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Life cycle assessment of global warming potential of feedstock recycling technologies: Case study of waste gasification and pyrolysis in an integrated inventory model for waste treatment and chemical production in Germany

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ABSTRACT

Feedstock recycling in the form of gasification and pyrolysis are promising alternatives to thermal treatment for the utilization of non-recyclable waste fractions. To date, the systemic consequences of their application for waste treatment and chemical production as well as associated environmental effects remain insufficiently investigated. To address this gap, this study introduces an integrated life cycle inventory model based on Germany's production system which encompassed the treatment of major post-consumer waste fractions (municipal solid waste and source-separated packaging waste) and the production of major base chemicals (lower olefins, BTX aromatics, methanol, ammonia and hydrogen). The utilized approach facilitates a prospective comparative assessment of feedstock recycling, conventional and PTX-based production routes under uniform system frameworks, accounting for differences in production characteristics (e.g. product yields and utility demands) with minimized allocation assumptions. An evaluation of the global warming potential shows that under assumptions resembling the current production system (Framework Status Quo), feedstock recycling pathways lead to a reduction in greenhouse gas emissions, with gasification exhibiting higher emission reduction (Framework Energy Integration), a higher greenhouse gas reduction is observed for feedstock recycling compared to PTX-based chemical production pathways.

FTFischer-Tropsch synthesisMTOMethanio to otenins synthesisGHGGreenhouse gasPTXElectricity-based production/products (Power-to-XGWPGlobal warming potentialRDFResidue derived fuelLCALife cycle assessmentRDF PPRDF incineration in RDF power plantLCILife cycle inventoryRMPRecyclable material rich in plasticsLCILife cycle impact assessmentWtEWaste-to-energy	Abbreviat ASU BTX CHP FT GHG GWP LCA LCI LCI	tions Air separation unit Benzene, toluene, xylenes Combined heat and power Fischer-Tropsch synthesis Greenhouse gas Global warming potential Life cycle assessment Life cycle inventory	LHV MPW MSW MSWI Mt MTO PTX RDF RDF PP RMP WtE	Lower heating value Mixed plastic waste Municipal solid waste MSW incineration Mega ton, 1 Mt = 10 ⁹ kg Methanol-to-olefins synthesis Electricity-based production/products (Power-to-X) Residue derived fuel RDF incineration in RDF power plant Recyclable material rich in plastics Waste-to-energy
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; ASU, Air separation unit; BTX, Benzene, toluene, xylenes; CHP, Combined heat and power; FT, Fischer-Tropsch synthesis; GHG, Greenhouse gas; GWP, Global warming potential; LCA, Life cycle assessment; LCI, Life cycle inventory; LCIA, Life cycle impact assessment; LHV, Lower heating value; MPW, Mixed plastic waste; MSW, Municipal solid waste; MSWI, MSW incineration; Mt, Mega ton, 1 Mt = 10^9 kg; MTO, Methanol-to-olefins synthesis; PTX, Electricity-based production/ products (Power-to-X); RDF, Refuse-derived fuel; RDF PP, RDF incineration in RDF power plant; RMP, Recyclable material rich in plastics; WtE, Waste-to-energy. * Corresponding author.

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1. Introduction

Chemical recycling is considered a potential complement to existing strategies (i.e. reuse and mechanical recycling) to realize a circular economy (Crippa et al., 2019; Lee et al., 2021). It encompasses a range of different technological pathways for reintegrating waste into the value chain. These range from solvent-based purification, depolymerization to feedstock recycling and differ in terms of applicable waste fractions, processes and technologies, products as well as product reintegration possibilities (Mamani-Soliz et al., 2020; Ragaert et al., 2017). Fig. 1 presents an exemplary overview of different routes/loops whereby waste plastics can be recirculated into the production pathway.

This investigation focuses on feedstock recycling technologies in the form of gasification and pyrolysis. Both technologies are characterized by their products (i.e. syngas and pyrolysis oil respectively) which are targeted for integration into base chemical production. Integration can take place in two ways. First, by directly replacing conventional feedstock, especially crude oil-based naphtha, liquified petroleum gas (LPG) and natural gas. Second, by replacing conventional base chemical production technologies (Ragaert et al., 2017).

In previous LCA investigations, gasification and pyrolysis are frequently evaluated as thermal treatment technologies for the production of electricity and/or heat (Arena et al., 2015; Astrup et al., 2015; Dong et al., 2018; Ramos et al., 2019; Zaman, 2013). Studies assessing their implementation as feedstock recycling technologies for chemical production is summarized in Table 1.

Two major problematic aspects can be identified from previous LCA studies. First, there is a lack of available inventory data for feedstock recycling processes due to limited large-scale technology application for chemical utilization (Hrbek, 2019). Further compounding the problem, inventory data is specific to the application case. Properties of waste-based feedstock can differ significantly (e.g. LHV of MSW is about 10 MJ/kg, LHV of mixed plastics is up to 40 MJ/kg), and process efficiencies and characteristics for pyrolysis and gasification are highly dependant on process technology and feedstock. Moreover, neither syngas nor pyrolysis oil are uniform products and differ greatly in quality and composition. Their characteristics - dependant on the specific technology, reaction conditions and applied feedstock - influence their further treatment (Punkkinen et al., 2017; Seidl et al., 2020; Seitz et al., 2020). Finally, both syngas and pyrolysis oil are applicable for the production of a variety of products in an integrated chemical production system (Keller et al., 2021). To fill inventory data gaps and ensure full energy and mass balances (Meys et al., 2020), process modelling is one suitable approach (Christensen et al., 2020; Tsoy et al., 2020).

Another issue is the variance in assessment scope and methodology. Previous investigations commonly apply an attributional approach to cumulate life cycle inventory (LCI). In *attributional LCI*, the aim is to describe relevant flows to and from a reference process and its associated processes under a normative rule. Allocation is applied to consider multifunctionality of processes and marginal processes for substitution and avoidance. Generally, the approach is applied to quantify the environmental impact and the distribution within the system, but is less suitable for decision support. In *consequential LCI*, the objective is the prediction of consequences of choices within a system based on developed scenarios, thereby addressing decision support. This requires a system modelling approach to reflect interactions of processes within the system. The application of allocation and association of marginal processes is avoided by system expansion. Furthermore, market constraints and effects of changing supply are considered (Brandão et al., 2017; Ekvall, 2020; Ekvall and Weidema, 2004).

The present investigation applies a consequential approach, by introducing an inventory system that expands the foreground system to the major processes for waste treatment and base chemical production. By introducing feedstock recycling processes in this assessment scope, it is possible to evaluate the systemic effects on the chemical production system with limited application of allocation. Based on this inventory model, a life cycle assessment with focus on global warming potential (GWP) is conducted, to assess gasification and pyrolysis-based pathways in comparison to conventional technologies for thermal waste treatment and chemical production, as well as power-to-X technologies (PTX) under uniform conditions. A sensitivity analysis enables additional insights into changes in GWP of feedstock recycling pathways as key assessment assumptions vary.

2. Material and methods

According to ISO standard 14040:2006 (DIN EN ISO 14040:2006, 2009), life cycle assessments include four steps: goal and scope definition, life cycle inventory analysis, life cycle impact assessment and interpretation.

2.1. Goal and scope definition

Goal and scope definition include the evaluation objective, description of the assessed system (system boundaries, functional unit, allocation procedure) and limitations of the assessment.

2.1.1. Objective

The study investigates the environmental impacts of feedstock recycling via the utilization of waste in the form of refuse derived fuel from (1) municipal solid waste (MSW), and (2) recyclable materials rich in plastics (RMP) in a scenario-based case study of Germany. Detailed



Fig. 1. Overview of different loops for plastics recycling in a circular economy (Crippa et al., 2019).

Overview of LCA investigations on feedstock recycling for chemical production.

Source	Technology	Waste fraction	Product application	Reference waste treatment
(Jeswani et al., 2021; Russ et al., 2020)	Pyrolysis	MPW*	Steam cracking	Material recycling; incineration; cement kiln
(Meys et al., 2020)	Liquefaction	Packaging waste (pure plastic)	Refinery input	Material recycling; incineration
(Somoza-Tornos et al., 2020)	Pyrolysis	Polyethylene	Ethylene substitution	Incineration; landfilling
(Broeren et al., 2019)	Pyrolysis; gasification	Recycling residue; MPW*	Fuels; syngas	Incineration; material recycling
(Keller et al., 2020)	Gasification	MSW	МТО	incineration
(Nuss et al., 2013)	Gasification	MSW*	Steam cracking (via FT*)	Sorting; incineration
(Perugini et al., 2005)	Hydrocracking	Polyolefins	Crude oil; heavy residue; naphtha; LPG	Sorting; incineration; landfilling

* MPW – mixed plastic waste, MSW – municipal solid waste, FT – Fischer-Tropsch synthesis.

definition and description of the investigated waste fractions are summarized in the supplementary material S1.

Basis for comparison are (1) conventional waste treatment technologies (incineration) and fossil-based chemical production pathways, as well as (2) PTX-based chemical production pathways. Main objective is the evaluation of changes in the foreground system (i.e. waste treatment and chemical production) with only unavoidable impacts on the background system (i.e. fossil feedstock and energy supply). Note that the study does not provide absolute environmental footprints for individual products, processes or industry sectors. The system boundaries are chosen to reflect the direct effects associated with the application of feedstock recycling technologies without allocation. The environmental impacts of different feedstock recycling pathways can therefore be derived as the deviation in a uniform production system with and without their application.

2.1.2. System description & boundaries

The investigation is based on an exemplary system framework for Germany. Waste treatment and base chemical production are defined as one common system. In consequential LCA, the functional unit of a product system refers to the quantified description of the performance requirements that the product system fulfils (Consequential-LCA, 2015). Thus, the *functional unit* consists of the assumed quantities for waste treatment and major base chemical production (see Table 2). Derivation of the considered waste quantities is specified in the supplementary material 1. Neither the generated waste quantities nor the base chemical demand is considered to be directly associated with the application of feedstock recycling technologies and both are maintained consistent throughout the investigation. Therefore, the inventory modelling is considered consequential, despite not taking societal or market developments into account.

The assessment is performed in two frameworks (see Fig. 2). The *Framework Status Quo* (FSQ) is based on a current production scenario for Germany. Conventional chemical production is based on fossil feedstock utilization (i.e. naphtha, natural gas, heavy fuel oil). Conventional waste treatment of the investigated waste fractions is associated with generation of electricity and steam for district heating. The system balance for electricity and heat (*baseline energy*) is determined by

Table 2

Assumptions for quantities for waste treatment and base chemical production.

Waste treatment	(Destatis, 2020)
Residual municipal solid waste (MSW) Recyclable materials rich in plastics (RMP) Base chemical production	10.519 Mt 3.928 Mt (VCI, 2019)
Lower olefins (C2 to C4)	9.368 Mt
BTX aromatics	2.592 Mt
Methanol	1.130 Mt
Ammonia	3.133 Mt
Hydrogen	0.471 Mt

a reference scenario without application of feedstock recycling and is maintained through all scenarios in the framework. With feedstock recycling application, energy generation from waste is decreased. The offset in the system balance is substituted by energy integration (*substitution energy*). In view of the political and regulatory developments for energy generation in Germany (i.e. priority and maximum feed-in of generated renewable energy, phase out of coal and nuclear energy) (IEA, 2020), natural gas-based electricity and steam supply is considered for substitution energy supply.

The Framework Energy Integration (FEI) assumes a future renewable energy supply to the process industry to reduce GHG emissions via Power-to-Heat (PTH) applications and hydrogen integration. The scope of additional electricity integration is unclear, since there is considerable uncertainty in renewable power projections. Germany's total renewable energy production in Germany in 2019 was 237 TWh, with 127 TWh being wind-based energy (B. Burger, 2020). To achieve carbon neutrality, the German chemical industry projects an annual energy demand of 685 TWh (Geres et al., 2019), almost three times the current renewable energy production. In contrast, system scenarios assume that 8 to 116 TWh renewable energy would be required for PTX and PTH applications in order for Germany to achieve its carbon reduction targets (Fraunhofer IWES, 2015). In this study, an exemplary electricity input of 100 TWh (wind-based) is assumed to reflect a significant, but not unlimited renewable energy availability. The assumption of constant waste treatment, base chemical production and energy balance are maintained.

2.1.3. Limitations

The study is restricted to two specified waste fractions and thus does not reflect the full quantitative potential of feedstock recycling. The study does not consider interactions of chemical production with refinery operation, especially regarding naphtha and heavy oil supply. No specific future renewable energy projection is considered, the applied electricity input (100 TWh wind-based energy) is a solely exemplary assumption. Further effects that are not considered include:

- Developments and projections for waste generation quantity, waste composition and base chemical demand.
- Developments in waste treatment technology (Mechanical-biological treatment, material recovery).
- Developments in conventional chemical production technology (e.g. electric heating).

2.2. Life cycle inventory analysis

The life cycle inventory encompasses foreground system processes (i. e. waste treatment, chemical production, utility processes), background system processes and the applied assessment scenarios. Fig. 3 provides an overview of the foreground system processes and the material streams in the inventory model.

Framework Status Quo (FSQ)

Framework Energy Integration (FEI)



Fig. 2. Illustration of applied system boundaries (foreground system).



Fig. 3. Overview of material processes and flows of the foreground system (thin lines: variable streams; capital lines: fixed streams).

Due to a limited number of industrial scale facilities, variation of applied feedstock and a general lack of applicable data (Punkkinen et al., 2017; Solis and Silveira, 2020), feedstock recycling technologies (i.e. gasification and pyrolysis) should be considered as emerging technologies. Their assessment is characterized by significant uncertainty and should therefore be considered prospective (JA Bergerson et al., 2020; Cucurachi et al., 2018). Data gaps can reasonably be bridged by process modelling, as demonstrated in the associated publication (Keller et al., 2021). But the resulting environmental impact should be considered as an impact potential, indicating how good the technology could perform in relation to the benchmark. This assumes perfect implementation of these technologies on industrial scale. The expected realistic technological performance is lower (Meys et al., 2020). While potential technological shortcomings and their impact on the environmental performance can be quantified and discussed by applying sensitivity analysis, this is beyond the scope of this publication.

2.2.1. Waste treatment

Waste treatment is modelled in EASETECH V3.1.7 (2019) (Clavreul et al., 2014; Lodato et al., 2020). Based on assumed waste compositions, the software enables the waste-fraction orientated mass flow balancing, ensuring material consistency in physical treatment steps and the

reproduction of feedstock compositions for chemical or energy utilization. Two major post-consumer waste fractions are considered in this study namely (1) residual municipal solid waste (MSW) and (2) recyclable materials rich in plastics (RMP). The latter consists mainly of source-separated lightweight packaging waste (i.e. yellow bag under the German Green Dot system).

MSW is either directly thermally treated in incineration plants (MSWI) or upgraded by mechanical-biological treatment (MBT). In MBT, a primarily organic waste fraction is separated from MSW and treated via aerobic fermentation for biogas production, which is subsequently utilized in a CHP unit. Fermentation residue is then treated in MSWI. Metallic components are extracted from the remaining non-organic fraction and a refuse derived fuel (RDF) is obtained after drying.

A material recovery facility (MRF) is considered the primary treatment step for RMP. Recovered materials (i.e. plastics and metals) from MRF are not further considered in this investigation, since they are subject of direct material recycling and the recovered quantities are constant in all frameworks and scenarios. The investigation focuses on the cumulated sorting residue i.e. RDF.

RDF from both MBT and MRF is conventionally utilized in RDF power plants for electricity and heat production. They are also suitable feedstock for chemical utilization via pyrolysis or gasification. Detailed descriptions, modelling assumptions and sources for waste fraction compositions and treatment steps are provided in the supplementary material S1.

2.2.2. Chemical production processes

LCI for all base chemical production processes is generated via detailed process modelling and balancing in order to ensure comparability, transparency as well as consistent mass and energy balances. Detailed modelling assumptions and LCI results are presented in the associated publication (Keller et al., 2021). Non-uniform intermediate products that vary heavily in their compositions and subsequent utilization characteristics (e.g. steam cracking raw product gas and pyrolysis gasoline, syngas, pyrolysis oil) are subject of process modelling and avoided in subsequent simplified mass flow balancing.

Conventional chemical production processes for the considered base chemicals (i.e. olefins, BTX aromatics, methanol, ammonia, hydrogen) include:

- (a) steam cracking of naphtha and LPG,
- (b) steam reforming of natural gas for the production of ammonia and hydrogen,
- (c) partial oxidation of heavy fuel oil for methanol production (which is the primary methanol source in Germany (Ellis, 2018), in contrast to the global prevalence of methanol production from natural gas reforming (Brinkmann et al., 2017)),
- (d) catalytic reforming of naphtha for the production of BTX aromatics.

Feedstock recycling in the form of gasification and pyrolysis of RDF can also be applied to produce the considered base chemicals. Gasification-based syngas can be to applied to replace conventional syngas applications (i.e. ammonia, hydrogen and methanol production). Further, it can be utilized for methanol-based olefin and BTX production (MTO/MTA) and naphtha production via Fischer-Tropsch synthesis.¹ Pyrolysis oil can be upgraded to steam cracking feedstock, or be applied directly for syngas production via partial oxidation. Note that the process characteristics (i.e. CO₂ emissions, product yields) for pathways based on gasification and partial oxidation processes for production of carbonaceous products (methanol, aromatics, olefins, FT naphtha) can be improved via external hydrogen integration (Ostadi et al., 2020; Seidl et al., 2020). The relevant processes are marked with *H*.

Lastly, Power-to-X processes considered in this study include methanol production from CO_2 and hydrogen, subsequent methanol utilization for MTO, and ammonia production from hydrogen and nitrogen.

2.2.3. Utility and background system inventory

An overview of balancing assumptions for required utility processes is provided in Table 3. Germany's electricity production mix is assumed for baseline electricity supply in the Framework Status Quo (JA Burger, 2020). Applied background datasets are summarized in Table 4.

2.2.4. System integration and scenarios

System mass flow balancing, LCI integration and Life cycle impact assessment (LCIA) is performed in GaBi 9.2.0. For both defined frameworks, a reference scenario without application of feedstock recycling pathways is defined. The impacts of feedstock recycling technologies are assessed based on scenarios with their application compared to respective reference scenario. In each respective scenario, the application range of alternative production processes can be restricted by available RDF quantity, target chemical production quantity and/or energy availability. An overview of the assessed scenarios is given Table 5. In Table 3

Modelling assumptions of utility processes.

Oxygen-ASU		
Oxygen purity	99.5 vol%	(Dave and Arné, 2016)
Electricity demand	0.245 kWh (el) / kg	
	Oxygen	
High-purity nitrogen	0.131 kg / kg Oxygen	
production		
Nitrogen-ASU		
Electricity demand	0.2 kWh (el) / kg Nitrogen	(Häring, 2007)
Alkali electrolysis		
Electricity demand	51.17 kWh (el) / kg	(Smolinka et al., 2018)
electrolysis	Hydrogen	
Electricity demand	0.63 kWh (el) / kg	(Wolfersdorf, 2017)
compression	Hydrogen	
Oxygen co-production	8.0 kg / kg Hydrogen	
Feed water supply for Power-to-I	Heat and electrolysis	
Feed water demand	1.175 t / t boiler feed	(Ras and Blottnitz,
	water	2012)
Electricity demand	0.66 kWh / t boiler feed	
	water	
Power-to-Heat		
Energy efficiency	0.99 MJ (th) / MJ (el)	(den Ouden et al.,
		2017)

Table 4

Overview of background processes.

Process Name in GaBi Database Naphtha DE: Naphtha at refinery, aggregated Natural gas DE: Natural gas mix, aggregated Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated Steam substitution DE: Process steam from natural gas 90%, aggregated Electricity substitution DE: Flexificity from patural gas aggregated	Process Name in GaBi Database Naphtha DE: Naphtha at refinery, aggregated Natural gas DE: Natural gas mix, aggregated Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated	Process Name in GaBi Database Naphtha DE: Naphtha at refinery, aggregated Natural gas DE: Natural gas mix, aggregated
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Diesel DE: Diesel mix at refinery, aggregated	Steam substitution DE: Process steam from natural gas 90%, aggregated DE: Electricity substitution DE: Electricity from natural gas aggregated	Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated
Caustic sodaDE: Sodium hydroxide (caustic soda) mix, aggregatedSulphuric acidDE: Sulphuric acid mix (96%), aggregated	Diesel DE: Diesel mix at refinery, aggregated Caustic soda DE: Sodium hydroxide (caustic soda) mix, aggregated Sulphuric acid DE: Sulphuric acid mix (96%), aggregated	Steam substitution DE: Process steam from natural gas 90%, aggregated Electricity substitution DE: Electricity from natural gas, aggregated Diesel DE: Diesel mix at refinery, aggregated Caustic soda DE: Sodium hydroxide (caustic soda) mix, aggregated Sulphuric acid DE: Sulphuric acid mix (96%), aggregated
	Diesel DE: Diesel mix at refinery, aggregated	Steam substitution DE: Process steam from natural gas 90%, aggregated Electricity substitution DE: Electricity from natural gas, aggregated Diesel DE: Diesel mix at refinery, aggregated
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Natural gas DE: Natural gas mix, aggregated Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated Steam substitution DE: Process steam from natural gas 90%, aggregated Elactricity substitution DE: Floating the subscience of the subsc	Natural gas DE: Natural gas mix, aggregated Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated	Natural gas DE: Natural gas mix, aggregated
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Process Name in GaBi Database Naphtha DE: Naphtha at refinery, aggregated Natural gas DE: Natural gas mix, aggregated Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated Steam substitution DE: Floeticity from natural gas 90%, aggregated DE: Process steam from natural gas 90%, aggregated DE: Floeticity from natural gas 90%, aggregated	Process Name in GaBi Database Naphtha DE: Naphtha at refinery, aggregated Natural gas DE: Natural gas mix, aggregated Heavy fuel oil DE: Heavy fuel oil at refinery, aggregated	Process Name in GaBi Database Naphtha DE: Naphtha at refinery, aggregated Natural gas DE: Natural gas mix, aggregated

5

Overview	of	assessed	scenarios

Framework Statu	ıs Quo (FSQ)
FSQ-Ref	Conventional chemical production, partial MSW treatment in MBT
FSQ-G1	RDF gasification for MTO synthesis
FSQ-G2	RDF gasification for methanol synthesis
FSQ-G3	RDF gasification for ammonia synthesis
FSQ-P1	RDF pyrolysis for upgrading, full hydrotreating
FSQ-P2	RDF pyrolysis for partial oxidation
FSQ-G1max	FSQ-G1, 100% MSW treatment in MBT
FSQ-P1max	FSQ-P1, 100% MSW treatment in MBT
Framework Ene	ergy Integration (FEI)
FEI-Ref	Energy integration for PTX MTO synthesis, partial MSW treatment
(PTX1)	in MBT
FEI-PTX2	Energy integration for PTX methanol synthesis
FEI-PTX3	Energy integration for PTX ammonia synthesis
FEI-G1	RDF gasification for MTO synthesis with H ₂ integration
FEI-G2	RDF gasification for methanol synthesis with H ₂ integration
FEI-P1	RDF pyrolysis for upgrading, full hydrotreating
FEI-G1max	FEI-G1, 100% MSW treatment in MBT
FEI-P1max	FEI-P1, 100% MSW treatment in MBT

marked max scenarios, the effect of a larger scope of chemical recycling application on the system is demonstrated by maximizing the amount of RDF generation by MBT of MSW (instead of direct incineration of large quantities of MSW). Note that the investigated scenarios do not include a full systematic variation, but only selected variations to demonstrate isolated effects.

The following assumptions for system integration are made:

¹ Note that LCI for MTA and Fischer-Tropsch processes is included in the associated publication Keller et al. (2021), but not considered in the present investigation to limit system complexity.

- Co-produced LPG is applied for steam cracking, external balancing is avoided.
- Co-produced fuel gas is balanced within the chemical production system, additional demand is met by natural gas supply.
- The external electricity and steam balance are benchmarked by the respective reference scenario and maintained constant in all scenarios within the framework.
- Pyrolysis residue is treated by MSWI.

For the Framework Status Quo:

- Electricity and steam are balanced by natural gas-based generation.
- Oxygen is supplied by cryogenic air separation unit (Oxygen-ASU).

For the Framework Energy Integration:

- Steam is balanced by application of Power-to-Heat.
- Hydrogen is generated by electrolysis.
- Concentrated CO₂ as feedstock is primarily provided by acid gas removal systems from syngas cleaning. Further demand is met by amine-based carbon capture from flue gas.
- High-purity nitrogen as feedstock is primarily provided from Oxygen-ASU. Further demand is met by air separation unit for nitrogen supply (Nitrogen-ASU).

2.3. Life cycle impact assessment analysis

The LCIA and results discussion in this study focuses on global warming potential (GWP). Life cycle GWP impacts are calculated in GaBi V9.2., IPCC AR5 GWP100 including biogenic carbon is applied (Myhre et al., 2014). LCIA results of other impact categories (i.e. fossil resource use, acidification, freshwater eutrophication, photochemical ozone formation, water use) are provided in the supplementary material S2, but are not discussed in this publication.

2.4. Sensitivity analysis

Selected key assessment assumptions are varied in the sensitivity analysis to assess the associated impacts on GWP for different chemical production pathways:

- Electrolysis efficiency
- Waste-to-Energy efficiency (MSWI and RDF incineration in RDF power plant i.e. RDF PP)
- Electricity substitution source

As with the scenarios with unrestricted RDF availability in Section 2.2.4, G1 and P1 scenarios are utilized as examples to illustrate the impacts of varying assumptions for the sensitivity analysis.

3. Results and discussion

LCI results of chemical and waste treatment processes for all considered scenarios are provided in the supplementary material S2.

The calculated compositions of RDF generated from MSW via MBT and RMP via MRF exhibit only minor deviations from a commercial RDF that is considered in this study (compare Table 6). Hence, they are assumed to be similar in this study. The calculated RDF composition is also consistent with other published compositions reported in literature (Montejo et al., 2011; Thomä and Widder (2012)).

The uniform system balance for electricity input and steam output is determined by the respective reference scenario for each framework. The available amount of RDF for feedstock recycling depends on the fraction of MSW that is directed to MBT. Hence, maximum RDF generation (i.e. MaxRDF) is associated with complete avoidance of direct MSW incineration (see Table 7).

3.1. LCIA GWP results of framework status Quo (FSQ)

The GWP results of the FSQ scenarios are summarized in Fig. 4 and Table 8. Note that only selected numbers are shown in the graph, full LCIA results are provided in supplementary material S2. The total GWP for the reference scenario of conventional chemical production and waste treatment is 46.03 Mt CO₂eq. GWP is observed not to be dominated by a single process, but by numerous main sources namely MSW incineration, naphtha steam cracking, naphtha supply, natural gas reforming for ammonia, RDF incineration and natural gas reforming for hydrogen.

Compared to the reference scenario, all feedstock recycling scenarios exhibit a reduced total GWP spanning a range of 2 Mt CO_2eq (i.e. 4.3%). This relative insensitivity in the overall system is determined by the limited availability of RDF in comparison to fossil feedstock input, but also the resulting systemic compensation between chemical production, waste treatment and energy generation. In addition to the absolute GWP balance, the specific GWP balance is calculated as the ratio of total GWP balance to RDF amount applied for chemical recycling. Note that both total and specific GWP balances are specific to the defined assessment framework and thus should not be applied outside of its scope.

Specifically, compared to the reference scenario, gasification-based MTO (FSQ-G1) – in replacing primarily naphtha steam cracking for olefin production – is observed to exhibit the largest total GWP reduction at 0.81 Mt CO₂eq. Gasification-based ammonia production (FSQ-G3) also displayed a similar total and specific GWP reduction to gasification-based MTO at 0.72 Mt CO₂eq and 0.25 t CO₂eq/ t RDF respectively. Gasification-based methanol production (FSQ-G2) shows the highest specific GWP reduction (0.84 t CO₂eq/ t RDF) by replacing GWP-intensive fuel oil partial oxidation. However, the RDF application scope (at only 0.96 Mt out of the available 2.85 Mt) is limited by methanol production quantities (i.e. low substitution potential).

Generally, pyrolysis and pyrolysis oil upgrading exhibit significantly less direct emissions in comparison to gasification-based processes. However, as pyrolysis products are being used to replace fossil feedstock

Table 7

Calculated balances for electricity, steam and produced RDF in the assessment frameworks.

Framework	Electricity	Steam	RDF
	Input	Output	applied
Status Quo (FSQ) Status Quo (FSQ) MaxRDF Energy Integration (FEI) Energy Integration (FEI) MaxRDF	4.66 TWh 4.66 TWh 100.00 TWh 100.00 TWh	39.95 Mt 39.95 Mt 21.61 Mt 21.61 Mt	2.85 Mt 4.76 Mt 2.85 Mt 4.76 Mt

Table 6

Comparison of applied waste composition with calculated RDF compositions from EASETECH modelling.

	Water wt%	Ash wt% (w	C f)	Н	0	N	Cl	S	Sum	LHV MJ / kg (wf)
Applied RDF (ECO20) (Recenso, 2019)	4.6	10.3	52.7	9.0	25.1	2.0	0.7	0.2	100.0	22.3
RDF from MRF (calculated)	5.0	11.4	58.5	8.7	19.6	0.7	1.0	0.1	100.0	23.2
RDF from MBT (calculated)	5.0	15.4	52.7	7.1	22.8	1.7	0.2	0.1	100.0	22.3



Fig. 4. Visualization of GWP results of the scenarios in Framework Status Quo.

Table 8

Summary of GWP results of scenarios in Framework Status Quo.

Scenario	FSQ-Ref	FSQ-G1	FSQ-G2	FSQ-G3	FSQ-P1	FSQ-P2	FSQ-G1max	FSQ-P1max
Total GWP [Mt CO ₂ eq]	46.03	45.22	45.22	45.31	45.56	45.58	43.99	44.55
GWP balance to reference [Mt CO2eq]		-0.81	-0.80	-0.72	-0.47	-0.45	-2.04	-1.48
RDF applied for feedstock recycling [Mt]		2.85	0.96	2.85	2.85	1.06	4.76	4.76
Specific GWP balance [t CO ₂ eq/ t RDF]		-0.28	-0.84	-0.25	-0.17	-0.43	-0.43	-0.31

in conventional processes (i.e. naphtha and LPG steam cracking), emissions associated with conventional processes will thus remain to contribute to the overall emissions along pyrolysis-based pathways. Hence, in contrast to gasification-based scenarios, pyrolysis-based scenarios (FSQ-P1 and FSQ-P2 at 0.47 and 0.45 Mt CO₂eq respectively) exhibit considerably lower total GWP reduction. Additional contributing factors are emissions associated with the incineration of pyrolysis residues, as well as additional hydrogen production demand to upgrade pyrolysis oil to a naphtha equivalent in FSQ-P1 (refer LCI results in supplementary material S2). Similar to FSQ-G2, pyrolysis-based methanol production (FSQ-P2) is observed to have a higher specific GWP reduction than the other investigated pyrolysis pathway. However, the total GWP reduction is also limited by the low production quantities of methanol, which limits the amount of RDF that could be applied (i.e. 1.06 Mt of available 2.85 Mt).

Based on the examples of FSQ-G1 and FSQ-P1, it is observed that via an increase of RDF production via MBT of MSW (i.e. FSQ-G1max and FSQ-P1max) to increase RDF availability for feedstock recycling, the total GWP reduction of both gasification and pyrolysis pathways also amplified (from 45.22 to 43.99 Mt CO₂eq and 45.56 to 44.55 Mt CO₂eq respectively). Moreover, the avoidance of MSW incineration also leads to an increase in the specific GWP reduction potential (from 0.28 to 0.43 CO_2 eq / t RDF and 0.17 to 0.31 t CO₂eq / t RDF respectively).

3.2. LCIA GWP results of framework energy integration (FEI)

With renewable energy integration of 100 TWh, the total GWP is reduced to 25.26 Mt CO_2eq (FEI reference scenario, see Fig. 5 and Table 9). This represents a reduction of 45.1% in total GWP compared to the FSQ reference scenario. Surplus electricity is utilized for the production of 1.60 Mt hydrogen via electrolysis, which is subsequently converted with CO_2 to produce lower olefins via methanol as intermediate (i.e. PTX to MTO). In the FEI-Ref scenario, the application of PTXbased MTO substitutes 34.7% of the total lower olefins production. The rest of the lower olefins demand is thus met by naphtha steam cracking (55.9%) and LPG steam cracking (9.4%).



Fig. 5. Visualization of GWP results of the scenarios in Framework Energy Integration.

Table 9

Summary of GWP results of scenarios in Framework Energy Integration.

Scenario	FEI-Ref (PTX1)	FEI-PTX2	FEI-PTX3	FEI-G1	FEI-G2	FEI-P1	FEI-G1max	FEI-P1max
GWP total [Mt CO ₂ eq] GWP balance to reference [Mt CO ₂ eq]	25.26	24.79 -0.47	23.77 -1.48	21.96 -3.30	24.11 -1.14	23.01 -2.25	19.48 -5.78	21.23 -4.02
Hydrogen applied in PTX MTO [Mt]	1.60	1.42	1.06	1.21	1.49	1.42	0.98	1.33
Hydrogen applied in alternative process [Mt]		0.16	0.60	0.33	0.08	0.07	0.56	0.11
RDF applied for feedstock recycling [Mt] Specific GWP balance [t CO ₂ eq/ t RDF]				2.85 -1.16	0.64 -1.79	2.85 -0.79	4.76 -1.21	4.76 -0.84

Comparing PTX technologies (FEI-Ref, FEI-PTX2 and FEI-PTX3), the scenario for ammonia production exhibits the highest GWP reduction at 1.48 Mt CO₂eq. However, the substitution potential of both methanol and ammonia is limited by their production quantities, thus they required only 0.16 and 0.60 Mt hydrogen respectively. The remaining hydrogen is assumed to be utilized for PTX-based MTO (i.e. 1.42 and 1.06 Mt hydrogen respectively).

By applying RDF gasification for olefin production with hydrogen integration (FR MTO H, in FEI-G1), the GWP reduction increases significantly compared to PTX-based MTO (i.e. 3.30 Mt CO₂eq). The application scope of gasification-based MTO is restricted by the available RDF quantity. Hence, only 1.21 Mt hydrogen is applied for lower olefins production via gasification (i.e. 15.6% of total olefin production). The remaining 0.39 Mt hydrogen is used to produce 26.2% of olefins via PTX-based MTO. Major contributors for GWP reduction are the avoided emissions from RDF incineration, process emissions and feedstock supply for naphtha cracking. The demand for CO₂ supply as feedstock via carbon capture also decreases from 7.23 to 4.33 Mt compared to the FEI reference scenario (40.1% reduction, compare LCI results in supplementary material S2).

Comparing feedstock recycling pathways, similar qualitative results are observed in FEI as in FSQ. Gasification-based methanol production (in FEI-G2) leads to higher specific GWP reduction (1.79 t CO₂eq / t RDF) compared to FEI-G1, but the application range is limited by the methanol production quantity. Compared to methanol production from PTX (FEI-PTX2), the GWP reduction is significantly higher despite the same quantitative application restriction resulting from the limited potential for methanol substitution (1.14 to 0.47 Mt CO₂eq). RDF pyrolysis for naphtha production (FEI-P1) shows lower specific GWP reduction (0.79 t CO₂eq / t RDF) than gasification routes, mainly due to pyrolysis residue incineration and emissions from naphtha cracking. Despite this, it still exhibits significant GWP reduction compared to the FEI reference scenario.

Based on the examples of FEI-G1 and FEI-P1, in increasing RDF

availability via MBT of MSW for feedstock recycling (i.e. FEI-G1max and FEI-P1max), GWP reduction is increased from 3.30 to 5.78 Mt CO₂eq as well as from 2.25 to 4.02 Mt CO₂eq respectively. A slight increase in specific GWP reduction for these two feedstock recycling pathways is also observable. This increase in GWP reduction is mainly due to the avoidance of MSW incineration. Note that despite maximized RDF quantities for feedstock recycling, GWP reduction remains limited by RDF availability. Hence, only 25.9% of total olefin production can be substituted via gasification in FEI-G1max and 21.9% of naphtha feedstock can be replaced via pyrolysis in FEI-P1max (compare supplementary material S2).

3.3. Sensitivity analysis

Sensitivity analyses are carried out – based on the examples of RDF gasification for MTO synthesis (G1), and RDF pyrolysis for upgrading with full hydrotreating (P1) – to illustrate the associated impacts on GWP of feedstock recycling routes resulting from changes in (a) electrolysis efficiency, (b) waste-to-energy efficiency, and (c) electricity substitution source (see Table 10). While (a) is relevant only for the FEI scenarios, (b) relates to the efficiency of MSWI and RDF PP and is thus relevant for both FSQ and FEI scenarios. In contrast, (c) applies only to FSQ scenarios as FEI scenarios are assumed to have an excess of renewable electricity.

a) Electrolysis efficiency

For both gasification and pyrolysis routes, the specific GWP reduction compared to the reference scenario is observed to decrease with higher electrolysis efficiency (i.e. lower electrolysis electricity demand) under FEI. This is determined by the larger applied quantity of hydrogen from electrolysis in PTX scenarios. Hence, the GWP reduction associated with in comparison to the reference scenario will reduce correspondingly.

a) Electricity efficiency MSW / RDF incineration

Under both FSQ and FEI conditions, higher electrical efficiencies of waste-to-energy (WTE) processes (i.e. MSW and RDF incineration) – while maintaining thermal energy efficiency – are associated with a significantly lower GWP reduction of feedstock recycling routes compared to the reference scenarios. This is because higher WTE efficiencies will lead to an increase in the applied baseline energy which will be substituted. Consequently, the GWP reduction associated with feedstock recycling routes in comparison to the reference scenarios will be reduced. Note that the impact is less significant under FEI conditions, where an increase in WTE electricity production is compensated by the applied baseline electricity.

a) Electricity substitution

Table 10

Results of sensitivity analysis.

Specific GWP balance [t CO ₂ eq/ t RDF] compared to the reference scenario					
Criteria	Assumptions	FSQ		FEI	
		G1	P1	G1	P1
a) Electrolysis electricity	160	-	-	-1.139	-0.739
demand [MJ / kg H ₂]	180	-	-	-1.157	-0.788
	200	-	-	-1.171	-0.828
b) Electrical efficiency	8% / 10%	-0.433	-0.315	-1.205	-0.836
MSW / RDF	11% / 15%	-0.283	-0.165	-1.157	-0.788
incineration	14% / 20%	-0.134	-0.016	-1.108	-0.739
c) Electricity substitution	Wind	-0.859	-0.692	-	-
	Natural gas	-0.283	-0.165	-	-
	Lignite	0.498	0.549	-	-

Electricity substitution is only relevant for FSQ scenarios as excess renewable energy is assumed for FEI scenarios. The integration of RDF in chemical production via feedstock recycling – instead of WTE – will require increased demand for external energy supply into the foreground system. While electricity substitution accounts for a major share of the total electricity demand in feedstock recycling scenarios (e.g. 42.8% in FSQ-G1, see supplementary material S2), it is leads to only a minor fraction of the total system GWP (3.7% in the FSQ-G1 scenario) (see Fig. 4). Nevertheless, the substitution electricity source is observed to have a significant impact on the GWP. Not surprisingly, the GWP reduction of feedstock recycling scenarios compared to reference scenario is observed to increase considerably if renewable energy (e.g. from wind) is assumed to be the substitution energy source. In contrast, if lignite-based electricity substitution is assumed, the GWP of feedstock recycling scenarios will exceed that of the reference scenario.

4. Conclusions

To address the challenges and gaps in LCA studies of feedstock recycling pathways, this study introduces an integrated life cycle inventory model which expanded the foreground system to include major processes for waste treatment and base chemical production. The objective is to support a consequential LCA of the GWP of feedstock recycling pathways (i.e. gasification and pyrolysis) in comparison to conventional and PTX-based chemical production pathways. The investigation is based on an exemplary system framework for Germany in terms of applied waste quantities and compositions, base chemical production quantities and energy supply. The assessment is performed within two frameworks namely (1) Framework Status Quo (FSQ) with current conventional waste treatment and chemical production processes as reference scenario, and (2) Framework Energy Integration (FEI) with an exemplary electricity input of 100 TWh and PTX process application as reference scenario.

Results indicate that for both FSQ & FEI conditions, GWP reduction is observed – despite systemic compensation between chemical production, waste treatment and energy generation – for feedstock recycling scenarios compared to the respective reference scenarios. It is shown that the application of feedstock recycling technologies has a limited impact on GHG emission reduction under current conditions (i.e. FSQ), but increases in future scenarios (i.e. FEI) in comparison to PTX applications.

However, the GWP reduction of feedstock recycling is limited by RDF availability under the applied system conditions. While methanol production via feedstock recycling exhibited the highest specific GWP reduction, the total GWP reduction for this route is limited by the low methanol production. Gasification-based production pathways exhibit higher GWP reduction compared to pyrolysis-based routes.

A sensitivity analysis enables additional insights into the impacts on GWP of feedstock recycling routes resulting from changes in electrolysis efficiency, waste-to-energy efficiency, and electricity substitution source. Results suggested that GWP in FSQ scenarios are significantly determined the electrical efficiency of thermal waste treatment and the source of electricity substitution. While GWP in FEI scenarios is also impacted – albeit to a lesser extent compared to FSQ – by the electricity efficiency of thermal waste treatment, feedstock recycling routes indicate a lower sensitivity to changes in electrolysis efficiency than PTX technologies. Consequently, it will be worthwhile to integrate a more detailed consideration of waste-to-energy technology advancements in future studies.

This investigation enables a perspective on the environmental viability of feedstock recycling processes through a comparative assessment with conventional and PTX-based production routes. To address existing methodological issues from previous LCA studies on feedstock recycling, the current assessment applied uniform system frameworks, accounting for differences in production characteristics (e. g. product yields and utility demands) with minimized allocation

assumptions. However, the investigation is prospective and results should be considered as impact potentials assuming optimal technology development and application. Despite uniform conditions for inventory generation for chemical processes via process modelling, a lack of largescale demonstration and missing industrial validation data leads to significant data uncertainty for feedstock recycling and PTX processes. To improve robustness of assessment results for significant conclusions to support decision processes, future research will benefit from improved data availability and extended uncertainty analysis of technology characteristics and performance.

CRediT authorship contribution statement

Florian Keller: Conceptualization, Methodology, Validation, Investigation, Writing – original draft, Visualization. **Raoul Lukas Voss:** Validation, Investigation, Writing – original draft. **Roh Pin Lee:** Conceptualization, Writing – review & editing, Project administration, Funding acquisition. **Bernd Meyer:** Conceptualization, Writing – review & editing, 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.

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Supplementary materials

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