THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Decarbonization in Carbon-Intensive Industries

An Assessment Framework for Enhanced Early-Stage Identification of Optimal Decarbonization Pathways

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CHALMERS UNIVERSITY OF TECHNOLOGY

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Abstract

Carbon-intensive industries currently account for a quarter of global annual CO_2 emissions. Achieving mandated climate targets necessitates the rapid implementation of decarbonization technologies in these industries. Such deployments typically involve substantial upfront investments amidst technical, economic, and policy uncertainties. Consequently, careful selection of decarbonization technologies or a combination thereof, coupled with measures like process electrification and energy efficiency, becomes increasingly crucial. In this context, numerous early-stage comparative assessment studies utilizing process integration and techno-economic methods to identify cost-optimal decarbonization technologies in unabated industries often overlook key considerations at the systems, plant, and site levels.

This thesis presents limitations in existing methodological approaches for comparing decarbonization pathways, spanning systems, plant, and site-level considerations. A generalized hybrid assessment framework was developed that addresses these limitations with individual framework methodologies developed in the appended papers. At the systems level, extended boundaries and exergy as a metric were used to compare two CO_2 capture technologies with inherently different exergy requirements per unit of CO_2 captured, considering plant owner and end-user perspectives. At the plant level, an iterative exergy-pinch analysis combined with techno-economic analysis was developed to identify promising process modifications in unabated process plants that maximize overall exergy utilization and CO_2 avoidance with successive designs towards net-zero emissions. Finally, a site-specific techno-economic analysis was developed incorporating site-specific factors expected to impact the final cost of CO_2 avoidance. These frameworks were demonstrated with industrial case studies on bio-CHP in a district heating system, propane dehydrogenation, and steam cracker plant, respectively.

The case study results show that preserving electric power in bio-CHP plants through the integration of amine-based CO_2 capture technology, complemented with industrial heat pumps, would not only ensure a greater potential for district heat delivery but also provide greater product flexibility in terms of both heat and power production, and negative CO_2 emissions. The iterative exergy-pinch analysis applied to the propane dehydrogenation plant revealed unconventional process modifications, resulting in a substantial reduction in CO_2 avoidance cost (58–70%) compared to CO_2 capture from the highly diluted flue gas stream from the unmodified process (167–181 €/tCO_2). Finally, utilizing site-specific techno-economic analysis, the cost escalation due to site-specific factors, in terms of CO_2 avoidance, was approximately 80% higher for the post-combustion CO_2 capture process (43 €/tCO_2) compared to the alternative of hydrogen-firing in the cracker furnaces, through the pre-combustion CO_2 capture process (24 €/tCO_2). These findings reveal that cost factors that are commonly neglected could significantly influence the choice of decarbonization technology at an early stage. In summary, the proposed assessment framework, combining these individual framework methodologies, can be utilized to obtain a comprehensive early-stage indication of the optimal decarbonization pathway for specific industrial sites.

Keywords: Industrial decarbonization, Carbon-intensive industries, CO_2 Capture, Process modeling, Process integration, Exergy and Pinch analyses, Cost estimation

List of publications

The thesis is based on the following appended papers, which are referred to in the text by their assigned Roman numerals:

I. Kumar, T. R.; Beiron, J.; Biermann, M.; Harvey, S.; & Thunman, H. "Plant and system-level performance of combined heat and power plants equipped with different carbon capture technologies," Applied Energy, **2023,** 338, 120927.

II. Kumar, T. R.; Beiron, J.; Marthala, V.R.R; Pettersson, L; Harvey, S.; & Thunman, H. "Combining exergy-pinch and techno-economic analyses for identifying feasible decarbonization opportunities in carbon-intensive process industry: Case study of a propylene production technology," Submitted for publication. **2024.**

III. Kumar, T. R.; Beiron, J.; Marthala, V.R.R; Pettersson, L; Harvey, S.; & Thunman, H. "Enhancing early-stage techno-economic comparative assessment with site-specific factors for decarbonization pathways in carbon-intensive process industry," Submitted for publication. **2024.**

Author contributions

Tharun Roshan Kumar is the principal author of all papers included in this thesis. Dr. Maximilian Biermann contributed to method development, modeling, discussions, and editing in **Paper I**. Dr. Johanna Beiron contributed to modeling in **Paper I**, method development in **Paper I** and **Paper III**, and discussion and editing of **Papers I–III**. Dr. V.R. Reddy Marthala and Lars Pettersson have contributed with discussions to **Papers II** and **III**. Professor Henrik Thunman is the main academic supervisor and contributed to the discussion and editing of **Papers I–III**. Professor Simon Harvey contributed to the discussions and editing of **Papers I–III**.

Other publications by the author related to the topic are listed below. However, these have not been included in the thesis as they fall outside the scope of the thesis.

- Kumar, T. R.; Mattisson, T.; & Rydén, M.; & Stenberg, V.; "Process Analysis of Chemical Looping Gasification of Biomass for Fischer–Tropsch Crude Production with Net-Negative CO_2 Emissions: Part 1," Energy & Fuels **2022**, 36 (17), 9687-9705.

- Kumar, T. R.; Mattisson, T.; & Rydén, M. "Techno-Economic Assessment of Chemical Looping Gasification of Biomass for Fischer–Tropsch Crude Production with Net-Negative CO_2 Emissions: Part 2," Energy & Fuels **2022**, 36 (17), 9706-9718.

- Saeed, M. N., Shahrivar, M., Surywanshi, G. D., Kumar, T. R., Mattisson, T., & Soleimanisalim, A. H. "Production of aviation fuel with negative emissions via chemical looping gasification of biogenic residues: Full chain process modelling and techno-economic analysis," Fuel Processing Technology, **2022**, 241, 107585.

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Tharun Roshan Kumar,
Göteborg, 7th March 2024

"Nothing will work unless you do."
Maya Angelou

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1 Introduction

Since the landmark Paris Agreement in 2015, global energy-related CO_2 emissions have steadily increased to 36.8 $GtCO_2/yr$ in 2022, of which 9.2 $GtCO_2/yr$ from carbon-intensive industries, constituting roughly 25% of the world's total carbon dioxide emissions [1]. To put these emission figures into perspective, given the current global CO_2 emission rate, it is likely that global temperatures will exceed the 1.5°C temperature rise above pre-industrial levels before the year 2030 and reach 2°C before mid-century [1,2]. In the European context, ambitious targets have been set in recent years for greenhouse gas emissions reduction. These targets include a reduction of 55% by 2030 and 90% by 2040 compared to the 1990 levels, aligning with the aim of achieving climate neutrality by 2050 [3,4]. A series of policy initiatives were introduced under the European Green Deal [5], which notably emphasized the role of industries in leading the transition towards climate neutrality and the aim of transitioning to circular systems in production and consumption.

Sectors such as iron and steel, refineries, cement, petrochemicals, pulp, and paper, which collectively contribute to approximately 94% of total industrial emissions in the EU [6], face significant challenges in meeting these stringent CO_2 mitigation targets within a rapidly evolving policy landscape. These challenges involve technical, economical, market, and institutional (policy) uncertainties [7]. Notably, the polluter's pay approach under the EU Emissions Trading Scheme (EU-ETS) and the upcoming Carbon Border Adjustment Mechanism[1] (CBAM) have put increased pressure on these industries to undergo a complete transformation of their operations, ensuring circular and sustainable emissions-free production. Currently, the aforementioned carbon-intensive industries are deemed at risk of carbon leakage and, therefore, benefit from free allowance under the EU-ETS [8,9]. However, this free allocation[2] will gradually be phased out when CBAM is phased in during the period 2026–2034 [10].

Considering these timelines, most carbon-intensive industries in the EU face a narrow implementation window of less than a decade, until 2034, to assess, choose, and deploy enabling process technologies for the transformation of existing industrial sites. Throughout this period, they must also concurrently adopt energy efficiency and decarbonization measures to achieve net-zero CO_2 emissions. Therefore, industrial processes are expected to transition into more circular and electrified systems, driven by the increasing need to decouple from fossil-resource extraction for the production of carbon-based materials. This shift is also supported by the diminishing carbon footprint of electricity with the growing share of variable renewable energy sources, such as wind power.

Near-term investments in the transformation of existing industrial assets through retrofits and the installation of low-carbon technologies will ultimately determine the achievable emissions reductions within these industries. Given that most of these installations will be first-of-their-kind, they are expected to incur significantly higher costs than installations implemented elsewhere in subsequent years. Therefore, the upcoming investment cycles become increasingly crucial for the careful selection of decarbonization technologies under these uncertainties and, more importantly, to ensure these industries comply with the mandated policies as well as retain their competitive advantage.

Pathways for industrial decarbonization include energy efficiency, end-of-pipe CO_2 capture, process electrification, oxyfuel combustion, and the transition to low-carbon fuels such as hydrogen or biogenic-based fuels. In this context, the selection of decarbonization technologies and the commitment to a

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[1] To prevent potential carbon leakage, i.e., relocation of industries to regions with less stringent climate regulations, by penalizing imports of materials and goods from carbon-intensive regions with less ambitious climate goals into the EU. Thereby, supporting the EU's climate mitigation actions and the competitiveness of European industry [62].

[2] The existing policy instrument under the EU-ETS which grants the right to emit greenhouse gas ($CO_{2,eq}$) to regulated industries to safeguards their competitiveness and address carbon leakage risks [6]

decarbonization pathway within specific industries often pose a paradox of choice or, conversely, a lack thereof, ultimately inducing decision paralysis.

Many emerging low-carbon technologies[3] currently remain unproven at industrially relevant scales and, therefore, would require several years before commercialization. Depending on their technological maturity, these technologies may exceed the targeted deployment timeframe, even in the most optimistic scenarios, thereby limiting their immediate role in the effort to curtail industrial CO_2 emissions. In contrast, the choice between mature technologies that can be retrofitted (e.g., absorption-based CO_2 capture from flue gases) and substitute process technologies nearing commercialization remains unclear, considering the limited time for near-term implementation and the uncertain potential for future cost reductions through technological improvements. This underscores the uncertainty related to both time and technology choices regarding their availability in relation to the 2050 net-zero CO_2 emissions target.

The incumbent assessment methods for process integration and techno-economic performance evaluation of various process technologies are bound by uncertainties stemming from the selection of system boundaries, limitations in identifying new process configurations or systems designs, and uncertainties regarding site-level conditions and future energy market conditions. Firstly, the choice of system boundaries during evaluation can significantly impact the selection of decarbonization technology, and the optimal solution may vary depending on whether the assessment is approached from the perspective of a plant owner or a systemic perspective. This highlights the need for careful consideration of system boundaries in the evaluation process. Throughout this thesis, the term 'system's perspective' indicates the perspective of the end-users of energy services and products, or more broadly, the societal perspective.

Secondly, while traditional process integration methods effectively minimize external thermal energy use in integrated industrial processes, they are not suitable for identifying innovative configurations involving unconventional integration measures. Given the projected increase in the availability of pure exergy, i.e., electricity, and the anticipated convergence in gas and electricity prices, unconventional integration measures, including process electrification and modifications at the core production units, will become more prevalent. Nonetheless, the ability to identify unique integration possibilities depends on the tools and methods available to the process designer. Therefore, there is a need for exergy-based methods to assess retrofit designs and develop novel integrated processes that achieve high CO_2 avoidance with minimal exergy loss. Finally, site-level factors, such as availability of space for new installations, interconnection costs, and other unaccounted site-level costs currently not considered in incumbent early-stage techno-economic methods, could alter the indication of the optimal decarbonization technology for decarbonizing a specific site, rendering the incumbent methods ineffective.

These limitations in the incumbent assessment methods are bound to not only risk delaying the implementation of decarbonization measures but also raise the possibility of implementing sub-optimal technological solutions at existing industrial sites. Consequently, establishing robust early-stage assessment methods for optimal technology selection from a wide range of technological options becomes crucial to prevent decision paralysis and facilitate the timely deployment of an effective set of decarbonization solutions.

1.1 Aim and scope

This work presents early-stage comparative assessment methods for evaluating mature CO_2 abatement systems and technology alternatives for existing carbon-intensive industries. This thesis aims to highlight the significance of carefully selecting system boundaries, as well as identifying and overcoming limitations of existing methods for process integration and techno-economic assessment methods. The overarching

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[3] Technological readiness levels (TRL) of <7.

goal is to facilitate the enhanced identification of optimal decarbonization technologies, providing valuable insights for decision-making for practical implementation within carbon-intensive industries as they transition toward achieving net-zero CO_2 emissions.

More specifically, this thesis presents a comprehensive assessment framework that combines individual framework methodologies that enable the technical and economic performance comparison of competing decarbonization solutions, with a focus on systems, plant, and site-level considerations for an enhanced indication of the optimal choice of decarbonization technology. This framework comprises the following methodologies:

- A methodology for evaluating decarbonization options in process plants, using exergy as a figure of merit with appropriate selection of system boundaries to highlight potential inconsistencies between the perspectives of the plant owner and the local energy system.
- An iterative combined exergy-pinch analysis with techno-economic analysis that enables the identification of promising process modifications required within the host process plant and the retrofitted decarbonization technology to maximize overall exergy utilization and CO_2 avoidance achieved within the plant.
- A site-specific techno-economic analysis method to enhance cost estimates derived from incumbent early-stage techno-economic assessment methods. This method incorporates pertinent site-specific factors, allowing for the rapid screening of various technologies even with limited site information. The objective is to obtain an enhanced early-stage and site-specific indication of the optimal decarbonization solution for a specific industrial site. Additionally, methods are developed for quantifying site-specific cost escalation factors, including energy supply options, the value of space available at existing sites for new installations, the opportunity cost based on the choice of technology and the timing of its deployment, site-layout dependent CO_2 interconnections, and the potential costs escalation due to forced downtimes and premature decommissioning of the selected decarbonization technology.

The developed methods are applied to case studies in the process industry, examining the best available CO_2 abatement options. Within the context of each case study plant, the following questions are addressed:

- What is the optimal CO_2 capture technology for large-scale BECCS deployment in bio-CHP plants operating within district heating systems, considering both the plant owner's and the end-users' perspectives on CO_2 capture technologies with inherently different exergy requirements per unit of CO_2 captured?
- What cost-effective decarbonization options are available for propylene production technology, which typically emits large amounts of CO_2 in a highly diluted flue gas stream?
- How do various site-specific factors influence the choice of decarbonization solution for a steam cracker plant, considering the potential cost escalation associated with these factors?

1.2 Outline of the thesis

The thesis consists of a summarizing essay and three appended papers. The summary contains six chapters. Chapter 1 introduces and contextualizes the appended papers, presenting the objectives of the undertaken work. Chapter 2 provides the background to this work, including an overview of existing methods to evaluate decarbonization pathways and their limitations. Chapter 3 presents an overview of the methodology, briefly introducing the case studies and frameworks developed in this work. Chapter 4 presents selected case study results, analyzing their implications, followed by the overall conclusions of this work in Chapter 5 and considerations for future work in Chapter 6. The scope of the appended papers is outlined below, with their relationships illustrated in the proposed assessment framework shown in Figure 2.

- **Paper I** provides a comprehensive evaluation of two inherently different carbon capture technologies. These technologies are modeled and integrated with a biomass-fired combined heat and power plant to assess their impact on plant operation. This paper highlights the significance of evaluating integrated decarbonized systems by extending system boundaries beyond the plant level. This extension helps identify optimal solutions from the local energy system's perspective, contrasting with the plant owner's perspective.

- **Paper II** presents a methodological framework that combines exergy and pinch analysis with techno-economic assessment to identify feasible decarbonization opportunities in carbon-intensive process plants. The paper emphasizes and presents an iterative method to conduct stepwise design of decarbonized process configurations that incur minimal exergy loss in their transition to net-zero CO_2 emissions. This method aims to identify promising process modifications in the existing process plant, as well as the retrofitted technology, considering an exergy-based approach. In doing so, it addresses the limitation of traditional thermal integration methods such as pinch analysis, which focuses solely on heat flows between different processes, thus inherently neglecting electrically driven process equipment. The developed methodological framework is demonstrated through an application to propane dehydrogenation technology, which presents a significant decarbonization challenge due to low CO_2 concentrations in highly diluted flue gas.

- **Paper III** introduces a methodological framework that builds upon standardized[4] techno-economic assessment methods by incorporating site-level factors, often overlooked in these approaches. These site-level factors can be pivotal in technology selection, as they may either impede or support the deployment of a specific decarbonization technology at a particular site. The paper presents methods to quantify relevant site-specific factors at an early stage, even with limited site-level information. This quantification enables the assessment of technology-specific retrofitability costs, accounting for both technology-specific attributes and site-level factors. Additionally, the paper introduces a generalized retrofitability assessment tool to evaluate qualitative site-specific factors. When coupled with quantified site-specific costs, this tool can facilitate clear identification of the cost-optimal decarbonization technology for a carbon-intensive industry. The developed methodological framework is demonstrated through a case study of a steam cracker plant.

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[4] 'Standardized' implies an established CCS costing methodology with common nomenclature and consistent cost escalation guidelines. More information on these methods can be found in Ref. [11].

2 Background

This chapter presents existing methods for comparative assessments of CO_2 mitigation options and their limitations in identifying optimal decarbonization technology in the context of different process industries. The theoretical backgrounds for the developed frameworks in this thesis are detailed in the appended papers, **Paper I–Paper III**.

2.1 Existing methodological approaches for comparative assessments of CO_2 mitigation options

Identifying optimal decarbonization technologies in the context of a specific industry is often addressed through a comparative assessment of various best available CO_2 capture technologies or alternative process routes incorporating emerging technologies. The objective of such comparative assessments is to provide an industry-specific[5] indication of the most cost-effective decarbonization pathway considering both retrofit and substitute technologies. The established methodological approach for such assessments typically involves selecting a reference process industry. Following this, system boundaries are defined, and a set of performance indicators is chosen to compare various technological options within the selected industry and its surrounding reference systems. Process modeling tools are then used to determine mass and energy balances, while process integration tools, such as pinch analysis, are applied to explore heat integration opportunities systematically. Finally, standardized bottom-up cost estimation methods are applied to quantify the economic performance of different proposed conceptual designs.

Table 1 provides a list of early-stage comparative assessments that have compared a wide range of decarbonization pathways in various carbon-intensive industries using the aforementioned methodology. It is worth noting here that essential economic performance indicators, such as CO_2 avoidance cost, consistently fall within a narrow range in these studies. These results are often subject to future market conditions, such as fuel, feedstock, electricity, and CO_2 prices, as well as local context considerations, such as access to CO_2 transportation and storage and grid connection capacity. Alternatively, if the comparative assessment reveals a clear differentiation, indicating the optimal decarbonization solution, the technology-specific attributes that might impede their direct integration or retrofit into these industries are seldom addressed. For example, Table 1 shows that oxyfuel combustion incurs the lowest CO_2 avoidance cost (CAC) for refineries, cement, and the petrochemical industry, suggesting it could be considered the cost-optimal solution. However, as highlighted by Hills et al. [12] in the context of retrofitting a cement plant, technology-specific attributes, such as the impact on product quality, operational complexity, and the process changes required in a host plant to minimize gas ingress and leakage from these units, along with other retrofitability aspects, must be considered at an early stage to differentiate proposed technological pathways as either new-build (substitute) or retrofit solutions.

These aspects raise questions about the efficacy and reliability of existing methods in clearly identifying a cost-optimal and technically superior decarbonization technology that can be effectively translated into actual implementation at specific industrial sites. If the differences in CO_2 avoidance costs across a wide range of available decarbonization technologies prove negligible, the process of selecting and deploying technology becomes straightforward, irrespective of the industry. Consequently, ongoing research and development in emerging decarbonization technologies are only justified if potential cost reductions are proven to be substantial, assuming realistic market conditions, in comparison to the best available

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[5] The term 'industry-specific' implies that only generalized process characteristics are considered from a reference process plant. However, no site-specific information are considered in these standardized early-stage techno-economic assessment methods.

technologies. In other words, the development of emerging technologies with comparable economic performance, even in highly optimistic techno-economic scenarios, could be discontinued forthwith.

Table 1: Overview of early-stage techno-economic studies that have applied process modeling and process integration methods with economic analysis with varied levels of detail to compare the overall economic viability of decarbonization via CO_2 capture pathways in different carbon-intensive industries.

Carbon-intensive industries	**Refs.**	**CO_2 capture pathways**	**Estimated CO_2 Avoidance Costs (€/tCO_2)[a]**	**Levelized CO_2 Avoidance Cost (€$_{2023}$/tCO_2)**
Iron & Steel	[13]	Post-combustion (MEA)	64	92
		Post-combustion (advanced amines)	41	59
		Overall reported CAC range for a set of decarbonization pathways	30–75	43–107
	[14]	Post-combustion (MEA), CO_2 captured from hot stoves and CHP	86–116	109–147
	[15]	Post-combustion (MEA), CO_2 captured from hot stoves, coke ovens, lime kilns, and CHP	100–150	127–190
	[16]	Post-combustion (MEA) CO_2 captured from hot stoves, coke ovens, lime kiln, and sinter	56–72	71–91
Refineries	[13]	Post-combustion (MEA)	72–118	103–169
		Oxyfuel combustion	54–55	77–79
	[17]	Post-combustion (MEA), CO_2 captured from hydrogen production unit stack	35–60	43–73
		Post-combustion (MEA), CO_2 captured from all stacks	59–101	72–123
	[18][c]	Post-combustion (MEA)	76–80 (69)	94–109
		Oxyfuel combustion with cryogenic air separation	59–62 (33–38)	45–84
		Oxyfuel combustion -membranes for air separation	52–57 (24–31)	33–77
		Pre-combustion	73–84 (87–90)	99–122
	[19]	Post-combustion (MEA), (T_{reb} = 90°C)	41–57	52–73
		Post-combustion (MEA), (T_{reb} = 120°C)	39–44	50–56
Cement	[20]	Post-combustion (MEA)	80	101
		Oxyfuel combustion	42	53
		Membrane-assisted CO_2 liquefaction	83	106
		Chilled ammonia process	66.2	84
		Calcium looping	52–59	66–75
	[21]	Post-combustion (MEA)	118	137
		Calcium looping	82	95
		CO_2 selective membrane	70	81
		Partial oxyfuel combustion	85	99
		Full oxyfuel combustion	61	71
	[13]	Post-combustion (MEA)	66–131	94–187
		Post-combustion (advanced amines)	37–52	53–74
		Partial oxyfuel combustion	43	62
		Full oxyfuel combustion	44	63
Petrochemical	[13]	Post-combustion (MEA)	118	169
		Pre-combustion (H_2-fired furnaces)	81	116
		Oxyfuel combustion	50–60	72–86
	[22][d]	Post-combustion (MEA), standalone NGCC	58–129	79–175
		Post-combustion (MEA), standalone NG boiler	35–47	48–64
		Post-combustion (MEA), standalone biomass boiler	41–59	56–80
		Post-combustion (MEA) use of current excess heat with industrial heat pumps	26–27	35–37

[a] Note, the reported cost data are from different cost-basis years. Comparing reported CAC estimates across studies within the same industry category is not recommended due to the inherent difference in economic assumptions.

[b] Full oxyfuel combustion includes the kiln and pre-calciner. Partial oxyfuel combustion is considered the pre-calciner alone.

[c] Cost estimates in brackets indicate long-term CAC estimates. The lower bound and upper bound CAC estimates correspond to two different refineries, with annual CO_2 emissions of 4.1 Mt/y and 2.2 Mt/y, respectively.

[d] Energy supply options for an amine-based CO_2 capture technology were compared. The lower and upper bound estimates correspond to an assumed specific reboiler duty range of 2.8–4.7 MJ/kgCO_2.

2.2 Identified limitations in existing process integration and techno-economic methods

2.2.1 Influence of system boundaries

System boundaries delineate the scope of the study, clearly specifying what is included and excluded from the evaluation. In the context of optimal technology selection for the decarbonization of unabated industrial processes, the optimal solution could vary depending on where the system boundaries are drawn. Figure 1 illustrates the hierarchy and interactions among the various system boundaries of a typical unabated process plant. Most studies conducted today, including those listed in Table 1, consistently adopt a system boundary at the plant level. This approach results in a comparative assessment of decarbonization technologies that inherently neglects systems-level considerations, potentially leading to unintended consequences from the societal perspective. On the other hand, failing to consider site-level constraints at existing industrial sites could potentially lead to impractical and uneconomical process designs.

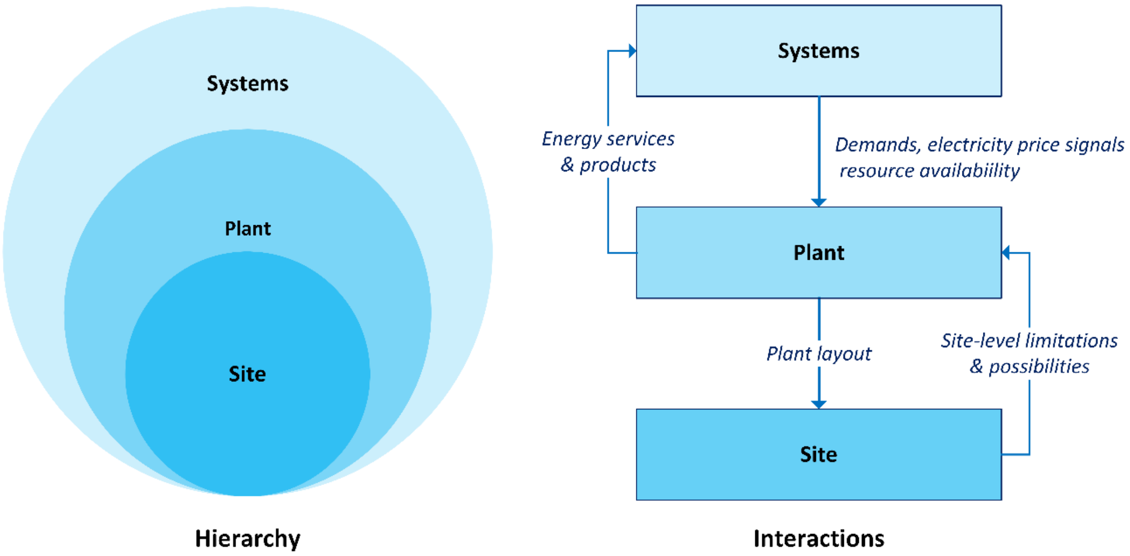

Figure 1: Hierarchy and interaction of the system boundaries for the evaluation of decarbonized industrial systems.

As shown in Figure 1, decarbonization measures in carbon-intensive industries will also impact the interaction, i.e., the exchange of material and energy flows between a specific plant and its local market and energy systems. For instance, most single-product process industries, such as steel and cement, would transform into multi-product sites, generating concentrated CO_2 alongside existent co-products like excess heat and electricity. This transformation implies that, as a consequence of decarbonization, decarbonized process plants will interact more with their local energy system compared to the unabated industrial systems. Given that most CCS technologies, i.e., post-combustion, oxyfuel, or pre-combustion technologies, have different energy and material requirements, their interactions with the local energy system would also vary significantly. Herein lies the first limitation addressed in this thesis – the impact of system boundaries on the early-stage indication of optimal decarbonization technology for existing industries.

Considering that the plant owners would likely initiate investments in CCS technologies, the plant-level evaluation is justifiable as the economic viability of CCS investments hinges on maximizing profits (or minimizing costs) through profitable transactions involving all possible main products and co-products. Therefore, from the plant owner's perspective, the choice of CCS technology is intricately tied to these economic considerations. In contrast, the end-users or consumers of energy services and products would expectedly lean towards CCS technology that maximizes overall CO_2 avoidance while incurring minimal cascading costs to the final product. This divergence in perspective underscores the complex interplay between economic feasibility and CO_2 reduction considerations within industrial decarbonization. Moreover, this inconsistency is expected to be aggravated in industrial energy systems, where the incumbent market or policy conditions favor the installation of a sub-optimal CCS technology at a specific site, which may maximize profit for the plant owner but, consequently, increase the end-users' cost of consumption. Therefore, there is a need for improved assessment methods that consider the expected changes in energy and material flows, especially with the possibility of increased process electrification at industrial sites. Systematic differentiation between various technological choices and a clear distinction between the perspectives of plant owners and systems concerning optimal decarbonization technology is also necessary. Some recent studies have addressed these aspects in the context of carefully selecting system boundaries for chemical process designs [23], life-cycle assessments for negative emissions [24], and cascading costs through value chains [25–28].

2.2.2 Targeting for minimal exergy loss toward net-zero CO_2 emissions process plant configurations

At an early stage of technology selection and integration studies, process integration methods such as pinch analysis are widely used to quantify minimum heating and cooling demands as well as the theoretical potential for maximum process heat recovery. Traditionally, these methods have been widely used to identify energy-saving measures in the process industries to minimize overall thermal energy losses in existing processes and, thereby, minimize primary energy use to reduce operational costs. Energy savings of up to 20–40% in the process industry have been achieved in the past using pinch analysis tools [29,30]. Pinch analysis has been extensively used to identify energy-efficient CO_2 mitigation options in integrated oil refinery concepts [19,31–33], biomass-based processes for the production of synthetic natural gas [34,35], platform chemicals [35–37], pulp mills [38], and steelmaking [39]. It was primarily used in these studies to improve the overall energy efficiency of these conceptual integrated processes and thereby reduce their primary energy use, resulting in avoided CO_2 emissions. These examples reveal that the primary driver for the application of pinch-based targeting methods has changed over time, from reducing operational costs to reducing overall CO_2 emissions. Furthermore, their application has expanded from evaluating retrofit designs in existing industries to evaluating grassroots designs of integrated processes.

However, one of the limitations of conventional pinch analysis is that it inherently neglects the performance of existing and newly retrofitted electricity-driven process equipment in the integrated processes. Inefficient electrically driven processes are only identified when a value is ascribed to them in economic or exergy terms at a later stage of the assessment. Here, inefficient electrically driven processes imply the utilization of electricity, a high-quality energy carrier, in process equipment with significantly higher exergy destruction, resulting from either process heating at relatively lower temperatures or motor-driven units with poor mechanical efficiencies. In addition, conventional heat integration studies cannot identify such inefficiencies, as they do not analyze exergy, which may lead to forgoing promising process modifications at successive design stages.

Consequently, pinch-based energy targeting methods are expected to be inadequate in identifying process modifications, especially when designing decarbonized industrial systems with increased process electrification. Process electrification has not been considered a decarbonization measure in the past due to the high cost and high carbon intensity in the power sector. Nonetheless, with rapidly declining grid carbon intensity [40], process electrification provides the possibility of both improving plant performance and reducing CO_2 emissions.

As a result, there is a clear need for exergy-based targeting methods that depart from the traditional methods of minimizing thermal energy consumption to methods that maximize exergy utilization within conceptual process designs of decarbonized process industries. Furthermore, such exergy-based targeting methods are essential for selecting optimal decarbonization technologies for process plants, which, in turn, provide greater societal value in terms of increasing the overall CO_2 avoidance achieved in the local energy system in which the process plant operates.

2.2.3 Site-specific techno-economic analysis

Early-stage techno-economic assessments serve as crucial inputs for various stakeholders, including governmental organizations, funding agencies, industry, and academia [11,41]. Within industries, these results lay the basis for a more comprehensive economic assessment with an increased level of engineering detail and project definition, incorporating site-level factors to estimate the expected cost of a selected decarbonization solution. A conclusive indication and selection of a specific decarbonization technology and pathway is typically followed up with securing project commitment and a final investment decision for the project.

While there is a plethora of early-stage TEA studies for various industrial sectors, some of which are listed in Table 1, these studies generally fall short of drawing concrete conclusions on the optimal decarbonization technology for the considered industry, indicating no standout winners in the compared set of decarbonization technologies. The limitation lies in the fact that the existing early-stage techno-economic methods are standardized to enable a fair comparison of different CCS technologies. The standardization of costing methodologies was motivated by the fact that reported cost estimates were highly heterogeneous, limiting their comparability with other reported results for the same industry. Concerted efforts have been made in the past to harmonize and standardize costing methods for differentiating between emerging and mature technologies [42], transferability and identifying limitations of costing methods from the CCS in the power industry to non-power industries [11], and application of uncertainty analysis methods for better understanding in the source of uncertainties in TEA studies. The key economic performance indicators used for comparisons, such as the CO_2 avoidance costs, often lie in a narrow range, well within the accuracy range (between 15% to +50% [43]) expected from the applied cost estimation methods. In addition, the inconclusiveness is often attributed to qualitative and quantitative uncertainties regarding operational experience, technology maturity, site-level factors, and local market conditions.

One of the primary limitations of standardized TEA methods is that site-related constraints and opportunities are often neglected to provide a fair comparison between different technological options. This limitation could lead to a severe disparity between the early-stage indication of cost-optimal CCS technology for a specific process industry and its actual (post-deployment) economic performance at a specific plant, which could also vary from plant to plant within the same industry sector, based on their site-related conditions. Recent developments in CCS in the power sector have shown that a majority of CCS projects applying large-scale amine-based post-combustion technology have been terminated (see database in Refs. [44,45]) due to significant project cost escalation at advanced design stages, unfavorable market conditions, or advancement in alternative decarbonization solutions that were initially deemed infeasible for a specific process industry. These deployment failures highlight the limitations of the existing early-stage standardized TEA methods in accurately identifying cost-optimal[6] decarbonization alternatives for process industries.

Roussanaly et al. [9,10] addressed these aspects with guidelines for CCS cost evaluation in the process industry that inherently has varying site-specific features. Martorell et al. [46] highlighted that site-related factors, often not considered in early-stage cost estimates, contribute significantly to the final capital cost required to realize the project. They compared differences in detailed cost estimates from front-end engineering design (FEED) studies for CO_2 capture via amine scrubbing for two natural gas combined-cycle power plants[7]. Site-specific factors such as site layout (determining the extent of flue gas conveying

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[6] 'Cost-optimal' hereinafter refers to a decarbonization option incurring the lowest CO_2 avoidance cost (compared to a non-exhaustive list of alternative decarbonization options) at a host process plant.

[7] With nominal capture capacities of 197 and 129 tCO_2/h, respectively.

equipment), resource availability (e.g., water), and steam supply alternatives were identified as key site-specific factors that may escalate capital cost estimates, compared to early-stage estimates using standardized TEA methods.

Transforming existing industrial infrastructure into decarbonized industrial systems necessitates a thorough evaluation of CO_2 abatement options tailored to the site-specific characteristics of each plant within an industrial sector at an early stage. The limitations of incumbent TEA methods raise questions about their reliability and utility in providing a conclusive indication of the optimal decarbonization pathway. Besides, these early-stage TEA studies ultimately form the basis for subsequent detailed evaluations of technology selections by the industry to execute their decarbonization projects within their limited timeframe.

This work addresses the need for rapid technology selection with limited site-level data at an early stage. An early-stage TEA method based on site-specific factors is introduced and generalized for applicability to other process industries. The introduced method aims to go beyond the incumbent industry-specific indication of optimal decarbonization technology, which is primarily based on the process and flue gas characteristics. This approach seeks to close the gap between cost estimates from early-stage TEA assessments and actual expected project costs, aiming to decrease upfront uncertainty regarding the choice of technology and thereby accelerate CCS deployment in these industries.

3 Methodology

This chapter is divided into four sections. Firstly, Section 3.1 introduces an overview of the methodology applied in this work, combining the individual framework methodologies developed in **Paper I–Paper III**. Within each paper, the limitations of existing methods identified (Section 2.2) are systematically addressed at multi-level system boundaries, progressing from systems-level to plant-level and site-level (Figure 1). Next, the selected industrial case studies are described in Section 3.2, followed by a detailed description of the developed framework methodologies in Section 3.3, and finally, the applied methods within these frameworks in Section 3.4.

3.1 Method Overview

Figure 2 illustrates the hybrid[8] assessment framework designed to overcome the identified limitations of incumbent process integration and techno-economic methods. The proposed framework is generalized for its applicability to other process industries. The overarching goal of this hybrid framework is to provide a comprehensive early-stage approach that considers influencing factors at each level of evaluation to obtain an enhanced indication of the optimal decarbonization pathway in a specific process plant.

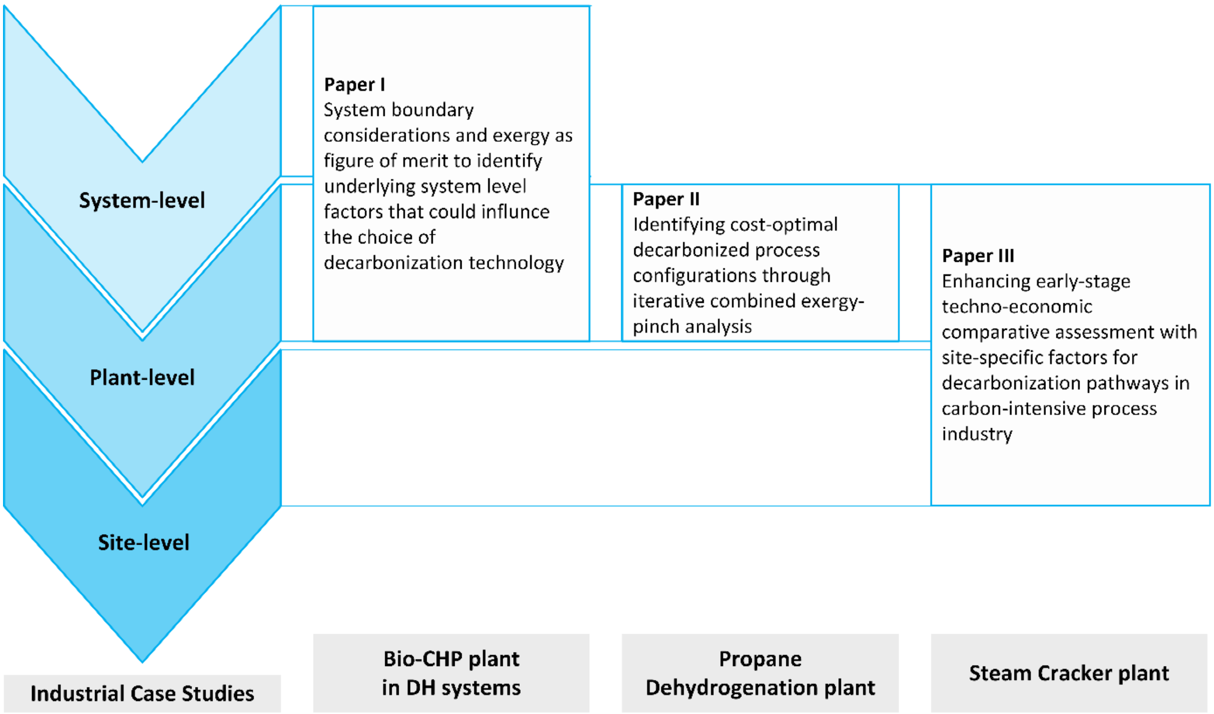

Figure 2: Hybrid assessment framework addressing limitation of incumbent process integration and techno-economic methods. Each of these limitations at different evaluation levels is addressed with the developed framework methodologies in **Paper I–Paper III**.

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[8] The term 'hybrid' implies that, although the methodology suggests a top-down approach, initiating with systems-level considerations followed by plant-level and site-level considerations, the methods employed within the individual methodological frameworks developed in **Papers I–III** take a bottom-up approach.

In this assessment framework, the following considerations must be taken into account at each level of evaluation:

- **Systems-level considerations:** Define the plant system boundary of the existing process plant and consider its interactions with its local energy system, i.e., its extended system boundary. Consider the possibility of pre-existing market or policy conditions that may facilitate the installation of a sub-optimal decarbonization technology. This technology may be perceived as cost-optimal at the plant level from the plant owner's perspective. However, it may be deemed sub-optimal from the societal perspective.

- **Plant-level considerations:** Within the plant system boundary, consider and evaluate the possibility of eliminating avoidable exergy losses with process modifications in installed process equipment as well as the new process equipment to be installed with the decarbonization technology. Additionally, investigate the possibility of replacing combustion exergy losses, which are typically one of the largest sources of inevitable exergy losses in industrial processes, with either direct electrification or indirect process integration measures. (discussed in Section 3.3.2).

- **Site-level considerations:** Constraints and opportunities at the site-level are expected to influence the final CO_2 avoidance from selected decarbonization technology. Therefore, consider and quantify the pertinent site-specific factors and their associated costs to derive site-specific indications for cost-optimal decarbonization solutions. The descriptions of site-specific factors and cost-quantification sub-methods can be found in **Paper III**. The total site-specific costs (in €/tCO_2), defined as the additional cost of retrofitting, can then serve as a cost escalation factor to determine the final CO_2 avoidance cost specific to each considered decarbonization technology. Additionally, qualitative site-specific factors could be assessed through expert elicitation using the *retrofitability assessment matrix*. This qualitative assessment, together with *site-specific CO_2 avoidance cost,* facilitates informed technology-selection for a specific process plant considering its site-level factors.

3.2 Selected case studies

In **Paper I**, the imperative to consider underlying systems-level factors, such as market or policy conditions that facilitate the deployment of a sub-optimal decarbonization technology, was addressed with a case study of a future BECCS plant in the Swedish district heating (DH) context (Ref. [47]). The case study presents the impact on the performance of the biomass-fired combined heat and power (bio-CHP) plant in a DH system when equipped with two different CO_2 capture processes with inherently different exergy requirements per unit of CO_2 captured from the flue gases (see Figure A1).

A detailed technology comparison was conducted to highlight the inconsistent views regarding the perceived value of high-quality energy carriers at both the plant and system levels. The case study includes a detailed performance comparison of the CO_2 capture technologies[9] as stand-alone processes and the respective bio-CHP plant performance. Lastly, the study examines their influence on end-users in the local DH system, considering the possibility of centralized and decentralized DH with industrial heat pumps and substitute technologies such as ground source heat pumps, respectively.

In **Paper II**, the need for alternative exergy-based methods, driven by the limitations of heat-flow analysis (pinch analysis), was addressed. An alternate exergy-based approach was introduced that combined an iterative combined exergy-pinch analysis (CEP) with techno-economic analysis. A propane dehydrogenation (PDH) plant using a state-of-the-art PDH process technology (CATOFIN® Lummus PDH technology)

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[9] End-of-pipe CO_2 capture technologies using benchmark monoethanolamine and potassium carbonate as capture solvents.

was selected as the industrial case study (see Figure A2) to identify promising process modifications that maximize overall exergy utilization and CO_2 avoidance when retrofitted with different decarbonization technologies. The selection of this process plant is motivated by its significant decarbonization challenge, where the direct retrofitting of end-of-pipe CO_2 capture is expected to be highly cost-intensive due to the substantial volume of flue gases to be handled and the ultra-low concentrations of CO_2 in the flue gas. Moreover, alternative decarbonization strategies for this technology (e.g., oxyfuel combustion) were considered unfeasible due to process-related constraints.

In **Paper III**, a steam cracker plant was selected as the industrial case study, where mature decarbonization options of end-of-pipe CO_2 capture CO_2 capture (Post-CCS) and hydrogen-firing via pre-combustion CO_2 capture (Pre-CCS) in the cracker furnaces were compared to assess the impact of site-specific factors on these technologies. The main material flows of these decarbonization pathways for the steam cracker plant are shown in Appendix A (Figure A4). Site-specific factors, such as existing energy supply options, space availability for installation of new equipment, site-layout constraints, annual maintenance schedules, and planned turnarounds, were considered. Furthermore, **Paper III** introduces a generalized retrofitability assessment matrix, which was employed to assess the qualitative site-specific factors of the steam cracker plant and technology-specific attributes through expert elicitation. Detailed process descriptions of the selected reference industries can be found in the respective appended papers.

3.3 Developed approaches

3.3.1 Impact of system boundaries on the choice of decarbonization technology

Inconsistencies between the plant and end-users' perspectives arise from the fact that the plant owner would typically prefer decarbonization technologies that incur the lowest specific CO_2 capture cost, which minimizes the impact on production costs, assuming that emitting CO_2 emissions into the atmosphere is no longer free. However, using CO_2 capture costs as a metric for technology comparison is also flawed, as it may favor technologies that capture larger amounts of on-site emissions. Conversely, from the end-users' perspective, decarbonization technologies that maximize CO_2 avoidance would be preferable, with minimal marginal cost increase in the final product, assuming that the plant owner passes on the costs to its consumers.

Three key considerations must be taken into account to observe these differences in perspectives. Firstly, one must assess and preserve the core functionality of the unabated process plant, whether it involves providing energy services or producing the primary product. Secondly, the use of exergy as a figure of merit is crucial for evaluating and comparing various decarbonization options that inherently have different exergy requirements. Thirdly, it is essential to consider the impact on plant performance resulting from the implementation and integration of decarbonization technology within the plant boundaries to establish what would be perceived as cost-optimal from the plant owner's perspective. Subsequently, expanding the system boundary to include the immediate consumers of energy services and products from the now-decarbonized plant becomes necessary. This expansion allows for the determination of which technology or process configuration retains the highest exergy efficiency within the broader system. Figure 3 depicts this system boundary expansion in the context of a bio-CHP plant operating within a district heating system.

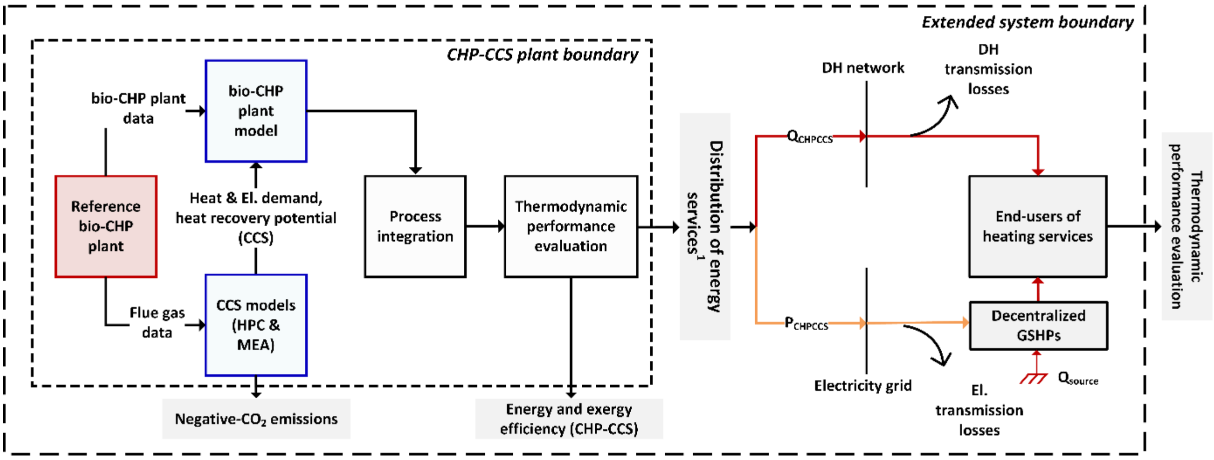

Figure 3: Overview of the methods and models used with the two system boundaries: bio-CHP plant boundaries and the extended system boundary (the local DH system). The reference bio-CHP plant (indicated in red) data are fed to the developed numeric models, indicated in blue. Finally, the applied methods and results obtained are highlighted in gray. [1]Delivered energy services of the bio-CHP plant equipped with CCS units (CHP-CCS), which includes the net district heating (DH) and power delivered to the bio-CHP plant's local DH system. Note that the electricity delivered to the grid is assumed to be consumed in decentralized ground-source heat pumps (GSHPs) to meet indoor climate needs, i.e., domestic hot water and space heating. Source: **Paper I**.

District heating networks are typically considered natural monopolies [48]. Therefore, CHP plants in DH networks primarily prioritize delivering district heat over electricity power production, as generated electric power is traded in the power market and is subject to competition and price volatility [49]. On the other hand, the immediate energy consumers in the DH network naturally assign a higher value to an energy carrier with higher exergy, i.e., electricity, as it can be utilized for various purposes (heating, cooling, and other power demands in households or industrial applications) in the local energy system. Moreover, electricity transmission losses are expected to be minimal compared to transmission losses incurred during DH supply over long distances. Therefore, the value assigned to the different energy services/carriers of an existing bio-CHP plant, i.e., heat and electricity, differs at the plant level than at the system level (energy consumers). Herein lies the underlying systems-level factor for this selected case study industry, which could potentially alter the optimal choice of decarbonization technology. Specifically, it pertains to the difference in the value assigned to electricity and heat, driving the bio-CHP plant operator in a DH system to opt for a carbon capture technology that minimizes the loss of heat sales. This observation can be confirmed by using exergy efficiency as the key performance indicator for comparing different CO_2 capture technologies within the bio-CHP plant boundary and subsequently within the local DH system.

3.3.2 Identifying promising decarbonization process configurations using iterative combined exergy-pinch analysis

The framework methodology proposed in **Paper II** incorporates a combined exergy-pinch analysis (CEP), which was initially proposed by Feng et al. [50] to set performance improvement targets based on the concepts of avoidable and inevitable exergy losses. The avoidable exergy losses within different process equipment in an existing system indicate the maximum potential for improvement that can be achieved with process modifications under current technical and economic conditions [50]. Feng et al. [50] introduced an exergy-energy (Ω-H) diagram to overcome the limitations of pinch analysis in representing only heat transfer processes. These concepts were adapted in **Paper II** to identify promising process modifications and subsequently evaluate process configurations resulting from retrofitting process technologies into unabated process plants. Additionally, an iterative approach for the CEP analysis was employed to assess the economic performance of each implemented process modification, aiming to achieve cost-effective CO_2 capture with minimal exergy losses.

In the CEP analysis proposed by Feng et al. [50], the process modification targeting method involved a merit order list where the largest avoidable exergy losses were addressed while neglecting inevitable exergy losses. As per their definition, the inevitable exergy loss is the exergy loss that cannot be avoided technically or economically. This definition implies that exergy losses, previously deemed inevitable, can be overcome with technological advancements and changes in economic conditions. However, this holds true until the exergy losses reach the minimum value that allows the process to function. Therefore, this limits the extent of process modifications that can be practically carried out to improve exergy efficiency.

Considering today's context of improving resource efficiency and minimizing CO_2 emissions, process modifications could be considered to replace existing combustion processes, which are typically one of the largest sources of inevitable exergy losses in industrial processes. For instance, process electrification for industrial process heating at higher temperature levels could provide the compounded advantage of avoiding direct CO_2 emissions on-site while minimizing exergy losses associated with combustion.

In this context, **Paper II** focused primarily on the inevitable exergy losses due to the combustion of carbon-based fuels, which are inherently irreversible and the primary cause of CO_2 emissions. To this end, an additional screening process was incorporated into the CEP analysis to address the inevitable exergy losses in combustion processes via methods to either replace or modify the existing combustion process. This screening step considers the temperature level at which the process requires energy and the possibility of minimizing combustion exergy losses through two approaches: (i) switching to electrified process heaters for process heating requirements >1000°C (dependent on heating element properties[10]), (ii) direct or indirect integration of an industrial gas turbine (GT), for process heating requirements below 800°C.

Direct electrification via heating elements directly converts the consumed electricity into heat at the desired temperature levels, with the possibility of uniform and controllable heat flux. The integration of gas-fired turbines is relatively straightforward because the advanced materials used in the turbines enable enhanced utilization of fuel exergy between the adiabatic flame temperature and the turbine outlet temperature. These efficiencies are typically lost in conventional process heaters and furnaces due to material constraints. With this, the merit order of process modifications starts with the combustion-related exergy losses first, followed by process equipment (reference process plant and the retrofitted decarbonization technology) with the largest to the smallest avoidable exergy loss. Finally, compatible process modifications are implemented to obtain promising decarbonized process configurations that incur minimal exergy losses in their transition to net-zero CO_2 emissions. The steps involved in the framework are described below in the context of an unabated propane dehydrogenation (PDH) plant, shown in Figure A2. However, they are generalizable and applicable to other process plants for techno-economic comparison of alternative process configurations with relevant decarbonization technologies, allowing for the determination of cost-optimal decarbonized process plant configuration, incurring minimal exergy loss. In Figure 4, PC_0 indicates the first decarbonized process configuration where a straightforward retrofit of the CCS plant is performed, with no modifications carried out in either the PDH plant or amine-based CC plant, which served as the basis for the first CEP analysis.

Each iteration of the CEP analysis involved the following steps: i) generation and analysis of the Ω-H diagram, ii) Identification of process modification options based on the generated Ω-H diagram, and the aforementioned merit-order list for process modifications that start with the screening steps to minimize or avoid the combustion exergy losses with direct electrification or direct process-to-process integration measures, iii) implementation of the identified process modifications compatible with the PDH process in order to minimize the avoidable exergy losses. Thereby, a first modified (decarbonized) process configuration (PC_1) is defined, with relatively lower exergy destruction compared to PC_0.

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[10] Maximum element temperature and maximum watt loading (W/m^2)

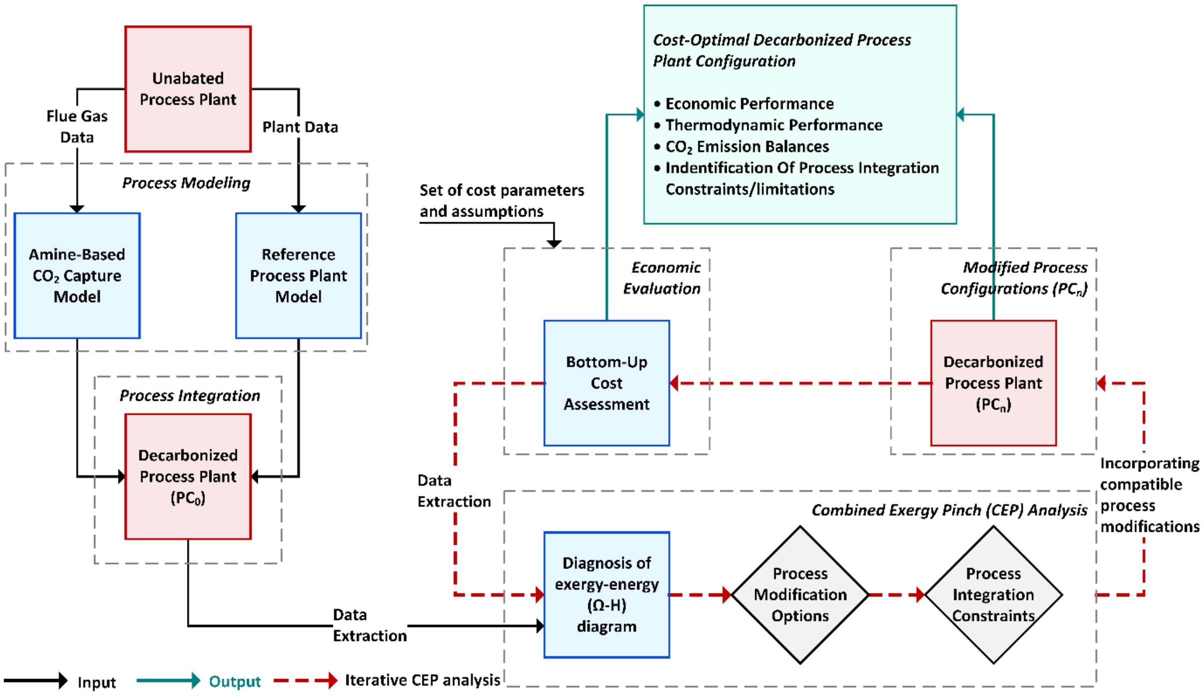

Figure 4: Overview of the applied methodological framework and method used – process modeling, process integration, combined-exergy pinch, and techno-economic analysis, indicated with gray-dashed boxes. Blue boxes indicate the numeric models developed. Gray-shaded boxes indicate the evaluation of process modification options. Process configurations and main outputs are indicated in red and green boxes. The integrated process configurations are abbreviated as 'PC,' which is suffixed with a number, indicating the number of stepwise implemented modifications. Source: **Paper II**

Finally, the iterations are repeated until an optimal decarbonized PDH process configuration (PC_n) is achieved, incurring relatively lower exergy losses and the lowest cost of CO_2 avoidance. The cost estimation methodology (Figure 7), incorporating cost parameters and assumptions from Refs [17,51–53], is applied. The step-by-step procedure of the CEP analysis, involving the generation and diagnosis of the (Ω-H) diagram, can be found in **Paper II**.

3.3.3 Site-specific techno-economic analysis

Paper III introduces a framework methodology that combines process modeling and integration tools, merging early-stage standardized techno-economic (s-TEA) methods with site-specific factors. The purpose is to clearly identify and differentiate the cost of decarbonization in terms of CO_2 avoidance of different decarbonization technologies at an early stage, as depicted in Figure 5. The framework accounts for site-level factors, providing an opportunity to improve early-stage identification of the optimal[11] decarbonization technology.

Site-specific conditions naturally vary among plants within the same industry. These site-specific factors encompass spatial and time-related constraints contingent upon the type and location of unabated carbon-intensive industries. In **Paper III**, these pertinent site-specific factors were identified and categorized as quantitative and qualitative factors. Generalized quantification methods and tools were developed for quantitative site-specific factors, such as energy supply costs, opportunity costs associated with the occupancy of available space on existing sites, site-layout-dependent CO_2 interconnections, and costs

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[11] The term 'cost-optimal' hereinafter refers to a decarbonization option incurring the lowest CO_2 avoidance cost (compared to a non-exhaustive list of alternative decarbonization options) at a host process plant

incurred due to forced downtime and premature decommissioning of newly installed equipment resulting from being locked into the end-of-life of the host plant.

These methods and tools can be applied with limited site-specific information from a particular site. Essential prerequisites for quantifying the impact of opportunity cost, site-layout dependent interconnections, and energy supply options, as well as for conducting site-specific TEA, include site-specific information such as the existing site-energy system, site layout, maintenance schedules, placement, and operation lifetimes of existing assets. Sensitivity analysis is recommended with respect to factors such as forced downtime and premature decommissioning due to the technology lock-in effect to evaluate the technology disparity in expected cost escalations. This evaluation considers delays during installation in the short term and deployment in the long term relative to the residual lifetime of the host plant.

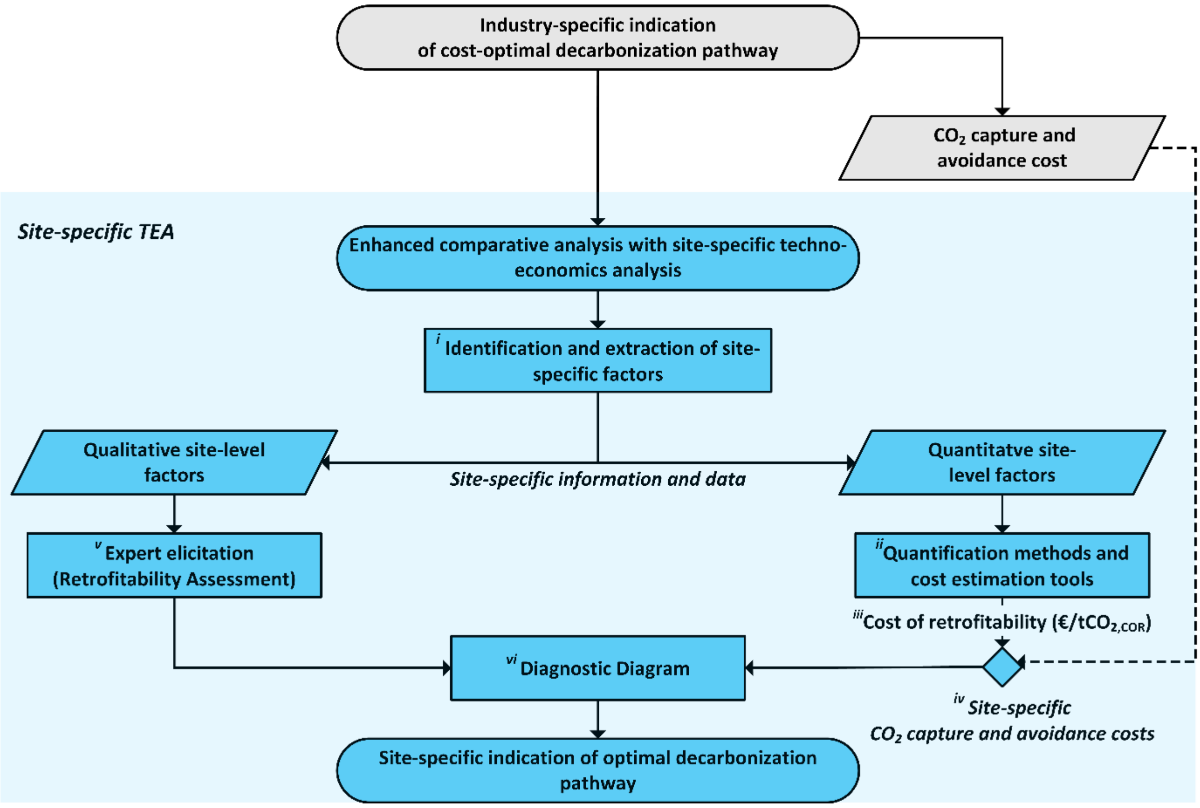

Figure 5: Overview of the framework methodology applied in **Paper III** to enhance the comparative analysis of different decarbonization pathways, incorporating site-specific factors. The oval symbol indicates the start/end of the framework methodology, parallelograms indicate input/calculated data, and rectangles indicate methods. Gray-shaded shapes indicate the application of the standardized TEA method, while the blue-shaded shapes indicate the site-specific techno-economic analysis. Adapted from **Paper III**.

Furthermore, a qualitative retrofitability assessment matrix is introduced in **Paper III** to evaluate factors that could potentially support or impede the integration of decarbonization technology at a specific industrial site. This assessment involves expert elicitation on factors such as dependence on external energy supply, reliance on external infrastructure (electricity grid, CO_2 pipelines), sensitivity to fuel and electricity prices, complexity of system integration, adaptability to future feedstock switches, availability of resources, alternative utilization of CO_2 capture equipment, and the feasibility of achieving 100% carbon recovery for CO_2 utilization pathways. Detailed descriptions of these site-specific factors, generalized quantification methods, cost estimation tools, and the retrofitability assessment matrix can be found in **Paper III**.

In general, the application of the developed framework involves the following steps: (i) identification and extraction of site-specific factors expected to influence the final cost of decarbonization for the studied process plant; (ii) quantification of site-specific factors with their respective estimation methods or tools;

(iii) estimation of the technology-specific cost of retrofitability (COR), the sum of all quantified site-specific factors, for each considered decarbonization technology; (iv) calculation of the site-specific cost of CO_2 capture (€/$tCO_{2,ss\text{-}cap}$) and avoidance (€/$tCO_{2,ss\text{-}avo}$)[12]; (v) evaluation of qualitative factors (technology and site-level attributes) via expert elicitation using the retrofitability assessment matrix; (vi) finally, compilation and visualization of the results from the qualitative retrofitability assessment, sensitivity analysis, and site-specific costs in a diagnostics diagram to obtain an enhanced visual indication of the optimal decarbonization pathway for a specific process plant. Outlined below is a brief description and quantification method for the considered site-specific factors.

Opportunity costs: The opportunity cost of the spatial footprint of decarbonization technologies (C_{OC}) can be estimated based on the opportunity cost of occupying available land with decarbonization technologies, where the alternative use would be to maintain current production with an added cost of emitting CO_2 or installing emerging low-carbon production technology at a later time. To this end, a space-value graph was conceptualized based on the process design hierarchy [54] to enable a generalized categorization of site layouts of different process industries with typically large spatial footprints. The space-value graph was complemented with a merit order for space utilization to determine where decarbonization equipment could be placed on-site at an early assessment stage. The merit order was classified based on the available space within an existing process plant and its type, i.e., brownfield or greenfield areas. Here, the unoccupied brownfield areas were prioritized first, followed by brownfield areas with redundant units (after removal/rearrangement) and greenfield areas. Finally, depending on the pre-determined placement of the decarbonization equipment within the plant, the opportunity cost was calculated as forgone revenue over the lifetime of the host plant. The space occupied by the decarbonization technologies prevents the installation of other alternatives for decarbonization with a delay in deployment or the possibility of operating the unabated process plant while incurring a direct operational cost in the form of EU-ETS permits price per tonne of CO_2 emitted. In contrast, early deployment of decarbonization technology avoids both CO_2 emissions and associated emissions costs. Therefore, a net present value method was used to discount the cumulative forgone annual revenue and avoided emissions costs related to the averaged EU-ETS allowance price for the residual lifetime of the host process plant, as these cash flows are not realized until the end of a specific year of decarbonized operation.

Site-layout-dependent interconnections: The site-layout-dependent interconnection costs, including the cost of flue gas ductwork, solvent, and CO_2 piping, are incurred as CAPEX during construction. This capital cost, expressed in terms of technology-specific annual CO_2 avoidance, represents the total specific CO_2 network costs ($C_{netw,}$ €/$tCO_{2,avo}$). A network design hierarchy, adapted from Berghout et al. [55], was followed to determine the design and technical specifications of each component of the local CO_2 transportation network. In **Paper III**, a simplified local CO_2 network cost estimation tool for the total CO_2 interconnection cost was developed based on the network design calculation method from Berghout et al. [55]. The main inputs to the network design calculations were the operating pressure and temperature of the pipeline, along with the flow rates and physicochemical properties of the liquid/gas transported. These costs were subject to limitations imposed by the site layout, for which an optimal network route within the plant boundary of the case study plant was determined using aerial images and additional site-specific information, such as available space within the plant where capture equipment could be accommodated and installed.

Forced downtime: The cost of forced downtime (C_{FD}) was quantified as lost revenue (M€) corresponding to the time a process plant is shut down, assuming that integration of decarbonization technologies renders the host process plant non-operational beyond the timeframe of the scheduled maintenance shutdown.

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[12] Site-specific costs are the sum of CO_2 capture and CO_2 avoidance cost, estimates from the bottom-up cost estimation method (Figure 7), and the estimated cost of retrofitability in terms of €/$tCO_{2,cap}$ or €/$tCO_{2,avo}$, respectively.

Depending on the year of operation and the process industry, this timeframe could last up to two to four weeks in a regular year and one to three months in a turnaround year. Given that it is rather challenging to foresee commissioning delays, an informed early-stage assessment can be performed with a sensitivity analysis on the lost revenue incurred due to forced downtime, which was accounted as CAPEX during the construction of the decarbonization technology.

Delay in CCS adoption/deployment (lock-in effect): The lock-in effect of the decarbonization pathway, with its host process plant, was demonstrated with the capital recovery factory applied to its total capital requirement, where the operational lifetime of the decarbonization technology was assumed to be equal to the residual lifetime of the decarbonized process plant, to obtain the cost of premature decommissioning (C_{PD}). Alternatively, a decarbonization technology with the possibility of operating as a stand-alone process plant, such as the process equipment in the Pre-CCS pathway, is not considered to be locked into its host process plant. Therefore, an operational lifetime equal to the design lifetime of the newly installed equipment was assumed. A sensitivity analysis was performed to visualize how CO_2 capture costs and avoidance costs escalate with a delay in installing and deploying decarbonization technologies that tend to be locked into their host plant.

The sum of all aforementioned site-specific costs was quantified as the technology-specific cost of retrofitability ($C_{COR,}$ €/$tCO_{2,avo}$), which excludes the technical costs of CO_2 avoidance (indicated in grey in Figure 5). The site-specific cost of avoidance ($CAC_{site\text{-}specific}$) was then calculated as the sum of the cost of retrofitability ($C_{COR,avo}$) to the technical cost of CO_2 avoidance, as shown in Equations 4–6. This work also investigated the impact of cost and emissions intensity of energy supply options for the considered decarbonization technologies. More detailed information and calculation procedures for these site-specific factors can be found in **Paper III**.

Retrofitability assessment matrix and diagnostic diagram: The retrofitability assessment matrix, detailed in the Supplementary Material of **Paper III**, incorporates generalized definitions for each qualitative factor, which are applicable to any unabated process plant of interest. The expert elicitation on site-specific factors involves a qualitative assessment based on their perception of the expected impact of a certain decarbonization technology on the host process plant. Each site-specific qualitative factor is assigned a 0–1 impact score. An impact score of 1 indicates that the evaluated factor has a higher overall impact on the host plant, implying higher perceived risks and unforeseen costs, while a score of 0 indicates no impact on the host plant.

The diagnostic diagram, presented as a spider plot with a 0–1 scale, combines the outcomes of quantified site-specific factors, sensitivity analysis, and the retrofitability assessment matrix. A greater spread on the spider plot for a decarbonization technology suggests a sub-optimal choice compared to other options depicted on the same plot. The spider plot serves the purpose of comparing factors that are not directly comparable with one another and are not individually the primary determinants in selecting the cost-optimal decarbonization technology. No weighting factors were assumed for the case study in **Paper III**. However, weighting factors could be assigned to the individual qualitative site-specific factors deemed important for other industrial sites. The calculation method used for plotting the site-specific factors on the diagnostic diagram is detailed in **Paper III**.

3.4 Applied methods

Figure 6 shows an overview of the methods applied and tools developed in this work. The following section briefly describes these methods, and for a detailed explanation of the developed methods and tools, the reader is referred to the appended papers.

	Applied methods and developed tools	Paper I	Paper II	Paper III
Systems level	Exergy analysis within local district heating system of the BECCS plant	■		
Plant/process level	**Process modeling**	■	■	■
	Amine-based CO_2 capture process	■		
	Hot-potassium carbonate process	■		
	Decarbonization pathways in propane dehydrogenation plant		■	
	Decarbonization pathways in steam cracker plant			■
	- *Aspen Plus*			■
	Process and heat integration analyses	■	■	■
	Pinch analysis	■	■	■
	- *Aspen Energy Analyzer*	■	■	■
	Exergy analysis	■	■	
	Combined exergy-pinch analysis		■	
	Graphical representation of Ω-H diagram		■	
	- *MATLAB*		■	
	Hybrid bottom-up and top-down cost estimation		■	■
	- *via cost literature & Aspen Process Economic Analyzer*		■	■
Site level	**Site-specific techno-economic analysis**			■
	Spatial footprint estimation for amine-based CO_2 capture technology			■
	- *Publicly available FEED-data derived spatial footprint correlations*			■
	Site-specific opportunity cost of occupying available space on-site			■
	-*Site-layout categorization and site-specific space-value functions based on process design hierarchy*			■
	- *Aerial images of industrial sites using Google Maps and ArcGIS*			■
	Local CO_2 network sizing and cost estimation tool			■
	- *MATLAB*			■
	Sensitivity analysis			■
	Impact of emissions intensity of energy supply			■
	Impact of energy supply costs on CO_2 avoidance costs			■
	Impact of site-layout constraints on opportunity costs			■
	- *MATLAB*			■
	Qualitative assessment			■
	Expert elicitation			■
	- via *Retrofitability Assessment Matrix*			■
	Combined assessment of site-specific quantitative and qualitative factors			■
	- *Diagnostic Diagram*			■

Figure 6: Overview of applied methods and developed tools and their links to the appended papers. The tools used are indicated in italics.

3.4.1 Steady-state process modeling

In **Paper I**, a process model of the reference bio-CHP plant was developed using EBSILON Professional with available plant data. Additionally, process simulation models of the MEA and HPC capture process adopting rigorous rate-based models with detailed reaction kinetics were developed and validated. The MEA model, representing the amine-based capture process with an aqueous solution of monoethanolamine (30 wt.% MEA), and the hot-potassium carbonate (30 wt.% K_2CO_3) process model were developed based on data sourced from previous works by Biermann et al. [56] and Gustafsson et al. [57], respectively. The models provide necessary mass and energy balances, including stream and equipment design data, that are subsequently used for process integration, as described in Section 3.4.2.

In **Paper II**, the reference air-regeneration train of the propane dehydrogenation process was modeled, which was subsequently integrated with an MEA process model for CO_2 capture from its flue gases. This model served as the reference decarbonized process model in which a CO_2 capture (CC) plant is retrofitted without any modifications to the existing process. The compatible process modifications identified through iterative combined-exergy pinch analysis were implemented until improvements in overall exergy efficiency, along with a reduction in CO_2 avoidance costs, were achieved relative to the preceding decarbonized process configuration.

In **Paper III**, the Post-CCS pathway was simulated using an updated MEA model with the flue gas data from reference cracker plant data, while the Pre-CCS process assumes an equilibrium-based model to simulate hydrogen production on-site valorizing methane-rich fuel gas obtained from the steam cracking process. Detailed modeling descriptions, system boundaries, process flowsheet diagrams, key assumptions, and process parameters of the process models can be found in the respective papers.

3.4.2 Process Integration

Grand composite curves (GCCs) and foreground–background analysis were applied for energy targeting in **Paper I–Paper III**. The GCCs graphically represent the net heat flows within a process at different temperature levels, including the minimum heat and cold utility demand of the process. In **Paper I**, energy targeting was performed to quantify and compare the heat recovery potential from the two CO_2 capture technologies, including compression and liquefaction units, for the DH network. In **Paper II–Paper III**, these methods were applied to estimate the heat recovery potential of different decarbonization technologies integrated into the case study plants and thereby estimate the on-site steam production capacity that could offset steam production from existing utility steam boilers.

3.4.3 Combined Exergy-Pinch Analysis

In **Paper II**, a combined exergy-pinch (CEP) analysis adapted from Feng et al. [50] was applied and modeled in MATLAB. The exergy-energy (**Ω**-H) diagram was used to visualize the total exergy losses in individual process equipment. In general, the energy level (**Ω**) of different process equipment units was defined as per Equation 1. Equations for steady-state flow systems, heat transfer, and work-driven equipment can be found in **Paper II**. The total exergy loss (EX_{total}) in each equipment was computed by calculating the area between the respective hot and cold exergy composite curves. The inevitable exergy loss (INE_{EX}) was quantified based on the definition of theoretical minimum exergy destruction required to drive a specific process [58]. The difference between the total and inevitable exergy losses thereby results in the avoidable exergy loss (AVO_{EX}) in each component as per Equation 2. Finally, the exergy efficiency was calculated as per Equation 3, as the ratio of the useful exergy output to the total exergy input for a given process configuration. Alternatively, the exergy efficiency can be calculated from the **Ω**-H diagram as the ratio of the total unshaded region over the total area[13] of the **Ω**-H diagram. Here, the $EX_{tot,area}$ denotes the total exergy losses (shaded region[14], see Figure A3) of all process equipment graphically assembled within the **Ω**-H diagram. The list of assumptions made in the CEP analysis can be found in **Paper II**.

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[13] The product of the lengths of the X-axis and Y-axis in the Ω-H diagram.

[14] The total exergy losses are differentiated as avoidable exergy losses, highlighted with diagonally-hatched lines, and inevitable exergy losses, highlighted with cross-hatched lines, respectively.

$$\text{General definition, } \Omega = \frac{\text{exergy}}{\text{energy}} \tag{1}$$

$$\mathrm{AVO_{EX}} = \mathrm{EX_{total}} - \mathrm{INE_{EX}} \tag{2}$$

$$\eta_{\text{exergy}} = \frac{\text{Useful exergy output}}{\text{Total exergy input}} = 1 - \frac{\mathrm{EX_{tot,area}}}{\text{Total area of } \Omega - \mathrm{H} \text{ diagram}} \tag{3}$$

3.4.4 Cost estimations

Figure 7 illustrates the hybrid[15] top-down/bottom-up capital cost estimation method, adapted from [17,51–53], applied in **Paper II** and **Paper III**. Default economic parameters, assumptions, and cost escalation factors, assumed based on the choice of technology and the context of the industrial case study, can be found in these appended papers. The top-down approach entails extracting data reported in the literature or using vendor data for a whole unit (including all associated equipment), typically reported as engineering procurement and construction costs. This approach was used for equipment for which ample cost data was available in the literature for entire subsystems of mature process technologies. A bottom-up approach involves using energy and material flow data from the developed process models to dimension each piece of equipment. The direct cost of each equipment was obtained from direct cost data or regressed direct cost functions derived from the Aspen Process Economic Analyzer. The regressed direct cost functions presented in Biermann et al. [17] were used for all major equipment sized from the developed process models in **Paper II** and **Paper III**. The total capital requirement (TCR) estimated from the hybrid top-down/bottom-up capital cost estimation method was annualized over the assumed plant lifetime or the design lifetime of the process technology.

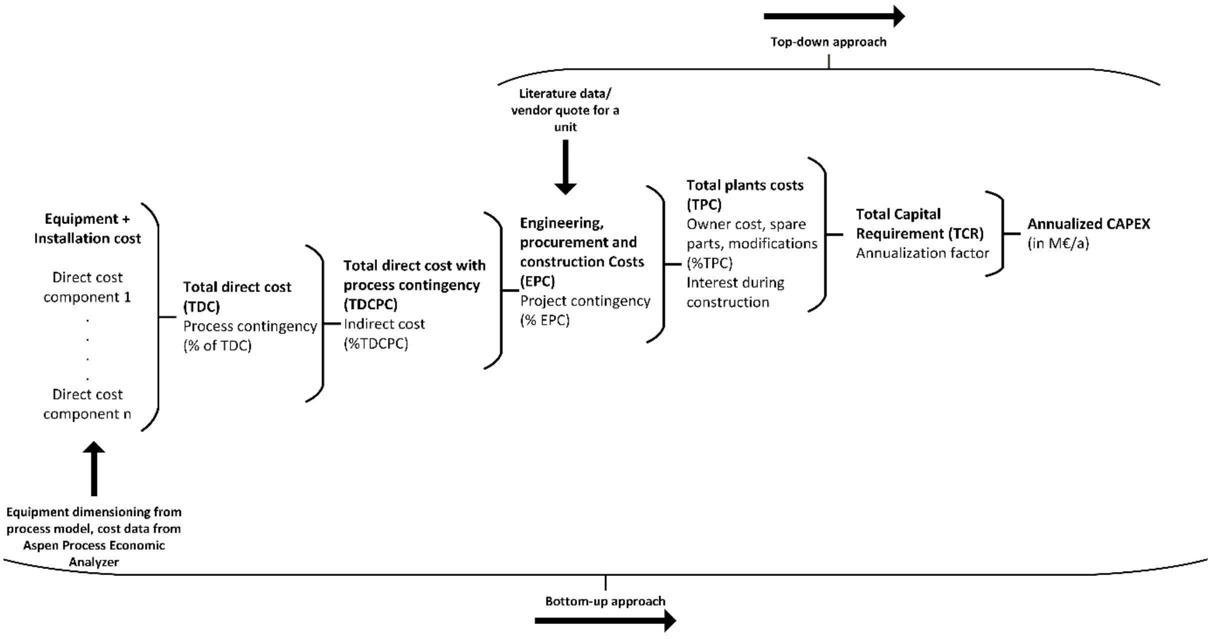

Figure 7: Capital cost estimation method. Arrow marks indicate the direction of cost escalation and input data from the Aspen Process Economic Analyzer. Illustration adapted from [17,51–53].

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[15] Disambiguation: This approach should not be confused with the hybrid costing method, proposed by Roussanaly et al. [11], which combines top-down model of technology learnings with bottom-up engineering-economic analysis for emerging/advanced technologies.

$$\mathrm{CAC} = \frac{\mathrm{CAPEX_{annualized}} + \mathrm{OPEX_{total}}}{\dot{m}_{\mathrm{CO2,avo}}} \left[\frac{€/\mathrm{a}}{\mathrm{tCO_{2,avo}/a}}\right] \tag{4}$$

$$\text{Cost of retrofitability } (\mathrm{C_{COR}}) = \mathrm{C_{OC}} + \mathrm{C_{netw}} + \mathrm{C_{FD}} + \mathrm{C_{PD}} \left[\frac{€}{\mathrm{tCO_{2,avo}}}\right] \tag{5}$$

$$\text{Site} - \text{specific cost of } \mathrm{CO_2} \text{ avoided } \left(\mathrm{CAC_{site-specific}}\right) = \mathrm{CAC} + \mathrm{C_{COR}} \left[\frac{€}{\mathrm{tCO_{2,avo}}}\right] \tag{6}$$

The estimation of CO_2 avoidance cost, shown in Equation 4, served as the key economic indicator for comparing different technological options. It is expressed as the ratio of the total annual costs of the CC and liquefaction plant to the total CO_2 emissions ($\dot{m}_{\mathrm{CO2,avo}}$) avoided annually. The avoided CO_2 emissions account for the indirect CO_2 emissions associated with the energy supply to the CC and liquefaction plant. The total annual cost comprises the annualized CAPEX and the annual operational costs, which include fixed costs (maintenance, insurance, and labor) and variable costs (fuel, electricity consumption, and other consumables).

4 Case study results and discussion

This chapter provides a summary of the main results obtained from applying the developed approaches to the selected case study plants in the appended papers. The chapter is divided into three sections that follow the proposed hybrid assessment framework (see Figure 1), progressing from 1) the influence of system boundaries in the context of BECCS in DH systems; 2) identifying exergy- and cost-optimal process configurations for the PDH plant using the iterative CEP analysis; and 3) the impact of site-specific factors of a steam cracker plant on the cost of decarbonization with different decarbonization technologies.

4.1 The influence of system boundaries – BECCS in DH systems

The choice of CO_2 capture technology from a plant owner's perspective: A detailed comparison of the CO_2 capture technologies comparison can be found in **Paper I**. In general, amine-based CO_2 capture technology using benchmark monoethanolamine solvent is inherently different from the hot-potassium carbonate (HPC) process in that flue gas compression is not required, and heat demand per tonne of captured CO_2 to regenerate the rich-amine solvent[16] is relatively higher. Figure 8 visualizes these technology-specific characteristics, and their impact on plant performance of the reference bio-CHP plant for the two investigated BECCS systems, namely, CHP-MEA and CHP-HPC plants. From an energy perspective, the CHP-HPC plant case incurs an energy penalty[17] of 9 percentage points, while the CHP-MEA plant incurs a significantly larger energy penalty of 15 percentage points. Although the total energy outputs for the CHP-MEA and CHP-HPC plants are estimated to fall within a narrow range of 345 to 366 MW_{th}, these cases exhibit different power-to-heat ratios.

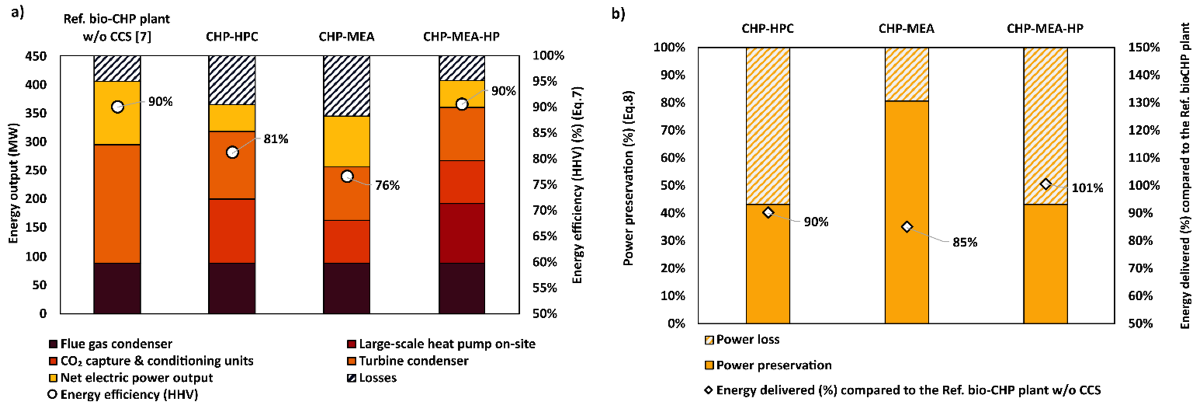

Figure 8: a) Comparison of net electric power outputs, shares of the net heat outputs from different heat recovery units, and energy efficiencies in the different studied BECCS configurations with the reference bio-CHP plant without CCS units. b) Power preservation and energy delivered in the studied BECCS configurations relative to the reference bio-CHP plant without CCS units. The equations indicated in the figure can be found in **Paper I**.

More specifically, considering turbine condensers, flue gas condensers, and recovered heat from the CO_2 capture and compression units, the CHP-HPC plant has the potential to deliver more heat (319 MW_{th}) compared to the CHP-MEA plant (256 MW_{th}) and the reference plant (296 MW_{th}), as shown in Figure 8a. A significantly higher power preservation[18] of 81% was estimated for the CHP-MEA plant, roughly twice that of the electric power output preserved with the CHP-HPC plant (43.2%), as shown in Figure 8b. Using

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[16] Low-pressure steam for driving the MEA capture process is extracted from the turbine at the deaerator stage, while the live steam is extracted from the boiler to drive the flue gas compressor and to supply heat to the HPC capture process. as shown in Figure A1.

[17] The percentage difference in the energy efficiency of the reference CHP plant following the integration of carbon capture and liquefaction processes.

[18] The ratio of the retained (or preserved) electric power output by the CHP plant following the integration of the carbon capture and liquefaction processes.

these energy performance indicators for technology comparison within the plant boundaries indicates that the HPC process is more favorable for a bio-CHP plant operating as a baseload unit in a DH system. This conclusion is especially true since the excess heat recovered at a relatively higher temperature from the flue gas compression could be utilized to increase the plant's DH delivery potential. Therefore, from a plant owner's perspective, the choice of CO_2 capture technology comes down to selecting between the loss of power production with the integration of the HPC capture process and the increased heat losses with the MEA capture process. Considering the pre-existing DH market conditions of the reference bio-CHP plant, where DH is prioritized over electricity production, a compelling argument can be made in favor of HPC capture technology as the optimal choice of decarbonization technology.

Figure 8 presents an additional case, the CHP-MEA-HP plant, considering that the retained electric power output in the CHP-MEA plant, roughly 80% compared to the reference plant (Figure 8b), could be consumed on-site in large-scale heat pumps to increase DH delivery. Here, to ensure a fair comparison, the difference[19] in electric power output between the CHP-MEA and CHP-HPC plants was assumed to be utilized by large-scale centralized heat pumps onsite (COP ~2.5) to recover low-temperature heat within the CHP-MEA plant. In other words, both CHP-MEA-HP and CHP-HPC plants have an equivalent electric power output of 47.5 MW, shown in Figure 8a. Consequently, the integration of large-scale heat pumps resulted in a significantly higher heat output (361 MWth), approximately 21% higher district heat delivery compared to the reference plant. This case highlights that the choice is not between a loss in power production or heat losses as a consequence of CCS integration. Instead, the choice lies in the optimal use of electricity generated on-site, specifically in heat pumps for recovering heat from the newly installed MEA process, instead of using it for flue gas compression in the HPC process. The CHP-MEA-HP solution was determined to be the most effective solution for maximizing heat delivery to the DH system, aligning with the plant owner's perspective.

The choice of CO_2 capture technology from the end-users' perspective: As described in Section 3.3.1, to observe the differences in plant and end-users' perspectives, first, the core functionality of the unabated plant must be retained. In this case, it is district heating, an energy service primarily used by the end-user for space heating and domestic hot water. Figure 8a illustrates the exergy efficiencies of the different BECCS configurations within the plant boundaries. In contrast, Figure 8b illustrates the case where all electric power delivered to the end-user was assumed to be consumed in GSHPs in a decentralized manner, as depicted in Figure 3.

Within the plant boundaries, the CHP-MEA plant expectedly yielded the highest exergy efficiency (approximately 35%), primarily due to its higher power preservation. As a result, the higher exergy efficiency confers this plant configuration with greater product flexibility, with the ability to vary the output load of a specific product by adapting product ratios between heat, power, and negative CO_2 emissions. The bio-CHP plant fitted with the MEA process and large-scale heat pumps, therefore, has greater availability of higher-exergy energy carrier, electricity, that could be strategically used in heat pumps to increase the total DH output (up to 41%) or delivered to the electricity grid. This conclusion highlights the limitation of using energy as a figure of merit for technology comparison within plant boundaries, which previously led to the conclusion that the HPC process could be the optimal decarbonization technology from the plant owner's perspective.

Figure 9b illustrates the estimated system exergy efficiency range, with an assumed Carnot efficiency (η_{Carnot}) in the range of 40%–60% for the GSHPs. The maximum values in the box plots indicate the exergy efficiencies estimated for a DH supply temperature of 86°C. The minimum and median values represent the exergy efficiencies corresponding to typical end-use supply temperatures, in the range of 30°–60°C for space heating and domestic hot water, respectively. The system exergy efficiency is highest when the retained

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[19] Note that, with this assumption, the preservation of electric power is equal (43.2%) for both CHP-HPC and CHP-MEA-HP cases, as shown in Figure 7b.

electric power output is consumed locally in decentralized GSHPs (CHP-MEA), followed by the case where it is consumed on-site in centralized large-scale heat pumps (CHP-MEA-HP), and finally, the CHP-HPC case, which exhibits the lowest system exergy efficiency owing to its lower power-to-heat ratio.

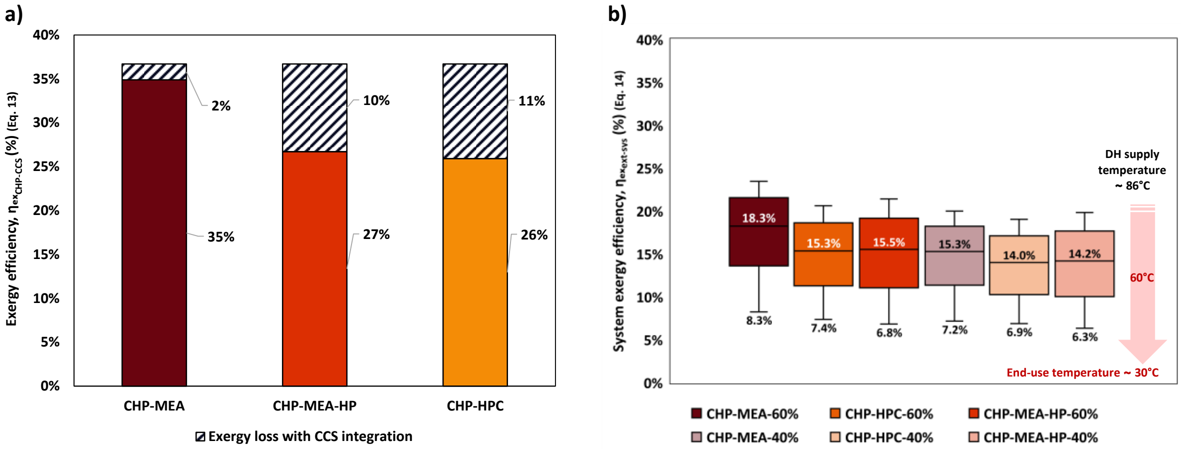

Figure 9: Exergy efficiencies of the CHP-CCS plant cases within their plant boundaries a) and extended system boundaries b), with GSHPs operating with Carnot efficiencies η_{Carnot} in the range of 40%–60%. Source: **Paper I**.

These results show that the retained electric power output in the CHP-MEA could be utilized in power-to-heat technologies, such as GSHPs in the local DH system, for increased levels of DH delivery, thereby avoiding DH distribution losses. Therefore, the CHP-MEA configuration coupled with the uptake of GSHPs in its local DH systems would be the optimal configuration for retaining the highest exergy efficiency in the broader system while achieving the same level of CO_2 reduction as the other BECSS configurations. From the end-users' perspective, this is the optimal configuration, which additionally confers them the option to choose between connecting to the DH network or installing a substitute technology for the same energy service. Additionally, they could optimize their indoor climate demand patterns by complementing their existing heating systems with heat pumps. This example of BECCS in DH systems highlights how plant owners' and end-users' perspectives on the optimal decarbonization technology or plant configuration could diverge, which could be identified using the framework described in Section 3.3.1.

4.2 Targeting minimal exergy losses in connection with net-zero CO_2 emissions in Propane Dehydrogenation Plants

The proposed framework, outlined in Section 3.3.2, was applied to a propane dehydrogenation (PDH) plant, resulting in the process configurations illustrated in Figure 10. Table 2 summarizes the implemented process modifications in each iteration of the CEP analysis and the corresponding influence on the composition of the flue gas at the end of the air-regeneration train. Figure A3 in the Appendix illustrates the exergy-energy diagram obtained for these process configurations. A detailed description of how the process modifications were identified from these figures can be found in **Paper II**.

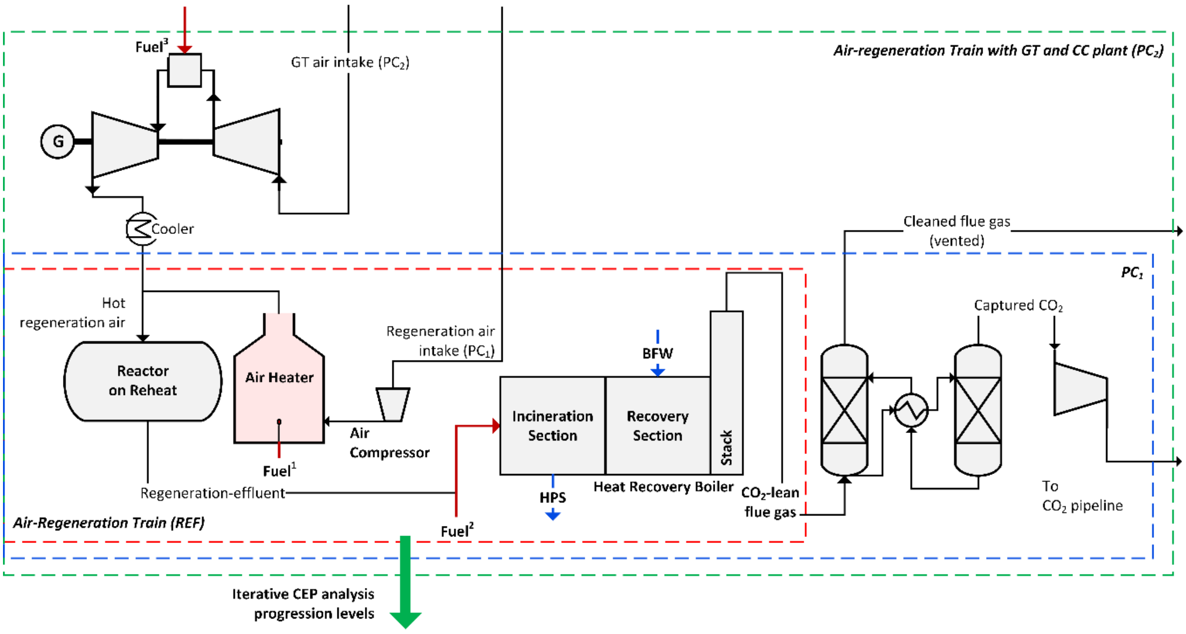

Figure 10: Simplified process flowsheet diagram of the different PDH process configurations and the progression of the CEP analysis. The CEP analysis starts with the reference air-regeneration train model (REF[20], indicated in red dashed box), followed by the PC_1 configuration (blue-dashed box), which includes the CC plant, and finally, the proposed PDH configuration (PC_2, indicated in green dashed box), which includes the industrial gas turbine from which the exhaust gases are used as the hot-regeneration air. In this case, the existing air compressor and air heater function as standby units. [1] The air heater utilizes H_2-rich fuel gas and natural gas at the main burners of the air heater in reference conditions, REF and PC_1, respectively. [2] Volatile organic compounds in the reactor effluent are incinerated with natural gas supplied through the main burners of the heat recovery boiler in all configurations.[3] The gas turbine utilizes natural gas as the primary fuel in the PC_2 configuration. Figure abbreviations: HPS – high-pressure steam, BFW – boiler feed water. Source: **Paper II**.

Figure 11 illustrates the technical and economic performance of the proposed process configuration (PC_2) identified with the framework methodology (Figure 4), incurring minimal exergy losses with retrofitted technologies for capturing CO_2 from the air-regeneration train. Figure 11a shows the exergy efficiency of the decarbonized process configurations (PC_1 and PC_2) related to the CO_2 avoidance achieved with them. Comparison of process configurations with CO_2 avoidance ensures a fair comparison, as the absolute amount of CO_2 generated in the air-regeneration train increases in the proposed configuration, indicated with green vertical lines, compared to the reference plant emissions, indicated with red vertical lines. The CO_2 emissions from the charge heater were included in Figure 11a, indicated with solid vertical lines, to show the CO_2 avoidance achieved in relation to the full scope of on-site CO_2 emissions.

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[20] In the reference air-regeneration train, the air heater utilizes hydrogen-rich fuel gases recovered from the propane dehydrogenation plant (without CO_2 capture plant).

Table 2: Description of implemented process modifications and corresponding process configurations.

Process configurations	Implemented process modification	Corresponding flue gas compositions prior to CO_2 capture	
		Main gas components	Vol.%
PC_0	Straightforward retrofit of CC plant to the reference PDH process[20] without process modifications.	CO_2	2.5
		O_2	13.4
		H_2O	11.2
		N_2 (balanced)	72.9
PC_1	Fuel switched from H_2-rich fuel gas to methane, followed by the retrofit of the amine-based CC plant.	CO_2	4.0
		O_2	12.2
		H_2O	9.0
		N_2 (balanced)	74.8
Proposed process configuration (PC_2)	Integration of industrial GT prior to the air heater. The GT exhaust gas is used as hot regeneration air in the air-regeneration of the PDH process.	CO_2	5.5
		O_2	8.9
		H_2O	10.8
		N_2 (balanced)	74.8

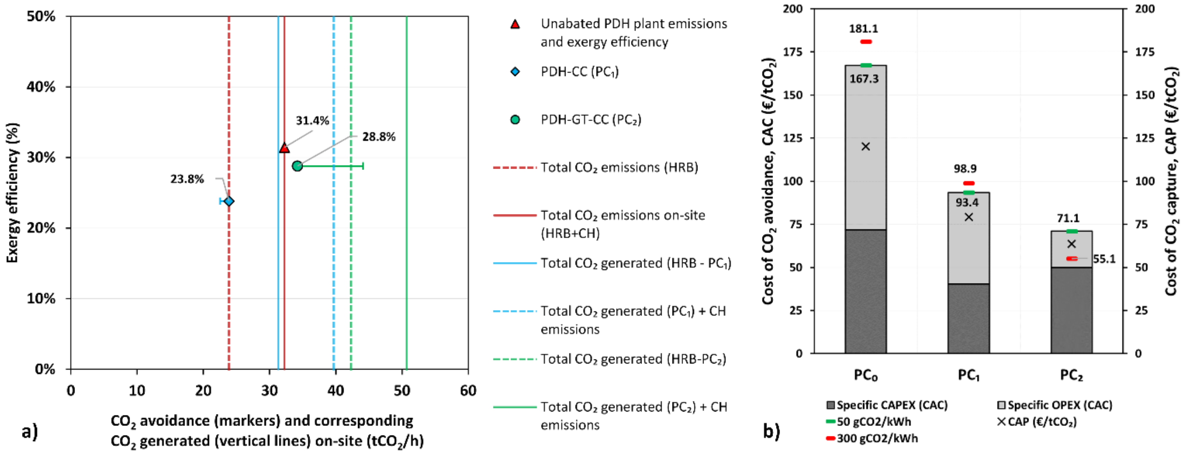

Figure 11: a) Comparison of exergetic performance of the PC_1 and PC_2 plant configurations related to their CO_2 avoidance, assuming a dedicated natural gas boiler to balance the steam demand in the CC plant and a reference grid carbon intensity of 50 gCO_2/kWh. Solid lines indicate CO_2 emissions generated in the air-regeneration train; Dashed lines indicate total CO_2 emissions on-site, including the charge heater emissions. The impact of grid emissions intensity on the overall CO_2 avoided is indicated with horizontal error bars; b) Economic performance of the different decarbonized process configurations of the PDH plant. Stacked bars indicate the cost of CO_2 avoidance (CAC) in terms of specific CAPEX and specific OPEX. The black markers indicate the cost of CO_2 captured (CAP), while green and red markers denote the avoidance cost, corresponding to a specific grid emissions intensity ranging from 50 to 300 gCO_2/kWh, respectively. Figure abbreviations: CC – carbon capture, CH – charge heater, HRB – heat recovery boiler stack, GT – gas turbine. Source: **Paper II**.

It is clear from Figure 11b that the integration of an industrial gas turbine in PC_2 results in greater CO_2 avoidance with a relatively low decrease in the exergy efficiency (28.8%) compared to the unabated air-regeneration train (31.4%). Although the fuel consumption increases slightly in the PC_2 configuration, the shift of fuel combustion from the existing air heater to the combustion chamber of the industrial GT results in greater fuel exergy utilization. This integration results in three favorable outcomes– first, the CO_2 concentration increases substantially prior to the carbon capture plant, which in turn minimizes the exergy loss in the CO_2 capture process. Second, the oxygen concentrations decrease slightly to 9 vol.% in PC_2 configuration (Table 2), which in turn minimizes the risk of capture solvent losses due to degradation at high O_2 concentrations. Finally, the generated electricity will have significantly lower carbon intensity as a result of CO_2 capture at the end of the air-regeneration train. Therefore, the higher CO_2 avoidance in PC_2

configuration results from the low-carbon electricity produced on-site, which can offset grid electricity emissions elsewhere. In Figure 11a, the horizontal positive error bars (green) indicate this potential, where an assumed EU average grid emissions intensity of 300 gCO_2/kWh results in a CO_2 avoidance greater than the total CO_2 emissions from the air-regeneration train in the PC_2 configuration, indicated with the green vertical dashed line. Conversely, higher grid emissions intensity (300 gCO_2/kWh) lowers the CO_2 avoidance achieved in the PC_1 configuration due to power consumption in the CC plant, as indicated by the negative error bar (in blue). Other inferences from Figure 11a can be found in **Paper II**.

Figure 11b illustrates the economic performance of the proposed configuration (PC_2) in comparison to the intermediary process configurations, which involve substantially diluted flue gas streams (Table 2). Expectedly, a significant and near-linear reduction in capture costs was observed in each subsequent process configuration due to increasing CO_2 concentrations and the absolute amount of CO_2 captured on-site (economy of scale). Interestingly, the difference between these process configurations is more pronounced when indirect CO_2 emissions (generated and avoided CO_2 emissions) and their corresponding costs are considered. The highly dilute flue gas stream from the reference PDH plant (PC_0) leads to an estimated avoidance cost of 167–181 €/$tCO_{2,avo}$ where the operational costs dominate due to the higher specific steam consumption. The PC_1 configuration[21] results in an avoidance cost reduction of 24%, therefore implying that the indirect CO_2 emissions from steam generated for the CO_2 captured plant are marginally offset by the slightly lower specific reboiler duty in PC_1 due to increased CO_2 concentrations in the flue gas, and the increased amounts of CO_2 captured on-site.

In contrast, the integration of GT confers a significant economic advantage over its counterpart configurations, owing to the benefits of increased CO_2 concentrations, resulting in lower steam reboiler duty and the generation of low-carbon electricity from the turbine. The impact of grid carbon intensity on the CO_2 avoidance costs is both significant and contrasting compared to PC_0 and PC_1. A typical operation of an amine-based capture plant inherently results in a higher CO_2 avoidance cost than CO_2 capture costs. However, through the integration of the GT, regions with higher grid carbon intensity could achieve lower avoidance costs for the PDH plant (55 €/$tCO_{2,avo}$) compared to the corresponding capture costs (63 €/$tCO_{2,cap}$), as a result of the CO_2 emissions that are offset in the local energy system with the low-carbon electricity generated from the PC_2 configuration. The proposed configuration achieves an overall reduction of CO_2 avoidance cost by 58–70% (55–71 €/$tCO_{2,avo}$) compared to the PC_0 configuration (167–181 €/$tCO_{2,avo}$), as identified through the developed framework applied in **Paper II**, which incorporates iterative CEP analysis. The potential and implications of the proposed PDH process can be found in **Paper II**.

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[21] Here, the fuel gas price was assumed to be equivalent to the assumed average natural gas prices (6 €/GJ). Therefore, the price difference between the fuel gas and the alternate hydrocarbon fuel (with H/C <4) would ultimately determine the cost reduction attainable with the fuel switch in PC_1 configuration.

4.3 The impact of site-specific factors on CO_2 avoidance costs – Steam Cracker Plant

Figure 12 shows the baseline avoidance cost estimates from the conventional cost estimation method (Figure 7), indicated with grey bars, to which site-specific cost factors (indicated with red solid floating bars), together representing the total cost of retrofitability, are added to obtain the site-specific cost of avoidance for the Post-CCS (blue) and Pre-CCS (green) pathways considered for the steam cracker plant. These cost escalations are estimated with the set of baseline site-specific assumptions (Table 6 in **Paper III**) to illustrate the differences at a conservative level. However, the full scope of expected cost escalation beyond the baseline site-specific assumptions is estimated with sensitivity analyses on each of these factors and their influence on the site-specific cost of CO_2 avoidance, which can be found in **Paper III**.

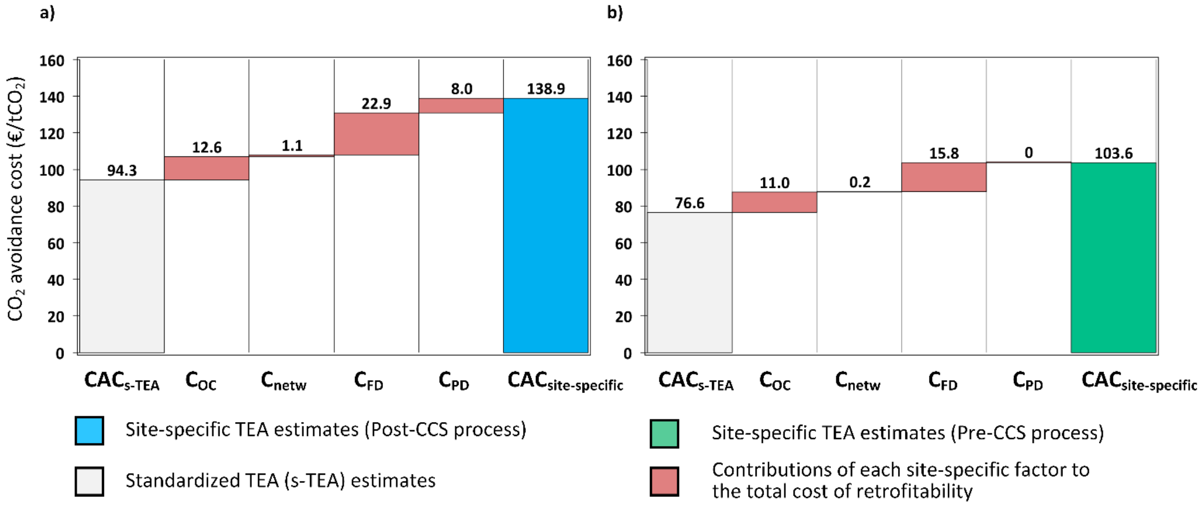

Figure 12: Site-specific cost of CO_2 avoidance for (a) Post-CCS and (b) Pre-CCS pathways. Note that the average value of opportunity costs (estimated for different space-value functions) was taken here, with the baseline assumptions listed in Table 6 in Paper III. Figure abbreviations: s-TEA – standardized TEA, C_{OC} – Opportunity cost, C_{netw} – CO_2 network costs, C_{FD} – Forced downtime costs, C_{PD} – Premature decommissioning costs due to technology lock-in effect. Source: **Paper III**.

The forced downtime has by far the largest impact on the site-specific CO_2 avoidance cost, followed by the opportunity costs of space available on-site and the technology-specific lock-in effect in relation to the residual lifetime of the case study plant. Accounting for avoided CO_2 emissions, the cost difference between the two decarbonization alternatives widens significantly, as shown in Figure 12a–b. The site-specific avoidance costs increase by roughly 47% and 35% for Post-CCS and Pre-CCS (139 €/$tCO_{2,avo}$ and 104 €/$tCO_{2,avo}$) from their respective baseline CO_2 avoidance cost estimates of 94 €/$tCO_{2,avo}$ and 76 €/$tCO_{2,avo}$. This difference is primarily due to technology-specific characteristics of the Pre-CCS pathway, which utilizes relatively smaller space on-site and remains unaffected by the lock-in effect, as the newly installed process equipment can be run as a standalone hydrogen production plant utilizing natural gas from the grid. Although higher installation complexity[22] and a higher probability of forced downtime can be expected for the Pre-CCS pathway, the incurred cost escalation remains lower than the estimated C_{FD} for the Post-CCS pathway owing to the significantly higher CO_2 avoidance achieved with the Pre-CCS option. With the inclusion of site-specific factors, the relative differences in CO_2 avoidance cost estimates between the two

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[22] For revamping existing fuel gas system to be compatible with pure hydrogen and replacing fuel gas burners to compatible burners for hydrogen firing in the existing cracker furnaces.

decarbonization options increase from approximately 20% with conventional TEA methods to 29% with the site-specific TEA method.

Figure 13 illustrates the diagnostic diagram that combines the quantitative site-specific results with qualitative assessment for site-specific factors to obtain an enhanced indication of the optimal decarbonization technology for the case study plant. Based on expert elicitation, qualitative factors such as flexibility to adapt to future feedstock switches, resource availability, alternative use of CO_2 capture equipment, the possibility of achieving 100% carbon recovery towards CO_2 utilization pathways, cost of retrofitability in terms of CO_2 avoidance, spatial footprint, and lower sensitivity to fuel prices generally favor the Pre-CCS process. In contrast, factors such as system integration complexity, sensitivity to electricity prices, dependence on external energy supply, and reliance on external infrastructure favor Post-CCS.

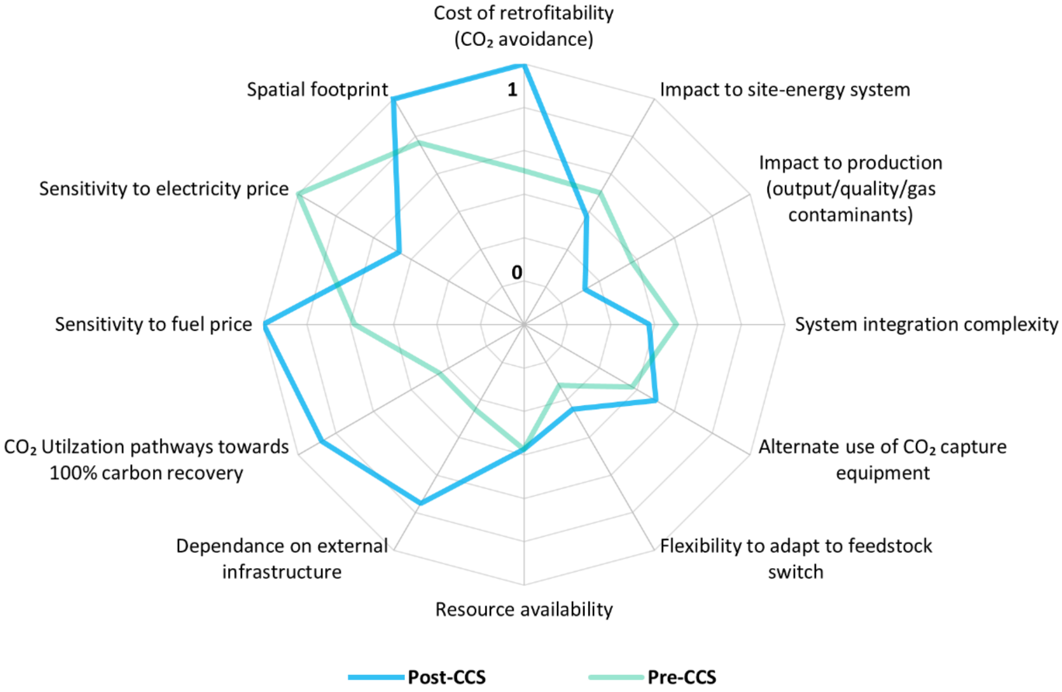

Figure 13: Diagnostic diagram combining quantitative site-specific factors (cost of retrofitability, spatial footprint, and sensitivity to fuel and electricity prices) with the results of the qualitative retrofitability assessment obtained through expert elicitation for the steam cracker plant. Averaged values of expert elicitation on qualitative factors are plotted. Source: **Paper III**.

Utilizing the diagnostic diagram, the Pre-CCS option, with an average score of 0.47 (showing a lower spread on the diagnostic diagram), was determined to be the preferable decarbonization alternative compared to the Post-CCS pathway, which has an average score of 0.59, implying relatively higher risks due to technical and economic uncertainty.

5 Conclusion

This thesis presented a hybrid assessment framework that combines a set of methodological frameworks that address the limitations of existing process integration and techno-economic assessment methods, thereby obtaining an enhanced early-stage indication of the optimal decarbonization solution for specific process plants, depending on its site-level, process-level, and systems-level contexts. The focus of this thesis was on bridging the gap between the early-stage assessment methods in identifying promising exergy- and cost-optimal decarbonization solutions that facilitate decision-making toward near-term implementation.

At the systems level, the potential inconsistency between the plant owner and the end-user was addressed using extended system boundaries with exergy as a figure of merit for comparing the impact of integrating different CO_2 capture technologies with inherently different exergy requirements (**Paper I**). At the plant level, the limitations of pinch-based heat flow analysis in identifying promising modifications in electrically driven units were addressed using an iterative combined exergy pinch analysis. The developed framework enabled the identification of process modifications in the existing plant as well as the retrofitted process technology with the objective of determining decarbonized process configurations that incur minimal exergy losses and the lowest CO_2 avoidance costs (**Paper II**). At the site level, the concept of site-specific techno-economic analysis, incorporating site-specific factors, was introduced. The developed framework unravels the site and technology-specific attributes expected to influence the final cost of decarbonization and thereby enhance the comparative assessment of decarbonization technologies for a specific industrial site (**Paper III**). These methodological frameworks were demonstrated through case studies of decarbonization in a bio-CHP plant operating in a district heating system (**Paper I**), a propane dehydrogenation plant (**Paper II**), and a steam cracker plant (**Paper III**).

In general, the case study results in **Paper I** highlight the advantage of choosing CO_2 capture technology that preserves the electric power production at future BECCS plants, which inherently offers greater product flexibility (i.e., the ability of the plant to vary a specific product output) and higher exergetic efficiency considering its local DH system. The integration of an amine-based CO_2 capture technology retained approximately 80% of the electric power output in the unabated bio-CHP plant. Complementing this BECCS configuration with large-scale heat pumps on-site could enable an increase in district heating output by 21% compared to the reference bio-CHP plant. In **Paper II**, the framework reveals how unconventional process modifications and integration measures in existing unabated propane dehydrogenation plants could lead to a substantial reduction in CO_2 avoidance cost (58–70%) compared to retrofitting the plant with end-of-pipe CO_2 capture technology (167–181 €/tCO_2).

In **Paper III**, the site-specific techno-economic analysis applied to the steam cracker plant highlights that the impact on current and potential future plant production has the highest impact on the total cost of retrofitability, followed by the opportunity cost of occupying valuable space available at existing sites. Considering site-specific factors of the reference steam cracker plant, the CO_2 avoidance cost is shown to be roughly 46% and 36% higher than the baseline estimates for the Post-CCS and Pre-CCS processes, respectively, of conventional bottom-up techno-economic assessment methods. Furthermore, the cost of retrofitability or the escalation of CO_2 avoidance costs due to site-specific factors is approximately 80% higher for the Post-CCS decarbonization process (43 €/tCO_2) than for the Pre-CCS process (24 €/tCO_2), highlighting the varied impact of these factors on different decarbonization technologies. The qualitative retrofitability assessment revealed pertinent site-specific factors such as sufficient access to external infrastructure such as the electricity grid and CO_2 supply infrastructure, the flexibility to adapt to future feedstock switches, the importance of decoupling the decarbonization technology and the process plant's lifetime, and the possibility of reaching 100% carbon recovery towards CO_2 utilization pathways as factors that could tip the favor from one decarbonization technology to another.

In summary, the proposed hybrid assessment framework, which integrates the individual frameworks developed in this thesis, can be utilized to acquire a comprehensive early-stage indication of the optimal decarbonization technological pathway. At the systems level, it is imperative to consider end-users' perspectives to avoid the selection of sub-optimal decarbonization technology at process plants and, in general, to achieve exergy-efficient industrial systems. Similarly, at the process level, it is essential to consider revamping and modifying exergy-destructive plant operations, such as fuel combustion, within existing plants. This approach involves iteratively conducting process modifications in both the decarbonization technology and the host process plant to enable achieving net-zero emissions at the lowest possible CO_2 avoidance cost when comparing various decarbonization alternatives.

Finally, site-level considerations offer the possibility of obtaining enhanced cost estimates for different process industry sites. These cost estimates not only provide insights into the hidden costs of decarbonization, often not considered in academic or advanced cost-engineering studies, but also facilitate informed technology selection. Moreover, integrating these enhanced site-specific cost estimates with the technology-specific cost of retrofitability into energy-systems level studies can improve the level of detail translated to higher-level analyses, such as national-level marginal abatement cost curves, and the cascading cost of decarbonization pathways on the final consumers, providing critical information for policymakers in this domain.

6 Future work

The hybrid assessment framework presented in this study provides a basis for conducting future comparative decarbonization studies that incorporate system, plant, and site-level considerations relevant to most carbon-intensive industries. This framework is built upon a combination of methodologies demonstrated with individual industrial case studies, chosen to best highlight the limitations in existing process integration and techno-economic methods.

Future efforts should prioritize implementing the proposed hybrid assessment framework (refer to Figure 2) across various industrial sectors. This will enable the observation of site-specific differences within and between industries and potentially differing perspectives between the plant owner and the end-users. Such an application is crucial for validating the relevance of the proposed approach for other industries and facilitating the identification of optimal decarbonization technological pathways suitable for implementation at the studied industrial site. Furthermore, applying this framework to a wide range of industrial case studies is necessary to identify limitations and uncertainties inherent to both the studied industry and the proposed approach. These limitations should then be addressed in future work to enhance the robustness of the proposed framework.

Some of the limitations concerning the developed methods, tools, and case study plants are discussed in the appended papers. In the context of this thesis, the following paragraph elucidates identified limitations at the respective evaluation levels of the proposed framework. It begins with site-level considerations and then zooms out into systems-level considerations.

Site-level: The site-specific techno-economic assessment method primarily prioritized mature CO_2 capture technologies, considering their lower uncertainties regarding their technical performance and cost structures compared to emerging process technologies. This approach allows for comparing technologies that are available for immediate deployment within the limited timeframe aimed at achieving net-zero CO_2 emissions. However, the optimal indication of the decarbonization pathway for any industrial site remains contingent on the number of decarbonization options compared in the assessment. The assessment would be otherwise incomplete without assessing the potential for emerging process technology that could achieve the same decarbonization target at a particular site.

Therefore, the proposed methodology could be further advanced by incorporating emerging technologies into the set of technologies compared within the framework. The inclusion of emerging technologies should be guided by a hybrid-costing method, integrating engineering-economic and experience curve methods for advanced technologies, as proposed by Roussanaly et al. [11]. This method accounts for future cost trajectories based on the maturity level and anticipated learning rates of emerging technologies. It is, however, important to ensure that accurate technology-specific attributes, such as physical spatial requirements, are available or estimated with reasonable accuracy for the emerging process technology.

In addition to these considerations, meticulous data curation of site-specific attributes is warranted to better understand the similarities and differences among sites, which are typically heterogeneous within the same industry category. This comprehensive approach enables the formulation of well-informed site-specific assumptions applicable to most process plants within the same industry category, thereby simplifying the assessment to some extent. Moreover, expanding the scope of the decarbonization options should be explored, with a focus on including measures such as feedstock-switch that could complement selected decarbonization technology in achieving relatively higher CO_2 avoidance. An example of how the optimal indication for the steam cracker plant could potentially change with a feedstock switch from steam cracking of naphtha to ethane is described in **Paper III**.

Plant-level: Decarbonization measures, such as direct process electrification of reactors or core-production units, are likely to affect the material and heat flow at a specific site. The methods developed in this work were demonstrated with industrial case studies, where no changes to the current consumption of feedstock or demand for products were assumed. Take, for example, the electrification of an existing steam cracker, which would increase the availability of fuel gas within the steam cracker plant that would need to be repurposed as a feedstock or a co-product. Furthermore, it could directly impact the available excess heat within the plant, as demonstrated in a case study by Wiertzema et al. [59] on an oxo-synthesis plant with electrified syngas production. Therefore, the consequence of direct process electrification in terms of changes to heat and material flows at existing sites must be accounted for in the proposed assessment framework.

Additionally, existing single-product industries are likely to transition into more complex multi-product industries with excess heat, repurposed fuel gas, and captured CO_2 as co-products along with their primary products. Therefore, the indication of optimal decarbonization technology from the assessment framework must be complemented with plant-level cost-optimization that considers the process technologies' ability to respond to uncertain energy market conditions and alternatively identify complementary process technologies that provide flexibility to plant operation. For example, **Paper I** highlighted the role of centralized large-scale heat pumps as a complementary process technology that could confer product (heat, power, and negative CO_2 emissions) flexibility to future BECCS plants in the DH system in response to fluctuating market conditions or demand levels. In this context, it will be essential to consider technology-specific attributes such as operational capacities and ramping rates. These considerations will help observe differences between various process technologies, particularly in their ability to respond to volatile electricity markets. Finally, the proposed framework could be expanded to evaluate the potential for integrating promising carbon dioxide removal technology[23] at industrial sites. This integration would enable the possibility of mitigating the residual industrial CO_2 emissions by leveraging the synergetic effects of co-location at large industrial clusters that typically offer large amounts of industrial waste heat, sufficient grid capacity, and potentially shared infrastructure such as a CO_2 purification and liquefaction plants, or CO_2 utilization plants, and transportation infrastructure such as CO_2 pipelines or access to port for ship transportation.

Systems-level: The case study in **Paper I** demonstrated how underlying market conditions in a DH system could potentially favor sub-optimal decarbonization solutions at the plant level. Although these disparities may be inconsequential in some industrial contexts with limited interaction with local energy systems, future efforts could still be focused on identifying similar disparities between plant and systems-level perspectives on identifying competing substitute technologies that provide the same energy services or products, contingent on either incumbent or proposed policies within this domain. Here, using exergy as a figure of merit is recommended to ensure a fair comparison between different decarbonization approaches. Finally, the proposed framework could be complemented with deep uncertainty[24] methods, applied at the plant or system level, to enable crafting robust and adaptive strategies, considering a broad spectrum of uncertainties that might impede the transition toward sustainable industrial decarbonization.

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[23] More specifically, adsorption-based direct air capture technologies that require low-grade waste heat at around 80–100°C (1600 kWh/tCO_2) and electricity (400 kWh/tCO_2) [63].

[24] Stenström et al. [64] applied Decision-Making under Deep Uncertainty (DMDU) methods to demonstrate how robustness against uncertainty could support investment decisions in BECCS deployment.

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Nomenclature

Abbreviations

AVO	Avoidable
Bio-CHP	Biomass-fired Combined Heat and Power plant
CAC	Cost of CO_2 avoided
CAP	Cost of CO_2 capture
CAPEX	Capital expenditure
CBAM	Carbon Border Adjustment Mechanism
CC	Carbon Capture
CCS	Carbon Capture and Storage
CEP	Combined Exergy-pinch
COR	Cost Of Retrofitability
DH	District Heating
EU-ETS	EU Emissions Trading Scheme
FEED	Front-End Engineering Design
GCC	Grand Composite Curve
GSHP	Ground Source Heat Pump
GT	Gas Turbine
HPC	Hot Potassium Carbonate
MEA	Monoethanolamine
NG	Natural Gas
NGCC	Natural Gas Combined Cycle power plant
PC	Process Configurations
PDH	Propane Dehydrogenation
Post-CCS	Post-combustion Carbon Capture and Storage
Pre-CCS	Pre-combustion Carbon Capture and Storage
s-TEA	Standardized Techno-Economic Analysis
TCR	Total Capital Requirement
TEA	Techno-Economic Analysis
TRL	Technology Readiness Levels

Symbols

AVO_{EX}	Avoidable exergy loss
INE_{EX}	Inevitable exergy loss
EX_{total}	Total exergy loss
Ω	Energy level (ratio of exergy and energy)
H	Energy input
C_{OC}	Opportunity costs
C_{netw}	Cost of all CO_2 interconnections within plant boundaries
C_{FD}	Cost of forced downtime
C_{PD}	Cost of premature decommissioning

Subscripts

avo	avoided
reb	reboiler temperature
source	heat source
n	number indicating the number of implemented process modifications
ss-cap	site-specific cost of CO_2 capture
ss-avo	site-specific cost of CO_2 avoidance

Appendix A. Reference process plants

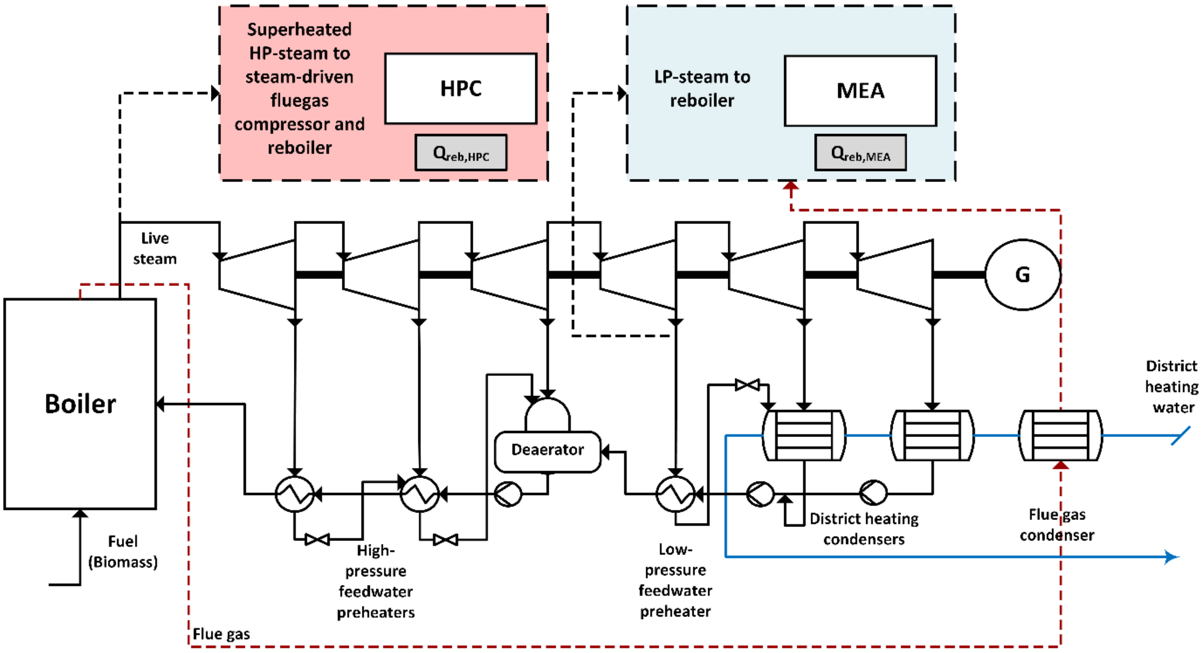

Figure A1: Process schematic of the CHP steam cycle modeled for the reference plant, adapted from Beiron et al. [3]. Note that only one of the two CCS units is considered when evaluating the CHP-MEA or CHP-HPC case. Note also that the flue gas condenser (shown in Fig. 2) is now placed outside the CCS unit blocks to represent the CHP steam cycle more accurately. Black dashed lines – steam extracted from the steam cycle to drive the corresponding CCS unit; Red dashed lines – flue gases from the boiler; Gray boxes – input data to the CHP steam cycle model. Source: **Paper I**.

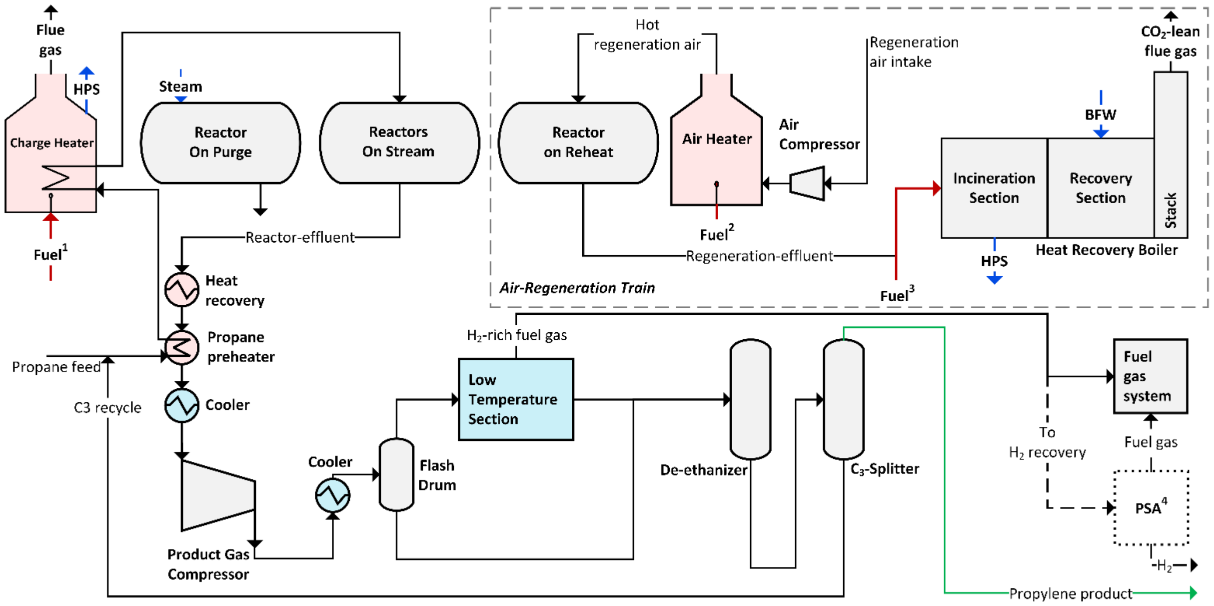

Figure A2: Simplified process flowsheet diagram of the propane dehydrogenation plant. The grey-dashed box indicates the system boundary of the air-regeneration train. Fuel inputs: [1] C_{4+}-liquid hydrocarbons and H_2-rich fuel gas. [2] H_2-rich fuel gas and natural gas at the main and pilot burners, respectively. [3] Volatile organic compounds in the reactor effluent are incinerated with natural gas supplied through the main burners. [4] Optional pressure-swing adsorption unit for hydrogen recovery; currently not in place at the reference plant. Source: **Paper II**.

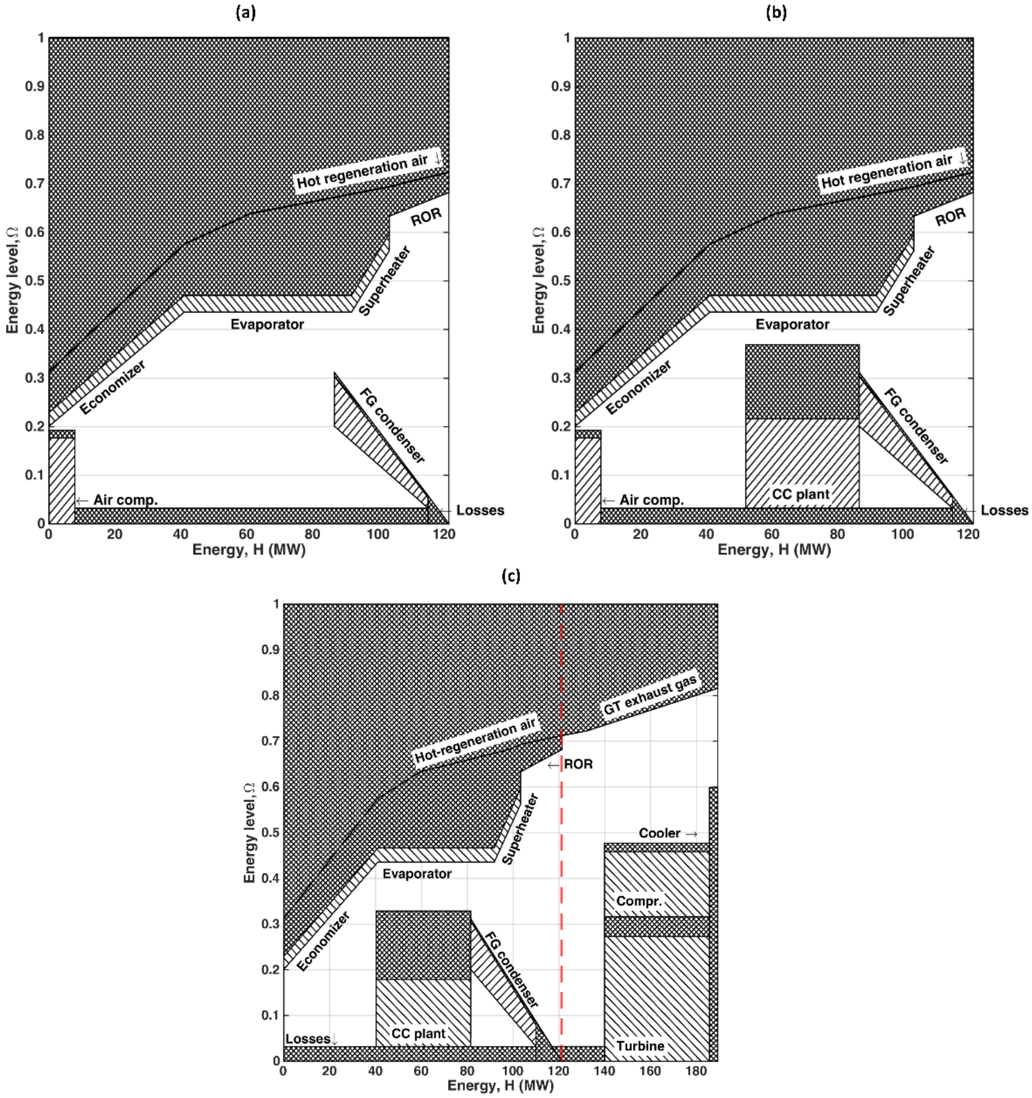

Figure A3: Ω-H diagram of the a) the reference air-regeneration train without the CC plant, b) PC$_1$ configuration and c) PC$_2$ configuration. Diagonal and cross-hatched regions indicate the avoidable and inevitable exergy losses, respectively. Note the increase in the X-axis scale in PC$_2$ configuration, with the integration of the industrial gas turbine. The red-dashed vertical line indicates the total exergy input to the previous process configurations, as depicted by the X-axis scales in sub-plots (a) and (b), prior to this modification. Source: **Paper II**.

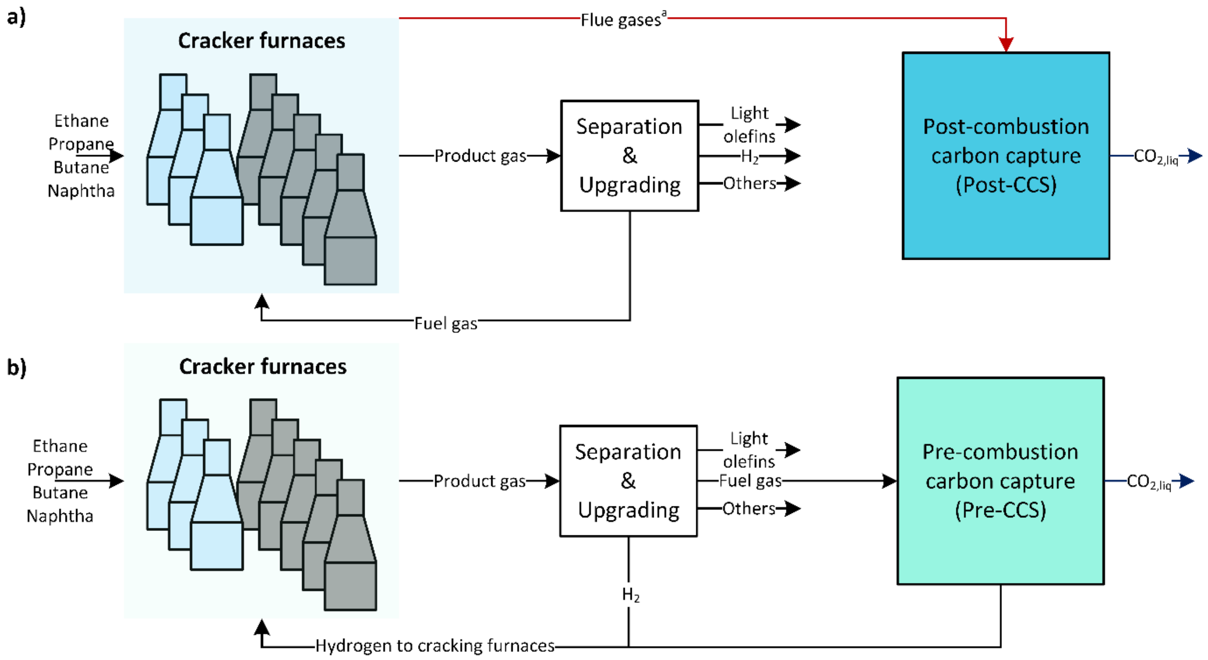

Figure A4: Overview of the main material flows in a steam cracker plant with (a) post-combustion (Post-CCS) and (b) pre-combustion (Pre-CCS) decarbonization pathways. [a] Flue gases include total plant CO_2 emissions. In Pre-CCS, CO_2 emissions from furnaces are entirely avoided; however, CO_2 emissions from NG-fired steam boilers remain (not shown in the Figure). Note that the output streams for both pathways are liquified CO_2 at the reported liquid CO_2 transport specification in the Northern Lights project (-26.5°C, 15 barg [60,61]). Source: **Paper III**.