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Research article

Climate footprint assessment of plastic waste pyrolysis and impacts on the Danish waste management system

M.B. Karlsson [a,*], L. Benedini [b], C.D. Jensen [b], A. Kamp [a], U.B. Henriksen [b], T.P. Thomsen [a]

[a] Roskilde University, Institute of People and Technology, Universitetsvej 1, 4000, Roskilde, Denmark
[b] Technical University of Denmark, Department of Chemical and Biochemical Engineering, CHEC Research Centre, Miljøvej, 2800, Kgs. Lyngby, Denmark

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ABSTRACT

Increased plastic recycling is necessary to reduce environmental impacts related to manufacturing and end-of-life of plastic products, however, mechanical recycling (MR) – currently the most widespread recycling option for plastic waste – is limited by quality requirements for inputs and reduced quality of outputs. In this study, pyrolysis of plastic waste is assessed against MR, municipal solid waste incineration (MSWI) and fuel substitution through climate footprint assessment (CFA) based on primary data from pyrolysis of plastic waste sourced from Danish waste producers. Results of the CFA are scaled to the Danish plastic waste resource in an impact assessment of current Danish plastic waste management, and scenarios are constructed to assess reductions through utilization of pyrolysis. Results of the CFA show highest benefits utilizing pyrolysis for monomer recovery (−1400 and −4800 kg CO_2e per ton polystyrene (PS) and polymethyl methacrylate (PMMA), respectively) and MR for single polymer polyolefins (−1000 kg CO_2e per ton PE). The two management options perform similarly with mixed plastic waste (200 kg CO_2e per ton plastic waste). MSWI has the highest impact (1600–2200 kg CO_2e per ton plastic waste) and should be avoided when alternatives are available. Scaling the results of the CFA to the full Danish plastic waste resource reveals an impact of 0.79 Mt CO_2e in year 2020 of current plastic waste management. Utilizing pyrolysis to manage MR residues reduces the system impact by 15%. Greater reductions are possible through increased separation of plastic from residual waste. The best performance is achieved through a combination of MR and pyrolysis.

1. Introduction

Plastic is a ubiquitous material in modern society. Due to the broad range of material properties achievable through different combinations of polymers and additives, plastics are used abundantly in products throughout all sectors of the economy. In 2019, 460 Mt of plastic was produced globally with a compound annual growth rate of 8.4% in the period 1950–2015 leading to a predicted doubling of production in less than 20 years (OECD, 2019; Geyer et al., 2017). With this substantial global production of plastics, several societal issues follow. If mismanaged at end-of-life, plastics accumulate in the biosphere, where harm to wildlife and ecosystems is well documented (McGlade, 2021). In addition to the problem of environmental plastic pollution, global plastic consumption has a large impact on climate change. With bio-based plastics constituting less than one percent of current plastic production, more than 99% of plastics are made from fossil oil and natural gas (European Bioplastics, 2022). Extraction, refining, and manufacturing processes related to plastic production are all large emitters of greenhouse gases contributing to climate change (Ford et al., 2022). At the end-of-life of a plastic product, if the plastic waste is not landfilled or mismanaged with the risk of contributing to plastic pollution, it is most commonly incinerated – releasing the stored fossil carbon to the atmosphere (Ford et al., 2022).

Globally, 22% of plastic waste is mismanaged, 49% is landfilled, 19% is incinerated and 9% is recycled. In the EU, 5% of plastic waste is mismanaged, 39% is landfilled, 44% is incinerated and 14% is recycled (OECD, 2022). With increasing focus on the phase-out of fossil carbon from the economy as a response to global climate change, political efforts towards increased recycling rates of plastics are accelerating. In the EU, a vision for plastics in the European circular economy has been formulated with a goal of increasing plastic recycling to reduce European dependence on imported fossil feedstocks (COM, 2018). Meanwhile, in 2017 China announced their intention to implement an import ban on several waste types including the most common plastic wastes. In

* Corresponding author.
E-mail address: mbok@ruc.dk (M.B. Karlsson).

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2020 China amended the ban to include import of all solid waste (WTO, 2017). In 2017, approximately half of the plastic waste generation in the EU was exported – more than 85% of the export going to China - further adding to the incentive to increase domestic European plastic recycling rates (COM, 2018).

Plastic recycling options are generally categorized in four levels ranked by value creation (Singh et al., 2016). All recycling options are limited in different regards.

Primary recycling includes closed-loop mechanical recycling (MR, see Table 1 for full list of abbreviations) mostly applicable to mono-polymer plastic waste flows where the plastic is generally recycled to the same quality as the input material. However, the need for single-polymer input greatly limits the feasibility of general plastic waste treatment (ibid.).

Secondary recycling includes open-loop MR and is currently the most common plastic recycling option. Open-loop MR consists of mechanical sorting of mixed polymer or even mixed waste flows into more homogenous flows of single polymers for reprocessing. Open-loop MR often leads to a loss of material quality and therefore the output material is often used in lower value applications (ibid.; Schyns and Shaver, 2020).

Tertiary recycling includes chemical recycling of plastic waste. Chemical recycling can refer to a range of technologies, however in this study focus is on pyrolysis. Commercial plastic waste pyrolysis most commonly consists of thermal treatment of mixed plastic waste for the production of a hydrocarbon feedstock used as a drop-in in existing oil refinery processes. For certain polymers, a high content of monomers can be recovered. In these cases, monomers can be directly polymerized into new plastics after purification (Gkaliou et al., 2023; Singh et al., 2016).

Finally, *quaternary recycling* includes incineration with energy recovery, where the high heating value of plastic waste is utilized to produce heat and/or electricity (Singh et al., 2016).

In a circular economy perspective, pyrolysis of plastic waste is often viewed as downcycling as the process produces a product of lower value than the input material. However, pyrolysis of plastic waste has the potential to circumvent the high quality requirements for input material in primary and secondary recycling. Not all plastic waste fractions are suited for pyrolysis. Most relevantly, pyrolysis of PVC can cause chlorine production which is highly corrosive to equipment and potentially hazardous. However, mixed plastic waste flows often contain amounts of PVC below critical levels and if the process is configured for robust operation pyrolysis can be utilized to recover value from plastic waste flows not suited for MR. Additionally, plastics produced from pyrolysis oil used as a drop-in at petrochemical refineries is identical to fossil-based virgin plastics, avoiding the problem of quality degradation observed in some MR. To assess these benefits, the life cycle impacts of plastic waste pyrolysis need to be compared against alternative management options. Several life cycle assessments (LCA) have been performed on plastic waste pyrolysis in recent years, however, very few studies are based on primary experimental data from pyrolysis of actual plastic waste samples (Das et al., 2022; Jeswani et al., 2021; Faraca et al., 2019; Benavides et al., 2017). Instead, the majority of LCA studies are based on literature data or data volunteered from commercial pyrolysis plants. The literature data used in plastic pyrolysis LCA studies is primarily results from lab-scale pyrolysis tests. Plastic feedstocks used for pyrolysis range from single pure polymers to mixed plastic waste with varying levels of contaminants. Modelling impacts from plastic pyrolysis requires data on carbon and energy content across plastic feedstock and pyrolysis products. This level of data quality is rarely available in existing literature references leading to generalized assumptions about e.g., pyrolysis oil and gas characteristics, which are highly sensitive (ibid.). Alternatively, using data volunteered from commercial pyrolysis plants is favorable as it maps directly onto the real-world praxis of plastic pyrolysis. However, there is a risk of inducing bias when working with commercial partners with economic incentives and the provided data often contains proprietary information making the complete life cycle inventory inaccessible for review.

This study is designed to close critical gaps in relation to knowledge of system level benefits of waste plastic pyrolysis through climate footprint assessment (CFA). Climate impacts of plastic waste pyrolysis are assessed in a life cycle perspective using a complete primary dataset generated from bench-scale pyrolysis of a range of real-world plastic waste fractions. A complete carbon and energy balance is calculated based on calorimetry and elemental analysis of feedstock and pyrolysis products for all fractions. Pyrolysis (PY) is compared against mechanical recycling (MR), municipal solid waste incineration (MSWI) and plastic waste used directly for fuel substitution in high temperature industrial processes enabling assessment of optimal waste management options for each plastic waste fraction. In addition, a national scale estimate of the Danish plastic waste resource is provided. Combining results from the CFA with the plastic waste resource estimate enables assessment of the current Danish plastic waste management system and identification of optimal utilization of plastic waste management options.

Table 1
List of abbreviations and definitions.

Abbreviation	Definition
ARC	Amager Recycling Center
CFA	Climate Footprint Assessment
EPA	Environmental Protection Agency
FU	Functional unit
GWP	Global Warming Potential
H/C ratio	Ratio of hydrogen to carbon
HDPE	High density polyethylene, polymer
HFO	Heavy fuel oil
HHV	Higher heating value
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
MMA	Methyl methacrylate
MR	Mechanical recycling
MSWI	Municipal solid waste incineration
NIR	Near infrared
PE	Polyethylene, polymer
PET	Polyethylene teraphthalate, polymer
PEX	Cross-linked polyethylene, polymer
PMMA	Polymethyl methacrylate, polymer
PP	Polypropylene, polymer
PS	Polystyrene, polymer
PVC	Polyvinylchloride, polymer
PY	Pyrolysis
SS	Source separated plastic waste
WPU	Waste Plastic Upcycling

2. Materials and methods

2.1. Climate footprint assessment

2.1.1. Goal and scope

The purpose of this study is to evaluate the impact of chemical recycling of plastic waste through pyrolysis on climate change in a Danish context. Using consequential CFA, the impacts of management of seven plastic waste fractions through pyrolysis are determined and compared with relevant reference scenarios. The results from the CFA are scaled to the Danish national quantities of plastic waste to provide estimates of the national potential of chemical recycling as well as optimized plastic waste management scenarios utilizing pyrolysis in conjunction with other plastic waste management options. The results of the CFA are intended to inform and guide the deployment of waste plastic pyrolysis in Denmark to maximize the environmental benefit of the technology. In addition, the study is intended to provide guidelines for maximizing climate benefit through choice of waste management method for plastic waste fractions in Denmark.

2.1.1.1. Functional unit. The functional unit chosen for the study is "Management of 1 ton plastic waste on wet basis collected in present day Denmark". A common alternative functional unit used in plastic waste pyrolysis assessment studies is based on the production of oil, e.g., "mass of pyrolysis oil produced" (Antelava et al., 2019). This is advantageous when comparing pyrolysis oil as a transport fuel with conventional alternatives. However, this would not allow for comparison with other waste management options. Therefore, the functional unit is defined as input of plastic waste, as this allows for comparison of different plastic waste fractions across multiple waste management options. Although the functional unit is defined on a wet basis, moisture content is insignificant in all samples.

2.1.1.2. Assessment method. Global warming potential (GWP) is assessed using characterization method IPCC AR6 GWP 100, excl biogenic CO_2 and IPCC AR6 GWP 20, excl biogenic CO_2. Assessment using GWP 20 is included to consider the potential of the modelled waste management options if a shorter time horizon for climate action is prioritized.

2.1.1.3. Plastic waste fractions. Seven plastic waste fractions are modelled in the CFA. The plastic waste fractions are described in Table 2. All modelled fractions are sourced from Danish waste producers except for MR - originating from Germany - which simulates the residues of European mechanical recycling of Danish plastic waste. The fractions are selected to cover the broad range of plastic wastes found in society. The fractions vary across multiple parameters including mixed and pure polymers, technical suitability for mechanical recycling and pyrolysis, quantity in society, environmental cost of production, etc. Additionally, fractions are specifically chosen to cover the major flows of plastic waste, i.e., source-separated plastic waste, plastic in residual waste, and residues from mechanical recycling of plastic waste.

2.1.1.4. Scenarios. The following four plastic waste management options are modelled.

Pyrolysis (PY): Plastic waste is pyrolysed producing oil, gas, and char. Oil substitutes naphtha at established refineries or in specific cases monomers. Energy in the gas drives the pyrolysis process. If energy content in the gas is insufficient, natural gas is used as supplement. Excess energy in the gas substitutes district heating. Char is transported and burned substituting fuel in high temperature industrial processes.

Municipal solid waste incineration (MSWI): The plastic waste is combusted in MSWI producing heat and electricity substituting Danish district heating- and electricity production.

Mechanical recycling (MR): Plastic waste is sorted and reprocessed at a European MR facility and residues are utilized in MSWI. The recyclable part of the plastic waste is reprocessed into granulate substituting virgin plastic polymers.

Fuel substitution: The plastic waste is combusted in a relevant high temperature industrial process, substituting energy production from a range of fuels. The following fuels are investigated: natural gas, fuel oil, coal, and lignite. The chosen fuels constitute a range of low to high environmental impact per energy content enabling assessment of plastic as a fuel substitute in a broad spectrum of use-cases.

Table 2
Description of modelled plastic waste fractions.

Fraction	Description
SS	Source-separated mixed plastic waste from industry
MR	Residues from German mechanical recycling consisting of mixed polymers with low level of non-plastic content.
Grass	Artificial grass from sports areas consisting of PE and PP
PE	Waste PE shielding from cable production
PEX	Waste PEX shielding from cable production
PS	Waste PS
PMMA	Mixed waste PMMA collected by plastic recycling company

Management of all seven fraction is modelled in four different scenarios except for MR, as only the fractions SS and PE are suitable for MR.

2.1.2. Life cycle inventory

2.1.2.1. General data. Mass distribution across pyrolysis products as well as carbon-, and energy balances for each plastic waste fraction are determined based on primary data as described in the supplementary information (SI 1). The GaBi database (Sphera, 2022) is used for modelling background system processes when substituting plastic, oil, fuel products and transportation using EU-28 mixes. For domestic transport - e.g., transport of plastic waste for MSWI - 100 km transport is assumed. For international transport - e.g., transport of plastic waste for MR or transport of pyrolysis oil for refinement - 500 km transport is assumed. Marginal Danish heat and electricity mixes are modelled for energy substitution based on Muños and Weidema (2021) and the Danish Energy Authority (2021). Detailed descriptions of background energy systems can be found in the supplementary information (SI 3).

2.1.2.2. Pyrolysis. The modelled pyrolysis process simulates a coupled batch reactor process as being developed by several companies e.g., Waste Plastic Upcycling (WPU) and Makeen Energy. The coupled batch process is a robust process where the plastic typically requires only little or no pre-treatment. The coupling of the batches consists of energy transfer between the batches.

The energy-, mass-, and carbon balances across the pyrolysis process are modelled using primary data from bench scale experiments on pyrolysis of different plastic waste samples (SI 1).

The fate of pyrolysis oil is modelled in two different scenarios. For the plastic waste fractions with potential for monomer production - i.e., PS and PMMA - the monomer fraction of the oil is modelled substituting the respective monomer. For all other samples the pyrolysis oil is used as a drop-in in established oil refineries. This simulates the current praxis of many plastic pyrolysis companies. The substitution value of the pyrolysis oil is determined by the quality of the oil. Miskolczi et al. (2004) found that pyrolysis of municipal plastic waste yielded an oil product with 65–70 wt% aliphatic hydrocarbons containing between 5 and 12 carbon atoms. Based on these findings, mass substitution of naphtha is considered the most reasonable.

The pyrolysis gas is used to drive the pyrolysis process in a coupled batch system design. For plastic waste fractions with excess energy in the gas product, surplus energy substitutes district heating. For fractions with insufficient energy to drive the pyrolysis process, the necessary energy is provided by natural gas.

Pyrolysis of plastic waste may lead to production of a char product. The quantity of char may vary substantially depending on the input material. The char product is assumed to be used for energy purposes in high temperature industrial processes.

2.1.2.3. MSWI. MSWI is modelled simply as production of heat and electricity through incineration of the plastic waste assuming total conversion of the carbon content to CO_2 during combustion. Impacts from auxiliary MSWI inputs and outputs related to flue gas cleaning and incineration residues were found to be insignificant in sensitivity analysis. Heat and electricity efficiencies are assumed at 75% and 14% on a higher heating value basis, respectively, based on data from project partner ARC, operating - among other facilities - the largest waste incineration plant in Denmark (Skibakken) located in the Copenhagen region.

2.1.2.4. Mechanical recycling. The MR scenario is modelled as a two-step process simulating an optical NIR-scanner sorting step followed by reprocessing of sorted polymers by float-sink separation with subsequent extrusion. Residues from recycling are incinerated in MSWI. Energy production from MSWI is modelled based on the measured

heating value of residues from commercial mechanical plastic recycling. The produced granulate substitutes virgin polymers.

Sorting and reprocessing efficiencies are based on Faraca et al. (2019). Specifically, three sequential NIR scanners are modelled targeting PP, PE, and PET, respectively, with sorting efficiencies and errors defined per scanner on a polymer level. Similarly, reprocessing efficiency is defined per polymer. This requires data on polymer distribution for the modelled plastic waste fractions. For the plastic waste fraction PE, this is trivial as the fraction is pure PE. For the plastic waste fraction SS, the polymer distribution is assumed similar to the source-separated plastic waste analyzed in Eriksen and Astrup (2019). Details related to polymer distribution can be found in the supplementary information (SI 4).

MR often leads to some level of degradation of the plastic quality depending on the feedstock (Ragaert et al., 2017). To account for loss of quality, a quality factor is applied to the substitution value of recycled polymers. Rigamonti et al. (2020) performed a review of existing LCA approaches to quantification of substitution value of recycled products. They reported substitution values of 0.69 for mixed plastic waste and 0.75 for pure HDPE. These values are used as quality factors in the CFA.

2.1.2.5. Fuel substitution. Plastic waste is often used as a fuel in high temperature industrial processes because of the high energy content when other recycling options are not feasible (Chaves et al., 2022). To account for the different potential fuels that can be substituted by plastic waste, four different fuel substitution scenarios are modelled. Fuel types are selected to cover a broad range of environmental impacts.

Fuel substitution scenarios are modelled assuming 1:1 substitution on a higher heating value basis. This assumption implies that the plastic waste can fulfill the same function per unit of energy as the substituted fuels. This could be exemplified by utilization of plastic waste in a rotary kiln fulfilling the need for high temperature process energy for cement production as a substitute for coal or lignite.

2.1.3. Uncertainty, sensitivity, and scenario analysis

Parameter uncertainty is assessed by Monte Carlo simulation based on the approach described by Bisinella et al. (2016). First, for all model parameters, contribution to total variance is determined. Then, parameters with the highest contribution to total variance, covering at least 90% of the total model variance are selected for Monte Carlo simulation.

In addition, several variation-scenarios are modelled investigating alternative model configurations and assumptions. For pyrolysis, a scenario is modelled extending the system boundary to include the end-use of pyrolysis oil. The impacts of combusting the oil for energy purposes are compared against similar combustion of naphtha and heavy fuel oil, simulating the use of pyrolysis oil as a transport fuel. For MR, a scenario is modelled utilizing residues in industrial fuel substitution instead of MSWI, as this is a common alternative praxis across Europe. Finally, the impact of alternative background energy-system assumptions is investigated.

2.2. Danish national scale assessment

To assess the potential climate impact of plastic waste pyrolysis in a Danish context, it is necessary to quantify the Danish plastic waste resource. With estimates of the Danish plastic waste resource and current management options, the results of the CFA can be used to assess the impacts of current plastic waste management in Denmark and form recommendations for improved plastic waste management.

It is necessary to define some central terms. Firstly, the term *plastic waste* refers to the total mass of a plastic waste fraction including contaminants, e.g., biomass. All quantities in the following section are reported by this definition. Additionally, a significant amount of plastic waste is in the form of *plastic in non-separable products*, e.g., aluminium bags with internal plastic coating. Plastics in non-separable products in the residual waste stream are excluded from this assessment as they are assumed unavailable as a resource for alternative management even in scenarios with additional sorting of residual waste. Finally, only nationally generated plastic waste is included in this analysis. Some Danish MSWI plants import waste from abroad; these quantities are excluded.

2.2.1. Plastic waste quantities

Quantities of plastic waste flows in Denmark are calculated based on existing datasets from the Danish Environmental Protection Agency (EPA) and Statistics Denmark (Miljøstyrelsen, 2020; Danmarks Statistik, 2021) in conjunction with sorting data on plastic content in residual waste fractions from Econet (Econet, 2015; Econet, 2019; Econet, 2020a; Econet, 2020b; Miljøstyrelsen, 2018). Data from the EPA is based on the Danish *waste data system*. This system requires all recipients of waste to report quantities and management method to the central system. Data from Statistics Denmark is based on import, export and production statistics. Given the different methodological approaches in the underlying datasets, the two estimates can be compared to assess uncertainty in the final estimate of national plastic waste quantities. Based on assessment of uncertainty related to the underlying datasets and key assumptions in the calculations, mean values with standard deviation are provided for the total Danish quantities of source-separated plastic waste and plastic waste in residual waste fractions. Details related to calculation of plastic waste quantities can be found in the supplementary information (SI 5).

2.2.2. National scale scenarios

Using the determined quantities of plastic waste, scenarios are modelled mapping the flow of plastic waste in the current waste management system and in four scenarios introducing pyrolysis as a waste management option. In the current waste management system, it is assumed that all source-separated plastic waste is sent for MR. There is some uncertainty as to the actual fate of source-separated plastic waste sent for recycling due to transparency issues in the value chain. As the purpose of this study is to compare the relative difference across waste management system configurations, the assumption is not regarded as critical. Scenarios are described in Table 3. Scenarios S1 and S2 integrate pyrolysis into the current waste management system. In S1, only residues from MR are utilized in pyrolysis. This is the minimal possible alteration of the current system. In S2, pyrolysis replaces MR as the management option for all source-separated plastic waste. In S3 and S4 central sorting of plastic in residual waste is simulated. These scenarios represent possible near-future waste management systems with increased plastic waste separation rates achieved through either technological or regulatory means, i.e., mechanical sorting or extended producer responsibility schemes etc.

2.2.3. Coupling CFA results with national scale scenarios

When assessing the impacts of each national scale scenario, flows of

Table 3
Descriptions of national scale scenarios.

Scenario	Description
Current	Plastic in residual waste utilized in MSWI. Source-separated plastic waste sent for MR. Residues utilized in MSWI.
S1	Plastic in residual waste utilized in MSWI. Source-separated plastic waste sent for MR. Residues utilized in pyrolysis.
S2	Plastic in residual waste utilized in MSWI. Source-separated plastic waste utilized in pyrolysis.
S3	Plastic in residual waste mechanically sorted. Source-separated plastic waste sent for MR. Sorted plastic from residual waste and residues from MR utilized in pyrolysis. Residues from residual waste sorting utilized in MSWI.
S4	Plastic in residual waste mechanically sorted. Sorted plastic from residual waste and source-separated plastic waste sent for MR. Residues from MR utilized in pyrolysis. Residues from residual waste sorting utilized in MSWI.

plastic waste are matched with corresponding plastic waste fractions from the CFA. Three flows of plastic waste are considered: plastic in residual waste, source-separated plastic waste, and residues from MR. The sample SS is used to simulate plastic in residual waste as it consists of mainly polymers with a polymer distribution representative of average Danish plastic waste based on Eriksen and Astrup (2019). For source-separated plastic waste, the composition of the SS sample is modified to match the content of non-plastic material found in Danish source-separated plastic waste (ibid.). Finally, the sample MR is used to simulate residues from mechanical recycling. See supplementary information for details regarding polymer distributions (SI 4).

Assessment of uncertainty related to the impact of national scale scenarios is performed through Monte Carlo simulation applying the uncertainties determined for Danish plastic waste quantities and the CFA results.

2.3. Plastic circularity potential

As an additional indicator of system performance, an assessment of plastic circularity potential is provided for both CFA and national scale scenarios. This indicator quantifies the potential recycled plastic output in each scenario. For MR, plastic circularity potential is calculated as the recycled plastic output applying the quality factor to account for quality degradation. For pyrolysis scenarios, 1:1 plastic production is assumed for the monomer fraction of pyrolysis oil. For non-monomer pyrolysis oil, plastic circularity potential is calculated assuming steam cracking of the oil followed by market average petrochemical refinement of the output. The ratio of steam cracking products ending in plastics is applied as the circularity potential for the oil. Assuming the oil is similar to naphtha, this ratio is approximately 0.6 based on material flow analysis of the chemical sector by Levi and Cullen (2018). Details related to calculation of circularity potentials can be found in the supplementary information (SI 6).

3. Results and discussion

3.1. Climate footprint assessment

The results of the CFA are shown in Fig. 1. Across all plastic waste fractions, MSWI scenarios show the highest impacts at 1600–2200 kg CO_2e per ton plastic waste. This is explained by the low substitution value of marginal heat and electricity in Denmark combined with the high impact of full conversion of the plastic carbon content to fossil CO_2. For MR, a clear advantage is seen for the plastic waste fraction PE over SS with an impact of −1000 compared to 200 kg CO_2e per ton plastic waste. The plastic waste fraction PE consists of pure waste PE from cable scrap as opposed to the mixed source-separated plastics of SS. This allows bypassing of the sorting step and the associated incineration of sorting residues, greatly reducing the impact of the scenario. For pyrolysis, PS and PMMA perform much better than other plastic waste fractions due to the high substitution value of recovered monomers. PS and PMMA pyrolysis show impacts of −1400 and −4800 kg CO_2e per ton plastic waste, respectively, while impacts from pyrolysis of non-monomer producing plastic waste fractions range from −300 to 300

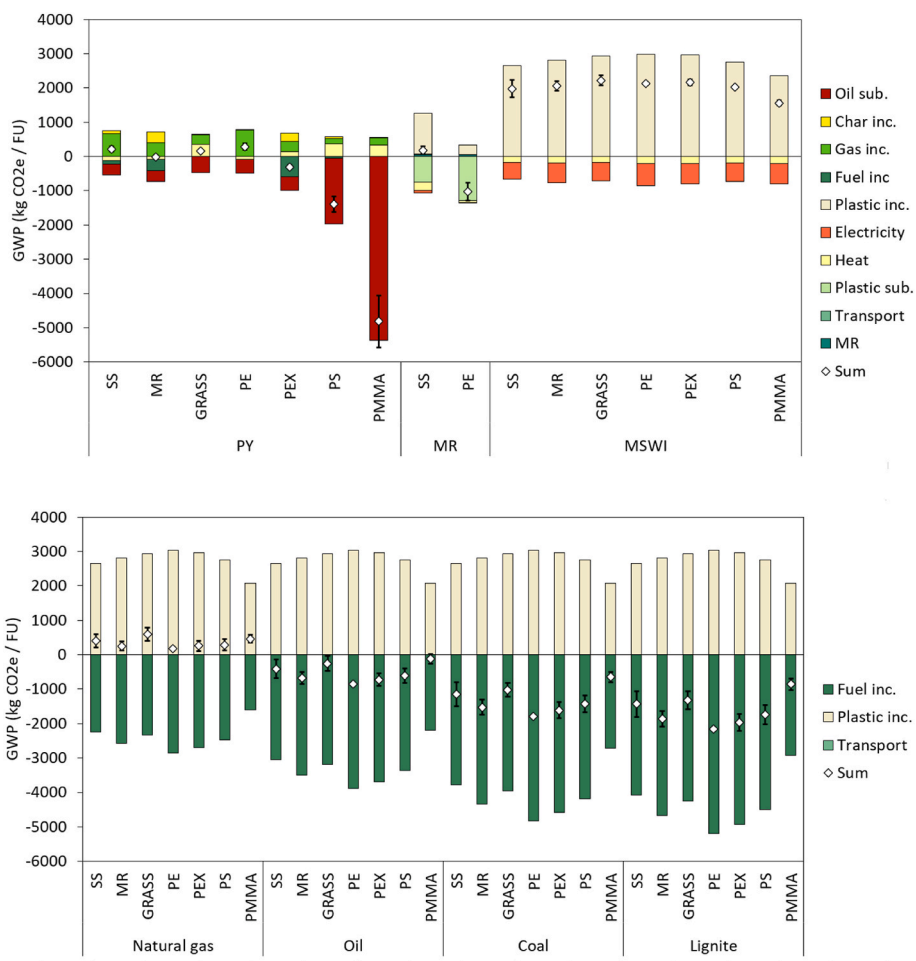

Fig. 1. CFA results for scenarios pyrolysis (PY), mechanical recycling (MR) and municipal solid waste incineration (MSWI) (above) as well as fuel substitution scenarios (below). FU: 1 ton plastic waste, as received. Error bars represent standard deviation from the net sum based on Monte Carlo simulation.

Table 4
Plastic circularity potentials for pyrolysis and mechanical recycling scenarios. Potential output of recycled plastic in tons.

Fraction	PY	MR
SS	0.34	0.40
MR	0.36	
Grass	0.52	
PE	0.44	0.68
PEX	0.44	
PS	0.74	
PMMA	0.83	

kg CO_2e per ton plastic waste. Fuel substitution scenarios show a clear ranking of fuels in terms of benefits from substitution. Average impacts from fuel substitution of natural gas, oil, coal, and lignite are 300, -500, −1000 and −1600 kg CO_2e per ton plastic waste, respectively. As such, natural gas is the only fuel with a net burden from substitution; all other fuels show a net benefit. This pattern relates mainly to the H/C ratio in the fuels versus plastics, and to a lesser extent with procurement emissions. Unless procurement emissions are excessive, a climate benefit is achieved through substitution of fossil fuels with higher H/C ratio plastics.

Comparing this study with existing literature results, GWP100 impacts in kg CO_2e per ton plastic waste range from −300 to 700 for pyrolysis with impacts around 600 kg CO_2e most frequent (Das et al., 2022; Jeswani et al., 2021; Faraca et al., 2019; Khoo, 2019; Gear et al., 2018; Shonfield, 2008). For MR impacts range between −700 and 900 (Faraca et al., 2019; Khoo, 2019; Shonfield, 2008), and for MSWI between 1000 and 1800 (Khoo, 2019; Gear et al., 2018). As such, this study generally confirms previous findings in terms of absolute impacts as well as ranking of plastic waste management options.

Table 5
Plastic circularity potential of national scale assessment scenarios. Potential output of recycled plastic in 1000 tons.

Scenario	Current	S1	S2	S3	S4
Oil		21	32	71	43
Granulate	38	38		38	97
Sum	38	58	32	109	140

Assessing the results with a 20-year time-horizon, the impact of selected processes drives the difference from GWP100 results. Using GWP20 as characterization method, methane emissions are weighted higher due to the shorter half-life of methane in the atmosphere compared to CO_2. This is especially pronounced in PY scenarios with naphtha or monomer substitution where methane emissions from petrochemical processes contribute substantially to the impacts. Naphtha, styrene, and methyl methacrylate (MMA) impacts increase by 49%, 48%, and 34%, respectively, using GWP20 compared to GWP100. Through the same mechanism, polymer substitution in MR scenarios is affected, however PE, PP and PET impacts increase only by ~8%. In MSWI scenarios, heat and electricity mixes are affected slightly with increased impacts of 5% and 7%, respectively, and in fuel substitution scenarios, impacts per MJ substituted fuel increase by 5%, 2%, 9%, and 2% for natural gas, oil, coal, and lignite, respectively. Across all scenarios assessing impacts with GWP20 results in lower net impacts with the highest relative decrease in PY scenarios. Results based on

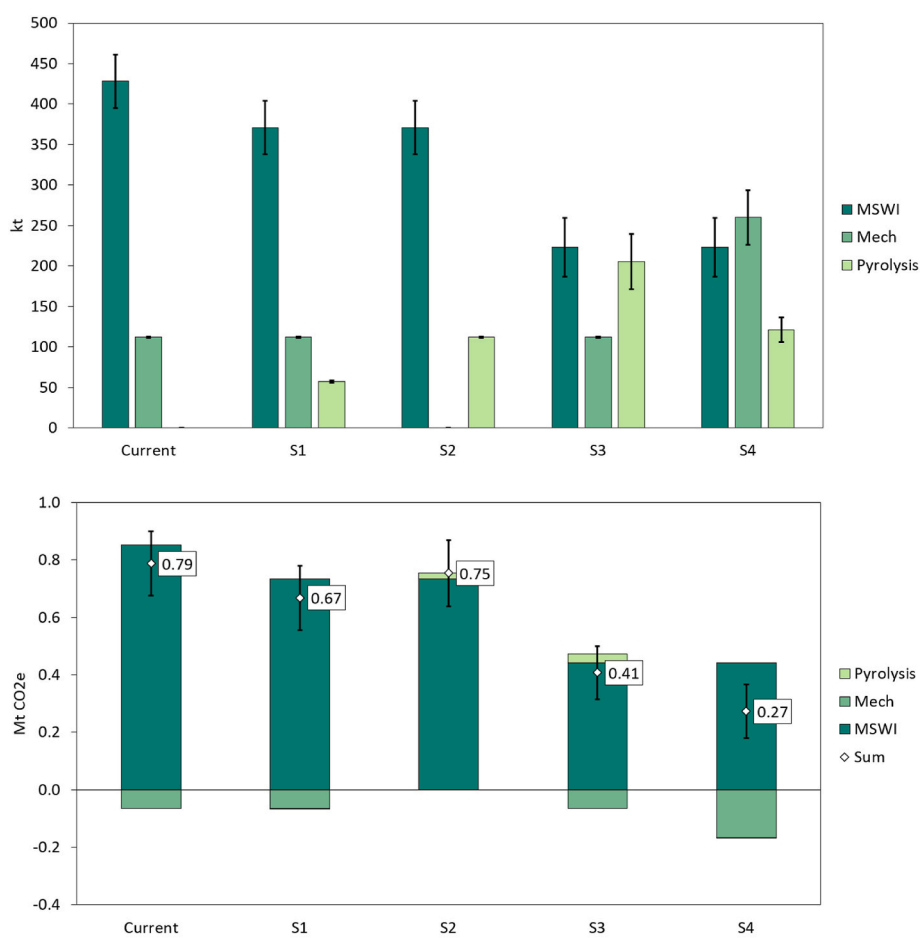

Fig. 2. *Above:* Quantities of plastic waste flows to management options across national scale assessment scenarios in 1000 tons. *Below:* Total impact of current Danish plastic waste management and scenarios including pyrolysis. In both diagrams, error bars represent standard deviation based on Monte Carlo simulation.

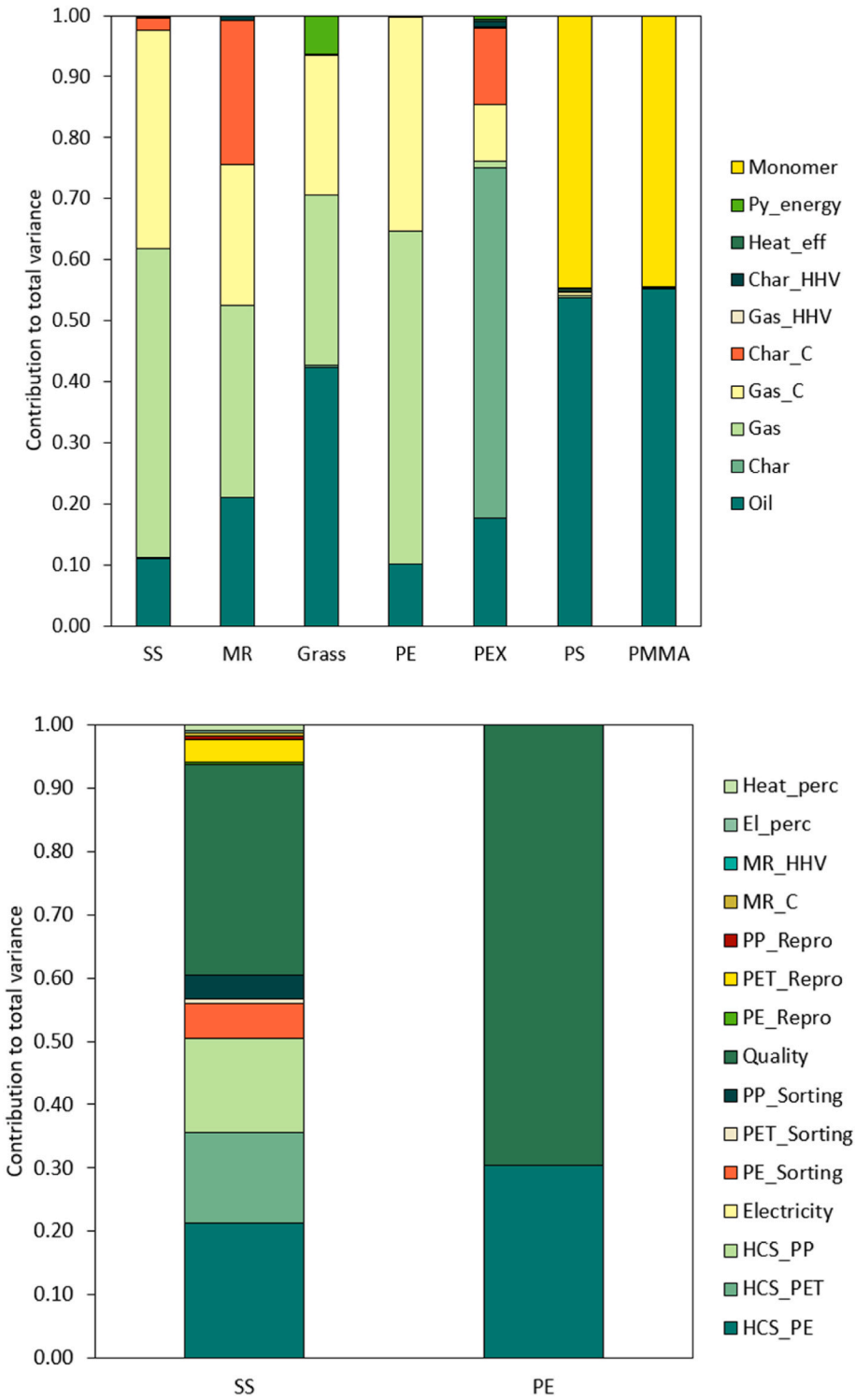

Fig. 3. Contribution to total variance of parameters in PY (above) and MR (below) scenarios.

assessment of impacts using GWP20 show that pyrolysis performance improves compared to other management options if a shorter time horizon for climate action is prioritized.

Based on the results of this study the following recommendations are formulated for policy makers and industry stakeholders to optimize plastic waste management options. The variation in impact between plastic waste fractions within waste management scenarios highlights the need for assessment of management options on a per case basis. MSWI of plastic waste should be avoided in low carbon energy systems whenever other waste management options are available. Single polymer polyolefins such as PE perform well in MR while technical polymers such as PS and PMMA with potential for monomer recovery perform very well in PY and should be prioritized accordingly. Mixed plastic waste management impacts are similar when comparing MR and PY, assuming MSWI of residues from recycling. However, utilizing MR residues for PY could decrease the impact further and potentially increase plastic circularity. Regarding fuel substitution, the specific carbon to energy ratio of the waste plastic must be confirmed to be lower than the

substituted fuel to achieve a benefit. For fuels with high carbon to energy ratios (e.g., coal and lignite), the benefit can be substantial, in some cases outperforming even single polymer MR. Therefore, when considering utilization of plastic waste as fuel substitution a specific assessment is necessary to determine impacts based on carbon to energy ratios of both plastic feedstock and substituted fuel. These recommendations can inform waste management policies and practices.

3.1.1. Plastic circularity potential

The output of recycled plastic in PY and MR scenarios is shown in Table 4. MR has a higher plastic circularity potential for the two modelled plastic waste fractions compared to PY. PY is, however, applicable to a broader range of plastic waste fractions, enabling management of waste fractions not suitable for MR. These results suggest that MR should be prioritized when applicable and potentially combined with PY of residues from recycling to maximize plastic circularity. Additionally, PY should be utilized to recover value from plastic waste fractions not suited for MR and specific effort should be put toward separation of technical polymers such as PS and PMMA for recovery through PY. The approach used to quantify plastic circularity potential for pyrolysis of non-monomer producing plastic waste fractions assumes market average petrochemical refinement of the oil product resulting in a plastic circularity factor of 0.6 applied to the oil output. To increase the incentive for recycling in the chemical industry, methods are currently being developed to enable recycling claims through mass balance certification (Ellen Macarthur Foundation, 2019). Such methods would enable mass balance-based guarantees that pyrolysis oil is used solely for new plastic production, in praxis raising the plastic circularity factor of the oil to one. Implementation of these methods should be pursued to incentivize greater circularity of plastic resources.

3.2. Danish national scale assessment

Total quantities of Danish plastic waste and impacts across waste management scenarios are shown in Fig. 2. Yearly, 370 kt plastic waste ends in Danish residual waste fractions and is incinerated in MSWI, while 110 kt are source-separated. Assuming MR of the source-separated plastic waste, an additional 60 kt residues are incinerated resulting in a total of 430 kt plastic waste to MSWI. Based on results from the CFA, management of these quantities cause an impact of 0.79 Mt CO_2e. The 60 kt residues contain a high content of polymers making them an ideal resource for pyrolysis. In S1 the system impact is decreased by 15% to 0.67 Mt CO_2e by utilizing recycling residues in pyrolysis. In S2, utilizing all source-separated plastic waste in pyrolysis instead of MR shows worse performance with an impact of 0.75 Mt CO_2e, a reduction of only 5% compared to the current management. Larger reductions are achieved in S3 and S4 where additional sorting of residual waste is modelled. At 40% sorting efficiency of plastic in residual waste, quantities to MSWI are reduced to 220 kt. The additional resource of 150 kt separated plastic waste is utilized for pyrolysis in S3, resulting in a reduction of 48% compared to current management with an impact of 0.41 Mt CO_2e. In S4, sorted plastic from residual waste is utilized in MR, achieving a reduction of 66% with an impact of 0.27 Mt CO_2e.

These results further highlight the foundational issue that plastic recycling options are limited by separation of plastic waste from residual waste fractions. In Denmark or other contries with low carbon energy systems, efficient separation either at the source by households and industry or centrally should be established to enable the potentials of existing and emerging waste management options including pyrolysis. It should be noted that the system boundaries of the national scale assessment exceed the Danish national geographical borders and as such these results cannot be translated directly to national political targets for waste sector emissions reductions or recycling rates. Instead, the results describe a potential to reduce the impact of plastic waste management across the full life cycle of plastic waste across national borders.

3.2.1. Plastic circularity potential

The circularity potential of all modelled national scale assessment scenarios is shown in Table 5. In the current plastic waste management system, 38 kt recycled plastic is produced through MR, factoring in quality degradation. This is improved to 58 kt in S1, utilizing pyrolysis in conjunction with MR but reduced to 32 kt in S2 where pyrolysis replaces MR. Much higher circularity potentials are achieved in S3 and S4 by increasing the resources available for recycling through residual waste sorting. There is great uncertainty related to the potential of residual waste sorting. The modelled scenarios simulate central mechanical sorting of residual waste; however, the modelled sorting efficiencies could also be achieved through alternative means, e.g., increased source separation efficiencies in households and industry. The feasibility of MR of plastics sorted from residual waste is not certain and depends on system configuration; however, pyrolysis is likely to be a feasible management option regardless of configuration. Therefore, pyrolysis of residual waste plastic is modelled in S3 and MR in S4. In S3, 109 kt recycled plastics are produced while 140 kt are produced in S4. These results highlight the potential of prioritizing MR when feasible and utilizing pyrolysis to manage recycling residues.

3.3. Sensitivity and scenario analysis

3.3.1. Parameter uncertainty

The contribution to total variance of parameters in PY and MR scenarios is shown in Fig. 3. Based on the combined uncertainty and sensitivity of parameters, contribution to total variance is calculated, describing how much of the total model variance can be attributed to a given parameter. In PY scenarios, variance of monomer producing plastic waste fractions is dominated by parameters for oil output and

Table 6
Scenario analysis. Absolute change in GWP from main model result across scenarios. Oil end-use scenarios model pyrolysis oil end-use as fuel, extending the system boundaries to account for emissions from combustion. Residues end-use scenarios model fuel substitution as end-use for MR residues. Alternative background energy scenarios model extreme case background energy-systems.

Scenario	Sub-scenario	SS	MR	Grass	PE	PEX	PS	PMMA
Oil end-use	HFO	244	265	31	−245	−165	1213	5454
	Naphtha	519	540	499	194	266	1705	5727
Residues end-use	Natural gas	−626			−130			
	Oil	−969			−203			
	Coal	−1280			−363			
	Lignite	−1403			−396			
Alternative background energy								
Coal & natural gas	PY	−700	−611		−624			
	MR	−1797			−2280			
	MSWI	−3615	−4529	−3836	−4691	−4613	−4490	−2170
Wind & biomass	PY	78	47		53			
	MR	253			−1			
	MSWI	524	216	480	583	385	428	617

monomer fraction of the oil due to the high substitution value of the monomers. For the non-monomer producing plastic waste fractions, most variance is attributable to parameters related to pyrolysis gas. Gas output and gas carbon content combined contribute 50–90% of total variance across these plastic waste fractions except for PEX where char output is very high. Following gas related parameters, output of oil is generally a sensitive parameter, contributing 10–40% of total variance for non-monomer producing fractions. This highlights the need for high data quality regarding pyrolysis product distribution and product characterization, especially carbon content of gas product.

In MR scenarios, the plastic waste fraction PE is completely determined by the parameters for PE content in the fraction and the quality factor because the fraction consists solely of PE. For the plastic waste fraction SS, parameters related to polymer distribution contribute half of the total variance. The quality factor has the second highest contribution at 33% and the remaining variance is attributable mainly to sorting and reprocessing efficiencies. This high contribution of the quality factor is critical. The parameter is applied to account for reduced substitution value of mechanically recycled polymers due to quality degradation. Methods for quantification include assessment of degradation of physical material properties and market-based assessments, however, no widespread consensus on quantification of the parameter exists.

Compared to PY and MR scenarios, MSWI and fuel substitution scenarios are defined by relatively few parameters. For MSWI, carbon content in plastic waste is generally the most important parameter contributing 60–90% of total variance followed by heat- and then electricity efficiency at 5–15 and 10–25%, respectively. For fuel substitution scenarios, plastic HHV generally contributes >90% of total variance due to the impact of fuel substitution in the results. This further underlines the need for high-quality characterization of plastic waste carbon- and energy content in the assessment of waste management options.

Given the sensitivity of parameters related to oil and gas output as well as gas carbon content in PY scenarios, the results of this study must be interpreted considering the limitations of the experimental data used. The use of primary data in this study allows assessment of specific plastic waste fractions for identification of high potential fractions. However, data for each fraction is based on pyrolysis of a small quantity of plastic waste without extensive optimization of process parameters. This risks underestimation of pyrolysis potential. LCA studies of plastic pyrolysis in the literature rarely apply primary data for pyrolysis parameters, instead referencing previous experimental data in the literature or volunteered data from commercial plastic pyrolysis. The latter approach is likely to better represent real world operation of plastic pyrolysis due to the large sample size of commercial plants and optimized operational parameters achieved through calibration over time. However, this approach risks over-estimation of pyrolysis potentials and there can arise issues of data transparency due to commercial interests. Future pilot- and full-scale studies are required to increase the quality and robustness of plastic waste pyrolysis LCA, ideally based on transparent LCI data from commercial plastic pyrolysis operation.

3.3.2. Scenario analysis

The results of the scenario analyses are summarized in Table 6. A few major findings can be determined from the results. From the results of oil end-use scenarios, it is obvious that monomer oil fractions should not be incinerated for energy. Additionally, for all plastic waste fractions but PE and PEX, fuel use performs worse than general naphtha substitution in the main model. Therefore, dedicated fuel production from plastic pyrolysis oil should be avoided when used as drop-in in refineries is possible. For residues end-use scenarios, a significant improvement in performance is observed through fuel substitution of recycling residues with increasing reductions depending on the impact of the substituted fuel. These findings suggest that when suitable high temperature industrial fossil fuel substitution is possible, it is preferable to MSWI as a management option for recycling residues in a Danish context. Finally, modelling with a high impact background energy-system based on coal and natural gas results in much greater reductions in all scenarios with energy substitution. This is especially pronounced in MSWI scenarios where energy production is the main product and to a lesser extent in MR scenarios with MSWI of recycling residues.

These findings highlight the climate benefits of pyrolysis regarding monomer production from technical polymers and the lost potential related to their inefficient management. In addition, the potential benefits of utilizing residues from MR for fuel substitution - given a carbon efficient energy system - and the benefits of energy production from plastic waste in countries with carbon intensive energy systems are significant.

4. Conclusions

Several key insights related to the climate impact of plastic waste management are identified from the results of this climate footprint assessment to inform plastic waste management policy and practice. MSWI should be avoided in a Danish context when alternative management options are available due to the low substitution value of Danish marginal energy products. Highest benefits are achieved through MR of single polymer plastic waste fractions and PY of monomer producing plastic waste fractions. For mixed plastic waste fractions, MR and PY performance is similar. Fuel substitution in high temperature industrial processes can be highly beneficial when substituting fossil fuels with higher carbon to energy ratios than the plastic waste in use, highlighting the need for assessment on a per case basis. Scaling the results of the CFA to the Danish plastic waste resource allows assessment of the current waste management system and scenarios introducing PY as a waste management option. An estimated 480 kt plastic waste is generated in Denmark each year, 110 kt of which is source-separated. Current management of this resource has an estimated impact of 0.79 Mt CO_2e. Optimizing the current system by utilizing residues from MR for PY reduces the impact by 15% to 0.67 Mt CO_2e. Increased separation of plastic waste enables higher reductions. Assuming 40% separation of plastic from the residual waste, reductions of 48–66% are achievable, corresponding to system impacts of 0.41-0.27 Mt CO_2e. Across scenarios, highest reductions and greatest plastic circularity potentials are achieved through utilization of MR and PY in combination. Data variations and assessment assumptions can have substantial impact on plastic waste CFA results. On the functional unit-based level, special awareness should be given to attaining high-quality data on pyrolysis product distribution and characterization as well as plastic waste carbon- and energy content. For MR systems, continued effort should be made toward developing robust methods for determination of substitution values of recycled polymers. From these system models, it is expected that improved pyrolysis system performance could be achieved through increased pyrolysis oil yield and attention towards separation of plastic waste with potential for monomer recovery.

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CRediT authorship contribution statement

M.B. Karlsson: Formal analysis, Investigation, Methodology, Writing - original draft, Writing - review & editing. **L. Benedini:** Data curation, Formal analysis, Investigation, Writing - review & editing. **C. D. Jensen:** Investigation. **A. Kamp:** Conceptualization, Supervision, Writing - review & editing. **U.B. Henriksen:** Resources, Supervision. **T. P. Thomsen:** Conceptualization, Funding acquisition, 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

No data was used for the research described in the article.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2023.119780.

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