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Assessing Chemical Recycling Regarding Sustainability Impacts and its Role in the Circular Economy

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Abstract

The transition from a linear to a circular economy to address the growing waste crisis, mitigate climate change, and conserve the planet's limited resources is given a top priority on the global agenda. The European Union seeks to lead the way by implementing its Circular Economy Action Plan as part of the European Green Deal. This action plan aims to promote high-quality recycling with the ultimate goals of transforming industrial value chains into value cycles, achieving net-zero emissions by 2050, and decoupling economic growth from resource consumption. However, the current recycling efforts primarily focus on source-separated plastics, while heterogeneous municipal solid waste fractions remain disregarded. This imbalance leads to significant resource depletion and greenhouse gas emissions as approximately 81% of heterogeneous municipal solid waste in the European Union is treated using landfilling or incineration.

Chemical recycling—a broader term for the reconversion of waste into molecular chemical building blocks to become feedstocks for the chemical industry—is now being brought to the table as a complement to conventional recycling that could reduce linear waste treatment and fossil feedstock use in numerous chemical production systems (i.e., plastics, fertilizers, pharmaceuticals). Specifically, gasification and pyrolysis represent potential chemical recycling solutions for heterogeneous waste streams due to their robustness towards feedstock impurities. As they own the potential to reduce greenhouse gas emissions and facilitate resource conservation by recirculating carbon into the chemical production, they may present sustainable circular economy alternatives to landfilling and incineration. However, because extant chemical recycling literature narrows down to isolated technical assessments of chemical recycling for pure plastic waste streams, science-based data on the environmental, economic, and social sustainability of applications to heterogeneous waste streams are lacking.

This dissertation aims to extend the literature with multidimensional, multidisciplinary, and systemic insights into chemical recycling sustainability including the development of corresponding assessment tools. Specifically, it applies process-based evaluations based on attributional life cycle assessments and techno-economic analyses to generate attributive plant-level data that support industrial decision-makers in inter-technology comparisons. Secondly, it uses systemic evaluations based on systemic life cycle assessments and agent-based modeling to generate consequential country-level data that provide policy-makers with information about the drivers and impacts of extensive chemical recycling deployments. As a case study, residual municipal solid waste treatment

and unsorted lightweight packaging treatment in Germany, a pioneering nation in the implementation of sustainable waste management policy, are examined.

The results indicate that chemical recycling can prospectively improve the environmental sustainability of waste management for heterogeneous streams compared to the linear modus operandi of treatment. Today, waste incineration with energy recovery seems to be a rather sustainable alternative to the conventional production of electricity and heat. However, this perception will gradually diminish as the German energy system shifts towards renewables due to the German Energy Transition (“Energiewende”). In contrast, the deployment of chemical recycling shows long-lasting positive impacts on climate change, terrestrial acidification, and fossil resource scarcity, in addition to positive labor effects. However, its diffusion depends on adjustments in regulatory framework conditions to bridge the cost gaps to waste incineration. In this regard, the introduction of a chemical recycling quota is effective and thought to accumulate more positive sustainability effects compared to the integration of municipal solid waste incineration into emission trading, which is currently being considered in Germany and the European Union.

By identifying heterogeneous waste streams as suitable feedstocks for chemical recycling, this dissertation extends the spectrum of technology applications now being discussed in the academic literature and socio-political debates. Specifically, detailed scenarios addressing potential roles for chemical recycling in the circular economy can support its integration into mature and regulated industrial sectors of waste management and chemical production. Additionally, developed models for life cycle assessment, techno-economic analysis, and life cycle sustainability assessment can serve as a blueprint for future comparative evaluations of chemical recycling to alternative waste treatment options. Summarized, this dissertation encourages scholars, industry representatives, and policy-makers to intensify their commitment to evaluating, developing, and promoting the global circular economy integrated with innovative chemical recycling techniques.

Content

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Abbreviations

CC	Climate change
CEAP	Circular Economy Action Plan
DPP	Dynamic payback period
EU	European Union
FCI	Fixed capital investment
GWP100	Global warming potential (100 years)
IF	Impact factor
LCCA	Levelized costs of carbon abatement
LCA	Life cycle assessment
LCSA	Life cycle sustainability assessment
LWP	Lightweight packaging
MSW	Municipal solid waste
NPV	Net present value
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
PVC	Polyvinyl chloride
RDF	Refuse-derived fuels
rMSW	Residual municipal solid waste
TEA	Techno-economic analysis
UN	United Nations

1. Introduction

1.1. Background

Prevalent linear economies follow a take-make-dispose logic in dealing with valuable natural resources as feedstocks for their production [1, 2]. Specifically, in linear economies, natural resources pass through the steps of extraction, production, distribution, and consumption before ending up as waste [3]. Today, waste is predominantly disposed of via landfilling and incineration, which leads to multiple environmental challenges including greenhouse gas emissions, resource depletion, and harm to human health and safety [2, 4, 5]. According to recent figures, only 47% of the waste produced by households in the European Union (EU) is recycled [6, 7]. Additionally, heterogeneous municipal solid waste (MSW) including residual MSW (rMSW) or residues from lightweight packaging (LWP) waste sorting only achieves a 9% recycling rate [8]. As MSW volumes in the EU and worldwide are expected to increase further, reaching a global annual production volume of 3.5 billion tonnes by 2050 (cf. Figure 1), waste treatment sustainability becomes an urgent priority for many nations and intergovernmental organizations [9, 10]. They aim to transform the linear modus operandi of environmental pollution and resource depletion into a modern circular economy[1] contributing to emission reduction and resource efficiency by reintegrating waste into production systems as a secondary resource [11, 12].

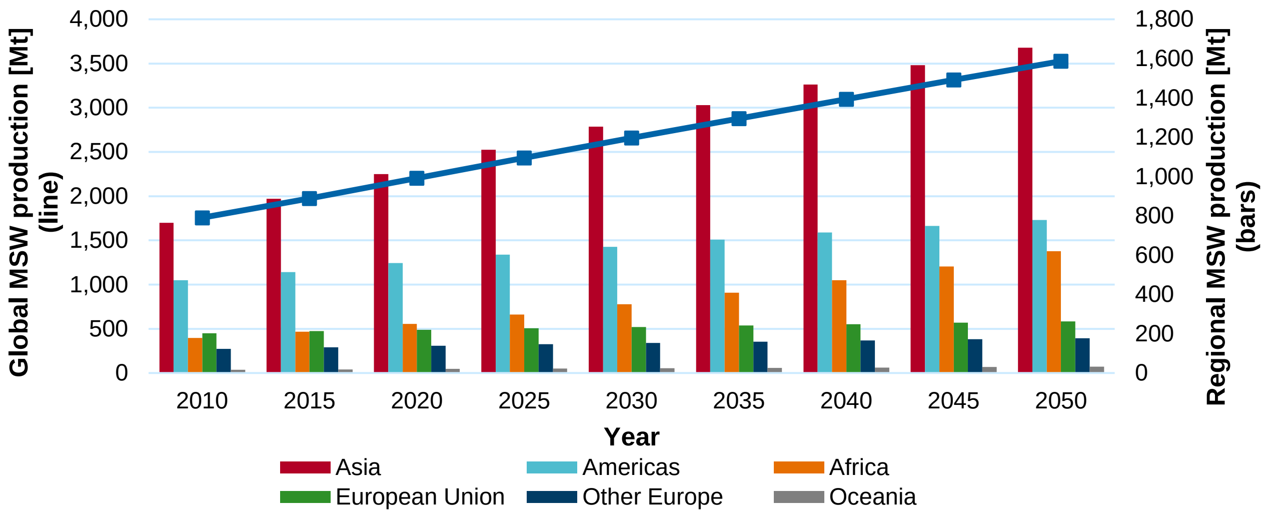

Figure 1: Global and regional production volumes for municipal solid waste (MSW) from 2010 until 2050 [9, 10]

[1] The circular economy perceives waste as a resource for production rather than an inevitable residue of consumption to reduce negative sustainability impacts and to build long-term resilience. Note that its global implementation is recognized, inter alia, by the United Nations (UN) as essential to achieving the UN Sustainable Development Goals [11, 2].

The EU aims to take a pioneering role in the transformation to a sustainable circular economy with its goals set in the Circular Economy Action Plan (CEAP) as part of the European Green Deal to decouple economic growth from resource use and to achieve net zero emissions by 2050 [13, 14]. A strategic cornerstone of the CEAP is the European Waste Hierarchy from reduce, reuse, recycle, and recover (e.g., energy recovery per incineration), to landfill that guides waste management legislation for the entire MSW spectrum (cf. Figure 2) [15, 16]. However, with the public and media focus on pure plastic waste streams, such as micro- and macroplastic on land and at sea, the European Commission and European member states currently primarily address the top of the hierarchy by incrementally phasing out single-use plastics, such as drinking straws or polystyrene beverage containers, and setting ambitious targets for plastic recycling [17]. In contrast, the circularity of heterogeneous post-consumer fractions that are currently mainly landfilled or incinerated represents a major challenge even for EU members that invest heavily in their waste management systems [18]. In upcoming years, efforts and regulations must be extended from reduction, reuse, and recycling of pure plastic waste to corresponding efforts for heterogeneous fractions based on innovative recycling concepts that complement the available mechanical recycling techniques[2] [20].

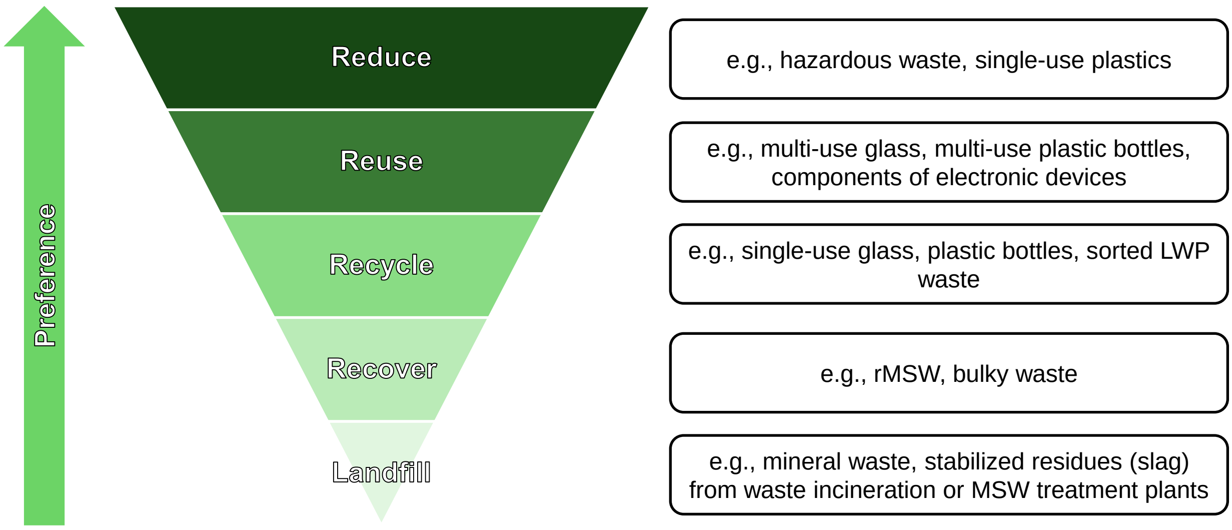

Figure 2: European Waste Hierarchy [18, 21]. LWP: Lightweight packaging. MSW: Municipal solid waste. rMSW: Residual MSW

[2] Mechanical recycling refers to the mechanical/thermal conversion of homogeneous and pure waste streams into secondary materials without changing the basic chemical structure as, for example, by the conservation of complete polymer structures in plastic waste [19]. The robustness of corresponding techniques against feedstock impurities is generally low, which disqualifies them for application to heterogeneous waste fractions [20].

Chemical recycling—breaking down carbonaceous waste into basic chemical building blocks to produce new chemical products with conventional quality including plastics, fertilizers, and pharmaceuticals—now receives increasing attention from science, industry, and civil society as a complement to mechanical recycling which is generally more robust and not prone to “downcycling”[3] [14, 22–31]. Based on Seidl et al. [22], chemical recycling processes can be classified into dissolution, depolymerization, pyrolysis, and gasification (cf. Table 1). While all processes share the principle of decomposing waste to the molecular level, significant differences exist in the individual process parameters. These include process temperatures, pressure conditions, catalyst requirements, and product outputs [22, 32, 33]. Concomitantly, processes show varying sensitivity to feedstock heterogeneity with gasification and pyrolysis being most robust due to, inter alia, high process temperatures [22]. Accordingly, gasification- and pyrolysis-based treatments could present alternatives to linear treatment for heterogeneous MSW fractions, such as residual MSW or unsorted lightweight packaging (LWP) waste [26, 34, 35]. The first practical applications for the respective concepts can be observed, for instance, in Germany, the Netherlands, or Canada [36–41]. To further illuminate these technological paths for decisions about regulation or large-scale investments, and to evaluate which role chemical recycling could play in the circular economy, robust quantitative insights into the impacts in the environmental, economic, and social realms are now being called for in political and public debates [29, 30, 42, 43].

Elaborate plant-focused and systemic technology assessments can support the identification of chances and pitfalls for gasification- and pyrolysis-based chemical recycling for heterogeneous waste fractions [20, 34, 44, 45]. The design of corresponding assessments needs to simultaneously consider technological facets concerning environmental, economic, and social sustainability, to effectively support academics, policy-makers, and industrial representatives in today’s complex socio-political decision arenas of waste treatment and chemical production [46–48]. Academic literature that has developed sophisticated product/technology assessment approaches, such as life cycle assessments (LCA), techno-economic analysis (TEA), life cycle sustainability assessments (LCSA), or computer-based system modeling could contribute a reliable compass for multidimensional sustainability evaluations of innovative chemical recycling concepts [49–51]. However, corresponding

[3] Downcycling refers to losses of material quality during mechanical recycling due to, e.g., impurity accumulations [19].

research, especially for the application of chemical recycling to heterogeneous waste, is still underrepresented and underdeveloped as discussed in the following section.

Table 1: Overview of waste treatment technologies [22, 32, 33]. MSW: Municipal solid waste. PE: Polyethylene. PET: Polyethylene terephthalate. PP: Polypropylene. PS: Polystyrene. PVC: Polyvinyl chloride. RDF: Refuse-derived fuels

	Mechanisms	Feedstock purity	Temp [°C]	Agents	Product	Emissions
Mechanical recycling	Agglutination, extrusion, cooling	High purity polymers (e.g., PS, PET)	200–240	-	Recyclates	
Chemical recycling						
Dissolution	Selective solvent-based dissolution	Target soluble polymers (e.g., PVC, PE)	90–280	Solvents	Polymers	
Depolymerization	Reverse polymerization	High purity polymers (e.g., PS, PET)	80–280	Solvents; enzymes	Monomers; oligomers	
Pyrolysis	Thermochemical decomposition	Mixed plastics (pref., PE, PP, PS)	350–550	-	Oil; hydrocarbons	
Gasification	Partial oxidation	Carbonaceous waste: mixed plastics, RDF; MSW, biomass	1000–1600	Gasifying agents (O_2, H_2O, CO_2)	Syngas (CO, H_2)	
Energy recovery	Incineration (full oxidation)	Carbonaceous waste: mixed plastics, RDF; MSW; biomass	1000	Fuel oil	Electricity/heat	

1.2. Literature review

To gain an overview of the extant literature on chemical recycling technology assessments, a systematic literature review is conducted to identify relevant studies in international peer-reviewed journals for subsequent analysis (cf. Annex for detailed review information). Figure 3 displays all 110 identified studies in a bar chart based on their year of publication, highlighting the growing academic interest in the field. While publication output is at a low level from 2000 to 2017, a significant increase can be observed after 2018, potentially caused by rising political and societal interest in the issues of linear plastic consumption, ocean waste, and microplastics [2, 11, 52]. Despite the growing engagement in the field, an in-depth analysis of all included studies reveals four key literature gaps that are presented in the following four subsections. Afterward, the four central objectives of this dissertation are derived from the identified gaps (cf. Section 1.3).

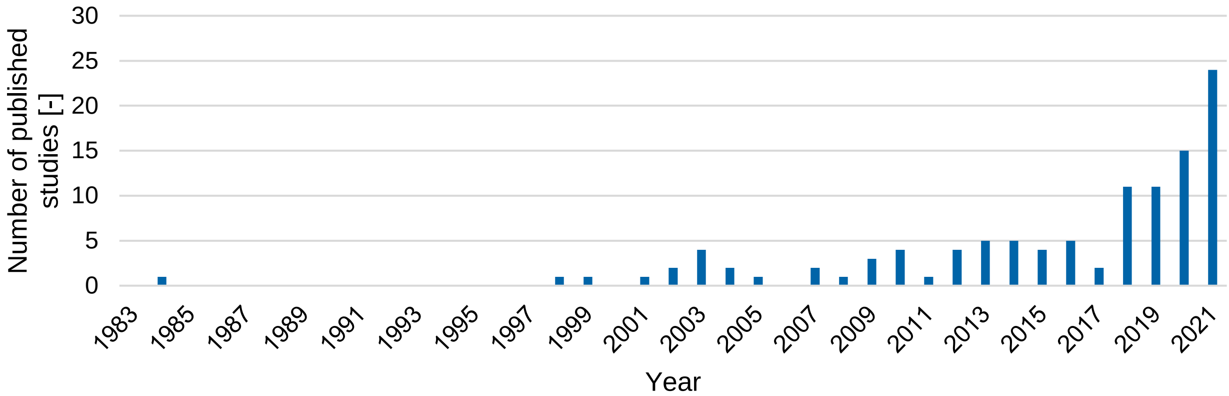

Figure 3: Published studies in chemical recycling technology assessments

1.2.1. Focus on assessments of chemical recycling for pure plastic streams using selective chemical recycling processes

The existing literature is dominated by assessments of chemical recycling for pure plastic streams using selective chemical processes that might put chemical recycling in competition with mechanical recycling (cf. Research gap 1 in Table 2). For instance, several studies assess and discuss chemical recycling as an alternative to conventional recycling and thereby neglect the fact that chemical recycling could complement conventional recycling rather than replacing it by focusing on applications to heterogeneous waste fractions. Specifically, assessments primarily address bottles made from polyethylene terephthalate (PET), foams made from polyurethane (PU), or pure streams of packaging waste made from polyethylene (PE) or polypropylene (PP) as feedstocks (cf. Figure 4). In summary, the analysis shows that n = 86 (~78%) studies exclusively address pure waste streams while only 24 (~22%) address heterogeneous streams. Additionally, a significant imbalance concerning the consideration of individual chemical recycling technologies can be observed (cf. Figure 4). Specifically, n = 41 (~37%) studies conduct technology assessments for depolymerization and n = 33 (~30%) studies for pyrolysis. In contrast, gasification and dissolution are only assessed n = 23 (~21%) and n = 12 (~11%) times, respectively. This focus on depolymerization and pyrolysis excludes alternative chemical recycling approaches such as gasification as potential technical solutions for chemical recycling. Because gasification is particularly applicable to heterogeneous and impure waste fractions due to high process temperatures (cf. Table 1), approaches to assess potentially attractive chemical recycling applications to waste fractions that are predominantly landfilled or incinerated today (cf. Section 1.1), are neglected in extant literature.

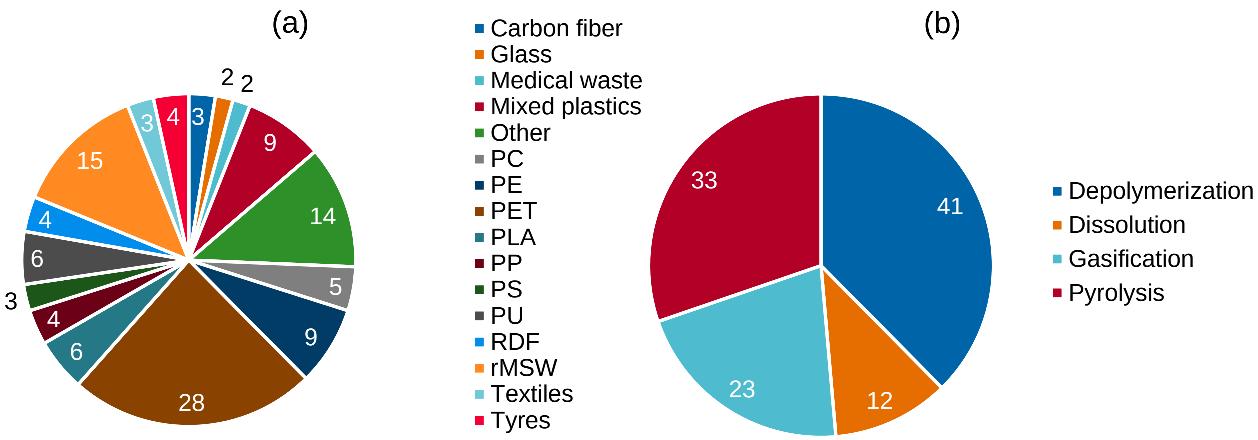

Figure 4: Addressed (a) waste fractions and (b) chemical recycling technologies. PC: Polycarbonate. PE: Polyethylene. PET: Polyethylene terephthalate. PLA: Polylactic acids. PP: Polypropylene. RDF: Refuse-derived fuels. rMSW: Residual municipal solid waste

1.2.2. Focus on unidimensional analyses of technical issues for chemical recycling technologies

A clear imbalance regarding the consideration of individual assessment dimensions is noticeable (cf. Research gap 2 in Table 2). Specifically, the majority of studies conduct unidimensional technical assessments of chemical recycling technologies while ignoring relevant sustainability factors. For instance, some articles thoroughly cover process performance analyses to determine optimal recycling process conditions (e.g., temperature, residence time, mass loss, the impact of waste input characteristics, plant design specifics) with experimental, process modeling, or numerical simulation approaches, but rarely discuss the process sustainability including the impacts on climate change or the economic viability of firms. Summarized, n = 79 (~72%) studies solely conduct technical process analyses or process modeling for chemical recycling technologies without reflecting on the environmental, economic, and social impacts of technology implementation via multidimensional and interdisciplinary research approaches (cf. Figure 5). This dominance of technical assessments significantly narrows down the potential of extant research to, for instance, support inter-technology decisions by industrial representatives to eventually balance their individual sustainability targets including greenhouse gas emission reduction.

1.2.3. Focus on isolated process evaluations that ignore systemic framework conditions and sustainability impacts

Applied methods in the literature neglect systemic framework conditions that will significantly impact whether or not chemical recycling will be applied on a larger scale and how systemic sustainability impacts unfold (cf. Research gap 3 in Table 2). Specifically, the vast majority of assessments (n = 93; ~85%) have a laboratory background that excludes relevant information on systemic boundaries for technology development and deployment, such as competition from other waste treatment and chemical production techniques in an industrial system. This emphasis on isolated laboratory assessments without the acquisition and integration of systemic knowledge impairs the generation of solid insights into the drivers and consequences of a country-level deployment of chemical recycling which are highly relevant for policy-makers.

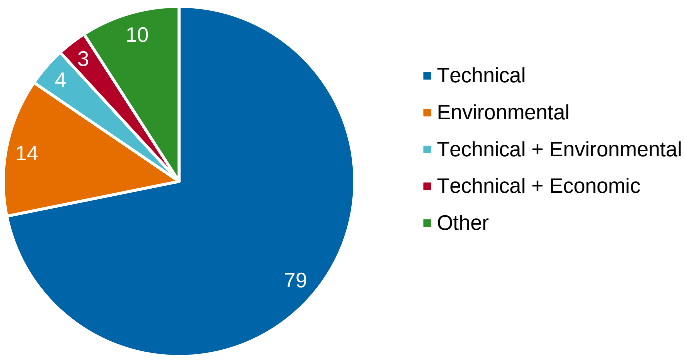

Figure 5: Addressed assessment dimensions

1.2.4. Lack of theoretical linkages between chemical recycling and the circular economy

To date, the existing literature fails to bridge the two worlds of chemical recycling and circular economy by comprehensively assessing reasonable roles for chemical recycling in the circular economy concerning the applied technology types (i.e., dissolution, depolymerization, pyrolysis, and gasification) in combination with targeted waste feedstocks (e.g., homogeneous/heterogeneous MSW fractions) and recycling products (e.g., methanol, naphtha, olefins) (cf. Research gap 4 in Table 2). A vast majority of the reviewed studies (n = 99; ~90%) fail to provide meaningful insights into the compatibility of individual chemical recycling applications with circular economy concepts as they either take an open-loop perspective without considering the uptake of recycling products (i.e., market demand for products) or fail to measure the contribution of developed concepts to central circular economy objectives including emission reduction, fossil resource-saving, and economic competitiveness. However, the corresponding information is invaluable to, among others, academics facing the challenge to theorize about potential transition pathways toward waste management sustainability and circularity.

1.3. Objectives

The four central objectives of this dissertation are presented in Table 2 and described in more detail below. First of all, this dissertation aims to extend the focus of extant literature on chemical recycling for pure plastic waste streams to chemical recycling for heterogeneous and “dirty” waste streams using robust chemical recycling processes (Objective 1 in Table 2). Specifically, technological options to address rMWS and unsorted LWP waste are explored, evaluated, and discussed to spotlight innovative and potentially more sustainable treatment alternatives to landfilling or incineration and to provide chemical recycling interest groups with a more comprehensive view of chemical recycling potentials.

The second objective is to extend the emphasis of current research on unidimensional analyses of technical characteristics for chemical recycling, such as product yields/qualities, to multidimensional analyses of chemical recycling sustainability represented by, inter alia, the impact on climate change, economic aspects, and the labor market (Objective 2 in Table 2). Specifically, methods for sustainability assessments based on LCA, TEA, and LCSA are developed and applied to generate practice-relevant information on the multifaceted sustainability impacts of chemical recycling and reliable tools to measure them.

The third objective is to extend the focus of current research from isolated laboratory evaluations on the plant-level to systemic assessments on the country-level to investigate scenarios of extensive chemical recycling deployments including the sustainability consequences of systemic change and challenges to technology diffusion (Objective 3 in Table 2). Specifically, systemic assessment approaches using computer-based simulations are developed, applied, and discussed to provide novel insights into potential barriers and accelerators for large-scale chemical recycling deployment. Concurrently, the effects of emergent regulatory and market dynamics as complex uncertainties in the diffusion process are highlighted.

The final objective is to link chemical recycling to the circular economy by identifying functional implementation options and evaluating chemical recycling technologies against the backdrop of national/transnational goals to implement the circular economy (Objective 4 in Table 2). Specifically, systemic models on the country-level are used to identify promising roles for chemical recycling to effectively close industrial material cycles and to achieve sustainability goals, such as climate neutrality or the decoupling of economic growth from resource use. Additionally, potential ways to harmonize chemical recycling with strategic cornerstones to circular economy achievement, such as the European Waste Hierarchy (cf. Figure 2), are discussed.

Table 2: Identified research gaps and associated objectives

No	Research gaps	Dissertation objectives
1	Focus on assessments for chemical recycling of pure plastic streams using selective chemical recycling processes	Explore applications for chemical recycling of heterogeneous and impure waste fractions using robust chemical recycling technologies
2	Focus on unidimensional analyses of technical issues for chemical recycling technologies with laboratory experiments	Develop and apply multidimensional tools to gain quantitative data on the environmental, economic, and social impacts of chemical recycling processes
3	Focus on isolated laboratory evaluations that ignore systemic framework conditions and sustainability impacts for country-level chemical recycling	Deliver tools to assess systemic chances and pitfalls for the integration of chemical recycling into existing waste treatment and chemical production systems
4	Lack of theoretical linkages between chemical recycling and the circular economy	Link chemical recycling to the circular economy by identifying functional implementation options

1.4. Outline

As shown in Table 3 and described in detail below, efforts to achieve the four pivotal objectives of this dissertation can be grouped into two categories, each represented by two publications in international, peer-reviewed journals: 1) initial multidimensional evaluations based on attributional LCA and TEA modeling principles that generate high-resolution data on the plant-level, and 2) extended systemic evaluations based on consequential system modeling principles that integrate plant-level insights into country-level assessment models. Note that Table 3 also includes two references to related assessment studies that are not the focus of this dissertation as they address slightly different aspects of the research object. Nonetheless, they can broaden the perspective on the overall topic with information about public perceptions of chemical recycling and the chemical recycling potential for promoting zero waste cities in China.

– <u>Plant-focused evaluations (cf. Section 2):</u> Publications 1 and 2 investigate chemical recycling for heterogeneous waste fractions on a plant-level using attributional LCA and TEA oriented towards ISO standard 14040:2006 [53]. Specifically, Publication 1 investigates the environmental impacts and economic aspects of chemical recycling for rMSW in Germany. Additionally, Publication 2 investigates the environmental impacts and economic aspects of chemical recycling for unsorted LWP waste in Germany. For both studies, detailed process inventories including mass/energy balances and labor needs are generated. Furthermore, inventories for current best-practice treatment pathways using waste incineration are developed to facilitate comparative assessments. As both publications provide quantitative information on chemical recycling for primarily heterogeneous waste fractions, they both contribute to the achievement of Research objective 1 (Feedstock spectrum extension). Additionally, as both studies conduct assessments for multidimensional sustainability impacts—environmental and economic—including the development of corresponding models/tools, they contribute to the achievement of Research objective 2 (Multidimensional analyses).

- Systemic evaluations (cf. Section 3): Publications 3 and 4 transfer previously generated knowledge on the plant-level to systemic computer models to facilitate insights into the preconditions and consequences of extensive chemical recycling deployments. Specifically, Publication 3 investigates the global warming impact of systemic gasification-based chemical recycling for rMSW and unsorted LWP waste in Germany compared to pyrolysis-based chemical recycling and the status quo of incineration-based treatment. The approach is considered consequential from the perspective of the system boundary definition as the modeled system includes both waste treatment and base chemical production in Germany (i.e., the model accurately reflects the impacts of extensive chemical recycling deployment on both the waste management industry and the base chemical production industry). Although the results can help political representatives to consider the trans-sectoral effects of technology diffusion to eventually achieve country-level sustainability goals, the drivers of and barriers to technology deployment that are highly significant for designing effective waste management regulations are not reflected. To fill this gap, Publication 4 includes a systemic model that couples LCSA—to generate insights into multiple environmental, economic, and social sustainability impacts—with agent-based modeling to replicate the technology diffusion process for chemical recycling realistically from the bottom up. Specifically, to eventually provide political representatives with information on the effectiveness of their regulatory measures, chemical production sites are represented as individuals in a competitive market environment of waste treatment that is, inter alia, affected by market and societal developments. As both modeled approaches 1) generate quantitative information on chemical recycling for heterogeneous waste fractions, 2) apply LCA/LCSA principles to attain insights about environmental, economic, and social impacts of chemical recycling, 3) conduct systemic evaluations using sophisticated computer simulations, and 4) help to identify roles for chemical recycling in the circular economy, they both contribute to the achievement of Research objectives 1 to 4, respectively.

In the following Sections 2 and 3, all four publications are thoroughly integrated into the framework of this dissertation by providing more detailed information on the individual study aims, applied methods, and obtained results.

Table 3: Published contributions. ALP: Author list position. CC: Climate change. CEL: Circular economy link. DPP: Dynamic payback period. FCI: Fixed capital investment. FRS: Fossil resource scarcity. FSE: Feedstock spectrum extension. IF: Impact factor. ILE: Impact on local employment. LCCA: Levelized cost of carbon abatement. MA: Multidimensional analyses. NPV: Net present value. NRY: Not rated yet. SA: System analyses. SC: System costs. TA: Terrestrial acidification.

No	Title	Journal (IF)	Year	ALP	Applied methods	Addressed objectives			
						FSE	MA	SA	CEL
Plant-focused evaluations (Section 2)									
1	Global warming potential and economic performance of gasification-based chemical recycling and incineration pathways for residual municipal solid waste treatment in Germany	Waste Management (8.8)	2021	1/5	Attributional life cycle assessments (Indicator: CC) and techno-economic analyses (Indicators: FCI, NPV, DPP, LCCA)	•	•		
2	Chemical recycling of plastic waste: Comparative evaluation of environmental and economic performances of gasification- and incineration-based treatment for lightweight packaging waste	Circular Economy and Sustainability (NRY)	2022	1/3	Attributional life cycle assessments (Indicators: CC, TA, FRS) and techno-economic analyses (Indicators: FCI, NPV, DPP, LCCA)	•	•		
Systemic evaluations (Section 3)									
3	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	Resources, Conservation & Recycling (13.7)	2022	2/4	Consequential systemic life cycle assessment (Indicator: CC) combining process and life cycle modeling	•	•	•	•
4	A consequential approach to life cycle sustainability assessment with an agent-based model to determine the potential contribution of chemical recycling to UN Sustainable Development Goals	Journal of Industrial Ecology (7.8)	2022	1/3	Consequential systemic life cycle sustainability assessment (Indicators: CC, TA, FRS, SC, ILE) combining process, life cycle, and agent-based modeling	•	•	•	•
Related publications									
(5)	Perception of chemical recycling and its role in the transition towards a circular carbon economy: A case study in Germany	Waste Management (8.8)	2021	3/3	Qualitative survey and semi-structured workshop discussions with stakeholders	•			
(6)	Sustainable waste management for zero waste cities in China: Potential, challenges and opportunities	Clean Energy (4.6)	2020	4/4	Literature and database research	•			•

2. Plant-focused evaluations

As a first step, high-resolution insights into the sustainability impacts of chemical recycling are generated by putting the assessment scope on individual plant configurations for the integration of chemical recycling with current waste treatment processes. The generated inventory data and attained assessment results stand alone, but also form the basis for subsequent systemic assessments (cf. Section 3).

2.1. Publication 1: Global warming potential and economic performance of gasification-based chemical recycling and incineration pathways for residual municipal solid waste treatment in Germany

The first scientific paper[4] was submitted to *Waste Management* by *Elsevier* in August 2020 and was accepted for publication in July 2021. The reported study assesses the multidimensional impacts of chemical recycling on rMSW in Germany, representing a heterogeneous fraction that is conventionally processed via incineration-based treatment pathways leading to significant greenhouse gas emissions and secondary resource depletion [54]. Today, chemical recycling is discussed as a solution to reuse carbon from rMSW as a resource for chemical production [55]. However, the current literature does not provide information about the associated environmental impacts or economic aspects as it focuses on technical assessments of chemical recycling for homogeneous plastic waste fractions (cf. Section 1.2). To address this gap, the central aims of this publication are to:

[4] Voss R, Lee RP, Seidl L, Keller F, and Fröhling M. 2021. Global warming potential and economic performance of gasification-based chemical recycling and incineration pathways for residual municipal solid waste treatment in Germany. Waste Management, Vol. 134: 206–19. doi: 10.1016/j.wasman.2021.07.040.

Contributions

Conceptualization: Voss R, Lee RP, Fröhling M; Methodology: Voss R, Fröhling M; Formal analysis and investigation: Voss R; Additional data contributions: Seidl L, Keller F; Writing – original draft preparation: Voss R; Writing – review and editing: Voss R, Lee RP, Fröhling M; Funding acquisition: Lee RP; Supervision: Lee RP, Fröhling M.

Note that the methods and results were presented and discussed at the *GOR-Online-Workshop 2020* at the *Ruhr-Universität Bochum:* Voss R, Lee RP, and Fröhling M 2010. Integrierte Bewertung des chemischen Recyclings von Restabfällen in Deutschland: Emissionsreduktionspotenzial und Kosten. Presented at the GOR-Online-Workshop 2020 at the Chair of Energy Systems and Energy Economics at Ruhr-Universität Bochum, 8 October, Bochum. https://www.ee.rub.de/webseitecs5/Aktuelles_Veranstaltungen/2020-10-06_PresentationDoktorandenSeminar_Voss.pdf.

- extend currently discussed feedstocks for chemical recycling by rMSW,
- deliver insights into the multidimensional impacts of chemical recycling (i.e., environmental and economic) and how to attain them,
- and generate valuable inventory data for rMSW treatment applicable to future investigations.

Methodologically, a comprehensive inventory dataset is developed based on technical reports by the German government, scientific publications, and LCA databases to assess three different treatment pathways (cf. Figure 6), namely:

- direct incineration of rMSW in a municipal solid waste incinerator for energy recovery,
- indirect incineration of refuse-derived fuels (RDF)—produced from rMSW in a mechanical-biological treatment plant—in an RDF power plant for energy recovery,
- and chemical recycling of RDF—produced from rMSW in a mechanical-biological treatment plant—in a gasification plant for the recovery of chemical intermediates.

Drawing on the generated dataset, a custom LCA model is subsequently developed with EASETECH V3.1.7., an LCA software provided by the Technical University of Denmark (DTU) that is well-established in research [56–58], to allow for conclusions to be made about the impact on climate change per indicator global warming potential (GWP100) as described in the Fifth Assessment Report of the IPCC [59, 60]. Additionally, a custom economic assessment model is developed in Microsoft Excel to assess the economic sustainability of all three pathways using the indicators fixed capital investment (FCI), net present value (NPV), dynamic payback period (DPP), and levelized cost of carbon abatement (LCCA) [61–64].

The publication provides relevant multidimensional insights into the chemical recycling of heterogeneous waste fractions and how to attain them. Specifically, the results indicate that chemical recycling of rMSW—the same as incineration-based energy recovery—can positively impact climate change via significant CO_2-eq emission reduction potentials. However, while waste incineration seems to be a rather sustainable alternative to the conventional production of electricity and heat today due to the high share of organic matter in German rMSW (~40%), this perception will gradually diminish as the German energy system shifts towards renewables in the course of the German Energy Transition ("Energiewende") [65, 66]. In contrast, the positive climate impacts of chemical recycling remain, as they are mainly achieved through reduced process emissions. From an economic perspective, the TEA results indicate that the implementation of chemical recycling plants requires higher fixed capital investments compared to incineration-based pathways and is associated with significant costs under the current market and regulatory conditions. However, a detailed sensitivity analysis reveals that a multi-pronged approach to increasing chemical recycling profitability including generous plant scaling, adjustments in the German greenhouse gas emission trading system, and price premium guarantees for chemical recycling products could significantly increase the chemical recycling profitability to the point where it outperforms conventional incineration-based treatment.

The central limitations of this research are 1) the restricted focus on the case of rMSW treatment in Germany and 2) the process scope that neglects the systemic framework conditions for chemical recycling deployment, such as environmental regulation. Nevertheless, as illustrated by Publication 2 in the next section, the developed methods and generated findings can be transferred to other cases, such as chemical recycling for unsorted LWP waste, in a straightforward way. Additionally, Publications 3 and 4 illustrate how the data and insights from this study can support assessment models on a country-level to generate knowledge about the systemic framework conditions for chemical recycling.

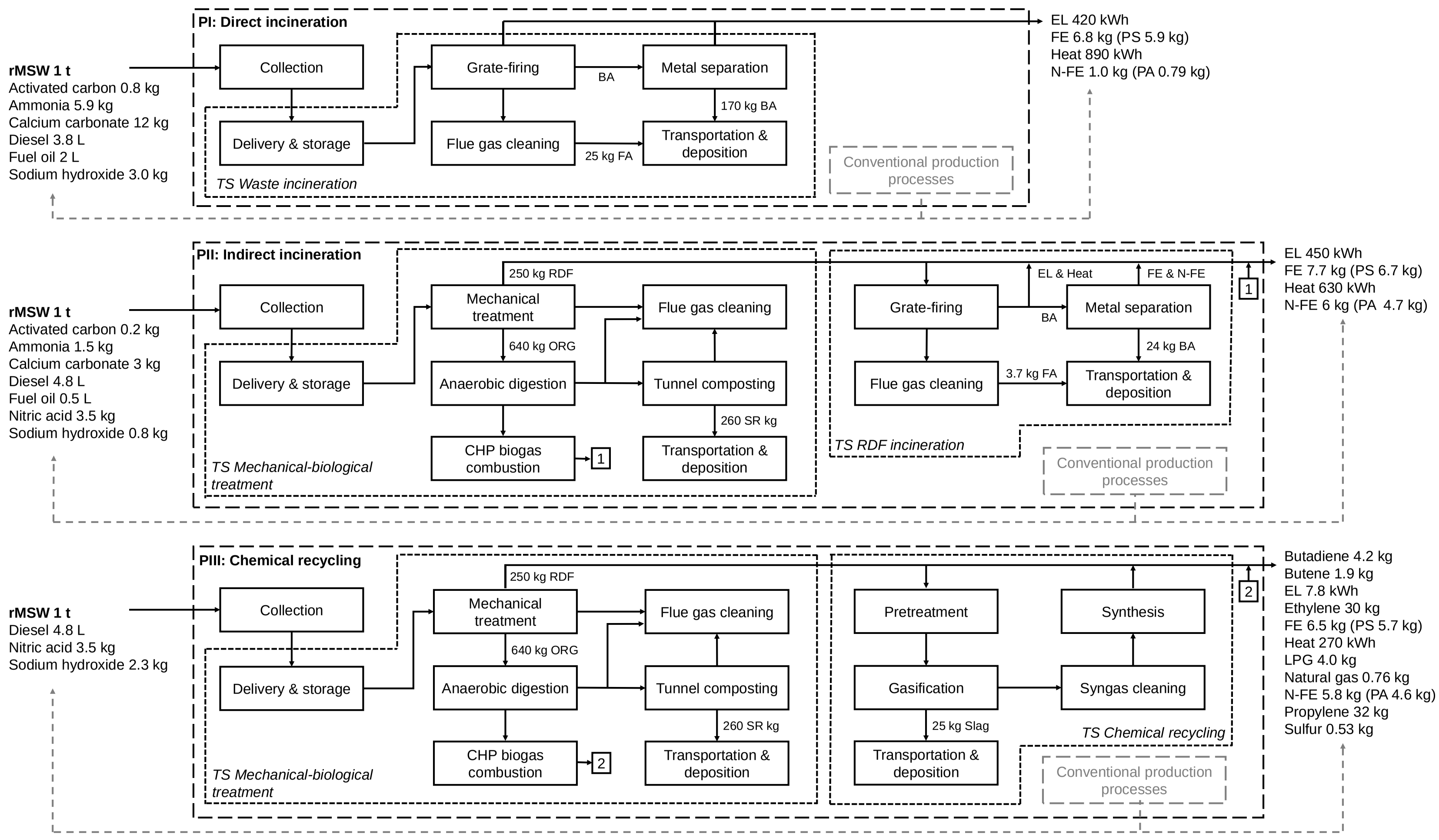

Figure 6: Illustration of the life cycle assessment system environment [34]. BA: Bottom ash. CHP: Combined heat and power generation. EL: Electricity. FA: Fly ash. FE: Ferrous scrap. LPG: Liquefied petroleum gas. N-FE: Non-ferrous scrap. ORG: Organics. PA: Primary aluminum. PS: Primary steel. RDF: Refuse-derived fuel. rMSW: Residual municipal solid waste. SR: Stabilized residues. TS: Treatment step. Values rounded to two significant digits. Energy figures are reported net of energy self-use. Substituted market products at substitution ratios <100% are indicated in brackets.

2.2. Publication 2: Chemical recycling of plastic waste: Comparative evaluation of environmental and economic performances of gasification- and incineration-based treatment for lightweight packaging waste

The second publication[5] was submitted to *Circular Economy and Sustainability* by *Springer* in July 2021 and was accepted in December 2021. The documented study builds on Publication 1 by adapting previously developed methods to the case of unsorted LWP waste treatment in Germany and extending them with measurements of additional environmental impacts (i.e., terrestrial acidification and fossil resource scarcity). LWP waste represents a mixed fraction of packaging waste (i.e., rich in polymers, but also includes paper and cardboard, ferrous and non-ferrous metals, and composites) that is either incinerated or processed in LWP sorting facilities to produce pure material fractions for conventional recycling in the EU [67]. As LWP sorting inevitably produces significant amounts of sorting residues for incineration (i.e., too small for sorting or do not meet the recycling quality standards), both routes are associated with significant greenhouse gas emissions and secondary resource depletion [68]. Chemical recycling could replace incineration for sorting residues to reintroduce contained carbon into chemical production systems [22, 55, 69]. However, insights into the sustainability impacts of the chemical recycling of unsorted LWP waste are lacking as the literature focuses on the technical issues of chemical recycling for pure plastic waste (cf. Section 1.2). To fill this gap, this research aims to:

- extend the currently discussed feedstocks for chemical recycling by unsorted LWP waste,
- derive knowledge about the additional multidimensional impacts of chemical recycling (i.e., terrestrial acidification and fossil resource scarcity) and on how to attain them, and
- generate valuable inventory data for LWP waste treatment applicable to future investigations.

[5] Voss R, Lee RP, and Fröhling M. 2022. Chemical Recycling of Plastic Waste: Comparative Evaluation of Environmental and Economic Performances of Gasification- and Incineration-based Treatment for Lightweight Packaging Waste. Circular Economy and Sustainability. doi: 10.1007/s43615-021-00145-7.

Contributions

Conceptualization: Voss R, Lee RP, Fröhling M; Methodology: Voss R, Fröhling M; Formal analysis and investigation: Voss R; Writing – original draft preparation: Voss R; Writing – review and editing: Voss R, Lee RP, Fröhling M; Funding acquisition: Lee RP; Supervision: Lee RP, Fröhling M.

Analogous to Publication 1, a comprehensive inventory dataset is developed from technical reports issued by the German government, scientific publications, and LCA databases to assess three different treatment pathways for unsorted LWP waste (cf. Figure 7) for the case of Germany, namely

- direct incineration of unsorted LWP waste in an RDF power plant for energy recovery,
- indirect incineration of sorting residues from unsorted LWP waste—produced in a materials recovery facility—in an RDF power plant for the recovery of energy and recyclable materials,
- and chemical recycling of sorting residues from unsorted LWP waste—produced in a materials recovery facility—in a gasification plant to recover recyclable materials and chemical intermediates.

Then, based on generated inventories, an LCA model using EASETECH V3.1.7. [60] and an economic assessment model using Microsoft Excel [63] are developed to assess the environmental (i.e., impacts on climate change, terrestrial acidification, and fossil resource scarcity) and economic sustainability (i.e., FCI, NPV, DPP, and LCCA) of the three pathway-reflecting treatment plants [60, 63].

The study results indicate that the central findings obtained concerning the chemical recycling of rMSW (cf. Section 2.1) can be transferred to the case of unsorted LWP waste. From the environmental perspective, chemical recycling can provide significant sustainability benefits by reducing impacts on climate change, terrestrial acidification, and fossil resource scarcity with low sensitivity to future transformations in the German energy system. Thus, chemical recycling increasingly outperforms conventional incineration-based treatment pathways as the energy system shifts towards renewables. Due to a higher carbon share in sorting residues from unsorted LWP waste, chemical recycling generates more greenhouse gas emission savings compared to rMSW applications. However, as in the case of rMSW, economic indicators including NPV and DPP point to the necessity of significant adjustments in regulatory and market conditions to make chemical recycling economically profitable for potential plant operators.

A central limitation of this research (as for Publication 1) results from the restricted and isolated single-plant perspective that neglects the systemic impacts of an extensive chemical recycling deployment on domestic waste treatment and chemical production systems. However, as illustrated in Publications 3 and 4 using sophisticated computer-based simulation models, the positive environmental sustainability impacts that are observed from a single-plant perspective are maintained if the assessment scope is extended to the country-level.

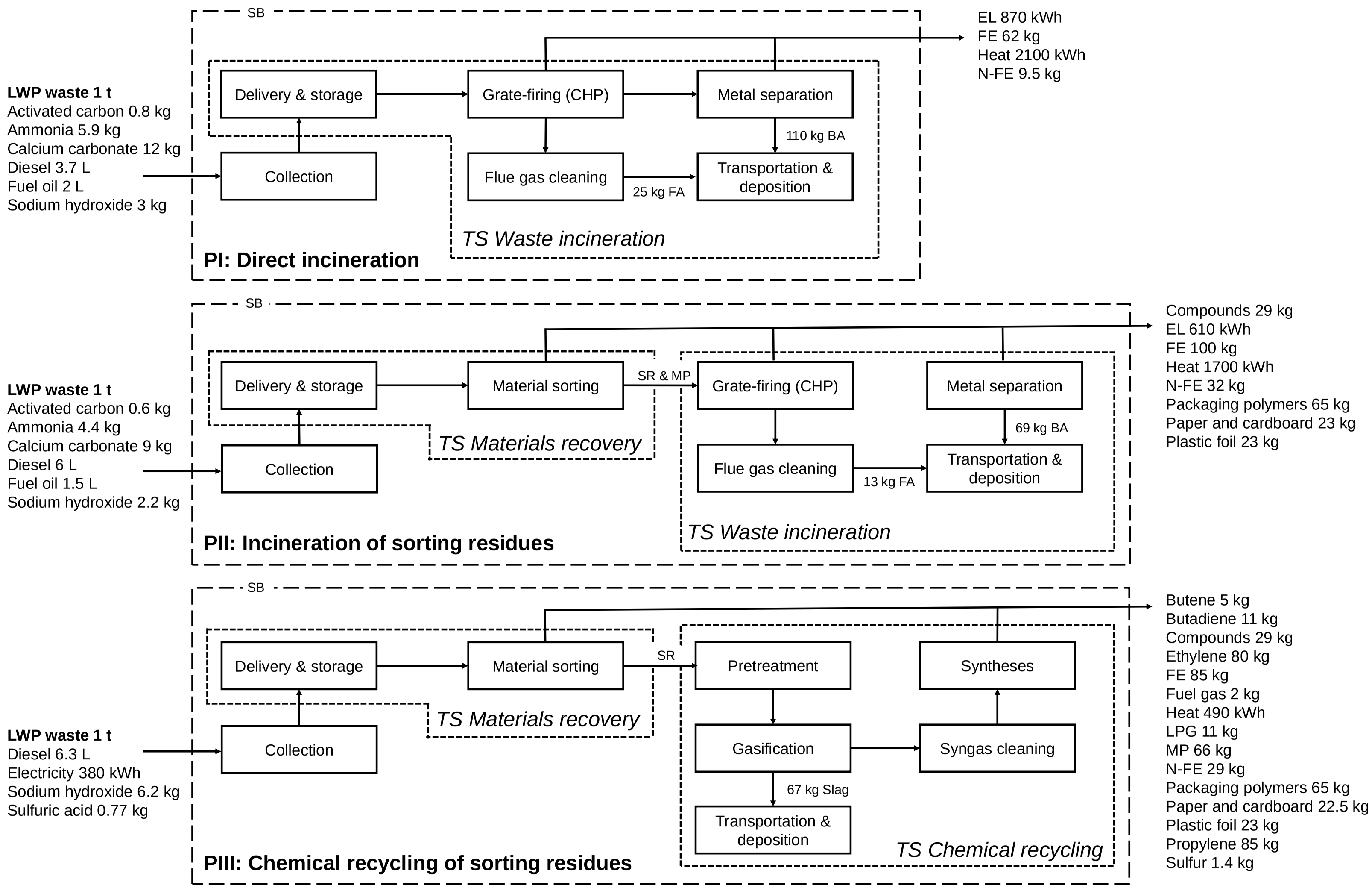

Figure 7: System environment and results of inventory calculations [45]. BA: Bottom ash. CHP: Combined heat and power generation. EL: Electricity. FA: Fly ash. FE: Ferrous scrap. LPG: Liquefied petroleum gas. LWP: Lightweight packaging. MP: Mixed plastics. N-FE: Non-ferrous scrap. SB: System boundaries. SR: Sorting residues. TS: Treatment step. Underlying values rounded to two significant digits. Energy figures are reported net of energy self-use

3. Systemic evaluations

In the second step, high-resolution insights into individual plant configurations for chemical recycling and waste treatment from Publications 1 and 2 are integrated into two individual system models to obtain data on potential country-level deployments of chemical recycling.

3.1. Publication 3: 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

The third scientific paper[6] was submitted to *Resources, Conservation and Recycling* by *Elsevier* in July 2021 and was accepted for publication in December 2021. The reported study assesses the impacts of chemical recycling on the global warming potential of German waste management and base chemical production. Chemical recycling is generally considered a potential building block for the systemic transformation from linear to circular value chains in Germany [28, 70]. However, extant assessments of environmental impacts from a life cycle perspective typically apply an attributional LCA principle that is less suitable for decision support as it disregards the consequences of systemic chemical recycling diffusion for the waste management and chemical production systems (e.g., reduction of energy recovery from waste and reduction of base chemical production from fossils) [71, 72]. To address this gap, this research conceptualizes, implements, and applies a consequential LCA model that reflects the interactions of the chemical recycling of rMSW and LWP waste with waste management and chemical production in Germany by integrating all relevant processes (i.e. chemical recycling, conventional waste treatment for heterogeneous waste, and conventional base chemical production) into a single model system. The aims of this research are to:

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[6] Keller F, Voss R, Lee RP, and Meyer B. 2022. 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. Resources, Conservation and Recycling, Vol. 179: 106106. doi: 10.1016/j.resconrec.2021.106106.

Contributions

Conceptualization: Keller F, Lee RP, Meyer B; Methodology: Keller F; Formal analysis and investigation: Keller F, Voss R; Writing – original draft preparation: Keller F, Voss R; Writing – review and editing: Lee RP, Meyer B; Funding acquisition: Lee RP; Supervision: Meyer B.

- provide additional insights into the chemical recycling of rMSW and unsorted LWP waste as heterogeneous waste streams,
- assess the systemic environmental impacts of chemical recycling on climate change and the design of corresponding assessment approaches, and
- to link chemical recycling to the circular economy in Germany.

The inventory data to model the relevant chemical recycling routes for rMSW and unsorted LWP waste using gasification and pyrolysis in Germany are drawn from Publications 1 and 2, and additional sources including custom process modeling using the flowsheet simulation software Aspen Plus V10 [73] that is widely applied in industry and delivers full energy and mass balances for chemical processes. Additionally, inventory data for conventional incineration-based waste treatment pathways and conventional fossil-based chemical production routes are derived from identical sources to facilitate a consequential systemic LCA. Subsequently, collected data are processed using EASETECH V3.1.7. and GaBi V9.2.0, a comprehensive LCA framework combining sophisticated modeling software with content LCA databases that is well-established in science and industry, to generate insights into the systemic impacts on climate change in 16 individual scenarios for the status quo and chemical recycling via gasification or pyrolysis [60, 74].

The developed LCA model shows that positive impacts on climate change can still be obtained for extensive deployments of chemical recycling that are associated with significant changes in mature and regulated systems of waste treatment and chemical production in Germany. Generally, gasification-based production pathways impact climate change more positively compared to investigated pyrolysis-based pathways. Interestingly, additional high-resolution insights can be obtained with the developed approach including the finding that the most significant total GWP reduction can be achieved if chemical recycling is integrated into the conventional value chain for olefins production in Germany. Ultimately, the results suggest that the energy recovery efficiencies for waste incineration and assumptions for conventional energy production significantly impact the greenhouse gas emissions of the modeled system. Consequently, it is very important to integrate detailed and robust insights into technological advancements in energy recovery efficiencies and conventional energy production in future LCA studies.

The central limitations of this research are 1) that sustainability assessments are limited to impacts on climate change, and 2) that the chosen scenario-based approach allows conclusions to be drawn about individual system states, but does not investigate the conditions under which these states could emerge (i.e., regulatory, societal, or market developments). However, as illustrated in Publication 4, by coupling LCSA with agent-based modeling, positive sustainability impacts extend to additional environmental and social categories, and transition pathways to the developed scenarios can be facilitated with targeted waste management regulations.

3.2. Publication 4: A consequential approach to life cycle sustainability assessment with an agent-based model to determine the potential contribution of chemical recycling to UN Sustainable Development Goals

The fourth publication[7] was submitted to *Industrial Ecology* by *Wiley* in October 2021 and was accepted in May 2022. The addressed study anticipates the multi-dimensional sustainability impacts (i.e., environmental, economic, and social) of gasification-based chemical recycling for rMSW in Germany until 2050 with a focus on the impact of different policy schemes as drivers for technology diffusion. As elaborated in previous publications, the gasification-based chemical recycling of rMSW represents a potential candidate for facilitating the transformation from linear to circular value chains in Germany and increasing the supply security of national chemical production systems by introducing domestic carbon resources into supply chains. However, systemic modeling approaches that can provide decision-makers with knowledge about the effectiveness of market developments or political regulations to support chemical recycling deployments are lacking (cf. Section 1.2). To address this gap, this research conceptualizes, implements, and applies an alternative consequential approach that combines LCSA with agent-based modeling to connect environmental regulations to market developments and ultimately investment decisions by industrial representatives. The aims of this research are to:

- provide additional insights into the chemical recycling of rMSW as a heterogeneous waste stream,

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[7] Voss R, Lee RP, and Fröhling M. 2022. A consequential approach to life cycle sustainability assessment with an agent-based model to determine the potential contribution of chemical recycling to UN Sustainable Development Goals. Industrial Ecology. doi: 10.1111/jiec.13303

Contributions

Conceptualization: Voss R; Methodology: Voss R; Formal analysis and investigation: Voss R; Writing – original draft preparation: Voss R; Writing – review and editing: Voss R, Lee RP, Fröhling M; Funding acquisition: Lee RP; Supervision: Lee RP, Fröhling M.

- deliver systemic insights into the multidimensional sustainability impacts (i.e., environmental, economic, and social) of an extensive chemical recycling deployment and show how they can be attained,
- learn about the regulatory- and market drivers of the technology diffusion process in detail, and
- link chemical recycling to the circular economy by identifying reasonable roles for chemical recycling and efficient regulatory instruments to promote technology deployment.

The inventory data to model the relevant treatment pathways of rMSW using gasification in Germany (cf. system environment in Figure 8) are drawn from Publication 1 and additional sources including technical reports by the German government, scientific publications, and LCA databases. The developed dataset is subsequently processed with a custom systemic computer model implemented in MATLAB V2019a [75], which is a widely-applied programming environment for complex numerical calculations and simulations, and couples LCSA—including process-based LCA, TEA, and social indicators—with agent-based modeling. As rMSW producers, waste treatment sites, and chemical sites are represented individually, the model captures temporal dynamics in the system resulting from agent interactions under changing regulatory/market framework conditions. Finally, to facilitate the interpretation of the sustainability impacts derived from the different scenarios of system development, the impact contributions to the UN Sustainable Development Goals are assessed [76, 77].

The results provide information that is complementary to that derived from previous studies on the multidimensional impacts of an extensive chemical recycling deployment for heterogeneous post-consumer waste fractions. In addition to further evidence of the positive environmental impact on climate change, the study results show positive impacts on terrestrial acidification and fossil resource scarcity. Additionally, the study highlights positive impacts on local employment as a relevant social indicator due to the labor intensity of chemical recycling including two treatment steps (i.e., mechanical-biological pretreatment plus chemical recycling). Accordingly, chemical recycling can represent an important building block to achieving sustainability goals in the framework of the UN Sustainable Development Goals or the European CEAP. However, the results also indicate that without regulatory action, waste producers, the chemical industry, and the waste management industry may follow their modi operandi of linear production/disposal, cementing incineration as the dominant treatment pathway for rMSW until 2050. To break out of the path-dependent lock-in, effective adjustments in the regulatory framework represent a sine qua non for the diffusion of chemical recycling technologies. These may include the implementation of a targeted recycling rate and—if the prices of European emission trading certificates rise sharply in the near future—the inclusion of rMSW incineration into emission trading. However, chemical recycling will come with increased economic costs for society due to the increased treatment gate fees that are passed on to the German population at €8 per capita per year until 2050 in the most beneficial case. This raises the questions of how high society's acceptance of such additional costs is and whether the costs can potentially be offset by efficiency gains in other steps of waste management, such as waste collection.

The central limitations of this study are represented by 1) simplifying assumptions regarding system external restrictions to chemical recycling deployment, such as political and social resistance, 2) the focus on rMSW treatment in Germany that could be associated with a limited transferability of the results to other cases, and 3) the restricted number of sustainability impact categories considered. However, future studies may address these shortcomings by building on the developed dataset and model approach.

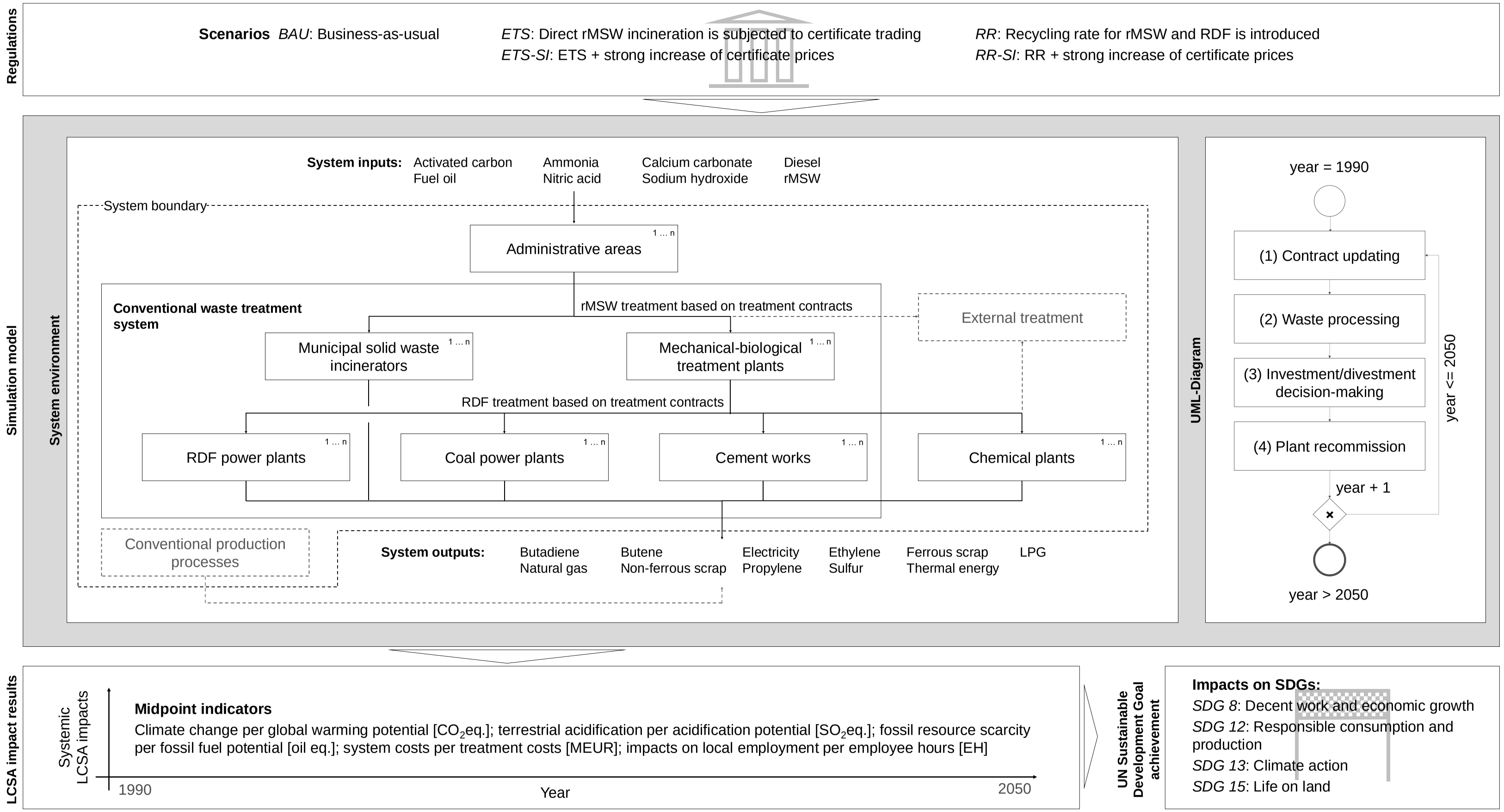

Figure 8: Consequential life cycle sustainability assessment (LCSA) system [44]. BAU: Business-as-usual. EH: Employee hour. ETS: Emission Trading System. LPG: Liquified petroleum gas. MEUR: Million euros. RDF: Refuse derived fuel. rMSW: Residual municipal solid waste. RR: Recycling rate. SDG: Sustainable Development Goal. SI: Strong increase

4. Discussion

4.1. Main insights and contributions

This dissertation deliberately extends current research by highlighting robust chemical recycling options for heterogeneous feedstocks (cf. Figure 9). It presents detailed sustainability analyses of chemical recycling options for rMSW in Germany as a heterogeneous waste fraction that is produced in significant amounts but has received little attention in the literature to date. Additionally, it presents corresponding insights for unsorted LWP waste that, despite a growing media interest in packaging waste on land and at sea, is rarely addressed in chemical recycling literature today (cf. Section 1.2.1). This shift/extension of the research is valuable for science and society as efficient mechanical recycling solutions to increase the circularity of pure plastic streams are commercially available and discussed extensively in the current literature [78–80]. In contrast, incineration or landfilling seem to represent the only treatment options for heterogeneous waste streams that are discussed in the extant literature or public debates. To illuminate chemical recycling alternatives to these linear treatment pathways as presented in this work facilitates the richness of future discussions about the benefits of chemical recycling and its manifold use cases.

This dissertation expands the existing literature focus on primarily technical assessments of chemical recycling (e.g., product yields and qualities) with data on multidimensional impacts (i.e., environmental, economic, and social sustainability), and provides sophisticated tools to assess them (cf. Figure 9). Specifically, the results indicate that the conceptualized, implemented, and applied LCA/TEA/LCSA models generate well-founded quantitative data on sustainability impacts concerning climate change, terrestrial acidification, fossil resource scarcity, economic viability, and labor intensity. In addition, the underlying systemic mechanisms and interrelationships that are most influential to the magnitude of these impacts are revealed and discussed. For instance, the results show that the environmental benefits of chemical recycling greatly depend on the reference energy system. Specifically, energy recovery via incineration-based treatment for heterogeneous waste receives significant credit for substituting conventional energy today. In contrast, the positive impacts of chemical recycling tend to be more decoupled from energy system characteristics. Thus, whether, where, and how chemical recycling will be applied sustainably also depends on how quickly and in which direction national and transnational energy systems transform in the coming years and decades. Additionally, the TEA assessments included in this work suggest that the economic viability of chemical recycling pathways for heterogeneous waste fractions depends on, inter alia, options for upscaling via centralization. As the presented pathways include a pretreatment step, their profitability could potentially be improved by systems that combine multiple pretreatment facilities with a single, upscaled recycling facility. It is important to note that comparable concepts have been discussed for second-generation biofuel or ethanol production [81–83]. Ultimately, the results indicate that there could be positive social impacts in terms of labor market effects due to the increased labor intensity of the two-stage recycling pathways that were assessed. The question is whether these benefits can be transferred to other chemical recycling concepts, for example, to source-separated industrial waste fractions that do not require a separate pretreatment step. This, and numerous other issues that need to be considered if chemical recycling is intensified in the future, are highlighted by the multidimensional and life-cycle-based sustainability analyses of this dissertation. The application and development of corresponding analyses represent a significant extension of the body of literature that has focused on unidimensional assessments of primarily technical aspects concerning this point (cf. Section 1.2.2). Indeed, this extension seems valuable as the importance of sustainability aspects in technological transformation processes increases with the rise of sustainability-related global challenges, such as climate change or the waste crisis [2, 84]. Specifically, multidimensional studies that address environmental, economic, and social aspects can help to reveal hidden synergies and trade-offs between different sustainability dimensions to achieve national and global sustainability goals faster than it would be possible with isolated assessments [85]. As all investigations are conducted in accordance with proven methodological standards, they can serve as a blueprint for future studies in chemical recycling assessment to further increase the assessment quality in the literature and the comparability among the results from different studies.

This research generates valuable knowledge about the drivers and sustainability impacts of a country-level diffusion of chemical recycling from a systemic perspective (cf. Figure 9). Specifically, the included studies facilitate a better understanding of the inner system mechanics of existing waste treatment and chemical production systems. They indicate that chemical recycling for heterogeneous waste fractions is not only suitable for highly specialized application contexts but holds the potential to be realized systemically as positive sustainability impacts persist when the technology is scaled-up. Additionally, due to a detailed agent-based approach that includes complex mechanisms of system development such as power dynamics and interactions between relevant system actors under changing regulatory and market conditions, this dissertation provides sensible quantitative data on individual drivers for chemical recycling diffusion (cf. Section 4.2). The results indicate that chemical recycling deployment is not a self-fire success due to increased costs, and careful regulatory design and clear communication to the public are needed to achieve acceptance and system change. By developing and applying complex models to sustainability assessment at the country-level, this dissertation adds efficient tools to the assessment toolbox in the extant chemical recycling literature. The approaches can also enhance related streams in the literature that address comparable recycling systems including the mechanical recycling of plastic waste or battery recycling [86, 87]. The advancement of plant-focused assessments that examine chemical recycling under isolated laboratory conditions to systemic assessments that investigate interactions with mature and regulated industrial systems represents a significant contribution to this research with findings that encourage the intensification of research and development in the field.

The results of this dissertation support the efforts to answer questions of whether and how to link chemical recycling to the circular economy (cf. Figure 9). Specifically, by developing multiple future scenarios for a country-level implementation of chemical recycling, and by comparing them to scenarios of conventional waste treatment, the included studies paint detailed pictures of the potential roles that chemical recycling could play in the circular economy. This is exceptionally useful, as the academic literature still struggles to find consistent chemical recycling definitions, which can be seen as a key prerequisite for the efficient and collective unification of chemical recycling with circular economy concepts. In particular, there is uncertainty about which concepts should be considered to be chemical recycling and under what circumstances, as several questions regarding systemic sustainability have not yet been resolved (cf., waste-to-fuels, waste-to-energy) [88–92]. The results from the complex modeling approaches of this dissertation support the answers concerning which chemical process routes chemical recycling should be implemented in, based on which technology, and with which feedstocks. Specifically, the results indicate that especially gasification-based chemical recycling for heterogeneous waste fractions to produce olefins provides environmental benefits including greenhouse gas reduction by replacing energy recovery with incineration. In providing these valuable and highly detailed insights into chemical recycling characteristics and potentials, this dissertation supports the closure of material cycles in the circular economy to subsequently achieve sustainability goals such as the United Nations Sustainable Development Goals including Goal 12 “Responsible consumption and production” and Goal 13 “Climate change” [2].

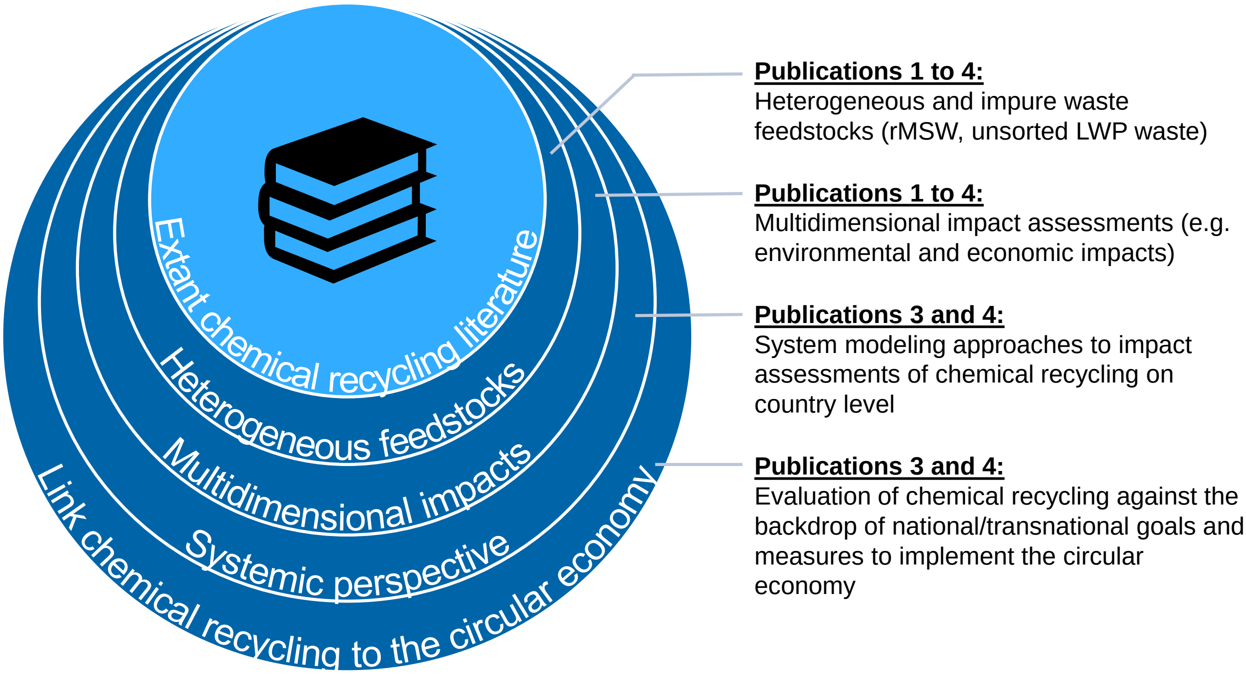

Figure 9: Research contributions to the existing body of literature on chemical recycling technologies. CR: Chemical recycling. LWP: Lightweight packaging. rMSW: Residual municipal solid waste

4.2. Implications for management and policy

The technological transitions required for sustainability bring about major societal changes that challenge established roles, rules, and business models in industry [93]. The results from this research provide decision-makers or technology developers in the chemical and waste management industries with attributive information on individual chemical recycling technology alternatives and the use of heterogenous waste feedstocks as alternative carbon sources to conventional fossil sources for chemical production. This information is highly valuable for industrial representatives as they face the question of whether applied linear waste treatment and chemical production routes are in line with the achievement of individual emission reduction targets and will enjoy public acceptance in the future when environmental awareness further increases in society. Even today, the chemical industry is confronted with increasing pressure from its customers to enhance the sustainability and circularity in its supply chains (e.g., demands by IKEA or Adidas) [94, 95]. The results from this research support the evaluation of technical alternatives that promote circular resource use by providing evidence of the positive sustainability impacts of chemical recycling. However, these positive environmental impacts are hampered by significant investment and operating costs as chemical recycling processes are more complex and labor-intensive than other approaches. This research indicates that the profitability of chemical recycling under current regulatory conditions depends, inter alia, on upscaling/centralization options and consumers' willingness to pay more for chemically recycled products. Thus, corresponding planning and information should play a pivotal role in future investment decisions.

From the policy perspective, governments and intergovernmental organizations are now discussing the recognition of chemical recycling as a possible technology option to support the achievement of future recycling targets and to increase the sustainability/circularity of chemical production [96, 97]. The results from this research provide regulators with consequential data to develop a solid understanding of the macro changes of chemical recycling diffusion and their consequences for ecosystems, economic systems, and society. They thereby facilitate a targeted harmonization of chemical recycling utilization with current waste management practices and regulatory frameworks such as the European Waste Hierarchy. Specifically, this dissertation provides evidence to perspectively integrate chemical recycling for heterogeneous streams into the hierarchy above energy recovery (cf. Figure 2). Furthermore, it can support decisions on the implementation of effective regulatory measures for technology promotion such as penalizing CO_2-eq emissions from municipal solid waste incineration, which is currently discussed at the EU level, or the implementation of targeted chemical recycling quotas [98, 99]. Specifically, the results from this research indicate a superiority of targeted recycling quotas as a practical recommendation for policy-makers concerned with the organization of waste management. Finally, the further use of the developed assessment tools to test additional policy measures may increase the quality of socio-political discussions and political actions to implement the circular economy.

5. Conclusions

Global challenges such as the growing waste crisis resulting from population growth, rising consumption, and the dominant take-make-dispose logic in existing linear economic systems increasingly threaten the viability of our global community. As chemical recycling could present a potential circular economy building block, it is currently discussed by academics, industry representatives, and in the socio-political realm. The insights obtained from this dissertation point to the potential of chemical recycling techniques and concepts for heterogeneous waste fractions to contribute to a closed-loop circular economy. Specifically, this dissertation provides novel data on, inter alia, CO_2-eq reduction and fossil resource saving potentials, economic profitability, and the labor impacts of chemical recycling. The developed and applied methods illustrate how multidimensional plant-focused and systemic evaluations can generate quantitative data to identify the advantages and disadvantages of possible technological applications. Despite these valuable contributions, the applied approaches have some limitations concerning transferability, result integration, data uncertainty, and assessment boundaries, that provide guidance for future research as outlined below.

– <u>Transferability of results to other case studies:</u> The case of Germany is at the center of all research activities. A central limitation is represented by the potential transference barriers of the findings to other geographical regions or research contexts. Specifically, Germany has complex waste management, chemical production, and energy systems in place that might differ significantly from corresponding systems in other countries or regions. Additional discrepancies in waste management regulation and legislation, especially outside Europe, might lead to reduced generalizability of assessment results and disqualify identified chemical recycling applications or regulatory adjustments in other cases. Thus, future research needs to investigate additional chemical recycling use cases for adjusted framework conditions to fully understand the potential roles in the transition toward the circular economy. Interesting objects of study include the Benelux region, which features modern waste management systems and well-developed chemical industries with comparable dependence on raw materials, where initial efforts to introduce chemical recycling on a large scale are apparent today [37, 100].

– <u>Integration of indicator results:</u> The studies from this research produce assessment results that are based on indicators from the environmental, economic, and social sustainability dimensions. A central limitation is represented by the largely disintegrated interpretation of the indicator results that were obtained. To acquire valuable integrated insights and explanations that cannot be obtained with an unintegrated view for comprehensive decision-making processes, innovative combinations and aggregations of methods and results are required [101]. In this research, a preliminary approach to integrated assessment can be seen in the determination of levelized costs of carbon abatement (cf. Sections 2.1 and 2.2) as suggested by Friedman et al. [62] which links the reduction of CO_2-eq emissions to economic costs. However, future research must further extend this to advance multidimensional assessments to interdimensional assessments in the field [101]. As presented in several studies in recent years [102–104], a coupling of LCA and LCSA approaches with multi-criteria decision analysis (MCDA) can improve decision-making by solving problems of decision complexity and conflicting impact categories [103, 105]. Specifically, MCDA could introduce impact category weighting by experts and subsequent aggregation to condense diverse and incommensurable impact category results to a single metric. As a corresponding metric would integrate expert knowledge about the significance of individual sustainability impacts into a science-based decision process, it could help to further bridge the science-policy gap between scientific data and untrained understanding of policy-makers.

– Data uncertainty management: Chemical recycling research and industrial deployment are in the early stages of development and application, resulting in a limited body of available data with reduced quality (cf. Section 1.2). A central limitation of this research is that quantitative results from plant-focused and systemic evaluations might be impaired by parameter uncertainty due to gaps in the extant literature and a lack of industrial validation data from industry-scale demonstrations. In particular, the data for chemical processes in this research mainly stem from computer-based process modeling that represents the inevitable first step of technology research and development but is highly theoretical compared to the actual experience with operating plants. Model functionality, scenario design uncertainty, and indicator relevance uncertainty lead to additional uncertainties in the research conclusions and associated decisions [106]. Future assessment approaches might benefit from extended data availability or improved quantitative uncertainty management (i.e., quantification of the overall assessment uncertainty) for chemical recycling technology characteristics and performances. The approaches for uncertainty management include detailed analyses of individual parameter variabilities and sophisticated probabilistic simulations based on, e.g., Monte Carlo Analysis [106–108]. The integration of corresponding or related approaches into complex and consequential LCA/LCSA models represents a particular challenge as potential solutions are associated with increased model complexity and might demand significant computing power. However, the corresponding efforts in future research can deliver additional robust conclusions on optimal technology development/deployment for chemical recycling in the circular economy [20].

– <u>Restricted assessment boundaries:</u> The publications of this research provide novel insights into the sustainability impacts of individual chemical recycling applications concerning specific technologies (i.e., gasification and/or pyrolysis) and feedstocks (i.e., residual MSW and unsorted LWP waste) as heterogeneous waste fractions. However, corresponding approaches still only map a small fraction of possible technology applications and systemic interactions between different waste treatment or chemical production routes. To extend future research with additional chemical recycling technologies (e.g., solvent-based purification, depolymerization), additional technological competitors (e.g., mechanical recycling technologies, incineration combined with carbon capture and storage, and substitution of conventional plastics with bio-based materials), and additional waste fractions (e.g., bulky waste, automotive waste, and source-separated industrial waste) facilitates an extended systemic assessment of chemical recycling to fully exploit the potential of corresponding chemical recycling concepts or to identify technological dead ends.

Notwithstanding these potential avenues for future studies, this dissertation provides significant contributions to multidimensional assessments of chemical recycling from plant-focused and systemic perspectives. It contributes to research, development, and deployment in the field by providing up-to-date knowledge and a deeper understanding of the sustainability impacts associated with chemical recycling as an important and novel emerging technology. Furthermore, it provides general methodological guidance for applications of LCA, TEA, LCSA, and sophisticated computer-based system modeling approaches for corresponding investigations in the future. The results support the hypothesis that chemical recycling is useful to address the global waste crisis, close material cycles for heterogeneous waste fractions, and support circular economy concepts, such as zero waste cities. Additionally, the insights obtained objectify the decision-making of political and industrial representatives concerned with waste management regulations to support the establishment of a take-make-recycling logic in dealing with valuable natural resources. Consequently, this research encourages scholars, industrial representatives, and policy-makers to increase their engagements in the field to reduce industrial emissions to net zero in the future and to permanently preserve fossil carbon resources in their natural deposits via innovative chemical recycling concepts.

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Annex

Literature review

The systematic literature review is conducted using the scientific electronic database Scopus, the largest abstract and citation database of peer-reviewed literature [109]. The search strategy involves the phrasing of a search string[8] that combines three keywords reflecting an assessment approach (e.g., “assessment,” “analysis,” and “evaluation”) with the term “chemical recycling” or one of four terms that directly address specific chemical technologies as introduced in Section 1.1 (i.e., “waste dissolution,” “waste depolymerization,” “waste pyrolysis,” or “waste gasification”). As a wild card, ‘*’ is applied to cover slight alterations in search term writing. To identify relevant studies among the search results, three main inclusion criteria are defined:

- only original and completed research is included, while literature reviews, comments, editorials, and incomplete studies are excluded. In this context, it is important to note that screening literature reviews in the field for additional studies is excluded, as it would interfere with drawing valid conclusions from the quantitative literature analysis in Section 1.2 due to a high level of specialization in extant reviews (e.g., “A review on the recycling of waste carbon fibre/glass fibre-reinforced composites: fibre recovery, properties and life-cycle analysis” by Karuppannan Gopalraj and Kärki [110]);
- the publication language is English;
- the study addresses technology assessments for chemical recycling. As defined in Section 1.1, chemical recycling refers to the dissolution, depolymerization, pyrolysis, and gasification processes to support the recirculation of waste into suitable chemical production systems. Applications of chemical recycling technologies to biomass are excluded (i.e., not considered as recycling). Additionally, processes targeting energy products including electricity, heat, or fuels (i.e., waste-to-fuels or waste-to-energy) are not considered chemical recycling.

It is also important to note that no cut-off criteria are applied to research methods, quality, and time span.

[8] (analysis OR assessment* OR evaluation*) AND (“chemical recycling” OR (waste Pre/0 (dissolution OR depolymerization OR pyrolysis OR gasification)))

General findings: The search strategy yields as many as 762 search results whereby 571 articles remain in the set after excluding duplicates, incomplete studies, reviews, comments, editorials, book chapters, etc. Then, after screening titles and abstracts, 138 articles are retained for a full-text review. A final set of 110 articles meets all inclusion criteria after the full-text review and is subjected to an in-depth analysis. Figure 10 resents a flow chart of the study selection process.

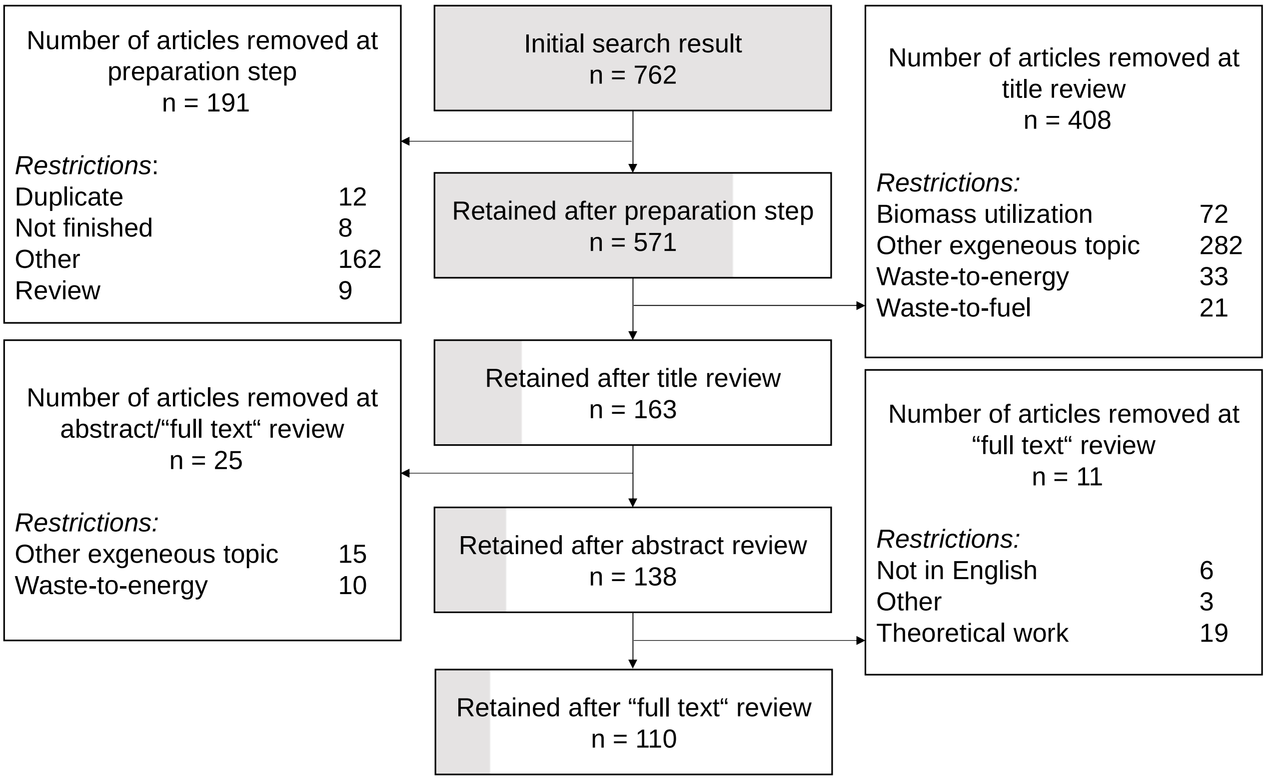

Figure 10: Detailed breakdown of the individual literature review steps