SUOMEN SOODAKATTILAYHDISTYS
FINNISH RECOVERY BOILER COMMITTEE

Suomen Soodakattilayhdistys ry

Lime Kiln alternative fuels and emissions/ Meesauunin päästöt erilaisilla polttoaineilla

Henna Hietanen, Jens Kohlmann
AFRY Finland Oy

17.11.2021

Raportti 4/2021

(16A0913-E0211)

Report

Author
Hietanen, Henna
Mobile
+358400883141
E-mail
henna.hietanen@afry.com

Date
13/09/2021
Project
101014962-001

Client
Finnish Recovery Boiler Committee

Lime Kiln alternative fuels and emissions

The pulp and paper industry is the fourth most energy-intensive in Europe. Reducing greenhouse gas emissions in the industry has a big impact on Europe's transition to a low carbon economy. This common goal has increased the use of renewable energy sources in pulp mills across Europe.

In pulp mills fossil fuels are primarily used in lime kiln operation, they are also used during upsets, start-up and shut-down, as well as sometimes for safety reasons to secure non-condensable gas combustion. In those cases the lime kiln is typically during normal operation the only source of fossil fuel-based emissions. Substituting fossil fuels with renewable fuels produced at the mill, would make typical kraft pulp mill operations possible to operate fossil fuel-free. Some of the renewable fuels originate from side streams of mill processes which improves even the self-sufficiency of mill.

This study compared the production and use of various renewable fuels in six Finnish pulp mills to define the effect in lime kiln operation, emissions and possible indirect environmental impacts. The study is entirely based on interviews and the data given by the mill representatives participating in the study.

AFRY Finland Oy

Contents

1 Lime kiln operation and fuel preparation

The modern kraft pulp mill uses fossil fuel for the operation of the lime kiln. Lime kiln is the biggest user of fossil fuels in kraft process and the only part of the pulp mill that needs substantial purchasing of fuel.

Common lime reburning kiln fuels are natural gas and fuel oil. In Finland at least these fuels have been used in lime kilns to replace fossil fuels:

- Wood powder
- Lignin
- Combustible gas from gasification process
- Hydrogen
- Turpentine
- Methanol
- Stripper off-gas (SOG)
- Non-condensable gas (NCG)
- Tall oil
- Tall oil pitch

The main alternative energy resources studied in this report are lignin, tall oil pitch, wood powder and combustible gas from bark gasification process. Each of these techniques has advantages and disadvantages.

Table 1 below shows basic data about the studied lime kilns. It shows that the kilns represent very different capacities, dimensions and ages. Only two of the kilns are originally designed for combustion of 100 % biofuels during normal operation. Other kilns have been upgraded to their present facility. Most of the upgrades towards more biofuel adequate lime kilns have been made during the 21st century. Thus, all the upgraded mills have over five years of operation experience with additional fuels.

Table 1 Basic lime kiln data from studied pulp mills.

Delivery Year	Capacity	Diameter	Length	Cross section	Volume	Surface area	Product/ Cross s.	Product/ Volume
	tCaO/d	m	m	m^2	m^3	m^2	$t/m^2,d$	$kg/m^3,h$
1951	150	3	70	5.3	372	572	28.3	16.8
1961	150	3	70	5.3	372	572	28.3	16.8
1991	450	4.15	120	11.0	1325	1414	40.7	14.1
1996	650	4.5	125	13.2	1650	1610	49.2	16.4
1998	575	4.25	115	11.6	1339	1391	49.4	17.9
2004	750	4.75	138	14.9	2051	1886	50.5	15.2
2017	1200	5.5	140	20.4	2860	2243	58.7	17.5

Table 2 below shows the higher heating values used in this report. For Combustible gas there is no value available. All the values are given by the mills.

Table 2 Used higher heating values

Fuel	higher heating value (HHV)	Unit
Heavy Fuel oil	41	MJ/kg
Natural Gas	36	MJ/m^3
Tall oil Pitch	37	MJ/kg
Lignin	28	MJ/kg
Wood powder	19	MJ/kg
Combustible gas	Not available	MJ/m^3
Methanol	20	MJ/m^3

Table 3 below shows statistics about lime kiln operation in six different Finnish pulp mills. There have also been listed causes for lime kiln operation disturbances and operational challenges.

Table 3 Statistics about lime kiln operation and equipment.

	A	B	C	D	E	F
Informed average amount of used alternative fuels	60 %	90 – 95 %	15 – 25 %	100 %	35 %	100 %
Calculated amount of alternative fuels from collected data	57 %	-	23 %	100 %	41 %	100 %
Used main alternative fuels at the moment	Wood Powder (Methanol)	Combustible gas	Tall oil pitch	Tall oil pitch (Methanol)	Lignin	Combustible gas (Tall oil pitch, Tall oil)
Introduction of alternative fuels in lime kiln	2015	2012	2014	2004	2014	2017
Lime losses from lime cycle	5 %	3 – 5 %	2,5 %	2 – 3 %	7 %	3 – 5 %
Biggest operational disturbances / challenges	Wood powder firing disturbances, lime dusting	Lime ring in kiln, flue gas duct blockages	Lime ring in kiln, brickwork damages	Lime cooler blockages	Lime ring in kiln	Lime cooler blockages, Lime ring in kiln
Low-NOx burner	No	No	Yes	No	No	No
Electrostatic precipitator	Yes	Yes	Yes	Yes	Yes, since 2020	Yes
Flue gas scrubber	No	Yes, since 2020(*	No	No	Yes, Alkali	No

(* Flue gas scrubber was purchased due to high dust emissions

1.1 Lignin

Lignin has a good heating value and is a good alternative fuel for lime kiln. Nevertheless, it needs an additional process to be separated from black liquor. First, the black liquor pH is lowered. When the pH drops, lignin is precipitated and separated from the liquor. After the separation lignin is washed, dried and stored. Lignin lean black liquor and wash water are returned to evaporation plant. Before combustion in lime kiln, lignin needs to be pulverised. The pulverised dry lignin needs to be stored and conveyed to the kiln in an environment with a low oxygen level to prevent dust explosion.

The usage of lignin as a lime kiln fuel has a higher impact on the rest of the mill than any other additional fuel presented in this report. The process uses sulphuric acid to purify the lignin. Most of the sulphur in the acid is returned to the mill. This affects the pulp mill sulphur balance and increases the need for sulphur removal. Depending on the amount of removed lignin, the black liquor heating value decreases. In mills that have thermally limited recovery boilers, lignin removal from black liquor has become an attractive option for increasing pulp production by allowing more black liquor to be processed through the boiler. In the case of lignin extraction from black liquor, the amount extracted lignin must be examined on a case-by-case basis.

In the example mill in this report, the lignin production is mainly targeted to sales for customers. In this example mill, lignin usage in the lime kilns is forecasted to be decreased. The main fuel used parallel with lignin is natural gas. This might also be the common trend in the future, lignin is more likely to be sold out than used as a lime kiln fuel.

Combustion of lignin is said to be very sensitive for disturbances. For this reason, there must also be support fuel available in order not to have too many disturbances in lime kiln operation. The maximum lignin feed is about 70 % from the needed lime kiln fuel power. Normally, the lignin ratio is based on available lignin. With powdered fuels, challenges occur also in storing and conveying. in the fuel storage, conveying and feeder systems dry powdered fuel increases the risk of dust explosion and blockages.

The amount of used lime kiln fuel (lignin and natural gas) as megawatts per day in year 2020 are shown in Chart 1 in order of magnitude. This chart is based on yearly defined data of one mill. Mill down time has been taken out. This Chart presents the amount of used lignin compared to total used kiln fuels. Also Maximum Continuous Rating (MCR) has been calculated and presented for total fuel and for lignin usage. For the total fuel usage calculated MCR is 23.3 MW/d and for lignin usage 16.3 MW/d.

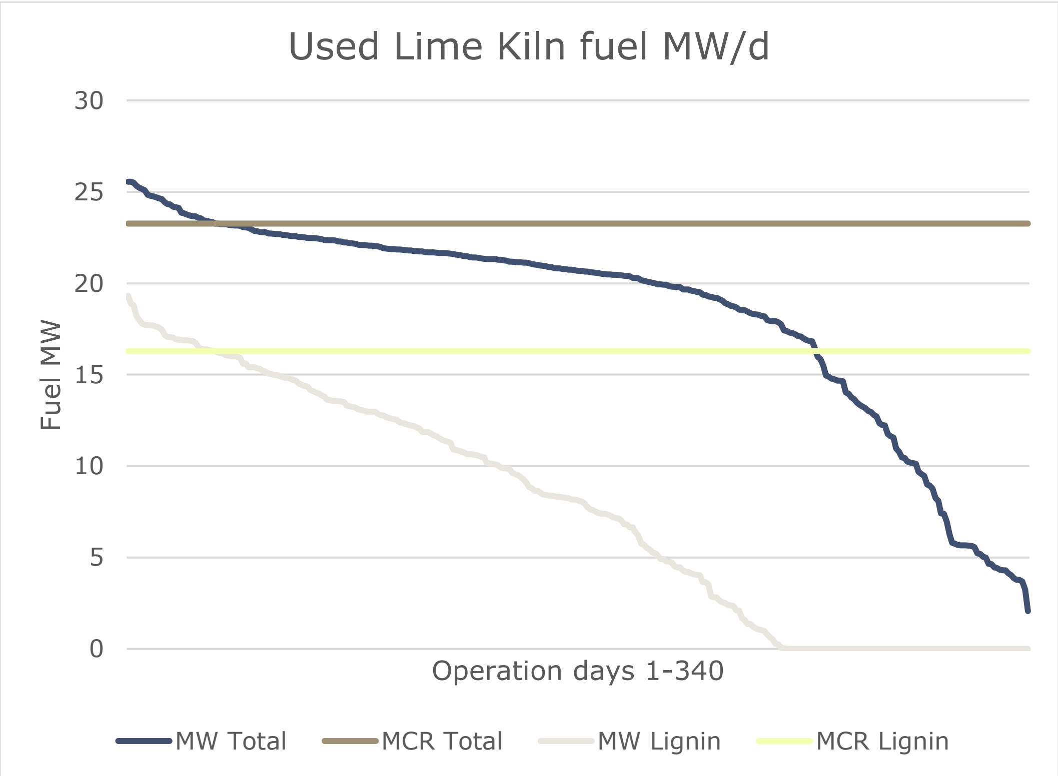

Chart 1 Used Lime kiln total fuels, lignin in MW/d

1.2 Tall oil pitch

Tall oil pitch can be seen as heavy fuel oil from an operational point of view. Generally, tall oil soap obtained as a side product has been refined to crude tall oil, which is sold to tall oil distillers. In distillation, tall oil pitch is first removed and it is often transported back to pulp mill to be used as a fuel. The major drawback is that, there is not enough tall oil pitch in pulp mill mass balance to produce all needed lime kiln fuel. There must always be other fuel sources available.

In the past, tall oil was more commonly used as an alternative fuel than tall oil pitch. Nowadays in Finland its use as a fuel has nearly stopped while excise tax is payable on tall oil and selling tall oil to distillers is very profitable.

Lime kiln can be operated purely with tall oil pitch or it can be used as a sub fuel. While combining tall oil pitch with natural gas or combustible gas, the different radiating heat between the fuels can cause troubles with too high temperatures in combustion zone. Without controlled combustion, this might lead to brickwork damages in the long run.

The amount of used lime kiln fuel (tall oil pitch and natural gas) as megawatts per day in year 2020 are shown in Chart 2 in order of magnitude. This chart is based on yearly defined data of one mill. Mill down time has been taken out. This Chart presents the amount of used tall oil pitch compared to total used kiln fuels. Also Maximum Continuous Rating (MCR) has been calculated and presented for total fuel and for tall oil pitch usage. For the total fuel usage calculated MCR is 46.0 MW/d and for tall oil usage 22.2 MW/d.

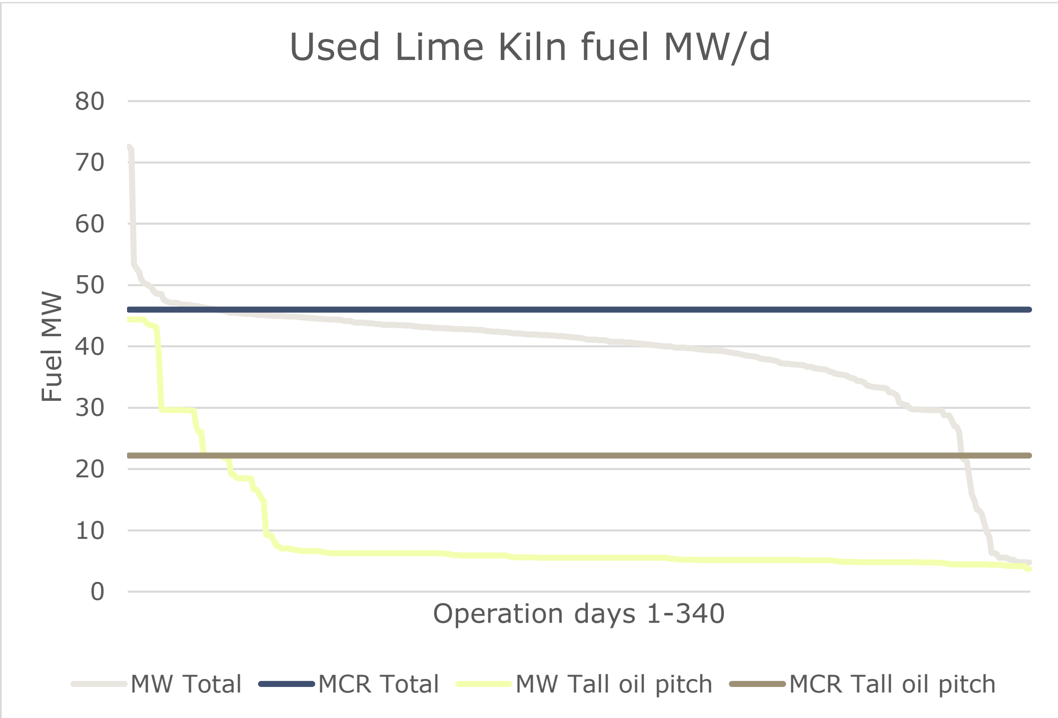

Chart 2 Used Lime kiln total fuels, tall oil pitch in MW/d

1.3 Wood powder

When wood chips and sawdust are used as a lime kiln fuel they need to be prepared as powder. The process starts from screening and drying of wood material. Most common dryers for biomass are belt drying systems. These dryers can utilize secondary heat from other process areas in addition to low pressure steam. The lower the moisture level in the dried wood the more efficient the kiln operation and higher kiln capacity. After drying, the wood material is ground with a hammer mill to reach appropriate particle size.

Handling wood powder has several safety related issues to be concerned. Exposure to excessive amounts of wood powder may irritate the eyes, nose, and throat. For some it may even cause allergic reactions. With unstable operation wood powder can easily cause a fire or an explosion. Especially small stones or metallic objects in grinder might cause sparks. To minimize these operational disturbances and safety risks the example mill has a short maintenance stop for wood handling equipment once a month.

Another fuel handling difficulty is controlling the moisture level in the dried wood. Online moisture measurement is inaccurate and conventional moisture evaluation is too slow for constant operation and altering process conditions. Too moist powder can block piping and process equipment, stopping fuel feed to the lime kiln. These sudden uncertainties have forced the mill in question to use continuous support fuel to have less disturbances in lime kiln operation. The wood powder equipment also needs space near the lime kiln which is not possible to implement in all mill sites.

As a lime kiln fuel, wood powder needs a lot of control and balancing. The flow is an important factor in stable kiln operation. Unstable flow will cause a variation in heat input to the burning zone, impacting product quality. As an advantage, wood powder firing can be implemented to any mill. Wood availability is seldom a challenge for pulp mills. Wood material can also be imported from sawmill side streams.

The amount of used lime kiln fuel (wood powder and heavy fuel oil) as megawatts per day in year 2020 are shown in Chart 3 in order of magnitude. This chart is based on yearly defined data of one mill. Mill down time has been taken out. This Chart presents the amount of used wood powder compared to total used kiln fuels. Also Maximum Continuous Rating (MCR) has been calculated and presented for total fuel and for wood powder usage. For the total fuel usage calculated MCR is 48.6 MW/d and for wood powder usage 34.8 MW/d.

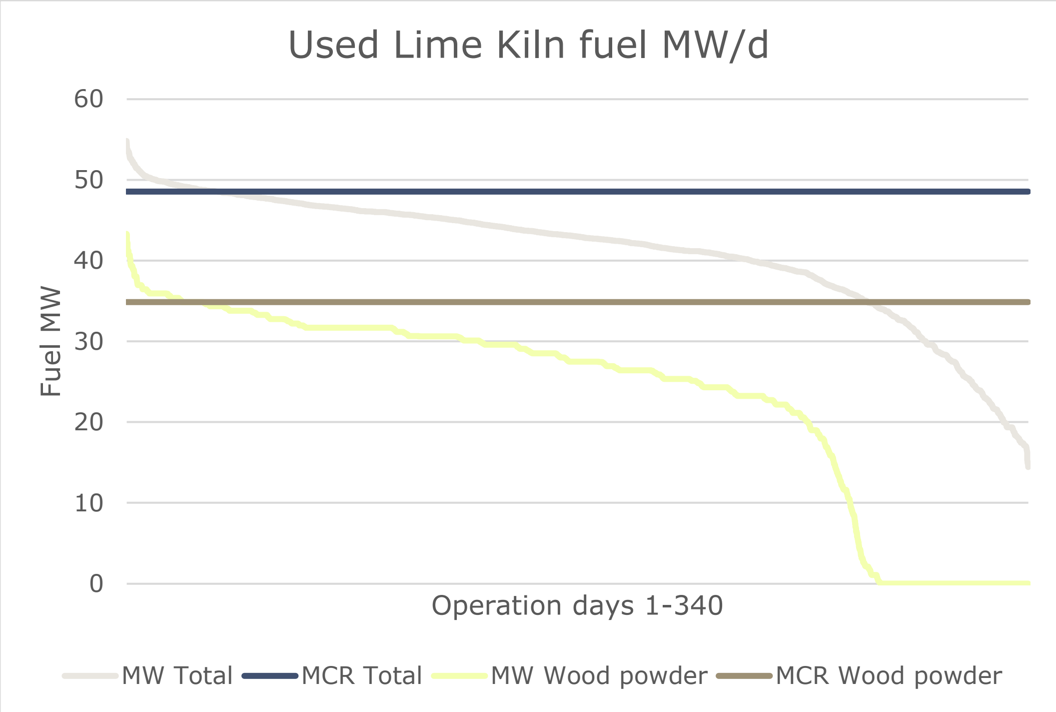

Chart 3 Used Lime kiln total fuels, wood powder in MW/d

1.4 Combustible gas from bark gasification

Good, stable operation is possible with gasification gas. With combustible gas lime kiln production capacity and heat rate are similar to those of natural gas. The advantage of gasification is that using the product gas is potentially more efficient than direct combustion of the original fuel because it can be combusted to produce higher temperatures. Additionally, various biomass fractions can be used, such as bark, chips and screening rejects.

Bark is commonly used in gasification process as a raw material. The bark is collected from the debarking and dried. Drying of biomass increases process efficiency and reduces the amount of flue gases. The dry bark is ground before actual gasification to reach even particle size. The drying and grinding processes develop easily a fire. Thus, fire safety must be taken into consideration. The process is also sensitive to impurities such as metals and sand.

Another downside is that the product gas cannot be stored. So, during process disturbances in fuel preparation substitutive fuel is needed for lime kiln. The fuel preparation equipment need also a significant amount of space near the lime kiln which is not possible to implement in all mill sites.

To produce stable product gas quality for lime kiln, biomass needs to be dried and ground to be uniform. Handling of biomass mixture during changing weather conditions and seasons is important. For example, bark quality changes a lot between different wood species and seasons. Quality changes in product gas can generate operational challenges in lime kiln such as increased NO_x emissions, unstable residual oxygen of flue gases, and temperature profile changes in lime kiln.

2 Non-process elements

The biofuels in the lime kiln can contribute with an increase of non-process elements (NPE). Table 4 below, shows amounts of magnesium, silicate and phosphorus measured from lime mud. To control the NPE levels all the mills have an option to take ESP ash out. The amount of make-up lime also indicates how much the mill actually has lime losses in lime cycle. The lime losses from lime cycle for each mill can be seen in Table 3.

Table 4 Measured NPEs in six different Finnish pulp mills

	A	B	C	D	E	F(*
	Wood Powder	Combustible gas	Tall oil pitch	Tall oil pitch	Lignin	Combustible gas
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
Magnesium	5 600	4 600	5 200	4 000	11 400	9 013
Silicate	470	1 155	-	1 200	500	-
Phosphorus	2 700	4 950	6 700	9 200	-	-

(From ESP ash*

The usage of alternative fuels for lime kiln has added a need for lime cycle opening. Wood residues contain a high amount of ash and non-process elements, such as aluminium, silica, phosphorus, and magnesium, which can be problematic in kiln combustion. Usually the extra purging is done at electrostatic precipitator. The impurities accumulate in the lime cycle causing ring formation in kiln and increased make-up lime demand. Also, a high level of ash may end up in the lime, which decreases lime quality.

Lignin consists of a relatively high level of sodium and sulphur. Also bark and wood powder have high NPEs. All of these could affect the lime kiln operation with ring and ball formation.

To find out an actual effect of lime kiln alternative fuels on non-process elements, would need more deeper study. These presented non-process element values are single measurements given by the mills.

3 Lime kiln emissions

The way emissions are monitored strongly influences the comparability of produced emission data. Depending on the purpose of monitoring and reporting (such as compliance control or environmental reporting) different reference periods (daily, monthly, yearly averages) are used. The degree of accuracy and completeness of different methods for the determination of emissions is also notable. Most of the emission values shown in Table 6 are directly measured, or calculated on the basis of direct measurements. All the values shown in Table 6 are presented as given by the mills, no alteration has been done by the author. One of the mills uses accredited private emission testing for authenticating of lime kiln emissions. These results of the single measurement have been compared with the monthly average given in the environmental permit limit value. Diffuse emissions are not taken into consideration.

All studied six mills have emission permits for lime kiln. Different limits for permits in year 2020 are shown in Table 5. All the mills have some permitted levels for sulphur, NO_x and dust emissions to atmosphere. Two of the mills have also limits for sulphur dioxide emissions. As shown in the measured emissions, none of the mills have trouble staying within the allowed limits. Most of the mills have had occasional troubles with dust and NO_x limits due to operational challenges. One of the mills does not have a continuous NO_x measurement.

Table 5 Lime Kiln emission permits 2020.

	A	B	C	D	E	F
Yearly average mg SO2/Nm3 (6 % O2)				200		70
Yearly average (TRS) mg S/Nm3 (6 % O2)	20	20	20	20	20	10
Yearly average mg NOx/Nm3 (6 % O2)	600	600		500		600
Yearly average mg dust/Nm3 (6 % O2)	30	50	50	60	30	30
kg SO2-S/ADt				0.07		
kg TRS-S/ADt				0.07		
kg dust/ADt		0.05				0.02
kg NOx/ADt		0.45	0.3		0.3	0.45

Table 6 Lime kiln measured emissions 2020.

	A	B	C	D	E	F
Yearly average mg SO2/Nm3 (6 % O2)	0.8	1.7	14.0	19.8	-	0.0
Yearly average (TRS) mg S/Nm3 (6 % O2)	2.7	3.1	6.0	1.7	-	0.0
Yearly average mg NOx/Nm3 (6 % O2)	187.0	263.1	290.0	88.9	-	297.4
Yearly average mg dust/Nm3 (6 % O2)	16.1	7.0	22.0	6.2	2.0	4.1
Yearly average mg CO/Nm3 (6 % O2)	10.4	16.0	25.0	0.0	-	7.0
kg SO2-S/CaO t	0.0080	0.0070	-	-	-	-
kg TRS-S/CaO t	0.0289	0.0080	-	-	-	-
kg dust/CaO t	-	0.0140	-	-	-	-
kg NOx/CaO t	-	0.6170	-	-	-	-

3.1 Specific emission

Specific emissions are defined as emissions of air pollutants (S, dust and NO_x) per Air Dry Ton of produced pulp. Four mills have given their annual average lime kiln specific emissions from last years. These values are shown in Table 7, which also shows as a comparison the best available technique (BAT) values from European Union.

Table 7 Lime kiln annual average specific emissions from years 2016 – 2020.

	2016	2017	2018	2019	2020	BAT(1
Lignin						
kg SO2-S/ADt	0.002	0.001	0.002	0.001	0.001	-
kg TRS-S/ADt	0.003	0.003	0.004	0.004	0.002	-
kg S/ADt	0.005	0.004	0.006	0.005	0.003	0.005 – 0.07
kg dust/ADt	0.07	0.04	0.07	0.04	0.02(2	0.005 – 0.03
kg NO2/ADt	0.12	0.11	0.13	0.11	0.11	0.1 – 0.2
Tall oil Pitch						
kg SO2-S/ADt	0.01	0.01	0.01	0.005	0.005	-
kg TRS-S/ADt	0.008	0.007	0.005	0.003	0.002	-
kg S/ADt	0.018	0.017	0.015	0.008	0.007	0.005 – 0.07
kg dust/ADt	0.12	0.007	0.009	0.011	0.01	0.005 – 0.02
kg NOx/ADt	0.14	0.13	0.12	0.17	0.16	0.1 – 0.2
Wood Powder						
kg SO2-S/ADt	-	0.002	0.002	0.003	0.003	-
kg TRS-S/ADt	-	0.016	0.006	0.006	0.013	-
kg S/ADt	-	0.018	0.008	0.009	0.016	0.055 – 0.12
Combustible gas from bark gasification						
kg S/ADt	-	-	0.00	0.00	0.00	0.005 – 0.07
kg dust/ADt	-	-	0.00	0.01	0.01	0.005 – 0.02
kg NOx/ADt	-	-	0.06	0.25	0.23	0.1 – 0.3

(1 European Commission, Integrated Pollution Prevention and Control (IPPC), Best Available Techniques (BAT) Reference Document for the Production of Pulp, Paper and Board. 2015

(2 Electrostatic precipitator since 2020

3.2 Measured emissions

As a comparison of alternative fuels and traditional fossil fuels, operational data from time before introducing alternative fuels to lime kiln would be needed. Unfortunately this type of data was not available or the data was not comparable to the current operation of the kiln. To have a proper comparison, a test run would be needed.

Based on the interviews, controlling the lime kiln emissions comes down to stable operation of the lime kiln; uniform fuel, handling of kiln temperature with combustion air and steady feed of fuel. Finding the balance in control might be difficult in the beginning while starting up the lime kiln with alternative fuels. Based on the common experience, difficulties to stay in current emission targets are more dependent on other process related issues than selected fuel.

When combusting fuels with high nitrogen levels, nitrous oxide enters the flue gas with the fuel. The formation of thermal NO_x is based on combustion temperature and the amount of excess combustion air. In some cases the experience shows that adding additional fuel with lower nitrogen levels than heavy fuel oil or natural gas, NO_x emissions can be lowered.

3.3 Indirect environmental impacts

Fuel preparation and purchase always create indirect environmental impacts. Lignin separation increases the use of chemicals at mill site, while wood powder creates reject and possible transportation must be calculated. In bark gasification, unreacted char, bed material and ash are removed from the bottom of the reactor with a discharge system. The used bed material can be sold as soil fertilizer or used as a soil improvement agent instead of landfill. In use of tall oil pitch, transportation between the mill and distillation plant plays a big role.

The combustion properties of the fuel have a major impact on the performance of the kiln. As shown in Table 2 fossil fuels and alternative fuels have different heating values and effect to lime kiln flue gas amounts. Thus, kiln fuel conversion may lead to increased fuel consumption and higher flue gas amount depending on fuel properties. The increase in flue gas flow can limit the production capacity of lime kiln. In cases where the kiln is already restricting the mill production, operating the lime kiln with alternative fuels may not be fully possible with existing equipment.

The requirement to open lime cycle more for NPE take out will increase the indirect environmental impact. Table 3 shows statistics about lime losses from lime cycle. In these six example mills the need for NPE take out varies from 2.5 % up to 7 % from the total lime amount. Opening the lime cycle increases the amount of needed make-up lime. External lime production has its own environmental impacts and those need to be evaluated together with the impact from increased transportation need. Also the lime that has been taken out of the process needs to be transported for reuse. Nevertheless, indirect environmental impacts of using alternative fuel must be examined on a case-by-case basis.

SUOMEN SOODAKATTILAYHDISTYS RY, RAPORTTISARJA

1/2020 Suomen Soodakattilayhdistys ry
Konemestaripäivä 23.1.2020, esitelmät
Sokos hotelli Lappee, Lappeenranta, UPM-Kymmene Oyj, Kaukaan tehtaat
(16A0913-E0201) 23.1.2020

2/2020 Suomen Soodakattilayhdistys ry
Sulakourujen materiaalitutkimus
Sanni Yli-Olli, Satu Tuurna, Aino Mäntyranta, Teknologian tutkimuskeskus VTT Oy
(16A0913-E0202) 18.2.2020

3/2020 Suomen Soodakattilayhdistys ry
Soodakattila-alan yhteistoiminta
Vuosikertomus 2019
(16A0913-E0203) 23.9.2020

4/2020 Suomen Soodakattilayhdistys ry
Biolietteen polton vaikutukset soodakattilassa ja lipeälinjalla
Lipeätyöryhmä
(16A0913-E0204) 29.9.2020

5/2020 Suomen Soodakattilayhdistys ry
Savukaasumäärän laskenta Suomen soodakattiloissa
Esa Vakkilainen, Kari Luostarinen, Simo Lyytinen, LUT-yliopisto
(16A0913-E0205) 29.9.2020

6/2020 Suomen Soodakattilayhdistys ry
Soodakattila-alan yhteistoiminta
Pöytäkirja. Vuosikokous 23.9.2020, Sokos hotelli Torni, Tampere
(16A0913-E0206) 23.9.2020

7/2020 Suomen Soodakattilayhdistys ry
Utilization of the forest industry sludges – trends and effects on modern mills
Katja Kuparinen, Satu Lipiäinen, Jussi Saari, Esa Vakkilainen, LUT-yliopisto
(16A0913-E0207) 16.12.2020

SUOMEN SOODAKATTILAYHDISTYS RY, RAPORTTISARJA

1/2021 Suomen Soodakattilayhdistys ry
Soodakattila-alan yhteistoiminta
Vuosikertomus 2020
(16A0913-E0208) 21.4.2021

2/2021 Suomen Soodakattilayhdistys ry
Soodakattila-alan yhteistoiminta
Pöytäkirja. Vuosikokous 21.4.2021, Teams-kokous ja AFRY Finland Oy, Vantaa
(16A0913-E0209) 21.4.2021

3/2021 Suomen Soodakattilayhdistys ry
Soodakattilapäivä 4.11.2021
Sokos hotelli Presidentti, Helsinki (16A0913-E0210) 4.11.2021

4/2021 Suomen Soodakattilayhdistys ry
Lime Kiln alternative fuels and emissions / Meesauunin päästöt erilaisilla polttoaineilla
Henna Hietanen, Jens Kohlmann, AFRY Finland Oy
(16A0913-E0211) 17.11.2021