packaging waste should rather be used for chemical recycling than to substitute natural gas or biomass in cement kilns.

3.1.3. Chemical recycling vs. mechanical recycling

The comparison between chemical recycling and mechanical recycling (see Fig. 4) can again be subdivided into two groups: (1) refinery feedstock and fuel production with negative environmental potentials and (2) monomer production and chemical upcycling with positive environmental potentials.

The results are mainly based on high global warming impacts avoided by mechanical recycling. To compete with mechanical recycling, chemical recycling must avoid high global warming impacts from conventional chemical production.

Chemical recycling to *refinery feedstock and gaseous fuels* avoids only small global warming impacts to produce crude petroleum oil/naphtha and natural gas, respectively. For chemical recycling to *liquid fuels*, environmental potentials are mostly negative, but differ strongly for PET and PS on the one hand and polyolefins on the other hand. Environmental potentials of PET (-2.19 kg CO_2-eq) and PS (-1.95 kg



negative potential	positive potential	Global warming impact [kg CO_2-eq]						Fossil resource depletion [kg oil eq]					
		refinery feedstock	gaseous fuels	gasoline	diesel	monomers	chemical upcycling	refinery feedstock	gaseous fuels	gasoline	diesel	monomers	chemical upcycling
(3) mechanical recycling	PET TPA			-2.19	-2.16	-0.08	1.13			-0.86	-0.83	-0.09	0.36
	PET DMT					0.59						0.15	
	LDPE	-0.38	-0.84	-0.06	0.00	0.39		0.04	-0.01	0.14	0.21	0.36	
	HDPE	-0.42	-0.89	-0.08	-0.03	0.37		0.02	-0.03	0.13	0.20	0.35	
	PP	-0.40	-0.91	-0.12	-0.07	0.46		0.00	-0.06	0.09	0.16	0.38	
	PS	-1.75	-2.68	-1.95	-1.90	0.21		-0.43	-0.61	-0.47	-0.40	0.32	

Fig. 4. Environmental potential for global warming impacts and fossil resource depletion of chemical recycling compared to mechanical recycling. Red indicates negative environmental potentials. Green indicates positive environmental potentials. White indicates values equal to zero. Grey indicates that chemical recycling does not exist or has been omitted. PET can be used to produce ethylene glycol and two types of monomers: terephthalic acid (PET TPA) and dimethyl terephthalate (PET DMT).

CO_2-eq) are clearly negative while the potentials for polyolefins are close to zero (e.g. 0.01 kg CO_2-eq for HDPE). Mechanical recycling of PET and PS avoids higher global warming impacts by substituting virgin polymer. In contrast, for LDPE, HDPE, and PP, mechanical recycling suffers from significant downcycling (Prognos AG, 2008) as reflected by the substitution factor of 0.7.

Monomer production from plastic packaging waste results mostly in positive environmental potentials if compared to mechanical recycling because substituting the conventional monomers avoids higher global warming impacts. Here, PET achieves the highest environmental potential, if dimethyl terephthalate and ethylene glycol (denoted PET DMT in Fig. 1) are produced.

For LDPE, HDPE, and PP, the global warming impact of avoided monomers is lower but still sufficient for positive environmental potentials, because downcycling during mechanical recycling reduces the savings in global warming impacts from virgin polymer production.

C*hemical upcycling* of PET packaging waste increases the environmental potential of up to 1.13 kg CO_2-eq per kg of plastic packaging waste.

In conclusion, in terms of global warming impacts, mechanical recycling is advantageous to chemical recycling for all plastic packaging wastes, if refinery feedstock and fuels are produced. Thus, mechanical recycling should be preferred if possible. In contrast, monomer production and chemical upcycling offer positive environmental potentials if the waste would otherwise be used for mechanical recycling.

For fossil resource depletion, the results slightly differ from global warming: Polyolefins have positive environmental potentials even for chemical recycling to refinery feedstock as well as gaseous and liquid fuels. For polyolefins, the environmental potentials for fossil resource depletion are up to 0.38 kg oil-eq for monomer production. In contrast, for PET, fuel production avoids little fossil resource depletion due to lower net calorific values of PET-derived fuels and thus, result in negative environmental potentials. Only the monomer production of dimethyl terephthalate and chemical upcycling of PET achieves positive environmental potentials for fossil resource depletion. For PS, qualitatively similar results are obtained for fossil resource depletion and global warming impacts.

3.2. Other environmental impacts

Besides global warming and fossil resource depletion, the environmental impacts most commonly assessed for plastic waste management are terrestrial acidification and marine/freshwater eutrophication. (Lazarevic et al., 2010) As for global warming and fossil resource depletion, we identify chemical recycling routes with positive and with negative environmental potentials for acidification and eutrophication. With respect to the specific routes, the findings for acidification and eutrophication differ from the results obtained for global warming impacts and fossil resource depletion indicating potential trade-off in environmental impacts. For instance, chemical recycling of waste currently used in municipal waste incinerators increases terrestrial acidification and freshwater/marine eutrophication impacts if refinery feedstock or fuels are produced. In contrast, all chemical recycling routes have the potential reduce terrestrial acidification if using plastic packaging waste currently used for energy recovery in cement kilns (cf. section S15 of the Supplementary Material for a detailed result presentation).

3.3. Environmental potentials vs. environmental benefits

It is important to emphasize that chemical recycling technologies with positive environmental potentials will not necessarily result in environmental benefits in real-case LCA assessments (cf. section 2.4). Additional environmental impacts are generated, for instance, by the separation and purification of chemical products (e.g. TPA and EG), lower conversion rates or additional compounds that must be heated or separated during or before chemical recycling. To understand the influence of additional environmental impacts, we performed a sensitivity analysis for the thermal energy demand and conversion rates of chemical recycling. We focus on monomer production and chemical upcycling because these achieve positive environmental potentials compared to all benchmark waste treatments (other routes see section S12 of the Supplementary Material).

The energy demand is varied between the minimum energy demand and 4 MJ per kg of treated plastic packaging waste. The proxy of 4 MJ represents the maximum energy demand that has been reported for 65 production processes for organic chemicals (Kim and Overcash, 2003). Conversion rates are varied between 0.7 and 1. The value of 0.7 represents the highest reported conversion rate for PS using metal oxides (Zhang et al., 1995).

Compared to *energy recovery in municipal solid waste incinerators*, chemical recycling has still positive environmental potentials for both global warming impact and fossil resource depletion over the complete range of the sensitivity study (cf. Fig. S8 and S9 in the Supplementary Material). Thus, plastic packaging waste should most likely not be used in municipal solid waste incinerators but rather be recycled chemically.

For *energy recovery in cement kilns*, monomer production and chemical upcycling of PET and PS achieve positive environmental potentials for both global warming impacts and fossil resource depletion over the complete parameter range (cf. Figure S8 and S9 in the Supplementary Material). Thus, PET and PS should rather be used for monomer production or chemical upcycling than for any energy recovery option. In contrast to PET and PS, the environmental potential for global warming impacts of PP turns negative if 0.39 MJ additional thermal energy is required, or the conversion rate decreases to 98.5 %. Thus, monomer production of PP probably will not achieve benefits regarding global warming impacts compared to energy recovery in cement kilns in a real-case LCA.

The comparison of chemical recycling to monomers and *mechanical recycling* reveals one major challenge (Fig. 5A): conversion rates have to be higher than a minimum value to achieve any environmental potential for global warming impacts. These minimum conversion rates range from 0.84 for PP to 0.91 for PS (Table 2 and Fig. 5).

The minimal conversion rates of polyolefins and PS are between 21% and 9% higher than the highest reported conversion rates of monomer production. Solely for PET, conversion rates are reported that are approx. 5% higher than the minimal conversion rates. To increase conversion rates, a separation step could be added after the reactor to recycle unconverted polymer back to the reactor. This separation step, however, would require additional process energy (Fig. 5B). Additional thermal energy would again lower the environmental potential of monomer production compared to mechanical recycling and thus, increase the minimal conversion rates of chemical recycling: Each additional megajoule of thermal energy would increase the minimal process yield by approx. 3% on average for all plastic packaging waste types. In conclusion, monomer production from plastic packaging waste that could be used in mechanical recycling represents a challenging task under more realistic conditions, and further improvements in chemical recycling technologies are needed. Such improvements should increase conversion rates of chemical recycling as well as energy efficiency.

3.4. Comparison of results with previous LCA studies

The results of the LCA-study and the sensitivity analysis show the challenge for chemical recycling to compete with the real-case benchmark waste treatment technologies, except waste incineration. Even under best-case conditions (e.g. with minimal environmental impacts), some chemical recycling technologies have higher global warming impacts than their benchmark waste treatments. These results are in line with previous LCA studies for chemical recycling:

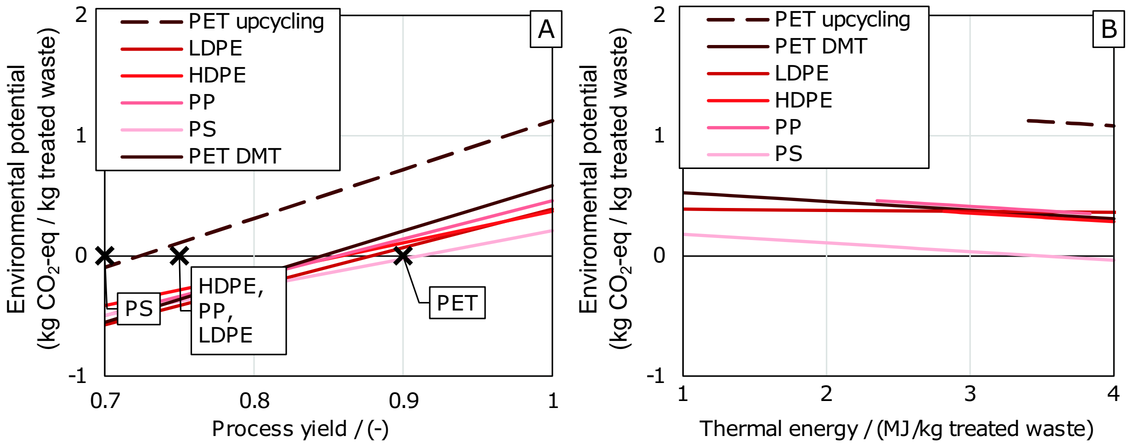


Fig. 5. Environmental potential for global warming of chemical recycling to monomers and PET upcycling in comparison to mechanical recycling. The values for the environmental potential (y-axis) depend on the process yield (left) or thermal energy demands of chemical recycling (right) (x-axis). The marked values (x) represent the highest reported values of conversion rates for monomer production by chemical recycling. Terephthalic acid production from PET is denoted PET TPA, while dimethyl terephthalate production is denoted PET DMT.

Table. 2
Minimal conversion rates and highest reported values of conversion rates for monomer production of chemical recycling.

parameter	HDPE	LDPE	PP	PET DMT	PS
minimal conversion rates	0.86	0.88	0.856	0.85	0.91
highest reported conversion rates	0.75	0.75	0.75	0.90	0.70

HDPE, LDPE, PP: (Milne et al., 1999); PET DMT: (Paszun and Spychaj, 1997); PS: (Zhang et al., 1995)

In 2000, an LCA-study by Patel et al. (2000) found for the treatment of mixed plastic waste that energy recovery in cement kilns as well as mechanical or chemical recycling reduces CO_2-emission compared to municipal waste incineration. Furthermore, their study suggests that substituting coal, a very carbon-intensive fuel, in cement kilns is advantageous to the substitution of natural gas in other industries. This finding is in line with the present work (cf. sections 3.2 and S11). Results of Perugini et al. (2005) further confirm that, for the case of PET waste, the combination of mechanical and chemical recycling is beneficial to energy recovery in waste incinerators. They show that mechanical recycling has lower environmental impacts than the combination of mechanical and chemical recycling. The results of these two studies have been confirmed by Lazarevic et al. (2010) in an extensive review of LCA-studies on plastic waste management scenarios in the context of a future European recycling society. In that review, 77 waste management scenarios were analyzed including mechanical and chemical recycling but also waste incineration and energy recovery in cement kilns. The review shows a clear preference of mechanical recycling and energy recovery in cement kilns over chemical recycling: for global warming impacts, all scenarios favored mechanical recycling and energy recovery in cement kilns. However, chemical recycling reduced global warming impacts compared to municipal waste incinerators in all scenarios. More recent studies by Maga et al., 2019 underline the benefits of chemical recycling of packaging waste over waste incinerators even for new polymer materials like PLA. In contrast to already available literature, this publication contributes by providing updated inventories for benchmark waste treatments and a comprehensive assessment of 5 major plastic waste streams based on a consistent LCA methodology applicable to all chemical recycling technologies. This consistent methodology allows the direct comparison across plastic waste streams and furthermore, to derive robust conclusions about the potential environmental benefits of chemical recycling technologies.

4. Conclusions

Recently, chemical recycling technologies have been advocated as enabling technologies for a transition to a circular economy for plastic packaging wastes. To determine whether chemical recycling could lead to environmental benefits, we introduce a consistent LCA-based method that calculates the maximal environmental benefits by chemical recycling technologies, denoted the *environmental potential of chemical recycling*. This method allows us to study the 5 major plastic packaging wastes PET, HDPE, LDPE, PP and PS and all currently discussed chemical recycling routes even at early stages of development.

Our results suggest that all chemical recycling pathways could reduce global warming impacts and fossil resource depletion if using sorted plastic packaging wastes otherwise treated in municipal solid waste incinerators. Current waste incinerators suffer from high emissions and low efficiencies in producing heat and electricity. The highest potential to reduce global warming impacts and fossil resource depletion is achieved for chemical upcycling of PET to cyclohexane di-methanol instead of its energy recovery in municipal solid waste incinerators: ideal chemical upcycling could avoid up to 4.2 kg CO_2-eq and 1.4 kg oil-eq per 1 kg of treated PET waste. Our sensitivity analysis reveals that global warming impacts and fossil resource depletion can still be reduced even for low conversion rates of 70% and high energy demands.

In contrast to using sorted plastic waste treated in municipal solid waste incinerators, plastic waste currently used for energy recovery in cement kilns or mechanical recycling needs a more careful analysis: Energy recovery in cement kilns and mechanical recycling avoids the use of lignite and the production of virgin polymers, respectively. If the objective is to reduce global warming impacts, sorted plastic packaging waste used in cement kilns or mechanical recycling should therefore not be converted to refinery feedstock or fuels. Both, have much lower credits for global warming impacts than substituting lignite or virgin polymers. However, if natural gas or biomass, e.g. waste wood, is used in cement kilns instead of plastic packaging waste, chemical recycling to refinery feedstock and fuels has the potential to reduce environmental impacts.

In contrast to refinery feedstock and fuels, chemical recycling of plastic packaging wastes could reduce global warming impacts and fossil resource depletion for most routes to monomers and for upcycling to value-added chemicals. However, a sensitivity analysis shows that environmental benefits regarding global warming impacts of chemical recycling to monomers would still require very energy-efficient processes and increased conversion rates compared to the current state of the art.

Considering terrestrial acidification as well as freshwater and marine eutrophication highlights the need to assess several environmental impacts to understand the full environmental consequences of implementing chemical recycling. For instance, chemical recycling of waste currently used in municipal waste incinerators increases terrestrial acidification and freshwater/marine eutrophication impacts if refinery feedstock or fuels are produced. In contrast, all chemical recycling routes have the potential to reduce terrestrial acidification if recycling plastic packaging waste currently used for energy recovery in cement kilns (cf. section S15 of the Supplementary Material). Even though trends often differ for the 5 assessed environmental impacts, some general conclusions can be drawn: in particular, (1) PET and PS treated in municipal waste incinerators should preferably be used for monomer production and chemical upcycling as all environmental impacts could potentially be reduced; in contrast, (2) PET and PS currently recycled mechanically should not be used for refinery feedstock or fuel production as all environmental impacts would increase. All other chemical recycling pathways show trade-offs between environmental impacts. The results clearly highlight that choosing a technology solely to reduce global warming impacts might increase other environmental impacts. However, as an overall trend, chemical recycling to monomers and chemical upcycling to value-added products tends to have the potential to reduce more environmental impacts than recycling to refinery feedstock and fuels.

Our study analyzed 75 waste treatment scenarios representing approximately 50 % of the global plastic waste. The analysis allows to exclude pathways offering no potential to reduce global warming impacts and to match waste types to chemical recycling technologies offering potential benefits. In line with previous LCA studies, our results highlight that it will be very challenging for chemical recycling to compete with real-case benchmark waste treatments if climate impacts should be reduced. Thus, we provide stakeholders of chemical recycling with the possibility to identify the most promising pathways at an early-stage of development based on a consistent LCA methodology. However, full-LCA studies are needed for all promising pathways, because previous LCA studies highlighted that it will be very challenging for chemical recycling to compete with energy recovery in cement kilns and mechanical recycling under more realistic conditions.

CRediT authorship contribution statement

Raoul Meys: Conceptualization, Methodology, Investigation, Data curation, Writing - original draft, Visualization, Validation, Writing - review & editing. **Felicitas Frick:** Investigation, Data curation. **Stefan Westhues:** Conceptualization. **André Sternberg:** Conceptualization, Writing - review & editing. **Jürgen Klankermayer:** Conceptualization. **André Bardow:** Conceptualization, Methodology, Writing - original draft, Supervision.

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.

Acknowledgements

A.B., A.S. and R.M. would like to acknowledge funding by the German Federal Ministry of Education and Research (BMBF project 033R150D).

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2020.105010.

References

Anuar Sharuddin, SD, Abnisa, F, Wan Daud, WMA, Aroua, MK, 2016. A review on pyrolysis of plastic wastes. Energy Convers. Manage. 115, 308–326. https://doi.org/10.1016/j.enconman.2016.02.037.

Baumann H, Tillman A-M. The hitch hikers's guide to LCA: An orientation in life cycle assessment methodology and application. Lund: Studentlitteratur; 2004.

BIO Intelligence Service. Study on an increased mechanical recycling target for plastics: Final report prepared for Plastics Recyclers Europe 2013.

Carta, D, Cao, G, D'Angeli, C, 2003. Chemical recycling of poly(ethylene terephthalate) (PET) by hydrolysis and glycolysis. Environ. Sci. Pollut. Res. Int. 10 (6), 390–394. https://doi.org/10.1065/espr2001.12.104.8.

Clark, JH, Farmer, TJ, Herrero-Davila, L, Sherwood, J, 2016. Circular economy design considerations for research and process development in the chemical sciences. Green Chem 18 (14), 3914–3934. https://doi.org/10.1039/C6GC00501B.

Connolly, D, Lund, H, Mathiesen, BV, Werner, S, Möller, B, Persson, U, et al., 2014. Heat Roadmap Europe: combining district heating with heat savings to decarbonise the EU energy system. Energy Policy 65, 475–489. https://doi.org/10.1016/j.enpol.2013.10.035.

consultic. Produktion, Verarbeitung und Verwertung von Kunststoffen in Deutschland 2015 - Kurzfassung [In German]; 2015.

Dehoust G, Christiani J. Analyse und Fortentwicklung der Verwertungsquoten für Wertstoffe (In German). Sammel- und Verwertungsquoten für Verpackungen und stoffgleiche Nichtverpackungen als Lenkungsinstrument zur Ressourcenschonung; 2012.

Doka G.Life cycle inventories of waste treatment services: ecoinvent report No. 13. Dübendorf; 2003.

Doka G.Updates to life cycle inventories of waste treatment services - part ii: waste incineration. Zurich; 2013.

DSD. Spezifikation der Wertstofffraktion (in German); 2018.

Ecoinvent. ecoinvent Data V. 3.5: Swiss Centre for Life Cycle Inventories, 2019.

Elordi, G, Olazar, M, Lopez, G, Amutio, M, Artetxe, M, Aguado, R, et al., 2009. Catalytic pyrolysis of HDPE in continuous mode over zeolite catalysts in a conical spouted bed reactor. J. Anal. Appl. Pyrolysis 85 (1-2), 345–351. https://doi.org/10.1016/j.jaap.2008.10.015.

Eriksson, O, Carlsson Reich, M, Frostell, B, Björklund, A, Assefa, G, Sundqvist, J-O, et al., 2005. Municipal solid waste management from a systems perspective. J. Cleaner Prod. 13 (3), 241–252. https://doi.org/10.1016/j.jclepro.2004.02.018.

Eriksson, O, Finnveden, G., 2009. Plastic waste as a fuel - CO2-neutral or not? Energy Environ. Sci. 2 (9), 907. https://doi.org/10.1039/b908135f.

Eriksson, O, Finnveden, G., 2017. Energy recovery from waste incineration—the importance of technology data and system boundaries on CO2 emissions. energies 10 (4), 539. https://doi.org/10.3390/en10040539.

Eriksson, O, Finnveden, G, Ekvall, T, Björklund, A, 2007. Life cycle assessment of fuels for district heating: a comparison of waste incineration, biomass- and natural gas combustion. Energy Policy 35 (2), 1346–1362. https://doi.org/10.1016/j.enpol.2006.04.005.

EU Commission. Directive (EU)2018/852 of the EP and of the Council of 30 may 2018 amending directive 94/62/ec on packaging and packaging waste, OJ L 150, 14.6. 2018, pp. 141–154; 2018.

European Comission. GREEN PAPER: On a European Strategy on Plastic Waste in the Environment 2013.

European Comission. Closing the loop - An EU action plan for the Circular Economy 2015.

Geyer, R, Jambeck, JR, Law, KL, 2017. Production, use, and fate of all plastics ever made. Sci. Adv. 3 (7), e1700782. https://doi.org/10.1126/sciadv.1700782.

Geyer, R, Kuczenski, B, Zink, T, Henderson, A, 2016. Common misconceptions about recycling. J. Ind. Ecol. 20 (5), 1010–1017. https://doi.org/10.1111/jiec.12355.

Guo, X, Xin, J, Lu, X, Ren, B, Zhang, S, 2015. Preparation of 1,4-cyclohexanedimethanol by selective hydrogenation of a waste PET monomer bis(2-hydroxyethylene terephthalate). RSC Adv 5 (1), 485–492. https://doi.org/10.1039/C4RA10783G.

Hong, M, Chen, EY-X., 2017. Chemically recyclable polymers: a circular economy approach to sustainability. Green Chem. 19 (16), 3692–3706. https://doi.org/10.1039/C7GC01496A.

Honus, S, Kumagai, S, Fedorko, G, Molnár, V, Yoshioka, T, 2018a. Pyrolysis gases produced from individual and mixed PE, PP, PS, PVC, and PET—part i: production and physical properties. Fuel 221, 346–360. https://doi.org/10.1016/j.fuel.2018.02.074.

Honus, S, Kumagai, S, Molnár, V, Fedorko, G, Yoshioka, T, 2018b. Pyrolysis gases produced from individual and mixed PE, PP, PS, PVC, and PET—Part II: Fuel characteristics. Fuel 221, 361–373. https://doi.org/10.1016/j.fuel.2018.02.075.

HTP GmbH & Co. KG. Confidential notification based on internal project reports 2017.

IEA, CSI. Technology Roadmap Low-Carbon Transition in the Cement Industry. Paris; 2018.

ISO 14040. Environmental management - Life cycle assessment - Principles and framework. Brussels, 2006.

ISO 14044. Environmental management - Life cycle assessment - Requirements and guidlines. Brussels, 2006.

Kalargaris, I, Tian, G, Gu, S, 2017. The utilisation of oils produced from plastic waste at different pyrolysis temperatures in a DI diesel engine. Energy 131, 179–185. https://doi.org/10.1016/j.energy.2017.05.024.

Kim, S, Overcash, M., 2003. Energy in chemical manufacturing processes: gate-to-gate information for life cycle assessment. J. Chem. Technol. Biotechnol. 78 (9), 995–1005. https://doi.org/10.1002/jctb.821.

Laurent, A, Bakas, I, Clavreul, J, Bernstad, A, Niero, M, Gentil, E, et al., 2014. Review of LCA studies of solid waste management systems–part I: lessons learned and perspectives. Waste Manage. (Oxford) 34 (3), 573–588. https://doi.org/10.1016/j.

wasman.2013.10.045. New York, N.Y.

Lazarevic, D, Aoustin, E, Buclet, N, Brandt, N, 2010. Plastic waste management in the context of a European recycling society: Comparing results and uncertainties in a life cycle perspective. Resour. Conserv. Recycl. 55 (2), 246–259. https://doi.org/10.1016/j.resconrec.2010.09.014.

Li, AM, Li, XD, Li, SQ, Ren, Y, Shang, N, Chi, Y, et al., 1999. Experimental studies on municipal solid waste pyrolysis in a laboratory-scale rotary kiln. Energy 24 (3), 209–218. https://doi.org/10.1016/S0360-5442(98)00095-4.

Lopez, G, Artetxe, M, Amutio, M, Bilbao, J, Olazar, M, 2017. Thermochemical routes for the valorization of waste polyolefinic plastics to produce fuels and chemicals. A review. Renewable and Sustainable Energy Rev. 73, 346–368. https://doi.org/10.1016/j.rser.2017.01.142.

Maga, D., Hiebel, M., Thonemann, N., 2019. Life cycle assessment of recycling options for polylactic acid. Resources, Conservation and Recycling 149, 86–96. https://doi.org/10.1016/j.resconrec.2019.05.018.

McDowall, W, Geng, Y, Huang, B, Barteková, E, Bleischwitz, R, Türkeli, S, et al., 2017. Circular Economy Policies in China and Europe. J. Ind. Ecol. 21 (3), 651–661. https://doi.org/10.1111/jiec.12597.

Michaud J-C, Farrant L, Jan O, Kjær B, Bakas I. Environmental benefits of recycling – 2010 update; 2010.

Milne, BJ, Behie, LA, Berruti, F, 1999. Recycling of waste plastics by ultrapyrolysis using an internally circulating fluidized bed reactor. J. Anal. Appl. Pyrolysis 51 (1-2), 157–166. https://doi.org/10.1016/S0165-2370(99)00014-5.

Sustain, Nat, 2018. Closing the plastics loop. Nat. Sustain. 1 (5), 205. https://doi.org/10.1038/s41893-018-0075-3.

Paszun, D, Spychaj, T., 1997. Chemical recycling of poly(ethylene terephthalate). Ind. Eng. Chem. Res. 36 (4), 1373–1383. https://doi.org/10.1021/ie960563c.

PlasticsEurope, editor. Plastics – the Facts 2018. An analysis of European plastics production, demand and waste data; 2018.

Prognos AG. Resource savings and CO2 reduction potential in waste management in Europe and the possible contribution to the CO2 reduction target in 2020. 2008.

Patel, M., Thienen, N.v., Jochem, E., Worrell, E., 2000. Recycling of plastics in Germany. Resources, Conservation and Recycling 29 (1–2), 65–90. https://doi.org/10.1016/S0921-3449(99)00058-0.

Perugini, F., Mastellone, M.L., Arena, U., 2005. A life cycle assessment of mechanical and feedstock recycling options for management of plastic packaging wastes. Environ. Prog. 24 (2), 137–154. https://doi.org/10.1002/ep.10078.

Ragaert, K, Delva, L, van Geem, K, 2017. Mechanical and chemical recycling of solid plastic waste. Waste Manage. (Oxford) 69, 24–58. https://doi.org/10.1016/j.wasman.2017.07.044. New York, N.Y.

Rahimi, A, García, JM., 2017. Chemical recycling of waste plastics for new materials production. Nat. Rev. Chem. 1 (6), 409. https://doi.org/10.1038/s41570-017-0046.

Reichel A, Schoenmakere M, Gillabel J. Circular economy in Europe: Developing the knowledge base. Luxembourg: Publications Office of the European Union; 2016.

Ritter, SK, 2011. BPA Is Indispensible For Making Plastics. Chem. Eng. News 89 (23). https://cen.acs.org/articles/89/i23/BPA-Indispensible-Making-Plastics.html

Shen, Li, Worrell, Ernst, Patel, Martin K, 2010. Open-loop recycling: A LCA case study of PET bottle-to-fibre recycling. Resour. Conserv. Recycl. 55 (0), 34–52. https://doi.org/10.1016/j.resconrec.2010.06.014.

The REMADE Institute, 2019. https://remadeinstitute.org/ (accessed May 03, 2019).

thinkstep, AG., 2019. Software-System and Database for Life Cycle Engineering. Leinfelden-Echterdingen, Germany. https://sphera.com/life-cycle-assessment-software/

Turner, SR., 2004. Development of amorphous copolyesters based on 1,4-cyclohexanedimethanol. J. Polym. Sci. A Polym. Chem. 42 (23), 5847–5852. https://doi.org/10.1002/pola.20460.

Westhues, S, Idel, J, Klankermayer, J, 2018. Molecular catalyst systems as key enablers for tailored polyesters and polycarbonate recycling concepts. Sci. Adv. 4 (8). https://doi.org/10.1126/sciadv.aat9669. eaat9669.

World Economic Forum, Ellen MacArthur Foundation. The New Plastics Economy: Catalysing action 2017.

Zeaiter, J., 2014. A process study on the pyrolysis of waste polyethylene. Fuel 133, 276–282. https://doi.org/10.1016/j.fuel.2014.05.028.

Zhang, Z, Hirose, T, Nishio, S, Morioka, Y, Azuma, N, Ueno, A, et al., 1995. Chemical recycling of waste polystyrene into styrene over solid acids and bases. Ind. Eng. Chem. Res. 34 (12), 4514–4519. https://doi.org/10.1021/ie00039a044.

Zheng, J, Suh, S., 2019. Strategies to reduce the global carbon footprint of plastics. Nat. Clim. Chang. 9 (5), 374–378. https://doi.org/10.1038/s41558-019-0459-z.