

Carbon Footprint and Techno-economic Analysis to Decarbonize the Production of Linerboard *via* Fuel Switching in the Lime Kiln and Boiler: Development of a Marginal Abatement Cost Curve

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The US Pulp and Paper (P&P) industry heavily relies on fossil sources, with lime kiln operations posing a significant challenge for achieving zero on-site fossil emissions. This study assesses the greenhouse gas (GHG) reduction potential and costs associated with alternative fuels in lime kiln operations for linerboard production. Various options, including bio-based fuels including pulverized biomass, gasification of biomass, crude tall oil, bio-methanol, and traditional fuels such as fuel oil and petcoke, were analyzed through detailed process simulations and Life Cycle Assessment. Results indicate that per ton of product, 2,789 kg of CO₂-eq is emitted, with 69% being biogenic CO₂ and 31% fossil CO₂-eq. Notably, replacing the natural gas boiler with a biomass boiler reduces Global Warming Potential (GWP) by 41%, while switching lime kiln fuel to biofuels achieves a 5.5% reduction. Combining a biomass boiler with pulverized biomass fuel use in the lime kiln yields a substantial 93.1% reduction in Scope 1 and 2 emissions, at a cost of \$76/ton of CO₂-eq avoided.

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Synopsis

The effect of switching fossil fuels with bioenergy to decarbonize the production of linerboard is revealed by an integrated environmental and economic evaluation and the construction of the Marginal Abatement Cost Curve

INTRODUCTION

The US Pulp and Paper (P&P) Industry has the third highest energy demand of all industrial sectors behind chemical manufacturing and petroleum/coal industries, with 8.7 trillion BTU per year (IEA 2022). Although most of the energy comes from renewables, the industry still has a high dependency on fossil fuels, which represent significant contributions to GHG emissions. The lime kiln is one of the larger users of fossil fuels. In the kiln, calcium carbonate is calcinated to regenerate calcium oxide, which is used to causticize sodium carbonate in the green liquor to form sodium hydroxide, reducing the demand for pulping chemicals in the system (Tran 2007).

The variation in the prices of fossil fuels and the commitment to reduce GHG emissions have driven the adaptation of renewable sources in the operation of lime kilns. For example, 90% of the energy demand in Swedish lime kilns is supplied by biofuels, including tall oil pitch (63%), wood and bark dust (24%), and methanol combined with non-condensable gases (NCGs) (3%). In Finland, 42% of the energy is supplied with biofuels, the most common being biomass gasification (18%), followed by tall oil pitch (13%), wood dust and lignin (8%), and methanol/NCGs (6%) (Berglin and Von 2022). Biofuels have shown little operational difference compared to fuel oil or natural gas (Berglin and Von 2022) and it is estimated the replacement of natural gas or fuel oil with bio-based fuels in lime kilns represents a 10% reduction in the GHG emitted by the European P&P industry (Taillon *et al.* 2018).

The US pulp and paper (P&P) industry needs to adopt more efficient technologies to match the energy performance of European mills. Compared to their European counterparts, US mills are generally less energy-efficient, consuming more energy per ton of product. European mills have achieved higher energy efficiency, allowing them to utilize biomass excesses and coproducts as energy sources in lime kiln operations. On the contrary, natural gas is the main fuel in lime kiln operations in the US. Before fracking for natural gas in the early 2000s, natural gas was so expensive that several mills burned bio-based coproducts available in the mill rather than using natural gas (Francey *et al.* 2009; Manning and Tran 2015; Hart 2020a,b). After widespread implementation of fracking, the price of natural gas decreased and pulp and paper mills began to implement more cheap natural gas fuels in their processes.

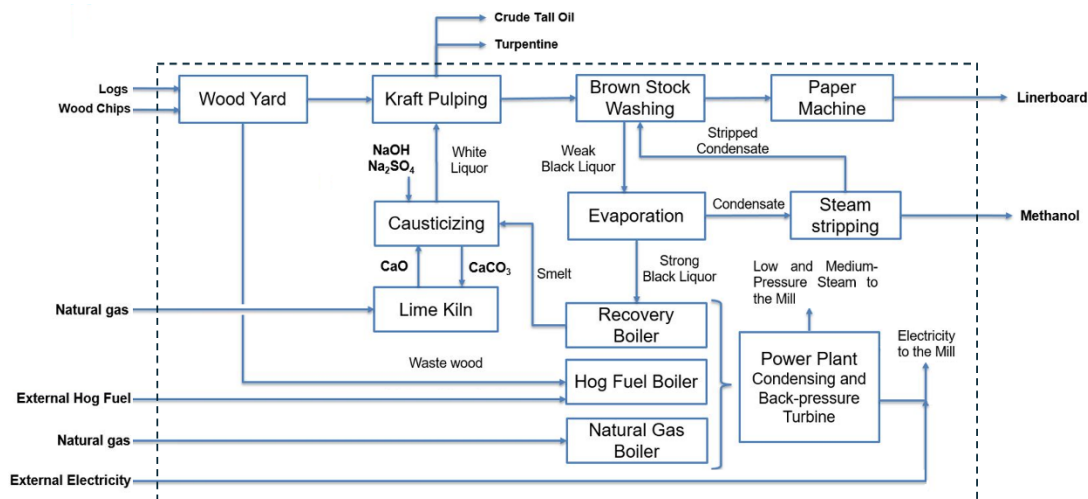
Recently, the US government has set the goal of 50 to 52% GHG reductions below 2005 levels by 2030, covering all sectors, followed by a net-zero emissions no later than 2050 (Kerry and McCarthy 2021). These ambitious goals and the unpredictable fluctuation in fossil fuel prices are leading the US P&P to incorporate technologies to reduce the GHG emissions.

The use of bio-based fuels may represent a reduction in on-site fossil emissions. Still, the transformation of raw materials into suitable lime kiln fuel (pulverized or gasified biomass) or the extraction and adaptation of secondary streams from the process (lignin, methanol, crude tall oil (CTO), or tall oil pitch (TOP)) implies indirect emissions that might diminish the benefit achieved. Moreover, the alternatives may represent an additional cost for the mill, making them less attractive or nonviable depending on operating conditions. While the use of bio-based fuels may represent a reduction in on-site fossil emissions, there are practical considerations such as the generation of ash, which can affect costs and efficiency by the buildup of insulating layers from deposits. Previous studies have shown the economic and environmental benefits of incorporating alternative fuels in lime kiln operations when surplus biomass and surplus electricity are available in the mill, it is possible to reduce GHG emissions and assure the economic viability of the alternatives (Kuparinen *et al.* 2016, 2017; Kuparinen and Vakkilainen 2017). However, these conditions are contrary to those faced by the US P&P industry.

The present study evaluated various renewable fuels for lime kiln operations in the production of linerboard, one of the largest and growing sectors in US P&P industry (Elhardt 2017). The alternatives include pulverized or gasified biomass, CTO, TOP, bio-methanol, turpentine, and lignin. Additionally, other traditional lime kiln fuels were evaluated (fuel oil, petcoke, and tire-derived fuel (TDF)), as well as the replacement of the natural gas boiler by a biomass boiler. The net fossil CO₂ reductions of the alternatives were determined through a detailed process mass and energy balance simulation using

MATERIALS AND METHODS

The mill in this work is a continuous linerboard unbleached mill, which is a virgin grade (new, unused wood fibers), with a production of 100 short ton per hour or 90.72 tons/h. The configuration and operating conditions were defined based on information reported in the literature and databases and industry experts' recommendations (Rydholm 1967; Grace *et al.* 1983; ResourceWise 2023; Fastmarkets 2023). Detailed information is included in the supporting information section (Appendix). Figure 1 shows the system boundary for the Cradle-to-Gate Life Cycle Assessment (LCA) developed and the main areas that compose the mill.



The life cycle inventory is based on the mass and energy balance for a mill configuration modeled in WinGEMS (Metso, version 5.3, Espoo, Finland), a specialized process simulation software for the P&P industry. The Ecoinvent database was used to determine the contribution of the upstream processes. The GWP was determined using the IPCC 2013 GWP 100a method, available in OpenLCA. The method expresses GHG emissions, in kilograms CO₂ equivalent, over a time horizon of 100 years. A mass allocation factor is used to allocate the GWP among the different coproducts in the system.

Evaluation of Alternatives to Reduce the GWP

The combustion of alternative lime kiln fuels, and the biomass boiler were incorporated into the base simulation model. The scenarios evaluated are in Table 1. For each scenario, the linerboard production remained the same; some of the fuels can substitute for 100% natural gas in the lime kiln (fuel oil, pulverized biomass, biomass gasification, CTO, and TOP), whereas others have limited substitution (methanol, turpentine, petcoke, and TDF) (Francey *et al.* 2009; Taillon *et al.* 2018; Hart 2020a,b). The GWP of the scenarios was estimated based on a Cradle-to-Gate LCA by implementing the IPCC 2013 GWP 100a method.

The alternatives were classified into four groups; the first was the replacement of the natural gas boiler with a biomass boiler to produce steam and electricity for the mill. The second group corresponds to external bio-based fuels that can displace 100% of the natural gas demand in the lime kiln. The third group corresponds to fuels that are available in the mill, such as CTO, methanol, and turpentine, or it can be extracted from the streams available in the mill, which is the case of lignin. The last group corresponds to other fossil fuels that can be burned in the lime kiln. The conditions for integrating each alternative are included in the supporting information section.

Table 1. Alternative Technologies to Reduce the GWP in the Production of Linerboard

Scenarios	Fuel Use		
	Min (%)	Medium (%)	Max (%)
Base case: Natural gas	-	-	100
1. Replacement of natural gas boiler with a biomass boiler			
2. External bio-based fuels			
2.1 Pulverized biomass	25	50	100
2.2 Biomass gasification	-	-	100
2.3 Tall oil pitch (TOP)	25	50	100
3. Bio-based products or bio-based streams available in the mill			
3.1 Crude tall oil (CTO)	25	50	100
3.2 Lignin	25	-	50
3.3 Methanol	-	-	10
3.4 Turpentine	-	-	10
4. Other fossil-based fuels			
4.1 Fuel oil	-	-	100
4.2 Petcoke	25	50	85
4.3 Tire-derived fuels (TDF)	-	-	15

RESULTS AND DISCUSSION

Carbon Footprint

To develop a representative picture of carbon footprint for linerboard production and to evaluate improvements in such, a detailed process simulation was developed in

WinGEMS. The operating conditions were based on both literature values and information from industrial experts. Baseline and various scenario mass and energy balance simulations were determined. The results for each case are listed in the supporting information section. These data, along with the LCI from the Ecoinvent database (Wernet *et al.* 2016), were entered into OpenLCA to estimate the GWP.

Figure 2 shows the total CO₂-eq emissions in the production of linerboard for the baseline case. A total of 69% of the total emissions correspond to biogenic CO₂; of these emissions, 82.3% came from black liquor combustion, the primary energy source in the process; 12.3% came from the biomass boiler that burns residual biomass from the woodyard and external hog fuel, and 5.4% came from the lime kiln. The lime kiln has both anthropogenic CO₂ from burning natural gas and biogenic CO₂ from the CaCO₃ conversion to CaO and CO₂. The biogenic CO₂ from CaCO₃ originates from Na₂CO₃ from the black liquor burnt in the recovery boiler. In this case, the ratio between the fossil and the biogenic CO₂ in the lime kiln is 66% biogenic to 34% fossil CO₂.

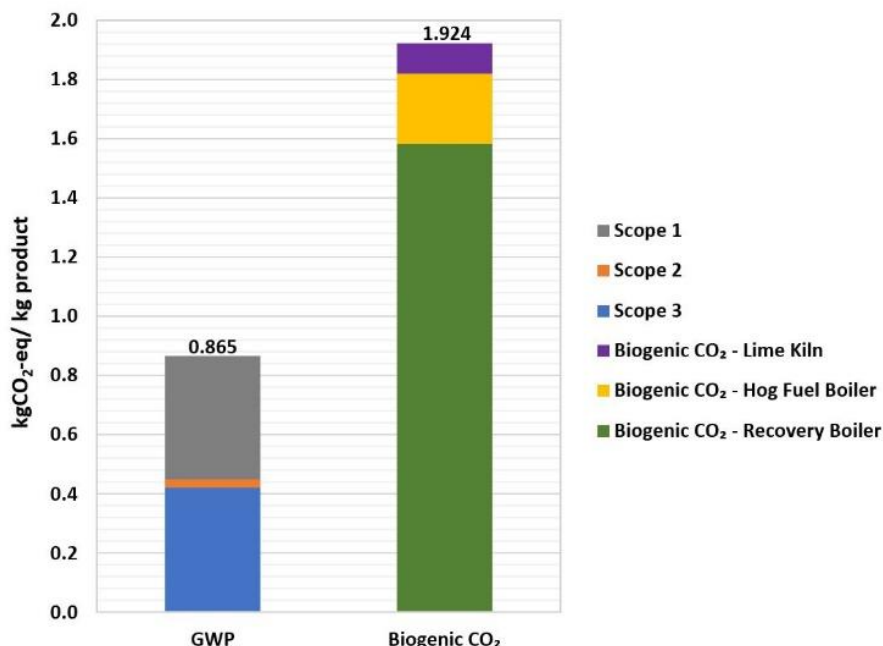


Fig. 2. CO₂-eq emissions in the production of one machine dry (10% moisture) kg of linerboard product

Regarding the GWP, the linerboard production has a total emission of 0.865 kg CO₂-eq / kg machine dry (MD) product (10% moisture content). Of these emissions, 48.1% are on-site emissions (Scope 1), 48.6% are indirect emissions from upstream processes and the disposal of waste (Scope 3), and 3.3% are from the purchase of electricity (Scope 2). Note the purchase of electricity is low because there is significant on-site production of electricity. The total emissions are similar to those reported in the literature for unbleached paperboard (0.714 kg CO₂-eq/kg product as an industry wide average) (Hart 2020b), and the process reported in Ecoinvent 3.8 as “containerboard production, linerboard, kraftliner-Rest of the world” (0.735 kg CO₂-eq/kg product) (Francey *et al.* 2009). The differences in the results arise from assumptions made in the simulation model and in the LCA model used herein. In the present study, the demand for raw materials and emissions are based on mass and energy balances from the process simulation, assuming standard operating

parameters in the industry for this type of pulp grade; in contrast, the referenced cases were based on a top-down approach, integrating average values of the industry to a production line level.

To have a detailed view of the sub-process contributions, a hotspot analysis was performed to identify critical sub-processes. Table 2 shows the detailed contribution of each process to the GWP.

Table 2. GWP Contribution of the Different Areas Involved in the Production of 1 kg of Linerboard

SCOPE	Process	Subprocess	GWP (kg CO ₂ -eq/kg product)	Contribution (%)
Scope 1	On-site emissions	Fossil CO ₂ Lime kiln	5.34*10 ⁻⁰²	6.17%
		Fossil CO ₂ Natural gas boiler	3.63*10 ⁻⁰¹	41.92%
Scope 2	Electricity from the grid	Electricity demand	2.85*10 ⁻⁰²	3.30%
Scope 3	Production of external fuels	External hog (Power plant)	3.48*10 ⁻⁰³	0.40%
		Natural gas - Boiler	4.75*10 ⁻⁰²	5.48%
		Natural Gas - Lime kiln	6.99*10 ⁻⁰³	0.81%
	Pulp biomass	Forestry activities (Logs)	7.28*10 ⁻⁰²	8.41%
		Wood chips	8.29*10 ⁻⁰²	9.58%
	Makeup chemicals	NaOH makeup	9.73*10 ⁻⁰³	1.12%
		Na ₂ SO ₄ makeup	9.39*10 ⁻⁰⁴	0.11%
		CaO	2.33*10 ⁻⁰²	2.69%
	Tall oil production	H ₂ SO ₄	3.76*10 ⁻⁰⁴	0.04%
	Transport	Transport biomass	1.53*10 ⁻⁰¹	17.66%
		Transport materials	6.16*10 ⁻⁰⁴	0.07%
	Waste disposal	Dregs	3.39*10 ⁻⁰³	0.39%
		Grits	4.46*10 ⁻⁰³	0.51%
		Ashes	2.00*10 ⁻⁰⁵	0.002%
		Sludge	1.14*10 ⁻⁰²	1.3%
TOTAL			8.65*10 ⁻⁰¹	100%

The red color indicates a high contribution, while green indicates low contribution. The on-site emissions are the primary source of GHG emissions in the system; 41.9% of the GWP is attributed to the fossil CO₂ from natural gas combustion for steam and electricity generation in the mill; whereas 6.2% comes from fossil CO₂ from natural gas combusted in the lime kiln. These emissions may be avoided by introducing renewable alternatives, such as a biomass boiler, or renewable fuels in the lime kiln. Likewise, pulpwood production corresponds to 18% of the GWP; these emissions come mainly from the combustion of fossil fuels in forestry operations such as harvesting, forwarding, and wood chipping. Pulpwood transport is an important contributor to the GWP, given the

transport distance from the field to mill (200 km) and the high biomass demand in the process (4.4 wet tons of wood total/1 MDT of linerboard).

In the present study, the emissions related to chemical manufacture are 0.034 kg of CO₂-eq/ kg of product or 4% of the total GWP. This is much lower than bleached grades of paper and board, as linerboard does not require bleaching chemicals. The GWP contribution from purchased chemicals has been reported as 0.101 kg CO₂-eq/kg of product for bleached market pulp (Tomberlin *et al.* 2020), 0.297 kg CO₂-eq/kg of product for bleached softwood fluff pulp (Buitrago-Tello *et al.* 2022), and 0.552 kg CO₂-eq/kg pulp for softwood acetate dissolving pulp (Echeverria *et al.* 2021). This difference is particularly due to the demand for sodium chlorate for the on-site production of chlorine dioxide (Tomberlin *et al.* 2020; Echeverria *et al.* 2021; Buitrago-Tello *et al.* 2022).

Given that on-site emissions are the main contributor to the GWP, the present study focused on alternatives to reduce Scope 1 emissions by introducing alternative fuels for energy production and lime kiln operations. It is worth mentioning that reducing emissions by the transport of pulp wood also requires attention, considering that variables, such as the location and aerial density of the biomass, and the transport media available in the supply chain can greatly affect the GWP contribution; however, this aspect is out of the scope of the present study.

The alternatives evaluated are listed in Table 1; the detailed GWP results for the scenarios are reported in the supporting information section. The GWP is reported in two ways. The first is aligned with the Greenhouse Gas Reporting Program (GHGRP) established by the EPA (EPA 2021), where only Scope 1 and Scope 2 emissions are considered. The second is a cradle-to-gate approach, where emissions Scopes 1, 2, and 3 are included in the GWP. Table 3 shows the change in the on-site emissions (Scope 1), the indirect emissions by the electricity demand (Scope 2), and the indirect emissions from other upstream processes (Scope 3) by implementing the alternative technologies. It also shows the net change by only considering emissions Scope 1 and 2 (GHGRP approach) and the total change by considering emissions Scope 1, 2, and 3 (cradle to gate approach).

Overall, the alternatives based on biofuels showed a reduction in the on-site emissions, particularly with the integration of the biomass boiler. However, the benefit achieved with these alternatives is reduced when the indirect emissions are considered (cradle-to-gate approach), especially for biomass gasification and lignin extraction.

Regarding switching natural gas for other fossil-based fuels, most alternatives represent an increase in the GWP; this increase is greatest by implementing petcoke with 85% replacement. These fossil-based scenarios are considered because these are possible fuels that can be used in the lime kiln and may have economic advantage. The use of petcoke and fuel oil has been shown to increase the fossil emissions in producing other paper grades, given the high carbon and low energy content compared to natural gas (Buitrago-Tello *et al.* 2022). The use of TDF does not represent a meaningful difference as, from a CO₂ perspective, it can be considered as substitute when the price is competitive compared with natural gas. Metals emissions from the wire reinforcements in tires may limit the total amount of TDF, which can be permitted for use in a kiln.

There are clear differences in the GWP when Scope 3 indirect emissions are considered. For the biomass boiler scenario, there is an 81.5% reduction for Scope 1+2 and only a 41.3% reduction when considering Scope 1+2+3 (Table 3). This difference arises mainly from the GWP associated with the production and transport of the biomass to the mill.

Table 3. Detailed Changes in the Emissions Scope 1, 2, and 3 by Implementing Alternative Fuels in Lime Kiln Operations and by Replacing the Natural Gas Boiler with Biomass Boiler Energy

Emission	Scope 1	Scope 2	Scope 1+2	Scope 3	Total	Total Biogenic
Base case (kg CO₂/kg machine dry linerboard)	0.416	0.029	0.445	0.421	0.865	1.924
Change in the Emissions (%)						
Biomass Boiler	-87.2%	1.2%	-81.5%	1.2%	41.3%	34.4%
Pulverized biomass (25%)	-3.1%	-0.2%	-3.0%	0.1%	-1.4%	1.3%
Pulverized biomass (50%)	-6.2%	0.0%	-5.8%	0.3%	-2.9%	2.5%
Pulverized biomass (100%)	-12.5%	1.4%	-11.6%	0.7%	-5.6%	5.0%
Biomass Gasification	-12.6%	9.7%	-11.2%	6.4%	-2.6%	6.3%
Tall oil pitch (25%)	-3.0%	-0.1%	-2.8%	0.5%	-1.2%	1.0%
Tall oil pitch (50%)	-6.0%	-0.7%	-5.6%	0.8%	-2.5%	1.9%
Tall oil pitch (100%)	-12.5%	-0.6%	-11.7%	1.5%	-5.3%	3.9%
Crude tall oil (25%)	-2.2%	0.6%	-2.0%	0.5%	-0.8%	1.9%
Crude tall oil (50%)	-5.0%	1.0%	-4.6%	0.5%	-2.1%	3.3%
Crude tall oil (100%)	-11.5%	-0.5%	-10.8%	-0.3%	-5.7%	7.1%
Lignin (25%)	-3.3%	-30.8%	-5.0%	4.8%	-0.2%	3.7%
Lignin (50%)	-6.5%	-19.4%	-7.3%	6.4%	-0.7%	4.2%
Methanol (10%)	-1.0%	0.6%	-0.9%	0.1%	-0.4%	0.7%
Turpentine (10%)	-0.9%	0.3%	-0.8%	0.1%	-0.4%	0.6%
Fuel Oil	5.2%	-5.1%	4.5%	0.3%	2.5%	0.0%
Petcoke (25%)	4.3%	0.7%	4.1%	0.2%	2.2%	0.0%
Petcoke (50%)	6.5%	0.2%	6.1%	0.2%	3.2%	0.0%
Petcoke (85%)	12.3%	0.1%	11.5%	0.5%	6.2%	0.0%
TDR (15%)	0.2%	0.8%	0.3%	-0.3%	0.0%	0.0%

Likewise, the reduction achieved in emissions Scope 1 and 2 by implementing bio-based fuels in the lime kiln is around 11% for some alternatives, including pulverized biomass-100%, biomass gasification, CTO-100%, and TOP-100%. This value corresponds to the potential reductions reported for the P&P in Europe by switching to alternative lime kiln fuels (Berglin and Von 2022). Nonetheless, the maximum reduction for these alternatives is 5.6% when the Scope 3 indirect emissions are considered (Pulverized biomass and CTO-100%). The use of turpentine and methanol offers a marginal reduction of total GWP (lower than 1%) despite these materials being available in the mill.

For lignin, the potential reduction is 7.3% considering only emissions Scope 1 and 2, but the indirect emissions reduce the benefit to a marginal value (0.7%). In addition, emissions Scope 2 are reduced from the scenario lignin-25% to lignin-50% due to a combined increase in the steam and electricity demand. Because the demand for electricity by the Lignoboost process is higher than the surplus electricity from the increment in the

steam demand, the Scope 2 emissions are reduced from a 25% substitution to a 50% substitution of natural gas by lignin.

Hotspot Analysis of the Alternatives

Understanding that reduction methods for Scopes 1 and 2 may have tradeoffs in increases in Scope 3, and to provide a more detailed view of the associated tradeoffs, a hotspot analysis was performed for sub-areas in the alternative scenarios that showed a reduction in the overall net GWP, considering the cradle-to-gate approach emissions Scope 1, 2, and 3. In this hotspot analysis, the relative contribution per area was defined based on the total GWP (Scope 1 2, and 3) in the base case as Eq. 1,

$$\frac{(CO_2eq_{ij} - CO_2eq_{i,bc})}{Total\ CO_2eq_{bc}} \times 100\% \quad (1)$$

where i corresponds to the area, j to the scenario, and bc to base case.

Table 4 shows the highest reduction achieved for each alternative, the hotspot results are included in Table S17. The maximum GWP reduction is achieved by the replacement of the natural gas boiler with a biomass boiler (41.3% reduction in the GWP). In this case, the fossil CO₂ emissions avoided from the natural gas combustion represent a 41.9% reduction, additionally the avoided demand of natural gas represents a Scope 3 reduction of 5.5%. Still, there are some areas that increase the GWP decreasing the net GWP savings somewhat.

Pulverized biomass is the alternative that offers the maximum reduction among the lime kiln fuels evaluated. In this case, the avoided emissions from the production and combustion of natural gas are realized but tempered by the indirect emissions associated with the procurement, transport, drying and pulverization of biomass. In this case, the reduction in the GWP increases with the amount of energy supplied by the pulverized biomass system, achieving a maximum reduction of 5.9% at 100% displacement of natural gas.

For biomass gasification, the avoided emissions by displacing natural gas are the same as for pulverized biomass. However, the lower HHV of the syngas (6.5 MJ/kg) (Rofouieeraghi 2012) compared to pulverized biomass (20.5 MJ/kg) (Valmet 2015), and a modest production ratio (0.9 kg syngas/ kg dry biomass) (Rofouieeraghi 2012) increases the demand of biomass, and therefore the indirect emissions.

Regarding TOP, this is a co-product of the distillation of CTO, with a HHV comparable to fuel oil (40.3 MJ /kg vs. 44.6 MJ /kg) (Francey 2009; Valmet 2015). Given this energy content and its bio-based origin, it might be expected to offer a better reduction in the GWP. Nevertheless, the indirect emission associated with the CTO distillation reduces the net benefit to a net 5.3% GWP reduction. Likewise, CTO has a lower energy content of 38.4 MJ/ kg (Lundqvist 2009), but it has the advantage of being available in the mill. Generally, it is more economically favorable to sell the CTO to the distilleries and buy back the tall oil pitch (Berglin and Von 2022); however, some mills still use this co-product as lime kiln fuel (Bajpai 2018). According to the results, the maximum reduction in the GWP by implementing CTO combustion in the lime kiln is 5.8%.

The extraction of lignin has various effects on the mass and energy balance. The lignin extraction implies a reduction in the black liquor solids to the recovery boiler. In the present model, the energy content of the extracted solids is countered by increasing the fuel demand in the biomass boiler. Additionally, the recirculation of liquor from the Lignoboost

process to the evaporator increases the steam demand, and consequently, the production of on-site electricity rises along with the increased steam production. This additional steam demand also contributes to the biomass demanded in the boiler. These changes in the energy balance are reflected in a reduction in the emissions Scope 2, and an increase in the biomass for energy production (Table 4).

Table 4. Hotspot Analysis for Alternatives that Represent a Reduction in the GWP for Linerboard Production. PV= Pulverized Biomass, BG= Biomass Gasification, TOP=Tall Oil Pitch, Crude Tall Oil=CTO, TP= Turpentine

Scope	Alternative Process	Biomass Boiler	PV (100%)	BG	TOP (100%)	CTO (100%)	Lignin (50%)	Methanol (10%)	TP (10%)
Scope 1	Fossil CO ₂ (Lime kiln)	-	-6.2%	-6.2%	-6.2%	-6.2%	-3.1%	-0.6%	-0.6%
	Fossil CO ₂ (Boiler)	-41.9%	0.1%	0.1%	0.2%	0.6%	-	0.1%	0.2%
Scope 2	Electricity (mill)	-	-	-	-	-	-1.5%	-	-
Scope 3	Chemicals	-	-	-	-	0.1%	1.3%	-	-
	Biomass (Energy)	3.3%	0.5%	1.9%	-	-	0.9%	-	-
	Natural gas production (Boiler)	-5.5%	-	-	-	0.1%	-	-	-
	Natural gas production (Lime Kiln)	-	-0.8%	-0.8%	-0.8%	-0.8%	-0.4%	-0.1%	-0.1%
	Alternative Fuel Production	-	-	0.7%	1.4%	-	0.9%	-	-
	Transport	2.8%	0.4%	1.7%	0.1%	0.3%	1.4%	0.1%	-
Net Reduction		-41.3%	-5.9%	-2.6%	-5.3%	-5.8%	-0.7%	-0.4%	-0.4%

The chemical balance is also affected by the Lignoboost process, a fraction of sodium is lost in the production of the lignin press cake (2.7 kg NaOH/ton). Additionally, there is sulfur added by the black liquor acidification with sulfuric acid; this acidulation reduces the demand of sodium sulfate (3.9 kg Na₂SO₄/ton reduction) makeup. However, the indirect emissions associated with sodium hydroxide are higher compared to sodium sulfate (1.4 kg CO₂-eq/kg NaOH vs 0.17 kg CO₂-eq/ kg Na₂SO₄). This results in increased indirect emissions from the pulping chemicals. Moreover, the Lignoboost process requires CO₂ (purchased from external sources in this simulation) and sulfuric acid for the precipitation of lignin, increasing the indirect emissions associated with chemicals. The extraction also implies other indirect emissions as electricity demanded in the lignin dryer and transport of additional materials.

Marginal Abatement Cost Curves

The alternatives were categorized by developing a Marginal Abatement Cost Curve (MACC). This curve shows the Cost of Avoided Carbon (CAC) in US \$/ton of CO₂-eq,

and the potential CO₂-eq reduction by implementing each technology for the established mill's production.

$$CAC = \frac{\text{Net Present Value}}{CO_{2eq} \text{ avoided in 10 years of operation}} \quad (2)$$

The MACC was built considering emissions Scope 1 and 2 (GHGRP approach), and the total emissions associated with the entire system (cradle-to-gate approach). Table 5 shows the total cost of implementing each technology, the changes in the annual operating and maintenance costs, and the NPV in an 11-year lifetime (the first year is for construction), with a 15% rate of return. In addition, the NPV and the CAC of each alternative was estimated considering two carbon-offset prices, \$11/ton and \$47/ton. These values are prices projected for 2030 and 2050, respectively (Bloomberg Finance 2022), and correspond to a market scenario where all types of carbon saving suppliers are allowed, including the offsets having avoided emissions (which is the case of the present study) rather than removing the carbon from the atmosphere (Bloomberg Finance 2022).

MACC-Emissions Scope 1 and 2

The MACC shown in Fig. 3a categorizes the alternatives according to the CAC, considering the onsite emissions (Scope 1 emissions) and the emissions derived from the production of the energy inputs (Scope 2 emissions). The width of each bar corresponds to the amount of CO_{2eq} avoided per year achieved by implementing the alternative. In addition, the total CO_{2eq} avoided per air-dry ton for each alternative is included in the green labels. The utilization of pulverized biomass and the combustion of TOP were found to be the most cost-effective method to reduce the GWP in the lime kiln at \$54 and \$78 per ton CO₂-eq avoided, respectively. This can be contrasted to another quote for carbon savings in a lime kiln used for cement production in Taiwan, of about \$26/per ton CO₂-eq (Huang and Wu 2021). The largest annual amount of carbon savings is through the implementation of the biomass boiler at a price of \$79/per ton CO₂-eq. Some of the other technologies have a high CAC, including gasification, methanol, turpentine, and lignin. Coproducts CTO and TOP do not show the same high CAC as the other coproducts such methanol, turpentine, and lignin.

Table 5. Capital Cost, Net Present Value, and Carbon Avoided Carbon for Alternatives to Reduce GWP in the Production of Linerboard

Scenario	ton CO ₂ -eq avoided /year (Scope 1+2)	Investment Capital (Millions)	Operating cost (millions/year)	NPV (Millions)			CAC (dollars / t CO ₂ -eq avoided)			Minimum offset price for a NPV = 0 (\$/ ton CO ₂ -eq avoided)
				\$0/ t CO ₂ -eq avoided	\$11/ t CO ₂ -eq avoided	\$47/ t CO ₂ -eq avoided	Offset price = \$0/ ton CO ₂ -eq avoided	Offset price = \$11/ ton CO ₂ -eq avoided	Offset price = \$47/ ton CO ₂ -eq avoided	
Pulverized biomass 100%	40,253	\$17.11	(\$0.08)	(\$21.66)	(\$19.00)	(\$10.27)	\$54	\$47	\$26	\$89
TOP	40,428	\$1.30	\$4.96	(\$31.52)	(\$28.84)	(\$20.08)	\$78	\$71	\$50	\$130
Biomass Boiler	281,753	\$178.85	(\$1.37)	(\$223.04)	(\$204.38)	(\$143.33)	\$79	\$73	\$51	\$132
CTO	37,416	\$1.30	\$5.25	(\$33.30)	(\$30.82)	(\$22.71)	\$89	\$82	\$61	\$148
Turpentine 10%	2,935	\$1.30	\$0.95	(\$7.38)	(\$7.18)	(\$6.55)	\$251	\$245	\$223	\$418
Methanol 10%	2,961	\$0.52	\$1.22	(\$8.01)	(\$7.81)	(\$7.17)	\$270	\$264	\$242	\$449
Lignin 50%	25,250	\$17.43	\$7.90	(\$70.06)	(\$68.39)	(\$62.91)	\$277	\$271	\$249	\$461
Biomass gasification	38,727	\$61.86	\$10.88	(\$145.49)	(\$142.92)	(\$134.53)	\$376	\$369	\$347	\$624
Pulverized biomass +biomass boiler	322,005	\$195.96	(\$1.44)	(\$244.70)	(\$223.38)	(\$153.61)	\$76	\$69	\$48	\$116

Note: The NPV and the CAC were estimated assuming three prices for the carbon offsets: \$0, \$11, and \$47 dollars for ton of CO₂-eq (Bloomberg Finance 2022)

Under this approach considering only scope 1 and 2, some lime kiln fuels have a CO₂-eq reduction ranging between 10.6 to 11.6%, including pulverized biomass, biomass gasification, CTO, and TOP (Table 3). However, pulverized biomass represents a low capital investment compared to biomass gasification and a low operating and maintenance cost compared with CTO and TOP; leading to a low NPV among these alternatives and consequently a low CAC (Table 5).

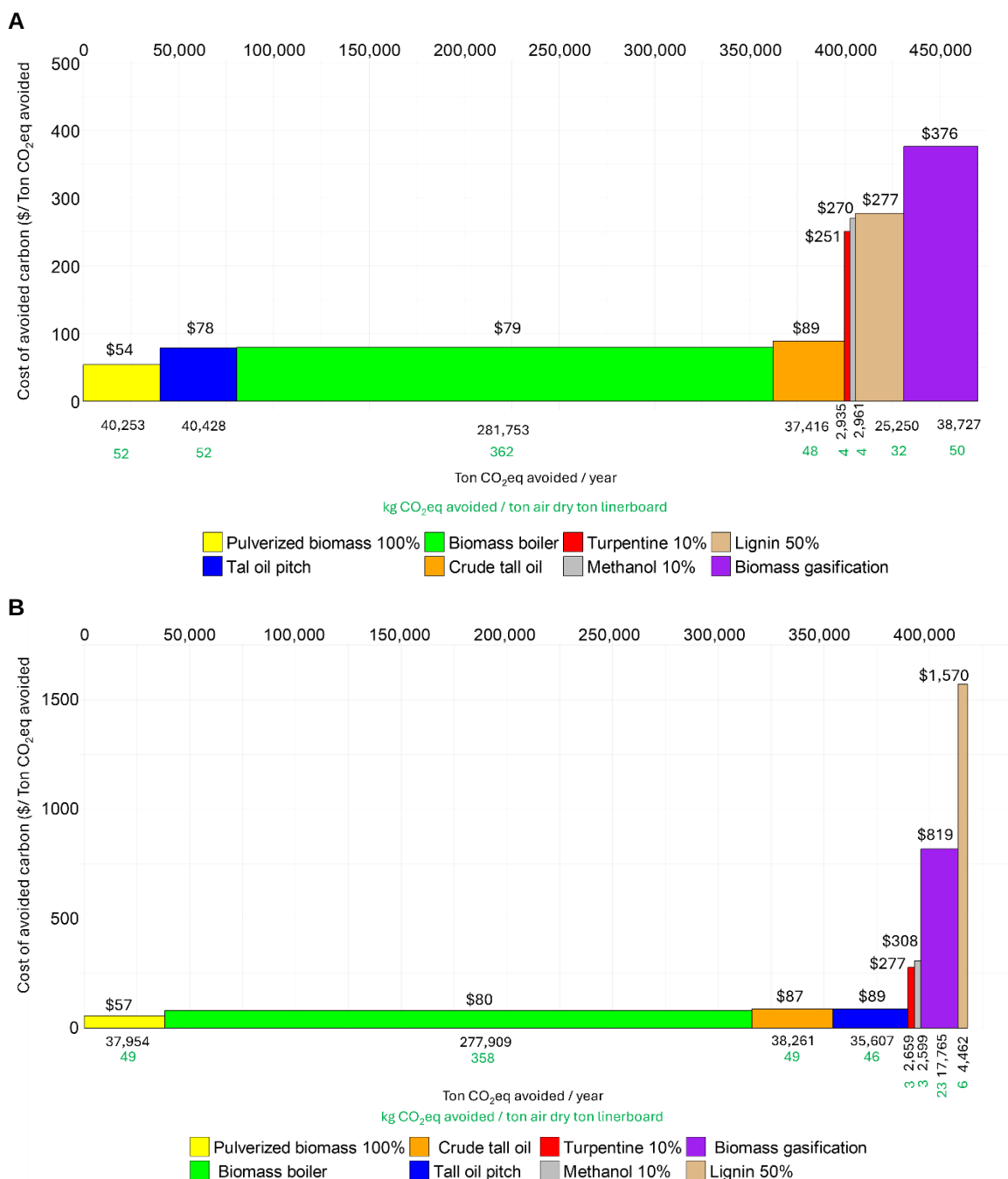


Fig. 3. Marginal abatement cost curve for alternatives to reduce the GHG emissions in the production of linerboard: a) CO₂ avoided based on scope 1 and 2, b) CO₂ avoided based on scope 1, 2, and 3. The production rate for the mill is 2,177 tons per day.

For CTO, the onsite CTO production is used to cover the energy demand in the lime kiln. The revenue lost by burning this biofuel instead of selling it as a coproduct is considered an operating cost in the analysis, which increases the NPV and consequently the CAC of this alternative. In contrast, for the TOP scenario, the CTO is sold to the market while the TOP demanded in the lime kiln is purchased at the same CTO price. The CTO lime kiln demand is 22,733 tons CTO/ year, while the TOP demand is 21,993 tons TOP/per year, which represents a higher operating cost for CTO and therefore a higher CAC than TOP. This result is reasonable given the price tendencies that CTO and TOP have shown in recent years (Niemeläinen 2018).

Regarding lignin combustion, the negative NPV is three times the value of the pulverized biomass negative NPV (Table 5); with lignin combustion having only a 7.3% reduction in emissions Scope 1 and 2 relative to the base case (Table 3), making this biofuel the less cost effective among the co-products. In contrast, the combustion of turpentine and methanol represents a low capital investment, given the few adaptations required in the lime kiln. Nonetheless, the high price in the market for these alternative fuels (\$750/ton and \$350/ton, respectively), and the low reduction in the GHG emissions makes the CAC higher compared to other alternatives with a high capital investment.

For alternative lime kiln fuels, the MACC shows that pulverized biomass is the most cost-effective alternative fuel, followed by TOP, CTO, turpentine 10%, methanol 10%, lignin 50%, and biomass gasification. This last alternative has a high demand for biomass, increasing the capacity required for biomass processing and drying, plus the gasifier. These components increase the capital investment resulting in a CAC superior among all the lime kiln alternatives.

Regarding the installation of the biomass boiler (working with an existing turbine), this alternative implies a high capital investment (\$179 million) and operating and maintenance costs; however, it offers the maximum reductions (81.5%) with a relatively low CAC of \$79/ ton of CO₂ avoided. Considering implementing both the pulverized biomass system in the lime kiln plus the installation of the biomass boiler, the total GHG emissions avoided per year are 322,006 tons of CO₂-eq per year, with a cost of US \$76 per ton.

MACC-Emissions Scope 1, 2, and 3

The total avoided emissions are reduced when Scope 3 emissions are considered along with Scope 1 and 2 for each alternative, increasing the CAC (Fig. 3b). This change is largest for biomass gasification and lignin. For biomass gasification, the CAC is more than doubled by the indirect emission from the biomass demand and other raw materials required in the gasification system. For lignin extraction, the CAC is 5.7 times higher by the indirect emissions associated with chemicals, including sodium hydroxide, sulfuric acid, and carbon dioxide. In addition, under this approach the CAC ranking changes, being more favorable for CTO than for TOP; this change is derived from the indirect emissions from CTO distillation into derived products, including TOP.

The MACC in this approach shows that the most cost-effective alternative lime kiln fuel is still pulverized biomass, followed by CTO, TOP, turpentine 10%, methanol 10%, biomass gasification, and lignin 50%. It is worth noting that the total GHG emissions avoided per year by implementing both the pulverized biomass plus implementing the biomass boiler at the same time are 315,863 tons of CO₂ per year, given a CAC of US \$77/ton, which is only one dollar above the CAC when Scope 1 and 2 emissions are considered.

Given that none of the alternatives offer a cost saving, the CAC analysis was performed assuming a revenue from the avoided CO₂-eq emissions. In this analysis only emissions in Scope 1 and 2 are considered, also two prices are assumed for the avoided emissions: a carbon offset price of \$11 per ton of CO₂-eq avoided, a price expected by 2030 under the current conditions of the market, and a price of \$47 per ton of CO₂-eq avoided, the expected value by 2050. These prices are values for alternatives that avoid emissions rather than removing them (Bloomberg Finance 2022). The NPV and the CAC for each offset price is shown in Table 5.

For the \$11 and \$47 offset prices, none of the alternatives showed a negative CAC; indicating that the alternatives represent a cost for the mill for the projected offset prices. Therefore, the minimum offset price in the market was calculated to obtain a NPV equal to zero (last column in Table 5). This minimum offset price was compared with the off-set prices assumed (\$11 and \$47/ton CO₂-eq), and also with the offset prices of alternatives that store or sequester carbon, in this case \$224/ton by 2029 and \$120/ton by 2050 (Bloomberg Finance 2022).

As shown in Table 5, the minimum offset prices are above \$11 and \$47/ton of CO₂-eq, the expected prices for alternatives that avoid carbon. Compared to the alternatives that store or sequester carbon, all the alternatives have a price above \$224/ ton, except for pulverized biomass, TOP, and biomass boiler. However, by 2050, technologies such as direct air capture will become more widely adopted, reducing the price to \$120/ton, a price lower than the minimum offset value of most of the alternatives considered in this study. The only alternative that may compete with direct air carbon capture technology is pulverized biomass, with an offset price of \$89/ton of CO₂-eq avoided (Table 5).

CONCLUSIONS

The U.S. pulp and paper industry is largely dependent on fossil fuels, with lime kiln operations representing a key challenge in achieving zero on-site fossil emissions. This study evaluates the GHG reduction potential and associated costs of alternative fuels for lime kiln operations in linerboard production, and the replacement of natural gas to cover the electricity and steam demand in the process. The alternative fuels for the lime kiln include external biomass and coproducts generated from mill operations.

For this pulp grade, 2,789 kg of CO₂-eq are emitted per ton of product, from which 1,924 kg corresponds to biogenic CO₂ (69%), and 854 kg (31%) corresponds to fossil CO₂-eq. Two major contributions to GWP are the natural gas boiler and the lime kiln. In this study, the replacement of the natural gas boiler by a biomass boiler represents a 41% reduction in the GWP, and fuel switching natural gas in the limekiln by biofuels achieves a 5.5% reduction.

The cost of the avoided carbon (CAC) was determined as 54 to 1600 \$/ton CO₂-eq for different alternative lime kiln fuels and the biomass boiler. Replacement of natural gas by biomass either in the lime kiln or the boiler has similar and very low CAC, 54 and 79 \$ /ton CO₂ avoided, respectively. The use of mill coproducts (turpentine/CTO// methanol/lignin) represent a higher CAC because of the high price of these coproducts in the market.

In constructing the marginal abatement cost curve to categorize the alternatives, Scopes 1, 2, and 3 emissions were considered, rather than only direct Scope 1 and 2 emissions. Some indirect emissions (Scope 3) can significantly increase the cost of

abatement. For example, in the case of biomass gasification and lignin as alternative fuels for lime kiln operations, the abatement cost is 2 and 5.5 times higher, respectively, compared to considering only Scope 1 and 2 emissions.

Finally, implementing the biomass boiler along with the pulverized biomass in the lime kiln represents a reduction of 93.1% in emissions Scope 1 and 2 (81.5% and 11.6%, respectively). These two technologies represent a total CAC of \$76/ ton of CO₂-eq avoided. The CAC can be further reduced if the mill gets a revenue from the CO₂ avoided. For instance, assuming a selling price of \$11 and \$47 per ton of CO₂-eq avoided, the total CAC is \$69 and \$48/ton of CO₂-eq avoided, respectively.

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SUPPLEMENTARY INFORMATION

Buitrago-Tello, R., Venditti, R. A., Jameel, H., Hart, P. W., and Ghosh, A. (2024). "Carbon footprint and techno-economic analysis to decarbonize the production of linerboard *via* fuel switching in the lime kiln and boiler: Development of a marginal abatement cost curve," *BioResources* 19(4), 7806–7823.

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INTRODUCTION

The present document compiles the assumptions included in the process simulation for producing linerboard using a continuous kraft pulping process, and the information used to estimate the GWP and the Marginal Abatement Cost Curve

LINEBOARD PRODUCTION PARAMETERS

Woodyard

The model includes the mass balance for the woodyard, including a debarker, chipper, and a chip screen (Fig. S1). The assumptions are included in Table S1:

Table S1. Main Assumptions for the Woodyard (Hart 2022; Fisher International Inc, n.d.)

Chips demand	Chips from roundwood - purchased wood chips ratio = 1:1 Biomass moisture content: 50% Temperature: 20 °C
Debarker yield	Softwood roundwood yield: 90% Bark free roundwood + 10% Bark
Chipper and screen yield	Chips yield: 95.5% Chips to pulping + 4.5 % Hog fuel

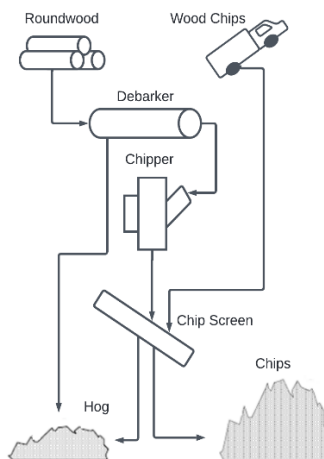


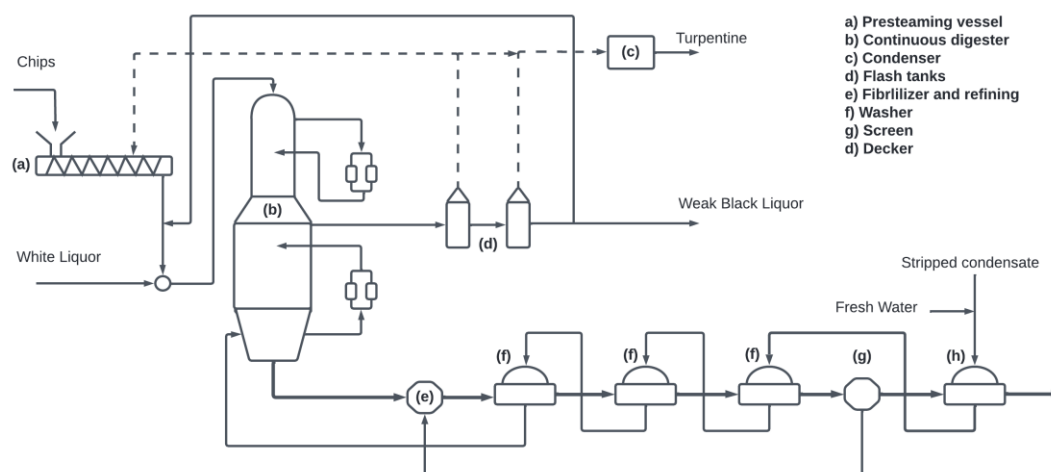
Fig. S1. Woodyard operations

Digester

Table S2 includes the assumptions for the continuous digester. The flashed steam from the weak black liquor is used to pre-steam the chips; the excess steam is sent to a condenser to extract turpentine (Fig. S2). After cooking, the pulp is washed using the stripped condensate from the weak black liquor evaporator and fresh water.

Table S2. Assumptions for the Pulping Area (Grace, Malcolm, and Kocurek 1983; Hart 2022)

Presteamming	Turpentine Formed = 0.9 gal / Ton pulp = 3.7 kg/ton pulp
White Liquor charge	Active alkaline on wood: 0.15 as NaOH Active alkaline concentration: 105 g/l as NaOH Sulfidity, on AA basis: 25% Reduction efficiency: 95% Causticizing efficiency: 82%
Digester conditions	Liquor to wood ratio = 3:1 Temperature: 160°C Yield: 56%
Pulp	Kappa: 110 (%lignin = 0.15 * kappa) Pulp composition: Cellulose: 68.5%, Lignin: 16.5%, Hemicellulose: 15%
Fibrilizer & Refining	Input pulp consistency: 6%
Screener	Input pulp consistency: 1%, Rejects are returned to fibrilizer (2% rejection , 35% consistency)
3 washing stages	Washing using stripped condensate Pulp consistency: 10% Efficiency Factor: 2 Outlet pulp consistency: 14%

**Fig. S2.** Continuous digester and washers

Multiple effect evaporator

A counter-current six-effect evaporator concentrates the weak black liquor to 55% solids; the heat transfer area and the outlet vapor temperature are assumed on each body. After the evaporator, the black liquor is sent to a concentrator to increase the solids content to 69%; the concentrator assumptions are shown in Table 3.

The black liquor is concentrated in a six-effect evaporator. The first body uses low-pressure steam to concentrate the liquor, and the clean condensate from this body is returned to the power plant. The rest of the bodies use the steam from the black liquor evaporation; the foul condensate from the bodies is sent to a stripping column. This column reduces the COD and separates methanol; the stripped condensate is used to wash the pulp in the washers (Fig. S2). Additionally, soap is extracted after the third body and is treated with sulfuric acid to produce crude tall oil; the yield of this reaction is 85% (Evdokimov *et al.* 2017).

Table S3. Concentrator Assumptions (Hart 2022)

Concentrator	69% solids content Condensate temperature: 85 °C Liquor temperature: 115 °C Steam economy: 1.8
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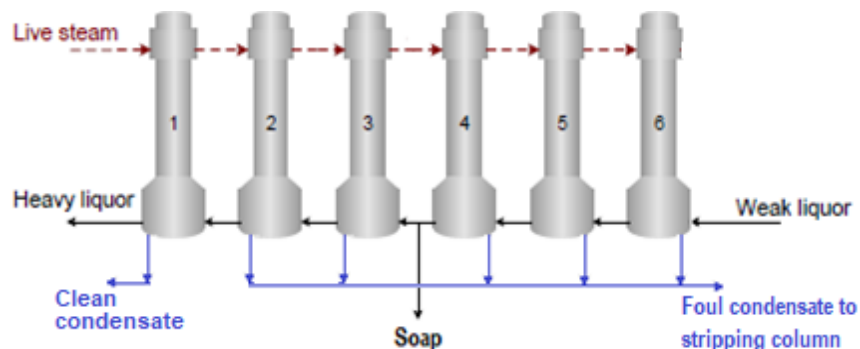


Fig. 3. Multiple-effect evaporator configuration

Condensate stripper

The fouling condensate from the evaporator and concentrator are steam stripped to extract the methanol and recirculate the condensate to wash the pulp.

Table S4. Condensate Stripper Assumptions (Valmet 2015)

Outlet stripped condensate	125 °C
Overhead vapor temperature	112 °C
Feed-to-steam ratio	5
%Methanol stripping	5 lb /Ton of pulp = 2.5 kg / ton of pulp

Recovery, biomass, and natural gas boiler

The heat and power demand are covered by burning the concentrated black liquor in the recovery boiler, burning the hog fuel from the woodyard and external hog fuel in a biomass boiler, and external natural gas in a natural gas boiler. The external hog fuel demand is controlled by setting the steam production at the hog fuel boiler at 10,000 lb of steam /hour.¹ Tables S4 to S6 show the main assumptions included in these boilers (Hart 2022; Fisher International Inc, n.d.; Valmet 2015; Grace, Malcolm, and Kocurek 1983).

Table S5. Recovery Boiler Assumptions

Excess air (%)	0.15
Fraction of the total chloride entering in the liquor which leaves in stack gas as NaCl	0.06
Loading factor	1*
Dregs per solids entering furnace	5
High heating value (HHV)	5200 Btu /lb
Reduction fraction	0.9
Smelt temperature	750 °C
Gas temperature	250 °C (Efficiency = 67%)
Pressure	850 psi
Temperature	850 F
Blowdown	5%

* The nominal loading is 500 kg of dry solids / m² / hr. The loading factor is the fraction of the nominal loading

Table S6. Hog Fuel Boiler Assumptions

Moisture in fuel	50%
Excess air	10%
Hog fuel composition (default value)	Carbon: 51.5 %, Hydrogen: 6.1%, Oxygen: 41.1%, Nitrogen:0.1%, Sulfur:0.1%, Inerts: 1.1%
High heating value (HHV)	4900 Mcal /mt
Outlet gas temperature	176.7°C (efficiency 72.6%)
External hog fuel demand	Controlled by setting the steam flow at 150,000 lb/h
Temperature	825 F
Pressure	850 psi
Blowdown	2%

Table S7. Natural Gas Boiler Assumptions

Excess air	10%
Natural Gas composition	Carbon: 74.8%, Hydrogen: 25.2%
High heating value (HHV)	13283.6 Mcal /mt
Combustion efficiency	85%

Natural gas demand	Controlled by setting the condensing turbine condensate at 10,000 lb/h
Temperature	825 F
Pressure	850 psi
Blowdown	2%

Back-pressure and condensing turbine

The model includes a back-pressure and a condensing turbine to generate electricity; the assumptions are included in Tables S8 and S9. The system has a sootblow of 1.5% of the total high-pressure steam. The rest of the high-pressure steam is distributed equally between the two turbines and is expanded to 160 psi. 50% of the mill's medium-pressure steam demand is extracted from the back-pressure turbine and 50% from the condensing turbine.

After the first extraction, the remaining steam is expanded to 60 psi; the low-pressure steam demand is controlled by the second extraction in the condensing turbine. In addition, the natural gas demand is controlled by setting the condensate flow in the condensing turbine as 10,000 lb/h.

Table S8. Back-Pressure Turbine Assumptions

First stage	160 psi, adiabatic efficiency 70%
Extraction	50% of the medium-pressure steam demand
Second stage	60 psi, adiabatic efficiency 70%

Table S9. Condensing Turbine Assumptions

First stage	160 psi, adiabatic efficiency 70%
First extraction	50% of the medium-pressure steam demand
Second stage	60 psi, adiabatic efficiency 70%
Second extraction	Controlled by the low-pressure steam demand
Third stage	20 psi, adiabatic efficiency 70%
Steam to condenser	10,000 lb/h

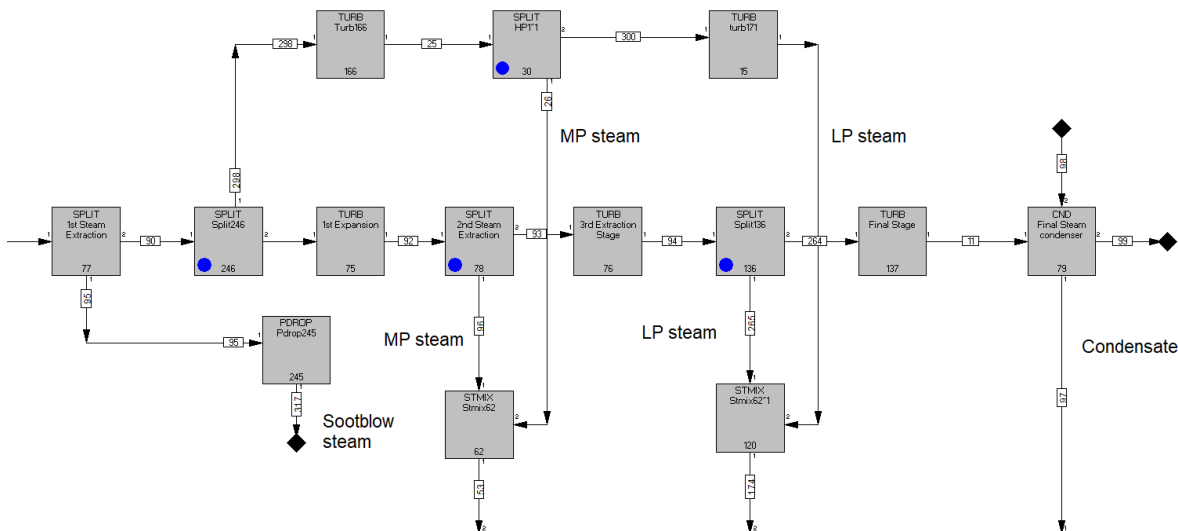


Fig. S3. Back-pressure and condensing turbine

Causticizing plant

The causticizing plant includes a smelt dissolving tank, a green liquor clarifier, the wash and extraction of dregs, the slaking and causticizing units, a white liquor clarifier, a mud washer and filter, a lime kiln, a lime kiln scrubber. The causticizing plant has a controller for the make-up sodium hydroxide and sodium sulfate demand, depending on the active alkaline and sulfidity set in the digester. Table S10 shows the assumptions for the different unit operations that compose the causticizing plant.

Table S10. Causticizing Plant Assumptions

Smelt dissolving tank	Intermediate smelt temperature = 250 °C $T = 12 \cdot (TGL) - 11 \cdot (TWL)$ where: TGL = temperature of green liquor (measured in the mill) TWL = temperature of weak liquor (measured in the mill)
Green Liquor Clarifier	Suspended solids in clarified liquor: 20ppm Underflow: 25% consistency
Dreg washer	Efficiency Factor (E) = 3, Consistency of outlet solids = 50%
Slaking and causticizing	- Make-up with CaO - Lime charge = 0.82, mole $[CaO + Ca(OH)_2]$/mole CO_3 in green liquor - Fraction of $CaSO_4$ dissolved, Default = 1 "A" in the formula, Density = $1 + (TDS \text{ as a fraction}) \cdot A$ (Default = 0.712) - Pressure in slaker and causticizer(s) (Default = 760 mm Hg) - Volume of vessel 1, 2, and 3 (or residence time) = 50 min - Rate constant for slaking at 95°C (suggested value: 0.18 minute⁻¹) (Activation energy of 11.2 kcal/mole assumed) - Rate constant for causticizing at 95°C (suggested value: 1.9 liters/mole minute) (Activation energy of 11.2 kcal/mole assumed)
Grits loss	0.1% liquor loss, 0.5% solids loss
White Liquor Clarifier	Suspended solids in clarified liquor: 100ppm, Underflow: 25% consistency
Mud Washer	Mud loss: 10%, Suspended solids in clarified liquor: 15ppm, Underflow: 40% consistency (mud)

Mud Filter	Input consistency: 25%, Efficiency Factor (E): 2, Consistency of outlet solids: 80%
Kiln Scrubber	- Na₂SO₄ dust removal efficiency: 99% - Na₂CO₃ dust removal efficiency: 99% - NaCl dust removal efficiency: 99% - Water demand: Controlled to adjust TTA white liquor to 113

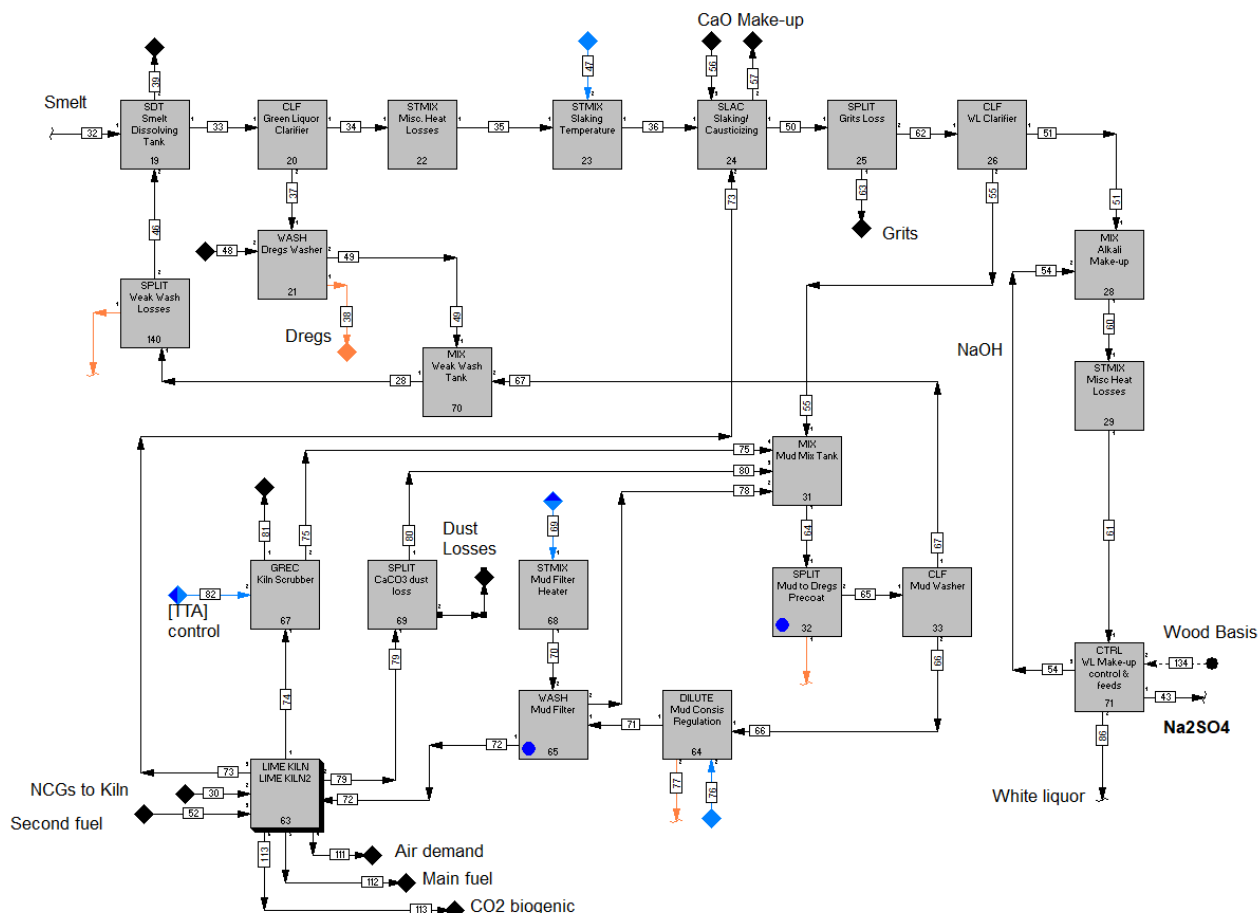


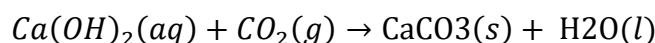
Fig. S4. Causticizing plant

Lime kiln model

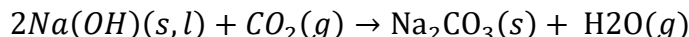
The causticizing plant includes a lime kiln simulation model that determines the mass and energy balance of drying and calcinating the lime mud. This balance is used to estimate the natural gas demand and the alternative fuels demand for the scenarios proposed in the present study.

The model includes different reactions to estimate the mass balance:

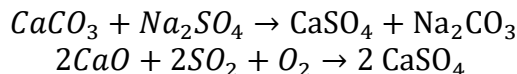
- The carbonation of the Ca(OH)₂ remaining in the lime mud, which is a small fraction of the suspended solids (~0.03)



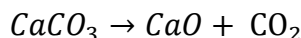
- b) The reaction of water-soluble sodium from the white liquor with CO₂ to form Na₂CO₃



- c) The formation of CaSO₄ by the presence of sulfur in the system



- d) And the calcination of CaCO₃



The energy balance is determined based on the sensible heat from the mud's initial temperature to 100°C, the energy required to dry the mud, the sensible heat from 100°C to 800°C, and the energy demand from the calcination.

The sensible heat corresponds to the change in the enthalpy of the primary mud components (CaCO₃, water, and CaO) using equation S1.(Lundqvist 2009) The mud's drying energy demand is determined assuming a heat of evaporation of 2257 kJ/kg of water evaporated.(Lundqvist 2009) The energy required for calcination is based on the heat of reaction (42657.9 kcal/kmol).(Lundqvist 2009)

$$\Delta H = \left(a_0(T_2 - T_1) + \frac{a_1}{2}(T_2^2 - T_1^2) + \frac{a_2}{3}(T_2^3 - T_1^3) + a_3(\ln(T_2) - \ln(T_1)) - a_4(T_2^{-1} - T_1^{-1}) \right) \text{ Eq. S1}$$

The values for the coefficient of the equation are in Table S11 (Lundqvist 2009).

Table S11. Heat Capacity Coefficients for Different Compounds

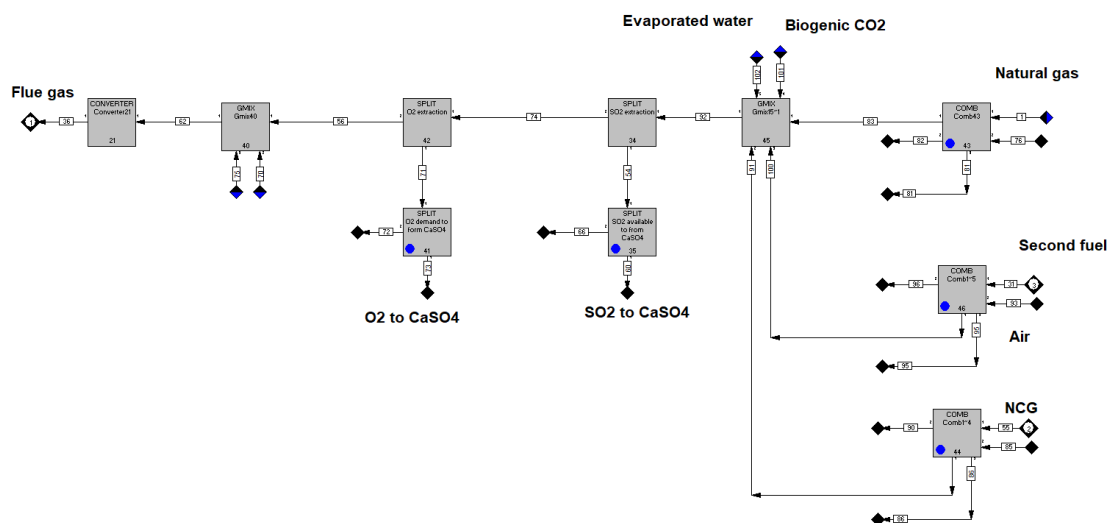
Component	a0	a1	a2	a3	a4
CaO	0.949	3.71	1.01	-27	-10574
CaCO ₃	1.009	25.36	0.64	-4.7	-20965
H ₂ O(l)	4.03	487	0	0	0

The fuel demand is determined based on the energy required for heating, drying, and calcinating the mud. The model includes three combustion blocks (Fig. S5), one for the natural gas feed, a second block for the alternative fuel (pulverized biomass, syngas, crude tall oil, *etc.*), and a third block in case the user wants to include an additional fuel for instance non-condensable gases (NCG). On each block, the user defines the moisture content, the high heating value, the composition (carbon, hydrogen, oxygen, nitrogen, sulfur, and inerts fraction), the exit gas temperature, and the heat loss. The fuel demand is determined based on the energy required to heat, dry, and calcinate the lime mud.

The evaporated water and the biogenic CO₂ from the CaCO₃ are mixed with the flue gases. Whereas the oxygen and SO₂ to form CaSO₄ are extracted. Table 10 summarizes the restrictions assumed in the lime kiln model.

Table S12. Lime kiln Model Assumptions

Limekiln	- Fraction excess air, typically 0.10 - Exit lime temperature, 800 °C - Exit gas temperature, 150 °C - Heat loss due to convection, 0.10 fraction - Percent availability of output lime, 85% - Weight fraction of total input Ca (mud and makeup) converted to CaSO₄, if sufficient sulfur is available. 0.06 - Fraction of incoming suspended solids in dust leaving the kiln, by weight. 0.1 - Fraction of incoming Na vaporized, by weight, 0.5
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**Fig. S5.** Lime kiln model

Paper machine

In the paper machine, the pulp is diluted to a 3% consistency before going to a tickler refiner/beater where 0.15% of the incoming fiber is lost; then, the pulp is sent to a primary and secondary cleaner to eliminate any undesirable material in the headbox. Next, the fibers are suspended in water to a 0.5% consistency and then sprayed in the headbox over the wire to orientate the fibers. Then the water is drained by gravity in the table rolls and by vacuum in suction boxes, couch, and trim to a 16% consistency. Finally, the sheet is pressed and dried to a final consistency of 90%. The pulp drier consumes low and medium-pressure steam in two sections: the seal pit that keeps water at 50°C using low-pressure steam, and the drying section that uses medium-pressure steam to evaporate the water in the pulp to the desired moisture content; a steam economy factor of 1.4 is assumed for this section.

ELECTRICITY DEMAND

The electricity demand was determined based on factors reported in the literature (Nilsson *et al.* 1995; Martin *et al.* 2000).

Table 12. Electricity Demand Per Area

Area or Unit Operation (kWh/ADt)		
Raw Materials Preparation	Debarking	20
	Chipping and conveyors	63
Pulping	Digester	43
	Washing and Screening	103
	Screening and storage	74
Chemical recovery	Black liquor concentration	66
	Causticizing & lime kiln	42
Powerhouse		125
Wastewater treatment		35
Other		15
Papermaking	Forming and pressing	238
	Drying section	21
TOTAL		845

LIFE CYCLE INVENTORY AND GWP

The net GWP for the scenarios under study were determined based on the inputs and outputs from the mass and energy balance (Table S13) the mass allocation factors for each scenario (Table S14) and the GWP of the upstream processes and waste treatment (Table S15). The GWP for each scenario was determined according to the following equation:

$$GWP_{scenario} = LBMAF_{scenario} * (Onsite\ emissions_{scenario} + \sum Flow_{i,scenario} * GWP_i)$$

LBMAF = Linerboard mass allocation factor

i = upstream process or waste generated in the system

The GWP for each upstream process and waste treatment was determined by transforming the Ecoinvent database life cycle inventory an data reported in the literature into CO₂-eq emissions through the method IPCC 2013 GWP 100-years included in OpenLCA 1.10.2.(“OpenLCA,” n.d.) The GWP for the scenarios considered are shown in Table S16.

ALTERNATIVE SCENARIOS

Biomass Boiler

In this scenario, it is assumed that the natural gas boiler is replaced by a biomass boiler that burns the internal hog fuel from the wood yard and external biomass to cover the steam demand in the mill, and the production level remains the same. It is also assumed that the back-pressure and the condensing turbine do not require any modification.

External bio-based fuels

This group includes external bio-based fuels that can cover the total energy demand in the lime kiln. In the case of pulverized biomass, the system includes a biomass dryer and a hammer mill with an electricity demand of 4.4 kWh/ton of water evaporated (Rofouieeraghi 2012) and 48.5 kWh/t of wood processed, respectively. (Wind *et al.* 2018) The external biomass is dried from 50% to 5% moisture content using the flue gasses from the recovery boiler as the heat source. (Hart 2020) The high heating value assumed for the pulverized biomass is 20.5 MJ/kg. (Valmet 2015) The lime kiln can be operated at different feed levels (Manning and Tran 2015), the present study assumes a 25, 50, and 100% displacement of natural gas.

Regarding the gasification system, this scenario includes a biomass dryer and a circulating fluidized bed (CFB) gasifier; the biomass is first dried to a 10% moisture content using the flue gasses from the recovery boiler as a heat source; the electricity demand factor is the same as for the previous scenario. The CFB gasifier's production ratio is 0.9 kg syngas/ kg dry biomass. (Rofouieeraghi 2012) The rest of the life cycle inventory as the electricity demand and other raw materials were adapted from the process available in the Ecoinvent database defined as "synthetic gas production, from wood, at fluidized bed gasifier-Rest of the world." (Wernet *et al.* 2016)

For the TOP scenario, the on-site CTO is sold and upgraded by distillation into different fractions, including tall oil fatty acids (38%), tall oil rosins (34%), distilled tall oil and heads (12%) and tall oil pitch (16%). (Aryan and Kraft 2021) The production of 1 ton of all these co-products is equivalent to 402 kg CO₂; (Cashman, Moran, and Gaglione 2016) therefore, it is assumed TOP on-site emissions are equivalent 64.1kg CO₂/ton, whereas the indirect emissions from CTO are based on the base case. The TOP is bought and transported to the mill to cover 25, 50, and 100% of the lime kiln energy demand.

Bio-based Products or Bio-based Streams Available in the Mill

This group includes the streams that are available in the mill (methanol, turpentine, CTO) and the extraction of lignin, which is a potential co-product in the production of kraft pulp. (Tomani 2010) In the case of CTO, the on-site CTO production is enough to supply 40% of the lime kiln energy demand. Therefore, for the scenario where CTO covers 25% of the lime kiln demand, the fraction sold to the market is lower compared to the base case, changing the mass allocation factors (see table S14). For the 50% and 100% scenarios, the CTO demand in the lime kiln is covered by the on-site CTO and external CTO. In these two scenarios, the GWP is mass allocated between the remaining products (linerboard,

turpentine, and methanol), and the GWP for the external CTO is based on the results from the base case.

Similarly, in the scenario Turpentine-10%, the on-site production is enough to cover 14% of the lime kiln energy demand, therefore the amount sold is lower compared to the base case, changing the mass allocation factors. In the case of methanol-10%, the onsite production covers 4.3% of the lime kiln energy demand, therefore, the remaining fraction is covered by external methanol, and the emissions are mass allocated between linerboard, turpentine, and crude tall oil. The GWP associated to the external methanol is based on the emissions allocated to the production of methanol in the base case.

In the lignin scenario, a fraction of the black liquor with a 36% solid content is extracted from the evaporator system. Then, the liquor is sent to a Lignoboost plant, where lignin is precipitated by reducing the pH with CO₂ and sulfuric acid.(Fredrik Öhman *et al.* 2013) The precipitated lignin is filtered, the filtrate is returned to the evaporator system, and the lignin with a 70% moisture content is dried to a 4% moisture content in a dryer that uses the flue gas from the recovery boiler.(Tomani 2010) Although the lime kiln can be operated with 100% lignin, the extraction of black liquor is limited to a level that lignin covers a maximum of 50% of the total lime kiln demand; this is to avoid an excess demand in the biomass boiler, which replaces the energy content of the extracted liquor.

Other fossil-based fuels

Natural gas is the predominant lime kiln fuel in the US and Canada, however there are other alternative fossil fuels in lime kiln operations. For instance one third of the kilns in the United States and Canada burns fuel oil.(Francey, Tran, and Jones 2009) Additionally, petcoke is an alternative fossil fuel used in 20 US lime kilns and replaces 25% to 85% of traditional fuel.(Francey, Tran, and Jones 2009) Tire-derived fuel has been also tested in lime kiln operation, replacing 15% of the natural gas demand (Hart, Hanson III Glenn M., and Manning 2021).

Table S13. Life Cycle Inventory for the Scenarios Analyzed in the Present Study

Inputs		mt/ adt linerboard								
		Nat gas	Biomass boiler	Syngas	Pulverized biomass			Tall Oil Pitch		
					25%	50%	100%	25%	50%	100%
Pulp Biomass	Logs (50% moisture)	2.33	2.33	2.33	2.33	2.33	2.33	2.33	2.33	2.33
	Chips	2.10	2.10	2.10	2.10	2.10	2.10	2.10	2.10	2.10
Chemicals	NaOH makeup	6.86*10 ⁻⁰³	6.86*10 ⁻⁰³	6.85*10 ⁻⁰³	6.88*10 ⁻⁰³	6.92*10 ⁻⁰³	6.97*10 ⁻⁰³	6.89*10 ⁻⁰³	6.92*10 ⁻⁰³	6.99*10 ⁻⁰³
	Na ₂ SO ₄ makeup	5.37*10 ⁻⁰³	5.37*10 ⁻⁰³	5.37*10 ⁻⁰³	5.31*10 ⁻⁰³	5.26*10 ⁻⁰³	5.14*10 ⁻⁰³	5.32*10 ⁻⁰³	5.26*10 ⁻⁰³	5.14*10 ⁻⁰³
	CaO makeup	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.02*10 ⁻⁰²	2.01*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²
	H ₂ SO ₄	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³
Power plant fuels	External hog (Power plant)	8.82*10 ⁻⁰²	8.03*10 ⁻⁰¹	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²
	Natural gas Boiler	1.35*10 ⁻⁰¹	-	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹
Lime kiln fuels	Natural Gas Lime kiln	1.98*10 ⁻⁰²	1.98*10 ⁻⁰²	-	1.49*10 ⁻⁰²	9.90*10 ⁻⁰³	-	1.49*10 ⁻⁰²	9.90*10 ⁻⁰³	-
	Tall Oil Pitch (external)	-	-	-	-	-	-	8.42*10 ⁻⁰³	1.47*10 ⁻⁰²	2.83*10 ⁻⁰²
	Methanol	-	-	-	-	-	-	-	-	-
	Syngas (Internal)	-	-	1.88*10 ⁻⁰¹	-	-	-	-	-	-
	Biomass (5% mc or 10% mc for syngas)	-	-	2.31*10 ⁻⁰¹	1.37*10 ⁻⁰²	2.74*10 ⁻⁰²	5.48*10 ⁻⁰²	-	-	-
Water demand	Water-Paper machine	16.20	16.20	16.20	16.20	16.20	16.20	16.20	16.20	16.20
	Water - Brownstock washing	5.46	5.46	5.47	5.47	5.47	5.47	5.46	5.47	5.47
	Water -Causticizing plant	3.75	3.75	3.75	3.74	3.75	3.75	3.75	3.75	3.75
	Water-Power plant	2.92	2.92	2.92	2.92	2.92	2.92	2.92	2.92	2.92
	Total Water demand	28.34	28.33	28.34	28.34	28.34	28.34	28.33	28.34	28.34
Electricity from Grid	Linerboard mill (MWh/mt pulp)	5.54*10 ⁻⁰²	5.61*10 ⁻⁰²	5.48*10 ⁻⁰²	5.52*10 ⁻⁰²	5.48*10 ⁻⁰²	5.58*10 ⁻⁰²	5.54*10 ⁻⁰²	5.51*10 ⁻⁰²	5.47*10 ⁻⁰²
	Alternative fuel (MWh/mt pulp)	-	-	6.08*10 ⁻⁰³	1.20*10 ⁻⁰⁴	2.40*10 ⁻⁰⁴	4.49*10 ⁻⁰⁴	-	-	-
Outputs		Natural gas	Biomass boiler	Syngas	Pulverized biomass			Tall Oil Pitch		
					25%	50%	100%	25%	50%	100%
Products	Pulp	1	1	1	1	1	1	1	1	1
	Turpentine	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³
	CTO	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²
	Methanol	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³
Emissions		Nat gas	Biomass boiler	Syngas	Pulverized biomass			Tall Oil Pitch		
					25%	50%	100%	25%	50%	100%
Onsite emissions	CO ₂ Limekiln Total	1.61*10 ⁻⁰¹	1.61*10 ⁻⁰¹	2.29*10 ⁻⁰¹	1.72*10 ⁻⁰¹	1.83*10 ⁻⁰¹	2.05*10 ⁻⁰¹	1.66*10 ⁻⁰¹	1.72*10 ⁻⁰¹	1.83*10 ⁻⁰¹
	CO ₂ Fossil -Lime Kiln	5.44*10 ⁻⁰²	5.44*10 ⁻⁰²	-	4.08*10 ⁻⁰²	2.7162*10 ⁻⁰²	-	4.10*10 ⁻⁰²	2.80*10 ⁻⁰²	-
	CO ₂ Biogenic -Lime Kiln fuel	-	-	1.23*10 ⁻⁰¹	2.46*10 ⁻⁰²	4.91*10 ⁻⁰²	9.83*10 ⁻⁰²	1.90*10 ⁻⁰²	3.73*10 ⁻⁰²	7.70*10 ⁻⁰²
	CO ₂ Biogenic Lime Kiln CaCO ₃	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹
	CO ₂ Biogenic Recovery Boiler	1.61	1.61	1.61	1.61	1.61	1.61	1.61	1.61	1.61

Waste	CO ₂ Biogenic Hogfuel	2.41*10 ⁻⁰¹	9.16*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹
	CO ₂ Fossil Natural gas	3.70*10 ⁻⁰¹	-	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.71*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.71*10 ⁻⁰¹	3.71*10 ⁻⁰¹
	Dregs	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²
	Grits	4.94*10 ⁻⁰³	4.94*10 ⁻⁰³	4.94*10 ⁻⁰³	4.93*10 ⁻⁰³	4.94*10 ⁻⁰³	4.93*10 ⁻⁰³	4.94*10 ⁻⁰³	4.93*10 ⁻⁰³	4.93*10 ⁻⁰³
Wastewater	Ashes	1.40*10 ⁻⁰³	5.34*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³
	Wastewater Paper machine	20.22	20.22	20.22	20.22	20.22	20.22	20.22	20.22	20.22
	Wastewater Causticizing	6.58*10 ⁻⁰¹	6.58*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹
	Total Wastewater	20.88	20.88	20.88	20.88	20.88	20.88	2.09*10+01	2.09*10+01	2.09*10+01
	Sludge	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²

Table S13. Life Cycle Inventory for the Scenarios Analyzed in the Present Study (continued)

Input		mt/ adt linerboard						
		Turpentine (10%)	Methanol (10%)	Crude Tall Oil			Lignin	
				25%	50%	100%	25%	50%
Pulp Biomass	Logs (50% moisture)	2.33	2.33	2.33	2.33	2.33	2.33	2.33
	Chips	2.10	2.10	2.10	2.10	2.10	2.10	2.10
Chemicals	NaOH makeup	6.86*10 ⁻⁰³	6.86*10 ⁻⁰³	6.86*10 ⁻⁰³	6.86*10 ⁻⁰³	6.86*10 ⁻⁰³	8.00*10 ⁻⁰³	9.13*10 ⁻⁰³
	Na ₂ SO ₄ makeup	5.37*10 ⁻⁰³	5.37*10 ⁻⁰³	5.37*10 ⁻⁰³	5.37*10 ⁻⁰³	5.37*10 ⁻⁰³	3.40*10 ⁻⁰³	1.45*10 ⁻⁰³
	CaO makeup	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.03*10 ⁻⁰²	2.02*10 ⁻⁰²	2.01*10 ⁻⁰²
	H ₂ SO ₄	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³
Power plant fuels	External hog (Power plant)	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²
	Natural gas Boiler	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹
Lime kiln fuels	Natural Gas Lime kiln	1.79*10 ⁻⁰²	1.79*10 ⁻⁰²	1.49*10 ⁻⁰²	9.92*10 ⁻⁰³	-	1.48*10 ⁻⁰²	9.83*10 ⁻⁰³
	CTO	-	-	-	2.59*10 ⁻⁰³	1.63*10 ⁻⁰²	-	-
	Methanol	-	3.35*10 ⁻⁰³	-	-	-	-	-
	Biomass to replace lignin	-	-	-	-	-	1.60*10 ⁻⁰¹	1.87*10 ⁻⁰¹
Lignin extraction	CO ₂ Lignoboost	-	-	-	-	-	4.93*10 ⁻⁰³	9.71*10 ⁻⁰³
	H ₂ SO ₄ Lignoboost	-	-	-	-	-	1.03*10 ⁻⁰³	2.06*10 ⁻⁰³
	Lignin (70%)	-	-	-	-	-	1.40*10 ⁻⁰²	2.79*10 ⁻⁰²
Water demand	Water-Paper machine	16.20	16.20	16.20	16.20	16.20	16.20	16.20
	Water - Brownstock washing	5.46	5.46	5.46	5.46	5.46	5.43	5.40
	Water -Causticizing plant	3.75	3.75	3.75	3.75	3.75	3.74	3.74
	Water-Power plant	2.92	2.92	2.92	2.92	2.82	4.23	4.22
	Water-Lignoboost	-	-	-	-	-	2.85*10 ⁻⁰²	5.67*10 ⁻⁰²
	Total Water demand	28.33	28.34	28.33	28.34	28.24	29.64	29.62
Electricity from Grid	Linerboard mill (MWh/mt pulp)	5.55*10 ⁻⁰²	5.56*10 ⁻⁰²	5.53*10 ⁻⁰²	5.50*10 ⁻⁰²	5.44*10 ⁻⁰²	3.07*10 ⁻⁰²	2.94*10 ⁻⁰²
	Alternative fuel (MWh/mt pulp)	-	-	-	-	-	7.69*10 ⁻⁰²	1.53*10 ⁻⁰²
Outputs		Turpentine (10%)	Methanol (10%)	Crude Tall Oil			Lignin	

Products				25%	50%	100%	25%	50%
		1	1	1	1	1	1	1
Products	Pulp	1	1	1	1	1	1	1
	Turpentine mt/mt pulp	9.63*10 ⁻⁰⁴	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³
	CTO	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	4.24*10 ⁻⁰³	-	-	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²
	Methanol	2.54*10 ⁻⁰³	-	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³
Emissions		Turpentine (10%)	Methanol (10%)	Crude Tall Oil			Lignin	
				25%	50%	100%	25%	50%
Onsite emissions	CO ₂ Limekiln Total	1.63*10 ⁻⁰¹	1.63*10 ⁻⁰¹	1.67*10 ⁻⁰¹	1.73*10 ⁻⁰¹	1.86*10 ⁻⁰¹	1.70*10 ⁻⁰¹	1.79*10 ⁻⁰¹
	CO ₂ Fossil -Lime Kiln	4.90*10 ⁻⁰²	4.90*10 ⁻⁰²	4.08*10 ⁻⁰²	2.72*10 ⁻⁰²		4.06*10 ⁻⁰²	2.70*10 ⁻⁰²
	CO ₂ Biogenic -Lime Kiln fuel	7.74*10 ⁻⁰³	8.10*10 ⁻⁰³	1.99*10 ⁻⁰²	3.98*10 ⁻⁰²	7.96*10 ⁻⁰²	2.35*10 ⁻⁰²	4.67*10 ⁻⁰²
	CO ₂ Biogenic Lime Kiln CaCO ₃	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.38*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹
	CO ₂ Biogenic Recovery Boiler	1.61	1.61	1.61	1.61	1.61	1.59	1.56
	CO ₂ Biogenic Hogfuel	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	3.17*10 ⁻⁰¹	3.29*10 ⁻⁰¹
	CO ₂ Fossil Natural gas	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.70*10 ⁻⁰¹
Waste	Dregs	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.03*10 ⁻⁰²	1.02*10 ⁻⁰²
	Grits	4.94*10 ⁻⁰³	4.94*10 ⁻⁰³	4.94*10 ⁻⁰³	4.94*10 ⁻⁰³	4.94*10 ⁻⁰³	4.92*10 ⁻⁰³	4.91*10 ⁻⁰³
	Ashes	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.84*10 ⁻⁰³	1.92*10 ⁻⁰³
	Wastewater Paper machine	20.22	20.22	20.22	20.22	20.22	20.22	20.22
	Wastewater Causticizing	6.57*10 ⁻⁰¹	6.58*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.56*10 ⁻⁰¹	6.58*10 ⁻⁰¹	6.59*10 ⁻⁰¹
	Total Wastewater	20.9	20.9	20.9	20.9	20.9	20.9	20.9
	Sludge	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²

Table S13. Life Cycle Inventory for the Scenarios Analyzed in the Present Study (continued)

Input		mt/ adt linerboard				
		Fuel Oil	Petcoke			Tired-derived fuels (15%)
			25%	50%	85%	
Pulp Biomass	Logs (50% moisture)	2.33	2.33	2.33	2.33	2.33
	Chips	2.10	2.10	2.10	2.10	2.10
Chemicals	NaOH makeup	7.02*10 ⁻⁰³	7.57*10 ⁻⁰³	8.28*10 ⁻⁰³	9.28*10 ⁻⁰³	7.01*10 ⁻⁰³
	Na ₂ SO ₄ makeup	5.06*10 ⁻⁰³	4.09*10 ⁻⁰³	2.82*10 ⁻⁰³	1.04*10 ⁻⁰³	5.11*10 ⁻⁰³
	CaO makeup	2.03*10 ⁻⁰²	2.02*10 ⁻⁰²	2.01*10 ⁻⁰²	1.99*10 ⁻⁰²	1.98*10 ⁻⁰²
	H ₂ SO ₄	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³	3.90*10 ⁻⁰³
Power plant fuels	External hog (Power plant)	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²	8.82*10 ⁻⁰²
	Natural gas Boiler	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.35*10 ⁻⁰¹	1.36*10 ⁻⁰¹	1.35*10 ⁻⁰¹
Lime kiln fuels	Natural Gas Lime kiln	-	1.68*10 ⁻⁰²	9.88*10 ⁻⁰³	4.93*10 ⁻⁰³	1.68*10 ⁻⁰²
	Fuel Oil	2.36*10 ⁻⁰²	-	-	-	-
	Tired-derived fuels	-	-	-	-	5.13*10 ⁻⁰³
	Petcoke	-	8.20*10 ⁻⁰³	1.64*10 ⁻⁰²	2.78*10 ⁻⁰²	-

Water demand	Water-Paper machine	16.20	16.20	16.20	16.20	16.20
	Water - Brownstock washing	5.47	5.47	5.48	5.49	5.47
	Water -Causticizing plant	3.74	3.74	3.73	3.73	3.74
	Water-Power plant	2.82	2.92	2.82	2.82	2.92
	Total Water demand	28.24	28.34	28.24	28.24	28.33
Electricity from Grid	Linerboard mill (MWh/mt pulp)	5.54*10 ⁻⁰²	5.56*10 ⁻⁰²	5.51*10 ⁻⁰²	5.48*10 ⁻⁰²	5.56*10 ⁻⁰²
	Alternative fuel (MWh/mt pulp)	-	2.13*10 ⁻⁰⁴	4.26*10 ⁻⁰⁴	7.22*10 ⁻⁰⁴	2.56*10 ⁻⁰⁴
Outputs		Fuel Oil	Petcoke			Tired-derived fuels (15%)
			25%	50%	85%	
Products	Pulp	1	1	1	1	1
	Turpentine mt/mt pulp	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³	3.35*10 ⁻⁰³
	CTO	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²	1.30*10 ⁻⁰²
	Methanol	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³	2.54*10 ⁻⁰³
Emissions		Fuel Oil	Petcoke			Tired-derived fuels (15%)
			25%	50%	85%	
Onsite emissions	CO2 Limekiln Total	1.81*10 ⁻⁰¹	2.25*10 ⁻⁰¹	1.87*10 ⁻⁰¹	2.11*10 ⁻⁰¹	1.62*10 ⁻⁰¹
	CO2 Fossil -Lime Kiln	7.477*10 ⁻⁰²	7.29*10 ⁻⁰²	8.04*10 ⁻⁰²	1.04*10 ⁻⁰¹	5.55*10 ⁻⁰²
	CO2 Biogenic Lime Kiln CaCO3	1.06*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.07*10 ⁻⁰¹	1.07*10 ⁻⁰¹	1.07*10 ⁻⁰¹
	CO2 Biogenic Recovery Boiler	1.61	1.61	1.61	1.61	1.61
	CO2 Biogenic Hogfuel	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹	2.41*10 ⁻⁰¹
	CO2 Fossil Natural gas	3.71*10 ⁻⁰¹	3.70*10 ⁻⁰¹	3.71*10 ⁻⁰¹	3.72*10 ⁻⁰¹	3.70*10 ⁻⁰¹
Waste	Dregs	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²	1.04*10 ⁻⁰²
	Grits	4.93*10 ⁻⁰³	4.92*10 ⁻⁰³	4.91*10 ⁻⁰³	4.89*10 ⁻⁰³	4.93*10 ⁻⁰³
	Ashes	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³	1.40*10 ⁻⁰³
	Wastewater Paper machine	20.22	20.22	20.22	20.22	20.22
	Wastewater Causticizing	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.57*10 ⁻⁰¹	6.56*10 ⁻⁰¹	6.58*10 ⁻⁰¹
	Total Wastewater	20.88	20.88	20.88	20.88	20.88
	Sludge	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²	1.01*10 ⁻⁰²

Table S14. Mass Allocation Factors

Product	Base case	Biomass boiler	External bio-based fuels*	Crude Tall Oil			Lignin		Methanol (10%)	Turpentine (10%)	Other fossil-based fuels
				25%	50%	100%	25%	50%			
Pulp	98.15%	98.15%	98.15%	99.00%	99.41%	99.41%	98.15 %	98.15 %	98.39%	98.38%	98.15%
Turpentine	0.33%	0.33%	0.33%	0.33%	0.33%	0.33%	0.33%	0.33%	0.33%	0.09%	0.33%
CTO	1.27%	1.27%	1.27%	0.42%	0.00%	0.00%	1.28%	1.28%	1.28%	1.28%	1.27%
Methanol	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%	0.00%	0.25%	0.25%

*Includes pulverized biomass, biomass Gasification and tall oil pitch

Table S15. GWP for the Upstream Processes and Wastes in the Production of Linerboard

Process	Value	Unit
Loblolly pine logs (dry basis). (Lan <i>et al.</i> 2020; Wernet <i>et al.</i> 2016)	6.36×10^{-02}	kgCO ₂ -eq/kg product
Softwood forestry, pine, sustainable forest management wood chips, wet, measured as dry mass Cutoff, U, RoW. (Wernet <i>et al.</i> 2016)	4.02×10^{-02}	kgCO ₂ -eq/kg product
Chlor-alkali electrolysis, diaphragm cell sodium hydroxide, without water, in 50% solution state Cutoff, U, RoW. (Wernet <i>et al.</i> 2016)	1.45	kgCO ₂ -eq/kg product
Sodium sulfate production, from natural sources sodium sulfate, anhydrite Cutoff, U, RoW. (Wernet <i>et al.</i> 2016)	1.78×10^{-01}	kgCO ₂ -eq/kg product
Quicklime production, milled, loose quicklime, milled, loose Cutoff, U, RoW.(Wernet <i>et al.</i> 2016)	1.17	kgCO ₂ -eq/kg product
Sulfuric acid production sulfuric acid Cutoff, U, RoW(Wernet <i>et al.</i> 2016)	9.84×10^{-02}	kgCO ₂ -eq/kg product
External hog (Power plant). softwood forestry, pine, sustainable forest management wood chips, wet, measured as dry mass Cutoff, U, RoW. (Wernet <i>et al.</i> 2016)	4.02×10^{-02}	kgCO ₂ -eq/kg product
Market for natural gas, high pressure natural gas, high pressure Cutoff, U, US. (Wernet <i>et al.</i> 2016)	3.59×10^{-01}	kgCO ₂ -eq/kg product
Heavy fuel oil production, petroleum refinery operation heavy fuel oil Cutoff, U, RoW.(Wernet <i>et al.</i> 2016)	3.23×10^{-01}	kgCO ₂ -eq/kg product
Biomass gasification. (Wernet <i>et al.</i> 2016; Rofouiceraghi 2012)	2.70×10^{-02}	kgCO ₂ -eq/kg biomass processed -10% moisture content

Biomass drying (from 50 % to 5% moisture content). (Wernet <i>et al.</i> 2016; Rofouieeraghi 2012)	$4.25 \cdot 10^{-3}$	kgCO ₂ -eq/ kg biomass processed -50% moisture content
Biomass drying (from 50 % to 10% moisture content). (Wernet <i>et al.</i> 2016; Rofouieeraghi 2012)	$3.90 \cdot 10^{-3}$	kgCO ₂ -eq/ kg biomass processed -50% moisture content
Biomass pulverization. ¹¹ (Wind <i>et al.</i> 2018)	$3.50 \cdot 10^{-4}$	kgCO ₂ -eq/kg biomass processed -5% moisture content
Transport, freight, lorry 16-32 metric ton, EURO5. ¹¹	$1.72 \cdot 10^{-1}$	t*km
Market for green liquor dregs green liquor dregs Cutoff, U, GLO. ¹¹	$3.32 \cdot 10^{-1}$	kgCO ₂ -eq/kg waste
Grits. Market for municipal solid waste municipal solid waste Cutoff, U, RoW. ¹¹	$9.20 \cdot 10^{-1}$	kgCO ₂ -eq/kg waste
Market for wood ash mixture, pure wood ash mixture, pure Cutoff, U, RoW. ¹¹	$1.45 \cdot 10^{-2}$	kgCO ₂ -eq/kg waste
Market for sludge from pulp and paper production sludge from pulp and paper production Cutoff, U, RoW. ¹¹	1.15	kgCO ₂ -eq/kg waste
Electricity, high voltage, production mix electricity, high voltage Cutoff, U - US-SERC. ¹¹	$5.24 \cdot 10^{-1}$	kg CO2 eq/ kWh
Tall oil Pitch. ¹¹ (Cashman, Moran, and Gaglione 2016)	$4.46 \cdot 10^{-1}$	kgCO ₂ -eq/kg product
Lignin drier (70% moisture content). (Wernet <i>et al.</i> 2016; Rofouieeraghi 2012)	$2.88 \cdot 10^{-1}$	kgCO ₂ -eq/kg lignin dried
CO ₂ for Lignoboost. Carbon dioxide production, liquid carbon dioxide, liquid Cutoff, U, RoW. ¹¹	$8.78 \cdot 10^{-1}$	kgCO ₂ -eq/kg product
Sulfuric acid production sulfuric acid Cutoff, U, RoW. ¹¹	$9.84 \cdot 10^{-2}$	kgCO ₂ -eq/kg product
Grinded petcoke. Petroleum coke production, petroleum refinery operation petroleum coke Cutoff, U, RoW ¹¹ (Ernst and Galitsky 2004)	$3.62 \cdot 10^{-1}$	kgCO ₂ -eq/kg product
Tired-derived fuels. ¹¹ (Feraldi <i>et al.</i> 2013)	$5.24 \cdot 10^{-2}$	kgCO ₂ -eq/kg product

Table S16. GWP for the Scenarios Analyzed in the Present Study (continued)

Process		kg CO ₂ -eq/kg product						
		Turpentine (10%)	Methanol (10%)	Crude Tall Oil			Lignin	
				25%	50%	100%	25%	50%
Pulp Biomass	Logs (50% moisture)	$7.30 \cdot 10^{-2}$	$7.30 \cdot 10^{-2}$	$7.34 \cdot 10^{-2}$	$7.37 \cdot 10^{-2}$	$7.37 \cdot 10^{-2}$	$7.28 \cdot 10^{-2}$	$7.28 \cdot 10^{-2}$
	Chips	$8.31 \cdot 10^{-2}$	$8.31 \cdot 10^{-2}$	$8.36 \cdot 10^{-2}$	$8.39 \cdot 10^{-2}$	$8.39 \cdot 10^{-2}$	$8.29 \cdot 10^{-2}$	$8.29 \cdot 10^{-2}$
Chemicals	NaOH makeup	$9.76 \cdot 10^{-3}$	$9.76 \cdot 10^{-3}$	$9.82 \cdot 10^{-3}$	$9.86 \cdot 10^{-3}$	$9.86 \cdot 10^{-3}$	$1.14 \cdot 10^{-2}$	$1.30 \cdot 10^{-2}$
	Na ₂ SO ₄ makeup	$9.42 \cdot 10^{-4}$	$9.42 \cdot 10^{-4}$	$9.48 \cdot 10^{-4}$	$9.51 \cdot 10^{-4}$	$9.52 \cdot 10^{-4}$	$5.94 \cdot 10^{-4}$	$2.53 \cdot 10^{-4}$
	CaO	$2.33 \cdot 10^{-2}$	$2.34 \cdot 10^{-2}$	$2.35 \cdot 10^{-2}$	$2.36 \cdot 10^{-2}$	$2.36 \cdot 10^{-2}$	$2.32 \cdot 10^{-2}$	$2.30 \cdot 10^{-2}$
	H ₂ SO ₄	$3.77 \cdot 10^{-4}$	$3.77 \cdot 10^{-4}$	$3.80 \cdot 10^{-4}$	$3.81 \cdot 10^{-4}$	$3.81 \cdot 10^{-4}$	$3.76 \cdot 10^{-4}$	$3.76 \cdot 10^{-4}$

Power plant fuels	External hog	3.49*10 ⁻⁰³	3.49*10 ⁻⁰³	3.51*10 ⁻⁰³	3.52*10 ⁻⁰³	3.52*10 ⁻⁰³	3.48*10 ⁻⁰³	3.48*10 ⁻⁰³
	Natural gas-boiler	4.76*10 ⁻⁰²	4.76*10 ⁻⁰²	4.80*10 ⁻⁰²	4.82*10 ⁻⁰²	4.82*10 ⁻⁰²	4.75*10 ⁻⁰²	4.75*10 ⁻⁰²
Lime kiln fuels*	Natural Gas-lime kiln	6.30*10 ⁻⁰³	6.30*10 ⁻⁰³	5.28*10 ⁻⁰³	3.54*10 ⁻⁰³	-	5.22*10 ⁻⁰³	3.46*10 ⁻⁰³
	Crude tall oil	-	-	-	2.90*10 ⁻⁰⁵	1.82*10 ⁻⁰⁴	-	-
	Methanol	-	7.25*10 ⁻⁰⁶	-	-	-	-	-
	Lignin Drying	-	-	-	-	-	3.95*10 ⁻⁰³	7.87*10 ⁻⁰³
	CO ₂ Lignoboost	-	-	-	-	-	4.25*10 ⁻⁰³	8.37*10 ⁻⁰³
	H ₂ SO ₄ Lignoboost	-	-	-	-	-	9.98*10 ⁻⁰⁵	1.99*10 ⁻⁰⁴
	Biomass to replace natural gas	-	-	-	-	-	6.31*10 ⁻⁰³	7.36*10 ⁻⁰³
Electricity	Linerboard mill	2.86*10 ⁻⁰²	2.87*10 ⁻⁰²	2.87*10 ⁻⁰²	2.87*10 ⁻⁰²	2.84*10 ⁻⁰²	1.58*10 ⁻⁰²	1.51*10 ⁻⁰²
Transport	Transport biomass	1.53*10 ⁻⁰¹	1.53*10 ⁻⁰¹	1.54*10 ⁻⁰¹	1.55*10 ⁻⁰¹	1.55*10 ⁻⁰¹	1.63*10 ⁻⁰¹	1.65*10 ⁻⁰¹
	Transport materials	6.17*10 ⁻⁰⁴	6.74*10 ⁻⁰⁴	6.21*10 ⁻⁰⁴	6.68*10 ⁻⁰⁴	9.02*10 ⁻⁰⁴	7.00*10 ⁻⁰⁴	7.83*10 ⁻⁰⁴
On-site emissions	CO ₂ fossil from fuel-lime kiln	4.82*10 ⁻⁰²	4.82*10 ⁻⁰²	4.04*10 ⁻⁰²	2.71*10 ⁻⁰²	-	3.99*10 ⁻⁰²	2.65*10 ⁻⁰²
	CO ₂ biogenic from fuel-lime kiln	7.61*10 ⁻⁰³	7.97*10 ⁻⁰³	1.97*10 ⁻⁰²	3.95*10 ⁻⁰²	7.91*10 ⁻⁰²	2.30*10 ⁻⁰²	4.59*10 ⁻⁰²
	CO ₂ biogenic from CaCO ₃ -lime kiln	1.05*10 ⁻⁰¹	1.05*10 ⁻⁰¹	1.05*10 ⁻⁰¹	1.06*10 ⁻⁰¹	1.38*10 ⁻⁰¹	1.04*10 ⁻⁰¹	1.04*10 ⁻⁰¹
	CO ₂ biogenic- recovery boiler	1.59	1.59	1.60	1.60	1.60	1.56	1.53
	CO ₂ biogenic-biomass boiler	2.37*10 ⁻⁰¹	2.37*10 ⁻⁰¹	2.39*10 ⁻⁰¹	2.40*10 ⁻⁰¹	2.40*10 ⁻⁰¹	3.11*10 ⁻⁰¹	3.23*10 ⁻⁰¹
	CO ₂ fossil - natural gas boiler	3.64*10 ⁻⁰¹	3.64*10 ⁻⁰¹	3.67*10 ⁻⁰¹	3.68*10 ⁻⁰¹	3.68*10 ⁻⁰¹	3.63*10 ⁻⁰¹	3.63*10 ⁻⁰¹
Waste	Dregs	3.40*10 ⁻⁰³	3.40*10 ⁻⁰³	3.42*10 ⁻⁰³	3.43*10 ⁻⁰³	3.43*10 ⁻⁰³	3.36*10 ⁻⁰³	3.34*10 ⁻⁰³
	Grits	4.47*10 ⁻⁰³	4.47*10 ⁻⁰³	4.49*10 ⁻⁰³	4.51*10 ⁻⁰³	4.51*10 ⁻⁰³	4.44*10 ⁻⁰³	4.43*10 ⁻⁰³
	Ashes	2.00*10 ⁻⁰⁵	2.00*10 ⁻⁰⁵	2.02*10 ⁻⁰⁵	2.02*10 ⁻⁰⁵	2.02*10 ⁻⁰⁵	2.62*10 ⁻⁰⁵	2.73*10 ⁻⁰⁵
	Sludge	1.14*10 ⁻⁰²	1.14*10 ⁻⁰²	1.15*10 ⁻⁰²	1.16*10 ⁻⁰²	1.16*10 ⁻⁰²	1.14*10 ⁻⁰²	1.14*10 ⁻⁰²
CO ₂ emissions	Total CO ₂ emissions	2.80	2.80	2.82	2.83	2.88	2.86	2.86
	GWP	8.62*10 ⁻⁰¹	8.62*10 ⁻⁰¹	8.58*10 ⁻⁰¹	8.47*10 ⁻⁰¹	8.16*10 ⁻⁰¹	8.63*10 ⁻⁰¹	8.60*10 ⁻⁰¹
	CO ₂ biogenic	1.94	1.94	1.96	1.99	2.06	2.00	2.00

*Includes the contribution from the electricity demanded by the alternative lime kiln fuel production

Table S16. GWP for the Scenarios Analyzed in the Present Study (continued)

Process		kg CO ₂ -eq/kg product				
		Fuel Oil	Petcoke			Tired-derived fuels (15%)
			25%	50%	85%	
Pulp Biomass	Logs (50% moisture)	7.28*10 ⁻⁰²	7.28*10 ⁻⁰²	7.28*10 ⁻⁰²	7.28*10 ⁻⁰²	7.28*10 ⁻⁰²
	Chips	8.29*10 ⁻⁰²	8.29*10 ⁻⁰²	8.29*10 ⁻⁰²	8.29*10 ⁻⁰²	8.29*10 ⁻⁰²
Chemicals	NaOH makeup	9.96*10 ⁻⁰³	1.07*10 ⁻⁰²	1.18*10 ⁻⁰²	1.32*10 ⁻⁰²	9.95*10 ⁻⁰³

	Na ₂ SO ₄ makeup	8.85*10 ⁻⁰⁴	7.16*10 ⁻⁰⁴	4.93*10 ⁻⁰⁴	1.83*10 ⁻⁰⁴	8.94*10 ⁻⁰⁴
	CaO	2.33*10 ⁻⁰²	2.32*10 ⁻⁰²	2.30*10 ⁻⁰²	2.28*10 ⁻⁰²	2.27*10 ⁻⁰²
	H ₂ SO ₄	3.76*10 ⁻⁰⁴	3.76*10 ⁻⁰⁴	3.76*10 ⁻⁰⁴	3.76*10 ⁻⁰⁴	3.76*10 ⁻⁰⁴
Power plant fuels	External hog	3.48*10 ⁻⁰³	3.48*10 ⁻⁰³	3.48*10 ⁻⁰³	3.48*10 ⁻⁰³	3.48*10 ⁻⁰³
	Natural gas-boiler	4.77*10 ⁻⁰²	4.74*10 ⁻⁰²	4.76*10 ⁻⁰²	4.78*10 ⁻⁰²	4.74*10 ⁻⁰²
Lime kiln fuels*	Natural Gas-lime kiln	-	5.93*10 ⁻⁰³	3.48*10 ⁻⁰³	1.74*10 ⁻⁰³	5.91*10 ⁻⁰³
	Fuel Oil	7.48*10 ⁻⁰³	-	-	-	-
	Tired-derived fuels	-	-	-	-	1.32*10 ⁻⁰⁴
	Petcoke	-	1.46*10 ⁻⁰³	2.91*10 ⁻⁰³	4.94*10 ⁻⁰³	-
Electricity	Electricity	2.85*10 ⁻⁰²	2.86*10 ⁻⁰²	2.84*10 ⁻⁰²	2.82*10 ⁻⁰²	2.86*10 ⁻⁰²
Transport	Transport biomass	1.53*10 ⁻⁰¹	1.53*10 ⁻⁰¹	1.53*10 ⁻⁰¹	1.53*10 ⁻⁰¹	1.53*10 ⁻⁰¹
	Transport materials	1.01*10 ⁻⁰³	7.43*10 ⁻⁰⁴	8.70*10 ⁻⁰⁴	1.05*10 ⁻⁰³	6.92*10 ⁻⁰⁴
On-site emissions	CO ₂ fossil from fuel-lime kiln	7.34*10 ⁻⁰²	7.15*10 ⁻⁰²	7.90*10 ⁻⁰²	1.02*10 ⁻⁰¹	5.45*10 ⁻⁰²
	CO ₂ biogenic from fuel-lime kiln	-	-	-	-	-
	CO ₂ biogenic from CaCO ₃ -lime kiln	1.04*10 ⁻⁰¹	1.04*10 ⁻⁰¹	1.05*10 ⁻⁰¹	1.05*10 ⁻⁰¹	1.05*10 ⁻⁰¹
	CO ₂ biogenic- recovery boiler	1.58	1.58	1.58	1.58	1.58
	CO ₂ biogenic-biomass boiler	2.37*10 ⁻⁰¹	2.37*10 ⁻⁰¹	2.37*10 ⁻⁰¹	2.37*10 ⁻⁰¹	2.37*10 ⁻⁰¹
	CO ₂ fossil - natural gas boiler	3.64*10 ⁻⁰¹	3.63*10 ⁻⁰¹	3.64*10 ⁻⁰¹	3.65*10 ⁻⁰¹	3.63*10 ⁻⁰¹
Waste	Dregs	3.39*10 ⁻⁰³	3.39*10 ⁻⁰³	3.38*10 ⁻⁰³	3.38*10 ⁻⁰³	3.39*10 ⁻⁰³
	Grits	4.45*10 ⁻⁰³	4.44*10 ⁻⁰³	4.43*10 ⁻⁰³	4.41*10 ⁻⁰³	4.45*10 ⁻⁰³
	Ashes	2.00*10 ⁻⁰⁵	2.00*10 ⁻⁰⁵	2.00*10 ⁻⁰⁵	2.00*10 ⁻⁰⁵	2.00*10 ⁻⁰⁵
	Sludge	1.14*10 ⁻⁰²	1.14*10 ⁻⁰²	1.14*10 ⁻⁰²	1.14*10 ⁻⁰²	1.14*10 ⁻⁰²
CO ₂ emissions	Total CO ₂ emissions	2.81*10+00	2.81*10+00	2.82*10+00	2.84*10+00	2.79*10+00
	GWP	8.88*10 ⁻⁰¹	8.85*10 ⁻⁰¹	8.93*10 ⁻⁰¹	9.19*10 ⁻⁰¹	8.65*10 ⁻⁰¹
	CO ₂ Biogenic	1.92	1.92	1.92	1.92	1.92

*Includes the contribution from the electricity demanded by the alternative lime kiln fuel production

Table S17. Hotspot Analysis for Alternatives that Represent a Reduction in the GWP for Linerboard Production

Scope	Alternative Process	Biomass Boiler	Pulverized Biomass (25%)	Pulverized Biomass (50%)	Pulverized Biomass (100%)	Biomass Gasification	Tall Oil Pitch (25%)	Tall Oil Pitch (50%)	Tall Oil Pitch (100%)	Crude Tall Oil (25%)	Crude Tall Oil (50%)	Crude Tall Oil (100%)	Lignin (25%)	Lignin (50%)	Methanol (10%)	Turpentine (10%)
Scope 1	Fossil CO ₂ (Lime kiln)	0.0%	-1.5%	-3.1%	-6.2%	-6.2%	-1.5%	-3.0%	-6.2%	-1.5%	-3.0%	-6.2%	-1.6%	-3.1%	-0.6%	-0.6%
	Fossil CO ₂ (Boiler)	-41.9%	0.0%	0.1%	0.1%	0.1%	0.1%	0.1%	0.2%	0.4%	0.6%	0.6%	0.0%	0.0%	0.1%	0.2%
Scope 2	Electricity (mill)	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	-1.5%	-1.5%	0.0%	0.0%
Scope 3	Chemicals	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.1%	0.1%	0.6%	1.3%	0.0%	0.0%
	Biomass (Energy)	3.3%	0.1%	0.2%	0.5%	1.9%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.7%	0.9%	0.0%	0.0%
	Natural gas production (Boiler)	-5.5%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.1%	0.1%	0.1%	0.0%	0.0%	0.0%	0.0%
	Natural gas production (Lime Kiln)	0.0%	-0.2%	-0.4%	-0.8%	-0.8%	-0.2%	-0.4%	-0.8%	-0.2%	-0.4%	-0.8%	-0.2%	-0.4%	-0.1%	-0.1%
	Alternative Fuel Production	0.0%	0.0%	0.0%	0.0%	0.7%	0.4%	0.7%	1.4%	0.0%	0.0%	0.0%	0.5%	0.9%	0.0%	0.0%
	Transport	2.8%	0.1%	0.2%	0.4%	1.7%	0.0%	0.0%	0.1%	0.2%	0.2%	0.3%	1.2%	1.4%	0.1%	0.0%
	Waste disposal	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
Net reduction		-41.3%	-1.5%	-3.0%	-5.9%	-2.6%	-1.2%	-2.5%	-5.3%	-1.0%	-2.3%	-5.8%	-0.2%	-0.7%	-0.4%	-0.4%

MARGINAL ABATEMENT COST CURVE

Capital Investment

The Marginal Abatement Cost Curve was developed based on the CO₂-eq reductions of each scenario and the Net Present Value (NPV). The NPV was determined assuming an implementation of 11 years, with the first year for construction, and a start of operation in the second year. 75% of the capital investment is made in the first year and 25% in the second year. The annual maintenance capital and the annual maintenance and repair cost is 1% and 2% of the cost of the equipment, respectively. The cash flow analysis considers a linear depreciation of 10% during the 10 years. For the scenarios where the coproduct is used as alternative fuel in the lime kiln (crude tall oil, methanol, turpentine), the revenue lost by burning the fuel is considered an operating cost in the analysis. The equipment cost is based on secondary sources (Laboratory and Laboratory 2013; Benali *et al.* 2016; Rofouieeraghi 2012) and recommendations of industry experts (Hart 2022). The cost for biomass gasification system, pulverized biomass and lignin extraction was adjusted to the capacity and year of investment assuming different scale exponents recommended by industry experts (0.6, 0.4, and 0.2, respectively) (Hart 2022). The equipment cost of the rest of the alternative fuels were provided for the year 2022, (Hart 2022) while the biomass boiler was adjusted from a total capital investment of \$158 million dollars for 2022, with a capacity of 400,000 lb steam/h (Hart 2022).

Table S18. Capital Investment of the Alternatives to Reduce the GHG Emissions in Linerboard Production

Direct Cost	% Total capital investment	Pulverized biomass 100% (Laboratory and Laboratory 2013; Rofouieeraghi 2012)	Gasification 100% (Laboratory and Laboratory 2013; Rofouieeraghi 2012)	Tall Oil Pitch 100% (Hart 2022)	Crude tall oil 100% (Hart 2022)	Methanol 10% (Hart 2022)	Turpentine 10% (Hart 2022)	Lignin 50% (Benali <i>et al.</i> 2016)	Biomass Boiler (Hart 2022)
- Purchased Equipment	100%	\$6.580	\$23.792	\$0.500	\$0.500	\$0.200	\$0.500	\$6.704	-
- Purchased Equipment Installation	13.3%	\$0.877	\$3.172	\$0.067	\$0.067	\$0.027	\$0.067	\$0.894	-
- Instrumentation and Controls	20.0%	\$1.316	\$4.758	\$0.100	\$0.100	\$0.040	\$0.100	\$1.341	-
- Piping	13.3%	\$0.877	\$3.172	\$0.067	\$0.067	\$0.027	\$0.067	\$0.894	-
- Electrical Systems	13.3%	\$0.877	\$3.172	\$0.067	\$0.067	\$0.027	\$0.067	\$0.894	-
- Service Facilities	6.7%	\$0.439	\$1.586	\$0.033	\$0.033	\$0.013	\$0.033	\$0.447	-
Sub-Total Direct Cost	166.7%	\$10.966	\$39.654	\$0.833	\$0.833	\$0.333	\$0.833	\$11.173	\$-
Indirect Cost		Pulverized biomass 100%	Gasification 100%	Tall Oil Pitch 100%	Crude tall oil 100%	Methanol 10%	Turpentine 10%	Lignin 50%	Biomass Boiler
- Engineering	20.0%	\$1.316	\$4.758	\$0.100	\$0.100	\$0.040	\$0.100	\$1.341	-
- Construction Expenses	20.0%	\$1.316	\$4.758	\$0.100	\$0.100	\$0.040	\$0.100	\$1.341	-
- Contractor Fee	13.3%	\$0.877	\$3.172	\$0.067	\$0.067	\$0.027	\$0.067	\$0.894	-
- Inflation	20.0%	\$1.316	\$4.758	\$0.100	\$0.100	\$0.040	\$0.100	\$1.341	-
- Contingency	20.0%	\$1.316	\$4.758	\$0.100	\$0.100	\$0.040	\$0.100	\$1.341	-
Sub-Total Indirect Cost	93%	\$6.141	\$22.206	\$0.467	\$0.467	\$0.187	\$0.467	\$6.257	-
TOTAL CAPITAL INVESTMENT	260%	\$17.107	\$61.860	\$1.300	\$1.300	\$0.520	\$1.300	\$17.429	\$178.852

Direct cost structure

The change in the operation cost was based on the mass and energy balance and the prices reported for the alternative fuels and chemicals. The prices are included in Table S19.

Table S19. Electricity and Chemical Prices

Variable	Cost
Electricity (Cents per Kilowatthour) (Administration, n.d.)	7.26
Natural gas (\$/1000 ft ³) (Administration, n.d.)	5.5*
biomass (hog fuel) (\$/ton) (Administration, n.d.)	34.4
biomass transport (\$-t km) (Stolaroff <i>et al.</i> 2021)	0.101
Crude tall Oil (US \$/mt) (Niemeläinen 2018; Stolaroff <i>et al.</i> 2021)	400
Tall Oil Pitch (US \$/mt) (Niemeläinen 2018)	400
NaOH (US \$/mt) ²	728
Na ₂ SO ₄ (US \$/mt) ²	88
H ₂ SO ₄ (US \$/mt) ²	88
Methanol (US \$/mt) (IHSMARKIT, n.d.)	350
Turpentine (US \$/mt) ²	716
CO ₂ (US \$/mt) (Inc. 2022)	250

*Electric power price

Figure S6 shows the direct cost structure for each alternative. In the case of pulverized biomass in the lime kiln and biomass boiler to replace the gas boiler, the displacement of natural gas represents a savings in the direct cost of \$0.08 and \$1.37 million dollars per year, respectively. This is reasonable given the prices reported by linerboard mills in the US; for natural gas, the average price in Q1 2022 is \$ 6.1/mmBTU, whereas for residual biomass, the price is \$ 2.8/mmBTU (Fisher International Inc, n.d.).

In contrast, biomass gasification has a direct cost of \$10.9 million dollars per year due to the low syngas-to-biomass ratio that increases biomass demand compared to pulverized biomass. In the case of lignin, the biomass demand to replace the extracted black liquor also increases the direct cost; the demand of chemicals to precipitate the lignin also is a variable that increases the direct cost to \$7.9 million dollars per year.

In the case of TOP, CTO, turpentine and methanol, the high cost of the external alternative fuel and the revenue lost by burning the on-site alternative fuel production countered the net savings, giving a net direct cost of \$4.96, \$5.25, \$0.95 and \$1.22 million dollars per year, respectively.

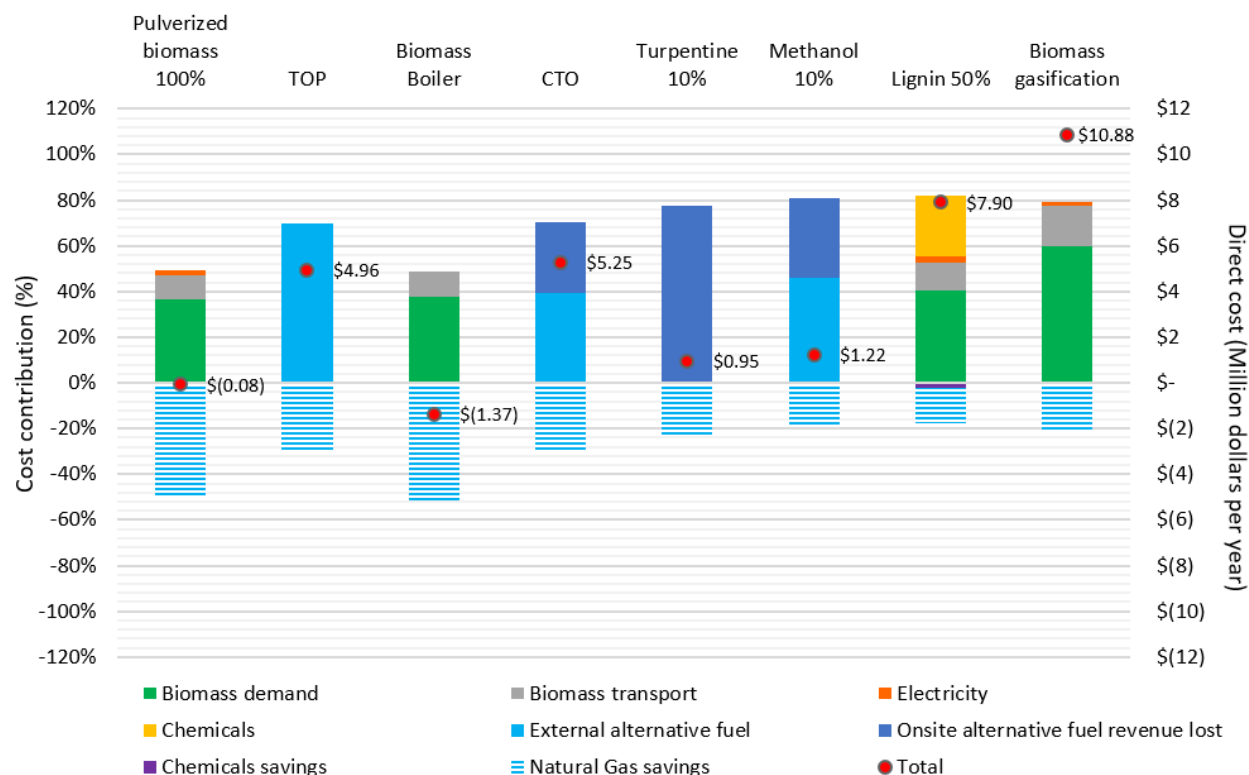


Fig. S6. Direct cost structure for alternatives to reduce the GHG emissions in the production of linerboard. External alternative fuel and onsite alternative fuel refer to crude tall oil, methanol, and turpentine

Sensitivity Analysis

To identify the economic variables that affect the CAC (or the NPV), a sensitivity analysis was performed varying $\pm 25\%$ the raw materials costs and the capital investment without including a revenue from the carbon offset (Fig. 7). In the case of pulverized biomass, the main variable is the natural gas price, with a variation of $\pm 26.7\%$ for the CAC, followed by capital investment with $\pm 25.5\%$, biomass cost with $\pm 19.3\%$, and biomass transport with $\pm 5.7\%$. Among the alternative fuels that imply mayor modifications in the mill (biomass gasification, lignin extraction and pulverized biomass) this is the one with the lowest capital investment, and mayor savings in the direct costs.

In the case of biomass boiler and biomass gasification; the capital investment is the factor that most affects the CAC, with a variation of $\pm 25.9\%$ for the biomass boiler, and $\pm 13.7\%$ for biomass gasification. The natural gas savings are more relevant for the biomass boiler with a variation of $\pm 17.6\%$ vs. a $\pm 4\%$ for biomass gasification, this is due the higher natural gas volume displaced on the first technology. The biomass price is also a variable with an important effect on the CAC, resulting in a variation of $\pm 12.9\%$ in the biomass boiler and $\pm 11.5\%$ in biomass gasification.

For lignin extraction, biomass is the most important factor rather than capital cost, with a variation of $\pm 10.7\%$ by the biomass cost and $\pm 3.1\%$ by its transport cost. The biomass relevance in the CAC is a consequence of two factors. First, the energy provided in the recovery boiler by the extracted lignin is covered by increasing the biomass demand in the boiler; however, lignin has a higher HHV (26.5 MJ/kg vs. 20.5 MJ/kg). In addition, the steam demand increases by the liquor

return from the Lignoboost process, increasing the biomass demand in the boiler. Therefore, the capital investment is slightly less relevant with a variation of $\pm 8.0\%$. Regarding the rest of the raw materials, natural gas savings has a variation of $\pm 4.2\%$, followed by CO_2 with a variation of $\pm 4.1\%$, NaOH with $\pm 2.8\%$; Na_2SO_4 savings with $\pm 0.5\%$, and H_2SO_4 with $\pm 0.3\%$.

Regarding the streams available in the mill as lime kiln fuel alternatives (CTO, TOP, turpentine, and methanol), these alternatives have low capital investment; however, the high price of these alternative fuels compared to natural gas affects the CAC considerably. For CTO and TOP, the CAC variation is around $\pm 42\%$ with the cost of the alternative fuel, whereas for the natural gas and the capital investment are around ± 18 and $\pm 1.3\%$, respectively. For turpentine and methanol, the variation in the CAC by the alternative fuel cost is $\pm 27.1\%$ and $\pm 30.1\%$; $\pm 7.8\%$ and $\pm 7.2\%$ with the natural gas cost; and $\pm 5.7\%$ and $\pm 2.1\%$ with the capital investment, respectively. In summary, the utilization of the streams available in the mill has low capital investment but the high price of these alternative fuels impacts the CAC considerably. For technologies that rely on biomass, as pulverized biomass, biomass boiler, and biomass gasification, the CAC has an important variation with the capital investment, followed by biomass. In the case of lignin extraction, the biomass demand is the variable that most impacts the CAC, given that the demand of biomass is increased to compensate the energy content of the extracted liquor. The savings in natural gas for all the alternatives is also relevant; in the case of pulverized biomass, the natural gas cost is the variable that most impacts the CAC.

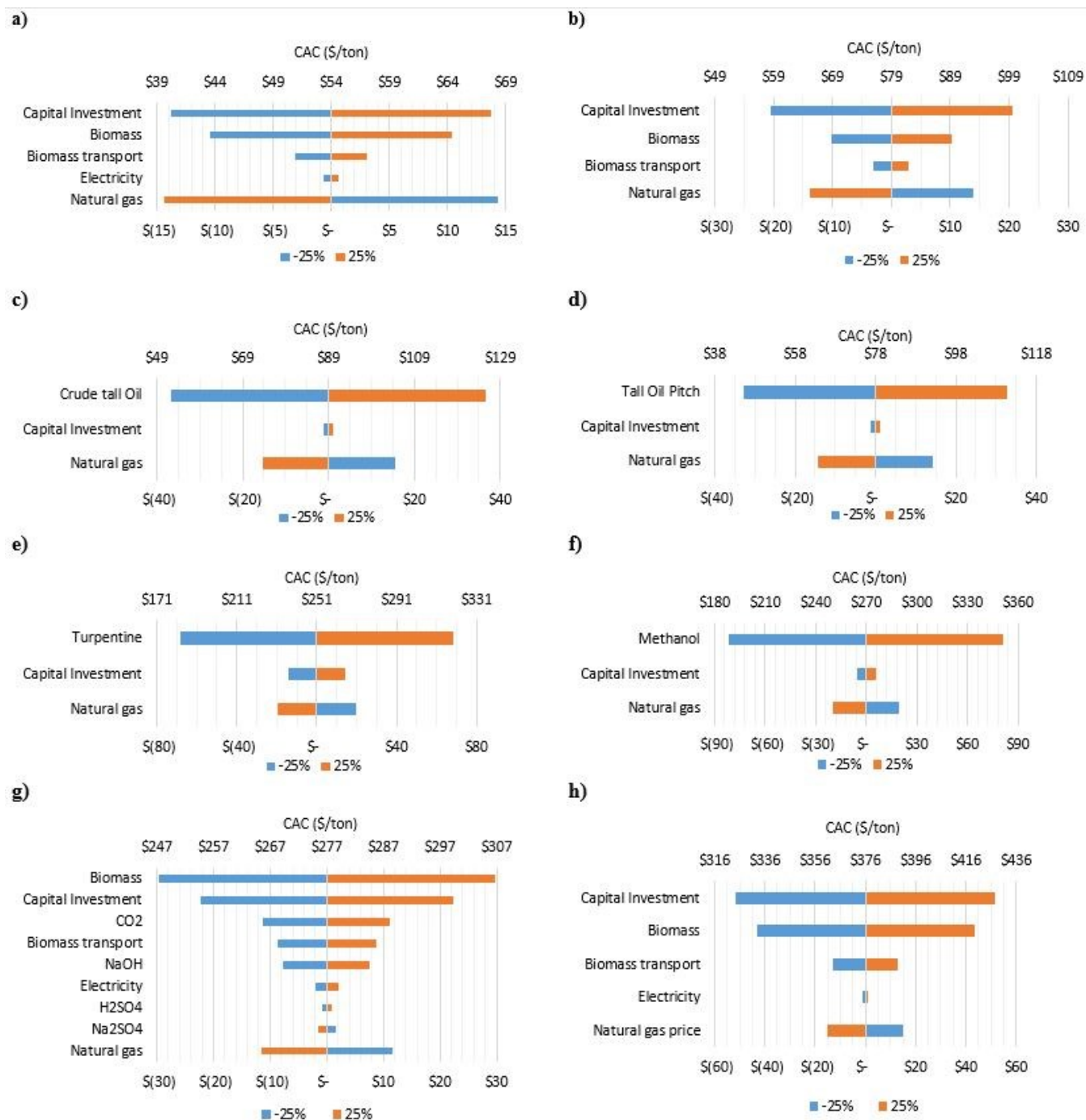


Fig. S7. Sensitivity analysis for the Cost of Carbon Avoided (Scope 1 & 2). Change in the variables is +/- 25% of the value assumed for each scenario. a) Pulverized biomass, b) Biomass boiler, c) Crude tall oil, d) Tall oil pitch, e) Turpentine, f) Methanol, g) Lignin extraction, h) Biomass gasification

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