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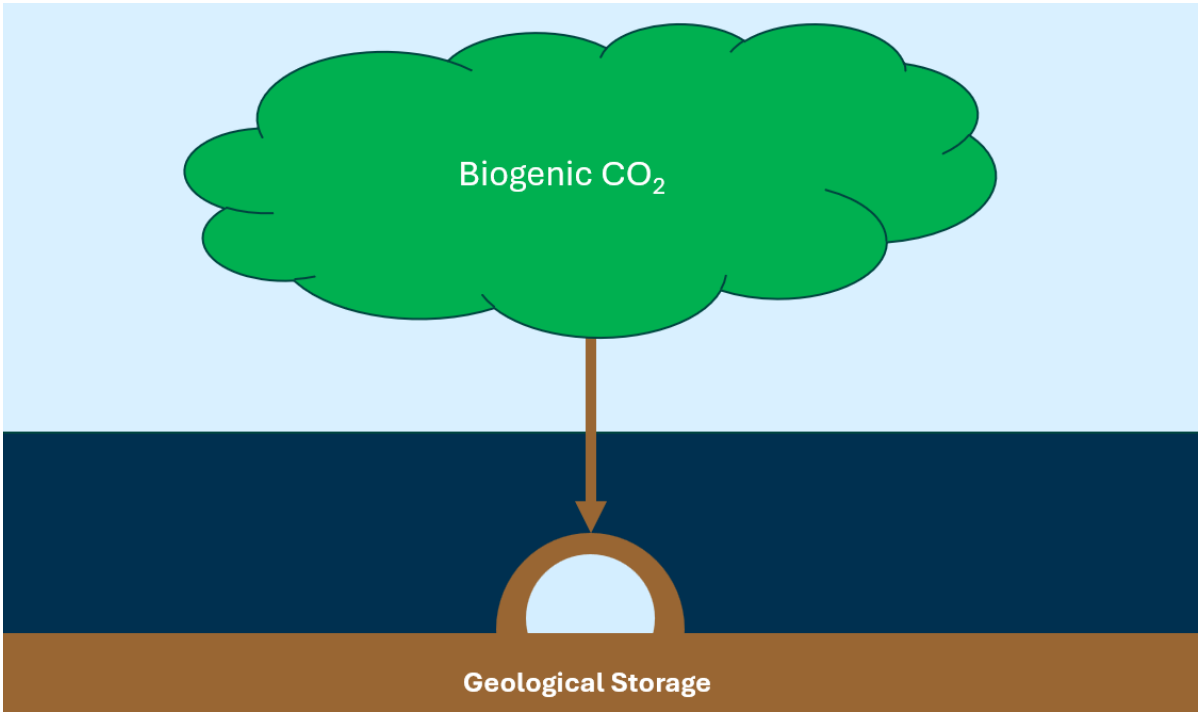

Techno-economic assessment of carbon capture for the pulp and paper industry

Master's thesis in Sustainable Energy Systems

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CHALMERS UNIVERSITY OF TECHNOLOGY
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MASTER'S THESIS 2024

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Abstract

The cost of capturing CO_2 in a recycled paper mill and a pulp mill was investigated by performing a techno-economic analysis. Two different post-combustion capture (PCC) technologies were explored: amine-based process (benchmark) and a carbonate-based process (novel). What sets the novel process apart from the benchmark process is its lower-regeneration temperature, allowing the possibility of meeting part of the reboiler heat using low-temperature excess process heat. In the Pulp Mill, the heat integration with both capture technologies was evaluated using pinch-analysis tools. It was found that the heat demand for capturing CO_2 in the Paper Mill could be met by a steam extraction, whereas in the Pulp Mill, the high-temperature excess heat from the recovery boilers was sufficient to meet the heat demand for capture. However, without additional fuel use, integrating carbon capture in this way would result in a loss in electricity generation for both the Paper and Pulp Mill.

Avoiding losses in electricity generation in the Pulp Mill was explored, leading to the development of several heat integration scenarios. This was done by optimizing the size of a simplified steam cycle to maximize electricity generation using existing or investing in extended back-pressure steam turbine capacity. Comparing the capture cost between the Paper Mill and Pulp Mill confirmed that site-specific factors such as the CO_2 flow (and its CO_2 concentration) and geographical location have a strong influence on the capture cost. With limited opportunity for heat recovery in the Paper Mill, comparing the capture cost of the novel process between the two sites indicated that excess heat availability is an important parameter for achieving a lower-capture cost. As pulp mills in the future may differ due to strategic developments in biomass resource utilization, the capture cost of a Pulp Mill with lignin extraction was also evaluated. This was found to be higher than a Pulp Mill without lignin extraction, indicating that carbon capture is clearly benefitted by favourable mill energy balances.

A sensitivity analysis was performed on the capture cost of the Pulp mill to evaluate the influence of different energy market conditions. In a low electricity price market, consuming more fuel to prioritize co-generation of back-pressure electricity did not result in a significantly higher capture cost compared to accepting losses in electricity generation. In a high electricity price market, investing in extended back-pressure steam turbine capacity and therefore producing more electricity than the Pulp mill without carbon capture achieved the lowest capture cost.

A bottom-engineering economic approach was used for estimating the CAPEX of the capture technologies where “Nth-of-kind” (NOAK) cost factors were utilized, assuming the retrofit will occur in a future where carbon capture integration with the pulp and paper industry has reached greater maturity. As carbon capture has yet to be demonstrated in the pulp and paper industry, the NOAK cost estimates presented in this work include significant uncertainties.

Key words: Pulp and paper industry, post-combustion carbon capture, heat integration, techno-economic assessment

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Eliette Lacaze-Masmonteil, Gothenburg, August 2024

"*And when you are surrounded by mysteries, as the saying goes, it's best to live life in the form of a question*" – David Brooks

Contents

Nomenclature

Below is a list of abbreviations and acronyms that have been used throughout this thesis, listed in alphabetical order:

AMP/PZ	AMP (2-amino-2-methyl-propanol) -PZ (piperazine)
APEA	Aspen Process Economizer Analyzer
BFB	Bubbling fluidized bed
Bio-CCS	Biogenic carbon capture and storage
BLG	Black liquor gasification
CAPEX	Capital expenditures
CCS	Carbon capture and storage
CDR	Carbon dioxide removal
CHP	Combined heat and power
DH	District heating
EPC	Equipment, procurement, and construction
FOAK	First-of-a-kind
GCC	Grand composite curve
HDC	Heat demand constrained (Scenario C)
MEA	Mono ethanol amine
NOAK	N^{th}-of-a-kind
O&M	Operational & maintenance cost
OPEX	Operational expenditures
PCC	Post combustion capture
RDF	Refused derived fuel
SRD	Specific reboiler duty
TC	Turbine constrained (Scenario B)
TCR	Total capital requirements
TDC	Total direct cost
TEA	Techno-economic assessment
TPC	Total plant cost
TRL	Technology readiness level

List of Tables

List of Figures

1 Introduction

1.1 Background

It is well known that the world needs to achieve ambitious levels of decarbonisation in the years to come to prevent catastrophic consequences from global warming. Responsible for 28% of the global CO_2 emissions in 2022 [1], one of the most challenging sectors to decarbonize is the industrial sector. While biogenic carbon capture and storage (Bio-CCS) has been identified as one solution for the decarbonization of industry in Europe, the cost of capturing carbon dioxide remains one the main barriers preventing its large-scale deployment. In the case of bio-CCS, other barriers identified by potential bio-CCS actors in Sweden and Finland are an absence of reliable long-term policy and the uncertainty of what a future negative emissions market will look like [2]. Moreover, there also exist uncertainty in future availability of biomass for such large-scale applications. These factors contribute to a high investment risk and thus hesitance to be an early adopter. It should also be noted that social acceptance may also be a barrier for both CCS and bio-CCS, but this varies from country to country. Actors in Sweden and Finland did not identify social acceptance as one of the main barriers to carbon capture [2], unlike France and Poland [3].

When it comes to estimating the cost of CCS for industry, there is a lack of alignment in the cost estimates reported within industrial sectors. This is partly due to the differences between each industrial site studied, such as excess heat availability, the concentration of carbon dioxide and other contaminants in the flue gas, the capture rate, and the size of the flue gas flow. Site differences aside, discrepancies can also emerge due to differences in methodological framework, cost metric definitions, input data quality, choice of economic parameters (plant lifetime or discount rate for example), assumptions in technology maturity (first-of-a-kind (FOAK) vs. N^{th}-of-a-kind (NOAK) cost estimates) and the system boundaries i.e. whether transportation and long-term storage is included or not [4] .

Using the cement industry as an example to further the point, one study in 2012 reported an avoidance cost of 40.6 \$/t CO_2 with 60 % of emissions captured [5]. In that same year, another study reported a cost of 23 \$/t CO_2 with 84% of emissions captured [6]. Both use oxy-fuel combustion with calcium looping as the capture technology. The wide range of these cost estimates introduces a large degree of uncertainty [7], making it difficult for decision-makers to make informed decisions regarding future CCS investments.

Despite the uncertainties of Bio-CCS, what is certain is that without the deliberate removal of carbon dioxide from the atmosphere, the world will not limit global warming by 1.5°C (with no or limited overshoot) [8]. As CO_2 emissions continue to rise, it is likely that all the 2°C pathways will share the same fate. Otherwise put, the world needs technological carbon dioxide removal (CDR) methods such as Bio-CCS and DACCS (Direct Air Carbon Capture and Storage) to meet its climate targets, with bio-CCS being one of the most mature options available.

A promising candidate for bio-CCS is the pulp and paper industry, as especially kraft pulp and paper mills have large point sources of biogenic CO_2 emissions. In addition, the concentration of CO_2 of the flue gas emitted is relatively high (13-20%), resulting in a lower capture cost in the case of post-combustion capture (PCC) [9]. The global capture potential from kraft pulp mills has been estimated at 137 Mt CO_2 /a [10]. In Europe, among 30 countries, the bio-CCS potential for pulp and paper plants is estimated at 62 (±5) Mt/a [11]. For reference, all pathways that limit global warming to 1.5°C with limited or no overshoot feature Bio-CCS deployment of up to 1000, 8000, and 16 000 MtCO_2 per year in 2030, 2050, and 2100, respectively [8]. The corresponding deployment for limiting warming to 2°C requires 80, 2750 and 8960 MtCO_2/yr. for these years, respectively [12]. It is worth noting here that dependence on Bio-CCS for CDR will only grow with further delay in deep emissions reduction, such that what is needed will likely go beyond technical and sustainable limits, emphasizing that Bio-CCS should not be regarded as the only solution to meet the climate targets.

What may affect the future potential for carbon capture in pulp and paper mills is the near-term implementation of methods for better usage of the biogenic carbon content of the feedstock. For example, lignin extraction where lignin can be used as feedstock for producing products with a long lifetime [13] could be one such option. Although this would not achieve the same levels of (biogenic) CO_2 reduction as carbon capture, the products made using lignin can displace fossil-based products [14], providing indirect reductions of fossil CO_2 emissions. The main and important difference between utilizing carbon in products via lignin extraction versus Bio-CCS is that Bio-CCS can ensure permanent long-term storage and thereby contribute to carbon dioxide removal (CDR) from the atmosphere (negative emissions), while lignin can contribute to emissions reductions through substitution effects.

A few studies have explored the implementation of carbon capture in pulp and paper mills where the choice of capture technology is typically amine-based post-combustion capture (PCC) [15], [16]. PCC is often the main choice not only because of its maturity but also because it is better suited for retrofits. The main drawback of this technology is that the temperature for solvent regeneration can range between 110-130°C [17], depending on the solvent. In the case of the pulp and paper industry, there is little opportunity to meet this heat demand using low-grade residual heat since most residual heat in such plants is only available below 100°C [18]. It is therefore of interest to explore whether a lower temperature PCC technology is a more economically attractive option compared to the amine-based technology.

Research efforts are therefore needed to identify the economic feasibility of integrating Bio-CCS with the pulp and paper industry. Moreover, there is a need to understand how different mill characteristics and capture technologies can influence capture costs.

1.2 Thesis Overview

1.2.1 Aim and scope

The aim of this study is to evaluate the techno-economics of bio-CCS for the pulp and paper industry using two mills in Europe as case studies, where a retrofit with two different PCC technologies (amine-based and carbonate-based) will be evaluated. One mill is based on a paper mill in Belgium whereas the other is based on a pulp mill in Sweden. The techno-economic assessment (TEA) will provide estimates of the capture cost for two different PCC technologies for the two mills where multiple heat integration scenarios have been explored for the Pulp Mill. Using these results, the work aims to determine the following:

1. What are the key characteristics of the two capture technologies and how does this influence the capture cost?
2. Which heat integration scenario results in a lower capture cost? How is this affected by the technology and different energy market conditions (electricity & fuel prices)?
3. If the Pulp Mill has lignin extraction, how does this influence the integration of carbon capture?

From the results of the TEA, the parameters that influence the capture cost of the Pulp Mill the most will be identified. These parameters will then be varied with the aim of providing a wider range of capture cost estimates, which could be applicable to other pulp mills in Europe rather than only one specific pulp mill.

1.2.2 Limitations

Some of the limitations of this study are related to the input data for the energy requirements and equipment cost estimates of the capture technologies investigated and the economic system boundaries, where the evaluation of the process ends after CO_2 compression and liquefaction. The energy requirements, such as cooling, heating (specific reboiler duty (SRD)) and electricity, were obtained through modelling where the different modelling tools have been utilized for the two capture technologies. In particular, the concentration assumed for the amine-based solvent has not been industrially proven, making the modelling assumptions for that technology highly theoretical. The carbonate-based technology on the other hand has been tested in commercial-scale demonstration plant and is in the industrialization phase for some applications. Consequently, for this technology, the design and cost estimations could be derived from industrial experience rather than only from theoretical studies.

As a result, despite having followed the same methodology when performing the TEA, the performance of the two technologies cannot be compared fairly. Moreover, only one design configuration was explored for each of the technologies, and neither were optimized for improved performance or lower CAPEX. Comparing optimized configurations of the two-capture technologies is definitely of interest for future work.

As for the economic boundaries, due to uncertainty of indirect emissions from other activities in the bio-CCS value chain, such as the activities before and after the mill (e.g. wood harvesting and transportation, CO_2 transport and leakages), combined with the cost uncertainty of future transportation infrastructure, calculating CO_2 avoidance cost would require making additional assumptions, adding another layer of uncertainty to the reported cost. In addition, given that the aim of this study is to obtain a general understanding of the capture cost for pulp and paper mills in Europe, the cost of transportation will vary significantly depending on the site-location. The capture cost is deemed appropriate for evaluating the cost efficiency of a capture technology, but for a better representation of the cost of achieving negative emissions, one should evaluate the cost of CO_2 avoided. As stated by Tanzer et al [19], negative emissions are only considered negative emissions if the following conditions are met:

1. The net result is a net decrease in CO_2 emissions despite the added effect of the activities along the CCS value chain.
2. The CO_2 is permanently stored.

With this in mind, and since this work will focus on direct emissions from the mill site and capture costs only, this work will use the term negative emissions sparingly.

2 Literature Review

2.1 Post Combustion Capture

Post combustion capture (PCC) has been selected as the capture technology in this work as it is better suited for retrofits. In the case of pulp and paper mills, specifically kraft mills, PCC may still be a better option whether it is to be implemented on an existing plant or it is a grass-roots design. Recovery boilers, representing a large source of CO_2 emissions in kraft mills, are not only used to generate steam but also to recover the cooking chemicals used in the pulping process. Therefore, complex chemical reactions occur, setting further constraints on the operating conditions. In the case of oxyfuel combustion, the change in the surrounding gas affects mass and heat transfer mechanisms [20]. Whether this could have a large influence on the reactions in the recovery boiler remains unknown as oxy-fuel combustion in kraft recovery boilers is still in the research and development phase.

Black liquor gasification (BLG) has been suggested as an alternative to a conventional recovery boiler where in addition to recovering cooking chemicals, syngas can be produced and utilized for production of transportation and chemicals. Alternatively, the syngas can be combusted in a gas-turbine to produce heat and power, forming what is known as a black liquor gasification combined cycle (BLGCC). Beforehand however, the CO_2 can be removed from the syngas when de-sulfurizing, effectively providing the opportunity for pre-combustion CO_2 capture (see Figure 5.3 in [21]). Onarheim et al. summarized results from literature and found that such a system could result in lower LP-steam and cooling requirement for CO_2 capture compared to MEA-

based post-combustion [21]. However, the pulp and paper industry has not been willing to adopt BLG technology due to associated risks with operating such a complex system. Moreover, BLGCC has not been demonstrated commercially and since this study is focused on near-term implementation of carbon capture, technologies commercially available or nearing commercial availability are of interest.

One example of such a technology is amine-based absorption PCC, which has been commercially demonstrated in other sectors (i.e. power generation, steel, natural gas processing [22] but not in the pulp and paper industry. Depending on the process conditions and the solvent, this requires a heat source at a temperature between 110 to 140°C, with a SRD ranging from 2.2 to 4.9 MJ/kg of CO_2 captured [17]. The most commonly used amine absorbent is monoethanolamine (MEA), but recent studies have shown that using amine blends, such as AMP (2-amino-2-methyl-propanol-PZ (piperazine)), can improve CO_2 capture while simultaneously reducing energy demand. One experimental study completed by Śpiewak et al. found that for similar process efficiency, the regeneration energy for AMP-PZ (30% AMP, 10% PZ) was lower than for MEA [23]. Additionally, in a pilot test campaign, it was found that a blend of AMP PZ (ca 26.7 % AMP and 12.9 % PZ) achieved a lower specific solvent regeneration heat demand than MEA [24]. Aside from improved energy efficiency, AMP-PZ also has a greater resistance to oxidative and thermal degradation compared to MEA [25]. As result of its lower degradation, combined with its potential to achieve a lower regeneration energy, AMP-PZ has been proposed by IEAGHG as the new benchmark solvent and therefore has been selected as such in this work.

In addition to reducing the SRD, research efforts have also focused on lowering the regeneration temperature of the absorbents. This is of particular interest for the pulp and paper industry where residual heat is often available below 100°C [15]. Without residual heat available, the heat demand of the capture process can only be met by low-pressure steam, which leads to a loss of power generation if additional fuel use is not implemented. The novel process investigated in this work offers the possibility to use residual heat to meet part of the reboiler duty as its regeneration temperature can be as low as 85°C. The solvent of this emerging PCC technology is a water-based solution, which contains potassium carbonate and an additive that acts as a reaction catalyst for improving the conversion of CO_2 into bicarbonate. This technology is nearing commercial availability with a full-scale pilot testing currently on-going in an industrial site.

Similarly to AMP-PZ, the carbonate-based solvent has a low-degradation rate. When comparing the cost of the carbonate-based solvent to AMP-PZ, potassium carbonate costs less than AMP-PZ, 0.46 €/kg vs. 8 €/kg (AMP) and 6 €/kg (PZ) [26]. As for the additive mentioned above, the cost of this has been found to be much higher than the cost of AMP-PZ. However, its concentration in the solution is significantly lower, which could make the cost of both solvents comparable. This of course depending on how much make-up solvent each process consumes. There are other key differences between the carbonate and AMP-PZ process, but these will be further discussed in their process description in Section 3.4.

2.2 Carbon Capture in the Pulp and Paper Industry

There exist several published studies related to the implementation of carbon capture in the pulp and paper industry, where only a few have evaluated the economic feasibility of capturing CO_2 in a pulp and paper mill. The focus of the literature review was on studies which looked at the implementation of PCC, as this the technology of choice for this work. The reported cost data obtained from literature has been summarized in Table 1. The studies which included transport and storage did not include the emissions from these activities in the capture cost. The point sources considered for capture in each work is detailed in the footnote of Table 1.

Table 1: Summary of cost data from literature for PCC in the pulp and paper industry

Year	Reference	Solvent and SRD	CO_2 Flow for capture (tons/year)	Cost (€/tonCO_2)	Capture Rate	Cost metric
2009	Hektor et al [15]	MEA (SRD 2880 kJ/kg CO2)	1 000 000 – 1 468 800[a]	25-53 (kraft mill) 20-65 (integrated pulp and paper mill)	90%	Avoidance cost; including the cost of storage and transportation by cost function.
2017	Onarheim et al [16]	MEA (SRD 3.1-3.2 MJ/kg	1 400 000-2 000 000[b]	52-66 (kraft mill) 71-89 (integrated pulp and board mill)	90%	Avoidance cost; difference between levelized cost of pulp production with CCS and without. Transport and storage included (Assumed 10 euro/tonCO_2)
2019	Nwaoha and Tontiwachwuthikul [27]	MEA (SRD 5.3 MJ/kg) And MEA-AMP blend (3.16-4.74 MJ/kgCO2)	147,402[c]	120-124[e] (AMP-MEA blend) 128-139[e] (MEA)	90%	$Capture\ cost = \frac{CAPEX + OPEX}{Captured\ CO_2}$

Table 1 continued

2022	Parkhi et al. [28]	MEA (SRD of 3.3 MJ/kg)	196,000[d]	65-77[e] (kraft mill)	90%	$Capture\ cost = \frac{CAPEX + OPEX}{Captured\ CO_2}$

[a] Emissions from recovery boiler considered for capture and emissions from integration (i.e. for the NGCC configuration, capture from flue gas of gas turbine was included)
[b] Different combination of flue gases for capture explored (lime kilns only, or lime kilns & recovery boiler) but emissions from use of auxiliary boiler to meet heat of capture demand not included for capture
[c] Only considered emissions from power boiler but mill equipped with
[d] Emissions from the Lime kiln of the stand-alone kraft mill studied in [16]
[e] Originally in $/ton$CO_2$ but converted to €/ tonCO_2 using exchange rate of 1$ = 0.94€

The most recent study completed by Parkhi et al. [28] only looked at the cost of capturing from the lime kiln of the stand-alone mill studied by Onarheim et al. in 2017 [16]. It was concluded that cost data reported by the two studies did not agree due to different methodologies followed for estimating CAPEX. The OPEX did not differ that much, despite the different methodology followed where Parkhi et al. assumed a cost for steam (9.45$/ton) and Onarheim et al. looked at potential for heat integration between the mill and the capture plant.

In the study completed in 2019, the focus was on the capture plant and how different process configuration modifications, such as rich solvent split, impacts the overall capture cost [27]. Two different solvents were also compared: MEA-AMP and MEA. The mill studied was a pulp mill with one recovery and power boiler where capture was only considered for the power boiler. As can be observed in Table 1, the capture cost is quite high compared to the other studies. With the cost metric definition differing between studies, as well as the fact that different cost methodologies were followed, it is difficult to understand why the cost reported by Nwaoha and Tontiwachwuthikul is significantly higher. The authors concluded that variable operational costs, namely the regeneration energy, contributes the most to the capture cost. In the case of the MEA, this is likely in part because the SRD is significantly higher than the SRD of the other studies detailed in Table 1. Otherwise, the higher cost could also be explained by their assumption that the heat demand for capture is met by steam with a cost of 16.8 US$/GJ (15.7 €/GJ), neglecting any opportunity for optimal heat integration between the plant and the capture process.

As for the study in 2009, Hektor et al explored different heat integration configurations for both an integrated and stand-alone pulp mill, where it was found that replacing the existing biomass boiler of the system with a NGCC resulted in the lowest CO_2 avoidance cost [15]. One of the configurations included using a heat pump to meet the capture's steam demand by producing LP steam using low-temperature excess heat. Hektor et al. found that the heat pump was not the most cost-effective option but only because it did not provide an increase in power production, unlike the other configurations explored. This is likely one of the reasons why the NGCC performed the best, given that this configuration produced almost 3 times as much power as the reference mill, converting the mill from being a net power importer to a net power exporter.

Another important takeaway from Hektor's work is that an integrated pulp and paper mill resulted in a higher CO_2 avoidance cost than a stand-alone pulp mill, when increasing the size of the onsite biomass boiler to meet the process heating demands of both the mills and the capture process. The study in 2017 by Onarheim et al. showed a similar result. As was concluded by both studies, this is due to the fact that there is less excess energy available in an integrated pulp and paper mill compared to stand-alone pulp mill. There was therefore a greater need for additional steam generation to cover the heat demand of the capture process, reflected in the capture cost through additional fuel use. A stand-alone pulp mill has been selected in this work in part due to the unfavorable energy balances of an integrated pulp and paper mill.

3 Process Description

3.1 Paper Mill

The modelled Paper Mill is based on the Langerbrugge mill located in northwest Belgium, which has an annual paper production of 540 000 t/a using recycled paper as a feedstock. The mill is equipped with two steam boiler combined heat and power (CHP) plants, generating approximately 180 MW of steam. This covers all of the mill's steam demand, as well as a approximately 70% of its electricity demand. The steam distribution network of the mill is shown below in Figure 1, where the network consists of four headers: two high-pressure (HP) headers, one low-pressure (LP) header and a condensate header.

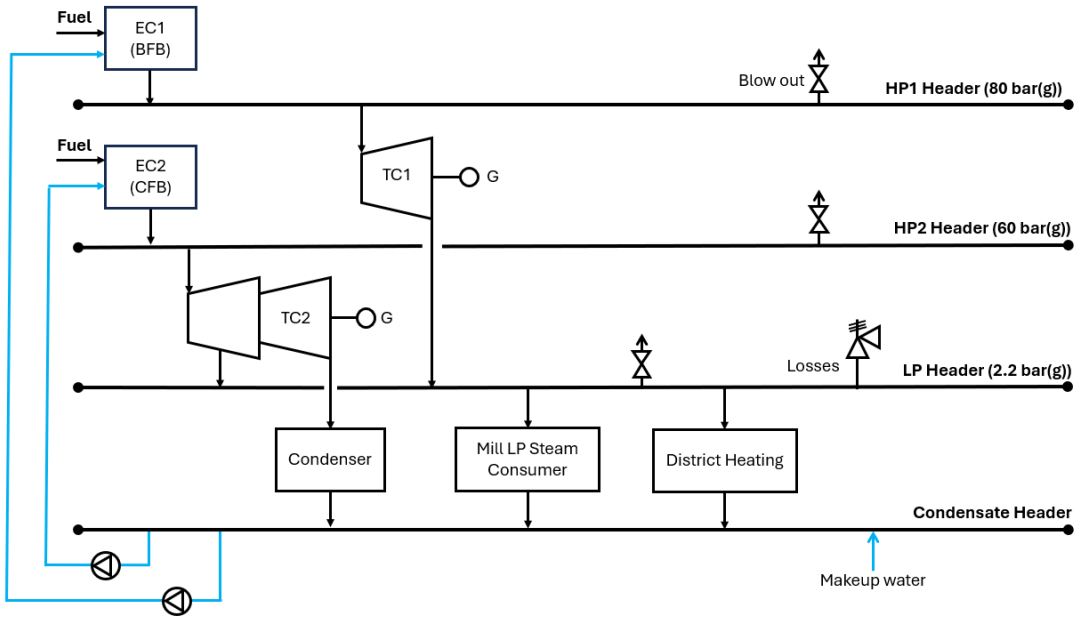

Figure 1: Simplified steam network for the Paper Mill where the first CHP plant consists of a bubbling fluidized bed (BFB), denoted as EC1, and a back-pressure turbine (T1). EC1 produces HP steam which is then fed into the back-pressure turbine to be fully expanded to the LP header. The second CHP plant consist of a circulating fluidized bed (CFB), denoted as EC2, and a condensing turbine. EC2 produces HP steam which is partly expanded to the LP header via an extraction. The remaining is expanded to a condensing stage where heat is rejected via an air-cooled cooling tower.

The real mill is also equipped with three auxiliary natural gas boilers, but these have been omitted from the steam network model as they are rarely operated. As for other key aspects of the steam network such as the accumulator tanks and pressure reducing valves for example, these have been omitted for simplicity. Further details on the two CHP plants are shown in Table 2 whereas the pressure, temperature and enthalpy specifications for the steam headers are presented in Table 3 .

Table 2: Specifications for the two CHP plants in the Paper Mill: Boiler and turbine details

Power Plant	Boiler Specifications			Turbine Specifications	
	Type	Fuel	Capacity (MW_{th})	Type	Capacity (MW_e)
CHP1	EC1: Bubbling fluidized bed	DIP sludge, waste wood	55.00	TG1: Back-pressure	11.00
CHP2	EC2: Circulating Fluidized bed	Waste wood, RDF, sewage sludge, biosludge	125.00	TG2: Condensing with steam extraction	43.00

Table 3: Steam header properties for the steam utility system of the Paper Mill

	Pressure [bar(g)]	Temperature [°C]	Enthalpy [kJ/kg]
High-pressure steam (EC1)	80	480	3348
High-pressure steam (EC2)	60	440	3283
Low-pressure steam*	2.2		

*Including only steam supplied by T1 and the steam extraction from T2

The Mill LP steam consumer shown in Figure 1 represents steam consumption for the mill's production processes, as well as the steam consumption for air-preheating and feedwater heating. The mill also delivers LP steam to a nearby plant, represented as district heating (DH). The amount of heat cooled in the turbine condenser of the second CHP plant, along with the steam and power production of the two CHP plants is detailed in Table 4. These values represent the average between summer and winter operation, where the operation of each season is based on data collected in February 2022 and July 2021, respectively. Seasonal variations in heat demand and supply which could affect the capture plant have been neglected for the analysis, based on the assumption that they are not significant.

Table 4: Key annual data of Paper Mill from 2021

Steam Production (MW)	Power Produced (MWe)	Electricity Purchased (MWe)	Heat rejected in Turbine Condenser (MW)
176	47	20	54

The fuel input of both boilers varies over time and includes de-inked plant (DIP) sludge, sewer sludge, waste wood and Refused Derived Fuel (RDF), or a combination of these fuels. As RDF is made partly from municipal solid waste (MSW), the CO_2 emissions of the mill are not fully biogenic. Based on what was reported in 2021, the total CO_2 emissions of the mill, including the auxiliary boiler, was 609 kton. Of these total emissions, 447 kton can be considered biogenic (73%) [29].

The capture process was sized only to capture from the flue gas of EC1 (CFB) and EC2 (BFB), where emissions from the auxiliary boiler have been neglected due to its intermittency and low flows. The CO_2 concentration and the size of the flue gas of each boiler are detailed in Table 5. It is assumed that all the flue gases will be mixed before being sent to a common absorber in the capture plant.

Table 5: Average flue gas data of streams considered for capture in the Paper Mill

	Flow (Nm^3/h dry)	CO_2 [Vol% dry] *	Operation (h/year)	Temperature [°C]
CHP1	100 000	15	8000	65
CHP2	200 000	12	8000	60
Total flow to capture plant	300 000	13	-	≈62

*CO2 concentration is not based on measured values but based on the total reported CO_2 emissions and flue gas volume flows.

3.2 Pulp Mill

The modelled Pulp Mill is based on a stand-alone Swedish kraft mill located on the east coast of Sweden, producing 500 kt/a of pulp using wood as a feedstock. The pulp produced is primarily fluff pulp which is used to make diapers, other hygiene products and non-woven material. The mill also produces paper pulp which is used to make liquid cardboard and special paper. As the mill is not an integrated pulp mill, these products are produced elsewhere. The mill's heating demand is met primarily by the production of steam using high-temperature excess heat from two recovery boilers. When the steam demand is greater than what can be supplied by the recovery boilers, steam is also produced in a power boiler. Aside from supplying heat to the mill, the steam is also fed through a back-pressure turbine, which meets approximately 76% of the mill's annual electricity demand. The steam distribution network of the mill is shown below, where the network consists of three headers: one HP header and two LP headers.

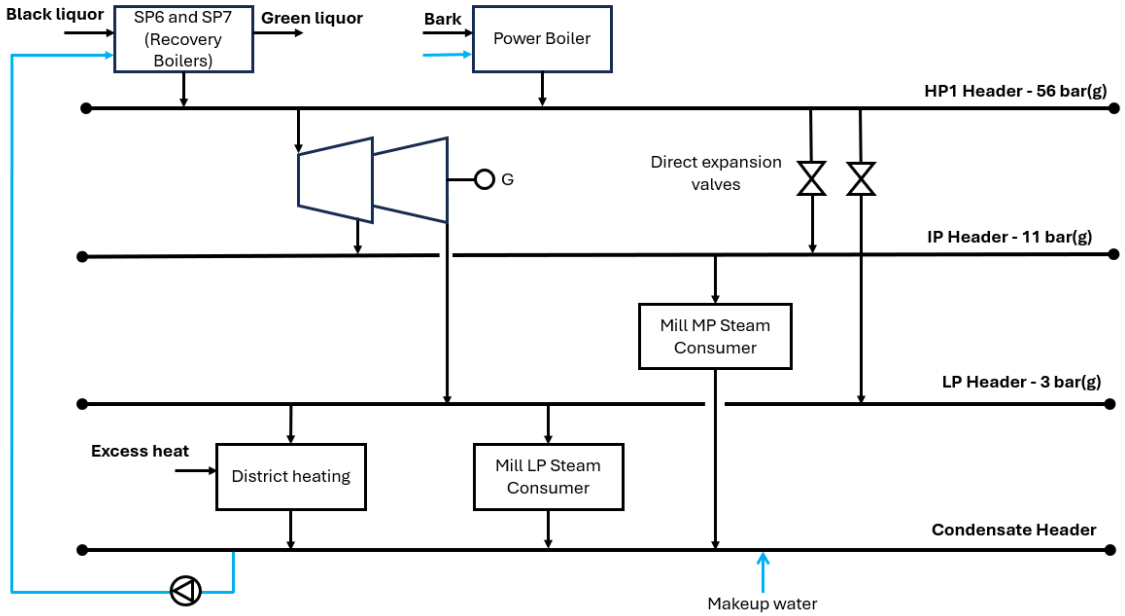

Figure 2: Simplified steam network of the Pulp Mill where there are two recovery boilers (SP6 and SP7) which produce steam from the combustion of black liquor. The back-pressure turbine receives HP steam which is partly expanded to the MP steam header via an extraction. The remaining is expanded to LP steam header. There is also the possibility of by-passing the turbines and passing the steam directly to MP and LP header via expansion valves. The mill is also equipped with a power boiler which is utilized when there is an additional need for steam which cannot be met by the recovery boilers.

What is not shown in the steam network in Figure 2 is the mixing of MP and LP steam, creating an intermediate steam level. This steam mixture is used in the last stage of the pulp making process for drying the pulp, which requires a heat supply at a temperature between the MP and LP steam level. Similarly to the steam network of the Paper Mill, the steam network in Figure 2 is a simplified representation of the Pulp Mill's actual steam network. Further details on the boilers and turbine of the mill are provided in Table 6 whereas the pressure and temperature specifications for the steam headers are presented in Table 7.

Table 6: Specification of the boilers used for heat and power production in the Pulp Mill.

Boiler Specifications			Turbine Specifications	
Type	Fuel	Capacity (MW_{th})	Type	Capacity (MW_e)
SP6 Recovery Boiler	Black liquor	100	BPT: Back-pressure turbine with extraction	46
SP7 Recovery Boiler	Black liquor	400		
Power Boiler	Bark (wood residue) and tall oil pitch*	95		

* Used for startup and occasionally as a marginal fuel when there is not enough bark to meet steam demand

Table 7: Steam properties for the steam utility system of the Pulp Mill

	Pressure [bar(g)]	Temperature [°C]
High-pressure steam	56	440
Medium-pressure steam	11	240
Low-pressure steam	3	145

The Mill MP and LP steam consumers shown in Figure 2 represent steam consumption for the mill's production processes, as well as steam consumption for non-process heating. Heat exchange between the mill's process streams and the steam mainly occurs through a heat exchanger but in some cases, the steam is mixed with process streams directly. The mill also supplies district heating to a nearby town, which is met using a combination of process excess heat and utility steam. Key annual data of the mill is shown in Table 8, where the values shown are annual averages based on monthly data collected for 2021.

Table 8: Key annual data of Paper Mill from 2021.

Primary steam Recovery Boilers (MW)	Power Produced (MWe)	Electricity Purchased (MWe)
286	36	11

It is important here to distinguish between the role of the recovery boiler and power boiler in the Pulp Mill. As can be observed in the simplified process flow of the pulping process in Figure 3, the power boiler is mainly fed with bark residue and its only role is to produce steam. The bark residue predominately comes from debarking the wood feedstock in wood handling section but at times bark or other wood processing residues is also purchased if the demand exceeds what can be supplied on-site. Bark can also be sold to external customers if on-site supply exceeds demand.

One role of the recovery boiler is to recover chemicals used in the cooking section of the pulping process. This is achieved by the chemical conversion of inorganic components in strong black liquor. The organic components of the black liquor, which are by-products from the pulping process, are instead combusted in the recovery boiler and used for steam production. The reason why it is important to make the distinction between the recovery boiler and the power boiler is because the high temperature heat from combustion of black liquor is process-inherent and therefore unavoidable (as long as conventional process technology is used for chemical recovery in the mill). This means that the CO_2 emissions of the recovery boiler are unavoidable in the current method of producing pulp.

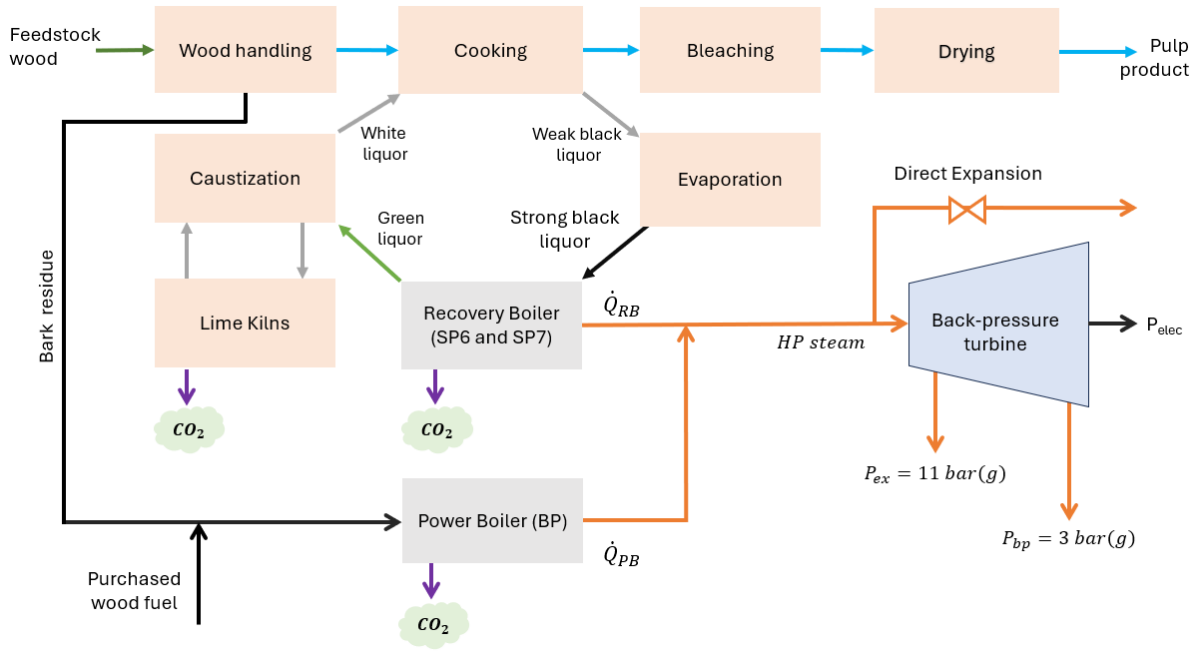

Figure 3: Simplified process flow chart for the kraft pulping process and how it interacts with the steam production network. The wood handling section involves debarking, de-icing (depending on the outside conditions) and a chipper. As for the cooking section, this includes air removal, white liquor impregnation and the digester. The evaporation section consists of a series of integrated evaporator effects which remove the water of the weak black liquor, improving the efficiency of the combustion.

As for the CO_2 emissions of the lime kilns, this is partly due to the use of tall oil pitch (biofuel formed by processing crude tall oil into tall oil diesel), which is supplied for the regeneration of burnt lime from calcium carbonate. Burnt lime is a key ingredient for last step for the regeneration of cooking chemicals. In addition to the combustion of tall oil pitch, there is also CO_2 generated through the oxidation of the carbonate. The emissions from the lime kiln are therefore also partly process-inherent and thus unavoidable.

There are two important take-aways from the pulping process flowsheet shown in Figure 3: (1) there are several point sources of CO_2 in a kraft mill and (2) part of these emissions are process-inherent and therefore unavoidable. The capture plant for both technologies was sized for capturing the emissions of the two recovery boilers (SP6 and SP7) and the lime kilns. These emissions are considered fully biogenic. Emissions from the power boiler have not been included in the capture process due the large operational variations of the power boiler. Additionally, as will be seen in the subsequent sections when looking to integrate the capture process, the load of the power boiler will vary depending on the heat integration scenario and therefore so will its CO_2 emissions. The CO_2 concentrations and the sizes of the flue gas streams from the lime kilns and both recovery boilers are detailed in Table 9.

Table 9: Average flue gas data of streams considered for capture in the Pulp Mill

	Flow (Nm^3/h dry)	CO_2 [Vol% dry]*	Operation (h/year)	Temperature [°C]
Lime kilns	45,000	22	8,400	300
Recovery Boiler SP6	100,000	12	8,400	54*
Recovery Boiler SP7	300,000	16	8,400	170
Total flow to capture	445,000	16	-	125

*The flue gas from SP6 enters a scrubber, hence the lower flue gas temperature compared to SP7

It is assumed that all the flue gases will be mixed before being sent to a common absorber in the capture plant, resulting in a total approximate CO_2 concentration of 16 %vol dry. Prior to entering the capture plant, the flue gases of the lime kilns and the recovery boiler SP7 will be mixed and cooled to 125° This heat will be recovered using a heat exchanger with the aim of utilizing it to meet some of the process heating demand. How this will be done in practicality has not been explored, however the cost of this heat exchanger will be considered in the capital costs estimations.

3.3 Pulp Mill with Lignin Extraction

This mill is based on the Pulp Mill described Section 3.2 where it is assumed that the mill has taken strategic steps towards better resource utilization by extraction lignin from black liquor, effectively converting more of the carbon from the feedstock into a valuable product. An extraction rate of 0.12 tonnes of lignin per air dried tonne of pulp was assumed for this work. This is within the feasible extraction rates reported in literature [30] and is similar to what has been seen in another Pulp Mill in Finland (0.13 tonne lignin/Adt) [31]. As shown in Figure 4, lignin extraction occurs before the strong liquor is sent to the recovery boilers. This has several impacts on the standard operation of the mill but the impact that it has on recovery boilers are in focus in this work as this directly affects the capture plant.

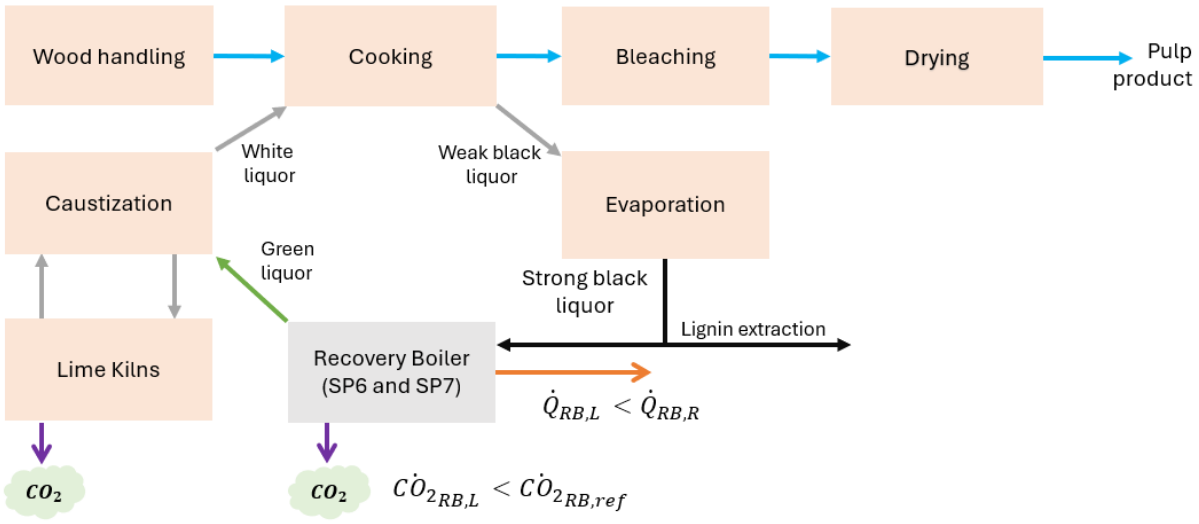

Figure 4: Simplified kraft pulping process with lignin extraction where the subscript RB, ref denotes the recovery boilers of the Pulp Mill described in Section 3.2 whereas RB, L denotes the recovery boilers for the Pulp Mill with lignin extraction.

Lignin is mainly composed of carbon (62.50 wt-%) and oxygen (29.15 wt-%), with some hydrogen, sulfur, and nitrogen ([30]). When extracted from the black liquor, it alters the properties of the black liquor such as the heating value and dry solids content. Notably, this reduces the head load into the recovery boilers, resulting in lower steam generation. According to a previous study completed by [32], steam generation is reduced by 5.4 MWh per tonne of lignin extracted. As shown in Table 10, this translates to an approximate 38 MW reduction in steam produced by the recovery boilers. In addition, as there are fewer carbon molecules in the black liquor, lignin extraction reduces the CO_2 emissions of the recovery boilers, thus resulting in less CO_2 available for capture.

Table 10: Changes in steam production and CO_2 emissions due to lignin extraction

	Recovery boilers steam production (MW)	CO_2 Flow (kg/s)
Pulp Mill without lignin extraction	286.0	36.5
Pulp Mill with lignin extraction	248.4	32.6

3.4 Capture Technologies

The PCC amine-based process using AMP-PZ solvent will henceforth be referred to as the benchmark process and the carbonate-based process will be referred to as the novel process. The benchmark process was designed and simulated using Aspen Plus v10 for the two mills described in Section 3.1 and 3.2, whereas the novel process was modelled using Protreat 6.6 (Optimized Gas Treating Inc. 2001). The simulation results, specifically the energy requirements for capturing and conditioning of each technology, was used as input data for the heat integration analysis presented in this work.

The carbon capture process for the Pulp Mill with lignin extraction was not simulated explicitly. Instead, based on estimations that the CO_2 concentration of the flue gases would remain the same, the specific energy requirements were assumed to be the same as for the mill without lignin extraction. Capital costs were also based on the estimations made for the mill without lignin extraction but scaled using an exponent to adjust for the lower CO_2 flow.

For the benchmark process, the solvent composition was chosen to be 33 wt% AMP – 12 wt% PZ (the composition is expressed in mass basis –$g_{APMP/PZ}/g_{solution}$). It is worth noting that this composition differs from that reported in literature, where 40% is indicated as the best concentration by most studies. The higher amine concentration assumed in this work benefits the capture efficiency and lowers the specific reboiler duty, but it has not been industrially tested and could potentially lead to problems with, for example, corrosion or foaming.

The two technologies are similar where CO_2 is captured by absorption, however as previously stated, the different solvents result in a different regeneration temperature in the stripper column. This lower regeneration temperature allows part of the novel process's heat demand to be met using hot water instead of low-pressure steam. Furthermore, the stripper column of the novel process operates at sub-atmospheric pressure. Consequently, once the CO_2 exits the capture plant, it is passed through a compressor for pre-compression to 1 bar(g), achieving the same exit conditions as the CO_2 captured in the benchmark process.

The last and important difference between both capture technologies is their energy requirements such as heating (SRD), cooling and electricity. Figure 5 shows the energy requirements of both the benchmark process and the novel process, which were estimated using their respective simulation models and modelling assumptions. Note that the results cannot be compared due to the different nature of the input data where, in particular, the benchmark results are purely theoretical. Also note that in the case of the Pulp Mill with lignin extraction, the energy requirements would be lower than what is shown in Figure 5 due to the reduction in the amount of CO_2 captured.

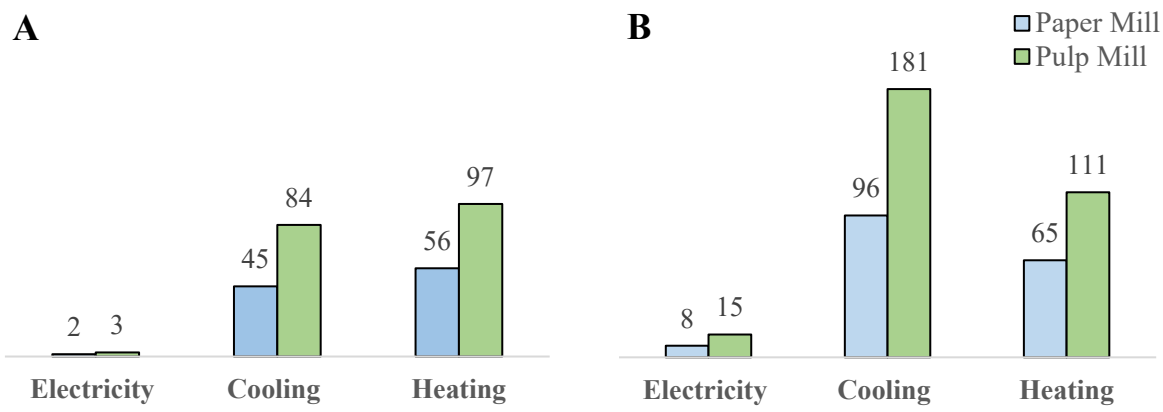

Figure 5: Energy requirements for the two capture technologies where (A) is the benchmark process (theoretical modelling approach) and (B) is the novel process (industrial design approach). Heating corresponds to the heat required for the reboiler. The values shown are in MW.

When comparing the specific energy requirements of the benchmark process between the two studied mills, it can be observed that they are comparable (see Table 11). The SRD of the Pulp Mill is slightly lower due the concentration of CO_2 in the flue gas being higher. As for the specific cooling duty, this is larger for the Pulp Mill due to the larger flows.

Table 11: The stripper reboiler temperature and specific energy requirements (kJ/kgCO_2) of the benchmark process for the two mills

	Stripper Reboiler Temperature [°C]	SRD	Specific Cooling Duty	Specific Electricity Consumption
Paper Mill	116.5	2980	2370	800
Pulp Mill	115.1	2950	2500	800

3.5 CO_2 Conditioning for Storage

The captured CO_2 exits both capture plants described in Section 3.4 at the same conditions (1 bar, 28°C). To be fit for buffer storage and transportation, the CO_2 undergoes a liquefaction process to 16 bar(a) and -26°C. These specifications are based on the Northern Lights requirements but this could be different for other storage sites. Shown in the simplified process flowsheet in Figure 6 , the captured CO_2 is first compressed by a multi-stage compressor, with intercoolers, before being liquefied by an ammonia refrigeration cycle. Aside from achieving an appropriate pressure for transportation, the liquefaction process also serves to remove impurities in the feed gas such as water and O_2, ensuring that the CO_2 meets the purity requirements for storage.

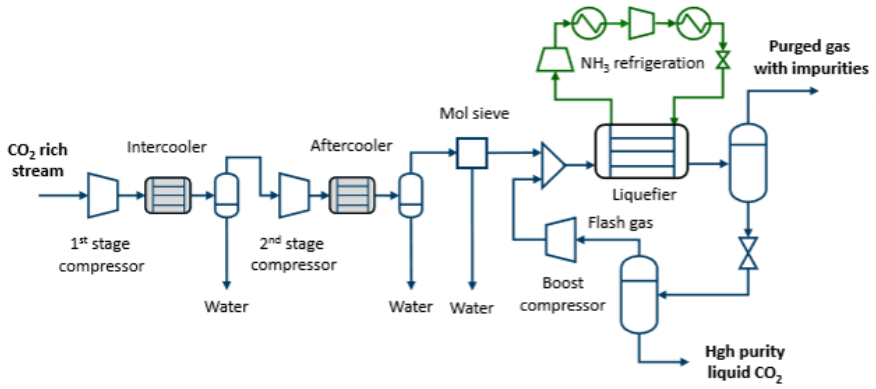

Figure 6: Simplified process flowsheet of liquefaction plant taken from [33]

The specific cooling requirements the liquefaction process are detailed in Table 12. The total cooling duty for liquefaction is equivalent for both technologies as equal amounts of CO_2 is captured. For more information regarding the process and how it was modelled, please refer to [34].

Table 12: Specific cooling duty for liquefaction process shown in Figure 6

	Specific cooling duty (kJ/kgCO_2)
Compressor train intercooling 1	119
Compressor train intercooling 2	113
Cooling refrigerant intercooling stage 1	35
Cooling refrigerant intercooling stage 2	405

4 Methodology

4.1 Integrating Carbon Capture with Reference Mills

In order to evaluate the cost of implementing carbon capture, a thermodynamic analysis was first performed to obtain the heat and power co-generation potential of the integrated system. In all cases (the Pulp or Paper Mill), the energy balance of the mill is directly affected by the need for LP steam to cover all or part (in the case of the novel process) of the heat demand of the stripper reboiler. This will typically come at a loss in power production (or possibly in an increased demand for fuel), in addition to increasing the mill cooling water and electricity demand. All of which influence the operational cost of carbon capture. The magnitude of the impact on the mill's energy balance depends on the technology and on the way in which the integration is implemented. The differences in the process heat flows, notably the unavoidable high-temperature excess heat from the recovery boilers, make the carbon capture integration for the Pulp Mill more complex than for the Paper Mill. The heat and power co-generation potential of the Pulp Mill once integrated with carbon capture was therefore evaluated using a different method based on a pinch analysis approach.

4.1.1 Paper mill

Due to limited opportunity for improved heat recovery, the heat demand for both capture processes can only be met by LP steam produced by the two co-generation plants (EC1 and EC2) of the Paper Mill. Namely, from the steam flow passed through the condensing stage turbine of EC2. The thermodynamic analysis consequently consisted of calculating the mass flow of the LP steam required to meet reboiler duty of the capture process, and then evaluating how this steam extraction influences the net power output of the mill. The mill described in Section 3.1 is representative of the base case mill used for quantifying the energy penalty of implementing carbon capture. It is worth noting here that using the same heat source to meet the heating demand for capture negates an advantage that the novel process has over the benchmark process, which is the possibility to use heat at a lower temperature.

4.1.2 Pulp mill

Using pinch analysis tools, a two-step process was followed where first the ideal heat integration potential between the mill and the capture process was determined, followed by estimating the effect that this integrated pulping and capture process has on power co-generation potential of the mill. This was done following a similar method that has been proposed by Svensson et al. for estimating the potential availability and the trade-offs for recovery of excess heat [18]. The trade-offs investigated in this work were minimizing fuel use or maximizing electricity production, similarly to what has been done in [35]. However, in this work, as will be discussed subsequently, limitations on power capacity have been considered when optimizing for maximum electricity production.

Pinch analysis is a well-established method used to minimize the utility requirements of an industrial process by identifying internal heat recovery potential. When looking to optimally integrate two processes, for instance a pulp mill and carbon capture, pinch analysis can also be used to determine ideal heat integration from an established heat recovery target. The use of pinch analysis tools in this work will be described based on the assumption that the reader is somewhat familiar with pinch analysis concepts. If this is not the case, the reader may refer to [36] for more details.

The ideal heat integration potential between the mill and the capture processes was determined using one of the main tools of pinch analysis, the grand composite curve (GCC). As detailed by Kemp [36], the steps involved in such an analysis include data extraction, selecting a minimum temperature difference for heat exchanger (ΔT_{min}), establishing energy targets through optimized heat cascades, and lastly representing the optimized heat cascade graphically in the GCC.

In this work, the optimal heat cascades were established assuming a ΔT_{min} of 10 K. For better visualization of the potential integration between the Pulp Mill and the capture process, the GCC for each process was drawn separately but on the same chart, with the GCC for the capture and conditioning plant mirrored. This is referred to as a split GCC, where the so-called background process includes the heat sources and sinks of the mill, whereas the foreground process includes the process streams for the capture and conditioning. The temperatures and cooling loads of the streams considered in this analysis can be found in Appendix A for reference.

It is important to note that the GCC is generated for maximum internal heat recovery without taking into consideration limitations on process layout. The downside of this approach is that it is unlikely that the ideal heat integration scenarios identified in this work can be fully realized in an actual process. Be that as it may, providing a detailed design of an optimized heat recovery system for a specific mill is not the aim of this work. This work aims to explore and compare different possible heat integration scenarios for a pulp mill. For these comparisons to be fair, they must all have the same reference point i.e. it would be unfair to compare the performance of an optimized integrated mill (one obtained from a GCC) to a non-optimized configuration (present day mill described in Section 3.2).This means that the by using pinch analysis tools, the base case mill for this analysis is a more energy efficient mill than what was described in Section 3.2. This somewhat follows the previous assumption that the implementation of carbon capture will occur in the future, where it is likely that the mill has implemented measures to improve its energy efficiency.

The effect that the integrated pulping and capture process has on power co-generation potential of the on-site steam cycle was then evaluated following a similar process as described above, where the background process refers to the integrated pulping and carbon process, and the foreground includes the process streams of the steam cycle. The size of the steam cycle was then optimized based on selected trade-offs between minimizing fuel use and maximizing electricity production. The way in which one was prioritized over the other when energy targeting was done in the same way as described in Section 2.4 of [35]. A simplified steam cycle, similar to what was shown in Figure 2 was assumed for heat integration where it was assumed that the turbines could by-pass to deliver LP steam to the process without co-generation of power.

In the case of minimizing fuel use, the heat cascades of the steam cycle were optimized with the aim of minimizing hot utility use (i.e. fuel use in the power boiler). This optimization scenario will henceforth be referred to as Scenario A. As for maximizing electricity generation, the additional LP steam demand incurred by the implementation of the capture plant, allows the potential to generate more back-pressure power than in the reference mill without carbon capture. However, this would require investing in an additional turbine as the Pulp Mill's existing turbine capacity is not enough to provide this additional back-pressure power. Two possible heat integration scenarios have therefore been explored when optimizing for maximum power generation: one that is constrained by existing turbine capacity (Scenario B) and one that constrained by the minimum heating demand of the mill, which has now increased due to the

implementation of carbon capture (Scenario C). The limit imposed on power generation was 46 MWe, as this is the size of the turbine of the existing Pulp Mill that is being modelled.

All of the energy targeting computations, including the integration of carbon capture with the mill were achieved using an in-house MATLAB-based tool called Mat4PI. The impact that each scenario has on fuel input and electricity production, relative to the base case Pulp Mill without carbon capture, can be observed in Table 13.

Table 13: Changes in heat input and electricity production for all heat integration scenarios relative to the base case for Pulp Mill without lignin extraction

Parameter	Scenario A1 Minimizing Fuel Use	Scenario B1 Maximizing Electricity Production – turbine constrained (TC)	Scenario C1 Maximizing electricity production – heat demand constrained (HDC)
Fuel Input	Unchanged	▲	▲
Electricity Production (46 MWe)	▼	Unchanged	▲

As for the lignin extraction, the same process described above was followed where the base case mill is a futuristic and efficient mill with lignin extraction. As was described in Section 3.3, extracting lignin reduces the available high temperature excess heat of the recovery boilers by reducing the carbon that can be combusted. Therefore, it may be possible that additional fuel will be required to ensure that there is sufficient heat to meet the process heating requirements of the mill and the heating demand for carbon capture, regardless of the technology. In addition, as a consequence of the reduction in excess heat, the power production of the Pulp Mill with lignin extraction without capture is reduced from 46 MWe to 40 MWe. The changes in heat input and electricity production of each scenario relative to the base case with lignin extraction is shown below in Table 14.

Table 14: Changes in heat input and electricity production for all heat integration scenarios relative to the base case for the Pulp Mill with lignin extraction

	Scenario A2 Minimizing Fuel Use	Scenario B2 Maximizing Electricity Production – turbine constrained (TC)	Scenario C2 Maximizing electricity production – heat demand constrained (HDC)
Fuel Input	Increased or unchanged	▲	▲
Electricity Production (40 MWe)	▼	▲	▲

The reader is encouraged to revisit these two tables later when reading the Results and Discussion as the aim here is to also define the nomenclature of each scenario clearly, where 1 and 2 differentiate between a mill without lignin extraction and a mill with lignin extraction, respectively. Instances where the scenarios are referred to without specifying 1 or 2, for example Scenario B, signifies that what is written applies to both of the pulp mills.

4.2 Economic Assessment

This works aims to follow, to the fullest extent, the current best practices for estimating the cost of implementing carbon capture and storage in industry. Such practices have been established by Roussanaly et al in a white paper [37] and specifically for industry in [38]. Following some of their recommendations, the author of this work hopes to facilitate comparisons with future work, while making reasons for any discrepancies obvious to the reader. Among other things, the authors of the white paper propose a framework for estimating future "Nth-of-a-kind" (NOAK) costs for technologies that have not yet reached commercial maturity. Two different methods are proposed depending on the purpose of the cost estimate. Since carbon capture has never been commercially demonstrated in the pulp and paper industry, the cost methodology used in this work more closely resembles the authors "What if" approach, whereby a bottom-up engineering-economic method was applied. Following the recommendation in [37], a more appropriate term for the costs obtained in this study are "*aspirational NOAK*" cost estimates.

The economic assessment was completed by evaluating the OPEX and CAPEX of the capture plant assuming fixed annual costs (annuity method), where the key economic indicator selected is the capture cost. Defined in Equation 1, this relates the cost needed to build and operate the capture and conditioning plant to the total amount of CO_2 captured and conditioned by the plant. This definition for the capture cost follows what has been recommended in [38].

$$Capture\ cost = \frac{\mathrm{C}_{CAPEX} + \mathrm{C}_{OPEX,fix} + (C_{electricity} + \mathrm{C}_{OPEX,other} + C_{fuel})}{CO_2\ captured} \quad (1)$$

where CO_2 captured is based on a 90% capture rate, $\mathrm{C}_{\mathrm{CAPEX}}$ is the annualized capital investment for the capture and conditioning plant and $\mathrm{C}_{OPEX,fix}$ is the fixed annual operational costs for capture and conditioning. The variable OPEX has been split into three terms where $C_{electricity}$ includes the electricity demand of the capture and conditioning plant, as well as the net change in electric power production due to the additional heat demand for capture. $C_{OPEX,other}$ includes cost of material consumptions (process, solvent, and cooling water), whereas C_{fuel} is the cost of additional fuel for steam production. Lastly, $\mathrm{C}_{OPEX,fix}$ includes maintenance, insurance, and labour cost. The parameters assumed for the operational costs are shown in Table 15. Note that the cost of the first fill of the solvent for both technologies was not included in the total cost estimations.

The CAPEX for the capture and conditioning plant of both technologies was evaluated using NOAK cost factors, since it was assumed that the retrofit will occur in a future, where CCS integration with the pulp and paper industry has reached a greater level of maturity. Moreover, performing NOAK cost estimates contributes to a more similar evaluation of the benchmark and the novel capture technology, which both currently have different technology readiness levels (TRLs). The contingency factors considered in the bottom-up estimation of the total capital requirements (TCR) cost for capture and conditioning are detailed in Table 15. The equipment cost and installation costs for the capture and conditioning plant were provided as input data to this work.

When evaluating the capture cost for a Pulp Mill with lignin extraction, the TCR of the capture and conditioning plant was multiplied by a scaling factor to account for the reduction in equipment size due to the reduction in CO_2 captured. The scaling factor was obtained from Equation 2 for scaling the CAPEX of a reference plant for a plant with different flue gas size but with the same CO_2 flue gas concentration.

$$C_i = C_{ref}\left(\frac{CO2_{emissions,i}}{CO2_{emissions,ref}}\right)^n \quad (2)$$

where the scaling exponent, *n*, was obtained from [9] and is dependent on the CO_2 flue gas concentration (0.61 for 16 vol% dry)

Table 15: Main parameters and assumptions for the economic assessment

Key economic parameters specific to each mill		
	Paper Mill (Belgium)	Pulp Mill (Sweden)
Electricity price, €/kWh	0.076[a]	0.045[b]
Operating hours, hours/year	8000	8400
Annual Labour cost, k€/year	60[c]	86[c]

Parameter	**Value**	**Unit**
Main process assumptions and parameters		
Capture Rate	90	%
Lignin scaling factor	0.93TCR	€
CAPEX[d]		
Plant lifetime	25	years
Discount rate	8	%
Construction time	2	years

Table 15 continued

From EC to TCR		
Total direct cost without process contingencies (TDC')[e]	EC+IC[e]	€
Total direct cost (TDC)	TDC'*1.10	€
Engineering, procurement, construction (EPC)	TDC*1.15	€
Total Plant Cost (TPC)	EPC*1.30	€
Total Capital Requirements (TCR)[f]	TPC*1.275	€
Overnight factor	1.18	-
Variable OPEX[g]		
AMP [26]	8	€/kg
PZ	6	€/kg
Additive	441.6	€/kg
Carbonate	0.46	€/kg
Cooling water[h]	0.039	€/m^3
Process water[h]	6.65	€/m^3
Wood Chips (for additional hot utility demand)	18	€/MWh
Fixed OPEX[h]		
Maintenance	2	%TPC
Labour	0.8	%TPC
Insurance and local property	2	%TPC
Administrative & support labour	30	%(O&M)
Operating Labour - Employees for capture	10	employees
Operating labour - Employees for conditioning	5	employees

[a] Median EU27 – Band IF 2020 [39]
[b] Sweden – Band IF 2020 [39]
[c] Labour cost levels for industry except construction: based on the Labour Cost Survey performed by Eurostat from [40] and includes compensation of employees plus taxes minus subsidies
[d] All NOAK cost factors assumed in this work (lifetime, discount rate and contingencies such as process, system, project and indirect cost) were taken from Table C.7 in [33]
[e] Equipment cost (EC) and installation cost (IC)
[f] Includes owner's cost, start up modification, start up spare parts and the overnight factor
[g] Obtained from literature (AMP/PZ [26] and wood chips [41])
[h] Also obtained from [33]

As for evaluating the cost of any additional equipment required for the integration of carbon capture with both mill, relevant cost factors are shown in Table 16. Note that all the cost are computed in $€_{2020}$. The cost of any additional heat exchangers required was estimated using Aspen Process Economizer Analyzer (APEA). In the case of the Pulp Mill, there is additional equipment cost depending on the heat integration scenario. For Scenario B and C, the additional hot utility demand is assumed to be covered by a biomass boiler using wood chips as a fuel. For Scenario C the cost of investing in additional turbine capacity is included in the TCR, in addition to investing in a biomass boiler.

Table 16: Equipment cost estimations (M$) for additional equipment required for integration

Equipment	Units for size calculation (S)	Cost function	Reference
Heat exchanger	Area of the HX (m^2)	Aspen Process Economizer Analyzer (APEA)	-
Biomass boiler (CFB)[a]	Heat output (MW_{th})	$C = 471\left(\frac{\dot{Q}_{MW_{th}}}{1521}\right)^{0.67}$	[42]
Turbine[b]	Power (kW)	$C = 15\,215 + 656S^{0.8}$	[43]

[a] Original equipment cost in $\$_{2021}$
[b] Original equipment cost in $\$_{2006}$

The equipment cost shown for the biomass boiler was originally developed by Martinez et al. [42], who used it to estimate the cost for a carbonator by approximating it as a circulating fluidized bed (CFB). Martinez et al. looked in literature for other cost functions and found that there was a lack of consistency between the equipment costs obtained using different cost functions reported, notably the scaling factor (shown as 0.67 in Table 16 above) varied significantly. Therefore, the cost function developed in their work is based on other reported cost functions but corrected using cost data from a realized project [44]. Whether this cost function yields a better approximation for the biomass boiler of this work than another is uncertain, adding to the uncertainty that already exists with the capital cost estimates for the capture and conditioning plant. Such uncertainty in the CAPEX will be addressed through a sensitivity analysis, describes in the subsequent section.

4.3 Sensitivity Analysis: Pulp Mill

Several assumptions were made in order to evaluate the economic performance of the scenarios previously identified, introducing a degree of uncertainty in the capture cost. These assumptions include the price of utilities (electricity, fuel, and cooling water), as well other raw materials (additive) and the NOAK cost factors utilized for capital cost estimations. The uncertainty lies in the fact that the future is uncertain where one cannot predict what will be the future price of electricity, nor the expected future cost of the capture technologies. Indeed, as carbon capture has never been demonstrated in the pulp and paper industry, it is not possible to accurately predict the cost of an n^{th} -of-a-kind deployment.

As a result of these uncertainties, two different sensitivity analyses were performed. One sensitivity analysis focused on the impact of future energy markets. For all scenarios and both technologies, the capture cost was evaluated for electricity prices between 20 and 300 €/MWh, at three different wood chip prices (9, 18 & 27 €/MWh). The motivation behind exploring a large range of electricity price is to demonstrate what the capture cost could be for a pulp located somewhere other than Belgium or Sweden. Following this, the lower and upper value were selected based on what has been observed in Europe over the past 10 years [45]. It is worth

noting that capture cost obtained from such an analysis would not be exactly representative of the capture cost for a mill in another location as other parameters which are dependent on location, such as labour cost, were kept constant.

The second sensitivity analysis focused on exploring what parameters have the most influence on the capture cost of the novel process. This was achieved by performing a single-variable sensitivity analysis where the capture cost was evaluated for different utility prices (electricity, fuel, and cooling water), % of annualized CAPEX and % of excess heat covering the SRD. This analysis was only performed for the novel process as excess heat availability varies between mill and will likely to be an important parameter in the capture cost of the novel process.

4.4 Integrated System: Changes in CO_2 Emissions

It has been previously stated that due to uncertainties regarding the rest of the CCS value chain, the cost of avoiding CO_2 has not been evaluated. Despite this, the author of this work believes that it is important to compare the heat integration scenarios not only regarding their economic feasibility (cost of capture) but also regarding their net CO_2 emissions. In the case of the Paper Mill, the loss in electricity generation, combined with the electricity demand of the capture and conditioning plant, result in indirect (fossil) emissions related to the import of electricity from the grid. The assumed emissions intensity for electricity in Belgium is shown in Table 17.

As for the Pulp Mill, the net CO_2 emissions of Scenario A, which is when fuel use is minimized, is similar to the integration of carbon capture with the Paper Mill. The mill would have additional indirect (fossil) emissions due to the need to import electricity from the grid. As for Scenario B, the net CO_2 emissions include the direct (biogenic) emissions due to the combustion of additional fuels used for steam production and the indirect (fossil) emissions from the electricity required for the capture and conditioning plant. Lastly in Scenario C, depending on the technology, the additional electricity gain may be enough to meet the electricity demand for capture and conditioning, and as well provide electricity for the other operations of the mill. If that is the case, this will result in a decrease in indirect fossil emissions as it would replace electricity provided by other generators. The upstream emissions associated with the additional biomass use in Scenario B and C (land-use changes, harvesting and transportation) have been neglected. The emissions intensity for electricity Sweden, as well as the emissions intensity of the fuel for the biomass boiler in Scenario B and C, is detailed in Table 17.

Table 17: Assumed data for calculating CO_2 emissions from electricity and biomass boiler

	Value	Unit
Belgium: electricity emissions intensity	230.70[a]	$kgCO_2/MWh$
Sweden: electricity emissions intensity	8.80[a]	$kgCO_2/MWh$
Wood chips	95[b]	$kgCO_2/GJ$

[a] EU27 emissions intensity used for Belgium and Sweden [46]
[b] Assumed wood bark with constant carbon content of 53.1 Carbon wt% (dry and ash free basis) from Table 5 in [47]

5 Results

5.1 Energy Required for the Integrated Mill and Capture System

5.1.1 Paper mill

The steam network of the integrated system (Paper Mill with carbon capture) is shown below in Figure 7. For both the benchmark and novel process, the condensing stage of the turbine TC2 is bypassed so that all the steam flow is passed through the 2.2 bar(g) extraction. Not all of the steam flow through the condensing stage is required to cover the reboiler duty of the capture plant in the case of the benchmark process, but the remaining steam flow in the turbine would be too low to operate the turbine stage. As for the novel process, all of the steam currently flowing through the condensing stage is needed to meet the reboiler duty.

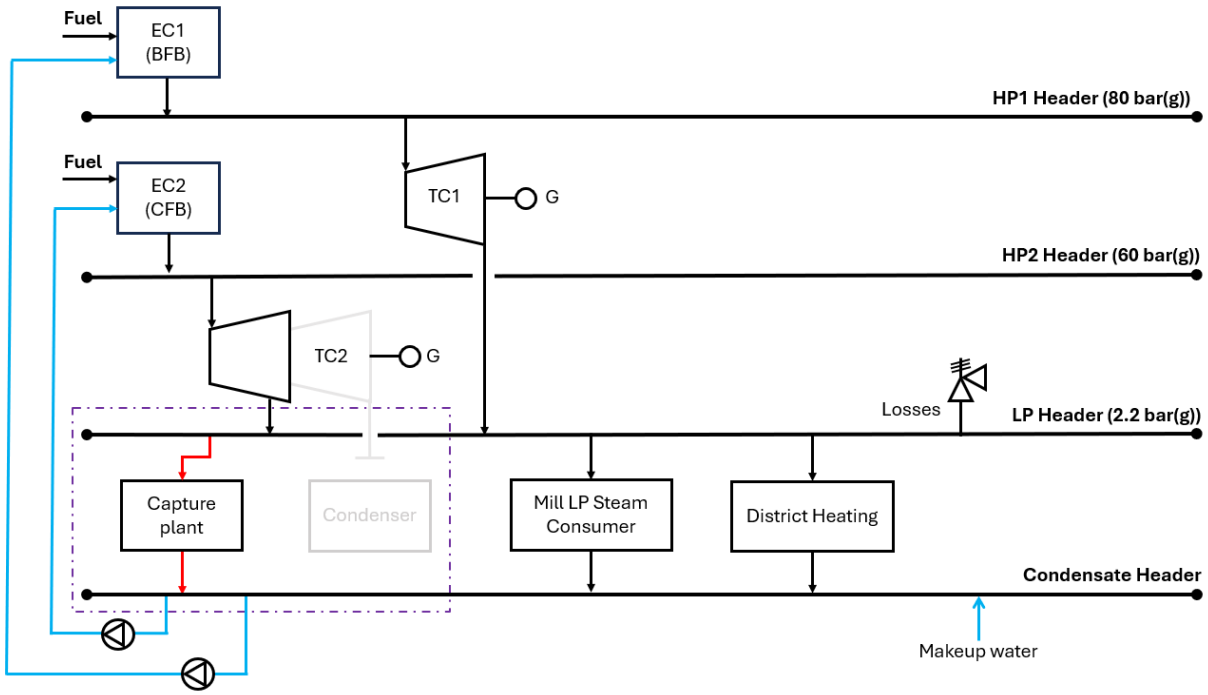

Figure 7: Modified steam network for the Paper Mill with carbon capture where the purple dashed box represents the main modifications to the system, which includes the bypass of condensing stage and the additional LP heat demand of the capture plant for both technologies.

Since the condensing stage is fully bypassed for both technologies, the loss in power generation is the same. Consequently, the specific cost for LP steam per tonne of CO_2 captured is the same despite their different SRDs. What sets the two technologies apart in this regard, and what is not reflected in the capture cost, is the excess heat (LP steam) remaining after integration. As detailed in Table 18, this is approximately 5 MW for the benchmark and 0 MW for the novel process. This would need to be cooled away as there is no use for this heat, thereby increasing the mill's cooling demand. Alternatively, this could used for district heating in the future,

providing an additional source of revenue to the Paper Mill. The integration of the benchmark process with the Paper Mill could have been optimized to avoid having additional excess heat leftover, for example reducing the fuel input. However, this would affect the capture plant as there would less CO_2 available for capture, requiring iterative re-modelling of the capture plant and steam system, therefore this was not further explored.

Table 18: Power production and excess heat availability for the integrated system (Paper Mill with carbon capture)

	Power produced (MWe)	Loss in power generation (MWe)	Heat cooled in Turbine Condenser (MW)	Excess LP Steam (MW)
Reference Paper Mill (no capture)	47.2	-	54*	-
Benchmark process & Paper Mill	34.9	12.3	0	4.9
Novel process & Paper Mill	34.9	12.3	0	0

*Approximate heat flow rejected to cooling tower based on the average annual values shown in Table 4.

As the temperature of the heat supply to the novel process is higher than 90°C, the LP steam extraction is used to produce hot water from 75°C to 85°C. Two heat exchangers (HX) were explored, one where the steam is condensed without subcooling and another with approximately 28 K of subcooling. The motivation behind exploring two different heat exchanger designs is due to the variation in excess steam available between seasons, where more steam is available during summer when there is no demand for district heating. The HX with 28 K subcooling respects the maximum available excess steam flow of the condensing stage of TC2 when the Paper Mill provides district heating.

Table 19: HX Specifications for heat exchange between mill and novel process

	HX Area (m^2)	Cost (M€)
HX no subcooling	418.2	0.27
HX 28 K subcooling	564.9	0.36

Despite the need for subcooling, the temperature of the feedwater after the heat exchanger would be higher than what it would be if the condensing stage were not bypassed. Given that there is already an existing steam demand for feedwater heating, it can therefore be assumed that the Paper Mill will not incur any additional cost for condensate pre-heating. The specifications of the two heat exchangers are detailed in Table 19, where it can be observed that the difference in cost between the two appears to be significant but was found negligible in the final cost estimate.

5.1.2 Pulp mill

The way in which the steam network of the Pulp Mill is affected by the integration of carbon capture depends on both the heat integration scenario (see Table 13 and Table 14) and on the capture technology. It is therefore difficult to comprehensively depict the modifications in steam network. In the case of the benchmark process, the reboiler duty is met completely by LP steam. As for the novel process, the heat integration results showed that approximately 38 MW excess heat (below approximately 100°C) could be used to meet the reboiler duty, with the remaining met by LP steam.

In scenario A, the high-temperature excess heat from the recovery boilers is not enough to meet the added heat demand of the capture process and maintain power production. Because of this, part of the high-temperature excess heat is utilized without co-generation of power. As shown in Figure 8, this power loss is lower when integrating the Pulp Mill with the novel process since it has a lower LP steam demand, thanks to the possibility to use low-temperature excess heat. Consequently, this is also why the novel process has a lower fuel demand compared to the benchmark process in Scenario B (24 MW vs. 5 MW). In this scenario, the Pulp Mill invests in a biomass boiler to ensure that there is enough heat to restore full power production and to meet the heating demands of integrated system. Furthermore, due to its lower LP steam consumption, the minimum heating demand of the mill once integrated with the novel process is lower compared to the benchmark process. This results in a smaller potential for electricity production in back-pressure steam turbines for the novel process in Scenario C (61 MW_e vs. 57 MW_e).

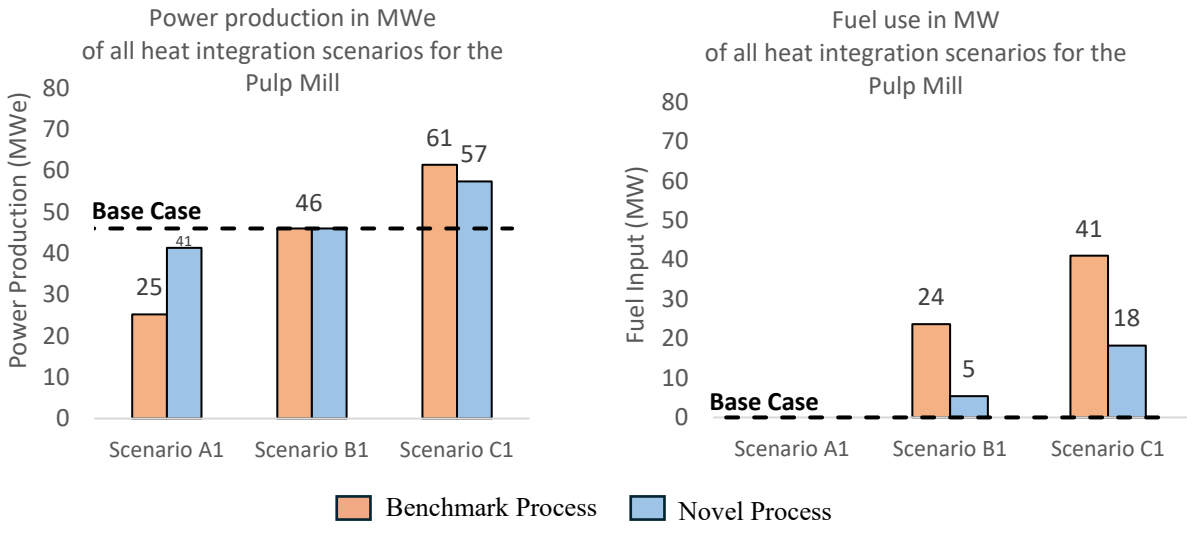

Figure 8: Power production (left) and fuel input (right) for the integrated system in all heat integration scenarios where Scenario A1-C1 refer to the mill without lignin extraction. The benchmark and novel process are shown in orange and blue, respectively. The electricity generation of the base case is 46 MWe where the fuel input is zero as there would be no need to increase fuel consumption without carbon capture.

When looking at the results for the Pulp Mill with lignin extraction in Figure 9, it can be observed that all the electricity production is lost for both capture technologies. What is not shown is that, in the case of the novel process, not all of the excess heat from the recovery boilers is required to meet the heat demand for capture, i.e. full flue gas heat recovery is not required. However, if the heat remaining were to be used for steam generation, the steam flow through the turbine would be below the minimum flow requirement for operating the turbine. Nevertheless, the greater excess heat availability from the recovery boiler reduces the amount of additional fuel needed to restore power production for the novel process compared to the benchmark process in Scenario B2 (55 MW vs. 36 MW).

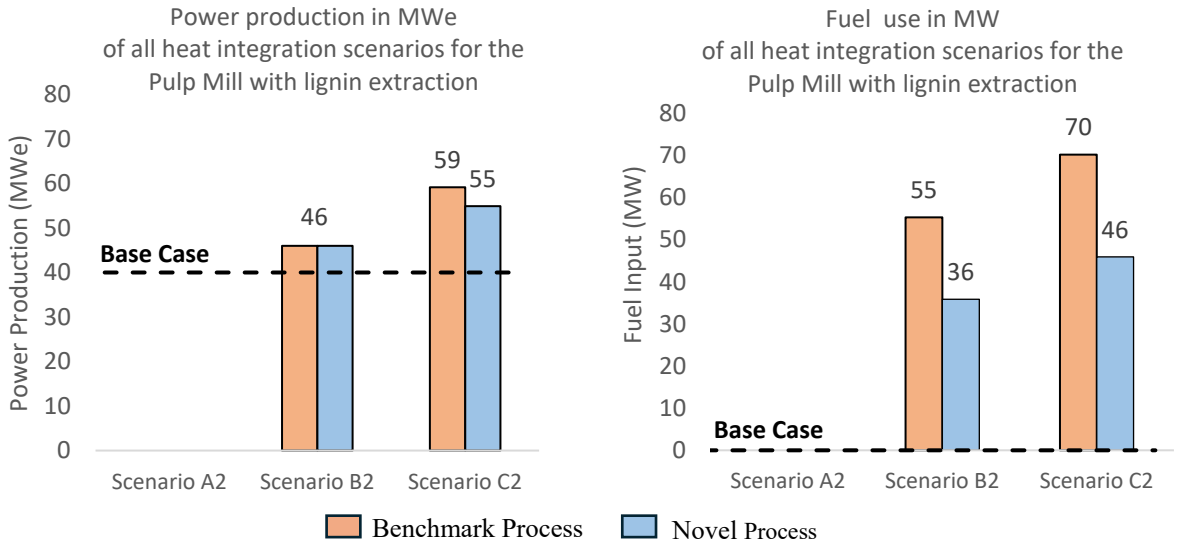

Figure 9: Power production (left) and fuel input (right) for the integrated system in all heat integration scenarios where Scenario A2-C2 to the mill with lignin extraction. Note that the power production of the base case for lignin extraction is lower than that of the power production for the mill without lignin extraction (40 vs. 46 MWe). As for the fuel input, this is zero as there would be no need to increase fuel without carbon capture.

One should also note that the power production in Scenario B2 is fully restored to what it could be in a mill without lignin extraction (46 MWe), resulting in a gain in electricity generation of 6 MWe. This increase in power generation could have been obtained even without the carbon capture integration, but would then require investment in a new boiler, with very small capacity and consequently high specific cost only to gain these few MW of electricity. In a way, the implementation of carbon capture on the mill with lignin extraction further justifies the investment in additional boiler capacity to restore full power generation as its more economically attractive to invest in a larger boiler (economy of scales).

Similarly to the Paper Mill, hot water is produced to supply heating to the novel process. However, two heat exchangers are required, one using a process stream as a heat source and another using LP steam. For simplification, the heat exchanger for recovering excess heat has been sized for recovering 72% of total actual excess heat recovered as this could be met by a single process stream. The specifications of the two heat exchangers are shown in Table 20, where the specifications for the heat exchanger for flue gas cooling has also been included. The flue gas cooler will be part of the capital cost for both technologies.

Table 20: HX specifications for excess heat recovery for the Pulp Mill

	HX Area (m^2)	Hot side	Cost (M€)
HX1 – Novel Process	387.6	LP steam	0.26
HX2 – Novel Process	484.8	Process Heat	0.29
HXFG – Flue gas cooler	381.6	Flue gas	0.75

5.2 Capture Cost

5.2.1 Impact of methodology and site differences

The economic performance of CO_2 capture cannot be directly compared in absolute numbers between the two sites, primarily since different methodologies have been applied to estimate effects on energy balances, but as well as because the sites are different. The impact that these differences have on the capture cost are apparent when comparing the share of the specific costs. As shown in Figure 10, variable OPEX for electricity (which covers electricity demand as well as loss in electricity generation) makes up the largest share of the capture cost for the benchmark process for both sites. For the Pulp Mill (Scenario A1), however, this cost is considerably smaller, contributing 21.4 €/tonCO_2 to the capture cost compared to 12.5 €/tonCO_2 for the Paper Mill (29% decrease). This is partly because the energy balance of the Pulp Mill is assumed to be more favourable for implementing carbon capture. Specifically, the energy balance is more favourable as maximum internal heat recovery was assumed when performing the heat integration, resulting in greater LP steam availability for capture. In addition, the impact of site differences, specifically geographical location, also contribute to a lower share in variable OPEX for electricity, as the price of electricity is lower in Sweden compared to Belgium. Site differences play a role in other aspects of the capture cost, such as the share of CAPEX for the capture and conditioning plant (and thereby fixed OPEX). This can be attributed to one of the most apparent differences between the two mills, which is the amount of CO_2 captured. The results show a clearly lower specific CAPEX for the Pulp Mill thus demonstrating the effect of economies of scale.

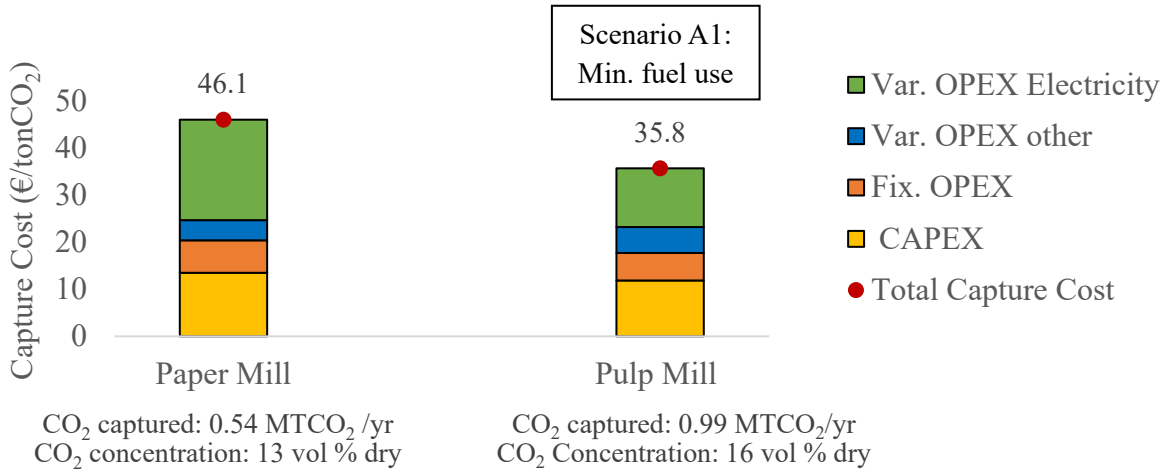

Figure 10: Breakdown of capture cost of the benchmark process for Paper Mill (left) and Pulp Mill (right) where the electricity is priced at approximately 80 €/MWh for the Paper Mill and 50 €/MWh for the Pulp Mill. Var. OPEX electricity includes the loss in electricity generation due to the use of LP steam and the electricity demand for capture and conditioning. Var. OPEX other includes materials such as process water, solvent, and cooling water. Conditioning includes the CAPEX and OPEX of the conditioning plant.

The economic performance of the benchmark capture process should not be directly compared to the performance of the novel process as energy requirements and capital costs were obtained from different sources, which might not be consistent in modelling assumptions. However, it is interesting to evaluate how the characteristics of the technologies affect the impact site differences can have on the capture cost. Notably, the influence of low-temperature excess heat availability on the economic performance of the novel process can be observed when comparing the cost of variable OPEX electricity of the two sites.

Unlike the Paper Mill, there is low-temperature process heat available in the Pulp Mill which can be used to meet part of the novel process's reboiler heat demand. As a result, the decrease in the share of the cost of variable OPEX electricity between two sites is even larger, see Figure 11. An approximate 57% decrease can be observed (from 29 €/tonCO_2 to 11 €/tonCO_2) for the novel process, compared to the 29% decrease observed for the Benchmark process (Figure 10). With regards to the other components of the capture cost, the same trend can be observed where the share of the specific CAPEX and fixed OPEX cost is lower in the Pulp Mill due to economy of scales. Contrary to the benchmark process, there is also an observed decrease in the other variable OPEX (includes cooling water demand and solvent consumption). This can be attributed to a lower solvent consumption per ton of CO_2 captured in the Pulp Mill, and as well as a lower specific consumption of cooling water per ton of CO_2 captured. The latter being because the cooling demand of the mill is reduced by recovering some low-temperature excess process heat, which was considered in the analysis by minimizing the cooling demand of the capture process.

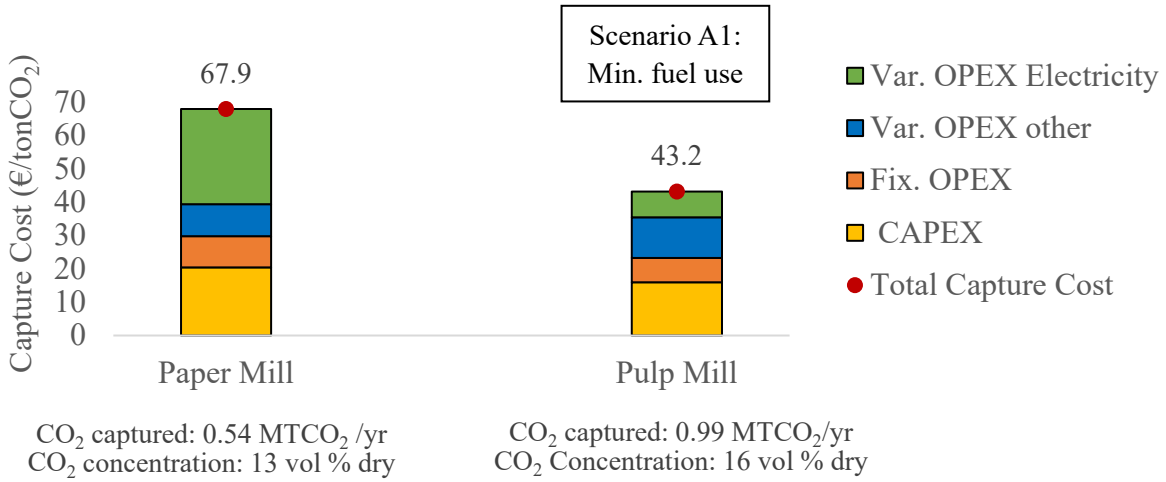

Figure 11: Breakdown of the capture cost of the novel process for the Paper Mill (left) and Pulp Mill (right) where the electricity is priced at approximately 80€/MWh for the Paper Mill and 50 €/MWh for the Pulp Mill. Var. OPEX electricity includes the loss in electricity generation due to the use of LP steam and the electricity demand for capture and conditioning. Var. OPEX other includes materials such as process water, solvent, and cooling water. Conditioning includes the CAPEX and OPEX of the conditioning plant.

5.2.2 Impact of heat integration scenarios

The breakdown of the capture cost of the benchmark process for the heat integration scenarios are shown in Figure 12 for the Pulp Mill without lignin extraction. In Scenario B1 and C1, it can be observed that there are two additional cost components in the capture cost. For Scenario B1, there is an additional CAPEX investment (shown in grey) for a biomass boiler with a corresponding variable OPEX fuel cost (shown in dark blue). For Scenario C1, the share of these two specific cost components is even larger. Not only because the load of the biomass boiler is larger but also because of the investment in additional turbine capacity. Note that the mill is not assumed to have any spare capacity in existing biomass boilers. In many mills, however, it can be expected that there is some capacity available to increase the load in existing boiler and thereby avoid additional CAPEX. Another notable difference between Scenario B1 and C1 is the share of the specific cost in variable OPEX electricity. The gain in electricity production in Scenario C1 is greater than the electricity required for capture and conditioning, resulting in a negative specific variable OPEX electricity cost. Despite these additional investments, the capture cost of Scenario B1 and C1 are only slightly larger than the capture cost of Scenario A1, indicating that the scenarios perform similarly in a low electricity price market.

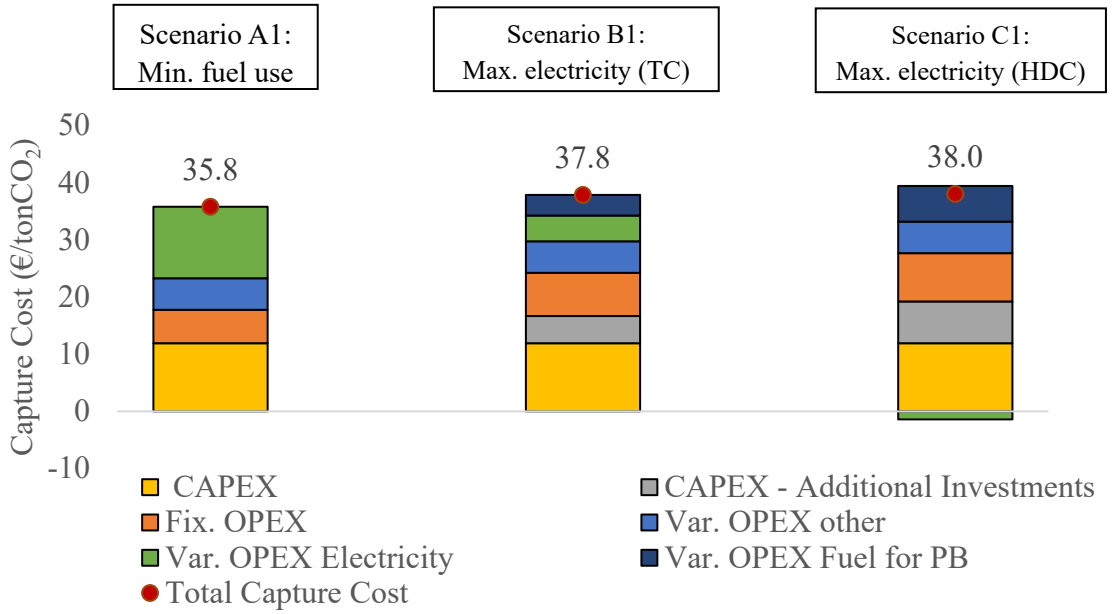

Figure 12: Benchmark process: breakdown of capture cost of the different heat integration scenarios for the Pulp Mill with lignin extraction where the Var. OPEX other includes the cost of materials (process water and solvent) and cooling water. Note that variable OPEX for electricity is negative in Scenario C1.

The breakdown of the capture cost of the novel process for the different heat integration scenarios is shown in Figure 13. The use of low-temperature excess heat minimizes the need for a utility boiler and therefore the share of specific additional CAPEX investment and variable OPEX fuel cost is smaller than what was observed for the benchmark process (Figure 13 above). As for Scenario C1, the electricity gained from maximizing back-pressure power is not enough to cover the electricity demand for capture and conditioning. What contributes the most to the cost of variable OPEX electricity is the capture technology's own high electricity demand. Similarly to the benchmark process, the heat integration scenarios perform similarly in a low electricity price market. It is worth noting as well that, although the two technologies cannot be directly compared as their energy requirements (cooling, electricity and reboiler heat) were obtained following different simulation methodologies, it can be observed that they both perform similarly in a low electricity price market.

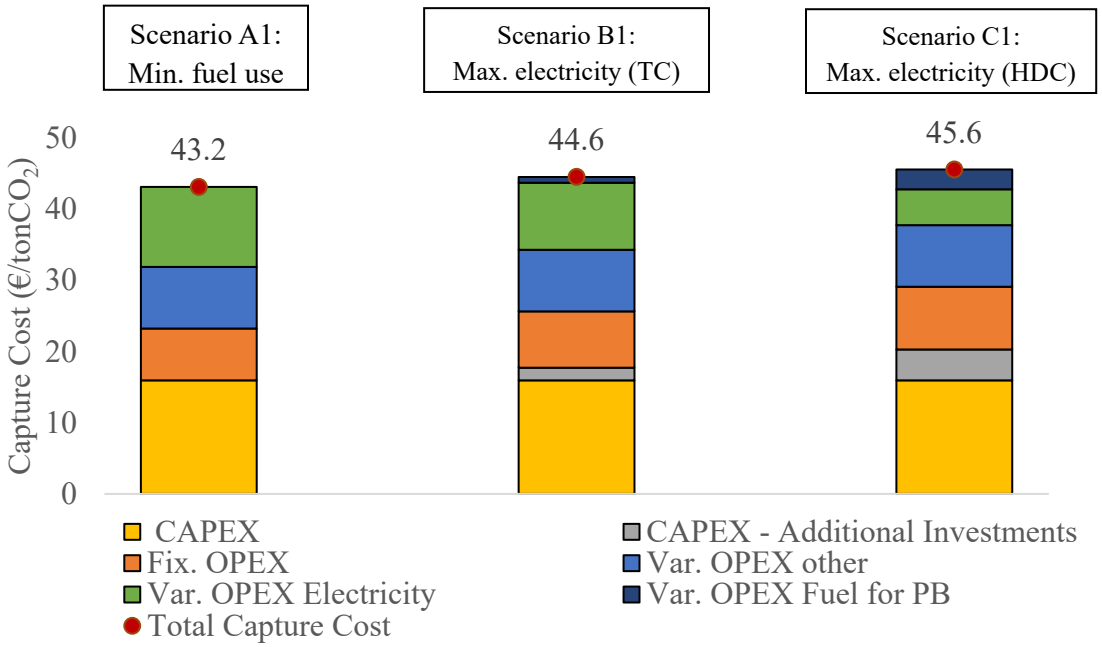

Figure 13: Novel process: breakdown of capture cost different heat integration scenarios for the Pulp Mill where the Var. OPEX other includes the cost of materials (process water and solvent) and cooling water.

For the Pulp Mill with lignin extraction, the capture cost for the different heat integration scenarios was only evaluated for the benchmark process. As shown in Figure 14, the capture cost increased by approximately 10 €/tonCO_2 for all the scenarios. This increase can be explained by the reduction in high-temperature excess heat from the recovery boilers, in turn creating a less favourable energy balance for heat integration with carbon capture. As was shown in Figure 10, this results in a complete loss in electricity generation and therefore, the specific cost for variable OPEX electricity is much larger in Scenario A2 compared to Scenario A1. Consequently, this also results in a higher biomass load required in Scenario B2 and C2, as shown by the larger specific cost for additional CAPEX investments and variable OPEX fuel.

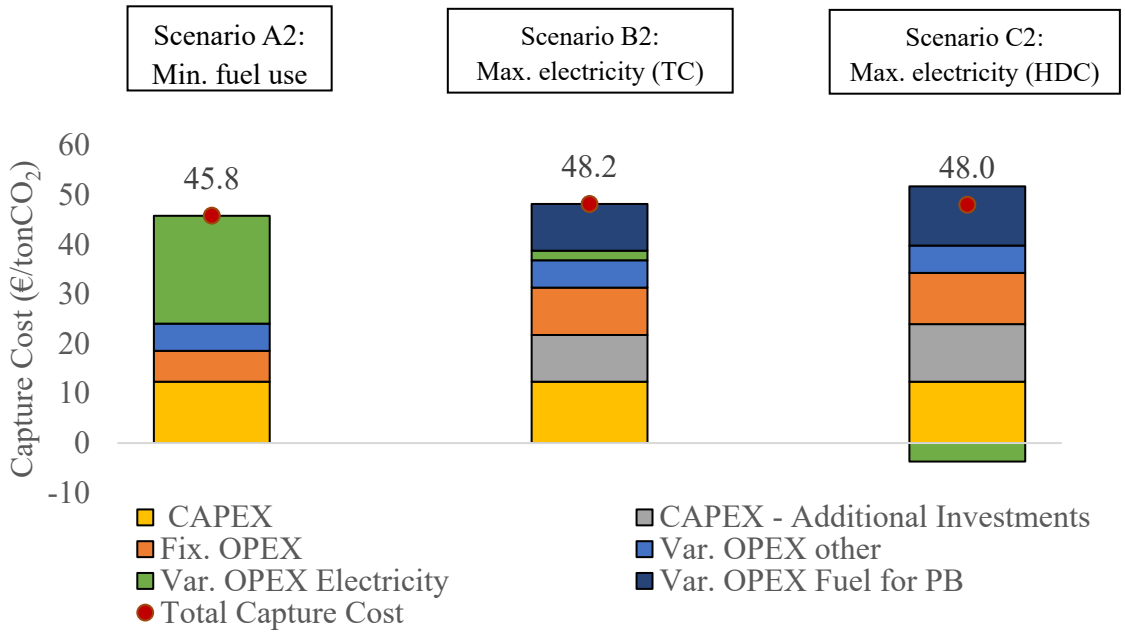

Figure 14: Benchmark process: breakdown of capture cost for different heat integration scenarios in a Pulp Mill with lignin extraction. Note that variable OPEX for electricity is also negative in Scenario C2.

While it costs more to capture CO_2 for the Pulp Mill with lignin extraction compared to the Pulp Mill without lignin extraction, it is worth noting that the analysis does neither consider the economic value of lignin extraction nor the potential climate benefit of substitution effects from lignin products. The results simply illustrate that the cost of carbon capture implementation is clearly favoured by favourable mill energy balances.

5.2.3 Impact of varying energy market conditions

The influence of electricity and fuel price on the capture cost for the benchmark process for the two pulp mills (with and without lignin extraction) can be observed in Figure 15. Focusing first on the Pulp Mill without lignin extraction, it can be observed that Scenario A1 is quite sensitive to changes in electricity price compared to other scenarios, as indicated by its slope. This is to be expected, given that in this scenario, the mill has lost almost half of its electricity production. Above an electricity price of approximately 80 €/MWh, the economic performance of all three scenarios is no longer comparable. When the price of electricity is high, it becomes worthwhile to invest in a biomass boiler to meet the heat demand of the capture process and maintain power production (Scenario B1). For Scenario C1, the capture cost decreases with an increasing price for electricity, achieving the lowest capture cost among the scenarios at approximately 60 €/MWh and above. The decreasing trend is due to the additional gain in electricity production, which resulted in a negative specific variable OPEX electricity cost (see Figure 12).

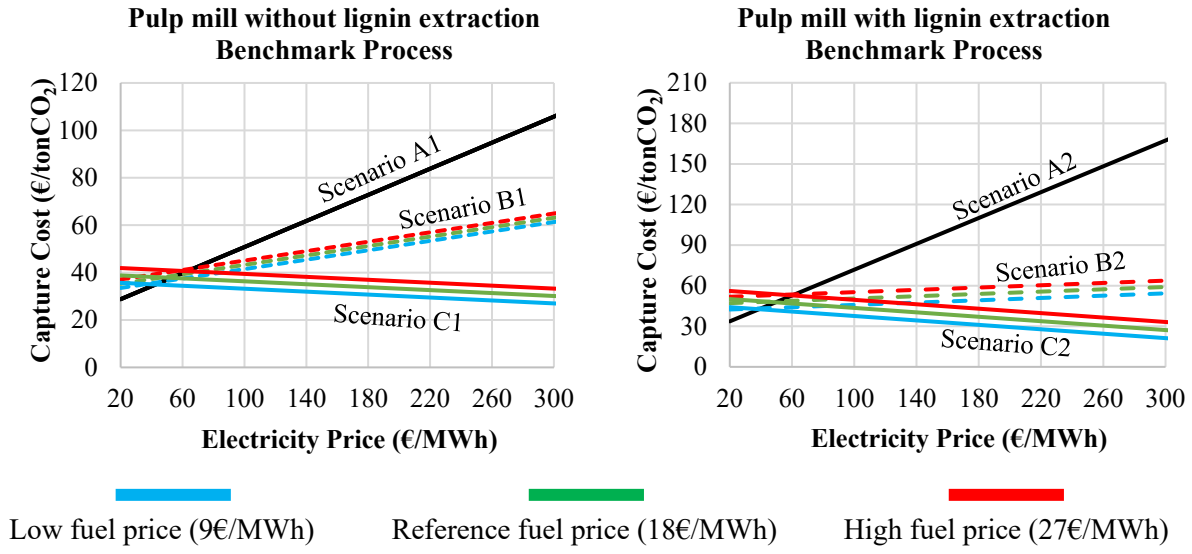

Figure 15: Sensitivity analysis on electricity and fuel prices for the capture cost of the benchmark process, where Scenario A (shown in black) is unaffected by changes in fuel prices since there is no additional fuel use. Recall that Scenario A is when fuel use is minimized, Scenario B is when electricity generation is maximised but constrained by existing turbine capacity and Scenario C is when (back-pressure) electricity is maximized but constrained based on the minimum heat demand.

Similar trends can be observed for the Pulp Mill with lignin extraction, shown on the right of Figure 16. Since all the electricity production is lost, Scenario A2 has higher sensitivity to variation in electricity price compared to Scenario A1. The opposite trend can be observed for Scenario B2, where the additional gain in electricity is almost enough to cover the electricity demand of the capture and conditioning plant, nearly removing the capture cost's dependency on electricity price. This gain in electricity being the 6 MW lost due to lignin extraction.

What can also be observed in Figure 15 is that the capture cost of the benchmark process appears to be affected minimally by changes in fuel price compared to changes in electricity price. This is seen by the proximity of the three different fuel price lines where red is a high fuel price, green is the reference fuel price and blue is a low fuel price. The fuel price was not varied as much as the electricity price, however comparing the impact that the same variation (+/- 50%) has on the capture cost confirms that the electricity price is indeed more influential. For example, in Scenario B1, going from 120 €/MWh to 180 €/MWh, the capture cost increases approximately by 5 €/tonCO_2. The capture cost from the reference fuel price to a higher fuel price result in less than 1 €/tonCO_2 increase. A higher dependency on fuel price can be observed in Scenario C (shown by the larger gap between the different fuel prices), which is to be expected given that this scenario has a larger additional fuel consumption compared to Scenario B.

The influence of energy market conditions on the heat integration scenarios for the novel process was also explored, as shown in Figure 16. All scenarios appear to be quite sensitive to variations in electricity price, as indicated by their slope. Scenario A1 and B1 perform quite similarly with B1 only achieving a slightly lower capture cost at around 100 €/MWh and above. This can be explained by the balance between what is lost in electricity generation (due to LP steam use) and what the capture technology consumes, where the latter is greater. Consequently, Scenario C1 performs the best in a high electricity price market (≈ 80 €/MWh and above) as the additional gain in electricity can be used to meet part of the electricity demand for capture and conditioning. Due to the minimal loss in electricity generation (and thereby less additional fuel consumption), compared to the benchmark process, the heat integration scenarios for the novel process are less influenced by variations in fuel price. This can be observed by comparing the proximity of the fuel price lines.

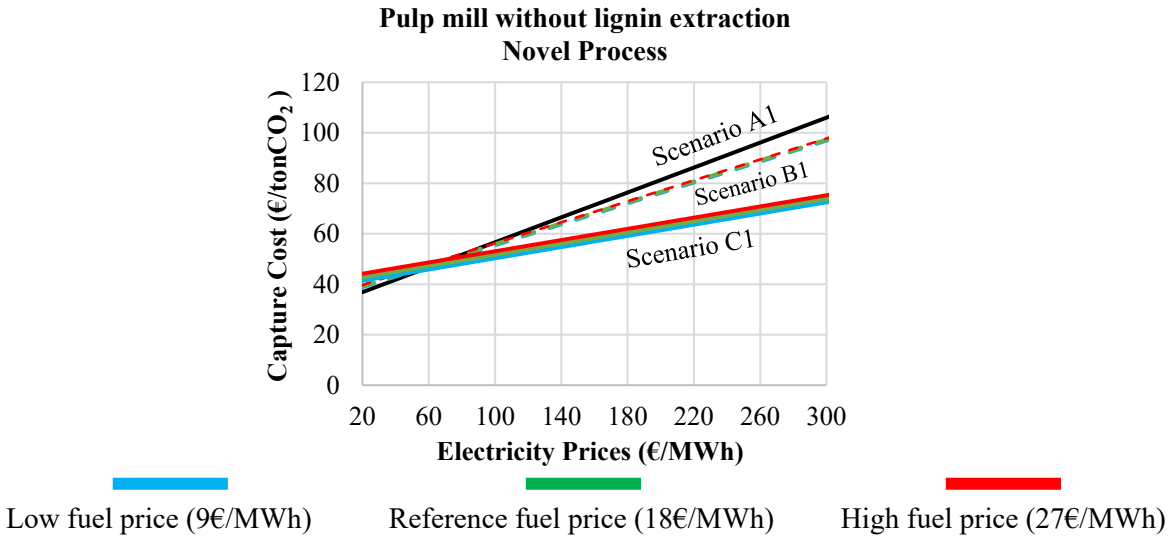

Figure 16: Sensitivity analysis on electricity and fuel prices for capture cost of the novel process, where Scenario A1 (shown in black) is unaffected by changes in fuel prices since there is no additional fuel use. Scenario A1 is when fuel use is minimized, Scenario B1 is when electricity generation is maximised with constraints on turbine capacity and Scenario C1 is when electricity is maximized with constraints based on heat demand.

5.3 CO_2 Emissions After Integration

The remaining CO_2 emissions after the implementation of carbon capture in the Paper Mill and the two pulp mills (with and without lignin extraction) is detailed in Table 21. What sets the heat integration scenarios apart are the additional biogenic emissions from the biomass boiler and the indirect (fossil) emissions from the change in electricity production. In Scenario A, indirect fossil fuel emissions from the grid are the highest. In Scenario C, biogenic emissions are the largest, and correspondingly indirect fossil emissions are the smallest. This scenario also happens to be the most economically feasible when the price of electricity is above around 60 €/MWh (from Figure 15).

Table 21: Resulting CO_2 emissions (in ton/hr) from integrating the Pulp Mill (with and without lignin extraction) with carbon capture. . In the case of Scenario C, the additional electricity produced would reduce the need to import electricity from the grid, thereby leading to a decrease in indirect fossil emissions (shown as negative for the benchmark process).

Pulp Mill without lignin extraction				
	Base Case	Scenario A1	Scenario B1	Scenario C1
$CO_{2,\ emitted\ (biogenic)}$	131.32	13.13	13.13	13.13
$CO_{2,\ captured\ (biogenic)}$	-	118.19	118.19	118.19
$CO_{2,\ biomass\ boiler\ (biogenic)}$	Benchmark	0	8.07	14.00
	Novel	0	1.84	6.22
$CO_{2,\ \Delta\ electricity\ (fossil)}$	Benchmark	0.29	0.10	-0.03
	Novel	0.26	0.22	0.12

Pulp Mill with lignin extraction				
	Base Case	Scenario A2	Scenario B2	Scenario C2
$CO_{2,\ emitted\ (biogenic)}$	117.52	11.75	11.75	11.75
$CO_{2,\ captured\ (biogenic)}$	-	105.77	105.77	105.77
$CO_{2,\ biomass\ boiler\ (biogenic)}$	Benchmark	0	18.87	23.96
	Novel	0	12.23	15.67
$CO_{2,\ \Delta\ electricity\ (fossil)}$	Benchmark	0.45	0.04	-0.07
	Novel	0.56	0.15	0.08

The total CO_2 emissions of the Pulp Mill with lignin extraction are lower than without lignin extraction as more carbon from the feedstock is converted into a product (lignin). Less CO_2 is therefore captured by the capture plant but the overall CO_2 emissions with capture are lower than for the Pulp Mill without lignin extraction (comparing Scenario A1 to A2). However, this effect is negated when comparing Scenario B and C, as there is a greater demand for the biomass boiler (therefore greater CO_2 emissions) due to the unfavourable energy balance of the mill.

The emissions from electricity in the Pulp Mill are estimated based on yearly electricity production and annual average CO_2 intensity of grid electricity (for Sweden). Consequently, this does not take into consideration marginal effects on electricity generation in the grid. The CO_2 intensity of grid electricity varies between countries and regions, making it difficult to state that one scenario is better than the other when comparing their net CO_2 emissions after capture. Indeed, whether it is better to accept the loss in electricity or to utilize more biomass to restore power generation is very much dependent on the system in which the mill operates. It could be argued that in countries where coal is on the margin, it would be better to restore power generation as it would avoid increasing indirect fossil fuel emissions, at the cost of emitting biogenic CO_2. On the other hand, as countries move towards a decarbonized economy, an emissions intensity similar to Sweden is likely to be norm in the rest of Europe, and therefore the argument for avoiding indirect fossil emissions from the grid will no longer be valid.

It is also important to consider substitution effects, where the additional use of biomass resources for electricity production would prevent it from being used for other applications such as production of biofuels or bio-based products. Another key aspect to consider is the amount of additional biomass available that could be consumed by the mill, something that also may be limited by an increased used of biomass for new applications, but also depends on the region where the mill is located. Specifically in Sweden, Karlsson et al. [48] found that the Bio-energy Carbon Capture and Storage (BECCS) potential in the Swedish pulp and paper industry would be limited in regions where there is competition for logging residues. An important takeaway here is that the implementation of carbon capture not only affects the mill's operation, but also the system in which the mill operates.

This effect of emissions intensity can be observed when comparing the emissions of the Paper Mill with carbon capture to that of Scenario A1 for the Pulp Mill, see Table 22. The electricity required includes what was lost in electricity generation due to LP steam consumption and the electricity demand of the capture and conditioning plant.

Table 22: Comparing CO_2 emissions (kg/hr) from the electricity required after integrating carbon capture in the Pulp Mill and the Paper Mill. The Paper Mill is located in Belgium (assumed electricity emissions intensity of 230.70 kgCO_2/MWh) and the Pulp Mill in Sweden (assumed electricity emissions intensity of 8.0 kgCO_2/MWh).

		Paper Mill	Pulp Mill (Scenario A1)
Electricity required (MW)	Benchmark	19	33
	Novel	25	29
$CO_{2_{\Delta\ electricity}}$	Benchmark	4,382	286
	Novel	5,850	257

5.4 Key Parameters Identified

The sensitivity analysis performed in Section 5.3 showed that the capture cost varies more with varying electricity prices than with varying fuel prices, indicating that the price of electricity is an important parameter in the economic analysis. Other influential parameters that have been identified include the capital cost estimates, excess heat availability and the other variable operational costs (cooling water and solvent).

The magnitude of the impact of these parameters on the capture cost of the novel process is shown in Figure 17, where only the capture cost of Scenario A1 and B1 were investigated. It is important to note that the sensitivity analysis performed is a single-variable analysis where one parameter was varied while the others were kept constant at their reference value (see Table 15). In the case of the price of electricity, fuel, and cooling water, as well as the annualized CAPEX, the minimum and maximum were chosen to be within reasonable limits according to historical data or assumptions used in literature. As for the cost of the additive, this is particularly uncertain as there is limited data available to the author's knowledge. A large span was therefore selected due to this cost uncertainty. It is worth noting that that the reference value for the annualized CAPEX is already an optimistic estimate, neglecting some potential additional costs (see Section

6.2). Consequently, the uncertainty span is strongly skewed towards higher CAPEX. Note also that this analysis was not performed for the benchmark process; however, the same could be said regarding its annualized CAPEX.

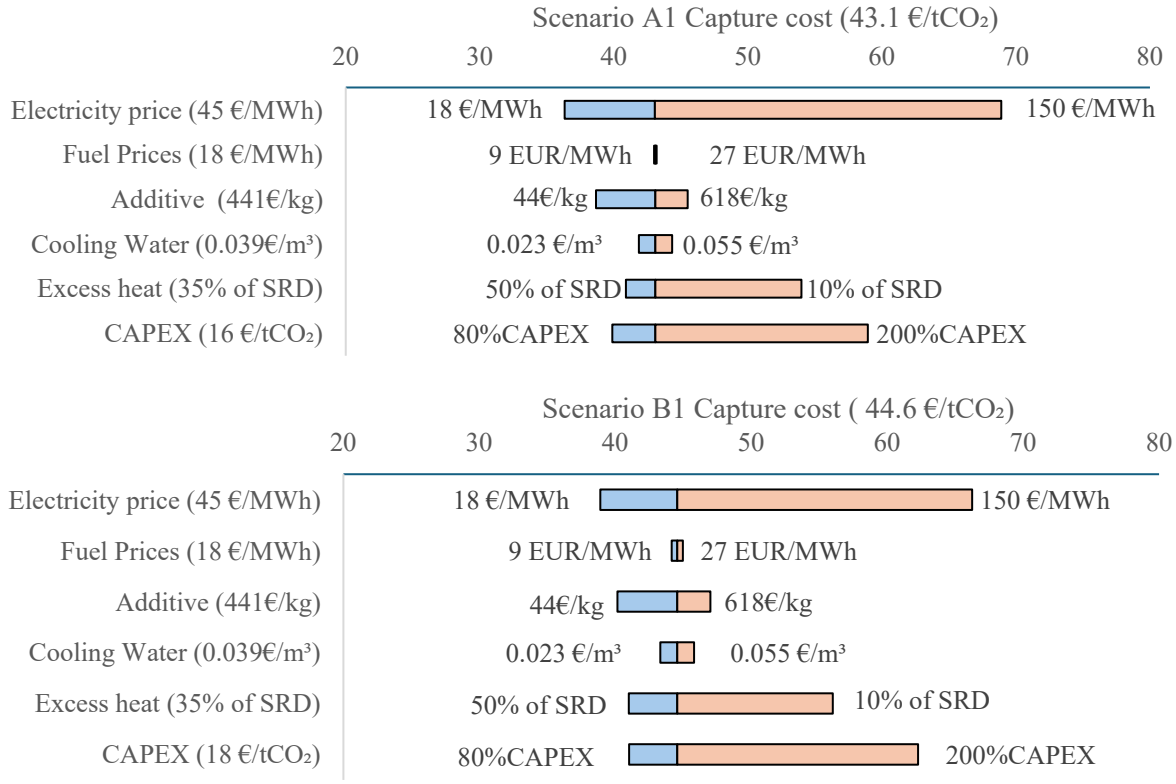

Figure 17: Single-variable sensitivity analysis on capture cost of the novel process with the reference values for the varied parameters shown in brackets on the left-hand side.

For both Scenario A1 and B1 increasing annualized CAPEX had a considerable effect on the capture cost, with a greater impact on Scenario B1 This is expected given that in Scenario B1, the CAPEX is higher due to the investment in a biomass boiler. Following that same logic, a change in the annualized CAPEX of Scenario C1 would result in a larger increase in the capture cost, given that additional CAPEX investments were even larger due to greater biomass boiler load and the additional turbine capacity. Overall, a 200% increase in annualized CAPEX, increased the capture cost of all scenarios by over 35%.

Unlike the other parameters investigated, the maximum % of SRD that can be covered by excess heat is directly limited by the Pulp Mill processes. Changes in excess heat were modelled by altering the heat load of the process streams in the mill that were assumed available for heat recovery. A clear limit exists, where an increase in excess heat availability is no longer reflected in the capture cost. With respect to Scenario A1, this limit occurs when the amount of excess heat used by the novel process minimizes the use of LP steam enough to restore all power generation. After this, due to limits imposed on power capacity, there is no further gain where the capture cost would remain constant. Similarly in Scenario B1, the biomass boiler size is dependent on

the excess heat availability where there would be no need for additional hot utility at a certain percentage of excess heat covering the SRD, since there is enough heat available from the recovery boilers for full power production.

It is difficult to say whether the excess heat availability would indeed be higher or lower for other pulp mills in Europe. However, in the future, it is likely that low-temperature excess heat will decrease as mill's invest in methods for improving their energy efficiency. On the other hand, better energy efficiency could result in LP steam surplus, which could still be used for meeting the heat demand of the capture process while equally generating power, should the mill choose to invest in additional turbine capacity. The way in which excess heat availability is analysed, i.e. how much excess heat a pulp mill is found to have, is also largely dependent on the methodology followed, where in this work, a highly theoretical approach has been followed.

There appears to be a slightly greater dependency between excess heat availability and the capture cost for Scenario B1 compared to Scenario A1. Less excess heat results in a greater demand in LP steam and therefore a greater loss in power production. In the case of Scenario B1, this means that the size of the biomass boiler (capital investments) and the fuel (operational costs) required to restore power production will be greater. The downside to a single-variable sensitivity analysis is that it does not capture the relationship between parameters. For example, at a higher electricity price, it is likely that excess heat availability would have greater influence on the capture cost of Scenario A1. This follows what was found in the sensitivity analysis performed in Section 5.2.3.

The impact of excess heat availability on Scenario C1 was not investigated but a decrease in excess heat would result in a greater use of LP steam. This would further increase the minimum heating demand of the mill, leading to greater potential in additional back-pressure electricity but a corresponding larger additional CAPEX investment due to the larger load of the biomass boiler and size of turbine. As for increasing excess heat availability, the same limit exists as was observed in Scenario A1 and B1. At a certain percentage of excess heat availability, there would be no need for a biomass boiler. This is because there is enough heat from the recovery boilers for full power production. In fact, due to the additional heat gain from flue gas cooling, the electricity generated would be greater than the base-case (46 MWe).

Variations in cooling water price and fuel price appear to have minimal effect on the capture cost compared to the other parameters analysed. As was previously stated, there is a stronger dependence on fuel price for Scenario C1 as this scenario has a higher fuel consumption than Scenario B1. This likely also explains why variations in electricity prices in Scenario C1 do not affect the capture cost as much as in Scenario A1 and B1 (confirmed by the slope of the capture cost of Scenario C1 in Figure 16). What has a greater influence on the capture cost compared to fuel and cooling water is the price of the additive. In this study, the cost of the additive was assumed but it is likely that it will decrease due to a scale up in production from a greater demand.

6 Discussion and Suggestions for Further Work

6.1 Flexible Operation

For the benchmark process, the results of the sensitivity analysis on electricity and fuel prices indicates there is a potential for flexible operation between heat integration scenarios, see Figure 15. For example, a mill owner could choose to operate in Scenario B when the price of electricity is high and in Scenario A when the price of electricity is low. Although Scenario C can provide a lower capture cost in a high electricity price market, this scenario carries a greater investment risk compared to Scenario B. If the price of biomass were to increase significantly, the pulp mill owner would likely cease the operation of the additional biomass boiler and turbine, resulting in greater number of stranded assets compared to Scenario B. The results of the sensitivity analysis for the novel process further indicate that such flexibility is only feasible if the performance of the capture technology differs significantly between heat integration scenarios.

For a Pulp Mill with lignin extraction, one could potentially investigate flexible lignin extraction based on excess heat availability. For example, when the mill's process heat demand is high, such as in the winter months, the lignin extraction rate could be reduced to minimize the cost of capture. Alternatively, one could optimize lignin extraction based on the energy market such that when the price of electricity is high, the lignin extraction rate could be reduced to minimize the losses in electricity generation. The future willingness to pay for biogenic CO_2, whether this is from a demand for negative emissions or from a carbon capture and utilisation (CCU) market such as for electrofuels, will be in large part be the deciding factor on whether to implement carbon capture or lignin extraction. As market demand for both are currently uncertain, it will be interesting to see in the future how they compare.

As for mills with unfavourable energy balances, such as integrated pulp and paper mills, partial capture may be more cost-effective option. Biermann et al. studied partial carbon capture by absorption and found that, depending on the market conditions, lower specific costs (€ per tonne of CO_2 captured) could be achieved compared to full capture [49]. Whether this would indeed be the case for integrated pulp and paper mills would need to be further explored.

6.2 Methodology Limitations

Several assumptions and simplifications were made when completing the TEA, resulting in uncertainty in the capture costs reported in this work. In the whitepaper for improved CCS cost estimation [37], the authors state that a bottom-up cost engineering approach, such as the one followed in this work, cannot provide an accurate prediction for the expected future cost of a carbon capture technology which has not yet reached commercial availability. This is the case for both the benchmark and novel process. It is worth noting that the low technological maturity also introduces uncertainty in the energy requirements as they were obtained from modelling and simulations rather than real operational data. Alternatively, the authors of the whitepaper recommend estimating capital cost by using learning rates from another technology that is similar. If there is no such technology, one can either use expert judgments or general characteristics from historical learning rate data for guidance (see pg. 46-47 of [37]). Whether such an approach could provide a better cost estimate is uncertain but may be worth exploring for future work.

The economic assessment performed also did not consider the full cost for heat recovery, nor site specific costs such as cost of retrofitability (i.e. flue gas piping) and spatial constraints. Quantified by Kumar in [50], foregoing the cost of retrofitability and spatial constrains introduces a considerable degree of uncertainty in the final cost estimates. Nevertheless, the aim of this work was not to provide detailed CO_2 capture cost estimates, but rather to propose a methodology for evaluating how certain site and technology characteristics could influence the economic performance. The reader should therefore not focus on the absolute numbers reported as they are likely to be optimistic estimates.

The last degree of uncertainty from the methodology comes from the use of a pinch-based energy targeting approach for evaluating the heat integration potential between the capture technologies and the Pulp Mill. Specifically, the amount of high- and low-temperature excess heat available to meet part of the capture heat demand of the novel process is uncertain as it was obtained using a theoretical approach. Given that the results in this work indicate this is a key parameter for achieving cost-effective carbon capture, how much excess heat is actually available for recovery in the Pulp Mill should be further investigated.

6.3 Bio-CCS Beyond Site Level

This work focused on the impacts that carbon capture has on the pulp and paper industry at a site-level where the impacts outside this boundary were not addressed in detail. Namely, challenges which could limit or prevent the implementation of bio-CCS such as the absence of policy or other financial incentives (e.g. voluntary markets for carbon removal credit) have not been addressed in this work.

The exact cost of implementing carbon capture in the pulp and paper industry may be uncertain but it is for certain that a mill owner will incur a cost. Without financial incentives for carbon removals or biogenic carbon pricing, such an implementation is unlikely to occur. It would be interesting to explore in future work what carbon removal credit price would make bio-CCS profitable. In addition, one could also explore whether policy could have influence on how carbon capture should be implemented in a pulp mill. For example, which heat integration scenario would be best if the pulp mill owner wanted to obtain carbon removal credits for capturing biogenic CO_2? Although all scenarios capture the same amount of CO_2, any on-site biogenic CO_2 emissions that are not captured or indirect emissions of fossil CO_2 due to increased demand for grid electricity could reduce the total negative emissions the pulp mill owner could claim. In addition to lack of policy, there are other challenges which prevent the deployment of bio-CCS in the pulp and paper industry in Europe such as biomass availability, social acceptance, cost, and availability of infrastructure for CO_2 transport and storage. Addressing these challenges was not the focus of this work but should be kept in mind when implementing bio-CCS.

7 Conclusions

A techno-economic assessment has been performed to estimate the CO_2 capture cost when integrating two different post-combustion CO_2 capture technologies with a Paper Mill and a Pulp Mill. The two capture technologies explored were an amine-based PCC technology (benchmark process) and a carbonate-based PCC technology (novel process). For the Pulp Mill, three different heat integration scenarios were explored: Scenario A (minimizing fuel use), Scenario B (maximizing electricity – turbine constrained) and Scenario C (maximizing electricity – heat demand constrained). The impact that future strategic developments for improvement utilisation of biomass resources may have on the implementation of carbon capture was investigated by evaluating the capture cost for a Pulp Mill with lignin extraction. The estimated capture costs are uncertain and not directly comparable between mills and technologies due to differences in methodology and assumptions. Accordingly, no numbers are reported in the conclusions. Nevertheless, several conclusions of a more qualitative character can be drawn, with the most important summarized as follows:

1) Site-specific factors have a strong influence on the capture cost. Specifically, the flue gas flow (and its CO_2 concentration), the site location and the energy balance of a mill play an integral role. This makes it difficult to generalize results from one mill to others, since accurate cost estimates for carbon capture would need to consider site-specific factors.
2) PCC technologies, such as the novel process studied in this work, that can utilize low-temperature excess process heat will require a utility boiler with a smaller load to avoid losses in co-generation of electricity. For such technologies, excess heat availability is an important parameter for achieving a lower capture cost.
3) The way in which carbon capture is integrated with a site and the system in which the site operates is of importance. At low electricity prices, Scenario A achieved the lowest capture cost but resulted in the highest indirect fossil fuel emissions from the grid. At high electricity prices, Scenario C achieved the lowest capture cost but resulted in the most biogenic CO_2 emissions from an additional biomass boiler. This was the case for both technologies. A pulp mill owner could potentially choose to operate in Scenario A at lower electricity prices and Scenario B at higher prices to reduce the capture cost. The trade-off here being a lower capacity factor for the biomass boiler and therefore a greater capture cost than what was obtained for Scenario B only.
4) The capture cost of the Pulp Mill with lignin extraction was higher than capture cost for the Pulp Mill without lignin extraction. However, this is only an indication that carbon capture implementation is clearly favoured by favourable mill energy balances.
5) The extent to which variations in annual CAPEX, excess heat availability, and the price of electricity and fuel affect the capture cost depends on the heat integration scenario. Overall, variations in these parameters, with the exception of cooling water and fuel price, were found to have a considerable impact on the capture cost.

Appendix A: Supplementary Data

The process stream data of the Pulp Mill utilized for pinch analysis is shown below. Key streams have been identified in the footnote.

Stream	Type	Tin	Tout	Load MW
Lime kiln Flue gas[a]	H	300.00	125.00	4.54
SP7 Flue Gas[a]	H	170.00	125.00	6.69
Hot stream 1	H	127.43	127.33	11.06
Hot stream 2	H	127.23	127.13	4.62
Hot stream 3	H	106.55	50.00	8.85
Hot stream 4	H	103.37	50.00	6.81
Black liquor Flash steam[b]	H	102.32	102.22	27.55
Hot stream 5	H	96.00	95.90	6.21
Hot stream 6	H	93.00	76.00	0.69
Hot stream 7	H	93.00	69.00	3.48
Hot stream 8	H	75.00	39.00	25.92
Hot stream 9	H	74.80	50.00	8.68
Hot stream 10	H	70.00	39.00	8.72
Hot stream 11	H	68.91	68.81	36.70
Hot stream 12	H	65.93	65.83	0.67
Hot stream 13	H	65.93	36.88	0.02
Hot stream 14	H	62.28	62.18	39.55
Hot stream 15	H	58.87	40.00	2.83
Hot stream 16	H	50.00	39.00	7.66
Hot stream 17	H	145.19	130.60	4.71
Cold stream 1	C	32.63	60.79	0.44
Cold stream 2 (MP steam consumer)	C	187.96	188.06	4.77
Cold stream 3 (MP steam consumer)	C	187.96	188.06	6.10
Cold stream 4 (MP steam consumer)	C	187.96	188.06	10.39
Cold stream 5 (MP steam consumer)	C	187.96	188.06	2.35
Cold stream 6 (MP steam consumer)	C	187.96	188.06	1.21
Cold stream 7[c]	C	155.50	155.60	17.09
Cold stream 8[c]	C	152.24	152.34	15.99
Cold stream 9[c]	C	151.84	151.94	6.08
Cold stream 10[c]	C	151.44	151.54	15.06
Cold stream 11 (LP steam consumer)	C	143.63	143.73	4.58
Cold stream 12 (LP steam consumer)	C	143.63	143.73	4.35
Cold stream 13 (LP steam consumer)	C	143.63	143.73	1.56
Cold stream 14 (LP steam consumer)	C	143.63	143.73	0.06
Cold stream 15 (LP steam consumer)	C	143.63	143.73	8.71
Cold stream 16	C	138.66	138.76	18.88
Cold stream 17	C	138.66	138.76	45.17

Cold stream 18	C	137.84	137.94	17.92
Cold stream 19	C	127.23	127.33	13.26
Cold stream 20	C	86.00	86.10	0.44
Cold stream 21	C	51.80	87.89	3.49
Cold stream 22	C	33.98	70.71	1.28
Cold stream 23	C	32.06	32.16	0.60
Cold stream 24	C	28.30	130.76	10.97
Cold stream 25	C	16.57	75.14	63.44
Cold stream 26	C	16.57	52.48	26.48
Cold stream 27	C	15.45	28.18	2.83
Cold stream 28	C	7.09	33.98	36.70
Recovery Boiler Heat Radiative SP7[d]	H	1 000.00	999.00	104.04 (90.35)
Recovery boiler Heat Convective SP7[d]	H	1 000.00	170.00	123.33 (107.10)
Recovery Boiler Radiative Heat SP6[d]	H	1 000.00	999.00	27.46 (23.85)
Recovery Boiler Convective Heat SP6[d]	H	1 000.00	170.00	31.21 (27.10)

[a] Flue gas for heat recovery
[b] Main process stream identified for providing excess heat to novel process
[c] Assumed to met by mixing LP and MP steam
[d] Excess heat from recovery boilers, values for lignin extraction shown in brackets

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