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Lignin extraction in chemical pulp mills: The role of flexible operation

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ABSTRACT

Lignin extracted from black liquor in chemical pulp mills can potentially replace fossil carbon feedstocks in fuels and materials, thereby increasing the economic and environmental added values of woody biomass. However, since lignin extraction reduces the electricity generation of the mill, the added value depends on the characteristics of the electricity market in which the mill operates. In this study, a model mill is exposed to two different electricity price profiles: the low and steady prices of south-central Sweden in Year 2019; and the high and volatile prices of the same region in Year 2022. For the model mill, investments in lignin extraction designed to increase pulp production are economically viable and have low levels of sensitivity to electricity price levels and price volatility. The viability of lignin extraction without increased pulp production depends on the relationship between the electricity and lignin prices. With stable electricity prices, or steady mill operation, a rule-of-thumb holds that for lignin extraction to be viable, the lignin price (€/t) must be 1.8-times the average electricity price (€/MWh) plus 40 €/t for the supply of chemicals. With volatile electricity prices and flexible operation of the recovery boiler, the mill can shift the loss in electricity sales to low-price hours, thereby saving 15–70 % of the operational costs of lignin extraction, as compared to steady operation. This effect can be further enhanced by increasing the capacity of the lignin extraction process or extending the size of the black liquor storage tank. The proposed flexibility measures allow the market-integrated pulp mill to export lignin to replace fossil carbon supplies in other sectors, while supporting the electricity system during hours with high demand and low supply.

1. Introduction

Climate change mitigation requires the phasing out of fossil fuels in the heat, power, and transport sectors, as well as fossil-free replacements for materials such as plastics and carbon fibres. With a limited biomass supply, this is expected to lead to increased competition for biogenic carbon atoms, increasing the economic value of by-products from the forest industry. In the electricity system in particular, a development towards high shares of weather-dependent renewable electricity sources, such as wind and solar power, and limited biomass supply motivates that the biomass from waste fractions of the forest industry is primarily used for peak electricity production during high-price hours.

In this context, the black liquor that is generated as a by-product in chemical pulp mills is a highly relevant resource. Usually, it is combusted to meet the internal energy demands of the mill and to supply excess energy as electricity or district heating to the market. In a comparison of the internal rates of return for different pathways involving the conversion of black liquor into steam, electricity, dimethyl ether, Fischer-Tropsch liquids, mixed alcohols, methanol, acetone, butanol, ethanol or lignin, lignin extraction was found to be the most-attractive pathway [1]. In addition, lignin extraction has been shown to be a cost-effective method for removing the bottleneck represented by the pulp mill's recovery boiler, so as to increase the pulp production capacity [2,3].

Lignin extraction plants have been commercially deployed in at least three pulp mills world-wide [4,5]. At these sites, the extracted lignin is mainly used as a fuel, e.g., to replace fossil fuels in lime kilns. However, lignin has a wide range of other applications, including the replacement of fossil carbon structures in plastics and carbon fibre materials [6]. If lignin-based products can be sold at the prices currently applicable to their fossil counter-parts, the value of lignin will increase considerably [7].

Black liquor from softwood with around 20 % of the lignin extracted has no significant difference in combustion properties compared to

Abbreviations: BB, bark boiler; LE, lignin extraction; Evap1, evaporation plant part 1; Evap2L, evaporation plant part 2, lignin stream; Evap2N, evaporation plant part 2, non-lignin stream; RB, recovery boiler; RBL, recovery boiler, lignin stream; RBN, recovery boiler, non-lignin stream; WY, wood yard; Kt, kilo-tonne 1 kt = 10^6 kg.

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black liquor without lignin extraction [8], and can be combusted with only minor operational changes in existing recovery boilers [9]. Higher extraction rates have been subject to academic studies but never implemented in real mills. Moosavifar et al. [10] studied black liquor with 70 % of the lignin extracted and found that the viscosity of the black liquor decreases with an increase in lignin extraction (for the same dry solids) and that the boiling point elevation is unaffected, which is important for the evaporation process. Calculations by Vakkilainen and Kivistö [11] show that softwood black liquor with 50 % lignin reduction still has combustion properties (heating value, carbon-to-hydrogen ratio, air requirement, flue gas flows, ash formation rates, etc.) within the ranges of black liquors that have been combusted in real mills. For two specific recovery boilers studied in [11,12], however, the temperature in the lower furnace is expected to start dropping at around 30 % of lignin extraction, which may be problematic for the regeneration of the chemicals, and the superheating limits of the respective boilers are expected to be reached at around 20 % and 40 % of lignin extraction. Both these limits could potentially be altered by increasing the total flow of black liquor or by investing in a new air system [11].

Several previous studies have assessed the impact on the mill's energy balances of integrating lignin extraction, and the economic impact from the consequential loss of electricity generation. Conventional mills that have integrated the LignoBoost [4] process have been studied by Olsson et al. [13], Tomani et al. [14], and Culbertson [15]. The similar concept of LignoForce [5] has been studied by Kannangara et al. [16]. Mesfun et al. [17] have investigated combined extraction of lignin and hemi-celluloses, while Lindgren et al. [18] have studied lignin extraction integrated with dissolving pulp production, and Skoglund et al. [19] have evaluated the combined integration of lignin extraction and carbon capture.

All of these studies have found that the relationship between electricity prices and lignin prices is decisive for the profitability of lignin extraction (in addition to the profitability of increased pulp production, where applicable). Although addressing the topic from different angles, all of the above-mentioned studies consider constant prices and steady-state operation. Since a large part of the cost for lignin extraction is linked to the value of the lost electricity production and since electricity prices are highly variable on an hourly time-scale (and will be even more variable in a future with an increased share of weather-dependent electricity generation), there is a need to complement the previous studies and perform analyses that include a temporal resolution equivalent to the clearance rate on electricity markets, in order to obtain a more-realistic picture of how the electricity price influences the profitability of lignin extraction.

Svensson et al. [20] have considered daily variations in the steam demands and electricity prices. They have shown that under certain market conditions, it is profitable for the mill to invest in both lignin extraction and additional steam turbine capacity, such that electricity production can be prioritised during periods of high electricity prices and lignin extraction can be prioritised during periods of low electricity prices.

In this study, the interactions between the industrial energy system and the electricity market are studied in greater detail with a 1-hour resolution. A plant-wide model that covers all the biomass flows from the wood input to the product outputs and the steam and electricity balances throughout the mill is extended to integrate a lignin extraction technology. The model is applied to investigate the technical conditions and economic implications of lignin extraction in two different contexts: (i) lignin extraction to remove the bottleneck of the recovery boiler to ensure increased pulp production; and (ii) lignin extraction for the sale of lignin as a product. The study covers cases with levels of lignin extraction that could be implemented with current technology, but also addresses exploratory cases with high levels of lignin extraction, to assess the value of a technology development in this direction.

2. Method

2.1. Model

The reference chemical pulp mill used in this work, which is illustrated in Fig. 1 and presented in detail in our previous paper [21], is a stand-alone mill with a capacity of 2000 ADT (air-dry tonnes) per day of bleached market pulp. The model is based on the steady-state description in Åforsk Model Mills 2010 [22], which represented the best-available technology at the time of its publication. The reference mill has been selected to represent present European mills, acknowledging that there are both older, less energy-efficient mills still in operation, and newer (or improved) mills that are more energy-efficient than the reference [23]. For the present study, modifications have been made to the model, so as to represent an installed lignin extraction process similar to LignoBoost [4] as an extra intermediate step within the evaporation plant.

Conversion from biomass to steam takes place in a recovery boiler, which combusts the black liquor, and in a separate bark boiler. Electricity is produced by two generators, which are connected to one back-pressure turbine and one condensing turbine, respectively. The generated electricity and steam are supplied to industrial processes throughout the mill, to the extent that the mill is self-sufficient in terms of heat and a net-exporter of electricity. The lime kiln is assumed to be fuelled with gasified bark. (The model does not include any potential need for complementary fuels or fuels for start-up, backup etc.)

The reference mill is modelled using a linear optimisation model with hourly resolution. The objective of the optimisation is to minimise the total cost of operation over the modelled time period, by varying the material, steam, and electricity flows throughout the mill, while maintaining all the energy and mass balances within the system.

2.1.1. Evaporation plant and lignin extraction

In the present work, the unit representing the evaporation plant process (purple box in Fig. 1) is split into two parts, termed evap1 and evap2, with the lignin separation process in between, as illustrated in Fig. 2. The amount of extracted lignin is decided by a set target for produced lignin over the full time period (typically 1 year).

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). The lignin extraction is integrated into the evaporation plant according to Eqs. (1) and (2), where $M^{in}_{p,t}$ and $M^{out}_{p,t}$ are the input and output mass flows, respectively, for each process p at each hour t, and M^{lig}_{t} is the lignin extracted at time t. The λ factor is the total share of lignin in the black liquor.

$$M^{in}_{evap2L,\,t} + M^{in}_{evap2N,\,t} + \quad M^{lig}_{t} \leq \quad M^{out}_{LE,\,t} \quad \forall t \epsilon T \tag{1}$$

$$M^{in}_{evap2L,\,t} + M^{lig}_{t} = \lambda * \quad M^{out}_{LE,\,t} \quad \forall t \epsilon T \tag{2}$$

The lignin production is limited by the capacity of the lignin extraction process, $m^{LE,cap}$, as well as by the maximum extractable lignin per black liquor dry solids, $\lambda^{LE,\max}$.

$$M^{lig}_{t} \leq m^{LE,cap} \quad \forall t \epsilon T \tag{3}$$

$$M^{lig}_{t} \leq \quad \lambda^{LE,\max} * M^{out}_{LE,\,t} \quad \forall t \epsilon T \tag{4}$$

The steam consumption $S^{use}_{evap2,t}$ in the second stage of the evaporation is given by Eq. (5), Where s^{gen}_{evap2L} and s^{gen}_{evap2N} are the amounts of steam consumed per material input to the process, for the lignin and non-lignin components of the black liquor, respectively. Due to higher steam demand that occurs when lignin has been extracted (lower dry solids per mass black liquor), s^{gen}_{evap2L} has a negative value.

$$S^{use}_{evap2,\,t} = s^{gen}_{evap2L} * M^{in}_{evap2L,\,t} + s^{gen}_{evap2N} * M^{in}_{evap2N,\,t} \; \forall t \epsilon T \tag{5}$$

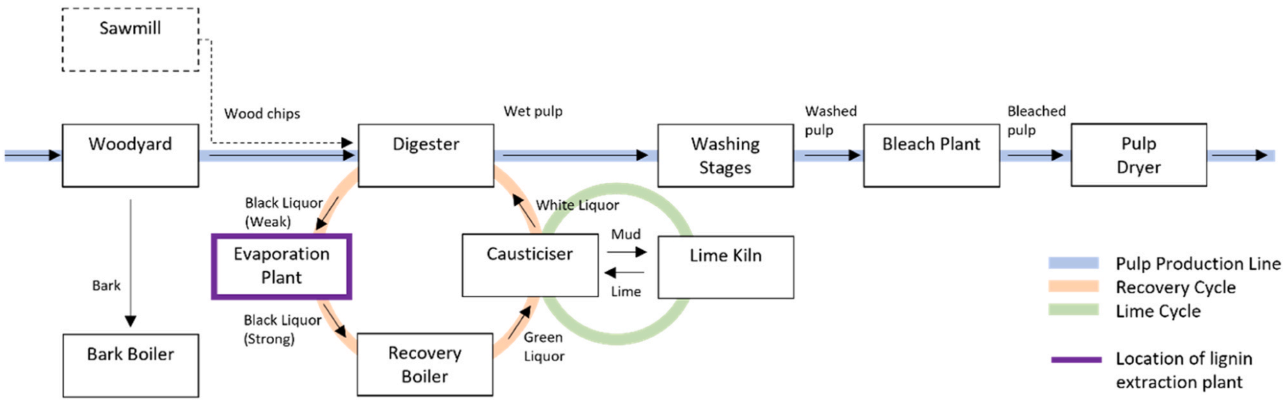

Fig. 1. Schematic overview of the material flows in the model mill.

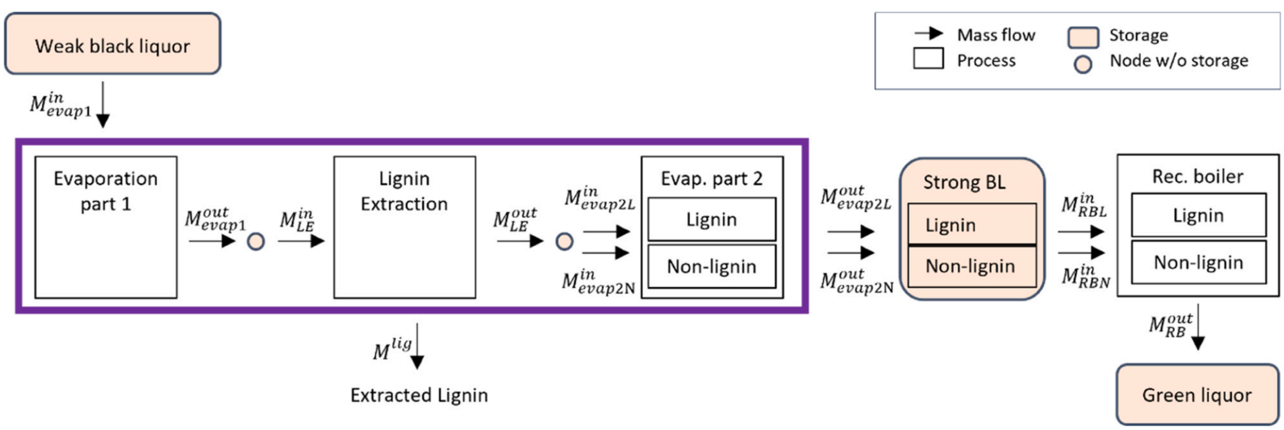

Fig. 2. Schematic overview of the modelled material flows between the evaporation plant, the lignin extraction process, and the recovery boiler. BL = Black liquor.

The strong black liquor storage unit between the evaporation plant and the recovery boiler is jointly managed for the lignin and non-lignin parts, according to Eqs. (6)–(8), where $M^{s}_{p,t}$ represents the storage level prior to process p at time t.

$$M^{s}_{RBL,\ t} = M^{s}_{RBL,\ t-1} + M^{out}_{evap2L,\ t} - M^{in}_{RBL,\ t} \quad \forall t\epsilon T \tag{6}$$

$$M^{s}_{RBN,\ t} = M^{s}_{RBN,\ t-1} + M^{out}_{evap2N,\ t} - M^{in}_{RBN,\ t} \quad \forall t\epsilon T \tag{7}$$

$$M^{s}_{RBL,\ t} + M^{s}_{RBN,\ t} \le m^{s,\max}_{RB} \quad \forall t\epsilon T \tag{8}$$

2.1.2. Recovery boiler

The recovery boiler is fed with black liquor from the storage tank, with controlled shares of lignin and non-lignin. The most fundamental limits to the recovery boiler operation are the minimum load level, the maximum load level (rated capacity), and the minimum share of lignin in the black liquor required to maintain the combustible properties of the liquor.

The minimum and maximum load levels of the boiler correspond to the energy input each hour (proportional to the hearth heat release rate). In the original model, the amount of high-pressure steam $S^{gen}_{RB,t}$ generated in the recovery boiler at each hour is proportional to the mass flow of black liquor, as given by Eq. (9), where s^{gen}_{RB} is the steam generated per material input. Due to the different heating values of the lignin and non-lignin components of the black liquor, the input to the recovery boiler $M^{in}_{RB,t}$ is here adjusted to represent the energy content of the fuel according to Eq. (10), where γ_L and γ_N are weights that are proportional to the respective heating values of the lignin and non-lignin components, respectively.

The minimum lignin content of the black liquor fed to the recovery boiler from the storage tank, $\lambda_{RB,\min}$, is restricted by the constraint in Eq. (11).

$$S^{gen}_{RB,\ t} = s^{gen}_{RB} * M^{in}_{RB,\ t} \quad \forall t\epsilon T \tag{9}$$

$$M^{in}_{RB,\ t} = \gamma_L * M^{in}_{RBL,\ t} + \gamma_N * M^{in}_{RBN,\ t} \quad \forall t\epsilon T \tag{10}$$

$$M^{in}_{RBL,\ t} \ge \lambda_{RB,\ \min} * \left(M^{in}_{RBL,\ t} + M^{in}_{RBN,\ t}\right) \quad \forall t\epsilon T \tag{11}$$

Furthermore, the recovery boiler has a limited ramping rate, allowing it at each hour to alter the load by at most half of its operational span. The green liquor passed on to the causticisation process is assumed to have the same quantity and properties as in the original process without lignin extraction.

2.1.3. Other model adaptations

Other adaptations made to the original model are that: (i) the objective function includes income for the produced pulp (instead of having a fixed production target); and (ii) the mill can trade bark with the surrounding system. Eq. (12) is the new objective function, representing the objective to minimise the total cost C of the mill. For each time-step t, $E^{use}_{p,t}$ is the electricity consumed in each process p, $E^{gen}_{\theta,t}$ is the electricity generated in each turbine θ, $M^{ext}_{p,t}$ represents the external supplies of wood, white liquor chemicals, lime and make-up water, and $M^{out}_{PD,t}$, M^{lig}_{t} and $M^{bark,sell}_{t}$ are the product outputs of dry pulp, lignin and bark, respectively, from the mill. Each of the inputs and outputs are associated with a cost c. The price for lignin, c^{lig} is the market price minus the input costs for CO_2 and chemicals. To avoid unrealistic trading patterns, the bark selling price $c^{bark,sell}$ is assumed to be 95 % of the external supply cost for bark c_{BB}^{ext}.

$$\min C = \sum_T \left[c_t^{el} \left(\sum_P E_{p,t}^{use} + \sum_\Theta E_{\theta,t}^{gen} \right) + \sum_P c_p^{ext} M_{p,t}^{ext} - c^{pulp} M_{PD,t}^{out} - c^{lig} M_t^{lig} s - c^{bark,sell} M_t^{bark,sell} \right] \tag{12}$$

Eq. (13) gives the mass balance constraint for the bark, where $M_{BB,t}^{s}$ is the bark storage level at time t, M_t^{kiln} is the bark used for the lime kiln, and a_{WY} is the input/output mass ratio in the wood yard (where the residual is bark).

$$M_{BB,t}^{s} = M_{BB,t-1}^{s} + (1 - a_{WY}) * M_{WY,t}^{in} + M_{BB,t}^{ext} - M_{BB,t}^{in} - M_t^{kiln} - M_t^{bark,sell} \quad \forall t \epsilon T \tag{13}$$

The model was implemented in GAMS [24] and solved using the IBM ILOG CPLEX Optimiser [25] on a standard laptop computer, with solution times of a few minutes per model run.

2.2. Assumptions and data

In this section, the assumptions and model input data related to the evaporation plant, lignin extraction process and recovery boiler are presented. The corresponding data for the rest of the model mill are provided in Appendix A – Model input data, and further described in our previous work [21].

The assumed capacities, through-put times and storage sizes are listed in Table 1. The lignin extraction process is assumed to be of the LignoBoost [4] type, as described by Tomani [26]. However, alternative processes exist, including an older technology from WestVaco, a process similar to LignoBoost called LignoForce, as well as the less mature Sequential Liquid-Lignin Recovery and Purification process (SLRP) [27]. The similarities and discrepancies between the processes have been discussed by Kienberger et al. [28] and Kihlman [29]. Kihlman has highlighted that the LignoBoost and LignoForce processes both increase the load on the evaporation plant, while the SLRP process does not.

In the model, the storage of weak black liquor (between the digester and the evaporation plant) is assumed to be unavailable for flexibility measures, so it has a size of 0 in the input data. Furthermore, there is no storage between the different parts of the evaporation plant and the lignin extraction plant. Thus, part 2 of the evaporation plant (*cf.* evap2 in Fig. 2) is implicitly restricted by the limits set for part 1. The energy consumption in the evaporation plant is adjusted to align with simulations of Olsson [30], accounting for an increased load in the modelled cases that include lignin extraction, in order to output strong black liquor with the same humidity (20 %) in all cases. The evaporation plant is assumed to have sufficient over-capacity to allow for the increased load.

Based on the work of Olsson [30], electricity consumption for the lignin extraction process is assumed to be 80 kWh per tonne of lignin extracted, and the duration of the extraction process is 3 h.

The assumed properties of the black liquor when the lignin content has been reduced is derived from the work of Vakkilainen and Kivistö [11] and is presented in Table 2. With more lignin extracted, a larger feed of black liquor is required to maintain the same boiler load. Extraction levels beyond 50 %, without increase in dry solids flow, are incompatible with the lower operational limit of the recovery boiler (at 70 % of the rated capacity of the boiler).

The assumed values of the maximum (momentary) extracted lignin, the minimum lignin content of the black liquor and the relative heating values of the black liquor components, used in Eqs. (4), (10) and (11), are given in Table 3. To allow for a wide range of cases to be studied, the minimum requirement for lignin content in the black liquor fed to the recovery boiler at each time step, $\lambda_{RB,min}$, is given a value corresponding to the maximum extractable lignin from the extraction process itself. This enables extraction levels up to the point where either the lower operational level of the boiler or the steam balance of the mill becomes limiting.

To study the economic impacts of electricity price levels and variations, two different price profiles (Year 2019 and Year 2022) for south-central Sweden (region SE3) are used in the study(Fig. 3). The data for Year 2019 represent typical historical conditions for the Nordic region, with higher prices during wintertime than during summertime and a yearly average price of around 40 €/MWh. The data for Year 2022 represent prices that are unusually high and volatile in the historical context, ranging from − 2 €/MWh to 800 €/MWh, with an average of 119 €/MWh. As indicated previously, future electricity prices are likely to be more volatile and may be intermediate to the two price profiles used in this study. Perfect market integration is assumed, meaning that no taxes, subsidies or other administrative costs are considered .

For the economic value of the extracted lignin, the price range of 100–500 €/t is used in the analysis. The price of 100 €/t corresponds to 14 €/MWh, which is close to the average market value of solid biomass in the Swedish fuel market in the period 2012–2021. Findings reported in the literature suggest a near-future value of kraft lignin in the range spanning from the current value to around 500 €/t (with a higher selling price of the lignin after further purification or processing into new products) [9]. Other product costs are implemented according to Table 4.

Information on the investment costs for the various lignin extraction technologies are scarce in the scientific literature. The estimates for LignoBoost reported by Olsson et al. [2] and Tomani et al. [14] were made prior to the first commercial deployment and are therefore uncertain. A press release from Stora Enso in Year 2013 reported an investment cost of 32 M€ (equivalent to 38.4 M€ in Year 2023) for a LignoBoost plant with an annual extraction capacity of 50,000 t (corresponding to 5.7 t/h, or roughly 10 % of the lignin in a mill of size similar to the model mill) [31]. Due to the large degree of uncertainty surrounding the required input data, no explicit investment optimisation is made in the present work. In the discussion of the results, annualised investment costs of 115 ± 45 €/(t/y) are used for comparison, where the intermediate value corresponds to the cost given by the above-mentioned press release from Stora Enso, combined with assumptions of 10 years of economic life-time and interest rate of 7.5 %.

Table 1
Technical assumptions made for the evaporation plant, lignin extraction process and recovery boiler, and the associated storage units up-stream of the respective process. All mass units are dry weight.

Process	Capacity [t/h]	Lower limit [% of capacity]	Duration [h]	Storage size [kt] (hours)
Evaporation plant, part 1	146	70	12	0
Evaporation plant, part 2	-	-	12	-
Lignin extraction plant	146	0	3	-
Recovery boiler	146	70	0	4 (27)

2.3. Scenarios

Two scenarios are studied for lignin extraction:

Bottleneck scenario. In this scenario, the pulp production capacity of the reference mill is limited by the recovery boiler, while there is 10 % over-capacity in all other processes (and greater over-capacities in the wood yard, pulp dryer and bark boiler; see Appendix A). The sizes of the intermediate storage units are selected to reflect a common real-world mill (see Appendix A). In this scenario, lignin extraction generates value for the mill by unlocking additional pulp production, as the bottleneck in the recovery boiler is reduced. The *Bottleneck* scenario is

Table 2
Assumptions for black liquor properties with various levels of lignin extracted. The lignin extraction levels are grouped into current, exploratory and conceptual technologies to highlight that the uncertainty of recovery boiler performance increases with decreasing lignin content in the black liquor. BL = black liquor, DS = dry solids.

	Current technology			*Exploratory*			*Conceptual*	
Lignin extracted	**0 %**	**10 %**	**20 %**	**30 %**	**40 %**	**50 %**	**60 %**	**70 %**
Lignin content (DS)	31 %	29 %	26 %	24 %	21 %	18 %	15 %	12 %
Total organic content (DS)	68 %	67 %	65 %	64 %	63 %	62 %	60 %	59 %
Inorganic content (DS)	32 %	33 %	35 %	36 %	37 %	38 %	40 %	41 %
Heating value of BL dry solids [MJ/kg]	14.1	13.8	13.4	13.0	12.6	12.1	11.6	11.1
BL feed with non-lignin stream unaltered [tDS/h]	146	132	127	123	119	114	110	106
BL feed at boiler rated capacity [tDS/h]	146	150	154	159	164	171	178	186
Boiler load at constant BL feed (DS)	100 %	97 %	95 %	92 %	89 %	86 %	82 %	78 %

Table 3
Assumptions made for the constants λ and γ.

Parameter	Description	Unit	Value
$\lambda_{ext,\max}$	Maximum (momentary) extracted lignin	%	70
$\lambda_{RB,\min}$	Minimum lignin content in strong black liquor fed to recovery boiler	%	12
γ_L	Constant proportional to heating value of lignin	-	1.79
γ_N	Constant proportional to heating value of non-lignin component of the black liquor	-	0.66

used to investigate a set of cases, as presented in Table 5. The base case without lignin extraction is compared with cases that have lignin extraction capacities in the range of 25–75 kt/year.

Balanced mill scenario. Here, the model mill is configured with over-capacities only for the wood yard, pulp dryer and bark boiler, with the remainder of the mill being perfectly balanced for steady operation. Adding the possibility for lignin extraction to this reference case does not increase the pulp production capacity. Thus, the value of lignin extraction rests solely in the income derived from selling lignin as a product. As lignin is extracted, over-capacity is unlocked in the recovery boiler, which when coupled with storage in the strong black liquor buffer tank can be used for flexible operation, so as to reduce the impact on the value of the sold electricity. The flexibility with respect to electricity prices can be further enhanced by additionally allowing over-capacity in the lignin extraction process itself or increasing the size of the black liquor storage.

The *Balanced mill* scenario is examined for different cases, spanning combinations of lignin extraction targets, lignin extraction capacity, and black liquor storage size, as presented in Table 5. The cases with 0–20 % lignin extraction are within the span which should be considered implementable in a real mill with current technology, while the target range 30–50 % rather serve to explore the potential economic implications of such operations, acknowledging that realisation of these cases may be problematic with the current technology.

Table 4
Assumptions related to the prices of traded commodities. €/t refers to dry-weight price.

Product	Value/Price [€/t]	Value/Price [€/MWh]
Wood	60	15
Pulp	1000	-
Bark	75	14

Table 5
Overview of the scenarios and cases used in this study. Each scenario spans a number of cases with different combinations of lignin extraction (LE) target, lignin extraction capacity, and black liquor (BL) storage size.

Scenario	LE annual target	LE capacity	BL storage size
Bottleneck	Unspecified	0, 25, 50, and 75 kt/year	Reference (4 kt)
Balanced mill	0–50 % of the total lignin content in the black liquor	LE target + 0 %, 5 %, and 10 % over-capacity	Reference × 1, 1.5, and 2

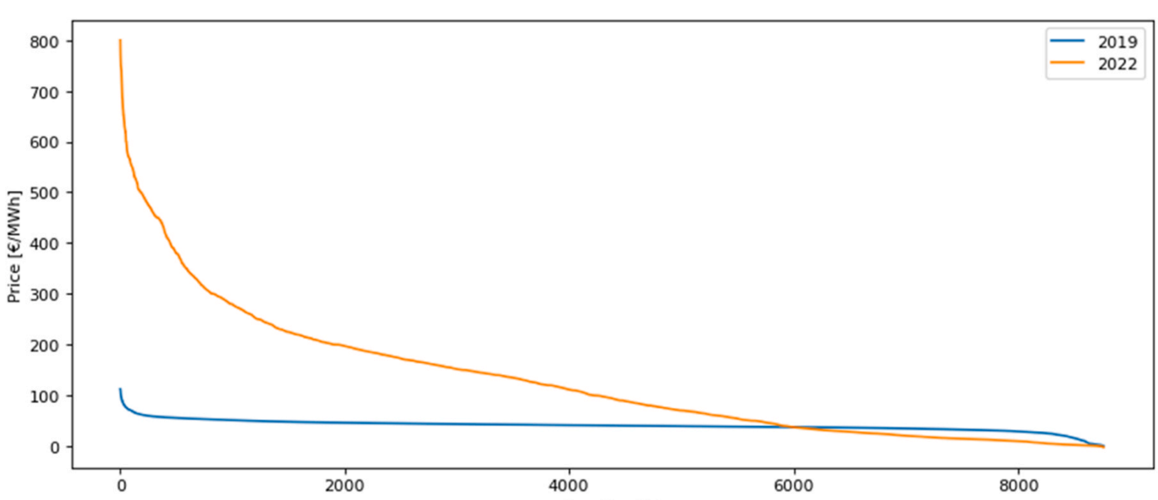

Fig. 3. Duration curves of the electricity prices from Year 2019 and Year 2022 for south-central Sweden (The SE3 region). The hours of each year have been sorted based on the electricity price, in descending order.

3. Results

3.1. Lignin extraction to relieve the bottleneck of the recovery boiler for increased pulp production

In the *Bottleneck* scenario, in which the model mill is configured with the recovery boiler as a bottleneck (10 % over-capacity for all the other processes), the modelling results show that for each tonne of lignin extracted each year, 0.9 tonnes of additional pulp are produced. The installed capacity of lignin extraction is used the maximum, up to 74.5 kt/year, or 17 % of lignin extraction. At this point, the other mill processes (including the digester) reach their maximum capacities, such that any additional increase in the lignin extraction capacity does not increase further the overall pulp production capacity of the mill. As the removed lignin is used to increase the materials input and pulp production, the recovery boiler also has a utilisation factor of 100 % in all cases.

Table 6 shows the key indicators for the *Bottleneck* scenario with lignin extraction capacities ranging from 0 to 75 kt/year. Although the steam output from the recovery boiler remains the same throughout all the cases, the electricity production from the mill is reduced as more steam is consumed internally within the mill's production processes, leaving less for the condensing turbine. In addition, the level of electricity consumption throughout the mill is increased.

In a situation with the electricity prices of Year 2019 and the assumed steady prices of bark, the mill is a net exporter of bark in all cases, since the bark boiler is already running at rated capacity for all hours in which the value of electricity production from the bark is higher than the selling price of bark. The amount of sold bark decreases with increasing level of lignin extraction, as bark is also used to cover the increasing process steam demands. For Year 2022, in which electricity prices are higher on average, while bark prices are assumed the same as for Year 2019, the mill is a net importer of bark in the base case and somewhat more electricity is produced.

Table 7 shows the value of the pulp production (579–637 M€) compared to the value of the sold electricity (24.6–22.2 M€), for increased levels of installed lignin extraction capacity (0–75 kt/year) and the electricity prices for Years 2019 and 2022. Even though the model does not take into account any income from the produced lignin, the mill's operating income increases with increased lignin extraction. This is because, for both years, the increased revenue from additional pulp production is several times higher than the revenue losses from electricity sales.

The marginal capacity cost shows the value of adding one more unit of lignin extraction. Given the investment cost range for extraction capacity, which is specified in the Section 2.2 as 115 ± 45 €/(t/year), investments should be recovered within only 1 or 2 years, as long as there is sufficient capacity in the other processes to increase pulp production and that the additional pulp can be off-set on the market. The value of installing additional lignin capacity decreases slightly with installed capacity (especially with the electricity prices of Year 2022). This is because the flexibility that existed due to over-capacity in certain electricity-consuming processes and the bark boiler before the overall increase of production, is diminished when the utilisation of these processes increases.

Table 6
Technical results for the Bottleneck scenario.

Extraction capacity [kt/year]	0	25	50	75
Cases investigated with Year 2019 electricity prices				
Pulp production [kt/year]	730	755	780	803
Lignin production [kt/year]	0	25	50	75
Bark import [kt/year]	0	0	0	0
Bark export [kt/year]	32.8	30.4	27.6	27.4
Electricity prod [GWh]	1125	1120	1112	1099
Electricity use [GWh]	527	541	554	567
Cases investigated with Year 2022 electricity prices				
Pulp production [kt/year]	730	755	780	803
Lignin production [kt/year]	0	25	50	75
Bark import [kt/year]	3.2	2.3	1.7	1.1
Bark export [kt/year]	0.7	0.7	0.6	0.3
Electricity production [GWh]	1155	1147	1141	1134
Electricity use [GWh]	527	541	554	567

Table 7
Economic results for the Bottleneck scenario.

Extraction capacity [kt/year]	0	25	50	75
Cases investigated with Year 2019 electricity prices				
Pulp value [M€]	579	599	619	637
Bark value [M€]	2.4	2.2	2.0	2.0
Net electricity value [M€]	24.6	24	23.3	22.2
Marginal capacity cost [€/(kg/h)]	6947	6811	6740	681
Cases investigated with Year 2022 electricity prices				
Pulp value [M€]	579	599	619	637
Bark value [M€]	− 0.2	− 0.2	− 0.1	− 0.1
Net electricity value [M€]	88.6	87.4	84.9	81.2
Marginal capacity cost [€/(kg/h)]	6888	6609	5976	533

3.2. Lignin extraction beyond removal of the bottleneck – impact of flexible recovery boiler operation

In the *Balanced mill* scenario, the level of pulp production cannot be increased further. Therefore, as shown in Table 8, each unit of lignin that is extracted leads to lower utilisation of the recovery boiler. The over-capacity that appears in the recovery boiler can, thus, be utilised for flexible boiler operation, within the limits imposed by the operational span of the boiler and the size of the strong black liquor buffer tank. The flexibility linked to recovery boiler operation, and thereby to the steam supply to the turbines, allows optimisation of electricity generation, such that the economic impact from extracting lignin can be minimised.

3.2.1. Implications of lignin extraction for the mill's electricity exports

Fig. 4 shows the load duration of the recovery boiler and Fig. 5 gives the corresponding net electricity production, in a case without lignin extraction (LE: 0 %) and in cases with different extraction targets (LE: 10–50 %), for the electricity prices in Year 2019. In the LE: 0 % case, the boiler is operating constantly at its rated capacity. The more lignin that is extracted, the more hours that the recovery boiler is operated at its minimum load limit (note that the y-axis in the figure does not start at zero). For 50 % lignin extraction, the recovery boiler is never operated at its rated capacity, and it operates at its minimum load limit for almost 7000 h, corresponding to 6400 full-load hours/year.

In an attempt to find whether the lower load limit of the boiler or the steam demands of the mill pose the tightest limitation to lignin extraction, the model becomes infeasible at extraction target of 56 %. At this target, the recovery boiler is pushed below its minimum operation level, even though there is still sufficient energy left in the black liquor to supply the internal demands at the mill. Thus, the results obtained from the assumed model mill configuration suggest that the upper limit of lignin extraction is around 56 % (assuming that the black liquor has combustible properties, see Section 2.2). Yet, some additional lignin could theoretically be extracted if stops are allowed in the recovery

Table 8
Pulp production, lignin production and recovery boiler utilisation for the lignin extraction targets in the range of 0–50 % of the total lignin in the black liquor, for the Balanced mill scenario.

Extraction target	0 %	10 %	20 %	30 %	40 %	50 %
Pulp production [kt/year]	730	730	730	730	730	730
Lignin production [kt/year]	0	40	79	119	158	198
Recovery boiler utilisation [%]	100	96	90	84	79	73

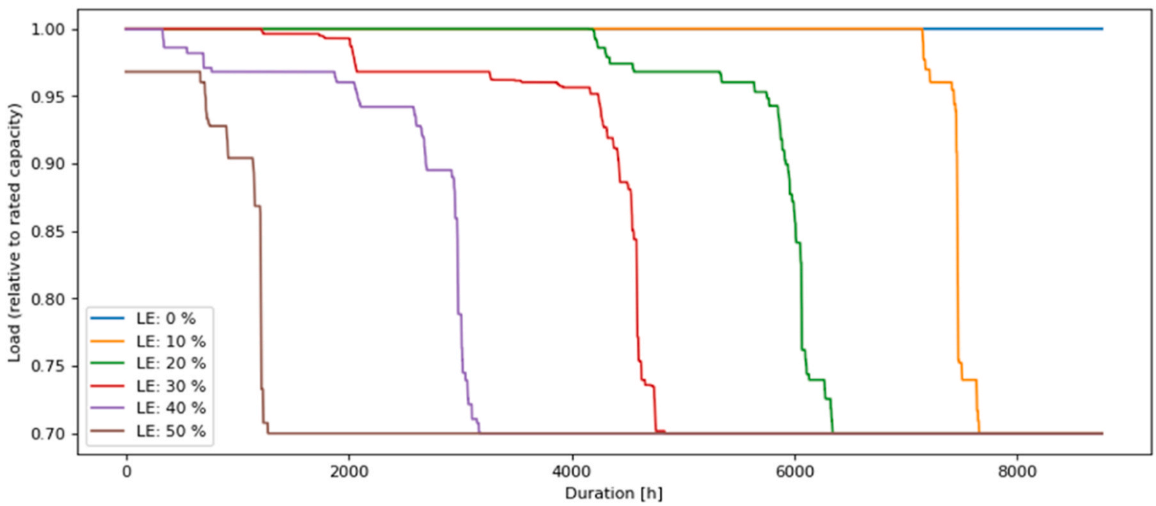

Fig. 4. Load duration curves for the recovery boiler for the five cases with different lignin extraction targets in the range of 0–50 % and electricity prices for Year 2019.

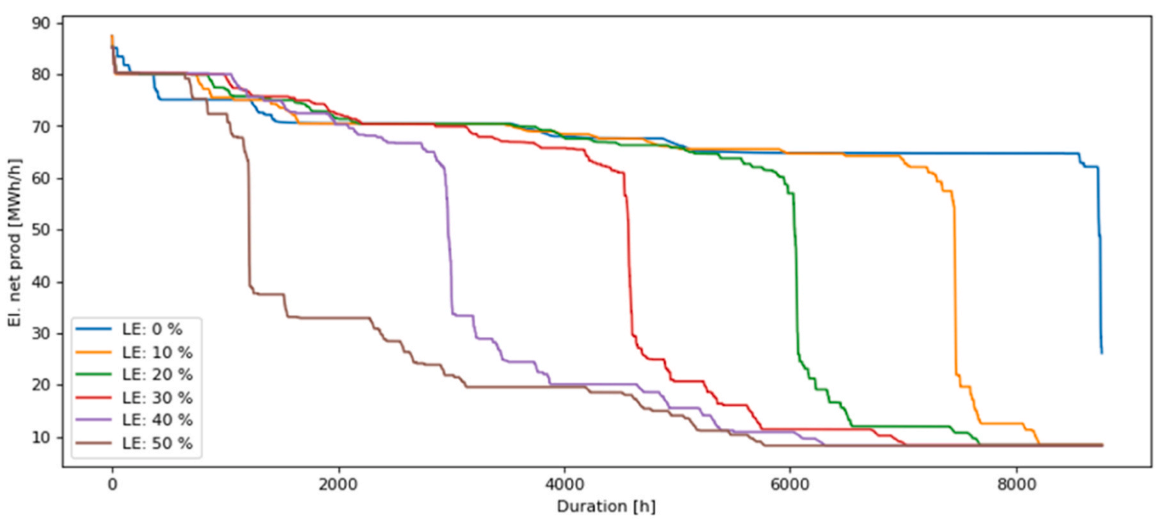

Fig. 5. Duration curves of the model mill's total net electricity production (production minus consumption) for the five cases with different lignin extraction targets in the range of 0–50 % and electricity prices for Year 2019.

boiler. However, that would immediately lead to a steam deficit in the steam-consuming processes throughout the mill.

Fig. 6 shows the impacts on aggregate net electricity production (production minus consumption; Panel a), average net electricity value (selling price; Panel b), and aggregated net electricity value (income from sales minus the costs for bought electricity; Panel c) for the electricity price profiles of Years 2019 and 2022. As can be seen in Fig. 6a, the amount of produced electricity declines linearly with increasing level of lignin extraction. As the operation of the model mill is optimised to reduce costs, the reduction in electricity generation is primarily

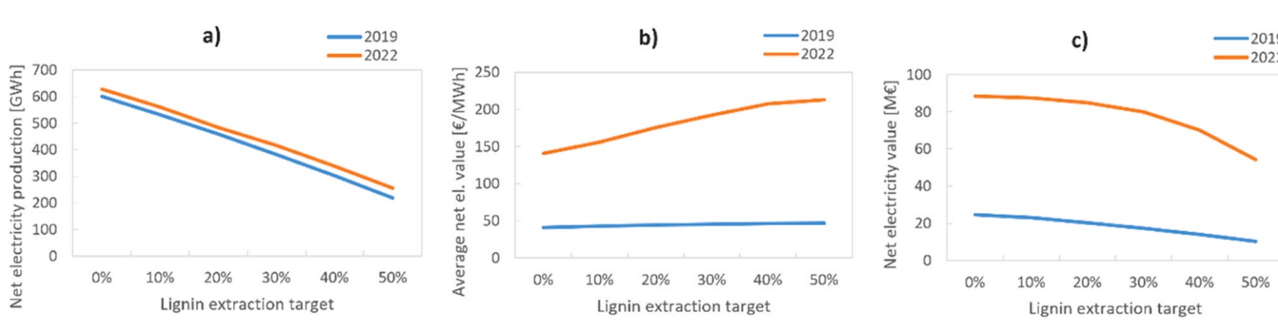

Fig. 6. Net electricity production (exported electricity; **a**), Average net electricity value (export value minus import value; **b**), and aggregated net electricity value (income from sales minus the costs for bought electricity; **c**) for the five cases with different lignin extraction targets in the range of 0–50 % and electricity prices and electricity prices for Years 2019 and 2022.

directed towards low-price hours. For Year 2019, in which year the electricity price profile is relatively flat, the average selling price of electricity is only slightly affected by the reduction in electricity production. In contrast, for Year 2022, the average selling price increases considerably as less electricity is produced during low-price hours, resulting in a weaker decline of the total net electricity value, for low targets of lignin extraction. This means that low shares of lignin can be extracted with a relatively small penalty with respect to the income from sold electricity. However, for the higher lignin extraction targets modelled, electricity production must be reduced also during more-valuable hours, leading to a flattening of the curve in Panel b and a sharper decline in the electricity value in Panel c.

Across the six cases (LE:0–50 %), the level of electricity consumption increases from 528 GWh to 544 GWh, for both Years, while the level of electricity generation drops with increasing level of lignin extraction, from 1127 GWh to 746 GWh for Year 2019 and from 1155 to 783 GWh for Year 2022. The reduction in net electricity production from lignin extraction, expressed in MWh per tonne of lignin extracted, thus comprises two parts: (i) on average, 1.7 MWh for the electricity that would have been produced from the lignin in the black liquor had it not been extracted; and (ii) around 0.1 MWh for the electricity that is consumed by the extraction process itself. As a rule-of-thumb, for lignin extraction to be more valuable than the corresponding electricity generation, the lignin prices (€/t) must thus be 1.8-times the electricity price (€/MWh), plus the operational costs of lignin extraction (here assumed to 40 €/t). For the higher levels of lignin extraction modelled, electricity generation is further reduced due to the increase in the energy demand of the evaporation plant and the decrease in turbine efficiency that occurs with reduced load.

Detailed technical and economic results related to electricity production and consumption, for the *Balanced mill* scenario, is presented in Appendix B, Table B1.

3.2.2. Implications of lignin extraction to the bark trade

Table 9 shows the import and export levels of bark for different lignin extraction capacities in the *Balanced mill* scenario. Similar to the *Bottleneck* scenario, the mill is a net exporter of bark when the electricity prices for Year 2019 are used and a net importer of bark when the process for Year 2022 are used. The more lignin that is extracted, thereby reducing the steam output from the recovery boiler, the less bark that is sold (2019) or the more bark that is purchased (2022). The utilisation of the bark boiler is increased accordingly. In a different hypothetical set-up, with a smaller bark boiler that is already running close to full utilisation in the base case, the steam balance of the mill would likely reach its operational limit at a lower level of lignin extraction.

3.2.3. Impacts of electricity prices and flexible recovery boiler operation on the production cost of lignin

Given that lignin extraction reduces the net sales of electricity and bark from the mill to the markets, the cost for producing lignin depends on the market price, and in the case of volatile prices, on the mill's ability to operate in a flexible manner. Fig. 7 presents a break-down of the average lignin production cost for each of the lignin extraction targets (10–50 %) and for the electricity prices of Years 2019 and 2022. The costs are divided into non-energy costs (primarily, for the supply of carbon dioxide), direct electricity costs (electricity consumed by the extraction process itself), and indirect costs from the reduction in net sales of electricity and bark. A steady-state case for each year is shown for comparison, assuming the average electricity price for the respective year and a reduction in electricity sales of 1.8 MWh for each tonne of lignin extracted, according to the derived rule-of-thumb. The assumption regarding average electricity prices is equivalent to an assumption of steady-state operation with varying electricity prices.

In nearly all the cases, the indirect electricity cost constitutes the largest part of the lignin production cost and, therefore, the electricity price exerts a strong impact on the lignin production cost. This is seen across the cases, when comparing the results obtained with the relatively low electricity prices of Year 2019 to the results obtained with the higher prices of Year 2022. However, one should bear in mind that energy prices and the values of feedstocks were significantly higher in Year 2022 than in Year 2019. Thus, the market price for lignin should also be expected to be higher for a year such as 2022.

Furthermore, when the recovery boiler is allowed to operate flexibly, instead of under the assumption of steady-state operation, the indirect electricity cost increases with higher extraction target. Compared to the steady-state assumption, the average cost of lignin is reduced by 15–70 % for the different targets, clearly highlighting the importance of using a time-resolved model for the assessment. The results show that in situations with volatile electricity prices, the cost of lignin extraction depends heavily on both the choice of extraction target and the mill's ability to implement flexibility measures.

Table 9
Bark trade and bark boiler (BB) utilisation for the Balanced mill scenario.

Extr. target [kt]	0 %	10 %	20 %	30 %	40 %	50 %
Year 2019						
Bark buy [kt]	0	0	0	0	0	0
Bark sell [kt]	31.4	28	19.8	13.7	9.4	7.2
BB util. [%]	41	45	52	57	61	63
Year 2022						
Bark buy [kt]	3.6	3.7	6.3	11.2	11.3	11.6
Bark sell [kt]	0.6	0.5	0.3	0	0	0
BB util. [%]	73	73	75	80	80	80

3.2.4. Marginal cost of lignin and the value of flexibility involving dimensioning the extraction process and black liquor storage

To illustrate in more detail how the electricity price affects the lignin production cost, Fig. 8 presents the marginal cost of lignin extraction in relation to the set extraction target. The graph shows the modelling results for a set of cases for each year. The blue lines indicate the cases with lignin extraction capacity equal to the target in the respective case, and black liquor storage of the reference size (the same cases as presented in the previous sections). The orange and green lines indicate those cases in which additional flexibility has been made available by adding 5 % and 10 % over-capacity to the lignin extraction process itself.

The different cases are compared with two hypothetical cases. The dashed line represents a steady-state case in which a constant lignin production cost is assumed based on the annual average electricity price (the same as in Fig. 7). The dotted line represents an ideal reference, representing a fully flexible mill, which can follow price variations in an optimal way without the model limitations regarding process capacities, storage sizes and through-put times. For the ideal case, the first unit of extracted lignin affects the mill's electricity generation only during the hour with the lowest electricity cost, and hours with higher electricity prices are used continuously up until the limit of maximum extraction, when electricity generation during even the most-expensive hour of the year must be sacrificed to fulfil the lignin extraction target.

With the electricity prices of Year 2019 (left panel in Fig. 8), the marginal cost of lignin is relatively flat, except at the very beginning and beyond the 50 % extraction target (shown in the figure only for the hypothetical case). For such electricity prices, a simplified calculation based on the derived rule-of-thumb with the average electricity price gives basically the same information. However, for Year 2022 (right panel), the graph shows that in the standard case (blue line) the model mill is sufficiently flexible to generate a large difference in cost between the lower and higher levels of extraction, pushing the cost below the steady-state alternative (256 €/t) for extraction levels up to around 17 %. Yet, it gives notably higher costs than the ideal reference line (dotted grey) at all extraction levels beyond 10 %.

With additional capacity in the lignin extraction unit (orange and

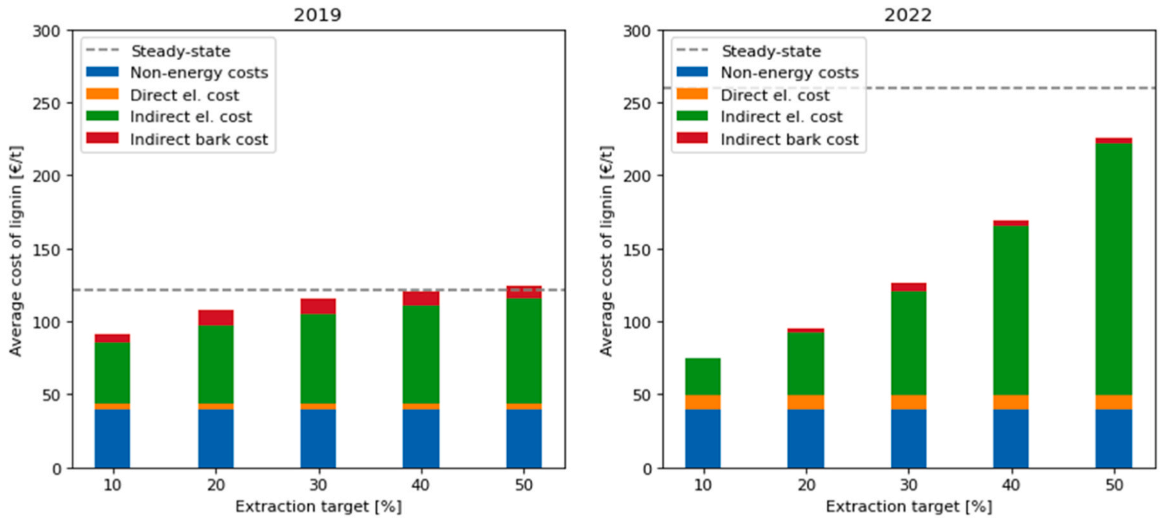

Fig. 7. Break-down of the average lignin production cost for the electricity prices of Years 2019 (left) and 2022 (right), for a set of cases with flexible recovery boiler operation spanning the different lignin extraction targets (10–50 %). Reference cases with steady-state operation are shown for comparison.

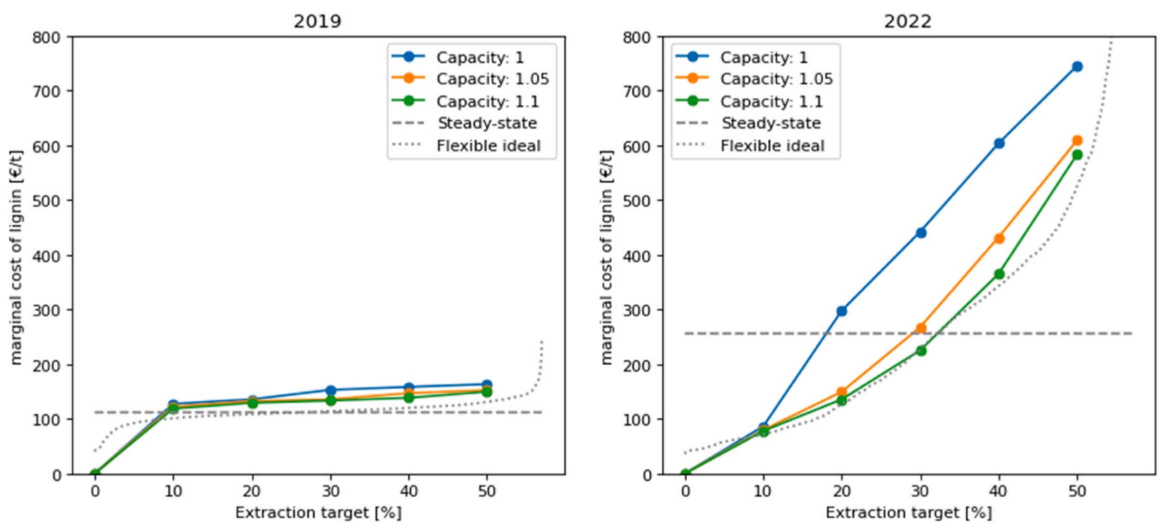

Fig. 8. Marginal cost of lignin, with flexible recovery boiler operation, for the electricity prices of Years 2019 (left) and 2022 (right), the five different lignin extraction targets ranging from 0 % to 50 %, and lignin extraction capacities equal to 1-, 1.05- and 1.1-times the target in each case. A steady-state case and an ideal fully flexible case are shown for comparison.

green lines in Fig. 8), the results migrate closer to the ideal reference. With 10 % over-capacity, the mill is flexible enough for near-perfect optimisation of the electricity generation for the price profile of Year 2022. Therefore, there is no value associated with increasing further the capacity. In this case, the marginal cost of lignin is lower than the steady-state alternative for all extraction levels up to around 30 %. At 30 % lignin extraction, the marginal cost of lignin is less than half that for the case without over-capacity (blue line). For Year 2019, the additional capacity also yields a profile closer to the reference line, although the effect in absolute numbers is small.

With the variable electricity prices of Year 2022, an effect similar to that obtained from increasing the extraction capacity can be achieved by instead increasing the size of the black liquor storage tank. With twice the reference storage size, the marginal cost of lignin is half that of the 1 × storage case for 20 % lignin extraction, matching the ideal reference, while at higher extraction levels, there is still a notable difference compared to the ideal. With 3 × the reference storage size (corresponding to 120 kt or 81 h of steady-state black liquor flow), the mill is sufficiently flexible to match the flexible ideal. For Year 2019, increasing the storage size has little impact.

3.2.5. *Revenues and investment cost of lignin extraction*

Fig. 9 presents the aggregated impact on the revenue of the mill from lignin, electricity, and bark trade, using the case without lignin extraction as baseline. The different lines represent different lignin price assumptions (range, 0–400 €/t). Annualised investment costs in the range of 115 ± 45 €/t annual lignin extraction capacity are shown for comparison (shaded section), under the assumption that investment costs scale linearly with investment size. In this simplified economic calculation, any combination of lignin price and investment cost is profitable if the revenue is greater than the investment, and the size of the profit is given by the difference between the costs and the revenue. Roughly speaking, this means that to cover the investment the lignin price must be 100–200 €/t higher than the operational costs.

For the flatter electricity price profile of Year 2019 (left panel in Fig. 9), the revenue increases linearly with increasing extraction target, from 10 % extraction onwards. Thus, any combination of electricity price, lignin price, and investment cost that supports a viable investment

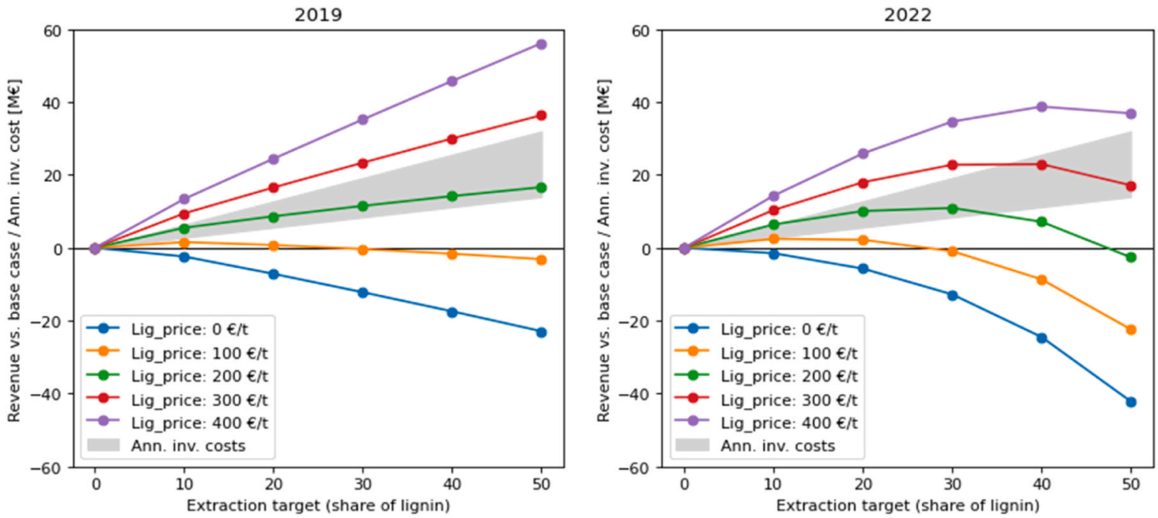

Fig. 9. Aggregated impact on mill revenue from lignin, electricity and bark trade, for the electricity prices of Years 2019 (left) and 2022 (right), for a set of cases with lignin extraction targets in the range of 0–50 %, with the case without lignin extraction used as baseline, and with different lignin price assumptions. The shaded section shows the annualised investment cost range of 115 ± 45 €/t, scaled linearly with the extraction target, for the purpose of comparison.

for low extraction levels, will do so also for higher extraction levels. For Year 2022 (right panel in Fig. 9), the picture is different. Due to the highly variable electricity prices, the revenue curves representing different lignin prices reach their respective maxima at different extraction targets. As a consequence, each combination of electricity price, lignin price and investment cost will define a unique optimum level of lignin extraction to maximise profits. These graphs represent the case without over-capacity in the lignin extraction plant and with the reference storage size; and the non-linear effect would be even more pronounced for a case with additional flexibility.

4. Discussion

The two scenarios used in the study are a stylistic representation of the two ways in which lignin extraction can produce value for the mill: (i) indirectly through increased pulp production capacity; and (ii) directly through the value of lignin as a product. In real mills, however, the conditions are likely to lie somewhere between these two cases. For example, the recovery boiler may not be a bottleneck in a technical sense, but rather the largest and most long-lived and expensive unit to invest in, and thus, entail the largest financial risk with respect to investments in additional capacity. Lignin extraction may, in such cases, be an attractive route to increase pulp production at lower risk, and an option that may be considered in the middle of an investment cycle for the recovery boiler, i.e., before reaching its technical lifetime. Another possible scenario is to have a mill that is configured as in the *Bottleneck* scenario, in which lignin is extracted at a higher level than the bottleneck limit, such that the recovery boiler capacity is not utilised fully. In this case, similar trade-offs between electricity production and lignin sales as those seen in the *Balanced mill* scenario will occur. The main differences compared to the *Balanced mill* scenario would be that the lignin concentration in the black liquor would be lower, potentially reaching the non-combustible limit earlier, and that the load in the evaporation plant would be higher, potentially resulting in a new bottleneck earlier.

The largest uncertainty regarding the applicability of the model results to a real case is the combustion properties of the lignin-reduced black liquor and the thermal performance of the recovery boiler at the high extraction targets, especially in combination with flexible operation. In light of this, it should be noted that the highest impact of flexible operation, which is the main focus of the study, is obtained at the lower extraction targets. For all lignin prices and investment costs examined together with the variable electricity prices (Fig. 9), the largest span between revenue and investment cost is found at extraction targets of 30 % or lower.

Furthermore, each mill is unique with regards to the installed capacities of the processes, boilers and turbines, the sizing of intermediate storage units, the steam pressure and temperature levels, and the measures of heat recovery. Most Nordic pulp mills are relatively old and are typically less-energy-efficient than model mills representing the best-available technology, like the one used in the present work. For the integration of a lignin extraction process, this means that real mills typically have lower levels of surplus energy for electricity generation, and therefore have a lower limit as to how much lignin that can be extracted while still maintaining the steam supply to the production processes. A lower maximum extraction limit also implies that the optimal extraction level with volatile electricity prices will be lower.

The technical limits of the operational spans for the processes and sizing of storage have, however, been selected to resemble as much as possible a typical real mill, based on dialogue with industry representatives. Thus, despite the unique characteristics of each real mill, the results clearly demonstrate how the smart implementation of lignin extraction can lead to a high level of flexibility in electricity trade. Although there are large variations in the mass and energy flows within the mill, the product outputs of pulp and lignin are totally or almost stable. This means that neither upstream nor downstream actors in the value chain are affected by the proposed measures, and deliveries can be contracted as if the mill was operating under steady-state conditions.

For typical historical, stable, electricity prices, the benefit of using a dynamic model compared to a steady-state model is negligible. However, given unstable electricity prices resembling those seen in Year 2022, the cost of lignin production may be highly over-estimated if the average electricity price is used, or if the mill is assumed to operate under steady-state conditions. While it is unclear as to how electricity prices will develop in the future, with an increasing share of weather-dependent electricity generation, it seems likely that the variations will resemble those of Year 2022, even if the average price may be lower.

The results show that with the relatively stable electricity prices of Year 2019, a lignin price that is slightly higher than the historical fuel price for solid biomass would be sufficient to motivate lignin extraction at the highest achievable level. This follows naturally from that fact that the value of combusting the lignin as black liquor in the recovery boiler or combusting it elsewhere at another time will generate the same amount of electricity. Since biomass prices are linked to electricity prices through its value as a fuel, it seems likely that the lignin price is also higher in situations with high electricity prices. If the market price for lignin is based on its value as a fuel and the electricity price profile is as flat as in Year 2019, the situation where lignin extraction is more valuable than the corresponding electricity generation would only occur for a few hours of exceptionally low electricity prices. However, since lignin is storable, the market price for lignin could be expected to match (or even exceed) the average electricity price. If electricity prices are volatile, such as for Year 2022, this means that it could be valuable to extract lignin during low-price hours and thereafter use it for electricity generation (internally or externally) during high-price hours. In this way, the extraction, storage, and combustion of lignin is used as an electricity storage system with a round-trip efficiency of around 85 %, when accounting for the electricity consumed by the extraction process (0.1 MWh el./t) and the additional steam demand in the evaporation plant when lignin is extracted (0.6 MWh steam/t). The round-trip efficiency might be improved by replacing the LignoBoost process with the SLRP process, which would not affect the steam demand in the evaporation plant.

With the emergence of new applications for lignin in transportation fuels or materials, it may be more profitable to extract lignin than to generate electricity, even during electricity peak-price hours. However, if fossil fuels are also limited, it is not certain that the willingness-to-pay will be lower in the electricity sector. From a climate change mitigation perspective, shifting electricity generation away from low-price or average hours (when renewable energy supply is sufficient to meet the demand) to replace the use of oil, gas, and coal during peak hours may contribute significantly to reducing greenhouse gas emissions from the electricity sector.

Whether lignin is best used for electricity generation or as a material depends on several factors that are beyond the scope of the individual mill. Nevertheless, it is safe to say that if the world changes in line with the climate targets and fossil sources of carbon are phased out, many different activities will be competing for the woody biomass. In such a future, it is unlikely that continuing with business-as-usual and combusting black liquor for heat and electricity generation during all hours of the year will be the most-cost-efficient or climate-efficient pathway. As the present study shows, lignin extraction allows for a larger share of the biomass to be utilised for the production of premium products or high-value electricity, as compared to the conventional pulp process, in alignment with the cascading principle. However, apart from the different applications for lignin, the pulp and paper sector has available alternative mature technologies, including black liquor gasification, carbon capture for storage or utilisation, and the shifting away from chemical pulp mills to thermo-mechanical or chem-thermo-mechanical pulp mills with a higher share of the wood ending up in the paper product. There are also wood-based bio-refinery concepts in or near commercial operation that are disconnected from the pulp and paper sector, e.g. the production of lignin and sugars via enzymatic hydrolysis [32]. It is our recommendation that future research should continue to study all these different pathways and compare them in terms of the added value created from the green carbon atoms and their respective contributions to climate change mitigation.

5. Conclusions

The chemical pulp mill model established in our previous work has been expanded to include an integrated lignin extraction technology. The time-resolved model enables an assessment of the dynamics in the mill when the processes, boilers and turbines respond flexibly to hourly electricity prices, which is shown to be especially important during periods of volatile electricity prices.

The results show that if the recovery boiler is the bottleneck in the pulp production process, i.e., if the capacity of the boiler is constraining the total output of the mill, eliminating the bottleneck of the boiler by extracting lignin from the black liquor represents an inexpensive opportunity to increase pulp production, with little dependence upon the market prices for electricity and lignin. For the model mill, producing 730,000 adt of pulp per year, the extraction of 75 kt lignin per year enables a 10 % increase in pulp production.

Whether or not there is an economic incentive for the mill to invest in lignin extraction capacity beyond eliminating the bottleneck, i.e., for selling lignin as a product, depends on the combination of lignin price, electricity price, and the investment cost for the lignin extraction technology. From this work, a rule-of-thumb is derived, which states that for lignin extraction to be more valuable than the corresponding electricity generation, the lignin price (€/t) must be 1.8-times the electricity price (€/MWh), plus the operational costs of lignin extraction (assumed to be 40 €/t).

With variable electricity prices, the economic case for lignin extraction is strengthened considerably if the recovery boiler is allowed to operate in a flexible manner in response to market signals, utilising the black liquor storage tank as a buffer. Through this measure, the lost income linked to electricity sales associated with lignin extraction is reduced, thereby lowering the average cost of lignin by 15–70 % compared to steady-state operation, where the highest impact is attained for the lowest extraction target.

Adding up to 10 % over-capacity to the lignin extraction plant further enhances the flexibility and, thereby, reduces the marginal cost of lignin by up to 50 %. A similar effect can be achieved by increasing the size of the black liquor storage by a factor of three, which also reduces the marginal cost of lignin by up to 50 %.

In summary, we show how market-integrated pulp mills can export lignin to replace fossil carbon supplies in other sectors, while still supporting the electricity system during hours with high demand and low supply.

CRediT authorship contribution statement

Simon Ingvarsson: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Mikael Odenberger:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Filip Johnsson:** Writing – review & editing, Validation, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Model input data

Tables A1–A5 present the technical and economic parameters that are assumed for the model mill, in addition to the data presented in the Section 2. The full model presentation can be found in our previous paper [21].

Table A1
Technical properties of the processes. The given capacities are steady-state values, and the flexibility spans are given in relation to the steady-state values. Mass units are dry-mass equivalents. The digester and causticiser have dual inputs and outputs, as shown in Fig. 1.

Process	Capacity	Duration	External supply limit	Storage size	Flexibility span
Unit	t/h	h	t/h	kt	%
Wood yard	136	0	∞	∞	0–120
Digester	119[a]/34	3	0/1	30/1.5	
Washing stages	89	2		-	
Bleach plant	85	6		-	
Pulp dryer	83	1		1	80–120
Causticiser	58/23	2	0/1	2	
Lime kiln	22	4		-	
Bark Boiler	9	0		5	55–140

[a] Excluding sawmill wood chips.

Table A2
Properties of the energy carriers included in the model.

Energy carrier[a]	Pressure [bar]	Temperature [°C]	Enthalpy [MWh/t]
HP steam	100	505	94.06
MP steam 2	25	275	81.78
MP steam 1	9	200	78.53
LP steam	3.5	150	76.33
Condensate 4	10	190	22.42
Condensate 3	6	160	18.75
Condensate 2	3.5	140	16.36
Condensate 1	1	100	11.64
Condensate 0	0.05	35	5.75
Feedwater	4	146	17.31
Make-up water	1	15	1.75

[a] HP = High-pressure, MP = Mid-pressure, LP = Low-pressure.

Table A3
Energy consumption (positive values) and generation (negative values) per processed material for each process. Unit-less entities are tonnes of steam/water per tonne of material.

	HP	MP2	MP	LP	Cond4	Cond3	Cond2	Cond1	Feedwater	Electricity [kWh/t]
Wood yard										28
Digester			0.28							22
Washing stages			0.03							113
Bleach plant			0.08							79
Pulp dryer				0.88				− 0.79		170
Evaporation, part 1			0.05	0.40			− 0.43			8
Evap2: lignin			− 0.11	− 0.86			0.93			
Evap2: non-lignin			0.13	0.97			− 1.04			8
Lignin extraction										8
Recovery boiler	− 4.05	0.45	0.25		− 0.37	− 0.13			4.07	
Causticiser										86
Lime kiln										
Bark boiler	− 5.45	0.06	0.19	0.28		− 0.46			5.47	
Other		− 0.04	0.03	0.13				− 0.06	0.03	179

Table A4
Properties of the modelled turbines. The given inlet and exhaust flows are steady-state values, and the flexibility spans for each turbine are shown in relation to the steady-state values. Corresponding flexibility spans are applied to the exhaust flows (upper limit only).

Turbine[a]	Level	Inlet [t/h]	Exhaust [t/h]	Coeff. α [MW/t]	Offset β [MW]	Flexibility span [%]
BPT	**HP**	431		0.101	− 13.5	50–120
	MP2		63	0.061		
	MP		111	0.071		
	LP		257			
CT	**HP**	210		0.352	− 6.6	20–120
	Cond0		210			

[a] BPT = Back-pressure turbine, CT = Condensing turbine.

Table A5
Cost assumptions made in this study.

Resource	Cost [€/t]
White liquor chemicals	370
Lime	120
Make-up water	0.2

Appendix B. Additional results

Table B1
Technical and economic results related to electricity production and consumption, for the Balanced mill scenario.

Year 2019						
Extraction target	**0 %**	**10 %**	**20 %**	**30 %**	**40 %**	**50 %**
Electricity production [GWh]	1127	1060	987	910	830	746
Electricity use [GWh]	528	531	534	538	541	544
Net electricity production [GWh]	599	529	453	372	289	202
Electricity production value [M€]	45.8	44.2	41.6	38.6	35.2	31.5
Electricity consumption value [M€]	21.2	21.3	21.5	21.6	21.7	21.8
Net electricity value [M€]	24.6	22.9	20.1	17.0	13.5	9.7
Average production price [€/MWh]	40.6	41.6	42.1	42.4	42.4	42.2
Average consumption price [€/MWh]	40.1	40.2	40.2	40.2	40.2	40.2
Reduced electricity production [MWh/t_lign]	-	1.7	1.78	1.83	1.89	1.93
Year 2022						
Extraction target	**0 %**	**10 %**	**20 %**	**30 %**	**40 %**	**50 %**
Electricity production [GWh]	1155	1089	1011	944	866	783
Electricity use [GWh]	528	531	534	538	541	544
Net electricity production [GWh]	627	558	477	406	325	239
Electricity production value [M€]	153	152.3	149.9	144.9	135	119.2
Electricity consumption value [M€]	64.6	65.3	65.7	66.0	66.4	66.8
Net electricity value [M€]	88.4	87.0	84.2	78.9	68.6	52.4
Average production price [€/MWh]	132.4	139.9	148.2	153.5	156	152.3
Average consumption price [€/MWh]	122.5	123.0	123.0	122.9	122.9	122.9
Reduced electricity production [MWh/t_lign]	-	1.68	1.84	1.78	1.84	1.88

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