



Life cycle assessment of Brazilian bleached eucalyptus kraft pulp: Integrating bleaching processes and biogenic carbon impacts

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ABSTRACT

Bleached eucalyptus kraft (BEK) pulp dominates global pulp production, yet the environmental impacts of its bleaching sequences in Brazil are not fully explored. Addressing this gap, we conducted a comparative life cycle assessment (LCA) of three bleaching sequences: conventional elemental chlorine-free (ECF), ECF with oxygen delignification, and ECF with oxygen delignification plus acid washing. We estimated the average global warming potential (GWP) for BEK delivered to the U.S. and examined how forest carbon cycle (FCC) elements, specifically biogenic GWP (GWPbio) and potential soil organic carbon (SOC) sequestration, influence GWP outcomes. Results show that the ECF sequence with oxygen delignification and acid washing reduces GWP by 11% and outperforms conventional ECF in 10 out of 11 environmental impact categories. The average GWP for Brazilian BEK delivered to the U.S. is 576 kg CO₂-eq/ton. Sensitivity analyses demonstrate that adding GWPbio increases GWP by 18%, whereas accounting for potential SOC sequestration reduces it by 39%. These findings highlight the necessity of optimizing bleaching processes and developing a standardized BEK LCA model for comparing the environmental impact of different fibers. This work sets a precedent for integrating FCC elements into LCAs and underscores the potential of SOC sequestration in mitigating climate change impacts.

1. Introduction

Bleached eucalyptus kraft (BEK) market pulp is in high demand due to its ability to produce the softest premium tissue products. Its short fibers, low coarseness, and minimal fines content create a flexible fiber web with a smooth surface, enhancing the tactile experience for consumers (De Assis et al., 2019). To achieve a balance of softness and strength in these premium tissues, BEK pulp is often mixed with northern bleached softwood kraft (NBSK) market pulp. NBSK's long fibers and intermediate coarseness contribute additional strength and durability to the final product (De Assis et al., 2019; FisherSolve and ResourceWise, 2024). Reflecting their significant roles in the global pulp and paper industry, Brazil accounts for approximately 60% of worldwide BEK pulp output, while Canada contributes about 33% of global NBSK market pulp production, according to the production database FisherSolve (FisherSolve and ResourceWise, 2024). To address the environmental concerns associated with BEK production, various life

cycle assessments (LCAs) have been conducted worldwide to evaluate its environmental impacts, focusing on different stages of the pulp-making process. Ryynanen and Nelson (RYYNANEN and NELSON, 1996) compared the bleaching sequences for BEK in an Australian Greenfield context, specifically examining elemental chlorine-free (ECF) and totally chlorine-free (TCF) processes. ECF bleaching involves using chlorine-based chemicals, while TCF bleaching avoids chlorine, utilizing alternative chemicals to achieve the desired pulp brightness. The authors highlighted the conversion process as the most significant environmental hotspot, with only minor differences between the ECF and TCF cases. In Chile, Gonzalez et al. (1998) compared the environmental impacts of BEK pulp derived from eucalyptus and pine pulp derived from pinewood using an ECF bleaching sequence, finding that eucalyptus pulp had a lower environmental impact, with the production stage contributing most to environmental burdens. In a follow-up study, Gonzalez et al. (2011) reported that technological improvements reduced the environmental impacts of water consumption, pollutant

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discharges, and air emissions in Chile. In Portugal, [Lopes et al. \(2003\)](#) assessed the environmental performance of printing and writing paper production, starting at the pulp production stage using an ECF sequence with oxygen delignification. Oxygen delignification is a pre-bleaching process that uses oxygen to remove lignin from the pulp, enhancing the efficiency of subsequent bleaching stages. The authors concluded that the production process was critical contributor to the environmental impacts, and that shifting from heavy fuel oil to natural gas generated improvements. In Thailand, [Jawjit et al. \(2006\)](#) identified pulp production, particularly the chemical recovery unit, as the most significant contributor to environmental impacts in the eucalyptus kraft pulp industry. The authors also noted the limitations of LCA methods in handling carbon sequestration in biobased products. In Spain, [Gonzalez-Garcia et al. \(González-García et al., 2009\)](#) performed an LCA of a TCF eucalyptus pulp mill, reporting that chemical production and on-site energy generation in the cooking and bleaching stages were the most significant contributors to environmental impacts. In China, [Xu and Becker \(2012\)](#) evaluated BEK production using an ECF sequence with oxygen delignification, and identified upstream processes, such as raw material and energy provision, as the most significant contributors to the total environmental burdens. The authors recommended incorporating biogenic global warming potential (GWPbio) in future assessments to account for the climate impacts of biomass. In Brazil, [Jour et al. \(2013\)](#) compared the carbon footprint of ECF with ECF ozone bleaching sequences, which incorporate an additional ozone bleaching stage to enhance pulp brightness. They found that the bleaching stage contributed 34%–41% of the total environmental impact and that ECF ozone bleaching sequences had the highest carbon footprint among the bleaching processes evaluated. More recently, [Brito et al. \(2023\)](#) evaluated the carbon footprint of premium tissue products in the U.S. This study presented results related to BEK production in Brazil and concluded that direct emissions from the pulp conversion process were the primary environmental contributors.

Despite significant progress in global LCAs, the environmental trade-offs between different bleaching sequences in Brazilian BEK production remain insufficiently explored ([Jour et al., 2013](#); [Brito et al., 2023](#)). More recent LCAs in Brazil have focused on individual technologies or production stages without comprehensively comparing the various bleaching sequences currently in use. Additionally, no effort has been made to establish a generic baseline of life cycle inventory (LCI) data and corresponding environmental impacts for BEK pulp production, referred to as the ‘average BEK model’, which is crucial for comparing the environmental impacts of conventional and alternative fibers in tissue production. Furthermore, existing studies often overlook vital factors like GWPbio and soil organic carbon (SOC) sequestration, which could significantly mitigate environmental impacts and are vital components of the forest carbon cycle (FCC). Therefore, a comprehensive comparison of bleaching sequences is essential for guiding more sustainable practices in BEK production. Establishing an average BEK model would provide a valuable reference for future comparisons of alternatives fiber sources. Incorporating recent methodologies like GWPbio ([Cherubini et al., 2011, 2013](#); [Guest et al., 2013](#)) and SOC sequestration potential ([Forfora et al., 2024](#)) will offer a more detailed understanding of the effects of the inclusion of FCC in LCAs ([Helin et al., 2013](#)).

To address these gaps, the primary objective of this study is to perform a comparative LCA of three bleaching sequences currently used in Brazilian BEK production. The first sequence, D₀-E_{op}-D₁-P (the base case), comprises D₀ (chlorine dioxide delignification), E_{op} (extraction enriched with oxygen and peroxide), D₁ (first brightening with chlorine dioxide), and P (peroxide bleaching). The second sequence, O/O-D₀-E_{op}-D₁-P, incorporates an additional O/O stage involving two-stage oxygen delignification before proceeding with the standard D₀-E_{op}-D₁-P process. The third sequence, O/O-A-D₀-E_{op}-D₁-P, adds an A stage, representing acid washing, to the O/O-D₀-E_{op}-D₁-P sequence. Secondary objectives of this work include developing an average BEK model and exploring

variability in global warming potential (GWP) results through sensitivity analyses incorporating GWPbio and SOC sequestration, providing deeper insights into the FCC's role in BEK production.

This article includes a methodology section, describing the LCA framework and detailed process operations for the current Brazilian BEK production with the prevalent bleaching sequences D₀-E_{op}-D₁-P, O/O-D₀-E_{op}-D₁-P, and O/O-A-D₀-E_{op}-D₁-P. The methodologies for incorporating GWPbio and SOC sequestration potential are also explained. Then, the results and discussion section presents a comparative LCA of the current bleaching sequences, the average LCA for BEK, and a sensitivity analysis of GWPbio and SOC factors. The limitations of these methodologies are also discussed. Finally, the article concludes with recommendations for improving practices in the pulp production industry to foster more sustainable operations.

2. Methods

This section describes the methodology used to perform the comparative LCA of the three currently used Brazilian bleaching sequences: D₀-E_{op}-D₁-P, O/O-D₀-E_{op}-D₁-P, and O/O-A-D₀-E_{op}-D₁-P. The section is organized into three parts. First, the LCA framework is outlined, covering the goal, scope, and system boundaries, including the LCI for eucalyptus cultivation, transportation to the U.S., kraft pulping, bleaching, chlorine dioxide generation technologies, and the selected life cycle impact assessment (LCIA) method. Next, the methodologies for the GWPbio and SOC sequestration potential are detailed. These GWPbio and SOC methodologies are explored as part of a sensitivity analysis to assess components of the FCC of eucalyptus plantations and to provide a more comprehensive environmental assessment.

2.1. LCA framework

The LCA of BEK pulp was conducted following ISO 14040-44 standards ([Finkbeiner et al., 2006](#)), covering four components: goal and scope definition, LCI, LCIA, and interpretation of results. The primary objective of this study is to assess and compare the environmental impacts of current bleaching sequences used in BEK production in Brazil. The declared unit is one air-dried ton (ADt, 10% moisture) of BEK pulp, with system boundaries extending from cradle to tissue mill gate, including transportation to the U.S. market. The inclusion of transportation to the U.S. reflects the primary end-use market for Brazilian BEK pulp, mainly for tissue production. Since the BEK pulp production process does not generate any co-products, the system is not multi-functional. Therefore, all environmental burdens are attributed solely to the production of BEK pulp. The first phase of the LCI considers eucalyptus cultivation, based on the work of [Forfora et al. \(2024\)](#). Eucalyptus plantations operate on a 7-year rotation cycle, yielding a biomass productivity of 15.1 oven-dried tons (ODT) per hectare per year. Throughout the cultivation process, fertilizers supply essential nutrients to the soil, while herbicides and insecticides are used to manage weeds and pests. Diesel fuel powers the machinery for planting, maintenance, and harvesting activities. After harvesting, the eucalyptus logs are debarked on-site and prepared for transport. With a moisture content of 40%, the logs are transported over an average distance of 61.2 km to the pulp mill using a standard 22-ton capacity truck ([Vivas et al., 2024](#)). Detailed inputs and outputs involved in the cultivation of eucalyptus are provided in [Table S1](#) of the supplementary material.

After harvesting and initial transportation, the eucalyptus is processed in pulp mills distributed across Brazil. Using *FisherSolve* ([FisherSolve and ResourceWise, 2024](#)), 14 Brazilian mills producing BEK pulp were identified. Primary mill data is collected by the *FisherSolve* research team, including production, paper grades, main equipment dimensions and age, fuels used, and whether the mill purchases or does not purchase electricity, among other details. With this information, and *FisherSolve* proprietary models, a mass and energy balance is performed for each mill. Each mill is optimized for the lowest

production cost, using researched prices for raw materials and energy based on the mill's location.

Fig. 1a and b show the production and geographic distribution of the 14 mills. These mills are located across various regions, and their proximity to ports is essential for efficiently transporting pulp to export markets. The mills employ an intermodal transportation system, combining rail and truck transport to move the BEK pulp to export ports. The primary export destination is Savannah, Georgia, a key location for bath tissue production in the U.S. The average transportation distances, methods, dimensions, capacities, and fuel economy are shown in Tables S2–S4. The logistics of transporting the pulp to the U.S., including internal transportation, are factored into the LCA to account for fuel use and emissions (Davis et al., 2017; ICF International, 2009).

The second part of the LCI focuses on the kraft pulping process, simulated using Valmet's WinGEMS software (Valmet WinGEMS simulation, 1990) (Fig. 2a). Wood chips are impregnated with an alkaline solution of sodium sulfate and sodium hydroxide (Na_2SO_4 and NaOH), or white liquor, in a continuous digester, operating at a pH of 11 and temperatures ranging from 130 °C to 160 °C for 1–1.5 h (Smook, 2016). Following digestion, the cooked chips are depressurized in the blow tank, releasing fibers. Fibers that remain intact (knots) are recirculated back to the digester. The pulp is then processed through screens, presses, and washers to remove residual chemicals and maximize the recovery of spent chemicals, minimizing the solids in mill effluents (Smook, 2016).

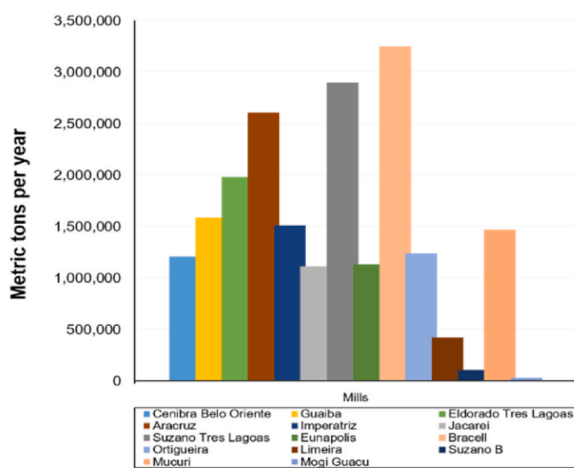
The separated lignin and chemicals form a black liquor solution, concentrated from 19% to 72% solids and combusted in the recovery boiler. This process generates heat and power, reduces sulfur compounds, and recovers chemicals for reuse in the pulping process (Smook, 2016). Green liquor, formed from the inorganic chemicals, is reacted with lime to regenerate NaOH in the causticizing plant, while lime mud is removed and white liquor is recirculated (Smook, 2016). After pulping, the cellulosic fibers contain residual lignin, which is removed through bleaching. This study considers three bleaching sequences, shown in Fig. 2b (FisherSolve and ResourceWise, 2024): 1) $\text{D}_0\text{-E}_{\text{op}}\text{-D}_1\text{-P}$, used in 9.8% of production, 2) $\text{O/O-D}_0\text{-E}_{\text{op}}\text{-D}_1\text{-P}$ applied in 34.1% of production, and 3) $\text{O/O-A-D}_0\text{-E}_{\text{op}}\text{-D}_1\text{-P}$, the most widely used sequence, accounting for 56.1%. These sequences have average technological ages of 24, 15, and 12 years, respectively for the Brazilian mills analyzed. They primarily differ in their initial delignification stages. The first sequence (D_0) uses chlorine dioxide (ClO_2) for lignin removal, while the second and third incorporate two-stage oxygen delignification, with the third adding an acid wash (A) to target hexenuronic acids (Smook,

2016).

Table 1 summarizes the average operational conditions for Brazilian BEK pulp mills, which are essential for understanding the environmental impacts of different bleaching sequences. These operational conditions, such as digester temperature, sulfidity, and energy self-sufficiency, directly affect the efficiency of the pulping and bleaching processes. For example, higher sulfidity improves the effectiveness of the chemical recovery process, and energy self-sufficiency highlights the mills' ability to generate sufficient power through combustion processes (e.g., using black liquor and wood waste in recovery boilers). The total energy required from combustion reflects the energy used for these processes related to steam and power generation. These values were estimated based on process simulations, which account for the fuel composition of the power boilers (wood-waste, gas, coal, fuel oil) described in Table 1. Table S5 shows the specifications required for process simulations.

In BEK pulp production, ClO_2 is a critical chemical used in the bleaching process to remove residual lignin from the pulp, enhancing pulp brightness and quality (Smook, 2016). Brazilian mills utilize three generations of ClO_2 production technologies: R-6, R-8, and R-10 (FisherSolve and ResourceWise, 2024). These technologies represent different levels of efficiency, environmental performance, and operational age. The R-6 process is the oldest generation of ClO_2 production, characterized by lower efficiency, leading to higher energy consumption and emissions. The R-8 process is an intermediate technology offering improved chemical and energy efficiency compared to R-6. The R-10 process is the latest and most efficient generation, with reduced energy and chemical consumption (Suhr et al., 2015; Deshwal and Lee, 2005). Each of these technologies significantly impacts the environmental performance of the bleaching sequences, with the more advanced R-10 stage resulting in lower energy consumption (Suhr et al., 2015) and reduced chemical waste compared to the older R-6 and R-8 technologies (Deshwal and Lee, 2005).

In our study, the efficiencies of the ClO_2 technology stages were modeled using production-weighted averages based on their prevalence in Brazilian mills: R-6 (18.4%), R-8 (43.3%), and R-10 (38.3%). The stoichiometry for the R-6 technology was obtained from Suhr et al. (2015), while the R-8 and R-10 technologies were modeled using WinGEMS software (Valmet WinGEMS simulation, 1990), with stoichiometry based on Deshwal et al. (Deshwal and Lee, 2005). For the R-8 and R-10 technologies, specific chemicals (e.g., sodium chlorate, sulfuric acid) and their consumption rates were incorporated according to each stage's stoichiometric relationships and process requirements. Fig. 3



a. Distribution of BEK market pulp production in Brazil



b. Geographic distribution of BEK market pulp production in Brazil and its nearby ports

Fig. 1. (a) Distribution of BEK market pulp production in Brazil. (b) Geographic distribution of BEK market pulp production in Brazil and its nearby ports (the number between parentheses represents the number of pulp mills in each state) (FisherSolve and ResourceWise, 2024).

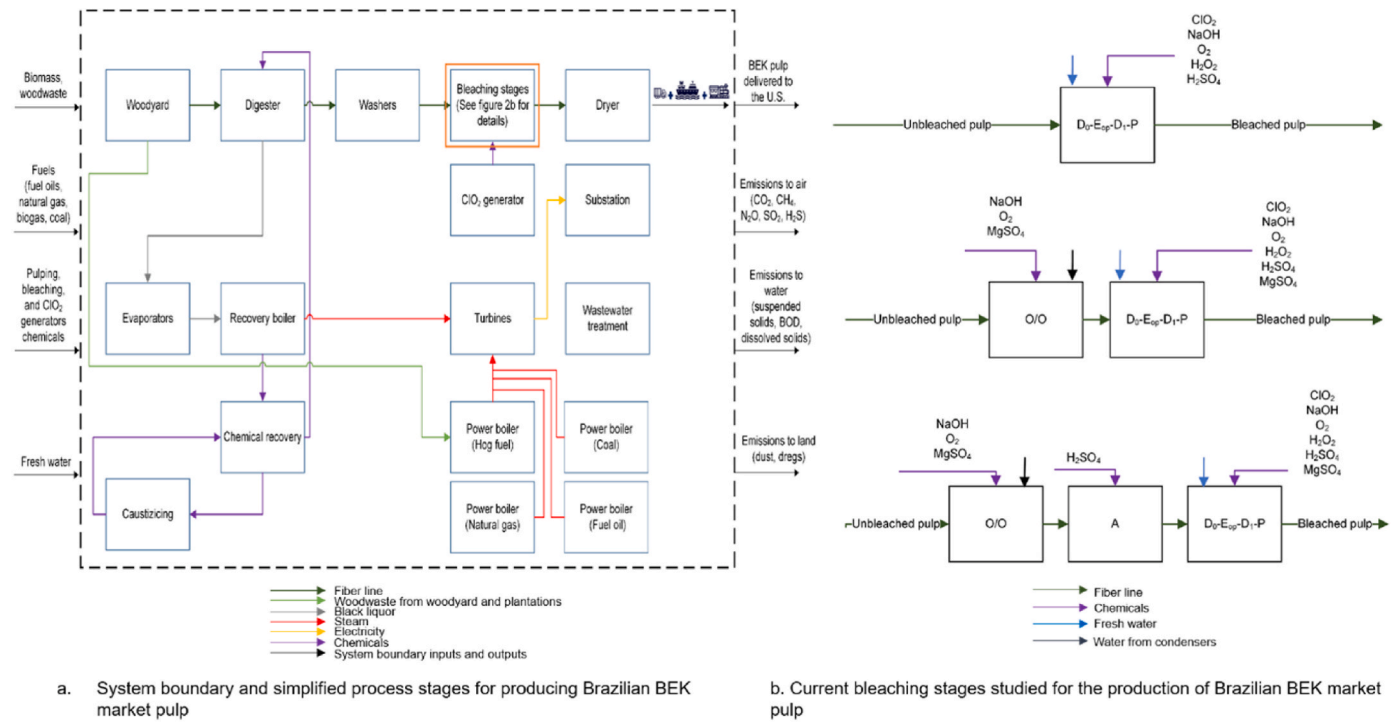


Fig. 2. (a) System boundary and simplified process stages for producing Brazilian BEK market pulp delivered to the U.S. (b) Current bleaching stages studied for the production of Brazilian BEK market pulp (FisherSolve and ResourceWise, 2024).

Table 1

Average operation conditions for Brazilian BEK pulp mills based on production-weighted averages (FisherSolve and ResourceWise, 2024).

Brazilian kraft pulp mills' average conditions	Bleaching sequences			
	D ₀ -E _{op} -D ₁ -P		O/O-D ₀ -E _{op} -D ₁ -P	
Digester temperature (°C)	160		160	
Sulfidity (%)	30		30	
Effective alkali charge (% Na ₂ O)	13.4		13.4	
Mill's power self-sufficiency (%)	100		100	
Digester yield (%)	50.5		50.5	
Pressure, recovery boiler (barg)	80.6		80.6	
Temperature, recovery boiler (°C)	481.1		481.1	
Pressure, power boiler (barg)	68.9		68.9	
Temperature, power boiler (°C)	465.9		465.9	
Fuel composition, power boiler (%)	Wood waste	83.4%	Wood waste	83.4%
	Gas	5.8%	Gas	5.8%
	Coal	7.9%	Coal	7.9%
	Number 6 oil	2.9%	Number 6 oil	2.9%
Total energy required for steam and power generation (GJ)	1486		1261	
Fuel composition, limekiln (%)	Gas	24.7%	Gas	24.7%
	Biogas	12.5%	Biogas	12.5%
	Number 6 oil	52.6%	Number 6 oil	52.6%
	Number 2 oil	10.2%	Number 2 oil	10.2%
Bleaching charges (% on OD pulp)	ClO ₂	2.3%	ClO ₂	1.3%
	NaOH	1.5%	NaOH	3.4%
	O ₂	0.5%	O ₂	3.5%
	H ₂ O ₂	0.7%	H ₂ O ₂	0.7%
	H ₂ SO ₄	0.3%	H ₂ SO ₄	0.1%
	MgSO ₄	0%	MgSO ₄	0.03%
Bleaching yield (%)	96.1		93.1	
Overall yield (%)	45.1		43.8	
ClO ₂ generators technology	R-6, R-8, R-10		R-6, R-8, R-10	
ClO ₂ production (kg/ADt)	21.1		12.4	
Mill power consumption (kWh/ADt)	974.2		986.9	

illustrates a simplified representation of the ClO₂ generation technologies considered in the LCA of Brazilian BEK pulp production.

The total power consumption is estimated using the values reported by Suhr et al. (2015), Echeverria et al. (2022), and Buitrago-Tello et al. (2022). The detailed power consumption for each bleaching sequence

can be found in Table S6 of the supplementary material.

The LCI shown in Table 2 provides a detailed breakdown of the inputs and outputs associated with the production of one ADt of BEK pulp for each of the three bleaching sequences studied: D₀-E_{op}-D₁-P (base case), O/O-D₀-E_{op}-D₁-P, and O/O-A-D₀-E_{op}-D₁-P.

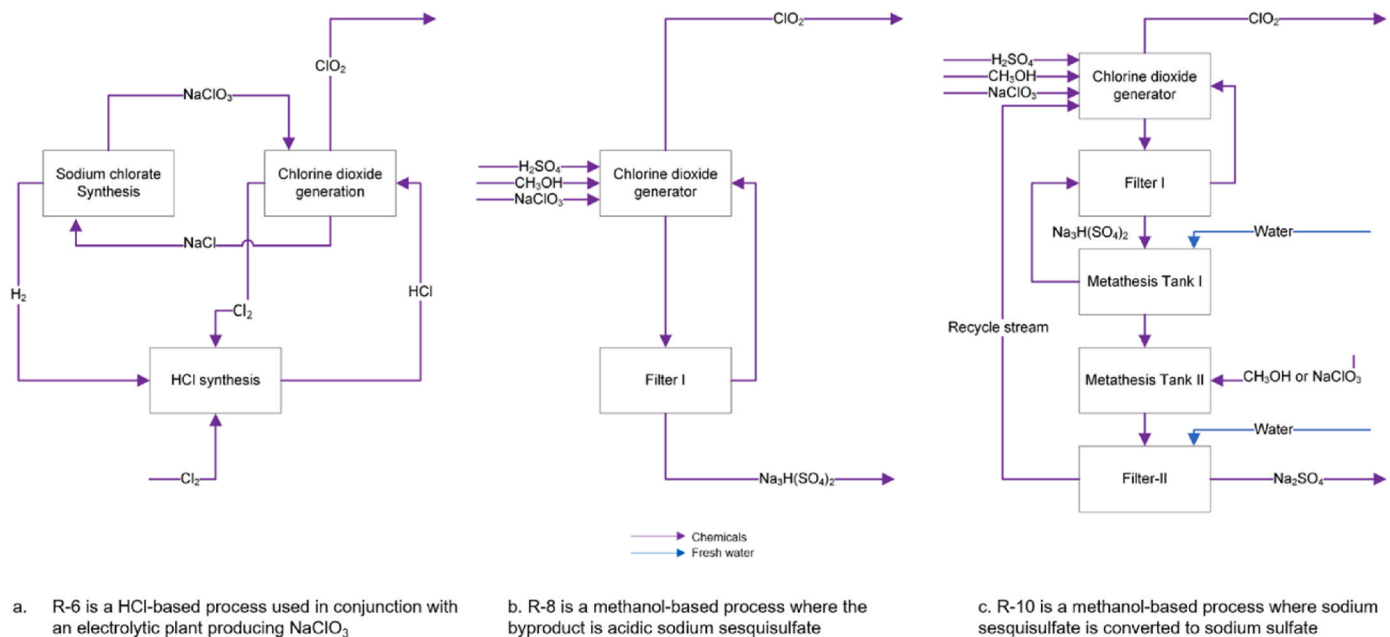


Fig. 3. ClO_2 generation technologies for the LCA of the Brazilian BEK pulp (a) R-6 process (b) R-8 process, and (c) R-10 process (Deshwal and Lee, 2005).

The LCIA was conducted using openLCA software (Ciroth, 2007), the ecoinvent 3.8 database (Wernet et al., 2016), and the Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) 2.1 methodology to assess ten environmental impact categories: GWP, acidification, eutrophication, ozone depletion, photochemical smog formation, ecotoxicity, human health cancer effects, human health non-cancer effects, respiratory effects, and fossil fuel depletion (Bare et al., 2012). Additionally, water consumption was included as a separate environmental indicator, although it is not part of the TRACI methodology. Water consumption data for the different bleaching sequences was gathered from mass balance process simulations and included in the analysis to provide a more comprehensive assessment of resource use. Emissions from transportation were calculated using the Environmental Protection Agency (EPA) factors (EPA, 2023a). Furthermore, the choice of TRACI 2.1 is justified by studies such as Cherubini et al. (2018), which demonstrate that while absolute LCIA values may vary across methods, scenario rankings generally remain consistent.

2.2. Biogenic global warming potential

The warming effects of CO_2 emissions significantly influence Earth's climate (Solomon et al., 2009). Solomon et al. (2009) highlight that increased atmospheric CO_2 concentrations result in climate changes that remain largely irreversible for at least 1000 years after emissions stop. This persistence is due to the slow atmospheric removal of CO_2 and the gradual heat loss to the ocean. Therefore, immediate and substantial reductions in greenhouse gas emissions are urgently needed to mitigate these long-term impacts. Incorporating the FCC into LCA's of eucalyptus plantations in Brazil is critical for understanding the overall carbon footprint. This approach allows for selecting feedstock and conversion technology combinations that reduce the warming effects of CO_2 emissions from forest products.

Helin et al. (2013) reviewed various methods for including the FCC in carbon footprint assessments. They suggested that when carbon dioxide sequestration lags behind carbon dioxide emissions from biomass, it causes a warming effect. The GWPbio factor measures the warming effects of the delay between carbon dioxide emission and sequestration. Moreover, the authors recommended using this indicator as it measures cumulative radiative forcing relative to a pulse emission of one mass unit

of fossil CO_2 , making the results easy to understand for a broad audience (fossil CO_2 equivalent).

According to Cherubini et al. (2011), the GWPbio represents the decay of CO_2 emitted from biomass as a function of the rotation period (delay between emission and sequestration) divided by the atmospheric decay of anthropogenic CO_2 emissions. The GWPbio can be described under equation (1).

$$GWP_{bio} = \frac{AGWP_{bio}}{AGWP_{CO_2}} = \frac{\int_0^{TH} \alpha_{CO_2} f(t) dt}{\int_0^{TH} \alpha_{CO_2} y(t) dt} \quad (1)$$

Where.

$f(t)$: Atmospheric decay of a pulse of biogenic CO_2 over time (kg CO_2/yr).

$y(t)$: Atmospheric decay of a pulse of anthropogenic CO_2 over time (kg CO_2/yr).

α_{CO_2} : Radiative efficiency of the gas of CO_2 (dimensionless).

TH: Time horizon (100 years for this study).

The GWPbio factor remains constant whether evaluated at the level of a single stand or across an entire landscape (Cherubini et al., 2013). Previous studies have demonstrated that the integrated impact of a pulse emission is equal to the instantaneous impact of continuous emissions (Peters et al., 2011; Aamaas et al., 2012; Azar and Johansson, 2012).

Furthermore, the GWPbio factor developed by Cherubini et al. (2011) did not account for when products store biogenic carbon. Therefore, in subsequent work, Guest et al. (2013) improved the atmospheric decay function of a pulse of biogenic CO_2 $f(t)$ by considering the storage time of the product and its end-of-life phase:

$$f(t) = \begin{cases} f_1(t) = -\int_0^t g(t')y(t-t')dt', & \text{for } 0 \leq t < \tau \\ f_2(t) = y(t-\tau) - \int_\tau^t g(t')y(t-t')dt', & \text{for } t \geq \tau \end{cases}$$

Where.

Table 2

LCI for the production of Brazilian BEK with the current bleaching sequences studied.

Functional Unit: 1 ADt of BEK market pulp (sold at 90% consistency)				
	Unit/ ADt	BEK (D ₀ -E _{op} - D ₁ -P)	BEK (O/O-D ₀ - E _{op} -D ₁ -P)	BEK (O/O-A- D ₀ -E _{op} -D ₁ -P)
Inputs				
Biomass	kg	2.06x10 ³	2.13x10 ³	2.13x10 ³
Woodwaste	kg	1.98x10 ²	1.49x10 ²	1.52x10 ²
NaOH (chemical recovery)	kg	0.53x10 ¹	0.76x10 ¹	0.72x10 ¹
NaOH (bleaching)	kg	2.47 x10 ¹	1.45x10 ¹	1.02x10 ¹
H ₂ O ₂	kg	6.40x10 ⁰	6.40x10 ⁰	6.40x10 ⁰
CaO	kg	1.35x10 ¹	1.46x10 ¹	1.46x10 ¹
NaClO ₃	kg	2.81x10 ¹	1.66x10 ¹	1.32x10 ¹
Cl ₂	kg	2.10x10 ⁰	1.20x10 ⁰	9.70x10 ⁻¹
CH ₃ OH	kg	2.90x10 ⁰	1.70x10 ⁰	1.40x10 ⁰
O ₂	kg	4.70x10 ⁰	3.35x10 ¹	3.35x10 ¹
H ₂ SO ₄	kg	1.76 x10 ¹	1.15x10 ¹	8.20x10 ⁰
MgSO ₄	kg	–	1.81x10 ⁰	1.81x10 ⁰
Natural gas	m ³	3.55x10 ¹	3.23x10 ¹	3.25x10 ¹
Biogas	m ³	4.00x10 ⁰	4.50x10 ⁰	4.50x10 ⁰
Fuel oil number 6	kg	2.13x10 ¹	2.13x10 ¹	2.14x10 ¹
Fuel oil number 2	kg	2.10x10 ⁰	2.30x10 ⁰	2.30x10 ⁰
Coal	kg	2.73x10 ¹	2.32x10 ¹	2.34x10 ¹
Freshwater	m ³	2.89x10 ¹	2.71x10 ¹	3.09x10 ¹
Outputs				
Pulp	ADt	1	1	1
CO ₂ biogenic, recovery boiler	kg	1.86x10 ³	1.97x10 ³	1.97x10 ³
CO ₂ biogenic, lime kiln	kg	1.05x10 ²	1.18x10 ²	1.18x10 ²
CO ₂ biogenic, hog fuel	kg	5.36x10 ²	4.55x10 ²	4.60x10 ²
CO ₂ fossil, power boiler	kg	1.58x10 ²	1.34x10 ²	1.36x10 ²
CO ₂ fossil, lime kiln	kg	6.87x10 ¹	7.69x10 ¹	7.70x10 ¹
CO ₂ fossil, R8, R10	kg	7.00x10 ⁻¹	4.00x10 ⁻¹	3.00x10 ⁻¹
SO ₂	kg	1.3x10 ⁰	1.1x10 ⁰	1.1x10 ⁰
H ₂ S	kg	1.20x10 ⁻¹	1.20x10 ⁻¹	1.30x10 ⁻¹
CH ₄	g	9.79x10 ¹	9.34x10 ¹	9.40x10 ¹
N ₂ O	g	3.25x10 ¹	2.99x10 ¹	3.01x10 ¹
BOD	kg	5.04x10 ¹	5.22x10 ¹	5.29x10 ¹
AOX	kg	8.50x10 ⁻¹	5.00x10 ⁻¹	4.00x10 ⁻¹
Dregs and grits	kg	6.6x10 ⁰	7.2x10 ⁰	7.2x10 ⁰
Mud inert	kg	2.00x10 ¹	2.17x10 ¹	2.17x10 ¹
Dust losses	kg	3.40x10 ⁻¹	4.00x10 ⁻¹	4.00x10 ⁻¹
Water effluent	m ³	2.75x10 ¹	2.52x10 ¹	2.87x10 ¹

$g(t)$: is the biomass growth model as a function of the rotation period (kg CO₂/yr).

τ : refers to the lifespan of the bio-based product (yrs.).

In this study, market pulp is intended for the U.S. market, where it will be used to produce consumer bath tissue (CBT). The assumption of a one-year end-of-life emission period is based on typical supply chain delays, usage, and the standard practice of applying wastewater sludge decomposition on land (EPA, 2023b). All emissions at the end of life are assumed to be CO₂, reflecting the most significant impact on GWP in this context. Under Brazil's current operations of eucalyptus pulp mills, the biomass rotation period is seven years, and the GWPbio factor used is 0.022, following the Guest et al. (2013) methodology.

2.3. Soil organic carbon sequestration potential

Soil holds the most significant amount of terrestrial organic carbon in the biosphere, containing more carbon than plants and the atmosphere combined (Schlesinger, 1977). As previously stated by Helin et al. (2013), Forfora et al. (2024), and Lan et al. (2024), it is essential to

include the SOC sequestration potential of biomass when performing LCA studies. SOC sequestration potential involves capturing carbon dioxide from the atmosphere and storing it in the soil through plant roots and organic matter, transforming biomass systems into carbon sinks and significantly reducing their overall GWP. To address this, Forfora et al. (2024) developed a methodology to include the SOC sequestration potential into the LCA framework described by equations (2)–(4).

$$C_{R_PP} = AGB * RSR * X_C * X_{PP} \quad (2)$$

$$Total\ C_{input} = \sum_i^n C_{input_i} = \sum_i^n C_{RPP_i} * S_{R_i} + C_{EPP_i} * S_{E_i} \quad (3)$$

$$C_{input} = \frac{Total\ C_{input}}{Rotation\ time} \quad (4)$$

Where.

C_{R_PP} : Carbon in coarse roots (ton C/ha)

AGB : Aboveground biomass (ton C/ha)

RSR : Root-to-shoot ratio (dimensionless)

X_C : Carbon mass fraction (dimensionless)

X_{PP} : Allocation factor (dimensionless)

$Total\ C_{input}$: Total carbon input to soil (ton C/ha)

n : Rotation time

i : Iteration index

S_{R_i} : Fraction of the coarse roots that are returned to the soil = 1 (dimensionless)

C_{EPP_i} : Carbon associated with rhizodeposition of extra roots = 0.65*

C_{R_PP} (ton C/ha)

S_{E_i} : Fraction of the extra roots that are returned to the soil = 1 (dimensionless)

C_{input} : Total carbon input normalized by year (ton C/ha.yr)

Following the methods described by Forfora et al. (2024), the baseline scenario involves using degraded lands to cultivate eucalyptus, which have the potential to sequester carbon, given that they are not yet saturated. The root-to-shoot ratio of eucalyptus is 0.21 (Mokany et al., 2005), and the total carbon inputs to soil were determined to originate solely from the coarse roots and rhizodeposition (Bolinder et al., 2007). The coarse roots of eucalyptus depend on the root-to-shot ratio, whereas the rhizodeposition was calculated at 65% of coarse roots (Forfora et al., 2024). For the analysis, a critical assumption is made regarding the stabilization ratio of 15% (Berti et al., 2016). This ratio signifies the fraction of carbon derived from belowground biomass that persists stably, thereby enhancing long-term soil carbon storage.

Regarding land utilization, Forfora et al. (2024) indicate that eucalyptus requires 0.1 ha to produce 1 ODT of biomass per year, representing 1.75 tons CO₂eq sequestered per hectare per year. Considering a rotation period of seven years for eucalyptus plantations, the calculated carbon sequestration is 12.3 tons CO₂/ha, equivalent to 115.8 kg of CO₂ eq per ODT of biomass. It is essential to note that this factor indicates the ability of eucalyptus biomass to enhance SOC during its initial rotation; however, this effect may diminish in subsequent rotations, especially if the soil approaches saturation, making it primarily useful for comparison with other biomass types (Forfora et al., 2024). The GWP estimation for a time horizon of 100 years for eucalyptus biomass assumes the permanence of biomass roots in the soil, the absence of natural disasters (e.g., forest fires), and the persistence of this soil carbon over a century (Forfora et al., 2024).

3. Results and discussion

This section examines Brazilian BEK pulp production, focusing on the environmental impacts of different bleaching sequences. It presents a combined analysis of the LCI and LCA results for the D₀-E_{op}-D₁-P, O/O-

D₀-E_{op}-D₁-P, and O/O-A-D₀-E_{op}-D₁-P bleaching sequences, highlighting the GWP, acidification, eutrophication, ozone depletion, photochemical smog formation, ecotoxicity, human health cancer effects, human health non-cancer effects, respiratory effects, fossil fuel depletion and water consumption. A sensitivity analysis then incorporates the FCC into the LCA by accounting for the GWBio and SOC sequestration potential of eucalyptus plantations. Finally, a discussion of the methodological limitations is presented.

3.1. LCA findings

The LCI data in Table 2 reveal that while the biomass input requirements for the O/O-A-D₀-E_{op}-D₁-P and O/O-D₀-E_{op}-D₁-P sequences are slightly higher than the base case, these sequences significantly reduce chemical usage. For example, oxygen consumption in the delignification stages is markedly higher, allowing for a 42% reduction in NaOH usage and a 53% reduction in sodium chlorate (NaClO₃) in the O/O-A-D₀-E_{op}-D₁-P sequence. These improvements highlight the increased chemical efficiency of oxygen delignification and acid wash stages.

Incorporating oxygen delignification reduces halogenated organic compounds (AOX) emissions, a significant environmental benefit, as these compounds pose risks to aquatic ecosystems. Some of these compounds tend to bioaccumulate, while others are carcinogens and mutagens, potentially causing harm to aquatic ecosystems (Savant et al., 2006). The D₀-E_{op}-D₁-P sequence, with its reliance on higher ClO₂ usage, generates 0.85 kg/ADt of AOX, the highest of the three sequences. In contrast, the O/O-D₀-E_{op}-D₁-P sequence reduces AOX emissions by 41%, while the O/O-A-D₀-E_{op}-D₁-P sequence achieves a 53% reduction. This reduction is primarily due to the removal of lignin in the oxygen delignification stages, reducing the need for chlorine-based chemicals and, thus, the formation of chlorinated organic pollutants.

Direct CO₂ emissions from fossil fuel combustion are highest in the D₀-E_{op}-D₁-P sequence due to its greater steam demand and reliance on fossil fuels like natural gas and coal. This sequence's lack of oxygen delignification requires additional energy to heat fresh water, leading to elevated CO₂ emissions from the power boilers. In contrast, oxygen delignification sequences reduce direct CO₂ emissions by recirculating hot water, which lowers steam demand and fossil fuel use, resulting in a significant reduction in fossil CO₂ emissions.

The increased fossil fuel consumption in the lime kiln for the oxygen delignification sequences is directly linked to higher NaOH demand. The chemical recovery process produces more calcium carbonate (CaCO₃), which must be calcined in the lime kiln. This process requires additional energy, increasing fossil fuel consumption and CO₂ emissions. Thus, the energy-intensive calcination process represents a fundamental trade-off in sequences with oxygen delignification despite their reduction in direct CO₂ emissions from other sources.

A notable trade-off in the O/O-A-D₀-E_{op}-D₁-P sequence is its higher freshwater consumption, which increases by 7% compared to the base case. This increase is primarily driven by the hot acid washing stage, which requires additional water inputs.

Fig. 4 illustrates the GWP results for the three bleaching sequences studied—D₀-E_{op}-D₁-P, O/O-D₀-E_{op}-D₁-P, and O/O-A-D₀-E_{op}-D₁-P—as well as the average GWP for BEK pulp production in Brazil delivered to the southeastern United States.

Fig. 4 shows that the O/O-A-D₀-E_{op}-D₁-P sequence achieves the lowest GWP compared to the other sequences. This reduction in GWP is primarily due to improved energy efficiency and lower chemical consumption, enabled by oxygen delignification and an additional acid wash stage. These process improvements reduce the need for NaOH and ClO₂, significantly decreasing chemical production and recovery emissions. Conversely, the D₀-E_{op}-D₁-P sequence exhibits the highest GWP contribution, mainly due to the absence of oxygen delignification, necessitating more chemicals to break down lignin. The higher consumption of NaOH and ClO₂ results in greater energy demand for

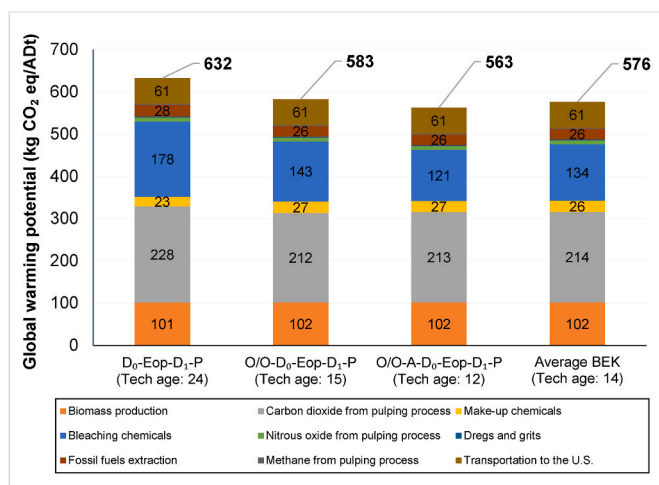


Fig. 4. GWP of Brazilian BEK market pulp delivered to the U.S. as a function of different bleaching sequences. Time horizon: 100 years. Tech age is the average age of the bleaching sequence technology.

chemical production and recovery, increasing overall emissions.

The O/O-D₀-E_{op}-D₁-P sequence offers a noticeable reduction in GWP. By incorporating oxygen delignification, this sequence removes a substantial portion of lignin earlier in the process, reducing the demand for harsh bleaching chemicals and lowering the associated emissions. The result significantly reduces bleaching-related GWP contributions compared to the base case.

Direct CO₂ emissions from fossil fuel combustion are also lowest in the O/O-A-D₀-E_{op}-D₁-P sequence. The combination of oxygen delignification and the acid wash stage reduces the steam demand, lowering the consumption of fossil fuels such as coal, natural gas, and fuel oil in the power boiler and lime kiln. This process efficiency minimizes direct CO₂ emissions across the bleaching sequences. In contrast, the D₀-E_{op}-D₁-P sequence has the highest direct CO₂ emissions, driven by higher steam demand and reliance on fossil fuels for energy generation. Without oxygen delignification, more energy is required to heat water and generate steam, leading to greater fossil fuel consumption.

The O/O-D₀-E_{op}-D₁-P sequence reduces direct CO₂ emissions from fossil fuel combustion by recirculating hot water from the delignification stage. This water recirculation reduces the need for additional heating and thus lowers fossil fuel consumption in the power boiler. Improved chemical recovery in this sequence also decreases energy demand in the lime kiln, contributing to lower direct CO₂ emissions.

The results shown in Fig. 4 also included an average BEK model that serves as a reference for future comparative assessments, particularly analyzing the environmental impacts of Brazilian BEK delivered to the U.S. for CBT production. This model represents a production-weighted average of the data collected from the three bleaching sequences used in Brazilian pulp mills: D₀-E_{op}-D₁-P (9.8%), O/O-D₀-E_{op}-D₁-P (34.1%), and O/O-A-D₀-E_{op}-D₁-P (56.1%).

Developing this average BEK model provides a valuable baseline for comparing the environmental impacts of different biomass types used in pulp production. By offering standardized LCI data for BEK pulp production (Table S7), the average BEK model can be employed to compare the environmental performance of various biomass sources for pulp production, including northern softwoods, non-wood fibers such as bamboo, agricultural residues like wheat straw, and dedicated energy crops like miscanthus and switchgrass. By revealing differences due to factors such as growth rates, harvesting practices, processing efficiencies, and process types, the model assists stakeholders in making informed decisions about raw material selection based on environmental considerations.

Fig. 5 comprehensively compares the environmental impact

categories for the three bleaching sequences—D₀-E_{op}-D₁-P, O/O-D₀-E_{op}-D₁-P, and O/O-A-D₀-E_{op}-D₁-P—along with the average BEK model.

The O/O-A-D₀-E_{op}-D₁-P sequence demonstrates the best overall environmental performance, showing lowest scores across most impact categories, particularly those linked to energy use and chemical consumption. This reduction in environmental burden is primarily due to the sequence's improved energy efficiency, stemming from the inclusion of both oxygen delignification and the acid wash stage. These process optimizations reduce the reliance on chemicals like NaOH and ClO₂, leading to lower emissions and resource use. However, one trade-off is the higher water consumption observed in this sequence, mainly due to the acid wash stage, which requires additional water to remove hexenuronic acids effectively. The D₀-E_{op}-D₁-P sequence, however, stands out for its higher environmental impact in several categories, particularly in terms of GWP, and impact categories related to energy use and chemical consumption. As depicted in Fig. 5, this sequence is characterized by its significant chemical usage and higher energy demand during the bleaching process, contributing to its elevated GWP and associated negative environmental effects. The lack of oxygen delignification means more chemicals are needed in the later stages to achieve the necessary brightness, ultimately increasing the environmental burden.

The O/O-D₀-E_{op}-D₁-P sequence exhibits moderate environmental performance compared to the base case and the sequence with oxygen delignification and acid washing. Including oxygen delignification in this sequence reduces the need for harsh bleaching chemicals, lowering emissions associated with chemical production and transportation.

As illustrated in Fig. 5, the average BEK model provides a broader view of the environmental footprint associated with Brazilian BEK production, reflecting the mix of technologies and practices used across the industry. The model captures these bleaching sequences' cumulative effects on energy use, chemical consumption, emissions, and resource utilization.

3.2. Sensitivity analysis for including the forest carbon cycle in LCA

This study introduces two methodologies to make LCA results related to GWP more comprehensive by integrating the GWPbio and SOC sequestration potential. These emerging approaches are informed by recent research, including Liu et al. (2017), which underscores the critical role that GWPbio plays in accounting for the climate impact of biogenic CO₂ emissions from biobased products. Additionally, studies by Lan et al. (2024) and Forfora et al. (2024) demonstrate the importance of considering SOC sequestration. Since these factors remain

underreported in many LCAs of BEK pulp (as summarized in Table S8), sensitivity analyses are crucial for understanding their effects on GWP results.

GWPbio was calculated over a 100-year time horizon. This approach captures the delayed warming effect due to the time lag between carbon dioxide emissions from biomass and its eventual sequestration, considering the storage period of the harvested biomass with subsequent oxidation, as developed by Guest et al. (2013). As shown in Fig. 6, incorporating GWPbio into the LCA increases the GWP of the average Brazilian BEK pulp by 18%.

Conversely, incorporating SOC sequestration potential into the LCA of eucalyptus plantations results in a 39% reduction in GWP. This methodology, developed by Forfora et al. (2024), underscores the possibility of eucalyptus plantations acting as carbon sinks, sequestering carbon in soils. This finding represents an opportunity to mitigate GHG emissions. As the results indicate, while GWPbio increases the climate impact of biomass emissions, SOC sequestration serves as a counterbalance, reducing the overall GWP.

It is important to note that the bleaching sequences studied do not directly influence GWPbio or SOC sequestration potential, as these factors are primarily related to the total biomass used. Differences in biomass requirements between the sequences are minimal (less than 1%, as shown in Table 2). Thus, the primary focus of this sensitivity analysis is on the cumulative impact of incorporating GWPbio and SOC sequestration into the GWP of the average Brazilian BEK pulp, as illustrated in Fig. 6.

Integrating GWPbio and SOC sequestration into the LCA framework offers a more holistic understanding of the environmental impacts associated with BEK pulp production. It underscores the importance of considering the FCC, as delayed carbon emissions and long-term sequestration shape the overall GWP of BEK pulp production.

In order to reduce the environmental impact of BEK pulp production, it is recommended that industry experts adopt a standardized protocol for measuring soil CO₂ efflux and integrate these measurements with SOC models to estimate soil carbon sequestration. This comprehensive approach involves measuring general site characteristics such as soil type, depth, texture, bulk density, and pH and recording land management practices and disturbance history. It also includes monitoring meso- and microclimate conditions, such as soil temperature and water content at multiple depths with horizontal replicates per site, precipitation, and wind speed, preferably measured near chamber vents. Collecting these parameters will enable industry experts to calculate, interpret, and upscale soil CO₂ efflux rates (Kutsch et al., 2009) and estimate SOC sequestration (Lan et al., 2024). Implementing this protocol not only enhances the precision of environmental impact assessments but also informs the development of effective strategies to mitigate emissions associated with BEK pulp production.

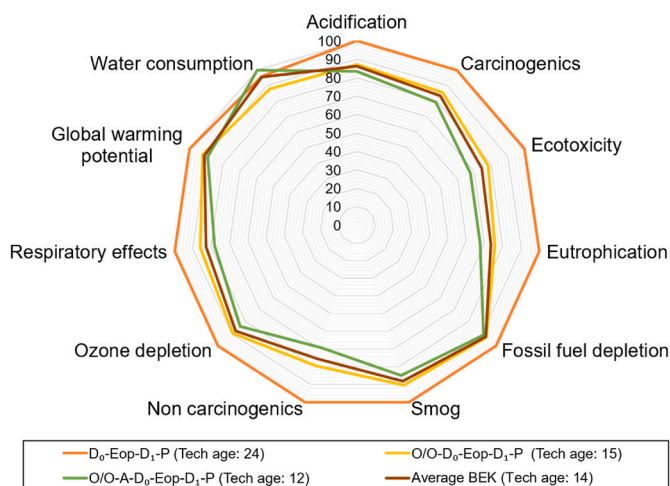


Fig. 5. Environmental categories for different bleaching stages of BEK pulp mill following the TRACI 2.1 impact assessment methodology. Tech age is the average age of the bleaching sequence technology.

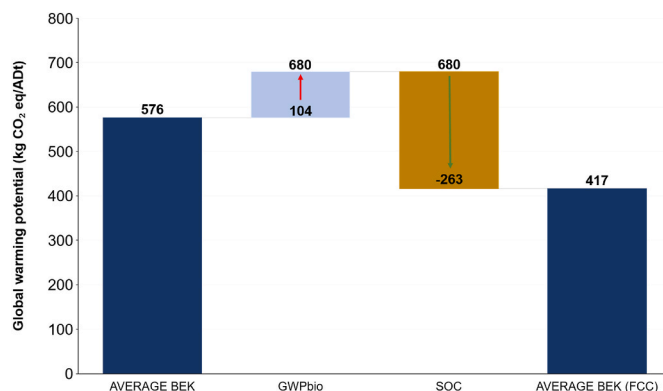


Fig. 6. Sensitivity analysis for including the FCC in LCA: GWPbio and SOC sequestration potential for Brazilian BEK market pulp delivered to the U.S.

3.3. Limitations

This study highlights several limitations associated with modeling the FCC within LCA. While the FCC plays a critical role in understanding the carbon dynamics of biobased systems, significant uncertainties remain. One of the key assumptions is that the GWPbio factor is based on immediate land revegetation after biomass harvest. However, if biomass residues decompose on-site, emissions could be underestimated due to delayed CO₂ release (Cherubini et al., 2011). The SOC sequestration model relies on a 15% stabilization ratio and considers only a single rotation on degraded lands. These assumptions may result in an overestimation of sequestration potential, particularly in regions where soils are nearing carbon saturation or multiple rotations occur (Forfora et al., 2024). The model also does not account for critical factors such as temperature, moisture, nutrient availability, and other soil properties which significantly influence carbon dynamics (Kutsch et al., 2009). Furthermore, processes like subsoil interactions, soil aggregation, and soil fauna activity (essential for long-term carbon stabilization) are omitted from the model (Kutsch et al., 2009; Paul et al., 1995; Smith et al., 1998; Jenkinson and Wild, 1988). Simplified interactions between soil types, climate, and land management practices also reduce the precision of sequestration estimates across various regions (Burke et al., 1989). Additionally, the analysis does not consider the specific properties of different soil types, which can greatly affect carbon storage capacity (Kelly et al., 1997). Another limitation is the assumption in the GWP calculations that biomass roots will remain in the soil for 100 years, unaffected by natural disturbances such as forest fires. The model assumes the persistence of soil carbon over a century (Forfora et al., 2024), which is difficult to guarantee under real-world conditions. Furthermore, according to the IPCC 2021 report, while CO₂ fluxes from vegetation and soils are a natural part of the carbon cycle, the biogeochemical processes governing these fluxes introduce substantial uncertainty. The complex interactions between ecosystems, climate conditions, and land management practices make it challenging to accurately predict long-term carbon sequestration (Intergovernmental Panel on Climate Change (IPCC), 2023).

4. Conclusion

This study underscores the significant potential for technological optimization in Brazilian BEK pulp production, mainly through adopting the O/O-A-D₀-E₀-D₁-P bleaching sequence. Our findings indicate that this sequence achieves the lowest GWP and improves environmental performance across acidification, eutrophication, ozone depletion, photochemical smog formation, ecotoxicity, human health cancer effects, human health non-cancer effects, respiratory effects, and fossil fuel depletion environmental impact categories. These improvements enhance sustainability and offer a clear path for operational efficiencies in mills that upgrade from outdated technologies. The trade-offs, such as increased water consumption, suggest areas for targeted innovation and resource management.

The average BEK model we developed provides a valuable benchmark for future assessments. By offering standardized LCI data for BEK pulp production, the model serves as a baseline against which alternative fibers and production processes can be evaluated. It can be employed to assess and compare the environmental performance of various biomass sources for pulp production, including natural forests, non-wood fibers, agricultural residues, and energy crops.

From a methodological standpoint, the integration of GWPbio and SOC sequestration into our LCA framework emphasizes the importance of the FCC in environmental evaluations. Although our sensitivity analysis highlights the potential of SOC sequestration to significantly lower GWP, the uncertainties linked to climate, land-use practices, and soil properties indicate the need for further refinement of SOC modeling. Methodological improvements in this area will help to enhance the accuracy of environmental impact assessments for bio-based products like

BEK pulp.

To further reduce the environmental impact of BEK pulp production, we recommend that industry experts adopt standardized protocols for measuring soil CO₂ efflux and integrate these measurements with SOC models to estimate soil carbon sequestration. This comprehensive approach involves detailed site assessments and soil and climate parameters monitoring, enabling accurate calculation and interpretation of soil CO₂ efflux rates and SOC sequestration. Implementing this protocol not only enhances the precision of environmental impact assessments but also informs the development of effective strategies to mitigate emissions associated with BEK pulp production.

Advancing SOC modeling, especially with attention to regional soil characteristics and long-term land management practices, will be critical for enhancing the sustainability of BEK pulp production. These refinements will also play a crucial role in advancing climate mitigation strategies within the broader industry. Continuing to innovate in these areas will be essential for shaping a more environmentally responsible pulp and paper industry.

CRedit authorship contribution statement

Rhonald Ortega: Writing – review & editing, Writing – original draft, Visualization, Project administration, Methodology, Investigation, Conceptualization. **Naycari Forfora:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Isabel Urdaneta:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Ivana Azuaje:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Keren A. Vivas:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Ramon E. Vera:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Jorge Franco:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Ryen Frazier:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Camilla Abbati:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Daniel Saloni:** Writing – review & editing, Supervision. **Hasan Jameel:** Writing – review & editing, Supervision. **Richard Venditti:** Writing – review & editing, Supervision. **Ronalds Gonzalez:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization.

Declaration of competing interest

The authors state no financial or personal conflicts of interest that could influence their work.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cesys.2024.100234>.

Data availability

Data will be made available on request.

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