Life cycle assessment of ECF bleaching sequences with focus on carbon footprint

PIA JOUR, KARIN HALLDÉN, AND EVA WACKERBERG

ABSTRACT: This paper presents a life cycle assessment (LCA) of bleached eucalyptus kraft pulp production in Brazil. The entire production system was investigated, starting with forestry and ending with bleached pulp at the gate of the pulp mill. Alternative bleaching sequences were compared for three different scenarios using somewhat different elemental chlorine-free (ECF) sequences: D_{hot}(EPO)DD, D_{hot}(EPO)DP, and aZeDP. The main difference between the scenarios investigated was the magnitude of the carbon footprint contribution from bleaching. For the base case and chemical island scenarios (both reflecting Brazilian conditions), the contribution was 15%-18% of the total carbon footprint. For the ecoinvent scenario, the corresponding share was 34%-41%. The ecoinvent scenario represents generic LCA data for bleaching chemicals. Ecoinvent is a public database commonly included in commercial LCA software. For each scenario, the alternative bleaching sequences studied resulted in similar carbon footprints of the bleached pulp. A comparison of the data from the different scenarios showed a large range of carbon footprints for the chemicals used for pulp bleaching. It is crucial to select data sets that are relevant in terms of geography and technology. The most dominant contributors to the carbon footprint of the unbleached pulp were forestry and pulp production. Although the focus has been on carbon footprints, the contributions to other environmental effects commonly included in LCAs were also assessed and only minor differences between the alternative bleaching sequences were found.

Application: This study shows how LCA can be applied to assess environmental impacts in the pulp and paper industry. Knowledge about the magnitude of the contributions made by the different parts of the value chain is useful as a basis for further improvements to reduce the environmental footprint.

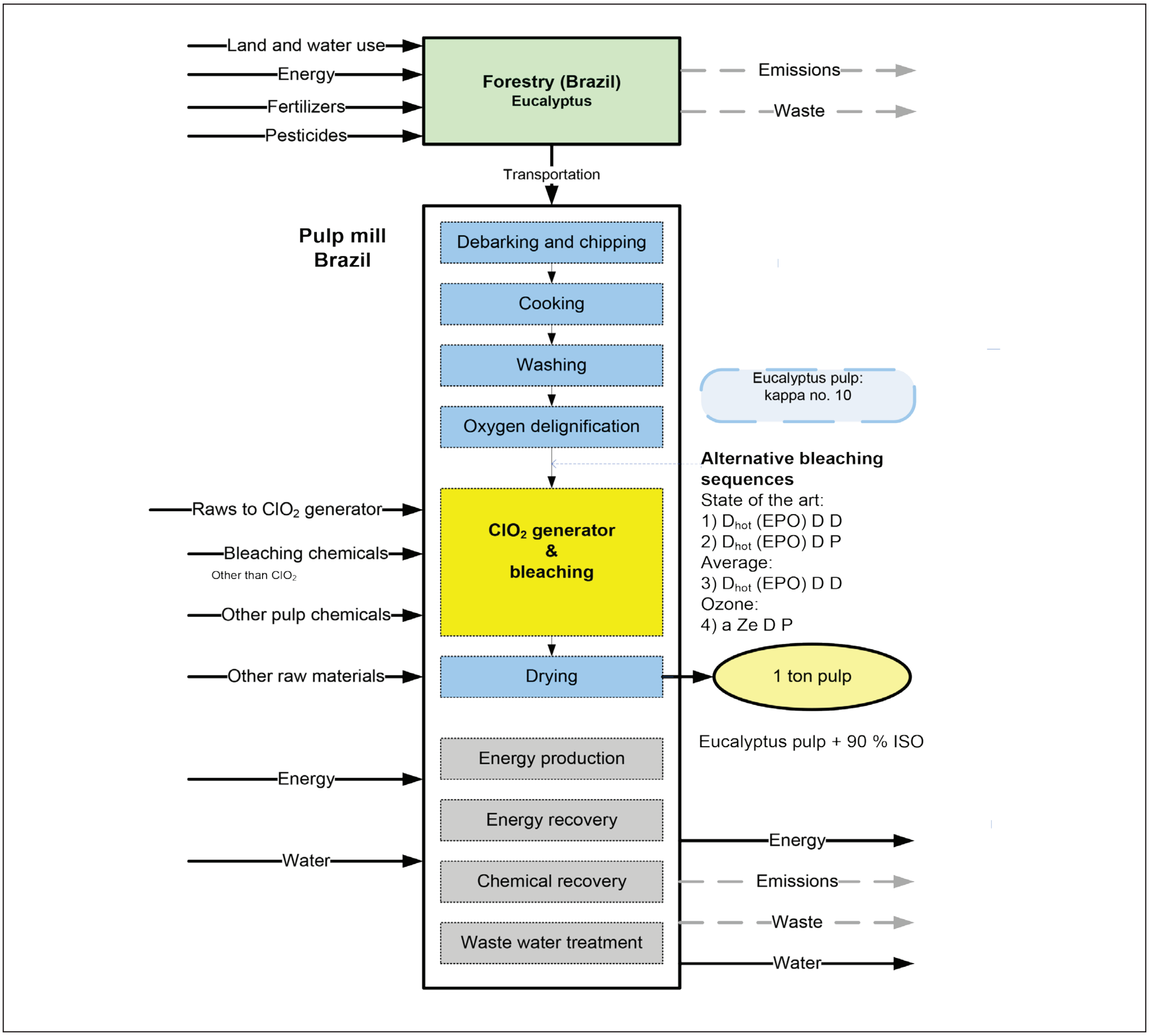
Substantial improvements have been made in recent decades when it comes to kraft pulp bleaching; the environmental impact per ton of bleached pulp caused by bleaching has been significantly reduced. Thirty years ago, molecular chlorine was commonly used for pulp bleaching. Nowadays, elemental chlorine-free (ECF) bleaching, using chlorine dioxide (ClO_2), is the dominant technology, with a market share of 87% of the bleached kraft pulp capacity worldwide [1]. Total elemental chlorine-free (TCF) bleaching has a market share of only 3%, and about 10% of the bleached kraft pulp is still bleached using chlorine or hypochlorite. Around 7% of the bleached kraft pulp is produced using ozone in an ECF or TCF bleaching sequence [1].

A number of life cycle assessments (LCAs) comparing alternative bleaching sequences have been published [2-5]. Ryynänen and Nelson [4] concluded that the difference in environmental performance between ECF and TCF bleached pulp is minor. In contrast, Hostachy [2] and Métais and Hostachy [3] concluded that ozone bleaching is significantly better than conventional ECF bleaching in terms of environmental performance. According to published LCAs of pulp and paper products, the relative significance of the bleaching (i.e., the bleaching process at the pulp mill and production of bleaching chemicals) varies depending on the type of environmental impact assessed [6,7].

The purpose of this study is to use LCA to quantify and compare the environmental impact of ECF bleaching sequences with and without ozone present. Although the focus is on the environmental effect climate change (measured as the carbon footprint), other types of environmental effects have also been included in the analysis. Knowledge of what type of environmental impact and the magnitude caused by the different parts of the pulp-making value chain is valuable when striving to minimize the negative environmental effects caused by pulp production. This is why the entire pulp production systems, starting with forestry and ending with the bleached pulp ready for distribution at the pulp mill gate, have been analyzed, rather than just the bleaching processes.

METHODS

For this study, we used the systems analysis method, LCA, performed according to ISO standards ISO 14040:2006 "Environmental management – life cycle assessment – principles and framework" and ISO 14044:2006 "Environmental management – Life cycle assessment - Requirements and guidelines," and, as far as possible, in line with the requirements of the new ISO standard regarding the



1. The system studied covers the production of bleached eucalyptus pulp in Brazil from the cradle to the gate of the pulp mill. Forestry and manufacturing of unbleached pulp were included in order to understand the relative significance of bleaching. Bleaching and manufacturing of bleaching chemicals were studied in greater detail. The functional unit was 1 ton of bleached pulp at the gate of the mill. In the chemical island scenario, the sodium chlorate plant is also located at the pulp mill site.

carbon footprint of products (ISO/TS 14067:2013 "Carbon footprint of products – Requirements and guidelines for quantification and communication"). The study was carried out using commercial LCA software (GaBi 4, PE International; Leinfelden-Echterdingen, Germany). The methodology used is described below. This study was also critically reviewed by experts in LCA of pulp and paper at the Technical Research Centre of Finland (VTT) to ensure that:

- Methods used to carry out the LCA are consistent with ISO 14040:2006 and ISO 14044:2006
- Data used is appropriate and reasonable
- Interpretations reflect the limitations identified and the goal of the study

Goal and scope

The goal of the study was to compare the environmental performance, focusing on the carbon footprint, of an oxygen delignified eucalyptus kraft pulp (kappa number 10) bleached to +90% ISO using the following alternative ECF sequences:

1) ECF state-of-the-art, D_{hot} (EPO)DD
2) ECF light state-of-the-art, D_{hot}(EPO)DP
3) ECF average, D_{hot}(EPO)DD
4) ECF ozone, aZeDP

Three scenarios were studied to investigate the consequences of using different data sets for the bleaching chemicals:

1) **Base case**: average origin of sodium chlorate used by

AkzoNobel's (hereafter known as "the supplier's") customers in Brazil (79% produced in Brazil and 21% imported).

2) **Chemical island**: sodium chlorate produced on-site at the pulp mill in a chemical island, which is the supplier's state-of-the-art chemicals production, management, and supply concept. One benefit of this concept is that sodium chlorate can be produced using biomass energy and that less transportation is needed.

3) **Ecoinvent**: Carbon footprint data sets for chlorine dioxide (with sodium chlorate upstream), hydrogen peroxide, and sodium hydroxide were collected from the ecoinvent database (ecoinvent; Zurich, Switzerland). Ecoinvent is a public LCA database commonly included in commercial LCA software.

Scenarios 1 and 2 describe modern technology and the situation in Brazil to the best of our knowledge. Scenario 3 is to be regarded as a sensitivity analysis. The study was conducted with a focus on the environmental effect climate change. The carbon footprint calculations carried out exclude the biogenic emissions of carbon dioxide. The environmental impact categories included and the method used to aggregate the quantified inputs and outputs are as follows:

- Climate change [8]
- Ozone depletion [9]
- Acidification [9]
- Eutrophication [9]
- Photochemical ozone formation [9]
- Primary energy use, renewable and non-renewable [9]

System boundary

This study is a cradle-to-gate LCA. **Figure 1** describes the system boundaries. The functional unit (basis of comparison) is 1 moisture-free metric ton of bleached pulp (+90% ISO) at the gate of the pulp mill in Brazil. Forestry and manufacturing of unbleached pulp are included to understand the relative significance of bleaching. The eucalyptus plantations and the pulp mill are located in Brazil. Bleaching and manufacturing of bleaching chemicals have been studied in greater detail. In this case, the chemical island scenario means that chlorine dioxide and sodium chlorate are produced at the pulp mill site. The production of sodium chlorate can therefore benefit from access to steam and electricity produced from biomass. The base case scenario also benefits, to some extent, from access to bio-based steam and electricity in the production of sodium chlorate, because 34% of the chlorate used by the supplier's average customer in Brazil is produced in chemical islands.

The ecoinvent scenario assumes sodium chlorate to be produced in a separate production unit and that there is a transport by lorry and train to the on-site chlorine dioxide generator. The ecoinvent data for sodium chlorate is based on information from European manufacturers. The electricity used in the production of sodium chlorate and chlorine dioxide in the ecoinvent calculations is based on a mix of European electricity providers and reflects a high percentage of fossil-based power.

The most important assumptions used in this study are the following:

- In the base case and chemical island scenarios, the chlorine dioxide is produced in a single vessel process-salt cake wash generator.
- The pulp mill's excess electricity can be exported to the national electricity grid in Brazil and replaces the production of an average Brazilian electricity mix.
- The oxygen and ozone used in the bleaching process is produced on-site with electricity produced from biomass (part of the pulp mill's excess energy).

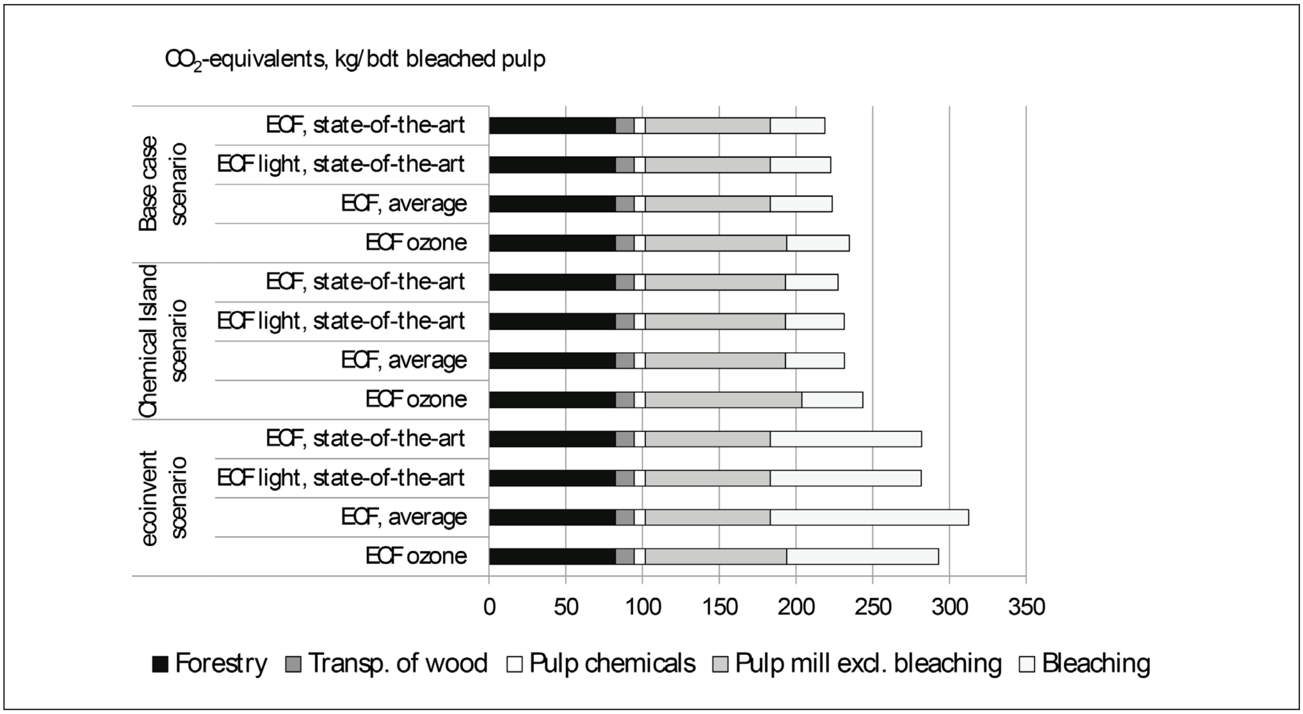
Data collection

When carrying out an LCA, vast amounts of information and data are needed. The relevance and quality of the data used

Chemicals	Chemical Charges (kg/moisture-free metric ton)			
	ECF State-of-the-Art	ECF Light State-of-the-Art	ECF Average D_{hot}(EPO)DD	ECF Ozone [1] aZeDP
Chlorine dioxide	9.9	7.6	14.8	8
Chlorine dioxide (as Cl)	26	20	39	21
Hydrogen peroxide	2	6	2	5
Oxygen	3	3	3	0
Sodium hydroxide	12	14	12	12
Sulfuric acid	6	6	6	18
Ozone	0	0	0	5

[1] Literature data from Hostachy [2].

I. The charge of chemicals in kilograms per moisture-free metric ton of pulp used in the calculations for the different elemental chlorine-free (ECF) sequences. The chemicals are given as 100%, except for sulfuric acid, where the concentration is 96%.



2. Carbon footprint contributions made by forestry, transportation of wood, production of pulp chemicals, pulp mill (excluding bleach plant) and bleaching. Bleaching includes both the bleaching process and production and transport of bleaching chemicals cradle-to-gate.

will influence the validity of the results. A detailed description of the type of data used for the different parts of the value chain can be found in Jour et al. [10]. The alternative bleaching sequences and their charges of chemicals are presented in **Table I**.

RESULTS AND DISCUSSION

Carbon footprint of the bleached pulp

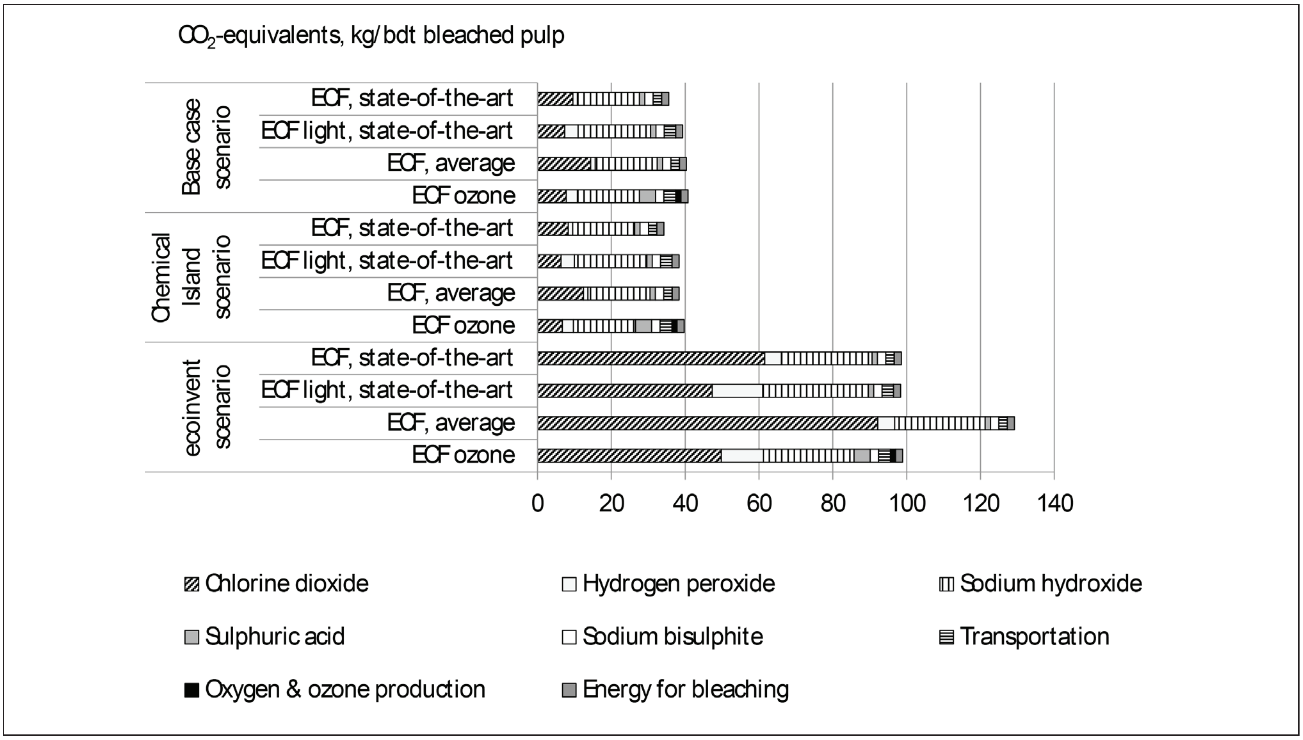
Figure 2 shows the total carbon footprint per ton of bleached pulp for the three scenarios studied. The sum of the contributions from forestry, transport of wood to the pulp mill, pulping chemicals, and the pulp mill—excluding bleaching—are modeled in the same way for the different bleaching sequences and scenarios in this study, except for the amount of pulp mill excess electricity available for export. The last segment of the bars (from left to right) represents the contribution made by bleaching, including the production of bleaching chemicals cradle-to-gate, transportation to the pulp mill, and contributions from the bleaching process itself.

The explanation for the base case and chemical island scenarios showing similar carbon footprints is that the sodium chlorate in both scenarios is produced using a high share of renewable energy. If a slightly higher share of fossil energy had been used for the base case sodium chlorate, the chemical island scenario would have been significantly better.

One assumption is that the mill is connected to the national power grid, which makes it possible to export the excess electricity. However, this option is not available to all mills in Brazil today. The amount of pulp mill excess electricity available for export varies between the scenarios. This influences the carbon footprint contribution made by the pulp mill, excluding bleaching. **Table II** shows the amounts of excess electricity and the carbon footprints for unbleached eucalyptus pulp (including forestry, transportation of wood, pulping chemicals,

	Excess Electricity (MJ/moisture-free metric ton pulp)	Carbon Footprint (kg CO_2/moisture-free metric ton pulp)
No export of excess electricity	—	226
100% export of excess electricity	1627	181
Chemical island	1270	191
Ozone bleaching	1241	192
Ozone bleaching and chemical island	884	202

II. Excess electricity and carbon footprint for unbleached eucalyptus kraft pulp produced in Brazil.



3. Carbon footprint (CO_2-equivalents) contributions from the production of the different chemicals, cradle-to-gate, used in the bleach plant and the bleaching process.

and the production of unbleached pulp) produced in Brazil. According to our calculations, the carbon footprint can vary between 181 kg CO_2-equivalents per ton of unbleached pulp (no electricity-consuming production of bleaching chemicals on site) and 202 kg CO_2-equivalents per ton of unbleached pulp (ozone and sodium chlorate production on site). The reason for the higher carbon footprint of the unbleached pulp when bleaching chemicals are produced on site is that less pulp mill excess electricity is available for export to the national grid. For the bleached pulp, this is compensated for by a smaller carbon footprint contribution from production of bleaching chemicals. This is discussed in greater detail by Jour et al. [10].

The carbon footprint contribution made by the pulp mill (excluding bleaching) is because of the production of steam and electricity. To generate energy, biomass and a small amount of fossil energy are needed. The fossil energy is mainly used for the lime kiln (366 kWh per ton of pulp [11]). In a chemical island, hydrogen from the chlorate plant can replace the fossil fuel; however, that has not been the case in this study. In the calculations, the fossil fuel is assumed to be natural gas.

In summary, the carbon footprint of bleached pulp is roughly attributed to the following:

- 26%-38% from forestry
- 4%-6% from transportation of wood
- 2%-3% from the production of pulping chemicals cradle-to-gate
- 26%-42% from the pulp mill excluding bleaching
- 15%-41% from bleaching

Even though the share of fossil energy used by the pulp mill is low, the resulting contribution to the carbon footprint is significant. Therefore, ways to reduce the amount of fossil energy needed would improve the carbon footprint of the kraft pulp production considerably.

State-of-the art ECF bleaching gives the smallest carbon footprint in all scenarios (Fig. 2 and **Fig. 3**). ECF ozone gives the largest carbon footprint in the base case and chemical island scenarios. In the ecoinvent scenario, the sequence ECF average shows the highest carbon footprint. The share of the total carbon footprint from bleaching is about 15%-18% for the base case and chemical island scenarios and 34%-41% in the ecoinvent scenario.

Among the greenhouse gases included in the analysis, carbon dioxide contributes the most (80%-85%) to the total carbon footprint, followed by nitrous oxide (~8%), methane (~6%), and non-methane volatile organic carbon (~3%). This is valid for all scenarios. Most of the nitrous oxide emissions are related to forestry (silviculture).

Carbon footprint of the bleach plant

In Fig. 3, the carbon footprint contribution from bleaching is further divided between the different chemicals used in the bleach plant, transportation of chemicals to the pulp mill, and the energy used in the bleaching process. Overall, the carbon footprint contributions made by bleaching are very similar: 34-41 kg CO_2-equivalents per ton of bleached pulp for all sequences in the base case and the chemical island scenarios. The bleaching sequence, ECF state-of-the-art, is the

Chemical (conc., %)	Base Case Scenario	Chemical Island Scenario	Ecoinvent Scenario	Minimum in Databases	Maximum in Databases
Chlorine dioxide (100%)	0.97 [1]	0.84 [1]	6.2 [3]	0.84 [1]	6.2 [3]
Hydrogen peroxide (70%)	0.42 [4]	0.42 [4]	1.6 [3]	0.38 [1]	3.7 [2]
Ozone	0.28 [5]	0.28 [5]	0.28 [5]	0.28 [5]	8 [3]
Sodium hydroxide (100%)	1.4 [2]	1.4 [2]	2.0 [3]	1.4 [2]	2.5 [3]
Sulfuric acid (96%)	0.24 [2]	0.24 [2]	0.24 [2]	0.14 [3]	0.24 [2]

[1] AkzoNobel internal data from own production units (2012); [2] data in the software supplied by PE International (GaBi 4); [3] ecoinvent (V 2.2) [11]; [4] AkzoNobel internal data from own production units (2012) with modified data using Brazilian electricity mix; [5] ecoinvent (V 2.2) [11] with modified data using electricity produced from biomass.

III. Carbon footprint (kg CO_2-equivalents per kg chemical) for the chemicals used in the bleach plant and the range, minimum and maximum, between data from different databases on hand.

best alternative; the worst, ECF ozone, is only 7% higher.

In the ecoinvent scenario, the contribution from bleaching is much higher: 98-129 kg CO_2-equivalents per ton of bleached pulp. ECF state-of-the-art, ECF light, and ECF ozone are all about the same. ECF average is the worst alternative because of the extreme carbon footprint data for chlorine dioxide in the ecoinvent database (**Table III**) and the relatively high use of chlorine dioxide. For all bleaching sequences, sodium hydroxide and chlorine dioxide contribute the most. The chlorine dioxide segment also includes the production of sulfuric acid, methanol, sodium chlorate, and transportation, if required, to the ClO_2 generator. Hydrogen peroxide gives the third greatest contribution to the carbon footprint in the ECF light and ECF ozone sequences because of the higher charges used in these sequences. For the ECF ozone sequence, a higher charge of sulfuric acid is needed, thus a higher carbon footprint contribution is observed. Furthermore, transportation includes the transport of all chemicals not produced on-site to the pulp mill. The carbon footprint contribution by transportation of bleaching chemicals to the pulp mill is clearly visible, but relatively small.

Carbon footprint data for the chemicals used in the bleach plant

Table III presents the carbon footprint data for the chemicals as used in the different scenarios. The data between the scenarios differs substantially. The base case and the chemical island scenarios are at the low end, while the ecoinvent scenario is at the high end. This is especially valid for chlorine dioxide, hydrogen peroxide, and sodium hydroxide.

For chlorine dioxide, the main reason for the differences is the electricity mix used in the production of sodium chlorate. The ecoinvent data are representative for European manufacturers and assume a European electricity mix with a fairly large share of fossil energy carriers. In the base case scenario, an average origin of sodium chlorate used by Akzo Nobel customers in Brazil is used. This sodium chlorate has been produced with a high share of renewable energy (hydro power and biomass). Electricity needed for sodium chlorate production in the chemical island scenario is produced from biomass. Table III also shows the minimum and maximum levels of data on hand for these chemicals.

Primary energy and other environmental impact categories

Primary energy demand is the total energy required by the system. It includes the extraction and processing of natural resources used to produce raw materials and energy carriers to produce electricity and heat. It also includes the total energy contained in raw materials, raw fuels, and other forms of energy (e.g., hydro power) going into the system. **Table IV** shows the primary energy demand for the alternative bleaching sequences for the three different scenarios. The share of renewable primary energy is also presented: 92%-93% for the base case and the chemical island scenarios and 89% for the ecoinvent scenario. The primary energy demand of the bleaching sequences did not differ significantly.

Although climate change is an important environmental effect, it is only one impact category in LCA. If all the focus is on one impact category, it might lead to sub-optimization. This is why other environmental effects commonly included in LCA, such as ozone depletion, acidification, eutrophication, and photochemical ozone formation, are also included in this study. The purpose of including those effects is to investigate whether there were any significant differences between the alternative bleaching sequences. Table IV shows that the differences were small between the bleaching sequences compared. This is the case for all of the environmental impact categories and for all scenarios evaluated.

Environmental effects, human toxicity, and eco-toxicity can also be included in LCA. However, because LCA is not the ideal method for handling these environmental effects, they have not been included here. When it comes to pulp bleaching, two types of analytical measures related to emissions to water are relevant to discuss with regard to toxicity: chemical oxygen demand (COD) and adsorbable organic halogen com-

Primary Energy	Primary Energy (GJ)	Share of Renewable Energy (%)	Ozone Depletion Potential (kg R11-eqv.)	Acidification Potential (kg SO_2-eqv.)	Eutrophication Potential (kg PO_4-eqv.)	Photochemical Ozone Formation (kg ethene-eqv.)
Base case scenario						
ECF, state-of-the-art	44	93	5.2E-06	3.3	1.2	0.46
ECF light, state-of-the-art	44	93	5.2E-06	3.3	1.2	0.46
ECF, average	45	92	5.4E-06	3.3	1.2	0.46
ECF ozone	46	92	6.0E-06	3.4	1.2	0.47
Chemical island scenario						
ECF, state-of-the-art	45	92	5.8E-06	3.3	1.2	0.46
ECF light, state-of-the-art	45	92	5.8E-06	3.3	1.2	0.46
ECF, average	46	92	5.8E-06	3.4	1.2	0.46
ECF ozone	46	92	6.5E-06	3.4	1.2	0.47
ecoinvent scenario						
ECF, state-of-the-art	44	89	1.1E-05	3.7	1.2	0.49
ECF light, state-of-the-art	44	89	1.2E-05	3.6	1.2	0.49
ECF, average	45	88	1.3E-05	3.9	1.2	0.51
ECF ozone	45	89	1.2E-05	3.7	1.2	0.50

IV. Total primary energy demand, share of renewable primary energy and results for other environmental impact categories included in the life cycle assessment per bone dry ton of bleached pulp.

pounds (AOX). COD emissions are included in the quantification of the contributions to the environmental effect of eutrophication.

The individual substances contributing to the analytical measures COD and AOX would be relevant to include in a risk assessment. This would provide more accurate information, rather than using LCA methods to assess the toxicity based on COD and AOX values only. A detailed risk analysis was beyond the scope of the study reported here. What was clear was that the levels of COD after external treatment were comparable for the different bleaching sequences studied, in the range of 7-9 kg/moisture-free metric tons of bleached pulp with the ECF ozone sequence at the lower end. AOX was below 0.2 kg/moisture-free metric ton of bleached pulp for all sequences.

CONCLUSIONS

The following conclusions from this analysis are valid for the production of ECF bleached eucalyptus kraft pulp in Brazil:

- Of the total carbon footprint of eucalyptus pulp, 15%-41% originated from bleaching, including the production of bleaching chemicals cradle-to-gate, transportation of bleaching chemicals to the pulp mill, and the bleaching process itself. For the base case and the chemical island scenarios, the contributions from bleaching were 15%-18%. For the ecoinvent scenario, the contribution from bleaching was 34%-41%.
- The alternative bleaching sequences studied resulted in rather similar carbon footprints for the bleached pulp in the base case and the chemical island scenarios. The ECF state-of-the-art sequence showed the lowest carbon footprint (219-227 kg CO_2-equivalents/moisture-free metric ton) and ECF ozone the highest (235-244 kg CO_2-equivalents/moisture-free metric ton).
- Using data from the public LCA database ecoinvent, state-of-the-art ECF and ECF light bleaching gave the lowest carbon footprints (282 kg CO_2 equivalents/moisture-free metric ton), followed by ECF ozone (293 kg CO_2 equivalents/moisture-free metric ton) and average ECF (312 kg CO_2 equivalents/moisture-free metric ton).
- There was a large range in the carbon footprint data of the chemicals used for pulp bleaching. It is crucial to select data that are relevant in terms of geography and technology. Sodium chlorate and sodium hydroxide each require large amounts of electricity when produced. The way electricity is produced strongly influences the carbon footprint of these two chemicals.
- The carbon footprint of bleached pulp is roughly made up of 26%-38% from forestry, 26%-42% from pulp mill excluding bleaching, 4%-6% from transportation of wood to pulp

mill, 2%-3% from pulping chemicals cradle-to-gate, and 15%-41% from bleaching. This assumes that the excess electricity from the mill is exported and replaces electricity produced by the national grid in Brazil.

• For environmental effects other than the carbon footprint, only minor differences between the alternative bleaching sequences were found. **TJ**

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ABOUT THE AUTHORS

We chose this subject for research to ensure a sustainable future business for the pulp industry. Sustainability is also of key importance for our company when it comes to our own production and the use of our chemicals by customers.

We and others have carried out life cycle assessments (LCAs; or carbon footprint calculations) before. In this study, we have focused on the use of bleaching chemicals in the production of eucalyptus market pulp and included a comparison between data found in the ecoinvent database and from our own production.

The most difficult part of the study was to make sure that the data used were relevant. To ensure this, the Technical Research Centre of Finland (VTT) was engaged to critically review the work. The most interesting part was to see the importance of collecting accurate data for the LCA.

Mills might benefit from the knowledge about the carbon footprint contribution from the different steps in the value chain when optimizing their production of bleached market pulp.

Jour

Halldén

Wackerberg

Our next steps are to communicate our data to the public LCA database ecoinvent, and to continue to communicate and discuss sustainability with our customers.

Jour is scientist and Wackerberg is manager, application RD&I, Akzo Nobel Pulp and Performance Chemicals AB, Bohus, Sweden. Halldén is eco-efficiency specialist, Akzo Nobel AB, Gothenburg, Sweden. Email Jour at pia.jour@akzonobel.com.