THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

New roles for chemical pulp mills in the future energy system

SIMON INGVARSSON

Department of Space, Earth and Environment

CHALMERS UNIVERSITY OF TECHNOLOGY

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SIMON INGVARSSON

Department of Space, Earth and Environment
Chalmers University of Technology
SE-412 96 Gothenburg
Sweden
Telephone + 46 (0)31-772 1000

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Abstract

Major transformations of the energy system are expected in the coming decades, in response to climate policy and reduced costs of renewable electricity sources. In this future, Swedish chemical pulp mills, as large processors of biogenic carbon and net suppliers of electricity, may have several roles to play. This thesis explores the conditions for chemical pulp mills to alter their position in the electricity system from being base-load providers to acting as flexibility providers, to introduce new biorefinery concepts that enable the production of advanced biofuels, materials and chemicals, and to provide negative emissions via bio-energy carbon capture and storage.

The analysis is carried out using a novel techno-economic model representing a typical pulp mill. The model is a linear optimisation model, optimising the operation of the mill, i.e., the load of all processes, boilers and turbines for each hour of the year, and, for a set of future scenarios, the investments in new technologies that can be added to the existing mill. The objective is to maximise the net revenue for the mill while maintaining the hourly balances of materials, steam and electricity throughout the mill. The outputs from the optimisation model are further processed to derive economic indicators, such as the product value generated from the mill and the price tipping points for profitability when comparing different options. The flows of carbon throughout the mill are also compared across the different scenarios.

The results show a potential for the mill to act as a flexibility provider for the electricity system, without any impact on pulp production. The flexibility is enabled by a combination of inter-dependent measures on the supply and demand sides. The main technical limitations to flexibility are the capacity limits of the flexible processes (minimum and maximum load levels), while storage units for intermediate products are typically large enough to handle variations on time-scales from hours to days. If, for an existing mill, supply-side flexibility is not already available in the recovery boiler, it can be unlocked by introducing a lignin extraction plant. This synergy increases the incentives for the mill to produce lignin as a new product, as it reduces the penalty on the electricity trade that is otherwise associated with lignin extraction.

When different options for the chemical pulp mill are compared, it is seen that the role that the mill plays in the energy system may evolve over the coming decades. In the short term, the proposed flexibility measures may facilitate the introduction of wind and solar power into the electricity sector and, thereby, support the electrification of the transport and industry sectors. In the longer perspective, an increasing demand for biomass may create incentives for more transformative changes, including the large-scale implementation of carbon capture, either for the production of e-fuels or to provide negative emissions.

Keywords: *Biorefinery, Carbon capture, Chemical pulp mill, Flexibility, Lignin extraction, Optimisation, Pulp and paper, Sector coupling*

List of publications

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

I. Ingvarsson, S., Odenberger, M., & Johnsson, F. (2023). The chemical pulp mill as a flexible prosumer of electricity. *Energy Conversion and Management: X*, *20*, 100401. https://doi.org/10.1016/J.ECMX.2023.100401

II. Ingvarsson, S., Odenberger, M., & Johnsson, F. (2024). Lignin extraction in chemical pulp mills: The role of flexible operation. *Sustainable Chemistry for the Environment, 7,* 100137. https://doi.org/10.1016/j.scenv.2024.100137

III. Ingvarsson, S., Odenberger, M., & Johnsson, F. (2024). Chemical pulp mills in future energy markets with variable electricity prices and increased demand for biogenic carbon. *Manuscript*.

Author contributions

Simon Ingvarsson is the principal author of **Papers I–III** and performed the modelling, analysis and draft writing for all three papers. Associate Professor Mikael Odenberger contributed with method development, editing and discussions to all the papers. Professor Filip Johnsson contributed with editing and discussions to all the papers.

Acknowledgements

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Simon Ingvarsson
Göteborg, September 2024

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1. Introduction

Swedish chemical pulp mills are nearly self-sufficient for their energy supply, owing to the use of biogenic by-products as fuels [1], [2]. Assuming that the woody biomass is sourced from a forest management system where the carbon stock is maintained, this means that they are close to carbon-neutral in their operations. Nonetheless, they may still have an important role to play in the mitigation of climate change due to: (i) their position as prosumers in the electricity system [1]; (ii) their potential to substitute emissions-intensive products in other sectors, such as transportation and construction [3], [4]; and (iii) the possibility that they could provide negative emissions via bio-energy carbon capture and storage (BECCS) [5].

For the electricity sector, an increasing share of variable renewable electricity generation is expected to introduce new challenges related to the matching of supply and demand, leading to higher volatility in the electricity price, as compared with the prices from the historical system [6]. This development calls for variation management strategies, such as energy storage and flexibility measures on both the supply and demand sides, to be implemented by actors within the system [7].

In the transportation sector, there will likely be a substantial demand for biofuels for maritime transport and aviation, whereas electrification will be the most cost-effective strategy to reduce emissions for road transport. If the biomass that is available for biofuels is limited, and fossil fuels should be phased out completely, there will also be a demand for electro-fuels (e-fuels) from these sectors [8].

BECCS could, if implemented on a large scale, and in a manner that does not lead to depletion of the carbon stock, help to reach the climate targets. Negative emissions achieved via BECCS can be used to compensate for residual emissions in hard-to-abate sectors, as well as to reach net-negative emissions for the whole economy eventually [9].

These new demands may increase the value of biogenic carbon, motivating the chemical pulp mill to alter its role in the electricity system from being a base-load provider to acting as a flexibility provider, and introducing new biorefinery concepts that enable the production of advanced biofuels, materials and chemicals. It is of value for both policymakers and stakeholders in the industry to acquire a clear understanding of the drivers, barriers and timing of this transition.

1.1. Aim and scope

This thesis explores different alternatives for the development of chemical pulp mills in the near future (up to around mid-century), under the assumption that the surrounding energy system evolves towards net-zero emissions. The scope covers both opportunities that can be implemented in the short term to provide flexibility to the electricity system, and transformative pathways in which the chemical pulp mill is developed towards becoming a multi-product biorefinery.

Given the relatively short time perspective in relation to the investment cycles of the industry sector, this study builds on conventional chemical pulp mills with typical process configurations. When the implementation of new technologies is considered, attention is focused on the technologies that have recently received the most interest from industrial actors. Additional technologies, which are established in the scientific literature, but which currently seem further away from being implemented, are treated in the *Background* and *Discussion* sections.

This thesis addresses the following questions:

- How can the chemical pulp mill contribute with flexibility measures to support the balance between supply and demand in an electricity system that has a high share of variable renewable electricity generation?
- What are the economic incentives and barriers that affect the ability of the chemical pulp mill to take on the role of a flexibility provider in the electricity system?
- How may the role of the chemical pulp mill in the energy system change over time in response to increased demands for bio-based products from other sectors?

1.2. Contribution of the included works

Three papers are appended to the thesis. **Paper I** serves as a proof-of-concept study for the modelling framework used in all the papers. In this paper, the technical and economic potentials of flexibility measures in a chemical pulp mill with current technology are assessed, and the technical limitations are analysed. **Paper II** focuses on the implementation of a lignin extraction technology, addressing how lignin extraction can be used to increase the production of the mill and how flexibility measures can reduce the trade-off between selling electricity and selling lignin. **Paper III** explores the future role of the chemical pulp mill in the energy system, considering the potential demands for electricity, transportation fuels and negative emissions. In this study, the investment options for a condensing turbine, a lignin extraction plant, and a carbon capture unit are compared.

2. Background and related work

In the following section, context is presented in relation to the different roles that the mill may take on in the studied future in response to new demands. Section 2.1 gives an overview of the technologies that are available to adapt chemical pulp mills towards multi-product biorefineries, producing advanced biofuels, materials and chemicals. Section 2.2 delves into the factors that are expected to drive the development of the electricity system and the ways in which the development will motivate the chemical pulp mill to act as a flexibility provider. Section 2.3 presents a review of previous studies on the interplay that occurs between the chemical pulp mill and the energy system.

2.1. From pulp mills to multi-product biorefineries

The scientific literature covers a wide range of available technologies that could be implemented with chemical pulp mills[1] to develop the mills towards becoming biorefineries, several of which have already been implemented in demonstration, pilot or commercial scale [10], [11]. A selection of these technologies is listed in Table 1, with a summary of their feedstocks and applications, and including references to techno-economic studies on how these technologies can be integrated into chemical pulp mills.

All of the reviewed technologies affect the energy balance of the mill, by using as feedstock the biomass that would otherwise have been used as fuels in boilers, and/or directly by energy consumption within the processes. The lost energy could be replaced by electricity, for example, by replacing conventional boilers with electric boilers or heat pumps for steam supply [12] or by replacing the conventional lime kiln with a gas-plasma calcination technology [13]. For the implementation of carbon capture, technologies that consume more electricity and less heat, such as the hot potassium carbonate process, could be considered [14][2].

Table 1. Examples of available technology options that can be integrated with chemical pulp mills.

Category	Technology / Process	Feedstock	Intermediate product	Final product (examples)	References (examples)
Thermochemical processes	Gasification	Black liquor Solid residuals*	Syngas	Advanced biofuels** Hydrogen Methane (biogas)	[15]–[17]
	Pyrolysis	Solid residuals*	Pyrolysis oil	Advanced biofuels**	[18]
Biochemical processes	Acid precipitation	Black liquor	Lignin	Advanced biofuels** Carbon fibres Chemicals***	[19]–[23]
	Hydrolysis	Solid residuals*	Sugars	Ethanol	[24]
Carbon capture processes	Chemical absorption	Flue gas	CO_2	E-fuels Negative emissions	[25]–[28]

**E.g. bark, wood chips and sawdust; **E.g. biodiesel, methanol and DME; ***E.g.* adhesives and dispersants.

[1]Chemical pulp can be made either through the kraft (sulphate) process or the sulphite process. However, this thesis only considers the kraft process (which dominates modern chemical pulping); the terms *chemical pulp* and *kraft pulp* are used interchangeably.

[2]Although, as discussed by Roshan Kumar et al. [14], it may be more efficient to use a heat-driven process in combination with a heat pump.

Furthermore, as proposed by Svensson and Ulmefors [29], one possibility to increase the share of the wood that ends up as high-value products is to increase the use of chemical-thermo-mechanical pulp (CTMP) mills, which have a higher yield per input wood than in chemical pulp mills (since lignin and hemi-cellulose are also included in the pulp). While chemical pulp cannot be directly replaced by CTMP since paper made from the different types of pulp have different characteristics, there is ongoing development to increase the share of CTMP in some applications.

2.2. An electricity system in transition

In parallel with the development of new markets for fuels, materials and chemicals, the electricity market is expected to be transformed in the coming decades. The demand for electricity in Sweden is projected to more than double up to Year 2050 compared to current levels, in scenarios with large-scale electrification (directly or via hydrogen production) of the iron and steel industry and transport sectors [30]. During the same period, several nuclear plants and a large proportion of the transmission grids are expected to reach their end of lifetime [2]. With electricity from wind power and solar PV now being less expensive to produce than electricity from fossil fuels [32], a cost-optimal future electricity supply mix in Sweden would be dominated by sources of variable renewable energy (VRE) [33].

Figure 1 shows the projected electricity price profiles (duration curves) for Year 2030 and Year 2050 for the SE3 region, derived from the optimisation model described by Öberg et al [34]. These price profiles represent a future development with new industrial demands and an increasing share of VREs in the supply mix, in comparison with the historical prices in Year 2017[3] for the same region. The increasing share of VREs in the future systems is reflected in the increasing variability of the projected prices for Year 2030 and Year 2050. Adding new nuclear plants to the Swedish generation mix would not significantly reduce the price volatility, due to the relatively small size of the Swedish system in relation to the interconnected north-European system of which it is a part [33].

It should be noted that since the price curves shown in Figure 1 are derived through an optimisation, the real prices are likely to be higher on average. As an example, the electricity prices for Year 2022 in the SE3 region were much higher and more-widespread than what would normally be the case for the current system, following unexpected price increases for natural gas and coal in neighbouring regions [35].

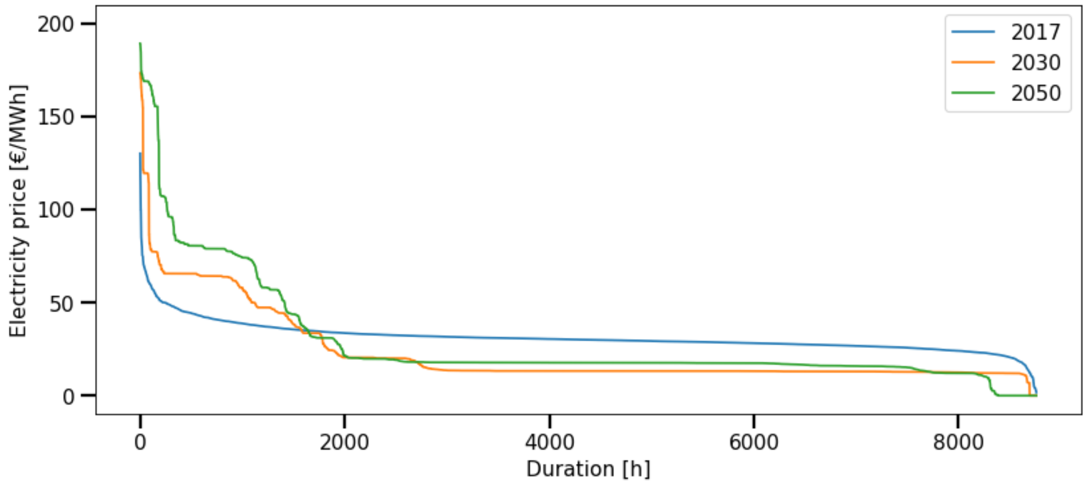


Figure 1. Duration curves of projected electricity prices for the SE3 region for Years 2030 and 2050, in comparison with the historical prices for the same region in Year 2017.

[3]Year 2017 was selected for comparison because the average price level was similar to that of the projected years.

Furthermore, since planned projects on both the supply side and demand side are expected to entail an annual electricity production or consumption level on the magnitude of terawatt-hours (TWh) [36], [37], any project that is not realised or that is added to the projections may severely disrupt the balance between the supply and demand. This may drive the prices higher and make them even more volatile, at least during the transition phase to a new stable system.

The highly variable electricity prices incentivise the application of flexibility measures that shift, absorb or complement the electricity load [6]. The most-rapid variations, with relevance for control of the grid frequency, can be cost-efficiently managed with stationary batteries [38]. For variations with durations longer than 1 hour, various flexibility measures may be viable, depending on the duration, frequency and amplitude of the variations and the system context. These measures include supply control, industry and household demand-side management, battery storage, and increased integration with the heating sector [7].

2.3. Previous studies on mill–energy system interactions

For increased integration of the chemical pulp mill and a future electricity system with high variability in the balance between the demand and available supply, a central aspect is the operational flexibility of the mill. A survey of flexibility measures in chemical pulp mills has been conducted by Jannasch et al. [39]. They have shown that to use a process for flexible operation in response to variable electricity prices, some kind of buffer must be available, both before and after the process. The process also requires an over-capacity compared to the average operation, so as to be able to catch up after a period of down-regulation (and conversely, the possibility to run on part-load after an up-regulation). Furthermore, they have proposed that supply-side and demand-side measures are inter-dependent; On the one hand, flexibility in steam-consuming production processes is limited by the capacities of boilers and turbines while, on the other hand, flexible operation of the boilers and turbines depends entirely on the demand-side processes, since the boilers are fuelled by black liquor and bark from the production processes.

Several modelling studies have been conducted, covering different aspects of flexible operation. Flexibility in the powerhouse of the mill (i.e., boilers, turbines and steam system) has been addressed previously [40]–[45][4]. However, none of those studies explicitly included the connection between the powerhouse and the production processes. Svensson [46] presented an optimisation model for investment decision in an individual mill, which has been used to assess the value of a diversified energy by-products portfolio for achieving more-flexible operation, including the possibility to extract lignin from the black liquor to sell as a product [47]. For carbon capture processes, possibilities for flexible operation in response to varying electricity prices, e.g., through the storage of absorbent chemicals, have been covered previously [48]–[51], although not specifically for pulp mill applications.

[4]See **Paper I** for a more-thorough review of these studies.

3. Method

The work presented in this thesis has been performed using techno-economic modelling methods, with the goal of bridging the gap between studies of industrial energy systems in the pulp and paper sector and studies of regional and national energy systems. To enable investigations of the interactions between the industry and the electricity system, all the analyses are carried out with an hourly resolution, matching the price fluctuations on the spot market for electricity.

Although the work primarily focuses on a single type of industry, i.e. the chemical pulp mill, the methodology has been developed with broader applicability in mind. The fundamental dynamics, including the drivers and restrictions for flexible operation, should be applicable to most process industries, with the highest relevance for those with integrated energy supply units. In addition to the work on chemical pulp mills covered by this thesis, the developed modelling framework has been adapted and applied by Kjellander [52] to model an electricity system-integrated sawmill with an internal heat supply.

The outputs from the models are further processed to derive economic indicators, such as the product value generated from the mill and the price tipping points for profitability when comparing different options. In **Paper III**, an indicator of carbon efficiency[5] is used as a measure to compare the mill's potential to replace fossil carbon across the different studied scenarios.

3.1. Optimisation model

The modelling activities in **Papers I–III** develop and apply the same modelling framework. The model is a linear optimisation model, representing a typical chemical pulp mill. In **Papers I** and **II**, the dimensioning of all processes in the mill is given exogenously to the model, while the model optimises the operational dispatch, i.e., the load of all processes, boilers and turbines at each time-step. In **Paper III**, the investments in certain units (the condensing turbine, lignin extraction plant and carbon capture system) are also optimised. In specific scenarios in **Paper I**, where the mill alternates between softwood and hardwood processing, a mixed integer linear programming (MILP) version of the model is used to optimise the timing of the operational campaigns.

The objective of the model is to maximise the net revenue (revenues minus costs) for the mill over a given time period (typically a year) by varying the material, steam, and electricity flows throughout the mill, while maintaining all the energy and mass balances within the system. Figure 2 shows an overview of the objective function, input parameters, constraints, and result variables included in the modelling. A detailed mathematical description of the model is given in **Paper I**, and further developments are described in **Paper II** and **Paper III**.

[5] In **Paper III**, carbon efficiency is defined as the fraction of the carbon input that leaves the mill in products.

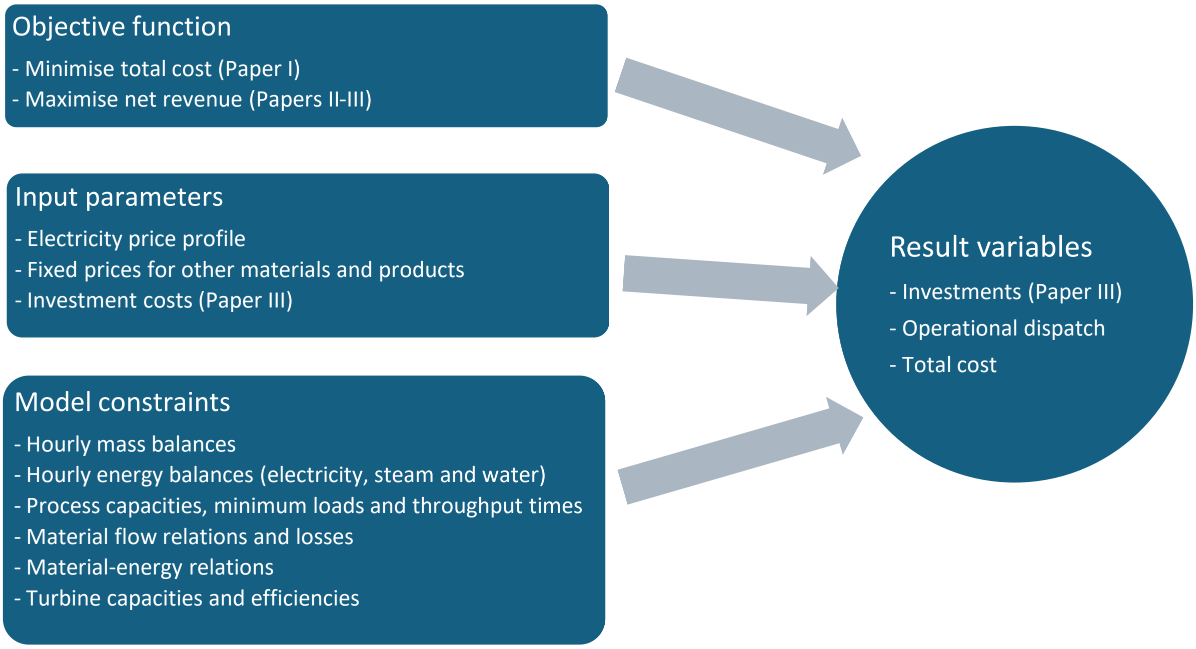


Figure 2. Overview of the objective function, input parameters, constraints and result variables included in the modelling.

Figure 3 shows an overview of the model mill, including the connections between production processes, boilers and storage units. In addition to the illustrated material flows, the model covers the flows of steam and electricity. Steam production takes place in the recovery boiler, which combusts the black liquor, and in a separate bark boiler. Steam is converted to electricity in one back-pressure turbine and, in some cases, one condensing turbine. The generated electricity and steam are supplied to industrial processes throughout the mill, and any surplus or deficit electricity is traded with the electricity market, which is simply included as an unlimited market with an endogenous price signal, under the assumption that the mill is a price-taker.

In **Paper II**, modifications are made to the model, so as to represent an installed lignin extraction process based on acid precipitation as an extra intermediate step within the evaporation plant. The unit representing the evaporation plant process is split into two parts, with the lignin separation process in between, as illustrated in Figure 4. The remaining lignin and the non-lignin parts of the black liquor are kept separate in the model, even though in reality they comprise a single physical flow.

Extracting lignin from the black liquor reduces the load in the recovery boiler, which the model can use either to increase the overall throughput of the mill (if the recovery boiler is a bottleneck to begin with) or to enable flexible operation of the recovery boiler. Three factors limit how much lignin can be extracted in the model: (i) the momentary maximum, as limited by the extraction process; (ii) the lowest lignin content of the black liquor for which it is assumed to be still combustible; and (iii) the overall steam balance of the mill.

In **Paper III**, the model mill is further extended with a carbon capture module that is connected to the three flue gas stacks: the recovery boiler, the bark boiler, and the lime kiln, as depicted in Figure 5. The module represents a post-combustion system that is based on chemical absorption, with separate absorbers for each flue gas stack, albeit with a joint stripper and liquefaction unit. For flexibility in relation to electricity price variations, the absorbers and the stripper can be operated independently from each other to the extent allowed by the size of the absorbent storage units.

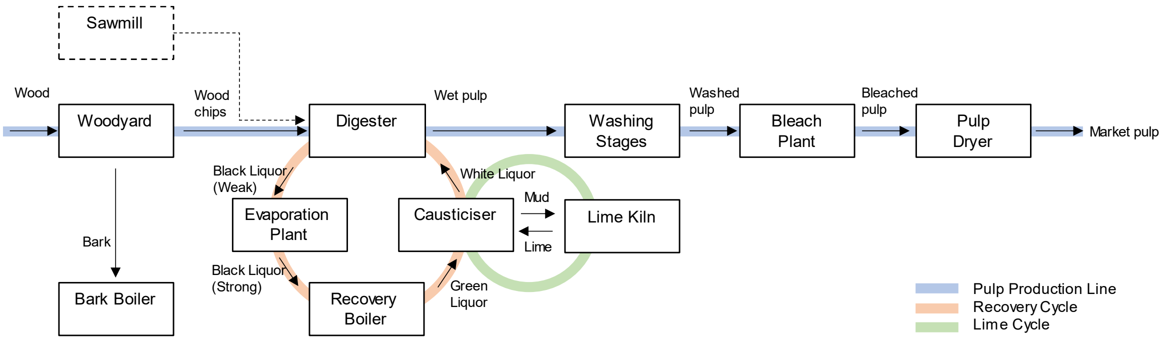


Figure 3. Schematic overview of the model mill. Source: Paper I.

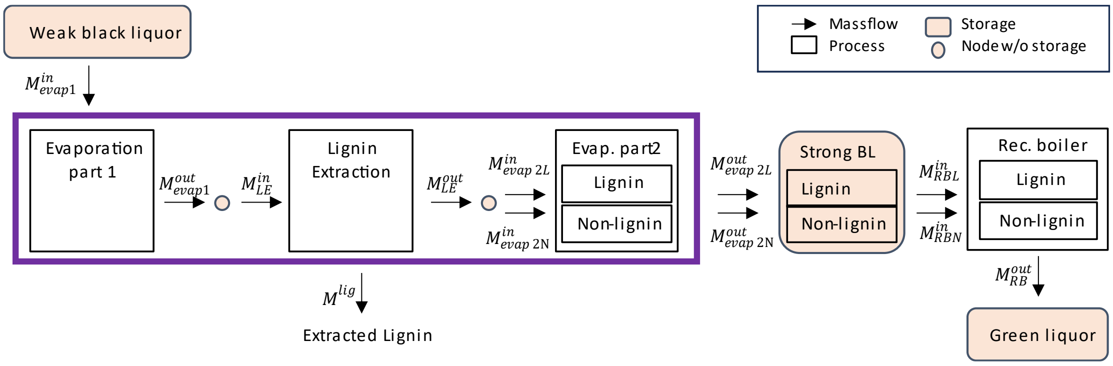


Figure 4. Schematic of the modelled evaporation plant and recovery boiler, where the evaporation plant includes an integrated lignin extraction process. BL, Black liquor. Source: Paper II.

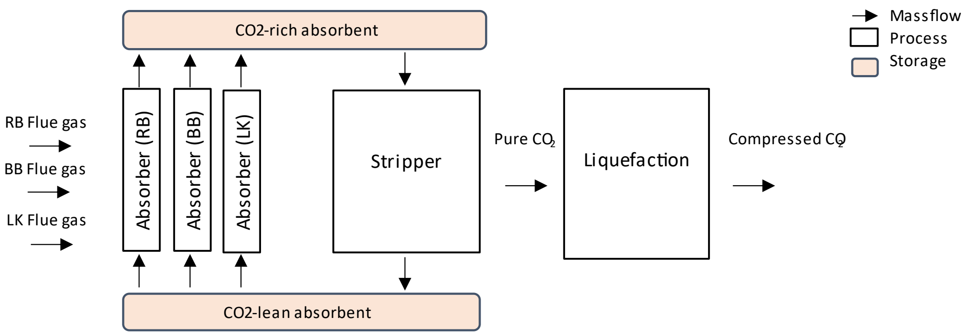


Figure 5. Schematic of the modelled carbon capture system. RB, Recovery boiler; BB, bark boiler; LK, lime kiln. Source: Paper III.

3.2. Assumptions and Data

The input data to the models are mainly based on steady-state descriptions from the literature, which have been complemented with assumptions for the dynamic aspects (mainly storage sizes and process throughput times) obtained through discussions with industry personnel.

For the conventional mill processes, the mass and energy balances are extracted from a bleached softwood market pulp mill presented in Åforsk Model Mills 2010 [53], which is a description that has been developed to represent a series of chemical pulp mills with the best-available technology at the time of its publication. It is a stand-alone mill with a capacity of 2,000 air-dry tonnes per day (ADt/d) of pulp. The mill is self-sufficient with regards to heat and is a net-exporter of electricity. The lime kiln is fuelled with gasified bark, such that no externally supplied fuels are needed.

Process throughput-times and storage sizes for the core model are assumed based on typical real mills in discussion with industry. For process over-capacities, the conditions vary between mills, so for the study, a set of cases covering different conditions is used.

The lignin extraction process is assumed to be similar to LignoBoost [54], and the input data for its model implementation are primarily based on the steady-state descriptions of Tomani et al. [20], Olsson [55] and Vakkilainen & Välimäki [56].

The carbon capture module is assumed to use monoethanolamine (MEA) as solvent and to have a maximum capture rate of 90%. The input data associated with carbon capture are mainly based on the steady-state descriptions of Onarheim et al. [26], [57].

For the future scenarios studied in **Paper III**, the electricity prices projected for Years 2030 and 2040 are supplied by Öberg et al. [34] and the future market prices for lignin and CO_2 are based on assumptions regarding the willingness-to-pay from the transportation sector or negative emissions buyers, as explained in detail in the paper.

3.3. Complementary methods

In **Papers II** and **III**, the optimisation modelling is in different ways complemented with idealistic calculations for the potential of flexible operation, given a certain electricity price profile. The idealistic approach ignores the limitations linked to some of the technological constraints (capacity sizes, storage volumes and time delays) of the model mill, as well as the detailed dynamics (frequency, duration, amplitude) of the electricity price variations. Nonetheless, it provides a rapid screening of the maximum economic potential of flexible operation and adds robustness to the results, since these analyses depend on fewer assumptions than the specific cases analysed by the optimisation procedure.

4. Results and discussion

In this section, the results are presented, analysed, and discussed. Sections 4.1–4.2 focus on the technical and economic aspects of flexible mill operation, mainly reviewing the results from **Papers I** and **II** and discussing how these are related, but also elaborating on some aspects that are not discussed in detail in the papers. Section 4.4 considers how the role of the chemical pulp mill may change over time, mainly based on the results from **Paper III**, while adding new insights based on the combined results from all three papers.

4.1. Technical potential and limitations for flexible operation

The technical potential and the limitations of flexible operation of conventional mill processes are investigated in **Paper I**, for a range of cases spanning from less-flexible to more-flexible. Figure 6 shows a duration curve of the net electricity production from the mill (a mill of producing 2000 ADt/d of pulp) for the modelled cases, together with a reference case that has steady-state operation. The results show that the mill remains a net producer of electricity during most of the operational hours, even with flexible operation. The span of the lowest to highest momentary net production ranges from 17 MW in the least-flexible scenario to 105 MW in the most-flexible scenario. Aggregated over the modelled year, 25–111 GWh of electricity are shifted from low-value to high-value hours.

To position these numbers in the context of the Swedish electricity system as a whole, the potentials are scaled with a factor of 10 to represent the 20 largest chemical pulp mills in Sweden[6]. The resulting potential is 200 MW to 1 GW in terms of capacity or 250 GWh to 1.1 TWh in terms of aggregated shifted electricity. Compared with the Swedish annual electricity consumption of 135 TWh (i.e., on average 15 GW), the potential is rather small but it is not negligible. If the numbers at the higher end of the range could be realised, the proposed measures could be of great importance locally or during hours when it is especially difficult to match the supply with the demand.

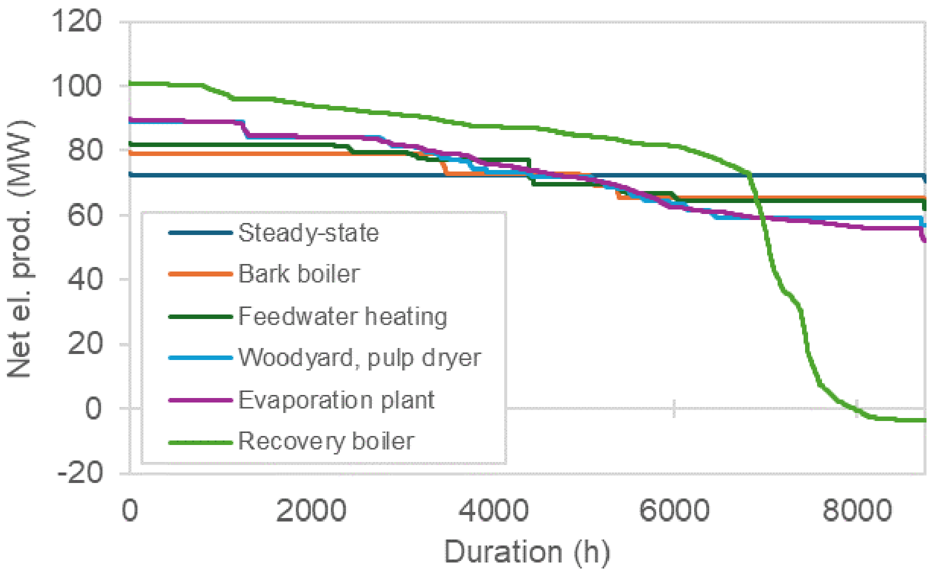


Figure 6. Duration curve of the net electricity production from the model mill (producing 2000 ADt/d of softwood market pulp) for the modelled cases and for a reference case with steady-state operation. The legend shows the unit(s) in which that flexibility is introduced in each case, in addition to the previous cases. Adapted from Paper I.

[6]Based on production data from Chalmers Industrial Case Study Portfolio (ChICaSP) [61].

The results from **Paper I** show that over-dimensioning the recovery boiler has a strong effect on the overall flexibility potential of the mill. In **Paper II**, the idea is presented that flexible operation of the recovery boiler can be enabled in an existing mill by adding a lignin extraction process, whereby lignin is removed from the black liquor before the black liquor enters the boiler. While the technology for lignin extraction is well known (and commercially deployed at several sites world-wide), the results from **Paper II** show that lignin extraction in combination with flexible recovery boiler operation can increase the product output from the mill with a low penalty the electricity trade, increasing the incentives for investments in lignin extraction. It should be noted, however, that lignin extraction with flexible recovery boiler operation does not increase the value of the electricity trade compared with the original mill configuration; instead, it minimises the loss of value from electricity trade that occurs due to the lignin extraction.

In both **Papers I** and **II**, the main technical limitations to flexibility are the capacity limits (minimum and maximum loads) of the flexible processes, turbines and boilers. The storage units for intermediate products are typically sufficiently large to handle variations on time-scales from hours to days but become limiting for storage on longer time scales. The flexibility is, therefore, mainly used to match the diurnal variations in the electricity price, although long-term storage of bark also enables a seasonal pattern of operation for the bark boiler.

4.2. Incentives for and barriers to flexible operation

Figure 7 shows the impacts on the mill economy of the proposed measures for flexible operation of conventional mill processes (based on results from **Paper I**), with the electricity prices of Year 2019. The results show that, already with these relatively stable electricity prices, the value of traded electricity can be increased by 1%–8% through increased integration with the electricity market. The For the model mill, producing 2,000 ADt/d of pulp, the economic gains correspond to 0.2–2.0 M€ per year. The gains are small in relation to the total income from electricity net export (around 26 M€ for the model mill) or the revenue from pulp sales (around 730 M€), but it should be noted that the benefits are achieved without investments in additional process capacity and without any impact on pulp production. Figure 7 shows the contributions to the value linked to having flexibility in the boilers (steam supply), steam consumption in the production processes and turbines (steam demand), and electricity consumption in the production processes (el. demand). In the most-flexible scenario, where over-capacity is available in the recovery boiler, steam supply flexibility dominates the value of the traded electricity. In the other scenarios, where flexibility of the steam supply, steam demand and electricity demand are all available, they contribute around 40%, 40% and 20% of the value, respectively.

The economic results presented in **Paper I** not only imply an economic synergy between the mill and the electricity system, but also show how additional value can be generated from the same biomass resource, even without shifting to alternative products. If the shifting of electricity generation from low-price hours to high-price hours lead to lower levels of fossil fuels being used for peak electricity production, the measures will also contribute to reducing greenhouse gas emissions from the electricity sector.

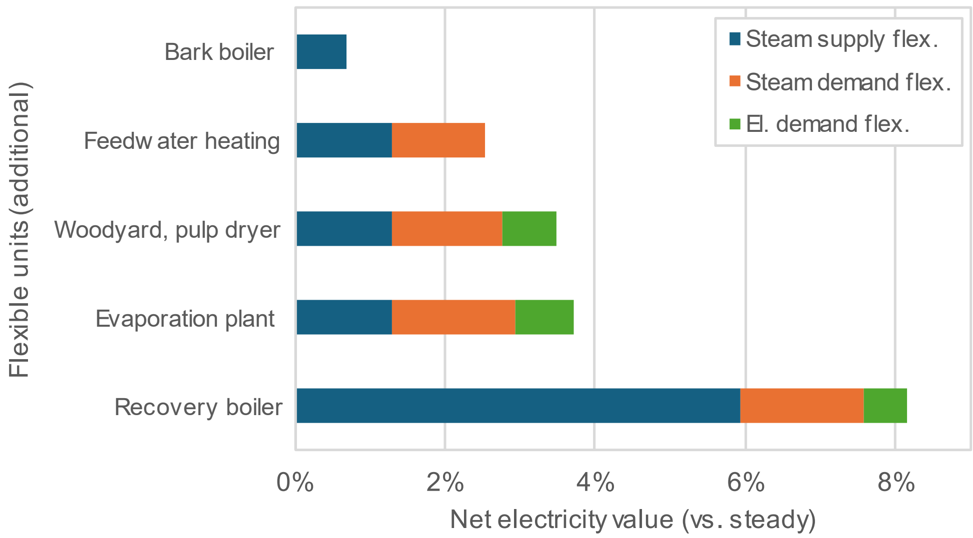


Figure 7. Impacts on mill economy of the proposed flexibility measures in cases modelled with the electricity prices of Year 2019. The flexibility measures are categorised as steam supply flexibility, steam demand flexibility and electricity demand flexibility. The y-axis labels list the unit(s) in which that flexibility is introduced in each case, in addition to the previous cases. Adapted from Paper I.

It should be noted that the modelled cases are stylistic, in the sense that they assume similar conditions for flexibility throughout the year. In reality, a capacity that allows flexible operation can also be available during parts of the day or year, e.g., due to different product campaigns (as shown in the additional cases in **Paper I**), seasonal variations (e.g., variations in heat demand due to climatic variations) or operations that are undertaken only on certain occasions, such as knife-changes in wood choppers or soot-blowing in boilers.

While the aim of **Paper I** was to study the potential and incentives for operating the processes flexibly when capacity and storage volumes are already available, it can also be discussed to what extent there is motivation to make additional investments in process capacity or storage for the purpose of flexible operation. According to the results presented above, the economic gains from the flexibility measures are relatively small in relation to the typical magnitudes of investments in production units, boilers and turbines. This suggests that spot market variations in general are not large enough to consider flexibility at the investment stage. However, when the model was run with the high and variable prices of Year 2022 (also presented in **Paper I**), the economic gains were 10-fold higher, although the average electricity price was only 3-fold higher. This supports the hypothesis that flexibility measures will be increasingly valuable in a future with more volatile electricity prices. While the electricity prices projected for Years 2030 and 2040 are not as high and volatile as those recorded in Year 2022, it is not inconceivable that future prices will turn out to be similar to those in the Year 2022 profile, at least for some period during the transition of the energy system in the coming decades. A site-specific assessment could be used to assess if such variable electricity prices can indeed motivate investments in additional capacity for specific processes.

In **Paper III**, investment optimisation is explicitly conducted for a condensing turbine, a lignin extraction plant, and a carbon capture plant under projected assumptions regarding electricity and product prices in Years 2030 and 2040. The model mill makes use of the flexibility that is inherent to existing mill processes, and benefits from the flexibility in the recovery boiler when lignin extraction is installed, although the model does not choose to invest in additional capacity for the purpose of part-time utilisation in any of the modelled cases (except for a minor investment in the condensing turbine in one case). However, it is shown in the paper that spot prices similar to those recorded for Year 2022 would dramatically change the calculations, motivating investments in combinations of lignin extraction and electricity generation or carbon capture and electricity generation, for part-time utilisation in response to the variable electricity prices, for a broad range of lignin and CO_2 prices.

If the price differences on the spot market are not sufficiently high to motivate investments in flexible technologies, even though there still is a demand for these services, e.g., to reduce the need for investments in transmission or distribution grids, one option is for the mill to participate in local markets for flexibility, which are currently being developed in several places around Sweden.

4.3. Combining flexibility measures with lignin extraction

In **Paper II**, it is shown how flexible operation of the recovery boiler can reduce the operational costs for lignin extraction. Figure 8 shows the average cost of lignin depending on the extraction target. The largest share of the cost for lignin is the indirect electricity cost, i.e., the lost income from electricity that was generated from the combustion of black liquor in the reference case without lignin extraction. For lower levels of lignin extraction, the indirect electricity cost is lower, since the lost electricity generation (and of less importance, the increased consumption of electricity) can be concentrated to low-price hours. With the electricity price profile of Year 2019, the value of the flexibility is relatively low, whereas for the Year 2022 prices, flexible recovery boiler operation is shown to save 15%–70% of the operational costs of lignin extraction, as compared with steady-state operation. If the price ranges for future years are expected to lie somewhere in between those of Years 2019 and 2022, then the Year 2022 numbers can be regarded as an upper limit for the potential.

Figure 9 presents the aggregated impact on the operational revenue of the mill from lignin, electricity, and bark trade, using the case without lignin extraction as the baseline. The operational revenue is compared with a range of possible annualized investment costs that should be covered by the net revenue for the investment to be profitable. For the flatter electricity price profile of Year 2019 (left panel), the level of revenue increases linearly with increasing extraction target, from 10% extraction onwards. For Year 2022 (right panel), the picture is different. While the amount of sold lignin increases with the extraction target, the revenue per unit of lignin decreases. Consequently, each combination of electricity price, lignin price and investment cost will define a unique optimum level of lignin extraction to ensure maximum profit, for the specific electricity price year.

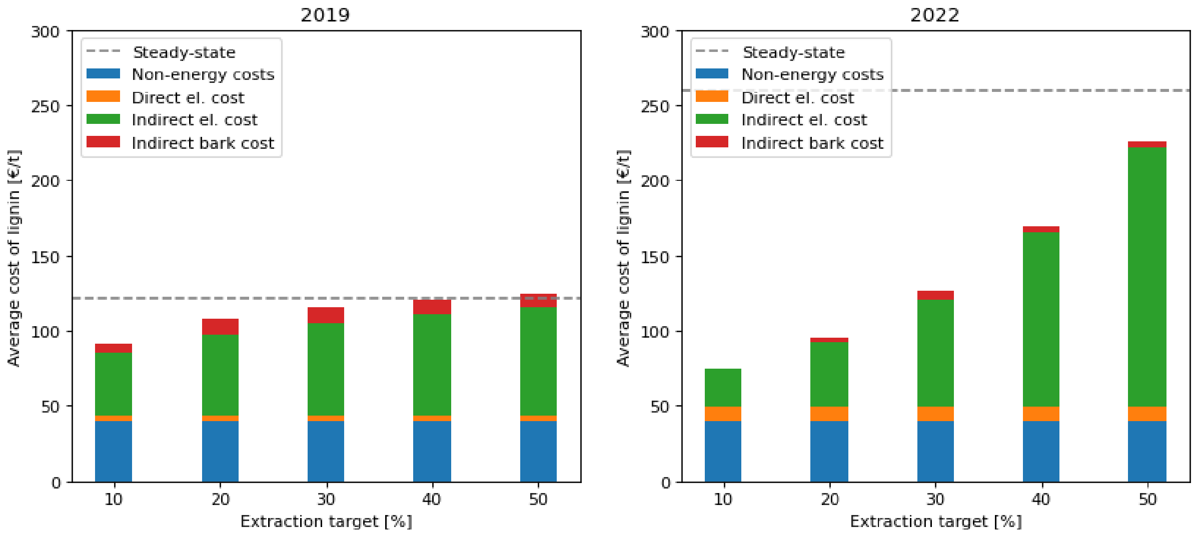


Figure 8. Average cost of lignin depending on the extraction target, for the electricity prices in Year 2019 (left panel) and Year 2022 (right panel). The indirect electricity cost (green bars) and indirect bark cost (red bars) represent the respective lost income levels from the sold electricity and bark that were generated in the reference case without lignin extraction. Investment costs are not included. Source: Paper II.

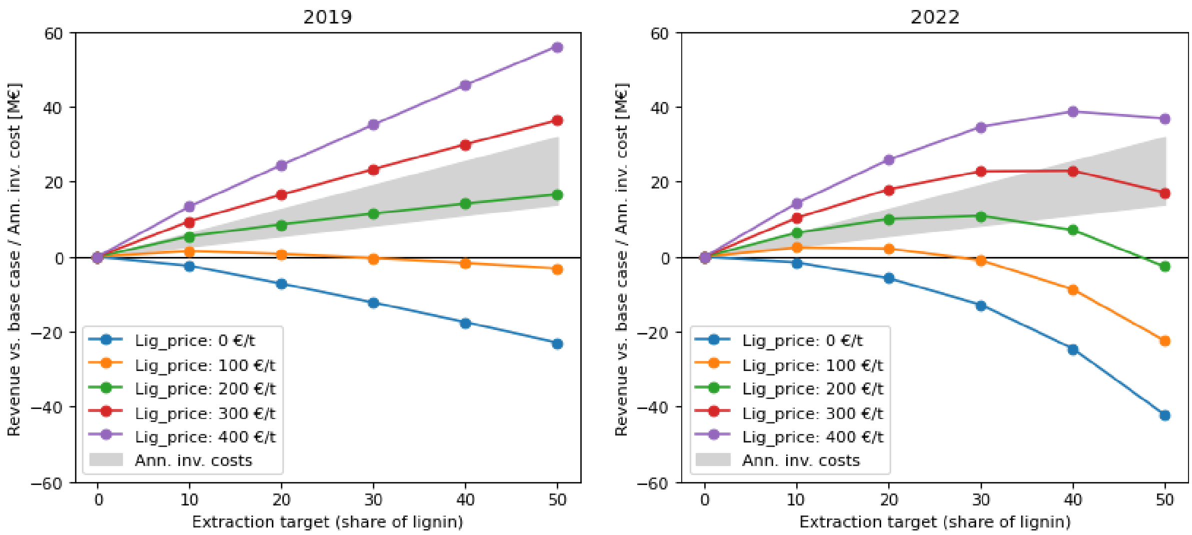


Figure 9. Aggregated impact on the operational revenue of the mill from lignin, electricity, and bark trade, using the case without lignin extraction as baseline, with the electricity prices of Year 2019 (left panel) and Year 2022 (right panel). A range of possible annualised investment costs are shown for comparison. Source: Paper II.

Furthermore, it is shown in **Paper II** that adding up to 10% of over-capacity in the lignin extraction plant further enhances the flexibility, thereby reducing the marginal cost of lignin by up to 50%, with the electricity prices of Year 2022. A similar effect can be achieved by increasing the size of the black liquor storage by a factor of 3, which also reduces the marginal cost of lignin by up to 50%.

4.4. The role of the chemical pulp mill in the energy system

Together, the three papers illustrate how the role of the mill in the electricity system could develop over the coming decades, if the electricity system develops towards an increasing share of variable renewable sources, while an increased demand for biogenic carbon drives the pulp mill towards investments in integrated biorefinery concepts.

The results from the reference scenario in **Paper III,** representing historical conditions, show that it has historically made economic sense to invest in an additional turbine for increased electricity generation, but not to invest in processes for the production of lignin or CO_2. In the reference scenario, the mill contributes to the energy system, mainly as a supplier of base-load electricity. However, the results in **Paper I** show, as explained above, that already with conventional mill processes, the mill can adopt a more active role on the electricity market than it has had historically, to act as an industrial-scale prosumer.

The role of the mill in the energy system may change more dramatically in the coming decades if the conditions on energy markets begin to favour the production of alternative by-products over the production of electricity. The results in **Paper II** show for which combinations of lignin price, electricity price, and investment cost for the lignin extraction technology that there is an economic incentive for the mill to invest in lignin extraction. From the results, a rule-of-thumb is obtained that lignin prices (€/t) must be 1.8-times the electricity price (€/MWh) plus the operational costs of lignin extraction (assumed to be 40 €/t), for lignin extraction to be more economically beneficial than the corresponding electricity generation.

Paper III contains analogous calculations for the comparison between carbon capture and electricity. Thus, for each tonne of CO_2 captured, 1.4 tonnes of low-pressure steam (1.08 MWh) are required, which could otherwise have been used to produce 0.17 MWh of electricity in the condensing turbine. In addition, the CO_2 liquefaction process requires 0.12 MWh of electricity per tonne of CO_2. Therefore, the willingness-to-pay for CO_2 (€/tCO_2) must be 0.29-times the electricity price (in €/MWh) plus the non-energy costs, for carbon capture to be more valuable than the corresponding electricity generation.

With the assumed capital costs being added to the analysis, the tipping points for investment decisions are calculated (Figure 10). The literature suggests that if kraft lignin is valued according to the equivalent fossil counterparts as a feedstock for chemicals and fuels, its market value may rise from the current value of around 100 €/t[7] to up to 350 €/t [58] or 500 €/t [59], which would then favour investments in lignin extraction over investments in electricity generation if average electricity prices remain around the current level (prices for region SE3 in Year 2019 are shown in the figure for comparison). Similarly, if the captured CO_2 is valued according to the credit price for emissions allowances within the EU ETS (minus the costs for CO_2 transport and storage), CO_2 market prices well above 100 €/t are not unlikely by Year 2040, which would outcompete electricity generation even at prices around the levels seen in Year 2022.

Comparing investments in carbon capture with investments in lignin extraction, Figure 10 shows that if CO_2 is sold at its production cost (around 50 €/t) the corresponding lignin price (150 €/t) is well within the price range for lignin applications. If, however, CO_2 is priced in line with the EU ETS, CO_2 prices at the ETS price for Year 2024 (75 €/t) [60] would match lignin prices >300 €/t, which is already near the maximum market value of kraft lignin (350 €/t) reported by Robinson et al. [58]. With CO_2 prices of 100 €/t, lignin prices must greatly exceed this level for lignin extraction to be the optimal investment.

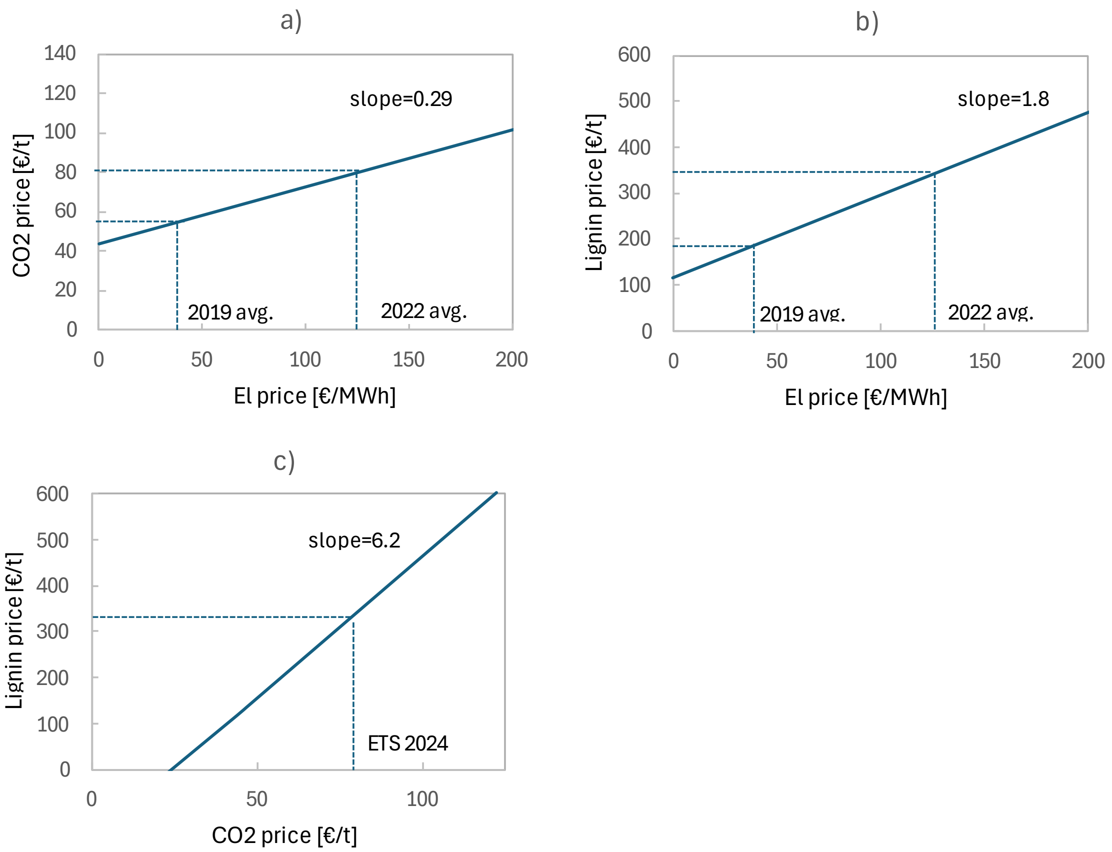


Figure 10. Tipping points for decisions to invest in: a) a condensing turbine or a carbon capture system; b) a condensing turbine and a lignin extraction plant; and c) a carbon capture system or a lignin extraction plant. The solid lines indicate the price combinations at which the economic incentives for both investment options are equivalent. Source: Paper III.

[7]If it is assumed that the price per MWh is equal to recent prices for solid by-products [62].

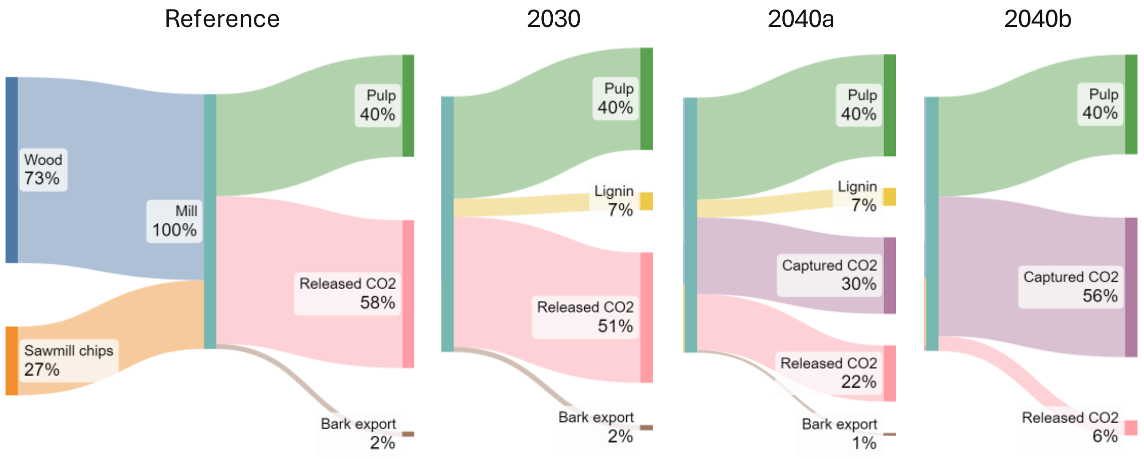
In **Paper III**, a set of scenarios is investigated, demonstrating a possible development in which the value of the biogenic by-products of the mill over time increases compared with the reference scenario, while electricity prices become more variable. Figure 11 shows the carbon balances over the mill, the carbon efficiency (defined as the share of the input carbon that leaves the mill in the form of products or as captured CO_2), and the net electricity generation from the mill.

In the Year 2030 scenario, an assumed demand for lignin from the maritime fuel sector sets the price for lignin. In this scenario, it is economically motivated for the mill to invest in both lignin extraction and a condensing turbine, slightly increasing the carbon efficiency while still maintaining a substantial electricity surplus.

In the Year 2040a scenario, lignin and CO_2 are valued according to their demands from biofuel and e-fuel applications, respectively (assuming that the need for e-fuels drives up the price also for biofuels). This incentivises the mill to prioritise increased carbon efficiency over electricity generation, leading to investments in lignin extraction and carbon capture. Lignin extraction is always favoured over carbon capture, although it is limited by technical constraints, which opens up possibilities for a combination of the two technologies.

In the Year 2040b scenario, CO_2 is priced according to its value in generating negative emissions via BECCS. This assumption implies a much higher price for CO_2 than in the Year 2040a scenario, such that lignin extraction cannot compete with carbon capture. In this scenario, the mill imports bark to increase further the output of captured CO_2.

These future scenarios describe a shift in the role of the pulp mill over time. In the reference and Year 2030 scenarios, the mill acts as a net electricity provider and flexibility actor, supporting the transition of the electricity sector (and, indirectly, the electrification of the heating, transportation and industry sectors) during this period. In the Year 2040 scenarios, in which the energy transition has reached the hard-to-abate sectors, it is more profitable for the mill to supply green carbon atoms than to supply electricity. Thus, it makes sense from both the economic and ecological perspectives that the role of the forestry sector in the wider energy system should eventually be as a supplier of green carbon atoms for hard-to-abate sectors (e.g., aviation, maritime transport, and plastics) and/or to provide negative emissions.



	Reference	2030	2040a	2040b
Overall carbon efficiency:	0.42	0.49 (+16%)	0.70 (+67%)	0.94 (+124%)
Net electricity generation:	641 GWh	428 GWh (-33%)	163 GWh (-75%)	246 GWh (-62%)

Figure 11.Carbon balances, carbon efficiency (share of the input carbon that leaves the mill in products or as captured CO_2), and net electricity generation from the mill in the four modelled scenarios. The percentages in parentheses indicate the differences compared to the Reference scenario. Source: Paper III.

If the value of the biogenic carbon is clearly higher than the value of electricity for most hours of the year, more-radical electrification measures could become relevant that were not available to the model mill in any of the presented modelling studies. Such possible measures include an electrified steam supply, gas-plasma calcination or integration of the electrolysis of hydrogen with the mill for the production of e-fuels.

It should be noted that the investigated scenarios are based on the assumptions that agreed policy measures on the European level for decarbonisation of the maritime and aviation sectors are implemented and followed as planned and/or that the allowance cap within the EU ETS is reduced to zero around mid-century (and in the 2040b scenario, that additional political decisions are taken to support the implementation of BECCS). In case that other political decisions are made, allowing for the continued use of fossil alternatives, the willingness-to-pay for bio-based fuels and products may be much lower than the assumptions on which this thesis is based. Although a market for these products may yet develop based on the possibility to sell carbon-neutral products at a premium value on a voluntary basis, it might not be sufficient to motivate any transformative changes in the pulp and paper sector.

Furthermore, if the suggested future scenarios are to be realised, a challenge is that the market conditions would change faster than it would take for the industry to alter roles, considering that investment cycles in the basic industries usually extend over decades. Thus, if the markets develop as suggested, it will be difficult for any actor in the system to take on the optimal role at every stage. Nevertheless, if decision-makers in the forest industry want to create the largest-possible value and contribute as much as possible to reaching climate targets, it is important that they understand how the markets and policy landscape are developing, not just in their own traditional value chain, but across all sectors of the economy.

5. Conclusion

This thesis and the appended papers present a novel methodology for techno-economic assessments of chemical pulp mills that covers the interplay between material, steam and electricity flows throughout the mill, as well as the interplay with the electricity market. The results demonstrate how existing and future processes within the chemical pulp mill can provide flexibility and increase integration with the energy system, and they provide insights into how the role of the pulp mill may change over time in response to an increased demand for biogenic carbon.

The aggregated potential for the flexible loads of conventional processes in Swedish mills, without any impact on pulp production, is estimated to be in the range of 200 MW to 1 GW. This range is broad due to a heavy dependence on site-specific conditions. The flexibility is enabled by a combination of inter-dependent measures on the supply and demand sides.

If, for an existing mill, supply-side flexibility is not already available in the recovery boiler, it can be unlocked by introducing a lignin extraction plant. This synergy increases the incentives for the mill to produce lignin as a new product, as it reduces the penalty on electricity trade that is otherwise associated with lignin extraction.

The main technical limitations in relation to flexibility are the capacity limits of the flexible processes (minimum and maximum load levels), while storage units for intermediate products are typically large enough to handle variations on time-scales from hours to days. Some intermediate products, including bark, can be stored over longer periods of time, enabling some potential for flexibility also on the seasonal time-scale.

In a near-future with high shares of variable electricity supply and/or bottlenecks in the transmission grid, there is a potential economic win-win situation for the mill and the electricity system if the mill can manage its processes and storages in a flexible manner in response to market signals. However, very large variations in the spot prices for electricity or complementary incentives are necessary if the flexibility is to make a considerable contribution to the mill's economy and influence the dimensioning of its investments.

The role that the chemical pulp mill plays in the energy system may change over the coming decades. In the short term, the proposed flexibility measures may facilitate the introduction of wind and solar power into the electricity sector and, thereby, support the electrification of the transport and industry sectors. In the longer perspective, an increasing demand for biomass in response to climate mitigation efforts in relation to hard-to-abate sectors and initiatives for negative emissions may create incentives for more-transformative changes. If bio-based products or negative emissions are highly valued in relation to electricity, the role of the mill in the electricity system may change from acting as a net supplier of electricity to being a net consumer of electricity.

5.1. Future work

The results and discussions arising from this thesis suggest various directions for the development of the pulp and paper sector. Thereby, it also indicates ways in which the associated research field could be expanded.

First, some gaps could be filled in, which can be addressed using the methods presented in this work. This work could entail full investment optimisation, in which the capacities of all the processes and storages would be endogenously dimensioned for a greenfield mill. This would elucidate which flexibility measures that are cost-efficient only if they are already available, and which ones that are worth the extra investment. Such an analysis could also include additional technologies, such as black liquor gasification, to see how they compare with lignin extraction and post-combustion carbon capture. Furthermore, it would be of interest to examine how the different technologies and flexibility measures perform in different electricity systems, such as wind power-dominated or solar power-dominated systems.

Second, the characteristics of the pulp mill identified in this work could be used to create a simpler representation of a pulp mill/biorefinery unit, which could be integrated with an electricity system model on the regional or inter-regional level. Such an approach would provide better assessments of the importance of the flexibility measures for the integration of renewable electricity sources and for reducing investments in grid capacity. In addition, it would highlight the new challenges that arise for the electricity system in high-electrification pathways with e-fuels, electrified steam supply, electric-arc-plasma calcination and/or black liquor gasification. On the regional level, integration with hydrogen clusters to create synergies, for example, with the petroleum industry or hydrogen-based steel industry, could be studied. Furthermore, integration of the mill into a larger system model would allow for studies of the feedback effects that occur between the mill and the system, to determine the point at which flexibility measures that are initially economically rational due to price variations eventually contribute to evening out the prices.

Third, future work should delve deeper into the development of markets for biomass and bio-based products, through market modelling with equilibrium models that include a broad range of sources of woody biomass, conversion technologies and applications. Such models could provide valuable insights into the willingness-to-pay for different products supplied by the forestry sector, the consequences of different suggested polices and market-based incentives for negative emissions, as well as the implications for the electricity system.

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