Life cycle inventory of pine and eucalyptus cellulose production in Chile: effect of process modifications.

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Abstract This work reports the life cycle inventory (LCI) of cellulose production in Chile, following a cradle-to-gate approach. Primary data have been used in this study, and cover 100% of pine and eucalyptus cellulose production capacity in Chile. Results show that pine based cellulose presents greater chemical and environmental loads than eucalyptus. Most fossil energy consumption takes place in raw materials transport, chemical manufacturing and limestone kilns in cellulose plants. Effluent discharges are associated with bleaching operations. New forestry practices and pulping and bleaching processes, introduced during the last decade, has led to significant reductions in water consumption, pollutant discharges, and air emissions. However, despite the sharp reduction in chemicals consumption due to process improvements and new technology, greater associated GHG emissions have been recorded as a result of the significant increase in the share of thermoelectric generation experienced during the last decade in Chile.

1 Introduction

Chile is a major exporter of bleached kraft cellulose, with an annual production around 5,000,000 ton per year, supported by more than 2 million hectare of pine and eucalyptus plantations. Forestry and industrial wood processing features large amounts of water and chemicals consumption, as well as significant effluent discharges and air emissions, and occupational risks. Since early 1990s, Chilean cellulose production capacity has doubled, leading to growing concerns about potential environmental impacts. Thus, industry has been under increasing pressure to improve process performance, according to modern standards. As a result, thorough technological upgrading has taken place in the last decade, and new mills feature state of the art processes. Indeed, all plants now feature elementary chlorine free (ECF) bleaching sequences, secondary effluent treatment and air emission abatement technologies.

Over 90% of cellulose plants are located within the Bio Bio region in southern Chile, where most forestry plantations are located [1]. Climate change has significantly affected rainfall, reducing water availability for industrial purposes. Moreover, nowadays water is a key issue for future development of cellulose production in the country. Additionally, reduction in hydroelectric capacity has led to massive introduction of thermoelectric power generation, with a considerable increase in greenhouse gases (GHG) emissions associated with electricity production. Within this context, different initiatives to reduce water and carbon footprints, effluent toxicity, and other environmental and occupational hazards along the cellulose value chain have been developed. This work presents the effect of process modifications on the environmental loads associated with cellulose production in Chile, following a life cycle approach.

2 Methodology

This works has been conducted within the ISO 14040-2006 methodological framework [2]. This LCI followed a cradle-to-gate approach, with the system boundaries set at the nursery where selected seedlings are grown, and at Talcahuano port (Bio Bio Region), where Chilean cellulose is shipped to international markets.

Primary data from 7 bleached kraft cellulose plants were obtained for the period 2008-2009, accounting for 100% of beached cellulose production in Chile. Data included raw materials, energy and chemicals consumption, air emissions, water discharges and solid waste.

Electricity LCI (Central Interconnected System, SIC) was considered taking 2008 as a reference year. Data from the National Electricity Distribution System (CEDEC), National Energy Commission, and yearly reports from main electricity generation and distribution companies were used to determine the electricity LCI.

Additionally, data from mills featuring chorine based bleaching sequences, typical of Chilean mills during the early 1990's, was also collected from historical records corresponding to three representative local plants [3]. The corresponding Chilean electricity LCI was also estimated for that reference year.

3 Results

The main processes included in the production system are described below.

3.1 *Process Description*

3.1.1 Forestry Subsystem

Nursing stage: Good seedling quality is a fundamental requirement to achieve acceptable forestry productivity. Current nursing practices in Chile include careful site selection, optimal nutrient addition, genetic improvement, disease control, controlled irrigation, and careful handling. Since *Eucalyptus globulus* is sensitive to low temperature, covered root methods are used, with 300 plants/m^2. In the case of *Pinus radiata*, bare-root procedures are also found, with 200 plants/m^2.

Establishment of Plantation: This stage is highly critical, since plants are transported from the protected nursing environment to the plantation site, where they will be subjected to environmental stress and competition from other species. The prospective site has to be thoroughly cleaned using manual, mechanical and/or chemical means to remove existing vegetation. In some cases, controlled burning is used to eliminate those residues. Deep ripping on compacted soils is usually carried out in order to improve the soil physical properties, and water retention capacity. Planting is carried out during winter, and beginning of spring. Plant density is selected according to the type of final product, and site quality. In Chile, the average initial stocking is around 1,600 trees/ha, both for pine and eucalyptus plantations. In some cases, preventive fertilization is conducted when required. Post-plantation chemical weed control is carried out in spring. During the first few years, competition by other species, and pests should be kept under control, since small tress may not be able to survive under those conditions.

Forest Management: This includes all activities carried out between the establishment of plantation and harvesting. Management practices depend on product objectives [4]. In Chile, pine plantations are managed to obtain knot-free wood, timber, and pulpwood. On the other hand, most eucalyptus plantations are aimed at producing pulpwood, with negligible uses in plywood, and board making. Pruning is necessary to produce knot-free timber. Thinning is carried out to stimulate growth by regulating stand density. In the case of pine plantations, non-commercial thinning is practiced at about 4-5 years, whereas commercial thinning is carried out at 7-14 years, mostly for pulp making, reaching a final density around 300-500 trees/ha. Depending on management, soil quality, and climate, 30-80 m^3/ha pulpwood could be obtained from commercial thinning.

Harvesting: Chilean *Pinus radiata* and *Eucalyptus globulus* harvest cycles are 20 and 10 years, respectively. Harvesting comprises felling, delimbing, bucking, transport to roadside, loading, and hauling. A wide range of techniques, and equipments are currently used. Intermediate processing operations, such as delimbing, bucking, or chipping, can take place at the stump, at roadside, or at the mill. Transport to roadside could be accomplished using animals or tractors, transported in the bunk of an off-road vehicle, or moved by cable. Depending on forest management practices, and soil quality, around 200-500 m^3/ha and 200-300 m^3/ha are harvested, for pinewood and eucalyptus, respectively. All major forestry companies maintain active reforestation programs, to keep a reliable biomass stock for long term industrial processing, and sustainability. Reforestation of harvested plots is conducted after appropriate site preparation.

3.1.2 Transportation Subsystem

Trucks (10-25 ton) are used for woody raw materials transport, whereas most chemicals and cellulose products are transported by rail. The distance between plantations and industrial sites ranges within 50-150 km, whereas most bulk chemicals (eg. sodium hydroxide and chlorate) are produced at industrial states in Talcahuano (Bio Bio Region), located at 100-150 km from mills.

3.1.3 Pulp Production

All leading Chilean companies feature a close integration between forestry and industrial processing. Around 50% harvested pinewood goes directly as a raw material for pulping, while the rest is sent to sawmills. In turn, sawmills generate pulping residues that represent 30% of total input. On the other hand, most harvested eucalyptus wood is used for pulping, with a negligible amount destined to board manufacturing.

Wood preparation: This includes wood debarking, and chipping. Chips are then screened, and stored. These operations involve considerable electrical power consumption. In modern plants, bark and other woody residues are used as fuel for steam and electricity production.

Digesting: Chips are treated at high temperature and pressure (160-180°C), with a NaOH and Na^2S solution at high pH (white liquor). This chemical attack leads to the dissolution of around 52-54 % and 46-48 % of initial wood, for pine and eucalyptus, respectively. Traditional cooking for pine achieved a kappa number around 30 and 25, for pine and eucalyptus pulp, respectively. Modern cellulose mills included in this study feature extended cooking, obtaining a crude pulp with kappa values around 24 and 14, for pine and eucalyptus, respectively. After cooking, crude fibres are separated from the black liquor, and washed to remove residual chemicals.

Oxygen delignification: This represents one of the major technological improvements. Modern plants incorporate an oxygen delignification stage, under alkaline conditions, which reduces lignin content by around 50-60%, to reach a kappa value in the range 11-13 and 6-8, for pine and eucalyptus, respectively.

Energy and Chemicals Recovery Systems: The black liquor has a high calorific potential and contains most of the digestion chemicals. The black liquor is concentrated using evaporators, and burnt in the recovery boiler, where all organic matter is oxidized to CO_2. The solid residue (Na_2CO_3 and Na_2S) is dissolved and mixed with CaO to regenerate the "white liquor" for digestion, leaving Na_2CO_3 as a residue. In turn, the CaO is regenerated by thermal treatment of Na_2CO_3 in a lime kiln.

Bleaching: Chileans plants move from chlorine-based sequences in the early 90's, to elementary chlorine free (ECF) bleaching sequences. These feature the use of ClO_2 as oxidation agent(D), and oxidative alkaline extraction, with O_2 and H_2O_2 (EOP). In the case of pine pulp, a $D_0E_{OP}D_1D_2$ sequence is normally used, whereas a shorter sequence ($D_0E_{OP}D_1$) is used for eucalyptus pulp. Liquid waste from bleaching contains chlorine compounds that are not suitable for burning in the recovery boiler due to potential corrosive effects. These effluents account for most of the contaminant load of final waste waters.

ClO_2 in-plant Generation: All surveyed plants generate ClO_2 by reduction of $NaClO_3$ under acid conditions (ie. in the presence of H_2SO_4), using CH_3OH as the reducing agent. It is interesting to note that $NaClO_3$ is produced by a local electro-chemical plant, that also generates NaOH, using NaCl as a common raw material.

Residual Gas Treatment and Disposal Systems: Gas emissions mainly come from the recovery boiler, the lime furnace, the power boiler, and vents. These are composed of particular matter, SO_2, mercaptans, and other volatile hazardous organic and inorganic pollutants (eg. chloroform, methanol, chlorine dioxide). In this study, gas emissions are characterized on the basis of CO_2, SO_2, TRS (total reduced sulphur), and HAP (Hazardous Airborne Pollutants). TRS is a generic parameter to account for mercaptans, and H_2S. HAP takes into account all other volatile hazardous waste. Due to legal pressures, abatement measures, such as, electrostatic precipitators, gas filters, scrubbers, and incinerators, have been introduced in all plants. The latter is used to incinerate non condensable gases. Gases from boilers are released to the atmosphere through 50-60 m chimneys.

Residual Liquid Treatment and Disposal Systems: Liquid wastes are generated from bleaching, and from general washing and cleaning operations. Bleaching effluents are highly coloured and contain dissolved organic and chlorinated organic compounds, and residual bleaching chemicals. Washing effluents may contain suspended solids, and other components from accidental spills. Environmental loads from final effluents are expressed in terms of generic parameters: COD, and AOX, to account for the total organic load and organo-chlorinated compounds, respectively. Effluent treatment systems feature pH neutralisation, cooling, primary sedimentation, and activated sludge.

Residual Solid Treatment and Disposal Systems: All woody residues from processing (eg. bark, knots) and effluent treatment sludge, are burnt in power boilers, to generate steam and electricity. Solid residues (eg. boiler ash, dregs, grits, sand, stones, spent oils, dirty chips, dust, etc) are disposed in in-plant controlled landfill.

3.2 Inventory results

Results on chemical and environmental loads associated to bleached kraft cellulose in Chile are summarized in Tables 1 and 2.

Table 1: **: Main chemical loads associated to BK Cellulose Production in Chile**

	Eucalyptus BK Pulp		Pine BK Pulp	
	1992	2008	1992	2008
	Kg /bone dry ton cellulose			
Sodium Hydroxide	22	25	47	35
Sodium Chlorate	17	24	28	34
Chlorine	27	0	55	0
Calcium Carbonate	16	15	30	18
Oxygen	0	23	0	30
Sulphuric Acid	16	21	23	33
Hydrogen Peroxide	2	3	1	4
Methanol	1	3	3	4
Forestry Biocides (various)	$2\ 10^{-3}$	$1\ 10^{-3}$	$4\ 10^{-3}$	$3\ 10^{-3}$
Fuel Oil+Diesel	90	85	104	93

In general, chemical loads associated with eucalyptus cellulose are much lower than pine based cellulose. This is explained by the lower lignin content and greater density of eucalyptus wood, as compared with pine. Additionally, technological improvements have significantly reduced bulk chemicals requirements in most cases. Sodium chlorate appears to increase due to the complete substitution of chlorine by chlorine dioxide as bleaching agent. This is in agreement with published LCI cellulose reports [5]

Table 2. Main environmental loads of BK Cellulose Production in Chile

	Eucalyptus BK Pulp		Pine BK Pulp	
	1992	2008	1992	2008
	Kg /bone dry ton cellulose			
Emissions to air				
CO_2 (fosil)	366	395	458	462
N_2O	0,002	0,004	0,004	0,005
CH_4	0,21	0,23	0,28	0,28
GHG	372	401	466	471
TRS	6.1	0.2	8,2	0.4
MP	0,05	0,05	0,08	0,07
Discharges to water				
COD	890	114	1190	146
AOX	4.2	0.1	7.6	0.1
NO_3^-	2	2	4	3
$PO_4^=$	1	1	2	1
$SO_4^=$	10	14	16	29
Natural Resources Depletion				
Crude oil, in ground	101	108	119	123
Natural gas, in ground (m^3)	6	8	9	10
Hard coal, in ground	19	20	34	27
Water from surface sources	81	32	98	34

Pine cellulose presents greater environmental loads than eucalyptus pulp. Liquid effluents are one of the main environmental aspect, and might seriously affect the quality of recipient waters. Since most cellulose production is located within the Bio Bio river basin, pollutants could seriously affect drinking and irrigation water.
Finally, it can be seen that process improvements since 1992 led to a significant increase in greenhouse gases. Manufacturing of bulk chemicals such as sodium hydroxide and chlorate heavily rely on electrochemical processing, with

considerable electricity consumption. Although overall bulk chemical consumption decreased since 1992, the Chilean electricity matrix experienced a drastic increase in the share of coal based thermoelectric plants, leading to a greater carbon footprint of cellulose. Only fossil GHG emissions are shown here.

4 Conclusions

This paper shows that process modifications have resulted in overall improvements in environmental performance. On the other hand, it shows the importance of the electricity matrix in the cellulose carbon footprint.

5 References

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