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Emission Factors for Combustion of Biomass Fuels in the Pulp and Paper Mills

Abstract

As one of the biggest wood consumers, pulp and paper mills are full participants in the $C \Rightarrow CO_2 \Rightarrow C$ cycle. Thanks to the production of climate-neutral biomass fuels recovered from pulping (including cellulose for textile applications, i.e. fibre manufacturing) and paper-making waste, carbon dioxide emission from biomass combustion as neutral is not counted in the inventory. In this paper, the main grades of biomass waste are defined. The Kyoto Protocol target on reduction of greenhouse gas (GHG) emissions and methods of emission factors from different biomass boilers and other sources are collected. At the end, we present and discuss the factors of the other gas emissions from recovery boilers, lime kilns and BFB boilers when burning fuel mixture of wood waste, sludge and peat.

Key words: CO_2 cycle, CO_2 neutral, neutral biomass, black liquor, bark, wood rejects, BFB (bubbling fluidised bed) and CFB (circulating fluidised bed) boilers, fuels properties, GHG (greenhouse gas) emissions, emission factors, methane, nitrous dioxide, SO_2 , H_2S , NO_x , TRS (Total Reduce Sulphur), CO.

fuel for pulp and paper mills (including cellulose manufacturing for textile purposes). Mills have switched from coal and bunker oil (which are heavy CO_2 sources) to clean natural gas or renewable biomass [1].

The pulp and paper industry is an energyintensive but energy-efficient industry. Its energy can represent up to 25% of manufacturing costs. Many pulp and paper mills generate more than half of their energy needs from climate-neutral biomass fuels recovered from industrial waste and process streams (Table 1). Energy-rich biomass, derived from wood chips, bark, sawdust and pulping liquors, and recovered from the harvesting and manufacturing process, is the result of atmospheric carbon dioxide amassed by trees during growth and transformed into organic carbon substances ($C \Rightarrow CO_2 \Rightarrow C$). Carbon dioxide emissions from biomass combustion as neutral are not counted in

Table 1. Shares of energy carriers in primary energy consumption in the member countries of the Confederation of European Paper Industries (CEPI) [2].

Fossil and non-fossil fuels	Share, %
Biomass	50
Gas	34
Oil	9
Coal	5
Other fossil	1
Other	1

the inventory. This definition results from the Protocol of Intergovernmental Panel on Climate Change (IPCC): "...Biomass consumption is assumed to equal its regrowth... In other words, biomass is equal to net zero because all carbon accounting is conducted within the forestry module." [2]. A wide definition of biomass covers all biodegradable products, waste and residues from the forestry and related industries.

Preface

The forest products industry plays an important and complex role in the global closed-carbon cycle. Forests which supply the industries' primary raw material also store significant amounts of carbon and provide much-needed products for society. Moreover, forests provide multiple environmental, social and economic benefits, and the industry's emissions should be viewed in the context of the overall forests products' carbon cycles. In Canada, over the past decade the forest products sector has moved biomass or wood waste to the top of the country's renewable fuels list. This greenhouse gas-friendly fuel now provides 57% of

 Table 2. Types of biomass wastes in the pulp and paper industry.

Definition	Sources of biomass wastes	Comments
Black liquor	Chemical pulp production (kraft technology)	Energy and chemicals recovered
Bark and wood rejects	Chemical and semi-chemical pulp as well as ground wood production	Energy, soil conditioner
Rejects of screening and cleaning processes	From process in: virgin pulp production waste paper treatment paper stock production	Landfill, energy, other solutions
Mechanical-chemical sludge	From white water treatment and mechanical and chemical effluent treatment	Landfill, energy, other solutions
Biological sludges	From biological effluent treatment	Landfill, energy, other solution are needed
Mixed sludges	From different sources	Landfill, energy, other solutions
De-inking sludge	From the de-inking processes of waste paper fibres	Landfill, energy, other solutions

Description of Biomass in Paper Industry [6-9]

The main grades of biomass wastes arising in the pulp and paper industry are grouped in Table 2.

Black liquor

The first step in the production of chemical pulp is the separation of the wood fibres from the lignin by cooking wood chips in a digester with a mixture of lye sodium sulphide. The cooking process is stopped when about 5% of the lignin remains. The chemicals used in the process circulate in a closed recovery system. The digester produces 'black liquor', i.e. a mixture of lye and organic substances removed from the wood chips. Following evaporation, this liquor is sent to the soda recovery boiler, and the organic material is burned as a fuel. The remaining, noncombustible chemicals dissolved in water is called 'green liquor'. It is converted to lye and called 'white liquor'. In the chemical recovery process, some 98% of the chemicals are recycled in the system.

Solid biomass

Biomass fuels to be burned can be divided into two categories: the mill's own waste fuel, and purchased primary fuels. Waste fuels like bark, water treatment sludges (primary and secondary) and de-inking sludge are all burned and utilised in bubbling fluidised bed (BFB) or circulating fluidised bed (CFB) boilers. However, the combined wood waste and sludges generated at the mill are not sufficient to meet the mill's steam demand, partly because of their low heating (calorific) value resulting from their high moisture and ash content (see Table 3).

Purchased supplementary fuel (peat, oil) is therefore added almost continuously,

for firing with the mill-generated wood waste and sludge. In addition, some wood waste and wood chips are brought from saw-mills and other surrounding industries.

Of the wood-type residues, it is the bark that is of significant in the pulp and paper industry. Bark has a high calorific value (18 MJ/kg), and is therefore an ideal energy source. Bark does not contain any pollutants to speak of and has a low inorganic content. The only factor to be considered is size, because of the risk of coarse inorganic matters being introduced into the process. Bark removed in the barker accounts for 5-10% of the weight of the log. Another 5% is lost in the chipper. Wood waste and sawdust are typically generated in the wood processing industry.

Rejects

Rejects comprise residues from various sources in the pulp and paper production. With the continual increase of world paper consumption, more and more waste paper of poor or unknown quality is entering the new material market. Sources of waste paper fibres (secondary fibres) are as follows:

- Mixed varied paper quality, boxboard cuttings and mill wrappings.
- News old newspapers, over-issue news, and ground wood and container plant cuttings.
- Pulp substitutes grades of white coloured kraft, white and semi-bleached sheets/cuttings, tab cards and trims and unprinted grades.
- Purchased de-inked de-inked white and coloured ledger, computer printout, ground wood, coated book and bleached sulphate sheet and cuttings.
- Other lightweight coated grades.

For this part of the waste paper stream, a solution other than fibre recovery should also be found. Although these residues are still largely dumped on landfill sites, a trend towards thermal disposal is crystallising. Due to their polyvinyl chloride (PVC) content, elevated chlorine concentrations may occur in the coarse fraction, which has to be taken into consideration in the plant design, especially with regard to dioxin emission.

Sludges

A distinction has to be made between de-inking sludges and the primary (chemical) and secondary (biological) sludge of the waste water treatment. The greatest increase in volume of the de-inking sludges is observed. Every tonne of recovered fibre creates 200 to 400 kg of sludge (dry weight). This sludge contains fibre, mineral fillers such as clay, calcium carbonate, ink and adhesives from the converting processes. The environmental fate of some of these components still remain insufficiently known.

By contrast, the sludge from paper mill waste-water treatment plants is much easier to dewater than sludge from municipal sewage treatment. As a rule of thumb, it can be stated that the higher the fibre content, the higher the dry substance content that can then be obtained. Sludge handling methods are available for all types of sludge produced by the present effluent treatment method. These include primary fibre and biological sludges, with a high content of organic matter, chemical sludge with a high content of organic matter, and chemical sludge with a high ash content. De-inking sludge is usually handled with the effluent sludges. Sludge handling includes dewatering which makes it easy to transport and dispose of. Maximum mechanical dewatering of the sludges prior to incineration is essential.

Table 3. Comparison of typical fuel properties [7].

Heat value and components		Unit	Bark	Mech. & biol. sludge	De-inking sludge	Peat	Coal
Heat Value - NHV as received		MJ/kg	5.9	4.2	2.8	9.2	24
Moisture	average	%	60	63	58	50	12
woisture	range	%	50 - 65	60 - 67	55 - 60	40 - 60	8 - 20
Ash (dry basis)	average	%	3	20	50	5	14
	range	%	2 - 5	15 - 25	45 - 55	3 - 8	8 - 15
	С	%	50.6	33.7	19.0	57.1	71.6
	Н	%	5.9	4.4	2.4	6.2	4.9
Elementary	S	%	-	0.3	0.05	0.2	0.6
analysis (dry basis)	N	%	0.5	0.7	1.0	1.9	1.9
	0	%	40.2	41.2	27.4	29.6	7.0
	CI	%	-	<0.01	-	-	-
	Ash	%	3	20	50	5	14

Estimation of GHG Emissions

The 1997 Kyoto Protocol set very ambitious targets for reducing energy consumption and the emission of greenhouse gases (GHG). Emissions included in the inventory were as follows:

Carbon dioxide emission from fossil fuel combustion. CO₂ emissions include those from production processes, as well as the use of companyowned vehicles and other equipment. Emissions are estimated using widelyaccepted emission factors, which are based on the carbon content of the fuel.

- Methane and nitrous oxide emissions from combustion processes, which are estimated using emission factors. Emissions of CH₄ and N₂O are usually very small compared to those of CO₂, and some inventory protocols do not address such emissions.
- Greenhouse gas emissions from mill landfills and wastes water treatment plants, which are estimated using mill-generated data, and are consistent with methods suggested by the Intergovernmental Panel on Climate Change (IPCC).

In early 2001, the pulp and paper industry, working through the International Council of Forest and Paper Associations (ICFPA), began a project to develop an international methodology for estimating greenhouse gas emissions. The calculation tools were recognised by governments as well as by domestic and international organisations, establishing protocols and developing calculation techniques as the appropriate tools for calculating greenhouse gas tools for the pulp and paper mills. The calculation tools are available at the following websites: www.ncasi.org, www.icpa.org, www.wri.org.

Greenhouse gas emissions (GHG) factors [10]

Although emission of CO₂ from biomass burning is almost excluded (depending on the use of fossil fuel) from GHG inventories, the methane CH₄ and nitrous oxide N₂O from biomass burning are sometimes included. If a company has reliable site-specific data allowing it to estimate CH₄ and N₂O emissions from biomass combustion, it should use this data. Otherwise, it will be necessary to use the most appropriate emission factors available. Unfortunately, there is little data on CH₄ and N₂O emissions from biomass boilers and recovery furnaces (see Table 4). The following table summarises the available information on methane and nitrous oxide emissions from biomass boilers. The variability in the data reflects the many different types and ages of boilers tested, the operating conditions, control equipment and fuel characteristics.

Where an emission factor description shown in Table 4 matches the conditions at a mill, the company may want to **Table 4.** Emission factors for CH_4 and N_2O from biomass combustion [10]; a) - converted from GCV to NCV assuming a 5% difference, GCV - gross calorific value, NCV - net calorific value, NCV=0.95 GCV; b) - excludes one very high number associated with low oxygen-high carbon monoxide conditions; c) - based on the heat content of 20 GJ/t dry solids, d) - based on the liquor heat content of 13.3 GJ/t dry solids.

Combustion unit	Emission Factor Description	CH ₄ kg/TJ	N₂O kg/TJ
	Wood, wood waste and other biomass and wastes	30	4
	Uncontrolled emissions from wood-fired stoker boilers	15	-
	unit Emission Factor Description unit Wood, wood waste and other biomass and wastes Uncontrolled emissions from wood-fired stoker boilers Average from wood residue combustion Average for circulating fluidised bed boilers (CFB) burning peat or bark Average for bubbling fluidised bed (BFB) boilers burning peat or bark Pre-1980 wood residue-fired stoker boilers sampled ahead of control devices Pre-1980 wood residue-fired stoker boilers sampled after wet scrubbers Wood fired boiler Wood as fuel Wood waste Data dispersion of emission factors Recovery furnace Recovery furnace Recovery furnace - Black liquor	9.5 ^{a)}	5.9 ^{a)}
Unit		1	8.8
		2 ^{b)}	<2
		8.2 ^{a)}	-
		2.7a)	-
	Wood fired boiler	41c)	3.1 ^{c)}
	Wood as fuel	24 ^{c)}	3.4 ^{c)}
	Wood waste	30	5
	12	4	
	Data dispersion of emission factors	1 - 40	1.4 - 75
	Pre-1980 wood residue-fired stoker boilers sampled after wet scrubbers Wood fired boiler Wood as fuel Wood waste Median emission factors for wood waste Data dispersion of emission factors Recovery furnace	<1	<1
_	Recovery furnace - black liquor	2.5 ^{d)}	-
	Black liquor	30	5
Turnaces	Median emission factors for wood waste	2.5	2.0
	Data dispersion of emission factors	1 - 17.7	1 - 21.4

Table 5. Emission factors for biomass fuels (according to the Japanese Ministry of the Environment) [10].

Combustion unit	Fuel	CH₄	N ₂ O		
Dailan	Black liquor	0.033 kg/ton dry solids	-		
Boiler	Wood	0.82 kg/ton dry solids	0.063 kg/ton dry solids		
Waste combustion	sludge from mill at 50% moisture	0.0097 kg/wet ton	0.45 kg/wet ton		

Table 6. Comparison of measured N_2O and CH_4 emissions with other emission factors (maximum values in brackets) [10]; 1) IPCC - Intergovernmental Panel on Climate Change.

Emission factor or emission	Unit	CFB- boilers	BFB- boilers	PC- boilers	Gas turbine plants	Recovery boilers	Heavy oil boiler
N ₂ O national factor	mg/MJ	30-39	27-30	2	1	1.4	2
N ₂ O IPCC- factor ¹⁾	mg/MJ	4	4	1.4	0.1	4	0.6
N ₂ O measured	mg/MJ	<22.6	<2	<1	<3	<1	<1
CH ₄ national factor	mg/MJ	11.1-22.4	14.5-21.3	4	3	1	8
CH ₄ IPCC- factor	mg/MJ	30	30	1	1	30	3
CH ₄ measured	mg/MJ 1		1 - 3 (72)	<1	<1 (26)	1	<1

Table 7. Data on methane emissions from wood residue boilers. Data has been converted from GCV to NCV, assuming a 5% difference.

Reference	Beiler Tyree	Number	Number Methane emissions, mg				
Reference	Boiler Types	of boilers	Min.	Max.	Average		
NCASI Air Technical Bulletin 109, Sept. 1980 (NCASI 1980)	1952 to 1976 vintage, stoker- fed boilers, sampled before particulate control devices	4	<1	20.4	8.2		
NCASI Technical Bulletin 455, April 1985 (NCASI 1985)	1952 to 1981 vintage, stoker-fed boilers, all but one sampled after wet scrubber	6	<1	4.5	2.7		

select that emission factor for estimating mill emission. In many cases, however, because of the ranges in emission factors and the limited ability to match emission factors to boiler design, operating conditions and fuels at this time, it is reasonable to use the median emission factors shown in Table 4.

The Japanese Ministry of Environment includes CH₄ and N₂O in its inventories, and uses the emission factors for biomass fuels seen in Table 5 [10]. Perhaps the most ambitious recent study was undertaken in Finland by Forum Power and Heay Oy. The report on Methane and Nitrous Oxide Emission in Finnish Energy Production was published in May 2001, and contains new data from 11 boilers and 2 recovery furnaces. Six of the 11 were fluidised bed boilers (CFB, BFB) burning biomass and/or peat, two were pulverised coal-fired boilers (PC), two were combined cycle gas turbine facilities, and one was a heavy oil boiler. In addition, the report summarises existing emission factors, noting that most of them had been proven to be unreliable (see Table 6).

In the late 1970s and early 1980s, the

Table 8. Gas emission factors for lime kiln (according to authors' data).

Factor sources	BAT (Best Available Techniques) see Table 10. Reference Document (BREF) [11]	Pulp and Paper Mills in Poland
SO ₂ sulphur dioxide oil burned process	 without condensing gases 3-30 mg/m³ with condensing gases 15-900 mg/m³ 	 limit <474 mg/m³ result 16-49 mg/m³
H ₂ S sulphured hydrogen	 standard <50mg/m³ temporary can be higher 	 limit <164 mg/m³ result 3-6 mg/m³
NO _x as NO ₂ Nitrogen peroxide	 oil burned 240-380 mg/m³ gas burned 380-600 mg/m³ 	 limit <800 mg/m³ result 99-175 mg/m³
TRS, Total reduced sulphur	-	 limit 2-7 mg/m³ result 0.12-0.25 kg/m³
CO carbon monoxide	-	 limit <348 mg/m³ result 18-164 mg/m³

 Table 9. Gas emission factors from recovery boiler (according to authors' data).
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Factor sources	BREF [11]	Pulp and Paper Mill in Poland
SO ₂	 without scrubber and the dry matter content in the black liquor 63-65%, 100-800 mg/m³ but at the 72-80%, 10-100 mg/m³ with scrubber and the dry matter content in the liquor 63-65%, 20-80 mg/m³ 	 limit <1068 mg/m³ result 20-420 mg/m³
H ₂ S	 standard time 90 percent <10 mg/m³ temporary can be higher 	 limit <9.02 mg/m³ result 1.0-3.4 mg/m³
NO _x as NO ₂	standard 100-260 mg/m ³	 limit <317 mg/m³ result 73.5-133.7 mg/m³
TRS	Result 0.5-1.3 kg/m ³	 limit <5.56 mg/m³ result 0.3-0.8 mg/m³
CO carbon monoxide	-	 limit <298 mg/m³ result 12-166 mg/m³

CO₂ emission from fossil fuel:

 (0.8×10^3) TJ/y × (77.4)t CO₂/TJ = 61,900 t CO₂/y

CH₄ emissions:

National Council for Air and Stream

Improvement (NCASI) developed meth-

ane data, the results of which are shown

in Table 7. The methane emissions fall

within the range of the newer Finnish

data, but tend to be a little higher on aver-

age. This is perhaps not surprising, since

the NCASI data were generated on older-

technology boilers during a period when

there was not as much emphasis on com-

bustion control. Nevertheless the data are

fairly consistent, and in any event, far

Example of GHG emission calculation

A mill has a 250,000 kg per hour steam

circulating fluidised bed (CFB) bark boil-

er. In one year, the boiler burns 6.9×10^6

GJ of bark and 0.8×103 TJ of residual

fuel oil. Because the boiler receives

supplemental fossil fuel, it is necessary

to estimate the CO₂ from the fossil fuel

use, and the CH₄ and N₂O emissions on

the basis of the total firing rate. The mill

decides to use the emission factor for

residual oil of 77.4 ton CO₂/TJ, and the

average emission factors are 1 kg CH₄/

TJ and 8.8 kg N₂O/TJ. Unburned carbon

from the residual fuel oil is assumed to

be negligible.

lower than the IPCC's default value.

for pulp and paper mill [10]

total heat input = (6.9×10^{6}) GJ/y + (0.8×10⁶)GJ/y = (7.7×10³)TJ/y (7.7×10³)TJ/y × 1kg CH₄/TJ = 7,700kg CH₄/y = 7.7t CH₄/y

N₂O emissions:

total heat input = 7.7×10^3 TJ/y (7.7×10^3)TJ/y $\times 8.8$ kg N₂O/TJ = 67,800kg N₂O/y

Other Gas Emission Factors

The most severe air pollution problems are usually associated with kraft pulp mills. The recovery boiler is a potential source of sulphur dioxide, but the emission level is generally below the nuisance level. SO_2 is the principal emission from sulphide mills, but this is because the odour threshold is about one thousand times higher for SO_2 than it is for reduced sulphur gases.

The major sources of TRS emission are digester blow & relief gases and multiple effect boiler exhaust gases. Over the last two decades, evaporator and recovery boiler manufacturers have improved their designs to achieve higher solids liquor for firing and better mixing within the combustion zone. These improvements serve to reduce TRS emissions. Relatively little attention has been directed toward the control of nitrogen oxides, which are byproducts of high-temperature combustion processes. However, both NO and NO₂ are known to have a photochemical impact on the atmosphere.

Fortunately, with respect to recovery boilers and lime kilns, it appears that the formation of nitrogen oxides can be adequately controlled by operating these combustion units with a minimum flame temperature and limited excess air. The emission factors of gases from the lime kiln and the recovery boiler are presented in Tables 8 and 9.

The development of fluidised-bed combustion technology seems to have very good perspectives for utilising low-quality fuel such as bark and other wood wastes. This kind of biomass has a relatively low content of sulphur (<0.1%), but SO₂ emission during its combustion is much lower than in the case of coal combustion. The emission factor of gases from bark and wood waste combustion in the BFB boiler are collected in Table 10. NO_x emissions in the BFB boiler can be reduced by maintaining the low combustion temperature between 700°C and 900°C. Another significant factor affecting the level of NO_x emission is the nitrogen content in the fuel. A long remain time of the fuel particles in the combustion chamber, together with proper mixing of the secondary air with volatile products, yields a low CO emission. Table 11 shows the results of heavy metal emission measurements made on the BFB boiler when burning the fuel mixture of sludge, wood waste and peat.

Most important for achieving low heavymetal emissions is having an effective dust cleaning system. NO_x emissions in wood/sludge combustion typically vary between 200 and 300 mg/Nm3, depending on the fuel's nitrogen and the firing conditions. Nitrogen content is relatively high in waste fuels, typically 1.5 to 2% (drying basis), and the fuel heat value is low. The N2O level only reaches several parts per million. Normally CO is below 100 and 200 mg/Nm3. Appropriate amounts of excess air are used. An important factor in keeping the CO level low with small excess air is the even fuel feed rate. Dioxin emission are 0.005 and 0.02 mg/Nm³, which is well below the common combustion limit of 0.1 mg/Nm³ in Europe for municipal waste incineration. The best combustion results and controllability are achieved when fuel moisture does not exceed 60%. The boiler can be operated at a 20% to 25% excess air level, thus maintaining the emission levels of unburned carbon CO and NO_x.

Summary

- In the pulp and paper industry, energy-rich biomass is obtained from wood chips, bark, sawdust and pulp-ing liquors.
- Combined wood waste and sludges generated at the mill are insufficient for combustion demand, because of the low heating value resulting from their high moisture and ash content.
- Carbon dioxide emission from biomass combustion as neutral is not counted in the GHG inventory.
- GHG emissions included in the inventory contain carbon dioxide CO₂, methane CH₄ and nitrous oxide N₂O. The last two are usually very small compared to CO₂, and some inventory protocols do not include them.
- Emissions are estimated by using

Table 10. Gas emission factors for bark combustion in BFB boiler [13].

Factor sources	Best Available Techniques (BAT), BAT Reference Document (BREF)	Power plant OKF-40 BFB boiler in Poland	BFB boiler in Pulp and Paper Mill in Poland
SO ₂ sulphur dioxide	5-15 mg/MJ of fuel coming into boiler	75-128 g/GJ of fuel coming into boiler	 limit <90 mg/m³ result 30-70 mg/m³
NO ₂ nitrogen peroxide	40-100 mg/MJ	90-119 g/GJ	 limit <370 mg/m³ result 270-370 mg/m³
CO carbon monoxide	-	10-350 g/GJ	 limit <200 mg/m³ result 33-82 mg/m³

Table 11. Heavy metal emissions [µg/MJ].

Test number	Hg	Cd	Cr	Cu	Ni	Pb	Zn	Sn	Co	Ash	Total
1	0.3	0.7	19.7	14.6	14.2	28.7	6.9	4.5	2.7	3.3	95.6
2	0.3	8.5	19.8	46.4	9.7	45.9	16.8	11.6	2.1	3.3	164.4
Average	0.3	4.6	19.8	30.5	11.9	37.3	11.9	8.0	2.4	3.3	130.0

widely accepted factors which are based on the carbon content of the fuel.

- GHG emissions from mills, landfills and waste water treatment plants are estimated using mill-generated data, and are estimated with methods suggested by the IPCC (www.ncasi.org).
- Though CO₂ from biomass burning is excluded from GHG inventories, depending on the use of fossil fuels, CH₄ and N₂O are sometimes included.
- Little data on CH₄ and N₂O emissions from biomass boilers and recovery boilers is available in world literature.
- The variability in the data reflects the many different types and gases of boilers tested, the generating conditions, control equipment and fuel characteristics.
- Other important gas emissions in the pulp and paper industry include SO₂, H₂S, NO_x, TRS and CO. Such a gas composition is descended from lime kiln and recovery boilers. Bark combustion in BFB boiler produces only SO₂, NO₂ and CO in small quantities.

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