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 CO_2-eq is emitted, with 69% being biogenic CO_2 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 CO_2-eq avoided.

DOI: 10.15376/biores.19.4.7806-7823

Keywords: Alternative lime kiln fuel; Biomass boiler; Life cycle assessment; Marginal abatement cost curve

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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 CO_2 reductions of the alternatives were determined through a detailed process mass and energy balance simulation using

WinGEMS. The alternatives are categorized by constructing a marginal carbon abatement cost curve (MACC), this MACC categorizes the alternatives by the cost of reducing 1 ton of CO_2-eq (carbon abatement cost) and shows the CO_2-eq reductions offered by each alternative. This study highlights operational conditions applicable to the US P&P sector, demonstrating the potential for significant carbon savings if these alternative fuels are adopted in US linerboard production. Implementing these best practices could result in substantial environmental and economic benefits, aligning the US industry with global sustainability standards.

MATERIALS AND METHODS

Definition of the Baseline

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.

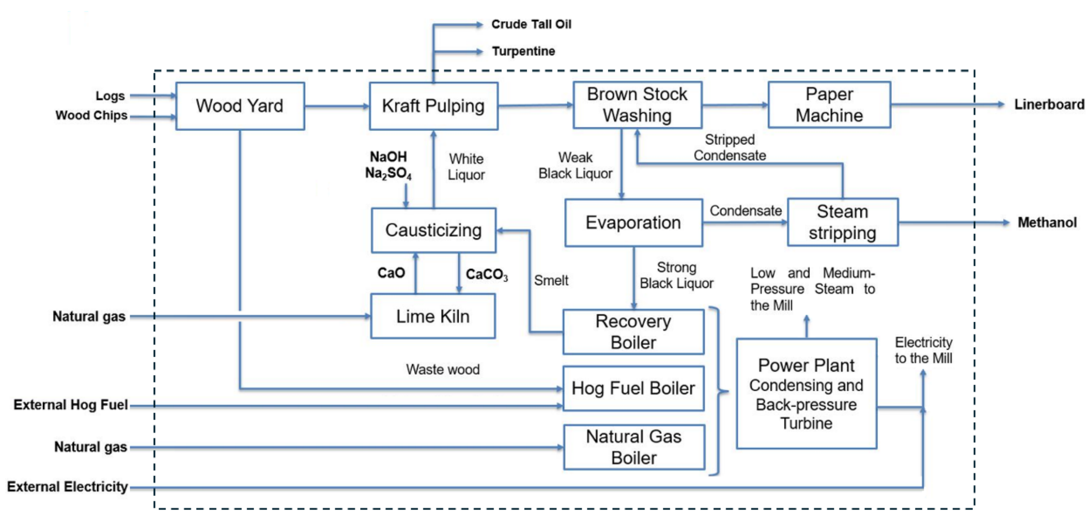

Fig. 1. System boundary for the Linerboard mill (base case)

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 CO_2 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 CO_2-eq emissions in the production of linerboard for the baseline case. A total of 69% of the total emissions correspond to biogenic CO_2; 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 CO_2 from burning natural gas and biogenic CO_2 from the $CaCO_3$ conversion to CaO and CO_2. The biogenic CO_2 from $CaCO_3$ originates from Na_2CO_3 from the black liquor burnt in the recovery boiler. In this case, the ratio between the fossil and the biogenic CO_2 in the lime kiln is 66% biogenic to 34% fossil CO_2.

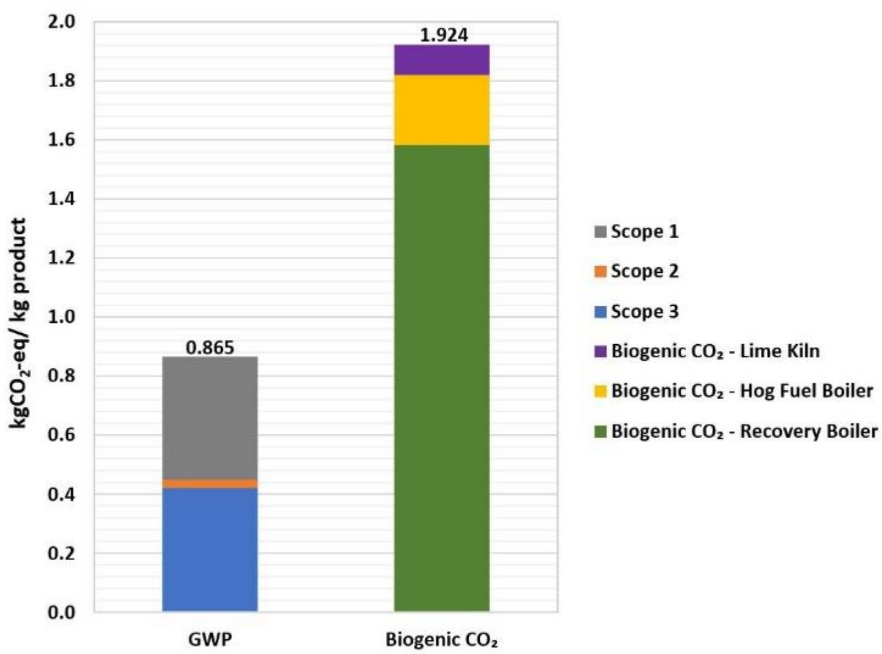

Fig. 2. CO_2-eq emissions in the production of one machine dry (10% moisture) kg of linerboard product

Regarding the GWP, the linerboard production has a total emission of 0.865 kg CO_2-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 CO_2-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 CO_2-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_2SO_4 makeup	$9.39*10^{-04}$	0.11%
		CaO	$2.33*10^{-02}$	2.69%
	Tall oil production	H_2SO_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 CO_2 from natural gas combustion for steam and electricity generation in the mill; whereas 6.2% comes from fossil CO_2 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 CO_2-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 CO_2-eq/kg of product for bleached market pulp (Tomberlin *et al.* 2020), 0.297 kg CO_{2eq}/kg of product for bleached softwood fluff pulp (Buitrago-Tello *et al.* 2022), and 0.552 kg CO_2-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 CO_2 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 CO_2/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 that 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,

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

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 CO_2 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 Na_2SO_4/ton reduction) makeup. However, the indirect emissions associated with sodium hydroxide are higher compared to sodium sulfate (1.4 kg CO_2-eq/kg NaOH *vs* 0.17 kg CO_2-eq/ kg Na_2SO_4). This results in increased indirect emissions from the pulping chemicals. Moreover, the Lignoboost process requires CO_2 (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 CO_2-eq, and the potential CO_2-eq reduction by implementing each technology for the established mill´s production.

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

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 CO_2-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 CO_2-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 CO_2-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 CO_2-eq (Bloomberg Finance 2022)

Under this approach considering only scope 1 and 2, some lime kiln fuels have a CO_2-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).

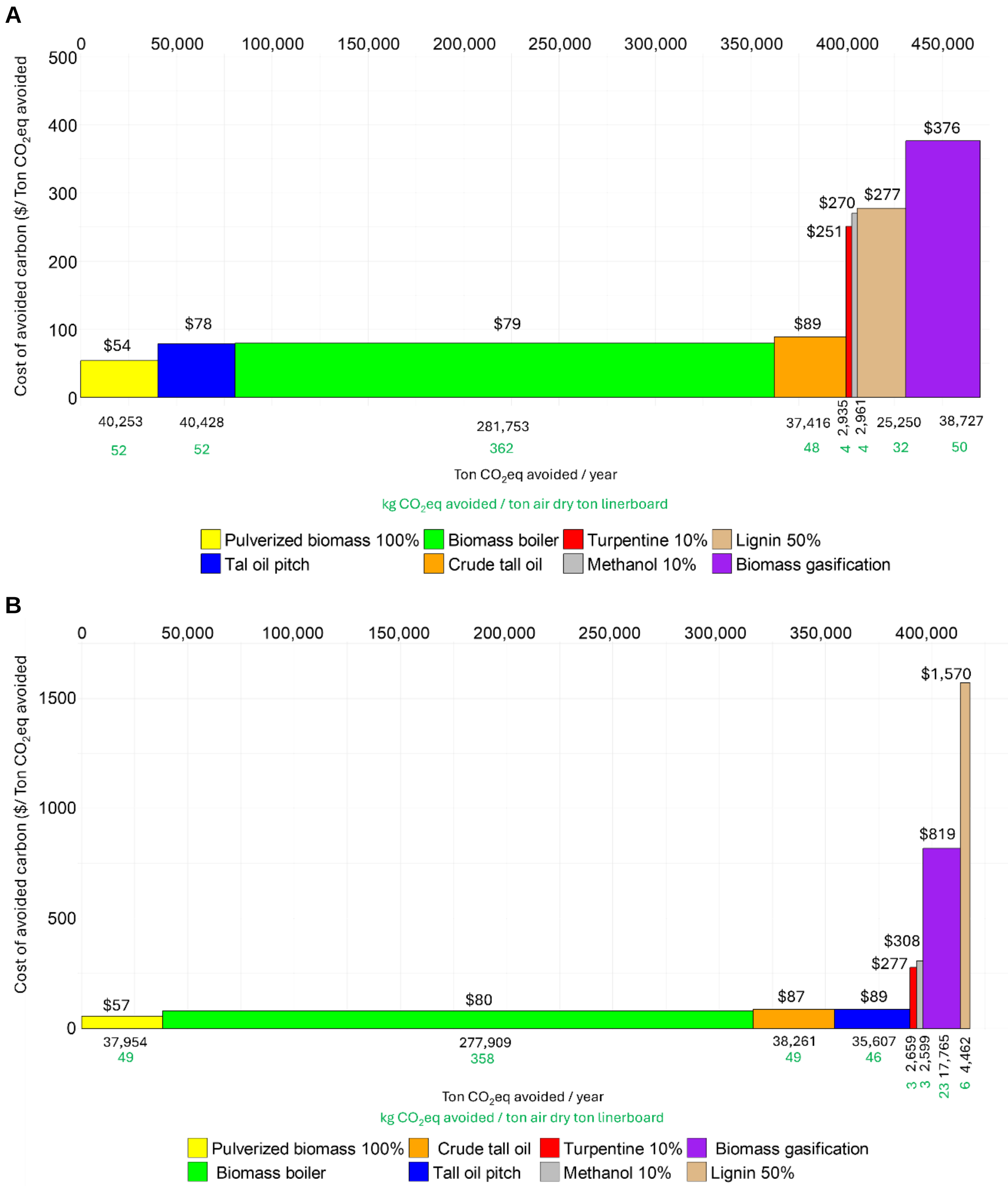

Fig. 3. Marginal abatement cost curve for alternatives to reduce the GHG emissions in the production of linerboard: a) CO_2 avoided based on scope 1 and 2, b) CO_2 avoided based on scope 1, 2, and 3. The production rate for the mill is 2,177 tons per day.

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 CO_2 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 CO_2-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 CO_2 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 CO_2-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 CO_2-eq avoided, a price expected by 2030 under the current conditions of the market, and a price of $47 per ton of CO_2-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 CO_2-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 CO_2-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 CO_2-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 CO_2-eq are emitted per ton of product, from which 1,924 kg corresponds to biogenic CO_2 (69%), and 854 kg (31%) corresponds to fossil CO_2-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 CO_2-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 CO_2 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 CO_2-eq avoided. The CAC can be further reduced if the mill gets a revenue from the CO_2 avoided. For instance, assuming a selling price of $11 and $47 per ton of CO_2-eq avoided, the total CAC is $69 and $48/ton of CO_2-eq avoided, respectively.

ACKNOWLEDGMENTS

The authors thank the Fulbright Program and the Colombian Institute of Educational Credit and Technical Studies Abroad (ICETEX) for providing the funding for Buitrago-Tello's doctoral studies under the "Fulbright-Pasaporte a la Ciencia" fellowship program. Support by the Forest Biomaterials of NCSU is also appreciated.

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Article submitted: November 29, 2023; Peer review completed: July 10, 2024; Revised version received and accepted: August 20, 2024; Published: August 30, 2024.
DOI: 10.15376/biores.19.4.7806-7823